# The transport and reaction behavior of arsenic in groundwater: Experiments and case studies

Inaugural – Dissertation

zur Erlangung der Doktorwürde der Naturwissenschaftlichen-Mathematischen Gesamtfakultät der Ruprecht – Karls – Universität Heidelberg

> vorgelegt von Diplom - Geologe Rouven Höhn aus Worms, Rheinland – Pfalz

Tag der mündlichen Prüfung: 22.07.2005

Gutachter:

Prof. Dr. Margot Isenbeck-Schröter PD. Dr. Laurence Noel Warr

### Abstract

The transport and reaction behavior of arsenic in the aquatic environment is of high importance, since arsenic in groundwater causes serious problems e.g. in Bangladesh. It is well known that elevated arsenic concentrations in drinking water lead to severe health risk. Various investigations have been carried out concerning the oxidation and adsorption of arsenic in the environment, but less is know about the reduction and release of arsenic.

A continuous injection tracer test was conducted at the USGS Cape Cod research site (Mass.,USA) in order to investigate natural reduction of As(V) to As(III) in an iron reducing environment. As(V) reduction was observed to take place under the conditions of this aquifer. Furthermore retardation of As(III) turned out to be minor than the one of As(V). Microbial investigations showed that arsenic reducing microorganism as well as iron reducers and sulfate reducers were present in the aquifer. Arsenic was assumed to be reduced by microbial reduction as well as by dissolved sulfide. To support this theory batch experiments with As(V) and sulfide were conducted. In preliminary experiments at pH 3, up to 30 % was reduced depending on the sulfide. Water samples buffered a pH 6.8 showed lower reduction to prevent arsenic reduction. In batch experiments with Fe(III), a simultaneous reduction of Fe(III) and As(V) was observed.

The third part of the work deals with the arsenic release from ore and mine dumps. For a better understanding of the flow and the reaction path of arsenic sediment samples from two sites on Sardinia highly influenced by ore and mine dumps were analyzed. In all sediment samples the total amount of arsenic as well as of Ca, Mn, Fe, S and Pb were measured. Different elution methods were conducted to make predictions of the mobility as well as the arsenic binding. In the first study area, the arsenic is transported in a stream in which the flow rate differs extremely over the year. High flow rates cause translocation processes of the tailing material and the stream sediments. During periods with low flow rates the arsenic is released in a wetland and in this case the transport is mostly controlled by geochemical conditions. Thus the As transport velocity is slow and the accumulation rates are much lower compared to the other study area.

### Zusammenfassung

Arsen verursacht in vielen Ländern große Probleme bei der Einhaltung der Trinkwassergrenzwerte. Es ist bekannt, dass Arsen Krankheiten verursacht. Aus diesem Grund ist es wichtig das Reaktions- und Transportverhalten von Arsen zu kennen.

Ein Tracer Test mit kontinuierlicher Injektion wurde auf dem Versuchsgelände des USGS auf Cape Cod (Mass., USA) durchgeführt, um die Reduktion von As(V) zu As(III) unter natürlichen Bedingungen in der Eisenreduktionszone zu beobachten. Außer der Reduktion von As(V) wurde ein schnellerer Transport von As(III) gegenüber dem von As(V) festgestellt. Mikrobiologische Untersuchungen an Proben aus dem Aquifer deuten auf eisen-, arsen- und sulfidreduzierende Organismen hin. In Anbetracht aller Ergebnisse kann das Arsen sowohl von Mikroorganismen als auch von Sulfid reduziert worden sein.

Um die Reduktion von Arsen durch Sulfid genauer zu untersuchen wurden im Labor Schüttelversuche durchgeführt. In ersten Experimenten bei einem pH-Wert von 3 wurde eine Reduktion, je nach eingegebener Sulfidkonzentration, von bis zu 30 % festgestellt. Bei einem pH-Wert von 6,8 waren die Reduktionsraten deutlich geringer. Bei einer Zugabe von Manganoxid war keine Reduktion zu As(III) erkennenbar. Wohingegen bei der Zugabe von Eisenoxid eine gleichzeitige Reduktion von Eisen und Arsen stattfand.

Der dritte Teil der Arbeit befasst sich mit der Freisetzung von Arsen aus Abraumhalden von Erzminen. Um mehr über das Reaktions- und Transportverhalten nach einer Freisetzung von Arsen zu erfahren wurden Sedimentproben aus zwei von Minen stark beeinflussten Gebieten analysiert. Des Weiteren wurden die Proben mit verschieden Chemikalien eluiert, um Aussagen über die Bindung und die Mobilität zu treffen. Im ersten Untersuchungsgebiet wird das Arsen als As(V) bei hoher Fließgeschwindigkeit des Baccu Locci Flusses partikulär an Mineralien und in Zeiten von geringer Fließgeschwindigkeit in gelöster Form transportiert. Im zweiten Untersuchungsgebiet wird Arsen in ein Feuchtgebiet freigesetzt und in gelöster Form transportiert. Die Ausbreitung vom Arsen ist dabei stark von den ändernden geochemischen Bedingungen des Feuchtgebietes abhängig. So wird es während oxischen Bedingungen als As(V) adsorbiert und während anoxischen Bedingungen zu As(III) reduziert und transportiert. Die Akkumulationsraten sind im zweiten Untersuchungsgebiet deutlich geringer als im Ersten. Index of Contents

1. Introduction	1
2. Arsenic in the atmosphere and the aquatic cycle	4
2.1. Atmospheric deposition	4
2.2. Rivers	4
2.3. Lake water	5
2.4. Groundwater	6
3. Processes concerning arsenic cycling in the environment	8
4. Intentions of the studies	. 11
4.1 Tracer test with As(V) to study transport and reaction rates in an iron-reducing environment a	at
the USGS Cape Cod site	. 11
4.2 Mobilization of arsenic from ore and mine dumps	. 12
4.3 Mobilization of arsenic from ore and mine dumps	. 12
5. Arsenic speciation in the presence of dissolved sulfide	. 14
6. Summary of results	. 20
6.1 Tracer test with As(V) to study transport and reaction rates in an iron-reducing environment a	at
the USGS Cape Cod site	. 20
6.2 Arsenic reduction by dissolved sulfide at MnO <sub>2</sub> and Fe <sub>2</sub> O <sub>3</sub> surfaces	. 21
6.3 Mobilization of arsenic from ore and mine dumps: Two case studies from Sardinia (Italy)	. 22
7. Tracer test with Arsenic (V) to study transport and reaction rates in an iron-reducing	
environment at the USGS Cape Cod Site (Mass., USA)	. 23
7.1 Abstract:	. 23
7.2 Introduction	. 24
7.3 Materials and Methods	. 26
7.4 Results and Discussion	. 31
7.5 Conclusions	. 46
7.6 Acknowledgements	. 47
8. Arsenic reduction by dissolved sulfide at $MnO_2$ and $Fe_2O_3$ surfaces	. 48
8.1 Abstract	. 48
8.2 Introduction	. 49
8.3 Materials and methods	. 50
8.4 Results and discussion	. 52
8.5. Conclusions	. 56
8.6 Acknowledgements	. 58

9. Mobilization of Arsenic from Ore and Mine Dumps: Two Case Studies from Sardinia (Italy). 59

9.1 Abstract	59
9.2 Introduction	60
9.3 Description of the study areas and sampling	62
9.4. Analytical methods	
9.5 Results and interpretation	67
9.6 Comparison of the two Sardinian Case Studies	76
9.7 Acknowledgements	77
10. Literature	78
11. Appendix	A1

# List of Figures

Fig.	1 a	a. Keratosis on the soles of an arsenic exposed Bangladeshi man. b. Black foot Disease from a
		patient in Taiwan1
Fig.	21	Partial Eh – pH stability diagram for dissolved arsenic species (VINK, 1996)
Fig.	3	Dionex series 4000i to separate As(III), As(V) and sulfide15
Fig.	4 \$	Separation with 3.45 g L <sup>-1</sup> Ammoniumdihydrogenphospahte
Fig.	5 \$	Separation with 12 g L <sup>-1</sup> Ammoniumdihydrogenphospahte
Fig.	6 \$	Separation with 6 g L <sup>-1</sup> Ammoniumdihydrogenphospahte
Fig.	7 \$	Separation of As(III), As(V) and sulfide in 5 bottles
Fig.	8 /	Automatic sampler collecting samples in five bottles
Fig.	97	Arsenic standards spiked with eluent
Fig.	10	Recovery of arsenic standards separated with IC and measured with FIAS - AAS
Fig.	11	Test site with the 10 multilevel sampling wells ( ) and two core locations ( )
Fig.	12	Preakthrough of bromide, nitrate and iron 1 m downstream
Fig.	13	Fe and Mn concentrations in M2 (injection well) and M3. The concentrations decreased during
		injection due to an input of small amounts of oxygen
Fig.	14	Map of wells with different H <sub>2</sub> S intensities
Fig.	15	Profiles of the arsenic plumes along the flow path directly after stopping the injection at the 30 <sup>th</sup>
		day. a) As(V) b) As(III)
Fig.	16	Profiles of the arsenic plumes along the flow path at the 45 <sup>th</sup> day. a) As(V); b) As(III)
Fig.	17	' Profiles of the arsenic plumes along the flow path at the 63 <sup>th</sup> day. a) As(V); b) As(III)
Fig.	18	Breakthrough curves of As(V) and As(III) measured in the injection well and in the three
		sampling wells in the levels with the highest concentrations. a) M2 (2.27 m MSL) b) M3
		(2.12 m MSL) c) M5 (2.24 m MSL) d) M9 (2.24 m MSL)
Fig.	19	As (tot) concentration from three cores after the sampling was stopped. C0 is a reference core
		taken upstream of the test area. CM2-3 and CM5 are in the flow path of the arsenic plume 43
Fig.	20	As(V) concentrations accumulated in the aquifer sediment at three core locations: C0:
		reference core, C2-3: between M2 and M3, C5: near M5; the four extraction methods
		represent different mobilities of As(V). a) $H_2O$ ; b) hydroxyl ammonium chloride; c) sodium
		ammonium hydrogen phosphate; d) HCI 44
Fig.	21	Schematic model of As(V) reduction by dissolved sulfide in an iron-dominated system 50
Fig.	22	As(III) production from As(V) reduction by different H <sub>2</sub> S concentrations in two different setups
		(* single container setup 2 with 500 ml bottle)
Fig.	23	$^3$ As(III) production from As(V) reduction by H <sub>2</sub> S and Na <sub>2</sub> S at pH 6.8
Fig.	24	Iron dissolution from iron oxides by sulfide
Fig.	25	As(III) formation from arsenate reduction by sulfide on iron oxides
Fig.	26	Schematic model of an iron-dominated system with sulfate reduction with possible overall
		reactions
Fig.	27	Schematic map of the Baccu Locci Stream region with indication of the sampling points 63

Fig.	g. 28 Schematic map of the Palude Sa Masa region with indication of the sampling points and the		
		flow path	65
Fig.	29	Handling of the samples and analyzed parameters	66
Fig.	30	As(V) concentrations in the supernatant water of the BLS sediment samples	68
Fig.	31	Iron concentration extracted by hydroxyl ammonium chloride	69
Fig.	32	Total arsenic concentrations in the sediments of the BLS	70
Fig.	33	Arsenic concentrations, given as percentage of the total amount, extracted from the BLS	
		sediments with (a) $H_2O$ ; (b) hydroxyl ammonium chloride; (c) sodium ammonium hydrogen	
		phosphate. Samples A and B are tailings	71
Fig.	34	(a) Arsenic release over time from a sediment sample by long-term extraction. (b) Arsenic	
		release over time from a tailing sample by long-term extraction.	72
Fig.	35	Correlation between iron and arsenic in samples 1-4	74
Fig.	36	Extracted $As(V)$ and $As(III)$ concentrations of the "source samples" calculated as percentage	of
		the total arsenic concentration.	75

## List of Tables

Tab. 1 The main arsenic containing minerals	3
Tab. 2 Recovery of arsenic standards in eluent solution	. 18
Tab. 3 Small scale hydrodynamic properties of the anoxic test site at Cape Cod estimated by the	
bromide breakthrough curves	. 32
Tab. 4 Background geochemical conditions in the anoxic zone at the Cape Cod aquifer	. 33
Tab. 5 Composition of the samples in the tests of As(V) reduction by dissolved sulfide at reactive	
surfaces	. 51
Tab. 6 Determined rate constants for arsenate reduction by sulfide	. 56
Tab. 7 Chemical composition of the supernatant water of the BLS sediment samples	. 68
Tab. 8 Total concentrations of calcium, iron, sulfur, lead and arsenic from the four groups of the	
Palude Sa Masa samples	. 74

### 1. Introduction

In Bangladesh approximately 30,000,000 people are affected by arsenic contamination of drinking water (WELCH and STOLLENWERK, 2003). Investigations particularly over the last few years have shown that health risk based on the presence of arsenic in the groundwater do not exclusively occur at of the Bengal Basin, but in other countries such as Mongolia, Taiwan, Ghana, Argentina, Chile, Mexico and Great Britain (Cornwall) as well (SMEDLEY and KINNIBURGH, 2002).

Values of the lethal dose of arsenic for human beings range from 0.1 - 0.3 g per 70 kg body weight (Holleman and Wiberg, 1990). Epidemiological studies demonstrated doubtlessly the carcinogenicity of arsenic (LEDERER, 1998). Typical symptoms of arsenic poisoning are: diaphoresis, muscle spasms, nausea, vomiting, abdominal pain, garlic odour to the breath, diarrhoea, anuria, dehydration, hypertension, cardiovascular collapse, aplastic anaemia and death. Dose and time of exposure will determine both the symptoms as well as their impact on human beings.





# Fig. 1 a. Keratosis on the soles of an arsenic exposed Bangladeshi man. b. Black foot Disease from a patient in Taiwan

The element arsenic is already known for a long time. Products containing arsenic as part of an alloy dating into the bronze time have been found (RIEDERER, 1987). Mining of arsenic has been reported from the Egyptians as well as the Chinese (AZCUE and NRIAGU, 1994). Arsenic was also mentioned by Aristotle. During that time arsenic sulfide was used to coat silver in order to give it a golden colour. In these

ancient times this yellow color was used to paint or to depilate. The origin of the word arsenic is dubious. It could either be from the Greek word arsenikon, which was used for the mineral auripigment ( $As_2S_3$ ), or from the Greek word arsenikos = male.

Arsenic is a metalloid and belongs to the fifth main group of the periodic table of elements, the nitrogen group. It has the chemical symbol As, the atomic number 33, an atomic mass of 74.9216 [g mol<sup>-1</sup>], a density of 5.727 g cm<sup>-3</sup> and an electronic configuration of [Ar] $3d^{10}4s^{2}4p^{3}$ . Arsenic can have the oxidation states V, III or –III. <sup>75</sup>As is the only stable isotope in nature. Radioactive isotopes range from <sup>67</sup>As to <sup>86</sup>As and show half-live-times between 1 s an 80 days.

Arsenic is a rare element in the earth crust accounting to approximately 5.5\*10<sup>-5</sup> percent by volume. It is a major component of more than 200 minerals, including elemental arsenic, sulfides, oxides, arsenates and arsenites (Tab. 1) and found e.g. in volcanic rocks, coal, the sea and mineral waters.

Mineral	Composition	Source
Elemental arsenic	As	Hydrothermal veins
Niccolite	NiAs	Vein deposits and norites
Realgar	AsS	Vein deposits, often with orpiment
Orniment	AsaSa	Hydrothermal veins, hot springs, volcanic
orpinient	10203	sublimation products
Cobaltite	CoAsS	High temperature deposits, metamorphic rocks
Arsenopyrite	FeAsS	The most abundant As mineral, mineral veins
Tennantite	$(Cu,Fe)_{12}As_4S_{13}$	Hydrothermal veins
Enargite	Cu <sub>3</sub> AsS <sub>4</sub>	Hydrothermal veins
	As <sub>2</sub> O <sub>3</sub>	Secondary minerale formed by oxidation of
Arsenolite		arsenopyrite, native arsenic and other As
		minerals
Claudetite	<u>م</u> هد)،	Secondary mineral formed by oxidation of
Olaudelite	A3203	realgar, arsenopyrite and other As minerals
Scorodite	FeAsO <sub>4</sub> *2 H <sub>2</sub> O	Secondary mineral
Annabergite	(Ni,Co) <sub>3</sub> (AsO <sub>4</sub> ) <sub>2</sub> * 8 H <sub>2</sub> O	Secondary mineral
Hoernesite	Mg <sub>3</sub> (AsO <sub>4</sub> ) <sub>2</sub> * 8 H <sub>2</sub> O	Secondary mineral, smelter wastes
Conichalcite	CaCu(AsO <sub>4</sub> )(OH)	Secondary mineral
Pharmacosiderite		Oxidation product of arsenopyrite and other As
Tharmacosidente	1 03(//304/2(011/3 011/20	minerals
Arsenian	Fe(S,As) <sub>2</sub>	Vein deposits

Most of the minerals are ore minerals or their alteration products. However these minerals are relatively rare in the environment. The most common arsenic sulfides are realgar ( $As_2S_2$ ), auripigment ( $As_2S_3$ ) and arsenopyrite (FeSAs) (SCHUMANN, 1985). But the most important natural source of arsenic is arsenian (arsenic rich pyrite Fe(S,As)<sub>2</sub>) (NORDSTROM, 2000). Arsenic is highly associated with deposits of many metals and therefore known as a good deposit indicator in prospection (KABATA-PENDIAS, 2001).

### 2. Arsenic in the atmosphere and the aquatic cycle

Arsenic e.g. impacted by mining activities or the use of arsenical pesticides may lead to severe environmental hazards if mobilized. It may be distributed by various media within the aquatic cycle and the atmosphere. Its potential impact on drinking water, commonly derived from ground- or surface water (rivers, lakes), is most important concerning the threat to human health. Groundwater e.g. may contain 200 - 1200  $\mu$ g L<sup>-1</sup> arsenic (AHMED et al., 2004; MCARTHUR et al., 2004; ZHENG et al., 2004), contrasting with the current WHO threshold value for arsenic in drinking water, which amounts to 10  $\mu$ g L<sup>-1</sup> (WHO, 1993).

#### 2.1. Atmospheric deposition

In unpolluted areas the concentration of arsenic in atmosphere is relativly low ranging from  $10^{-5}$  to  $10^{-3} \ \mu g \ m^{-3}$ . Wind, erosion, volcanic emissions, marine aerosols and pollution represent the main sources. The most important anthropogenic emissions result from smelter operations and fossil fuel burning. The atmospheric arsenic mainly consists of As<sub>2</sub>O<sub>3</sub> dust particles (CULLEN and REIMER, 1989). In general the concentrations of arsenic in the atmospheric deposition are relatively low. Normally rainwater has concentrations less than 0.5  $\mu g L^{-1}$  (BARBARIS and BETTERTIN, 1996; SCUDLARK and CHURCH, 1988). Andreae (ANDREAE, 1980) reported arsenic concentrations of 0.5  $\mu g L^{-1}$ , when the rainwater could have been affected by smelting and coal burning.

### 2.2. Rivers

The arsenic concentration in river water varies according to the composition of surface recharge, the contribution of baseflow and the bedrock lithology. Background concentrations range from  $0.1 - 0.8 \ \mu g \ L^{-1}$  (ANDREAE et al., 1983; FROELICH et al., 1985; SEYLER and MARTIN, 1991).

High arsenic concentrations in rivers have been reported from areas with geothermal sources or arsenic contaminated groundwater. Typical concentrations of arsenic in river waters from geothermal areas vary around  $10 - 70 \ \mu g \ L^{-1}$  (MCLAREN and KIM, 1995; NIMICK et al., 1998; ROBINSON et al., 1995). In the Madison River

(Wyoming and Montana), arsenic concentrations up to 370 µg L<sup>-1</sup> have been reported as a result of the geothermal activity in the Yellowstone area (NIMICK et al., 1998). Some rivers show seasonal variations in the arsenic concentration. The Madison River has the highest arsenic concentration during its low-stand phases (NIMICK et al., 1998). In the Waikato River (New Zealand) the highest arsenic concentration has also been found in the summer months. An increase of the arsenic concentration has been successfully correlated to a temperature controlled microbial reduction of arsenic (McLAREN and KIM, 1995).

A significant increase of arsenic concentration may also occur as a consequence of pollution from industrial or sewage runoff. Andreae and Andreae (ANDREAE and ANDREAE, 1989) found arsenic concentrations up to  $30 \ \mu g \ L^{-1}$  in the Zenne River, Belgium, which was affected by industrial sources and sewage.

The arsenic concentration in rivers can be increased by mine waste and tailings. Azcue and Nriagu (Azcue and NRIAGU, 1995) report concentrations up to 0.7  $\mu$ g L<sup>-1</sup> from the Moira River, Ontario upstream of a gold mine. Downstream the concentration increased up to 23  $\mu$ g L<sup>-1</sup> as a result of tailings outwash caused by the mine. (Azcue et al., 1994) found concentrations up to 556  $\mu$ g L<sup>-1</sup> (in the average 17.5  $\mu$ g L<sup>-1</sup>) in streams close to mine tailings in British Columbia. Smedley et al. (SMEDLEY et al., 2001) reports high arsenic concentrations (around 200 - 300  $\mu$ g L<sup>-1</sup>) in surface waters related to tin- and goldmines but furthermore those pollutions display relatively high concentrations of arsenic, in most cases the area affected is relatively small. This is the result of the strong adsorption affinity of arsenic to oxidic minerals, especially iron oxide. The sources for arsenic in mines are mostly Fe(II) sulfide minerals.

### 2.3. Lake water

In general arsenic concentrations in lake waters are relatively low (<1  $\mu$ g L<sup>-1</sup>) (Azcue and NRIAGU, 1995). Only if the rivers running into the lakes are contaminated by geothermal- or mine activity arsenic concentration may increase, especially in mining areas up to 500  $\mu$ g L<sup>-1</sup>(BRIGHT et al., 1996). In geothermally active areas concentrations up to 1000  $\mu$ g L<sup>-1</sup> have been reported (BENSON and SPENCER, 1983). Sometimes the arsenic contamination in lakes affected by mining is not detectable, because iron oxides adsorb the arsenic under neutral to mildly acidic conditions. In

Canadian lake waters affected by mining arsenic concentrations, similar to those unaffected by mining, around 0.3  $\mu$ g L<sup>-1</sup> were found (AZCUE et al., 1994).

High arsenic concentrations are also found in some alkaline basins as a result of extreme evaporation. In Mono Lake California, USA, concentrations from 10 to 15 mg L<sup>-1</sup> of dissolved arsenic at pH values of 9.5 - 10 have been reported as a result of inputs from geothermal springs and the weathering of volcanic rocks followed by evaporation. In this lake, a simultaneous reduction of arsenate and sulphate by microorganisms was observed (OREMLAND et al., 2000).

In lake sediments, the oxidation of arsenic has been attributed to manganese oxides. The elimination of the dissolved oxygen and the suppression of microbial activity had no effect on the oxidation rate, whereas a removal of the manganese oxides by hydroxylamine, hydrochloride or sodium acetate lowered the oxidation rate (OSCARSON et al., 1980; OSCARSON et al., 1981).

### 2.4. Groundwater

Arsenic concentration in the groundwater used as drinking water is important for human health. Drinking arsenic enriched groundwater is the primary source of arsenic uptake. Therefore a lot of investigations concerning the transport behavior of arsenic in groundwater have been conducted. The World Health Organization has set a guideline for drinking water with a arsenic concentration of 10  $\mu$ g L<sup>-1</sup> (WHO, 1993). In most countries the background concentration of arsenic in groundwater is less then 10  $\mu$ g L<sup>-1</sup> and sometimes even substantially lower (WELCH et al., 2000). Arsenic concentrations of groundwater samples reported in the literature display a wide range from 0 – 5000  $\mu$ g L<sup>-1</sup>. High concentrations of arsenic are found in a variety of environments, affected by geothermal-, mining-, industrial activities and geogenic release.

In Bangladesh and Bengal more than 30.000.000 people are affected by arsenic rich groundwater (HARVEY et al., 2002; SMEDLEY and KINNIBURGH, 2002). In the last decade a lot of investigations have been made to elucidate the origin and the arsenic flow path in this region (WELCH and STOLLENWERK, 2003). All studies dealing with Bangladesh are in good agreement that the arsenic originates from natural background sources. Nevertheless the processes leading to the release of arsenic are not yet fully understood. Ravenscroft et al. (RAVENSCROFT et al., 2001) reported

three different theories concerning the release of arsenic. 1. arsenic release by oxidation of arsenic-bearing pyrite (*Oxidation Theory,* (MALLICK and RAJAGOPAL, 1996; MANDAL et al., 1996), 2. arsenic release by competitive exchange of phosphate from fertilizers (ACHARYYA et al., 2000) and 3. arsenic release by reduction of iron hydroxides under anoxic conditions (Reduction Theory, (BHATTACHARYA et al., 1997; MCARTHUR et al., 2001; NICKSON et al., 1998; NICKSON et al., 2000)). Ravenscroft et al. (RAVENSCROFT et al., 2001) confirm the *Reduction Theorie* and correlated the arsenic distribution in the Bengal Delta with peat deposits of Pleistocene and Holocene age.

It is important to know the ongoing processes in the environment therefore the next chapter goes more in detail concerning the reactions of arsenic.

### 3. Processes concerning arsenic cycling in the environment

The distribution of arsenic in the environment is depending on the partitioning between the aqueous and solid phase. The main processes controlling the distribution are:

- mineral precipitation  $\leftrightarrow$  dissolution.
- adsorption ↔ desorption
- oxidation  $\leftrightarrow$  reduction

Arsenic occurs in several oxidation states (-III, 0, +III and +V). In the environment it is mainly found in inorganic form as arsenite (As III) or arsenate (As V). Redox milieu and pH are the most important factors controlling the arsenic speciation. In an oxic environment the dominant species is the As(V) existing as H<sub>3</sub>AsO<sub>4</sub> at pH < 2, at pH between 2 and 6.9 as H<sub>2</sub>AsO<sub>4</sub><sup>-</sup> or as HAsO<sub>4</sub><sup>2-</sup> once the pH exceeds 6.9. Under reducing conditions and pH less than 9.2 the uncharged arsenic species H<sub>3</sub>AsO<sub>3</sub> will be predominant. In Fig. 2 the species distribution is given in pure water as a function of pH and Eh.



Fig. 2 Partial Eh – pH stability diagram for dissolved arsenic species (VINK, 1996).

In experiments with contaminated soils under different pH and Eh conditions the distribution of the two arsenic species was observed (MASSCHELEYN et al., 1991). A higher concentration of arsenite under reducing conditions and a higher concentration of dissolved As(III) in the solutions at lower pH were demonstrated. The dominance of As(III) in strong reducing aquifers (iron- and sulfate reduction) could be seen in reducing arsenic rich groundwaters from Bangladesh. The arsenic species have As(III) / As (total) ratios between 0.1 - 0.9 (DPHE, 1999), usually in the range of 0.5 - 0.6. In the Huhhot Basin (Inner Mongolia) As(III) / As (total) ratios between 0.7 - 0.9 were found (SMEDLEY et al., 2001).

In the absence of oxygen, As(III) is oxidized very fast at MnO<sub>2</sub> surfaces (CHIU and HERING, 2000; MOORE et al., 1990; SCOTT and MORGAN, 1995; TOURNASSAT et al., 2002). Manganese oxides are used in order to improof the removeability of As(III) from drinking water (DRIEHAUS et al., 1995). Wherease the oxidation at FeOOH is relatively slow (TRETNER et al., 2005). Also with pure oxygen at pH 7 As(III) is oxidized very slow (CHERRY et al., 1979). After 2.5 months just 5-7 % of the arsenic was oxidized. In water equilibrated with atmospheric air a half life for arsenic oxidation of 1 - 3 years was measured (EARY and SCHRAMKE, 1990).

Less is known about the solid phase reduction of arsenic. There are some studies with sediments in soils which showed under reducing conditions (Eh < 100 mV) an arsenic reduction in days or several weeks (MASSCHELEYN et al., 1991; REYNOLDS et al., 1999). Masscheleyn et al. (MASSCHELEYN et al., 1991) found a release of arsenic before the iron was released, implying reductive desorption from iron oxides rather than reductive solution.

In the presence of high concentrations of dissolved sulfur, arsenic sulfide species can be of importance. Reducing acidic conditions favor the precipitation of orpiment  $(As_2S_3)$  and realgar (AsS). Thus high arsenic concentrations are not to be expected in waters with high concentrations of free sulfides (MOORE et al., 1988).

Authigenic pyrite is found in sediments of many rivers, lakes and aquifers (WOLTHERS et al., 2000). Under aerobic conditions pyrite is not stable and gets oxidized. Thereby iron, sulfate and trace constituents like arsenic are released and acidity is increased. These processes called "acid mine drainage" (AMD) could be observed in mine areas. The ongoing processes of two case studies are discussed

more intensively in the chapter: Mobilization of Arsenic from Ore and Mine dumps: Two Case Studies from Sardinia (Italy).

In sediments arsenic is mainly bound to metal oxides such as iron and manganese oxides (AMITA et al., 1999; BOWELL, 1994; CHIU and HERING, 2000; MANNING et al., 2002; RÜDE, 2001; SCOTT and MORGAN, 1995; SUN and DONER, 1998; TOURNASSAT et al., 2002; WALTHAM and EICK, 2002). Based on their wide distribution and strong sorption affinity, iron oxides are the most important adsorbents of arsenic in the environment. It has been reported that within 24 h 93 % of arsenate sorbed onto goethite at a pH of 6. (O'REILLY et al., 2001a). Fe(III) precipitation seems to enhance the amount of As(V) sorbed by a significant factor (Fuller et al., 1993).

One reason for high arsenic concentration in groundwater could be the release of adsorbed arsenic during iron reduction. Therefore processes causing changes in the iron redox chemistry need to be considered since they directly affect the mobility of arsenic.

### 4. Intentions of the studies

Three different investigations are described in this dissertation. The first was a field experiment conducted at the Cape Cod aquifer (Massachusetts, USA) in cooperation with Dr. James Davis and Dr Douglas Kent from the U.S. Geological Survey. In this experiment As(V) was injected into the iron reducing zone to investigate arsenic reduction.

The findings of this field experiment led to the assumption, that arsenic is pathly reduced by sulfide. To study this hypothesis batch experiments were conducted in the laboratory to examine reduction of arsenic by sulfide in cooperation with Prof. Dr. Dirk Wallschläger (Trent University, Canada).

During these laboratory experiments with arsenic and dissolved sulfide a new method had to be developed because of analytical problems. This analytical method is not described in the manuscript (chapter 8) but in chapter 5.

In the third part of the work two areas affected by arsenic release from ore and mine dumps were studied in cooperation with Prof. Dr. Maria Boni (Università di Napoli "Federico II", Italy) and Prof. Dr. Franco Frau (Università di Cagliari, Italy). Such areas are highly contaminated with arsenic and thus perfect to improve the knowledge from experiments.

# 4.1 Tracer test with As(V) to study transport and reaction rates in an iron-reducing environment at the USGS Cape Cod site

For a better understanding of the processes driving the reduction of As(V), a tracer test was conducted in the iron reducing zone of the Cape Cod aquifer. The aquifer is split into an oxic, suboxic and anoxic zone because of an inflow of sewage. Properties of the aquifer are well known because of the numerous investigations (BARBER et al., 1992; KENT and FOX, 2004; LEBLANC et al., 1991). The aquifer material mainly consists of quartz (95%) with coatings of iron, manganese and aluminum hydroxides. The intention of the test was to observe a reduction of As(V) to As(III) in an iron reducing environment, to find out possible reduction partners and to examine the transport behavior of As(III) and As(V) in an iron reducing environment. In the literature, there are a couple of evidences about arsenic reduction by microorganisms (NIGGEMEYER et al., 2001; ZOBRIST et al., 1998; ZOBRIST et al., 2000).

A continuous injection method for the tracer test was used to give arsenic reducing microorganism the chance to grow. Water samples were taken over a period of 3 months with assistance from Steffen Jann, Dr. Volker Niedan and Susanne Stadler. After the experiment was finished, three cores were taken to study the aquifer material. Dr. James Davis and Dr. Dougles Kent from the USGS supported the experiment with know-how and material. All samples were measured in the laboratories of the University of Heidelberg. Analytical work supported by Stefan Rheinberger, Christian Scholz, Andreas Tretner and Volker Wild. Microbial studies of the test site were performed at the MIT. In cooperation with Rasmus Jakobsen, H<sub>2</sub> concentrations were measured in the anoxic zone of the aquifer.

The methods and the results of the tracer test are discussed in more detail in chapters 6 and 7: Tracer test with Arsenic (V) to study transport and reaction rates in an iron-reducing environment at the USGS Cape Cod Site (Mass., USA).

### 4.2 Mobilization of arsenic from ore and mine dumps

The results of the Cape Cod tracer test led to another aspect of As(V) reduction. Beside the reduction of arsenic by microorganism a second possible process is the reduction by dissolved sulfide. Batch experiments with sulfide and As(V) were conducted in the laboratory to study As(V) reduction and to give hints concerning the importance of this process in natural environments. In a natural system, such as the Cape Cod aquifer, As(V) is bound onto oxidic minerals before it is reduced. In order to study changes of the reduction rate under different conditions arsenic experiments were made at different pH-condition and in the absence or presence of oxidic minerals under inert atmosphere. Some samples were sent to Prof. Dr. Dirk Wallschläger (Trent University, Canada) who analyzed the samples with regard to arsenic sulfide complexes. The experimentL setup, the methods and the results of these experiments are described in the chapters 6 and 8: Arsenic reduction by dissolved sulfide at MnO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub> surfaces.

### 4.3 Mobilization of arsenic from ore and mine dumps

Mobilization of arsenic from ore and mine dumps: Two case studies from Sardinia (Italy)

The third part of the work deals with two field studies where arsenic and sulfide are released from ore and mine dumps. World wide, a lot of regions are affected by this problem known as acid mine drainage "AMD". In the majority of cases arsenic concentrations in such areas are relatively high. This project was carried out to study the arsenic distribution under different conditions and to improve the knowledge of possible reactions. The two sites differ in hydraulic, physical and geochemical conditions. The first study area is influenced by the Baccu Locci stream. The question was whether the arsenic is dislocated by particle transport or which geochemical reactions governed the arsenic distribution. The study of the Baccu Locci area was done in cooperation with Dr. Franco Frau and Silvia Pontis (University of Cagliari, Italy) Silvia Pontis did the sampling and supported laboratory work at Heidelberg University.

In the Paluda Sa Masa region the wetland sediments are contaminated with arsenic. The question in this area was whether concentrations and arsenic mobility of samples give indications of possible sources and distribution processes. The investigations were conducted with Prof Dr. Maria Boni and her student Marcello Romano who did the sampling (Universita di Napoli, Italy). The results of both case studies are describes and compared in chapters 6 and 9: Mobilization of Arsenic from Ore and Mine Dumps: Two Case Studies from Sardinia (Italy).

### 5. Arsenic speciation in the presence of dissolved sulfide

All samples from the Cape Cod tracer test were measured with a FIAS-AAS (flow injection analysis system - atomic absorption spectrometer) (AAS: Perkin-Elmer 4100) (FIAS: Perkin-Elmer 200) by determination of As(III) and As(tot) in two measurements. After both measurements the As(V) concentration was calculated (1).

As(V) = As(tot) - As(III) (1)

This calculation was made iteratively, to consider the small amounts of interference between As(III) and As(V). The interference was defined by measuring different standards from the contrariwise species during each method.

At first, As(III) was measured. In the FIAS the samples were mixed with 2 g L<sup>-1</sup> NaBH<sub>4</sub>, 0.2 g L<sup>-1</sup> NaOH and 4 M HCI. The flow rate of the sample was 2 ml min<sup>-1</sup>, NaBH<sub>4</sub> + NaOH 1.6 ml min<sup>-1</sup> and HCI 1.6 ml min<sup>-1</sup>. In a gas liquid separator arsine was purged and transported in an argon flow to the quartz cuvette (argon flow rate 80 ml min<sup>-1</sup>). The cuvette converts the arsine at a temperature of 900 C° to elemental arsenic, which is detected with an AAS (Perkin-Elmer AAS 4100) at a wavelength of 193.7 nm (RÜDE, 1996). Afterwards the As(tot) concentration was measured. For this measurement the samples had to be reduced first to As(III) using a solution of 100 g L<sup>-1</sup> KI and 100 g L<sup>-1</sup> ascorbic acid mixed with 6 M HCI and sample in the ratio of 1:4:5. After 30 minutes the samples were measured as described above.

After the first test with sulfide we noticed that the measuring method did not work in the presence of sulfide. The sulfide damaged the quartz cuvette and so the arsine could not be converted to elemental arsenic any more.

To solve this problem an IC (ion chromatography) (Dionex series 4000i) was connected with an ICP-OES (Inductive coupled plasma optical emission spectroscopy). The detection limit for arsenic measurment with the OES is too high (in the range of mg L<sup>-1</sup>). Thus the IC had to be coupled with the FIAS-AAS. Normally the samples were reduced over a period of 30 minutes with the reduction solution to As(III). But this was not practicable working with coupled instruments. To shorten the reduction time did not work either. The problem could be solved by separating the samples with an IC (Fig. 3) and measure them afterwards with the "old" method using the FIAS-AAS.



Fig. 3 Dionex series 4000i to separate As(III), As(V) and sulfide.

For the separation a guard column (Dionex IonPac AG7 (4 x 50 mm)) and an analytical column (IonPac AS7 (4 x 250 mm)) were used. Ammonium dihydrogen phosphate ( $H_6NO_4P$  99%) turned out to be the best eluent for the separation of As(III), As(V) and sulfide.

In order to optimize the concentration of the eluent, the IC was coupled with the ICP-OES. High concentrations of arsenic standards (10 mg  $L^{-1}$ ) were tested with different concentrations of the eluent (3.45 g  $L^{-1}$ , 12 g  $L^{-1}$  and 6 g  $L^{-1}$ ).



Fig. 4 Separation with 3.45 g L<sup>-1</sup> Ammoniumdihydrogenphospahte



Fig. 5 Separation with 12 g L<sup>-1</sup> Ammoniumdihydrogenphospahte



Fig. 6 Separation with 6 g L<sup>-1</sup> Ammoniumdihydrogenphospahte

Using a concentration of 3.45 g L<sup>-1</sup>, the separation time of the two arsenic species was too long (Fig. 4). After increasing the eluent concentration up to 12 g L<sup>-1</sup> the two peaks overlapped (Fig. 5). A clear separation of the two arsenic species was received at an eluent concentration of 6 g L<sup>-1</sup> and a flow rate of 0.9 ml min<sup>-1</sup> (Fig. 6).

In the next step the retention time of sulfide with these settings was tested. The sample was split into different bottles after the IC, and the sulfide was measured in the bottles with a probe. The retention time of sulfide was longer than that of the two arsenic species (Fig. 7).



Fig. 7 Separation of As(III), As(V) and sulfide in 5 bottles

To simplify the separation method an automatic sampler controlled by a computer was installed behind the IC and each sample was divided temporally into five bottles (Fig. 8). In the second bottle the As(III) species was detected, in the fourth the As(V) species and in the fifth was the sulfide.



Fig. 8 Automatic sampler collecting samples in five bottles.

The next step was to control changes in the arsenic species by the eluent. Arsenic standards (5  $\mu$ g L<sup>-1</sup> and 10  $\mu$ g L<sup>-1</sup> of As(III) and As(V)) were spiked with the eluent and afterwards threefold measured with the FIAS-AAS. Each arsenic species was arranged three times in two concentrations (Fig. 9).



#### Fig. 9 Arsenic standards spiked with eluent.

	5 µg/l	
As(V)	90,0 ± 2,7 %	99,2 ± 2,2 %
As(III)	99,6 ± 2,2 %	98,1 ± 2,2 %

Tab. 2 Recovery of arsenic standards in eluent solution

No negative effect at the arsenic species was found by spiking the eluent to the standards. The recovery rate of the spiked arsenic standards was between 90 % and 99.6 % (Tab. 2).

The next step was to recover arsenic standards after the run through the IC. The dilution factor was calculated from the flow rate, the sample loop and the time of collecting the sample (2 and 3)

dilution factor = 
$$\frac{\text{flow rate} \cdot \text{time of sampling}}{\text{sample loop}}$$
 (2)  
dilution factor =  $\frac{\left[\frac{\text{ml}}{\text{min}}\right] \cdot \left[\text{min}\right]}{[\text{ml}]}$   $\frac{0.9 \left[\frac{\text{ml}}{\text{min}}\right] \cdot 6 \left[\text{min}\right]}{0.141 \left[\text{ml}\right]} = 38.3$  (3)

The dilution factor from the IC was 38.3. In the first tests the arsenate was underestimated. After changing the sampling time for As(V) to six minutes and using new standards the recovery was more than 90 %. An error of 10 % by the measurement was also calculated by the Cape Cod samples with the "old method"

so the new setup was used to measure the sample from the test with dissolved sulfide (Fig. 10).



Fig. 10 Recovery of arsenic standards separated with IC and measured with FIAS - AAS

### 6. Summary of results

The combination of laboratory studies, field experiments and field investigations leads to a comprehensive overview concerning the reaction and transport behavior of arsenic in the aquatic environment. The following summary of results gives a short overview of the main results and conclusions. Each study was conducted in cooperation with international scientists and the manuscripts have been submitted to international scientific journals.

# 6.1 Tracer test with As(V) to study transport and reaction rates in an iron-reducing environment at the USGS Cape Cod site

The tracer test with As(V) in the iron reducing zone at the Cape Cod site contributes the first results concerning the question of As(V) reduction and transport in a real aquifer system under controlled input conditions. During this experiment, a reduction of the injected As(V) to As(III) was observed. As(III) was significantly more mobile under the anoxic conditions of the Cape Cod aquifer than As(V), which accumulated at the aquifer material.

A continuous injection was chosen to create stable conditions for a period of four weeks in order to improve growth of arsenic reducing microorganisms. MPN (Most Probable Number) tests of sulfate-, iron-, and arsenic reducers conducted at the MIT showed that populations of each were present at the Cape Cod site.

The experiment has to be divided into two periods. During the input period, nitrate as well as small amounts of oxygen entered the aquifer changing the iron reducing conditions into oxic/suboxic conditions. This led to a fast oxidation of dissolved iron. Following the precipitation of iron hydroxides phosphate as well as the injected As(V) accumulated in the input area. After the injection was stopped, the anoxic conditions re-established and the adsorbed As(V) was released and reduced under anoxic conditions. The As(III) was more mobile under the anoxic conditions than As(V), which accumulated at the aquifer material. The As(V) was predominant in extractions from the core material originating from the test area. Distribution of the As(V) at the sediment material illustrate the contact zone with the arsenic plume.

During the sampling  $H_2S$  was smelt in some tubes indicating an ongoing sulphate reduction. Even though it could not be detected in the water, a sulphate reduction can be supported by  $H_2$  measurements.

Thus we concluded that both the reduction of As(V) by microorganisms as well as by dissolved sulfide could be important processes in the Cape Cod aquifer.

### 6.2 Arsenic reduction by dissolved sulfide at MnO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub> surfaces

To improve this theory several batch experiments were conducted with As(V) and dissolved sulfide. Indications about a reduction of arsenic by sulfide could be found in the literature (CHERRY et al., 1979; ROCHETTE et al., 2000). The first tests were conducted with low concentrations of H<sub>2</sub>S gas and As(V) in an unbuffered solution. After 30 days 7-30% of the As(V) was reduced to As(III) depending on the injected sulfide concentration. In the following experiments the solution was buffered at a pH of 6.8 and Na<sub>2</sub>S salt as well as H<sub>2</sub>S gas were used. At this pH the reduction rate of As(V) was 20-50 times lower then in the experiments at pH 3. To adapt the experiments more to the natural conditions observed in the Cape Cod aquifer, the As(V) was first adsorbed at manganese or iron oxides. In the samples with manganese oxides no significant reduction of As(V) could be observed. In contrast in the setup with iron oxides a slight production of As(III) was detected. In the test with Fe<sub>2</sub>O<sub>3</sub> the reduction rates of As(V) were lower than in solutions due to the fact that the oxides were also reduced, thus the availability of sulfide was lower.

These experiments support the concept that As(V) reduction by dissolved sulfide was important during the Cape Cod anoxic tracer test. We conclude that sulfide is one important factor in this environment.

In some studies from the Bengal delta indications could be found about an ongoing sulfide reduction parallel to arsenic reduction (AHMED et al., 2004; ZHENG et al., 2004). This can be supported by our results. We conclude that environments where small amounts of sulfide may reach iron reducing conditions, a reduction of As(V) is obvious (MCARTHUR et al., 2004; RAVENSCROFT et al., 2001; WAGNER et al., 2005).

# 6.3 Mobilization of arsenic from ore and mine dumps: Two case studies from Sardinia (Italy)

The findings from the two case studies on Sardinia generate various conclusions about the factors governing the distribution of arsenic originating from ore and mine dumps. In both areas the acidity is totally buffered by the carbonate rocks so that pH values were between 7 and 8. Therefore, the direct affect of acid mine drainage, which is acidity, was absent. The distribution of arsenic is highly influenced by the transport conditions. A river, like in the Baccu Locci study area with its high erosion power and dominant physical transport is able to leach and transport high amounts of arsenic mostly bound to minerals. Due to the oxic conditions in both the stream water and the sediments, only As(V) could be observed with high concentrations decreasing over the transport distance. Results from different extraction methods showed an increase of mobility over the distance. The flow rate of the river varies extremely over the year and thus the amount of transported arsenic varies too. During times with high flow rates sediment transport dominants the translocation of arsenic whereas with low flow rates the arsenic is mainly transported in the dissolved fraction.

In the other study area arsenic distribution in a wetland was studied. The arsenic concentrations were about 20 times lower compared to the Baccu Locci region. The wetland is mainly fed by two creeks which have low concentrations of arsenic. The arsenic sources in this study area are the mine dumps on the hills in the north-west of the wetland.

The distribution of arsenic in the wetland is affected by external influences. In regions where the wetland is influenced by mine dumps the arsenic concentration is high, whereas in regions where the wetland is affected by rivers the arsenic concentration is relatively low. As in the Baccu Locci area, the concentration of arsenic in the wetland decreases with flow distance, whereas the mobility increases. Also on this site In the arsenic transport is extremely influenced by seasonal changes of hydrological conditions. During rain periods the sediments become water saturated so that the redox conditions get anoxic and iron reduction occurs. Together with the iron also the As(V) is reduced and transported as As(III) which is more mobile. During dry periods, iron and arsenic are oxidized and precipitate. Thus the arsenic transport is slow and the accumulation rates are much lower compared to the Baccu Locci area.

7. Tracer test with Arsenic (V) to study transport and reaction rates in an iron-reducing environment at the USGS Cape Cod Site (Mass., USA)

R. Höhn<sup>1</sup>, M. Isenbeck-Schröter<sup>1</sup>, D. Kent<sup>2</sup>, J. Davis<sup>2</sup>, R. Jakobsen<sup>3</sup>, S. Jann<sup>4</sup>,
V. Niedan<sup>5</sup>, C. Scholz<sup>1</sup>, S. Stadler<sup>6</sup>, A. Tretner<sup>7</sup>
<sup>1</sup>Institute of Environmental Geochemistry, University of Heidelberg
<sup>2</sup>United States Geological Survey, Menlo Park, CA
<sup>3</sup>E&R; Technical University of Denmark, Lyngby
<sup>4</sup>Institute of Soil Science, Technical University Munich

<sup>5</sup>Lonza, Switzerland

<sup>6</sup>Technical University Bergakademie Freiberg

<sup>7</sup>Inorganic and Isotope Geochemistry, GeoForschungsZentrum Potsdam

## 7.1 Abstract:

Transport of arsenic(V) in groundwater can be affected by oxidation-reduction (redox) and sorption reactions. To study transport and reaction rates under field conditions, a small-scale tracer test was performed in an anoxic, iron-reducing zone of a sandy aguifer at the USGS research site on Cape Cod, Massachusetts, USA. For four weeks, a stream of groundwater with added As(V) (6.7 µM) and bromide (1.6 mM), which is chemically inert, was injected in order to observe the reduction of As(V) to As(III) and other reactions influencing arsenic transport. Breakthrough of bromide (Br<sup>-</sup>), As(V), and As(III) was observed at various locations downstream of the injection over a period of four months. Transport velocities of the Br<sup>-</sup> as well as the two arsenic species were characterized. After a short lag period, nitrate and small concentrations of dissolved oxygen in the stream of injected groundwater oxidized ferrous iron, resulting in precipitation of hydrous iron oxides. During the period of active iron oxidation, As(V) concentrations decreased to near the detection limit, indicating that As(V) was bound onto the freshly formed hydrous iron oxides. Once dissolved iron concentrations decreased to below detection, nitrate concentrations increased and As(V) was observed to undergo extensive retardation during transport.

Approximately 1 week after terminating the injection, disappearance of nitrate and oxygen and increasing concentrations of dissolved iron indicated that anoxic conditions had been re-established in the vicinity of the injection. Shortly after anoxic conditions had been re-established, decreases in As(V) concentrations and increases in As(III) concentrations were observed within 1 meter of the injection. A period followed during which decreases in As(V) concentrations and increases in As(III) concentrations was observed in this region, after which As(V) and As(III) concentrations remained relatively constant through the end of the experiment. The latter results suggest that a quasi steady state was achieved between slow release of As(V) from hydrous Fe oxide precipitates and loss of As(V) by reduction to As(III) and transport processes. Downstream of the near-injection region, As(V) transport was dominated by reduction and extensive adsorption. Arsenic(III) generated by the reduction of As(V) underwent adsorption during transport. Sediment assays indicated the presence of As(V) reducing microorganisms, indicating the possible importance of As(V) reduction organisms; elevated concentrations of hydrogen above background values near the injection and the possible existence of low concentrations of sulfide suggest that reduction by sulfide generated by sulphate reduction may have contributed to the observed As(V) reduction. These findings demonstrate that oxidation of Fe(II) in anoxic groundwater by nitrate or dissolved oxygen can greatly attenuate concentrations of As(V) and As(III) over relatively short time periods, that As(V) sequestered in hydrous iron oxide precipitates can be released under anoxic conditions, and that reduction of As(V) to As(III) can be fast enough to cause significant decreases in As(V) concentrations over short transport distances.

## 7.2 Introduction

High natural arsenic content in groundwater is often correlated with high iron concentrations due to iron reducing conditions (CUMMINGS et al., 1999; HAURY et al., 2000; NIGGEMEYER et al., 2001; ZOBRIST et al., 2000). The arsenic in these groundwaters is supposed to be in the reduced arsenic (III) (As(III)) form. It is well known that the As(III) is mobile and leads to severe drinking water problems. This is especially the case in countries, where the raw water is used without control or clean up, e.g. India, Bangladesh and other developing countries (HARVEY et al., 2002;

RAVENSCROFT et al., 2001; SMEDLEY and KINNIBURGH, 2002; WELCH and STOLLENWERK, 2003). The arsenic sources are mostly geogenetic, e.g. arsenic bearing pyrite (COURTIN-NOMADE et al., 2003; FUKUSHI et al., 2003; GALÁN et al., 2003; JOHNSON and THORNTON, 1987). The transport and reaction pathways of the arsenic into the groundwater are not yet sufficiently investigated. One possible pathway is the oxidation of pyrite, finally to Fe(III), sulfate and arsenic (V) (As(V)). The iron (III) is fixed in hydroxides or hydroxosulfates (e.g. hydrous ferric oxide, goethite, jarosite) and As(V) is sorbed onto the surfaces or incorporated into these minerals (AMITA et al., 1999; BOWELL, 1994; DZOMBAK and MOREL, 1990; FULLER et al., 1993; MANNING et al., 2002; O'REILLY et al., 2001a; RÜDE, 2001; XIAOHUA and DONER, 1996). When iron reducing conditions establish, high amounts of As(III) are released into the water together with the Fe(II).

Microorganisms which use As(V) as terminal electron acceptor during the degradation of organic matter have been identified in arsenic-rich aquatic environments and in laboratory investigations (AHMANN et al., 1997; DOWDLE et al., 1996). Chemical reduction processes by redox sensitive phenolic substances which also reduce Fe(III), have been observed, too (HAURY et al., 2000). Also the sulfide originating from sulfate reduction processes can reduce the Fe(III) as well as the As(V) (ROCHETTE et al., 2000; VAN CAPPELLEN and WANG, 1996).

In order to study the As(V) reduction in an iron reducing environment under field conditions, a tracer test was performed at the United States Geological Survey (USGS) groundwater research site on Cape Cod, near Falmouth, Massachusetts, USA. The aim of the investigation was to find out possible reaction partners and reaction rates and to characterize the transport behavior of the As(V) and As(III) species under anoxic conditions. This study was part of a series of small scale field experiments to investigate the influence of redox and adsorption reactions on the transport of As in groundwater. First results of these tests including two tracer tests where As(III) was injected into oxic and suboxic groundwater are described elsewhere (HÖHN et al., 2001; STADLER et al., 2001).

## 7.3 Materials and Methods

### 7.3.1 Site description

The hydraulic properties and the geochemical conditions of the test site Cape Cod have been characterized in detail (DAVIS et al., 1991; LEBLANC et al., 1991). A shallow, unconfined aguifer resides in the uppermost 30 m of the sediments, which consist of permeable, stratified glacial sand and gravel deposits. Hydraulic conductivities are in the range of  $10^{-3}$  m s<sup>-1</sup>, the mean gradient is 0.0015 and the effective porosity is 0.39. These conditions result in a mean flow velocity of 0.3 to 0.4 m d<sup>-1</sup>. The sediment mineralogy is dominated by quartz with minor feldspars and other accessory minerals. Carbonate minerals are absent as demonstrated by the low calcium concentrations maintained along long flow paths under acidic conditions (Bau et al., 2004). Surfaces of sediment grains are coated with iron, manganese and aluminum hydroxypolymere coatings, which cause relatively high sorption of trace elements (COSTON et al., 1995). Land disposal of dilute sewage effluent to the shallow, unconfined aguifer has resulted in the development of a plume of sewagecontaminated groundwater, which has distinct redox zones. The experiment discussed herein was conducted in the core of the sewage plume, which is anoxic, with up to 500 µM dissolved Fe(II). The pH values are slightly acid ranging from 6.5 to 6.8, dissolved phosphate concentrations are elevated up to 140 µM. Background concentrations of As(III) in the anoxic groundwater are up to 0.15  $\mu$ M.

The site was instrumented with an array of 10 multilevel sampling wells (MLS) (Fig. 11). Each MLS consists of a cluster of 15 polyethylene tubes (LEBLANC et al., 1991), spaced approximately 20 cm apart in altitudes of 2.7 to -0.2 m above sea level. Sampling verified that all ports were located in the anoxic zone.


Fig. 11 Test site with the 10 multilevel sampling wells ( $\bigcirc$ ) and two core locations ( $\bigstar$ )

#### 7.3.2 Tracer test

The experiment was run as a natural gradient test with a long-term injection of the tracer solution. Methods were modified from those described by (SMITH et al., 1996). Briefly, over a period of four weeks groundwater amended with As(V) (6.7  $\mu$ M) and Br<sup>-</sup> (1.6 mM) was injected continuously with an injection rate of approximately 25 l h<sup>-1</sup> into the port of MLS F625-M2 (Fig. 11) at an altitude of 2.27 m. The groundwater used for the injection was pumped from the suboxic zone in order to have no dissolved Fe(II) and low (< 5  $\mu$ M) concentrations of dissolved oxygen. Specifically, groundwater was pumped from 2 ports (altitudes 10.0 and 9.7 meters to sea level, respectively) of an MLS approximately 10 meters northwest of the injection MLS (USGS Cape Cod site MLS 8-18) using a 2-head peristaltic pump with a 600 rpm head. This feed solution was pumped through a flow-through cell outfitted with a conductivity probe, after which a concentrated solution of Br and As(V) (sodium salts, with pH adjusted to 6.4) was pumped into the stream using a piston pump. The flow rate of the piston pump was adjusted to provide the desired concentrations of Br and

As(V) in the stream of groundwater. The stream passed through a flow meter and a second flow cell equipped with a conductivity probe before entering the tube the MLS used as the injection port. The stream was outfitted with "T" fittings at the inlet and outlet ends so the groundwater could be sampled periodically before and after tracers were added. Tracer input was monitored continuously by conductivity and flow-meter measurements. Samples were collected over time at the injection well (M2) and at wells one, three and five meters downstream at different depths.

#### 7.3.3 Groundwater sampling and preservation

Samples were collected using peristaltic pumps (SAVOIE and LEBLANC, 1998). Samples for laboratory determination of Br<sup>-</sup>, As(V), As(III), Fe(II) and Mn(II) were filtered (0.45  $\mu$ m). Samples for arsenic and cation analysis were acidified with HCl to pH < 2 and stored at 4 - 6°C. The samples were measured within two weeks after sampling. Samples for field determination of pH, dissolved oxygen, sulfide, as well as pH, alkalinity and dissolved oxygen were taken and measured directly in the field laboratory.

 $H_2$  was measured after the injection in the As affected part of M2 and M3, using the bubble-stripping method of Chapelle and McMahon (CHAPELLE and McMAHON, 1991), optimized for small sampling flow. Detection of  $H_2$  in the equilibrated bubble gas was done using an RGD2 detector from TraceAnalytical (the use of brand names is for identification purposes only and should not be taken to construe endorsement by the U. S. Geological Survey).

Monitoring was terminated 105 days after the end of the injection.

#### 7.3.4 Sediment sampling and analysis

After the sampling was stopped, cores were taken from two locations on the test site and a reference core was taken upstream of the input well (Fig. 11). Core CM2-3 was collected on November 9, 2000 (138 days after start of injection) and the other cores were collected on November 8, 2000. Sediment samples were collected in plastic core liners using a wireline coring apparatus (ZAPICO et al., 1987). Sediments to be used for extractions were preserved by freezing. Sediments to be used for microbial assays were chilled until use.

The cores used for digestions were deep frozen during transport. Before the geochemical analysis, the cores were cut into 10 cm slices in a glove box and digested using different methods. The core material was digested using different methods in order to characterize the total content, the strength of binding of arsenic, and the speciation with respect to arsenic. Before the microwave digestion (MLS-ETHOS Plus) the samples were dried (120 °C for 24 h) and ground. Afterwards 0.1 g sample was extracted with 2 ml HNO<sub>3</sub>, 0.5 ml H<sub>2</sub>O<sub>2</sub> and 0.5 HF. The microwave was programmed for 35 minutes heating up to 200 °C. After the digestion the samples were diluted up to 20 ml with H<sub>2</sub>O (ultra pure) and total arsenic concentrations were measured.

In former studies it could be demonstrated, that the different redox species are not affected by the following four extractions (TRETNER, 2002):

- 1. The extraction with water dissolves the highly soluble part of arsenic giving an idea of how much arsenic can be washed out.
- The extraction with a large excess of sodium hydrogen phosphate (pH 5) over arsenic releases strongly adsorbed As(V) and As(III) (ALAM et al., 2001; KEON et al., 2001).
- The extraction with hydroxylamine hydrochloride releases arsenic from the easy reducible fraction including manganese and amorphous hydrous ferric oxides.
- The extraction with HCl releases the arsenic from the crystalline fraction of iron hydroxides by etching the surface(HUERTA-DIAZ and MORSE, 1992; KEON et al., 2001).

Over a period of 24 hours, extractions were made with 2.5 g sample and 30 ml extraction solution. Reagents used to prepare extraction solutions were as follows:  $H_2O$  (ultra pure), 1 M sodium ammonium hydrogen phosphate, 1 M hydroxyl ammonium chloride and 1 M HCl. After 24 hours the solutions were filtered (0.45 µm) and analyzed for As(III) and total As.

#### 7.3.5 Analytical methods

Specific conductance, pH values, and dissolved oxygen concentrations were determined in the field. On occasion, ortho-phosphate concentrations and sulfide were determined colourimetrically in the field using CHEMetrics sampling kits.

Phosphate was determined to a limit of quantitation of approximately 1  $\mu$ M, a precision of approximately 5 percent, and an accuracy of approximately 15 percent (McCobb et al., 2003). The limit of quantitation for dissolved sulfide measurements was approximately 1.6  $\mu$ M; precision and accuracy were similar to those for the phosphate measurements.

As(III) and total arsenic content were measured using atomic absorption spectrometry (AAS, Perkin Elmer 4100) coupled to a hydride unit (FIAS 200). The species separation was done by two measurement steps. First samples were analyzed with respect to As(III) using optimized hydride generation conditions by the combination of HCl and NaBH<sub>4</sub> so that only the As(III) was reduced. To get the total arsenic (As(tot)) concentration, samples were reduced by KJ-solution to As(III) before the second measurement. The limit of quantification for both measurements was 13 nM with a precision of 10 %. As(V) contents were calculated from As(tot) and As(III) concentrations. The stability of the arsenic species in the groundwater samples was controlled measuring As-spiked water samples from Cape Cod after different storage periods.

Anions were analyzed with an ion chromatographic system (series 4000i, Dionex) and conductivity detection using AG4A-SC (Dionex) and AS4A-SC (Dionex) columns and a mixture of  $1.8 \text{ mM Na}_2\text{CO}_3$  and  $1.7 \text{ mM Na}\text{HCO}_3$  solutions as eluent. Cations were measured using standard flame atomic absorption methods (Perkin Elmer AAS 4100).

Phosphate concentrations determined by ion chromatography (IC) in samples with high Fe(II) concentrations that had not been acidified were not reliable and are not reported. In samples with high Fe(II) concentrations not preserved by acidification, Fe(II) was oxidized upon exposure to the atmosphere resulting in the precipitation of hydrous ferric oxide, which removes phosphate from solution. Phosphate concentrations determined by IC were only reported for samples with low concentrations of Fe(II).

Phosphate concentrations reported for samples with high Fe(II) concentrations were determined colorimetrically in the field (McCOBB et al., 2003). This method has been shown to produce phosphate concentrations in good agreement with those determined on samples preserved by acidification (McCOBB et al., 2003).

## 7.4 Results and Discussion

## 7.4.1 Injection

Injectate concentrations were monitored by the increase in conductivity between the incoming and injected groundwater. Bromide concentration was found to be a linear function of the increase in conductivity owing to the injected tracers. On the 6<sup>th</sup> day of injection, pumping of the stream of injected groundwater stopped for 8 hours because of equipment failure. In addition, during the first several days of the injection, the piston pump failed to inject the tracer cocktail into the stream of injected groundwater. This problem was overcome by modifying the plumbing to provide a head of 1-2 meters of water upstream of the piston pump in order to maintain prime on the pump. Thereafter, random variability in the pumping rate of the peristaltic pump and the piton pump provided small sources of variability in the injected concentrations of Br and As(V). This variability resulted in injectate concentrations to vary from average values by approximately 20 percent during the course of the injected.

## 7.4.2 Transport processes

The small scale hydrodynamic characteristics like the transport velocities, the dispersivity coefficients as well as the longitudinal dispersivities of the aquifer were estimated from the bromide breakthrough curves at the three MLSs downstream of the injection well as described by equations 4-6 (SCHRÖTER, 1984):

$$v_a = \frac{x}{t_{C \max/2}}$$

vamean transport velocity [m d-1]xdistance [m]t<sub>Cmax/2</sub>time, when half of the maximum concentration is reached [d]

(4)

$$D_{l} = \frac{1}{8} * \frac{\left(v_{a} * (t_{85} - t_{15})\right)^{2}}{t_{50}}$$
(5)

D<sub>I</sub>: longitudinal dispersivity coefficient [m<sup>2</sup> d<sup>-1</sup>]

 $t_{85}$  : time of the passage of 85 % of the tracer mass [d]

 $t_{50}$  : time of the passage of 50 % of the tracer mass [d]

 $t_{15}\,\colon\,$  time of the passage of 15 % of the tracer mass [d]

$$\alpha_l = \frac{D_l}{v_a} \tag{6}$$

 $\alpha_l$ : longitudinal dispersivity [m]

The small scale transport velocities in the anoxic zone are significantly higher than predicted from the average hydraulic conditions observed for the whole site (LEBLANC et al., 1991) (Tab. 3). They are similar to those from tracer tests conducted concurrently at nearby locations (STADLER et al., in prep.). The observed vertical variations in a profile of a few decimeters are in the range of 30 % and reflect the variation of hydraulic conductivities. This variability, as well as the discrepancy between velocities determined in small-scale tests, is consistent with the small-scale variability in the hydraulic conductivity structure of the aquifer (Hess et al., 2002).

Tab. 3 Small scale hydrodynamic properties of the anoxic test site at Cape Cod estimated by the bromide breakthrough curves

	Level	Altitude to	distance from		_	
Well		MSL <sup>1</sup> [m]	injection <sup>2</sup> [m]	Va	D <sub>I</sub> [m²/s]	α <sub>l</sub> [m]
M3	02	2.53	1.03	0.53	0.02	0.04
M3	03	2.33	1.00	0.56	0.03	0.06
M3	04	2.12	1.02	0.77	0.01	0.014
M3	05	1.92	1.08	0.54	0.004	0.007
M3	06	1.72	1.17	0.58	0.03	0.06
M3	07	1.51	1.29	0.48	0.01	0.026
M3	08	1.31	1.43	0.51	0.006	0.01
M5	02	2.45	3.00	0.59	0.13	0.21
M5	03	2.24	3.00	0.63	0.009	0.02
M5	04	2.05	3.01	0.63	0.14	0.22
M5	06	1.64	3.07	0.53	0.01	0.02
M5	07	1.43	3.12	0.54	0.01	0.02
M5	08	1.23	3.18	0.43	0.004	0.008
M6	02	2.45	3.17	0.55	0.06	0.1
M6	03	2.24	3.16	0.49	0.04	0.09
M6	04	2.05	3.17	0.4	0.001	0.003
M9	02	2.45	5.00	0.46	0.06	0.13
M9	03	2.24	5.00	0.52	0.03	0.05
M9	04	2.05	5.00	0.64	0.03	0.05
M9	05	1.84	5.02	0.42	0.03	0.08

Longitudinal dispersivities are in the range of centimeters and are in good agreement with values given for glacial sands and flow distances of a few meters (SCHRÖTER, 1984).

### 7.4.3 Background chemical conditions

Chemical conditions in the region of the aquifer where tracers were injected were typical of those reported previously for the anoxic zone of the sewage plume (KENT et al., 1994). Groundwater was anoxic, had no nitrate, elevated concentrations of Fe(II), and no detectable sulfide (Tab. 4). Concentrations of sewage-derived phosphate were 40  $\mu$ M. Arsenic concentrations ranged from 0.13 to 0.26  $\mu$ M with at least 80 percent as As(III). Values of pH in the range 6.6 to 7.0 are consistent with those expected for the sewage effluent after biodegradation has consumed the dissolved oxygen and nitrate initially in the sewage effluent and additional biodegradation by iron-reducing bacteria is occurring (ABRAMS and LOAGUE, 2000; ABRAMS et al., 1998).

Dissolved  $H_2$  concentrations were 0.2 to 0.4 nM, similar to those determined elsewhere in the anoxic zone.

	unit	min.	max.	mean	standard deviation
pН		6.12	6.71	6.47	0.14
conductivity	[µS/cm]	201	271	22.1	15.3
alkalinity	[meq/L]	0.45	0.79	0.54	0.11
Na	[µM]	532	730	586	45.0
K	[µM]	196	333	266	35.8
Ca	[µM]	173	285	203	27.7
Mg	[µM]	62.5	137	87.1	23.5
Fe	[µM]	120	462	244	74.2
Mn	[µM]	2.95	5.83	4.19	0.89
As(III)	[µM]	0.04	0.25	0.13	0.05
As(V)	[µM]	0	0.04	0.02	0.01
CI.	[µM]	65.5	1374	534	248
NO <sub>3</sub> <sup>-</sup>	[µM]	<8x10 <sup>-3</sup>	<8x10 <sup>-3</sup>	<8x10 <sup>-3</sup>	-
PO <sub>4</sub>	[µM]	32.7	49.9	42.2	5.55
SO4	[µM]	13.9	179	69.2	43.1
H₂S	[µM]	<1.6	<1.6	<1.6	-
H₂	[nM]	0.15	0.49	0.27	0.08

Tab. 4 Background geochemical conditions in the anoxic zone at the Cape Cod aquifer

## 7.4.4 Chemical perturbation caused by injection

The injection had a major impact on ambient groundwater chemical conditions (Fig. 12). Ambient groundwater had no nitrate or dissolved oxygen, approximately 200  $\mu$ M Fe, and 40  $\mu$ M phosphate. Injected groundwater had approximately 100  $\mu$ M nitrate, 10 - 30  $\mu$ M dissolved oxygen, 50  $\mu$ M phosphate, and no Fe (Tab. 4). Dissolved Fe

remained at approximately 200 µM for several days after Br<sup>-</sup> had broken through, indicating that dissolved Fe concentrations were maintained by a reservoir of sediment-bound Fe(II). Sediment-bound Fe(II) was most probably Fe(II) adsorbed onto the sediments (APPELO et al., 2002). After approximately 9 days, Fe concentrations decreased slowly with time until 16 days after beginning the injection, at which point they had decreased below the detection limit. The slow increase in Fe concentrations observed after the end of the injection likely resulted from the slow build-up of adsorbed Fe(II) on the sediments the occurred as Fe was transported from upgradient regions of the aquifer that had not been perturbed by the injection.



Fig. 12 Breakthrough of bromide, nitrate and iron 1 m downstream

Nitrate concentrations equivalent to those in the injected groundwater were observed several days after the beginning of the injection. Nine days after the beginning of the injection, nitrate concentrations were below detection. Nitrate concentrations began to increase after Fe concentrations had become negligible, indicating that the reservoir of adsorbed Fe had been depleted. Dissolved oxygen was not detected. Rapid disappearance of nitrate and the slow increase in Fe concentrations observed shortly after the end of the injection signal the reestablishment of anoxic conditions (Fig. 12).

The data are consistent with the hypothesis that dissolved oxygen and nitrate injected into the anoxic zone oxidized Fe(II), which precipitated as hydrous ferric oxide or other Fe(III) or mixed Fe(II)-Fe(III) oxyhydroxides. The time-scale of the abiotic oxidation of aqueous Fe(II) by dissolved oxygen at pH 6 is on the order of 10 hours (EARY and RAI, 1988), which is fast enough to account for the observed loss of dissolved oxygen over the 2-day transport time from the injection port to the sampling

port 1 meter downgradient. Abiotic oxidation of Fe(II) by nitrate is very slow under the prevailing conditions (OTTLEY et al., 1997). Microbial oxidation of Fe(II) by nitrate in aquifers and sediments is well documented, (BÖHLKE et al., 2002; POSTMA et al., 1991; SENN and HEMOND, 2002; STRAUB et al., 1996). Some of the loss of nitrate observed during transport might have resulted nitrate reduction coupled to organic carbon compounds as electron donors (SMITH et al., 1991). However, the observation that nitrate concentrations only increased after the reservoir of sediment-bound Fe(II) was depleted indicates that nitrate reduction coupled to Fe(II) oxidation was an important process during the tracer test. The cause of the lag period prior to disappearance of nitrate is unknown, but could have been a result of the time required to build up a significant population of microorganisms capable of coupling nitrate reduction to Fe(II) oxidation. The rate of nitrate consumption required to remove approximately 100 µM nitrate over the 2-day transport time was much faster than indicated by results of previous field studies (BÖHLKE et al., 2002; POSTMA et al., 1991), where the source of Fe(II) was pyrite and other crystalline Fe(II)-containing minerals.

Phosphate was present at a concentration of approximately 50  $\mu$ M in the both the ambient and injected groundwater (Tab. 4). The absence of phosphate at the observation port 1 meter downgradient until 32 days after the start of the injection (Fig. 12) shows that phosphate was removed during transport. The removal of phosphate likely resulted from adsorption onto freshly precipitated hydrous ferric oxide or other Fe(III)-containing phase. Adsorption is an important processes limiting phosphate transport in aquifers (ISENBECK-SCHRÖTER et al., 1993; WALTER et al., 1996) and phosphate would be expected to adsorb extensively on any freshly precipitated, Fe(III)-containing phase that formed as a result of oxidation of adsorbed Fe(II). These results illustrate that rapid oxidation of Fe(II) and precipitation of Fe(III)-containing oxyhydroxides can occur during transport and that these precipitates can strongly influence solute concentrations locally.

The injection also caused a decrease in pH from ambient values in the range 6.6 to 6.8 to 6.2 to 6.4. The latter values were characteristic of those in the injected groundwater with tracers.



Fig. 13 Fe and Mn concentrations in M2 (injection well) and M3. The concentrations decreased during injection due to an input of small amounts of oxygen

Anoxic conditions had been re-established within the region between the injection MLS and sampling port 1 meter downgradient by 45 days after beginning the injection (Fig. 13). By this time, nitrate and dissolved oxygen concentrations had been diminished by transport and reaction with Fe(II) to below detection. Once anoxic conditions had been re-established at the sampling port 1 meter downgradient from the injection, Fe concentrations began to increase steadily. The slow increase in Fe(II) concentrations prior to the rapid increase observed after approximately 70 days likely resulted from adsorption of Fe(II) during transport. Similar trends were observed at sampling ports further downstream.

Once anoxic conditions had been re-established, the smell of sulfide could be detected in the initial groundwater pumped from MLS sampling ports. The intensity of the smell was greater in sampling ports outside the path of the tracer cloud (M4, M6, M8, and M10, Fig. 14) than those in the path of the tracer cloud (M2, M3, M5, and M9, Fig. 14). The smell could not be detected after purging. Measurements of dissolved sulfide conducted in late September showed that dissolved sulfide concentrations both in and out of the path of the tracer cloud were below the limit of quantitation (1.6  $\mu$ M). Sulfide concentrations in equilibrium with the poorly ordered iron sulfide phase mackinawite at 12 °C, an ionic strength of 0.003, and an Fe(II) concentration of 200  $\mu$ M, calculated from the thermodynamic data of (BENNING et al., 2000), should exceed the limit of quantitation at pH values below approximately 6.7. Dissolved sulfide concentrations determined in low-temperature sedimentary environments often exceed those in equilibrium with mackinawite (CHEN et al., 2003). Thus, one would expect that sulfide concentrations exceed the limit of quantification if

present in the aquifer, especially in the path of the tracer cloud where pH values had been decreased below those characteristic of the anoxic zone. Nevertheless, dissolved sulfide could also be consumed by redox reactions at the surface of iron hydroxy oxides and thus be decreased below the detection limit. This could be shown in laboratory studies of Höhn et al. (HÖHN et al., 2005b). Whether sulfate reduction was occurring in the aquifer or just in the column of water left standing in the MLS tubes is unknown.



Fig. 14 Map of wells with different  $H_2S$  intensities.

## 7.4.5 Spatial snapshots of As(V) and As(III)

The most important result of our test is that a portion of As(V) was reduced to As(III) in the iron reducing zone of the Cape Cod aquifer. The development and movement of the plumes of As(V) and As(III) are figured using an interpolated twodimensional profile along the flow path at different times after the injection stopped (Fig. 15, Fig. 16, Fig. 17). Fig. 15 shows the As(V) and As(III) distributions directly after the input had been stopped at the  $30^{th}$  day. The As(V) formed a plume with concentrations nearly up to the input around the injection well and a spreading downstream. At this time no As(V) reduction could be observed. The As(III) distribution (Fig. 15 b) represents the background concentration.



Fig. 15 Profiles of the arsenic plumes along the flow path directly after stopping the injection at the  $30^{th}$  day. a) As(V) b) As(III).

At the 45<sup>th</sup> day the As(V) plume was evenly distributed around M2 and M3 (Fig. 16 a). The concentration at the injection well decreased to 2.8  $\mu$ M indicating a desorption took place. Around the injection well, the aquifer material was highly loaded with As(V) because of the accumulation at iron hydroxides. At the same time the significant increase of As(III) concentration at M2 and M3 demonstrated that reduction of As(V) to As(III) had started (Fig. 16 b).



Fig. 16 Profiles of the arsenic plumes along the flow path at the 45<sup>th</sup> day. a) As(V); b) As(III).

63 days after the input started both plumes have the same extension and only differ in the maximum concentrations (Fig. 17 a and b). This similarity suggests an in-situ reduction of As(V).

While the plumes moved with water flow, they also tended to drop downwards because of density effects.



Fig. 17 Profiles of the arsenic plumes along the flow path at the 63<sup>th</sup> day. a) As(V); b) As(III).

#### 7.4.6 Breakthrough curves

Breakthrough curves for As(V) and As(III) are presented for the injection port and for sampling ports with the highest concentrations of injected tracers at each of the MLS downstream of the injection (Fig. 18).



Fig. 18 Breakthrough curves of As(V) and As(III) measured in the injection well and in the three sampling wells in the levels with the highest concentrations. a) M2 (2.27 m MSL) b) M3 (2.12 m MSL) c) M5 (2.24 m MSL) d) M9 (2.24 m MSL).

At the sampling port 1 meter downstream from the injection, As(V) concentrations began to increase above background values 4 days after the start of the injection and peaked at 1.3  $\mu$ M from day 6 to 8 (Fig. 18b). From day 8 to 11, As(V) concentrations decreased to near the detection limit. As(V) concentrations remained near the detection limit between days 11 and 14, after which time they increased steadily until day 27. As(V) concentrations peaked at 4.8 to 5.1  $\mu$ M from 27 to 28 days after the start of the injection Arsenic(V) concentrations decreased from 5.1 to 4.2  $\mu$ M from days 29 and 34. As(V) concentrations exhibited steep increases and decreases between days 34 and 39, reaching a maximum concentration of 7.4  $\mu$ M on day 37. As(V) concentrations decreased slowly between days 39 and 60, after which time they remained near 2  $\mu$ M until sampling ended 120 days after the start of the injection.

Concentrations of As(III) remained near background values (0.02 to 0.07  $\mu$ M) at the sampling port 1 meter downstream until 46 days after the injection (Fig. 18b). Forty-five days after the start of the injection, at which time anoxic conditions had been reestablished in the region between the injection MLS and sampling port 1 meter downstream, concentrations of As(III) began to increase above background values. The As(III) breakthrough curve exhibited a broad peak, with maximum concentrations of 1.6 to 1.7  $\mu$ M. After approximately 80 days after the start of the injection, As(III) concentrations remained between 0.4 and 0.7  $\mu$ M.

The trends in the complex As(V) breakthrough curve and the As(III) breakthrough curve 1 meter downstream of the injection (Fig. 18b) are consistent with the observed changes in chemical conditions during and after the injection and the impact of these changes on As(V) adsorption and transport. Prior to 45 days after the start of the injection, As(V) reduction was not a significant reaction and, therefore, did not influence the observed trends in As(V) concentration. During this period, As(V) concentrations were influenced by adsorption at sediment surfaces and interactions with hydrous Fe oxide precipitates formed by oxidation of adsorbed Fe(II). In the pH range 6 to 7, As(V) adsorption is strongly influenced by pH and phosphate concentrations (DIXIT and HERING, 2003; HIEMSTRA and VAN RIEMSDIJK, 1999; HINGSTON et al., 1971; MANNING and GOLDBERG, 1996). In the presence of significant concentrations of dissolved and adsorbed phosphate, such as exist in the anoxic zone (PARKHURST et al., 2003; STOLLENWERK, 1996; WALTER et al., 1996), As(V) adsorbs extensively but competition for adsorption sites with phosphate results in the

existence of significant concentrations of As(V) in aqueous solution (KENT and FOX, 2004). Thus, during the first several days after the start of the injection, adsorption onto the sediments in the presence of adsorbed phosphate resulted in retarded transport of As(V).

By 9-10 days after the start of the injection, significant decreases in Fe concentrations were observed indicating that oxidation of Fe(II) and precipitation of hydrous Fe oxides began to have a major impact on dissolved Fe concentrations (Fig. 12 and Fig. 13. A decrease in As(V) concentrations coincided with the decrease in Fe concentrations (Fig. 18b, Fig. 12 and Fig. 13). Decreases in As(V) concentrations during this time to values near the detection limit are consistent with results of previous studies of adsorption of As(V) onto and occlusion into precipitating hydrous Fe oxides (FORD, 2002; FULLER et al., 1993). In addition to direct uptake of As(V) during precipitation of hydrous Fe oxides, removal of phosphate by adsorption and occlusion would be expected to decrease As(V) mobility. In the absence of phosphate, As(V) adsorption onto hydrous Fe oxides would be expected to be sufficiently extensive to decrease greatly As(V) mobility (KENT and FOX, 2004). This explains the extensive delay in breakthrough of As(V) concentrations at values near those in the injectate 1 meter downstream of the injection. The steep fluctuations in As(V) concentration observed 1 meter downstream after the end of the injection period but before the re-establishment of anoxic conditions likely resulted from changes in chemical conditions such as pH, phosphate concentrations, and ionic strength following the end of the injection. It is likely that fluctuations in concentrations of injected tracers and the degree of mixing between the injected stream of the groundwater and ambient groundwater caused the minor changes in As(V) concentrations observed during breakthrough.

At the injection port and the sampling port 1 meter downgradient, elevated concentrations of dissolved and sediment-bound As(V) were present prior to reestablishment of anoxic conditions approximately 35 days after the start of the injection. At both locations, As(III) concentrations began to increase on day 45, approximately 10 days after anoxic conditions had been re-established. Thus, it appears that throughout this region, reduction of significant quantities of As(V) to yield a measurable increase in dissolved As(III) had occurred.

At the sampling port 2.5 meters downgradient from the injection (M5), anoxic conditions had been re-established approximately 41 days after the start of the

injection but elevated concentrations of As(V) had not yet arrived (Fig. 18c). Breakthrough of As(III) concentrations above background occurred approximately 60 days after the start of the injection. The 15-day interval between appearance of elevated concentrations of As(III) 1 meter downgradient and 2.5 meters downgradient shows the occurrence of extensive retardation of As(III) by adsorption. Iron oxides and hydrous oxides have a much higher affinity for adsorption of As(III) than aluminum oxides and silicates (ARAI et al., 2001; LIN and PULS, 2000; MANNING and GOLDBERG, 1997). Thus, the 15 days required for transport of As(III) along the 1.5 meter flow path from M3 to M5 indicates that the quantity of freshly precipitated hydrous Fe oxides in this region increased the reactive surface area sufficiently to cause the observed retardation of As(III).

Breakthrough of As(III) and As(V) 2.5 meters downstream from the injection started at approximately the same time. It is possible that, under the perturbed and variable chemical conditions present along the path of the tracer cloud through the anoxic zone, that adsorption of As(V) was more extensive than that of As(III). However, if reduction of As(V) to As(III) was occurring at a rate comparable to the time-scale of As(V) transport, the leading edge of As(V) breakthrough curve could have been eroded. This could contribute to the greater apparent retardation of As(V) as compared to As(III) both at 2.5 meters downgradient (Fig. 18c) and 4.5 meters downgradient (Fig. 18d).

Time histories for As(V) and As(III) from both the injection port and sampling port 1 meter downstream exhibited extensive tailing. Tailing in the As(V) time history could result from slow rates of diffusion of As(V) out of freshly precipitated hydrous Fe oxides (FULLER et al., 1993) or release of occluded As(V) upon reductive dissolution of the fresh hydrous Fe oxides (CUMMINGS et al., 1999; FORD, 2002). Assuming that the rate of reduction of As(V) was proportional to its concentration, a quasi steady state would eventually be achieved between rates of release and reduction of As(V) to As(III).

#### 7.4.7 Extractions

Directly after the water sampling was stopped, cores were collected at three different locations of the test site (compare Fig. 11). Cores CM2-3 and CM 5 were collected in region traversed by the As(V) tracer cloud. Core C0 (not shown in Fig.

11) was taken upstream of the tracer test site to determine quantities of naturally occurring arsenic extracted using the various methods.

The total concentrations of naturally occurring arsenic (Fig. 19) are in the range of arsenic concentration determined in granitic rocks from New England (AYOTTE et al., 2003), which are likely to be similar to the source material for these sediments. In addition, they are similar to those determined in sediments collected above the sewage-contaminated zone at a site approximately 40 meters away (KENT and FOX, 2004) using USEPA method 3050B, which is similar but does not require using hydrofluoric acid.



Fig. 19 As (tot) concentration from three cores after the sampling was stopped. C0 is a reference core taken upstream of the test area. CM2-3 and CM5 are in the flow path of the arsenic plume

Total arsenic concentrations in sediments collected adjacent to the injection location (core CM2-3) were significantly above background values (Fig. 19). Sediments collected at the elevation of the injection port had the highest concentration of arsenic, which was over twice the typical background concentration. This is consistent with the hypothesis that, once extensive oxidation of Fe(II) by nitrate began, As(V) was immobilized close to the injection location by incorporation into hydrous Fe oxide precipitates. It also demonstrates that significant quantities of arsenic remained bound in these precipitates several months after anoxic conditions had been re-established. Total arsenic concentrations in sediments collected approximately 2 meters downgradient of the injection (CM5) were similar to background concentrations at most levels (Fig. 19). This would be expected if most of the sequestration of injected As(V) occurred in the vicinity of the injection.

Extractions of sediments collected from the path of the tracer cloud with water, hydroxylamine hydrochloride, and phosphate yielded arsenic concentrations

significantly above background values (Fig. 20 a-d). The agreement between the trends in arsenic extractable from sediments in the path of the tracer cloud by phosphate and by hydroxylamine hydrochloride indicates that these methods primarily extract adsorbed arsenic, as has been previously suggested (KENT and FOX, 2004; KEON et al., 2001). Significant concentrations of adsorbed arsenic remained associated with the sediments 4 months after cessation of the As(V) injection. In the absence of co-precipitation of As(V) with hydrous Fe oxides, adsorbed As(V) would be expected to be substantially depleted by desorption owing to the elevated phosphate concentrations (DARLAND and INSKEEP, 1997; STADLER et al., 2001). This observation supports the hypothesis that a quasi steady state was achieved near the end of the experiment between transport of As(V) away for the vicinity of the injection and diffusion of As(V) occluded in hydrous Fe oxides as well as reductive dissolution of hydrous Fe oxides. This is consistent with expectations based on results of experimental studies with synthetic ferrihydrite (FORD, 2002; FULLER et al., 1993).



Fig. 20 As(V) concentrations accumulated in the aquifer sediment at three core locations: C0: reference core, C2-3: between M2 and M3, C5: near M5; the four extraction methods represent different mobilities of As(V). a)  $H_2O$ ; b) hydroxyl ammonium chloride; c) sodium ammonium hydrogen phosphate; d) HCl

#### 7.4.8 Microbial analysis and H<sub>2</sub> measurements

Most probable (MPN) number analysis (HARRINGTON et al., 1998) was conducted on sediment samples to estimate microbial populations capable of Fe reduction, As(V) reduction, and sulfate reduction. Sediment samples were obtained from the anoxic zone outside of the region impacted by the tracer tests and from the lower elevations (below 1.9 meters to sea level) from cores CM2-3 and CM5. Iron reducing microorganisms were the most abundant, with MPN values in the range 140 to 350 cells per gram. As(V) reducers were in the range 5 to 46 cells per gram. A low density of sulfate reducers (1 cell per gram) was also observed. MPN values for autotrophic, Fe-oxidizing denitrifiers were not determined. Higher MPN values for As(V) reducers were determined in the core from outside the path of the tracer cloud than those in the path. Whether this constitutes a significant impact of the injection on microbial populations in the anoxic zone or reflects the heterogeneous distribution of microbial populations requires further study.

The MPN data suggest the importance of As(V) reducing microorganisms in the As(V) reduction observed during the tracer test. Microbial reduction of As(V) coupled to oxidation of organic carbon or hydrogen has been shown to be an important process in some systems (AHMANN et al., 1997; OREMLAND et al., 2000). However, an experimental study has suggested that sulfide, if present at sufficiently high concentrations, could reduce As(V) under ambient pH values (HÖHN et al., 2005b). Hydrogen concentrations determined September 24-25, 2000 (92-93 days after the start of the injection) in most sampling ports in the path of the tracer cloud were 0.2 to 0.4 nM, which has been suggested to be characteristic of Fe reducing microorganisms (CHAPELLE et al., 1995; LOVLEY et al., 1994). However, concentrations of 0.5 and 0.8 nM were determined at sampling ports directly above and directly below the injection port. These elevated values might indicate increased activity of microorganisms using a terminal electron acceptor with a lower energy yield than that for hydrous Fe oxide, such as As(V) or sulfate (CHRISTENSEN et al., 2000). Thus, whether As(V) was reduced by As(V) reducing microorganisms or sulfide cannot be determined conclusively.

### 7.5 Conclusions

The anoxic tracer test with As(V) at the Cape Cod site in Mass. (USA) contributes the first experimental results concerning the question of As(V) reduction and transport in a real aquifer environment. As could be shown above, part of the As(V) input was reduced and transported in the As(III) form. This species was significantly more mobile under the anoxic conditions than As(V), which accumulated at the aquifer material. Hence we have to discuss the question, which geochemical conditions favor this reaction in the studied aquifer system.

The experiment has to be divided into two periods, the input period and the release period. During the injection and for a short period afterwards, nitrate and small amounts of oxygen injected along with As(V) oxidized Fe(II) associated with the sediments. After a short lag period, Fe(II) oxidation became sufficiently fast to remove 100  $\mu$ M nitrate during the 2-3 day period required for transport 1 meter. Previous field studies conducted in systems with elevated nitrate concentrations owing to agricultural practices observed oxidation of pyrite and other crystalline Fe(II) containing minerals, but at much slower rates than those observed here (BöHLKE et al., 2002; POSTMA et al., 1991). During the period when oxidative precipitation of hydrous Fe oxides occurred, virtually all of the injected As(V) was incorporated into the precipitates. Once the supply of Fe(II) had been depleted, formation of precipitates ceased and the injected As(V) was transported slowly downstream. These results corroborate the strong linkage between nitrate-induced oxidation of Fe(II) and attenuation of arsenic concentrations suggested by the results of previous field studies (HARVEY et al., 2002; SENN and HEMOND, 2002).

During input phosphate concentrations in the water decreased below the detection limit and also most of the injected As(V) was bound to the aquifer material. These observations lead to the conclusion that the removal of As(V) and phosphate occurs rapidly compared to the time scale of transport, when Fe<sup>2+</sup> is oxidized.

The iron and As(V)-pool, which had accumulated in the input area during injection, then was released after the input had been stopped. While the geochemical conditions became anoxic again, significant amounts of arsenic were reduced to As(III). These observations indicate that a reduction of As(V) to As(III) can occur rapidly once electron acceptors like oxygen and nitrate are depleted.

In previous investigations the reduction of As(V) was driven by arsenic reducing microorganisms (MYERS et al., 1973; OREMLAND et al., 2000). The continuous injection during our tracer test was chosen in order to create stable conditions over a period of several weeks for a possible development of an arsenic reducing microbial population. In microbial investigations on core material from the test site arsenic reducing microorganisms could be detected. Thus, microbial reduction of As(V) could be an important process during the release period of the tracer experiment.

During the sampling,  $H_2S$  was smelt in some tubes of the MLS indicating a sulfate reduction going on. Dissolved sulfide could not be detected in the water, whereas  $H_2$  data measured in the anoxic zone during the tracer experiment support sulfate reduction. As the system is iron dominated, high concentrations of Fe<sup>2+</sup> are released into the groundwater (VAN CAPPELLEN and WANG, 1996) limiting the dissolved sulfide concentrations to a low level because of the low solubility of FeS. Sulfide also reacts fast at the surfaces of iron hydroxides, where it is consumed by oxidation. Studies of Höhn et al. (2005) support the theory that  $H_2S$  could also be a reducing agent of As(V) adsorbed to ironhydroxide. Thus, we concluded that sulfate reduction could be another important process in the Cape Cod aquifer.

Arsenic transport is governed by redox, adsorption, and, possibly, co-precipitation reactions and, therefore, cannot be adequately quantified or characterized by  $K_d$  values or retardation coefficients. In the experiment it could be observed that As(III) moves faster than As(V). Predicting the reactions and transport of the two arsenic species requires the application of well-constrained multispecies reactive transport model (PARKHURST and APPELO, 1999).

## 7.6 Acknowledgements

This work was supported by the "Deutsche Forschungsgemeinschaft", the USGS U.S. Geological Survey Toxic Substances Hydrology Program, and U.S. Geological Survey National Research Program. Vanja Klepac-Ceraj and Dr. Martin Polz of Massachusetts Institute of Technology are greatfully acknowledged for making possible the microbiological assays of sediment samples. We thank Denis LeBlanc and Kathryn Hess for their support in the field as well as Stefan Rheinberger and Volker Wild for their support in the laboratory.

# 8. Arsenic reduction by dissolved sulfide at $MnO_2$ and $Fe_2O_3$ surfaces

Höhn Rouven<sup>1</sup>\*, Isenbeck-Schröter Margot<sup>1</sup>, Wallschläger Dirk<sup>2</sup>

<sup>1</sup>Institute of Environmental Geochemistry, University of Heidelberg

<sup>2</sup>Environmental & Resource Studies Program, Trent University

#### 8.1 Abstract

The mobility and toxicity of arsenic in soil and aquatic environments depend on its redox speciation. Therefore, it is important to know reaction partners and pathways for the redox-processes of arsenic. In iron-reducing environments, arsenic is generally mobile. Dissolved sulfide (H<sub>2</sub>S or HS<sup>-</sup>) may be present in small amounts in such an environment and is a strong reducing agent. Therefore, reactions between arsenic and sulfide may strongly influence arsenic mobility. In this study, we conducted batch tests on the reduction of As(V) by low concentrations of dissolved sulfide in the presence of Fe<sub>2</sub>O<sub>3</sub> or MnO<sub>2</sub> serving as reactive surfaces. Sulfide was added as H<sub>2</sub>S gas or Na<sub>2</sub>S salt and samples were buffered to pH 6.8. First investigations without minerals using different amounts of H<sub>2</sub>S gas confirmed that the arsenic reduction rate depended on initial sulfide concentration and strongly on pH. In samples with MnO<sub>2</sub>, no significant As(III) release into solution was observed, probably due to the Mn(IV) acting as an oxidant for sulfide and As(III). By contrast, sulfide reduced As(V) bound to Fe<sub>2</sub>O<sub>3</sub>, but reduction rates were significantly lower than in mineral-free solutions, presumably due to sulfide consumption by Fe(III). There was no evidence of soluble arsenic-sulfur compounds in solution after the experiments, probably because all sulfide was consumed by oxidation.

#### 8.2 Introduction

In Bangladesh and Bengal more than 30,000,000 people are affected by arseniccontaminated drinking water (HARVEY et al., 2002; SMEDLEY and KINNIBURGH, 2002; SWARTZ et al., 2004; WINSTON et al., 2003). Worldwide the problem is even more serious. It is well known that arsenic ingestions cause severe health problems, including skin cancer. In the last decade, huge scientific effort has been made to elucidate the origin and the geochemical context of high arsenic concentrations in groundwater (Welch and Stollenwerk, 2003).

During a tracer test in the Cape Cod aquifer, the reduction of arsenate (As(V)) to arsenite (As(III)) was observed under iron-reducing conditions (Höhn et al., 2005a). No arsenic-reducing bacteria could be identified, but during sampling, small amounts of H<sub>2</sub>S were observed. These observations led to the concept of iron and arsenic reduction by dissolved sulfide (Fig. 21). The redox conditions during the experiment could consequently be characterized as sulfate reduction in an iron-dominated system.

Similar conditions have been described in other systems, e.g. in the Römö aquifer (JAKOBSEN et al., 1998), where small amounts of sulfide are released into the ironreducing zone and may also reduce ironoxid minerals (CANFIELD et al., 1992; DOS SANTOS AFONSO and STUMM, 1992). In groundwater samples from Bengal, arsenic concentrations correlate with ferrous iron concentrations (WAGNER et al., 2005), indicating iron reducing conditions; in those sediments, sulfur isotope studies also indicated sulfate reduction conditions.

The batch tests presented here were carried out to investigate such a system under controlled conditions in the laboratory. The reduction of As(V) at  $MnO_2$  and  $Fe_2O_3$  surfaces by dissolved sulfide was studied in experiments using high ratios of ferric iron and As(V) to sulfide in order to obtain an analogous model of sulfide entering or being formed in an iron- dominated system (Fig. 21).



Fig. 21 Schematic model of As(V) reduction by dissolved sulfide in an iron-dominated system

First, the reduction of As(V) by dissolved sulfide was investigated in unbuffered solutions (modified after Rochette et al. 2000), to determine the best experimental setup as well as the required reaction period. In the next step, As(V) reduction was studied in solutions buffered to pH 6.8 using dissolved H<sub>2</sub>S gas as well as Na<sub>2</sub>S. Investigations at mineral surfaces were performed with MnO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub> in order to simulate Cape Cod conditions (pH 6.8) in a simplified system. The ratios of Mn and Fe to As were chosen such that the whole amount of As(V) could be adsorbed on the mineral surfaces.

#### 8.3 Materials and methods

**8.3.1 Chemicals:** The following chemicals were used for the experiments: Iron (III) oxide ( $Fe_2O_3$ , 99 %, Merck), manganese (IV) oxide ( $MnO_2$ , 99.999 %, Alfa Aesar),  $H_2S$  gas (purity: "highest"), sodium sulfide ( $Na_2S * x H_2O$  (x= 7-9) Merck extra pure), As(V) ( $H_3AsO_4$ ), Tris ( $C_4H_{11}NO_3$  99.9 %, ICN ultra pure) and hydrochloric acid (conc. HCI). Solutions were prepared in degassed and double deionized water under inert atmosphere conditions (argon-filled glove box). Tris buffer was prepared by adjusting 0.1 M Tris solution to pH 6.8 with hydrochloric acid.

## 8.3.2 Preliminary test of As(V) reduction by dissolved sulfide in aqueous solution

All experiments were conducted in an argon filled glovebox.

Setup 1 (multiple containers): 20 headspace vials (size 25 mL) were filled with 20 mL of  $H_2O$  and As(V) was added to produce a starting concentration of 890  $\mu$ M. The vials were degassed with nitrogen and 75, 150 and 300  $\mu$ M of  $H_2S$  gas were

injected through a septum using GC-syringes. The test was conducted over a period of 21 days at room temperature (22°C). During each sampling, one bottle was opened and the two arsenic species were measured immediately.

Setup 2 (single container): One 500 ml glass bottle containing 890  $\mu$ M As(V) and 150  $\mu$ M H<sub>2</sub>S gas was prepared and sampled over a period of 21 days.

We found that As(III) production in the multiple container setup 1 showed greater variability than the single container setup 2, likely due to inter-sample variability. Therefore setup 2 was chosen for the following tests.

### 8.3.3 Tests of As(V) reduction by dissolved sulfide at reactive surfaces

The compositions of these samples (500 ml glass bottles) is given in Tab. 5; the single container setup was used. In the first 24 hours of the tests the As(V) was adsorbed onto the mineral surfaces. Afterwards the sulfide was added to the samples. The bottles were sampled every second day over a period of 30 days. The experiments were conducted at room temperature (22°C) under inert atmospheric conditions in an argon-filled glove box and shaken once a day.

Tab. 5 Composition of the samples in the tests of As(V) reduction by dissolved sulfide at reactive surfaces

sample Nr	As (V)	Tris	H₂S	Na₂S	MnO <sub>2</sub>	$Fe_2O_3$
1	100µM					
2	100µM	0.2mM				
3	100µM	0.2mM	100µM			
4	100µM	0.2mM		100µM		
5	100µM	0.2mM	1000µM		0.1M	
6	100µM	0.2mM		1000µM	0.1M	
7	100µM	0.2mM	1000µM			0.1M
8	100µM	0.2mM		1000µM		0.1M
9	100µM	0.2mM			0.1M	
10	100µM	0.2mM				0.1M
11		0.2mM		100µM	0.1M	
12		0.2mM	100µM			0.1M

## 8.3.4 Analytical methods

To monitor the formation of As(III) during the reduction experiments, arsenic species were separated with by ion chromatography (Dionex series 4000i) and measured off-line by flow injection-atomic absorption spectrometry (FI-AAS; Perkin Elmer FIAS 200 and AAS 4100). Sulfide had to be separated from the samples

before measuring with the FIAS-AAS to prevent damage to the quartz cuvette. The separation method was developed using standards of As(III) (133 $\mu$ M) and As(V) (133 $\mu$ M) and sulfide (1.5 mM). The eluent consisted of 6 g L<sup>-1</sup> ammonium dihydrogenphosphate ((NH<sub>4</sub>)H<sub>2</sub>PO<sub>4</sub> 99 %) with a flow rate of 0.9 ml min<sup>-1</sup>. The guard column was a Dionex IonPac AG7 (4 x 50 mm) and the analytical column an IonPac AS7 (4 x 250 mm).

All samples were pre-reduced using  $100 \text{ g L}^{-1}$  KI and  $100 \text{ g L}^{-1}$  ascorbic acid solutions. Samples were then mixed with 6 M HCl at a ratio of 1:4:5. In the FIAS the samples were mixed with 2 g L<sup>-1</sup> NaBH<sub>4</sub>, 0.2 g L<sup>-1</sup> NaOH and 1.2 M HCl. The FI flow rates were: sample 2 ml min<sup>-1</sup>, NaBH<sub>4</sub> + NaOH 1.6 ml min<sup>-1</sup> and HCl 1.6 ml min<sup>-1</sup>. In a gas-liquid separator arsine was purged from solution and transported by an argon flow to the quartz cuvette (argon flow rate 80 ml min<sup>-1</sup>). Arsine was converted to elemental arsenic at a temperature of 900 C°, and detected at a wavelength of 193.7 nm (RÜDE, 1996).

At the end of the reduction experiments, selected samples were sent deep-frozen by express service to Canada, and analyzed for reduced arsenic-thio species by ion chromatography-inductively-coupled plasma-mass spectrometry (IC-ICP-MS) at Trent University (WALLSCHLÄGER et al., 2002).

Dissolved iron and manganese were measured with a flame AAS (Analytic Jena; vario 6), and interpreted as Fe(II) or Mn(II) based on geochemical considerations.

#### 8.4 Results and discussion

## 8.4.1 Determination of reduction rates of As(V) in unbuffered aqueous solution

In the preliminary test we investigated the effect of different  $H_2S$  concentrations onto the arsenate reduction over a period of 21 days. The samples were not buffered and had pH-values between 3.01 and 3.12. The initial sulfide / arsenate ratios were approximately 1:12, 1:6 and 1:3. After 21 days 7-30 % of the As(V) was reduced to As(III) depending on the gaseous  $H_2S$  concentration (Fig. 22); sulfide could no longer be detected in any sample.



Fig. 22 As(III) production from As(V) reduction by different  $H_2S$  concentrations in two different setups (\* single container setup 2 with 500 ml bottle)

For the determination of reduction rate constants, it was assumed that the reduction of arsenate by sulfide is a bimolecular reaction, which follows the second order rate law

-d c[As(V)]/dt = k c[As(V)] c[S(-II)] (7)

In the absence of other reaction partners, equation (7) can be integrated based on the stochiometry of the reduction reaction, which we assumed to involve complete oxidation of sulfide to sulfate according to

$$4 H_2 AsO_4^{-} + H_2 S + 2 H^+ \leftrightarrow 4 H_3 AsO_3 + SO_4^{2-}$$
(8)

The arsenite concentrations measured throughout each experiment were used to calculate the remaining arsenate and sulfide concentrations according to equation (8), and graphic analysis of the calculated arsenate and sulfide concentrations yielded the rate constant k for each experiment (Tab. 6). Since the formed arsenite may be re-oxidized in solution over time, especially at circumneutral pH, arsenite concentrations were initially plotted against time, and the kinetic analyses only included data up to the points when arsenite concentrations started decreasing again.

For the reduction of arsenate by sulfide under acidic conditions, rate constants were obtained at different sulfide concentrations, so it was possible to express k as a function of the starting sulfide concentration  $c[S(-II)]_0$ . Our data suggest that k was not independent of  $c[S(-II)]_0$ , indicating that the reaction kinetics were not as simple as assumed in equation (7), but since the correct rate law for our experiments was unknown, we still employed the above formalism to derive the rate constants for each

experiment. Also, the known pH–dependence of the reaction (which is discussed later) was not incorporated into the kinetic model.

## 8.4.2 Reaction rates of As(V) reduction by dissolved sulfide in a buffered system

At pH 6.8, As(V) reduction was observed both in samples injected with  $H_2S$  gas and in Na<sub>2</sub>S solution (Fig. 23), but the reduction rate constants were 10-40 times lower than at pH 3 (Tab. 6).



Fig. 23 As(III) production from As(V) reduction by H<sub>2</sub>S and Na<sub>2</sub>S at pH 6.8

#### 8.4.3 Determination of reduction rates of As(V) at reactive surfaces

In the third test we studied As(V) reduction by sulfide (H<sub>2</sub>S gas and Na<sub>2</sub>S salt) on MnO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub> mineral surfaces at pH 6.8. In the samples with MnO<sub>2</sub> the concentration of dissolved As(V) was very low, because it adsorbed onto the mineral surface (as shown by sample 9, Tab. 5, i.e. in the absence of sulfide): after 24 hours only 1-2 % of the injected As(V) (100  $\mu$ M) were left in solution. During this experiment, As(III) concentrations were always below the detection limit. It is possible that some As(V) was reduced by sulfide in this experiment, but if so, the formed As(III) was re-oxidized by the manganese oxide immediately (CHIU and HERING, 2000; MOORE et al., 1990; SCOTT and MORGAN, 1995; TOURNASSAT et al., 2002; TRETNER, 2002). The concentrations of Mn<sup>2+</sup> in solution were not significantly different from the corresponding control (sample 11 in Tab. 5).

In samples with  $Fe_2O_3$  a significant amount of  $Fe^{2+}$  is released into solution (Fig. 24), due to reduction by dissolved sulfide according to reaction (9). After 10 days, a concentration plateau is reached.

4  $Fe_2O_3 + HS^- + 15 H^+ \leftrightarrow 8 Fe^{2+} + SO_4^{2-} + 8 H_2O$  (9)



Fig. 24 Iron dissolution from iron oxides by sulfide

In the presence of Fe<sub>2</sub>O<sub>3</sub> only 0.3 - 0.5  $\mu$ M of the As(V) (100  $\mu$ M) remains dissolved after 24 hours, but the injected sulfide led to As(III) production (Fig. 25).



Fig. 25 As(III) formation from arsenate reduction by sulfide on iron oxides

The total amount of As(III) formed by Na<sub>2</sub>S was higher than from H<sub>2</sub>S. In both experiments, the As(III) concentration peaked after 10 days and declined afterwards. Both As(III) and Fe<sup>2+</sup> are generated during sulfide oxidation, so one fraction of the total sulfide is used to reduce Fe(III) (reaction 3) and the other fraction is used to reduce As(V) (reaction 2).

In the kinetic evaluation of the experiments in the presence of  $Fe_2O_3$ , it cannot be ruled out that some of the formed As(III) is re-oxidized by Fe(III) (TRETNER et al., 2005). It is also possible that reduced As(III) is adsorbed onto the surface and therefore not detectable in solution. Therefore, the determined As(V) reduction rate constants shown in Tab. 6 must be interpreted as minimum net reduction speeds for these experiments.

[As(V)]₀ [µmol L <sup>-1</sup> ]	sulfide reductant [µmol L <sup>-1</sup> ]	рН	surface	k [L mol <sup>-1</sup> d <sup>-1</sup> ]
890	75 H <sub>2</sub> S	3	none	308
890	150 H <sub>2</sub> S	3	none	145
890	300 H <sub>2</sub> S	3	none	82
890	150 H <sub>2</sub> S *	3	none	143
100	100 H <sub>2</sub> S	6.8	none	7.3
100	100 Na₂S	6.8	none	8.5
100	1,000 H <sub>2</sub> S	6.8	$Fe_2O_3$	0.04
100	1,000 H <sub>2</sub> S	6.8	$Fe_2O_3$	0.12

Tab. 6 Determined rate constants for arsenate reduction by sulfide

Measurements of arsenic-sulfide complexes gave no indication of the formation of such complexes in the samples. Sulfide was used up by the reactions in the samples and could not be detected after the experiments.

#### 8.5. Conclusions

In a typical-iron reducing environment, iron oxide minerals are reduced by microbes oxidizing organic matter (BERNER, 1981; FROELICH et al., 1979). Ferrous iron is released together with any arsenic bound to the iron oxide minerals, but cannot reduce As(V) for thermodynamic reasons (ZOBRIST et al., 1998).

Strongly sulfidic environments form a sink for As(III), which can be incorporated into pyrite or precipitates (BELZILE and LEBEL, 1986; EARY, 1992; MOORE et al., 1988; SMITH et al., 2003). Additionally, dissolved As(III) forms various soluble arsenic-thio species under such conditions (WILKIN et al., 2003), which have also previously been invoked in the reduction of As(V) by sulfide (ROCHETTE et al., 2000). However, we only observed very small concentrations of such species in these experiments, suggesting that they are either not formed to a significant extent (probably because our sulfide/arsenic ratios were too low), or that they are not stable in solution over the experimental period.

In this investigation, we did not focus on such a typical sulfidic environment, but wanted to study a system where small sulfide concentrations enter or are formed in an iron-dominated system (Fig. 26). It has been demonstrated previously that iron and sulfate reduction can take place simultaneously in certain parts of an aquifer (JAKOBSEN et al., 1998). However, our study suggests that the formed sulfide may

not be detectable, but will be oxidized to other sulfur species by ferric iron and also by As(V) (Fig. 26). Small amounts of sulfide are released into the iron reducing zone and may also reduce iron itself (CANFIELD et al., 1992; DOS SANTOS AFONSO and STUMM, 1992; POULTON et al., 2004).





The experiments shown here were conducted with relatively low concentrations of sulfide and high concentrations of iron oxides and As(V). We found that the reduction rate of As(V) is correlated with the sulfide concentrations, and that the reduction is enhanced by low pH values, confirming previous results (ROCHETTE et al., 2000). In that study, however, no arsenic reduction was observed within 7 days at pH 7.

In the presence of MnO<sub>2</sub> or Fe<sub>2</sub>O<sub>3</sub> surfaces, the reduction of As(V) was suppressed compared to the homogeneous solution reactions. This was probably due to the fact that the oxide minerals were also reduced by sulfide, thus reducing sulfide concentrations available for As(V) reduction. In the test with MnO<sub>2</sub>, manganese reduction seemed to prevent arsenic reduction. The reason for this could be that As(III) is quickly re-oxidized at manganese oxide surfaces (CHIU and HERING, 2000; MANNING et al., 2002; NESBITT et al., 1998). In the presence of Fe(III), a simultaneous reduction of Fe(III) and As(V) was observed. The As(III) concentrations decreased after the sulfide was consumed, probably due to a slow re-oxidation of As(III) at the ferric iron surface (HAURY et al., 2000; SUN and DONER, 1998; TRETNER et al., 2005). It is also possible that reduction rate of As(III) is underestimated because of As(III) adsorbed onto the surface of the oxides (and therefore not detectable in the solution).

In our opinion sulfide could be the reducing agent of As(V) in the experiments conducted in the Cape Cod aquifer (HÖHN et al., 2005a), where it was shown that the iron-reducing conditions were enhanced by sulfate reduction.

### 8.6 Acknowledgements

This work was financially supported by the "Deutsche Forschungsgemeinschaft". We thank Sebastian Pfeil, Stefan Rheinberger and Christian Scholz for their assistance in the laboratory as well as fruitful discussions. Errol Lewars and Keith Oldham provided assistance with the integration of the kinetic rate law.

## 9. Mobilization of Arsenic from Ore and Mine Dumps: Two Case Studies from Sardinia (Italy)

Rouven Höhn<sup>a\*</sup>, Margot Isenbeck-Schröter<sup>a</sup>, Maria Boni<sup>b</sup>, Franco Frau<sup>c</sup>, Silvia Pontis<sup>c</sup>

<sup>a</sup>Institute of Environmental Geochemistry, University of Heidelberg, Germany <sup>b</sup>Dipartimento di Geofisica e Vulcanologia, Università di Napoli "Federico II", Italy <sup>c</sup>Dipartimento di Scienze della Terra, Università di Cagliari, Italy

<sup>\*</sup> corresponding author: rouven.hoehn@ugc.uni-heidelberg.de; Fax: +49 (0) 6221 545228

## 9.1 Abstract

Mine deposits often cause arsenic contaminations originating from arsenic bearing ore minerals like arsenopyrite, pyrite and other sulfides. High amounts of arsenic can be released into the environment by erosion as well as by geochemical weathering together with the primary products of iron sulfides oxidation such as acidity, iron and sulfate. Our study aims to analyse and compare two sites located in Sardinia (Italy) with different transport and reaction pathways of arsenic. In the first case we studied sediments from a stream channel, whereas the second case study describes a wetland. The two hydraulic situations reveal different physical and geochemical transport conditions which influence the sort and intensity of arsenic accumulation in the sediments. In the case of the *Baccu Locci Stream* the transport of As(V) is mainly depending on the flow rate, which differs extremely over the year. High flow rates cause translocation processes of the tailing material and the stream sediments. During periods with low flow rates the arsenic transport is dominated by the dissolved fraction of As(V). Also in the Sa Masa wetland varying hydraulic situations play an important role concerning the transport of arsenic, but in this case the influence is mainly geochemical. During rain periods the sediments become water saturated so

that the redox conditions get anoxic and iron reduction occurs. Together with the iron also arsenic is reduced, released and transported as As(III). In summer the sediments dry out and the iron as well as the arsenic get oxidized again and accumulate.

#### 9.2 Introduction

The cycling of arsenic in the aquatic environment is of high importance, as arsenic causes serious problems in groundwater of countries such as India, Bangladesh, Vietnam, Mongolia, Taiwan, Ghana, Argentina, Chile and Mexico (BUNDSCHUH et al., 2004; HARVEY et al., 2002; SMEDLEY and KINNIBURGH, 2002). It is well known that elevated arsenic concentrations in drinking water lead to severe various health problems. Therefore in the last decade a lot of investigations have been made to understand the environmental behavior of arsenic (WELCH and STOLLENWERK, 2003). Most arsenic contaminations originate from arsenic bearing ore minerals such as metal sulfides. In cases where these minerals are present in mine deposits high amounts of arsenic can be released into the environment by erosion of the material as well as by geochemical weathering (COURTIN-NOMADE et al., 2003; FUKUSHI et al., 2003; GALÁN et al., 2003; JOHNSON and THORNTON, 1987).

The predominant forms of arsenic in surface and ground waters are the inorganic arsenite (As(III)) and arsenate (As(V)). The speciation may reflect the redox conditions in the water (CHERRY et al., 1979), if thermodynamic equilibrium conditions can be assumed. Lots of studies showed that the mobility of arsenic in the environment is mainly affected by sorption processes. Especially the anionic As(V) species form strong surface complexes at iron hydroxide surfaces (AMITA et al., 1999; BOWELL, 1994; DZOMBAK and MOREL, 1990; FULLER et al., 1993; MANNING et al., 2002; O'REILLY et al., 2001b; RÜDE, 2001; XIAOHUA and DONER, 1996).

Associated with metal ores and coal deposits different arsenic bearing minerals can occur, among which pyrite is the most common. During the mining process pyrite bearing material is stored in mine dumps and thus exposed to atmosphere. Pyrite weathering by oxidation leads to a couple of environmental threats, which are commonly summarized as "acid mine drainage" (JAMBOR and BLOWES, 1998; NICHOLSON et al., 1988; NORDSTROM and ALPERS, 1999; SIMON et al., 1999).

The primary products of pyrite oxidation are acidity,  $Fe^{2+}$  and sulfate, which react in the environment depending on the geochemical conditions. In the case of carbonate rocks, buffer reactions dissolve calcite and dolomite, whereas gypsum and siderite, as well as different iron oxides and iron hydroxy sulfates, are formed. Heavy metals and metalloids associated with pyrite are involved in diverse reactions and mineral formations. In the case of arsenic the release from an arsenian pyrite can be described by the following overall reaction (10):

$$Fe(S_{1-x}As_x)_2 + (7-3x)/2 O_2 + (1+x)H_2O \leftrightarrow Fe^{2+} + (2-2x)SO_4^{2-} + (2x)H_3AsO_3 + (2-4x)H^+$$
(10)

In a carbonate system part of the ferrous iron is oxidized by oxygen and precipitated as iron hydroxide or iron hydroxy sulfate, generating acidity. Also arsenic is oxidized, either by manganese oxides (11) or by ferric iron (NESBITT et al., 1998; TRETNER et al., 2005) and successively bound to iron hydroxides by surface complexation (12) or coprecipitation.

$$H3AsO3 + MnO2 \leftrightarrow Mn2 + HAsO42 + H2O$$
(11)

$$\equiv \text{FeOH}^{\circ} + \text{HAsO}_4^{2-} + \text{H}^+ \leftrightarrow \equiv \text{FeHAsO}_4^{-} + \text{H}_2\text{O}$$
(12)

The following study aims to analyze the transport and reaction pathways of arsenic originating from mine deposits on two sites located in Sardinia (Italy). The two study sites differ in the geological and hydrological situations leading to different distribution patterns of arsenic.

First of all we evaluated the local distribution of total arsenic concentrations linked to the geochemical properties of the samples from each study area, in order to reconstruct the sources and find out possible sinks of arsenic. In addition we conducted four different extractions to get an idea about the binding forces as well as the redox species of arsenic in the samples. In former studies it could be demonstrated that the different redox species are not affected by the following four extractions (TRETNER, 2002):

- 1. The extraction with water dissolves the highly soluble part of arsenic giving an idea of how much arsenic can be washed out.
- The extraction with sodium hydrogen phosphate exchanges the arsenic which is complexed at the surface because of the same symmetry and size of H<sub>2</sub>PO<sub>4</sub><sup>-</sup> and H<sub>2</sub>PO<sub>4</sub><sup>2-</sup> compared to H<sub>2</sub>AsO<sub>4</sub><sup>-</sup> and HAsO<sub>4</sub><sup>2-</sup>, respectively (ALAM et al., 2001).
- The extraction with hydroxyl ammonium chloride releases the arsenic from the easy reducible fraction including manganese and amorphous ferric oxides.
- The extraction with HCl releases the arsenic from the crystalline fraction of iron hydroxides, by etching the surface, and from the carbonate fraction (HUERTA-DIAZ and MORSE, 1992; KEON et al., 2001).

Long term elution tests were carried out with four samples from the Baccu Locci Stream area (two stream sediments and two tailing samples) using  $H_2O$  (ultra pure) over a period of 37 days. These tests were conducted in order to observe changes in the release of arsenic over a long period.

#### 9.3 Description of the study areas and sampling

#### 9.3.1 Baccu Locci Stream

The first case study area is represented by the Baccu Locci Stream (BLS) catchment affected by past mining. A detailed hydrogeochemical and mineralogical study of the area has been performed by Frau & Ardau (FRAU and ARDAU, 2003; FRAU and ARDAU, 2004).

The Baccu Locci Pb-As abandoned mine is located in the Sarrabus-Gerrei mining district, Quirra region, near the town of Villaputzu (SE Sardinia, Italy). About a century (1873-1965) of exploitation for Pb and As has produced a visible environmental impact, mainly due to the unwise practice of discharging tailings from the flotation plant, built in 1949, directly into the Baccu Locci Stream. This has caused wide scattering of highly contaminated materials over the whole catchment.

Primary metalliferous mineralization is composed of prevalent arsenopyrite and galena in about equal proportions, and minor amounts of sphalerite, pyrite,
chalcopyrite and pyrrhotite; gangue minerals are mainly quartz, minor calcite and siderite, and rare fluorite.



Fig. 27 Schematic map of the Baccu Locci Stream region with indication of the sampling points

Past mining was developed mainly underground. Some waste-rock dumps, subjected to strong erosion, are still present along the upper BLS, which drains the mined area sensu stricto (Fig. 27). Arsenic contained in waste-rock materials mainly occurs as arsenopyrite and scorodite, or as sorbed species on Fe(III)-hydroxides. Tailings produced by the flotation plant were discharged directly in the BLS and transported downstream as far as the alluvial plain. Nowadays, these tailings are visible as both hardpans and layered loose materials interbedded with stream sediments on the banks of the medium-lower BLS course, but above all occur mixed with and covered by stream bed sediments. Arsenic contained in tailings mainly occurs as sorbed and/or co-precipitated species in Fe(III)-hydroxides (arsenical ferrihydrites with various Fe/As molar ratios) which form coatings around silicate

grains (quartz, K-feldspar, chlorite, biotite and muscovite), indicating a precipitation process which likely took place straight after the mine slurry discharge from the flotation plant into the BLS.

The BLS is characterized by a torrential regime (discharge: <0.01-100 m<sup>3</sup>/s), with alternating of minimum flows and periodic, sometimes catastrophic, floods. During the prolonged dry periods, the stream waters flow underground for long stretches, and the bed sediments remain exposed to the air.

The BLS waters have pH = 7-8, Eh (redox potential) = 0.4-0.5 V, TDS (total dissolved solids) = 0.2-0.4 g  $l^{-1}$  and Ca-Mg-SO<sub>4</sub> to Ca-Mg-HCO<sub>3</sub> composition; As concentrations up to 1 mg  $l^{-1}$  were measured.

Twelve sediment samples were taken from the river channel upstream and downstream of the flotation plant in about 10 cm depth (Fig. 1). The sample numbers reflect increasing flow distance from the source of the BLS. Two samples of tailings were also taken on the river banks (samples A and B). The water saturated samples were stored at 4°C until analysis.

#### 9.3.2 Palude Sa Masa

The second test site is a wetland (Palude Sa Masa) in SW Sardinia in the Iglesias mining area, near the small village of Gonnesa. The Sa Masa wetland is located at the mouth of the small Rio Sa Masa, derived from the joining of both Rio Sa Crabiola and Rio di Gonnesa which drain the old Zn-Pb mining district of the Iglesias Valley.

The biggest mines of the whole island of Sardinia occurred in the hills around the town of Iglesias and their tailings and dumps dot the landscape. The upper part of the sulfide ore has been repeatedly subjected to supergene oxidation, which caused the deposition of Zn-Pb carbonates and silicates, as well as the partial immobilization of Fe and Mn in deep reaching gossans. Arsenic is present in both primary and secondary ores, even if much lower amounts as in the Baccu Locci area. A preliminary geochemical study of the stream sediments in the Iglesiente area has been performed by Boni et al. (BONI et al., 1999).

The wetland is fed by groundwater, by runoff from the hills and, in former times, also by the two streams Rio Sa Crabiola and Rio di Gonnesa, which are channeled nowadays. During summer the groundwater level drops and the soil falls periodically

dry. After rainfall in autumn the groundwater level rises again and the soil gets humid. The hills in the north have only minor vegetation and bear a lot of mine dumps, which are easily washed out during the rain period. These mine dumps are supposed to be the main arsenic source. The catchments of the rivers also have several mine dumps, so that the rivers transport contaminated material and water to the wetland. The soil consists of silty river sediments which are contaminated with heavy metals and arsenic. Today the seasonally varying soil moisture, which leads to varying redox conditions, seems to be the most important factor mediating the arsenic distribution in the wetland.



# Fig. 28 Schematic map of the Palude Sa Masa region with indication of the sampling points and the flow path

Two sediment samples (sample no 13 and 14) were taken near the river channels upstream of the wetland in order to get an idea about the arsenic input into the wetland from the sources in the river catchment. Eleven soil samples were randomly taken from the wetland (Fig. 28). One additional sample (12) was taken from the beach near the river mouth. All samples were collected during the dry season.

#### 9.4. Analytical methods

At first the supernatant water from the saturated sediment samples was filtered and analyzed. Extraction methods and long term extraction tests were conducted with fresh sediment and soil samples. Total element concentrations were analyzed using dried and ground sediment and soil samples after microwave digestion. The handling of the samples as well as all parameters measured in the different samples are illustrated in Fig. 29.



#### Fig. 29 Handling of the samples and analyzed parameters

The anions Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup> were measured with an ion chromatograph (Dionex DX-120) using an AS-14-A analytical column and a carbonate/bicarbonate eluent with a flow rate of 1 ml min<sup>-1</sup>. The elements Fe, Mn, Mg, Na, Ca, Zn, Pb, Cu, Cd and K were measured with an ICP-OES (Varian VISTA-MPX axial). The arsenic species were determined with a FIAS-AAS. At first only As(III) was measured. For total arsenic concentration reduction solution was added to the water samples. The reduction solution was composed of 100 g l<sup>-1</sup> KI and 100 g l<sup>-1</sup> ascorbic acid and mixed with 6 M HCl and water sample using a volume ratio of 1:4:5. In the FIAS (Perkin-Elmer 200) the sample solution was mixed with 2 g l<sup>-1</sup> NaBH<sub>4</sub>, 0.2 g l<sup>-1</sup> NaOH and 1.2 M HCl. The flow rate of the sample solution was 2 ml min<sup>-1</sup>, NaBH<sub>4</sub> + NaOH 1.6 ml min<sup>-1</sup> and HCl 1.6 ml min<sup>-1</sup>. In a gas liquid separator arsine was purged and transported in an argon flow to the quartz cuvette (argon flow rate 80 ml min<sup>-1</sup>). The cuvette converts the arsine at a temperature of 900 C° to elemental arsenic, which is detected with an AAS (Perkin-Elmer AAS 4100) at a wavelength of 193.7 nm.

Before the microwave digestion (MLS-ETHOS Plus) the solid samples were dried (120  $^{\circ}$ C for 24 h) and ground. Afterwards 0.1 g sample amount was added to 2 ml

 $HNO_3$ , 0.5 ml  $H_2O_2$  and 0.5 HF. The microwave was programmed for 35 minutes heating up to 200 °C. After the digestion the solubilised samples were diluted up to 20 ml with  $H_2O$  (ultra pure).

Over a period of 24 hours, extractions were made with 2.5 g sample amount and 30 ml extraction solution. As extraction solution we used four different chemicals:  $H_2O$  (ultra pure), 1 M sodium ammonium hydrogen phosphate, 1 M hydroxyl ammonium chloride and 1 M HCl. After 24 hours the extraction solutions were filtered (0.45µm) and analyzed.

Selected solid samples were extracted with  $H_2O$  (ultra pure) over a period of 38 days in order to observe the long-term behavior of arsenic release. Two tailing samples and two sediment samples from the BLS (4, 6) with high arsenic concentration were chosen for this test. The water was changed every 24 hours, filtered and arsenic species were measured immediately.

#### 9.5 Results and interpretation

#### 9.5.1 Baccu Locci Stream

The water quality of the BLS in equilibrium with the sediment is evaluated from the supernatant of the water saturated sediment samples (Tab. 7). Results of PHREEQC calculations indicate that all water samples are saturated with respect to calcite and also to dolomite further downstream (e.g. sample 11), due to the carbonate content of the rocks. Calcium and bicarbonate contents as well as pH-values vary depending on the dilution of the water from the tributaries. The influence of the flotation plant (i.e. the presence of tailings mixed with the bed sediments) can be deduced from the sulfate and arsenic contents of the water. The overall concentrations of sulfate are significantly high with values between 3.8 and 1.6 mM, showing a general decreasing trend downstream. However, the water is under saturated with respect to gypsum, which is a typical secondary mineral of sulfides weathering. Other typical products of iron sulfides weathering, such as acidity and high iron content, can not be detected in the water. The acidity is buffered by reactions between the water and the carbonate rocks, whereas Fe<sup>2+</sup> is oxidized to Fe<sup>3+</sup> by atmospheric oxygen and subsequently removed by precipitation as hydroxide. Only in the first sample, which was taken near

the waste rock dumps upstream of the flotation plant, relatively high concentrations of iron and manganese were measured. We assume that this was a result of an in-situ arsenopyrite oxidation under reduced redox conditions occurred in the sample during storage.

sample	km *	рН	Са	Mg	Κ	Na	Zn	CI	NO <sub>3</sub>	SO4 <sup>2-</sup>	Fe	Mn	HCO <sub>3</sub> <sup>-</sup> **	As(III)	As(V)
							[1	nmol/	1]					[µmol/l	]
1	1.30	7.53	4.01	2.39	0.08	2.23	0.010	3.09	0.07	3.87	0.050	0.029	4.21	0.05	4.36
2	1.99	7.55	3.41	1.13	0.07	1.09	0.017	1.16	0.17	3.84	n.n.	n.n.	1.22	0.03	2.44
3	2.31	7.36	2.24	1.29	0.13	1.07	0.010	1.12	0.12	2.75	n.n.	n.n.	1.53	0.02	1.50
4	2.82	7.19	2.85	1.23	0.08	1.21	0.067	1.25	0.08	3.04	n.n.	0.002	2.04	0.09	5.20
5	3.64	7.86	2.78	0.87	0.07	1.50	0.002	1.23	0.03	3.33	n.n.	n.n.	0.95	0.14	7.88
6	4.31	7.8	2.76	1.36	0.07	1.52	0.002	1.57	0.09	3.62	n.n.	n.n.	0.92	0.18	14.10
7	5.17	8.18	2.95	1.30	0.08	1.80	0.005	1.75	0.05	3.13	n.n.	n.n.	2.30	0.20	13.73
8	5.60	7.95	2.87	1.12	0.07	1.70	0.003	1.95	0.07	2.56	n.n.	n.n.	2.61	0.15	13.80
9	5.93	7.8	1.87	0.99	0.08	1.69	0.002	1.93	0.11	1.85	n.n.	n.n.	1.76	0.13	7.51
10	6.41	7.73	1.92	0.93	0.09	1.60	0.002	1.89	0.20	1.66	0.002	n.n.	1.97	0.12	7.15
11	7.08	7.94	2.57	1.29	0.18	2.02	0.002	2.22	0.07	2.14	0.003	0.001	3.34	0.16	9.66
12	8.51	7.84	2.08	1.28	0.07	2.03	0.001	2.31	0.14	1.92	0.002	n.n.	2.54	0.10	7.80

Tab. 7 Chemical composition of the supernatant water of the BLS sediment samples

The As(V) content in the supernatant is elevated in all samples with values between 1.5 and 14  $\mu$ M and maximum concentrations directly downstream of the flotation plant (Fig. 30). The species distribution shows only minor concentrations of As(III), whereas As(V) is the predominant species (Tab. 7).





The sediment was characterized with respect to the iron content. The total iron content of the sediment is relatively high with values between 2.2 wt. % and 3.5 wt. % showing a lightly decreasing trend downstream. The sources of the iron are the waste rock dumps and the tailings, respectively occurring upstream and downstream of the flotation plant. When arsenopyrite oxidation takes place, Fe(II) may be transported as dissolved fraction and subject to geochemical reactions. This reaction may be the oxidation by oxygen or manganese oxides and following precipitation as iron hydroxide forming coatings on the sediment particles or the formation of siderite in reduced sediments. Ferric iron might also be physically transported in particle form. Results of the extraction tests described above indicate a higher mobility of iron downstream, where the flotation tailings mixed with the stream sediments occur. This can be deduced from the increase of the fraction of easily reducible iron extracted by hydroxyl ammonium chloride (Fig. 31).



Fig. 31 Iron concentration extracted by hydroxyl ammonium chloride

The total manganese content of the sediments was in the range of 400 to 700 mg kg<sup>-1</sup>. The total arsenic content in the sediments is very high with values up to 3700 mg kg<sup>-1</sup>. The distribution of arsenic in the stream sediments exhibits a close relationship to the possible sources and shows the peak concentration immediately downstream of the flotation plant (Fig. 32) from which the tailings were discharged into the stream. The gradual decrease in arsenic concentration is then due to decreasing amounts of tailings mixed with the stream sediments as the distance from the flotation plant increases.



Fig. 32 Total arsenic concentrations in the sediments of the BLS

Thus the flotation plant has been the most important source of arsenic contamination. In fact, total arsenic concentrations in the two tailing samples are as high as 20000 and 24000 mg kg<sup>1-</sup>.

The mobility of arsenic was evaluated using the As concentrations extracted by the four solutions which represent different mobility fractions of arsenic: soluble form (water), exchangeable form (sodium hydrogen phosphate), bound to easily reducible minerals (hydroxyl ammonium chloride) and crystalline iron hydroxide and carbonate fraction (hydrochloric acid). In all extracted solutions the amount of As(III) was negligible.

By comparing the total amount of arsenic in the solid samples with the amount dissolved by  $H_2O$  we get an idea of the most mobile arsenic fraction [%] (Fig. 33a). The highly soluble fraction in the stream sediments is between 0.05 and 0.5 % of the total content. The mobility increases significantly downstream. The arsenic found in the stream sediments tends to be ten to hundred times more mobile than the arsenic in the tailings.



Fig. 33 Arsenic concentrations, given as percentage of the total amount, extracted from the BLS sediments with (a)  $H_2O$ ; (b) hydroxyl ammonium chloride; (c) sodium ammonium hydrogen phosphate. Samples A and B are tailings.

This trend of increasing mobility can also be seen in the exchangeable and in the easily reducible fractions (Fig. 33b and Fig. 33c). The amounts of these fractions are about 10-100 times higher than the soluble form. Both fractions represent the amount of arsenic bound to amorphous iron hydroxides, which are recently precipitated or have been physically transported in the stream. This can also be deduced by the close correlation between the distribution of easily reducible iron (Fig. 31) and the respective arsenic fraction (Fig. 33b). The highest arsenic amount comprising 30 to 65 % is anyway bound to crystalline iron hydroxides.

Further information about the binding forms and the ongoing processes in the sediments and the tailings was drawn from long-term extraction tests with water

using selected samples with high arsenic concentrations. The release of arsenic with time is shown for one sediment and one tailing sample as typical examples (Fig. 34a and Fig. 34b). It is important to stress that interaction of water with stream sediments and tailings produces near-neutral and acidic pH conditions, respectively.



Fig. 34 (a) Arsenic release over time from a sediment sample by long-term extraction. (b) Arsenic release over time from a tailing sample by long-term extraction.

Also in the long-term experiments only As(V) was observed. The sediment sample showed a permanent decrease in As(V) concentration (Fig. 34a). This figures the leaching from the sediment surface without further geochemical reactions of arsenic bearing minerals. In contrast the trend of arsenic release with time from the tailing sample is inverse (Fig. 34b). After a sharp drop in concentration in the first days it slightly increases indicating ongoing geochemical reactions in the material. Probably an in-situ arsenopyrite oxidation leads to a continuing arsenic release.

As shown above, the redox conditions in the BLS are oxic and the arsenic is observed as As(V). The mobility of As and Fe increases with flow distance. The transport behavior in the BLS catchment is primarily influenced by the flow rate of the stream, which varies seasonally. During high flow rates the transport of sediment and tailing material leads to a physical transport of As bound to iron. Whereas during low flow rates leaching processes are dominant and the arsenic is mainly transported as dissolved fraction.

#### 9.5.2 Palude Sa Masa

The geochemistry of the sediments in the Sa Masa wetland is strongly influenced by the exploitation of several sulfide ores in the area, so that the contents of various heavy metals, e.g. Pb and Cd, are very high. The samples can be subdivided into four groups depending on their distance to the stream lines and the original mine sites. These groups differ in the element concentrations of sulfur (pyrite), calcium (calcite), iron, manganese, lead and arsenic:

- samples influenced directly by the mined ore deposits on the hills reflecting the **local sources** (1-4)
- sediments mainly influenced by Rio Sa Crabiola (5,6)
- sediments downstream of Rio Gonnesa, Rio Sa Masa and the local sources (7-12)
- sediment load of the rivers upstream of the local mine deposits (13-14)

The sediment load of both rivers upstream of the wetland differs notably. The Rio Sa Crabiola sample (13) has low concentrations of calcium, iron and manganese (Tab. 8). In spite of the sulfur content is about 1 %, the load of heavy metals and arsenic is relatively low compared to the other samples. In contrast the load of heavy metals and arsenic of the Rio Gonnesa sample (14) is much higher and even combined with higher calcium, iron, sulfur and manganese concentrations.

The arsenic concentrations in the supernatant water of these two samples are in contrast to the contents in the sediments. Sample 13 has a relatively high concentration whereas sample 14 from Rio Gonnesa has nearly no dissolved arsenic. These differences can be explained by the different redox conditions in the two samples. In sample 13 we found a high As(III) portion as well as a significant concentration of Fe<sup>2+</sup> indicating pyrite oxidation in the sample. In sample 14 no Fe<sup>2+</sup> was detectable. Extractions of the sediments with water showed the same effect.

group	sample	Ca [%]	Mn [mg/kg]	Fe [%]	S [%]*	Pb [mg/kg]	As [mg/kg]
	1	4.03	1196	3.21	1.39	11133	73.1
samples reflecting	2	10.9	1617	3.12	0.52	19534	57.9
local sources	3	16.2	2606	1.51	0.29	10461	22.4
	4	7.53	1889	7.72	2.10	9671	156
samples	5	13.4	1622	1.36	0.54	7700	9.4
influenced by Rio sa Crabiola	6	6.95	764	3.08	0.03	122	21.7
	7	14.4	2217	5.09	1.05	9409	30.2
samples	8	14.6	1255	3.71	0.91	5850	32.5
downstream of	9	2.11	1422	4.73	0.05	286	53.1
Rio di Gonnesa and	10	2.16	1032	4.77	0.10	415	46.1
local sources	11	4.97	275	1.11	0.24	2380	10.5
	12	8.41	522	1.40	n.m.	1299	24.0
samples upstream of	13	1.31	1537	0.99	0.83	710	1.6
Paluda sa Masa	14	8.06	210	4.67	1.47	12195	38.5

Tab. 8 Total concentrations of calcium, iron, sulfur, lead and arsenic from the four groups of the Palude Sa Masa samples

The input from local sources can be pointed out by the samples 1 to 4, with a maximum As concentration of  $156 \text{ mg kg}^{-1}$  (sample 4) (Tab. 8). Also Pb concentrations in this area are extremely high with values up to 19 g kg<sup>-1</sup> (sample 2). In these samples arsenic is closely correlated with iron (Fig. 35) and sulfur indicating that these elements come from the same mineral, probably arsenic bearing pyrite and its fresh oxidation products.



Fig. 35 Correlation between iron and arsenic in samples 1-4

The mobility of arsenic in these samples is relatively high, except from the water extraction. All extracts have the highest portion of As(III) compared to the other sample groups. Most of the mobile arsenic is in an exchangeable form (Fig. 36). The

in situ As-bearing pyrite oxidation leads to the release of As(III) which is then partly oxidized to As(V) and both are bound to amorphous iron hydroxide surfaces.

As shown in Fig. 36, As(III) is more mobile and less strongly adsorbed onto iron hydroxides than As(V). Sample 2 exhibits a different speciation for As(III) and As(V). Most of As(III) is in an exchangeable form, whereas As(V) prevalently occurs in a strongly bound form.



Fig. 36 Extracted As(V) and As(III) concentrations of the "source samples" calculated as percentage of the total arsenic concentration.

The geochemistry of the samples 7-12 is influenced by the mine deposits on the hills and their dumps (local sources) as well as by Rio Gonnesa. The sediments have varying calcium contents (from 2 % up to 15 %), which is correlated with varying sulfur ( $r^2 = 0.8$ ) and lead ( $r^2 = 0.8$ ) contents. This could be an indication for gypsum formation in the sediment, as well as for the presence of anglesite or/and galena. The high iron concentrations in samples 7-10 significantly decrease downstream (samples 11-12). The arsenic load is relatively homogenous in samples 7-10 and the correlation to the iron content is less obvious than in the source samples ( $r^2 = 0.7$ ). Samples 11 and 12 are located further downstream of the sources and the absolute concentrations. Downstream we found significantly lower As(III) concentrations in the extracts than in the source samples, but we could observe an increase of mobility with flow path in the dissolved, easily reducible and the exchangeable fractions.

Samples 5 - 6 are located in the former infiltration area of the Rio Sa Crabiola and not directly downstream of the local sources. Corresponding to this fact the concentrations of the elements of interest are relatively low (Tab. 8). In sample 5 the lead concentration is higher than in sample 6 correlated to higher calcium and sulfur concentrations, whereas in sample 6 we found more arsenic load together with slightly higher iron content. In these samples we observed only minor As(III), whereas the mobility of As(V) was in between the source and the downstream samples.

As shown above, the distribution and mobility of arsenic in the Sa Masa wetland is influenced by different factors such as the hydraulic system, the location of mine deposits (sources) (Fig. 28) as well as the mineralogy of the sediments. A factor which should also be taken into consideration is the seasonal variation of soil humidity. This variation leads to changing redox conditions in the sediments, with iron reduction under wet conditions and iron oxidation during dry periods. The iron cycle is closely linked to the release or the accumulation of arsenic and thus is the main factor driving the transport of arsenic in this wetland.

#### 9.6 Comparison of the two Sardinian Case Studies

Worldwide a lot of anthropogenic arsenic contamination can be attributed to mining activities. Therefore the understanding of reaction pathways and transport processes of arsenic in such environments is important. These processes differ notably depending on the geological, geochemical and hydrological framework, and have thus to be investigated in case studies.

The two sites presented here have been affected by mining for a long time. In the first case we studied sediments from a stream channel, whereas the second case study describes a wetland. The two hydraulic situations reveal different physical and geochemical transport conditions which influence the sort and intensity of arsenic accumulation in the sediments by a factor of 20. In both systems natural acidity buffering plays an important role, as shown by pH values between 7 and 8 in stream waters. Therefore the direct effect of the acid mine drainage is almost absent but is still visible in the environment by typical secondary mineral formation and oxidation of the products.

In the case of the BLS the transport of As(V) is mainly depending on the flow rate, which differs extremely over the year. High flow rates cause translocation processes of the tailing material and the stream sediments. During periods with low flow rates the arsenic transport is dominated by the dissolved fraction. This originates from the tailings as well as from the stream sediments. Due to the oxic conditions in both the stream water and the sediments, only As(V) can be observed.

Also in the Sa Masa wetland varying hydraulic situations play an important role concerning the transport of arsenic, but in this case the influence is mostly geochemical. During rain periods the sediments become water saturated so that the redox conditions get anoxic and iron reduction occurs. Together with the iron also arsenic is reduced, released and transported as As(III) which is more mobile and less easily adsorbed onto iron hydroxides than As(V). In summer the sediments dry out and the iron as well as the arsenic get oxidized again. The arsenic then is bound to the freshly precipitated ferric iron hydroxides until the seasonal cycle starts again. Thus the arsenic transport velocity is slow and the accumulation rates are much lower compared to the BLS site.

### 9.7 Acknowledgements

We thank Marcello Romano for taking the samples in the Sa Masa wetland as well as Luca Fanfani.

#### 10. Literature

- Abrams R. H. and Loague K. (2000) A compartmentalized solute transport model for redox zones in contaminated aquifers 2. Fieldscale simulations. *Water Resour. Res.* 36, 2015-2029.
- Abrams R. H., Loague K., and Kent D. B. (1998) Development and testing of a compartmentalized reaction network model for redox zones in contaminated aquifers. *Water Resour. Res.* **34**, 1531-1541.
- Acharyya S. K., Lahiri S., Raymahashay B. C., and Bhowmik A. (2000) Arsenic toxicity in groundwater in parts of Bengal Basin, in India and Bangladesh: role of Quaternary stratigraphy and Holocene sea-level fluctuation. *Environ. Geol.* **39**, 1127-1137.
- Ahmann D., Krumholz L. R., Hemond H. F., Lovley D. R., and Morel F. M. M. (1997)
   Microbial mobilization of arsenic from sediments of the Aberjona Watershed.
   *Environ. Sci. Technol.* **31**, 2923-2930.
- Ahmed K. M., Bhattacharya P., Hasan M. A., Akhter S. H., Alam S. M. M., Bhuyian M. A. H., Imam M. B., Khan A. A., and Sracek O. (2004) Arsenic enrichment in groundwater of the alluvial aquifers in Bangladesh: an overview. *Appl. Geochem.* **19**, 181-200.
- Alam M. G. M., Tokunaga S., and Maekawa T. (2001) Extraction of arsenic in a sythetic arsenic-contaminated soil using phosphate. *Chemosphere* **43**, 1035-1041.
- Amita J., Raven K. P., and Loeppert R. H. (1999) Arsenite and Arsenate Adsorption on Ferrihydrite: Surface Charege Reduction and Net OH<sup>-</sup> Release Stoichiometry. *Environ. Sci. Technol.* 33, 1179-1184.
- Andreae M. O. (1980) Arsenic in rain and the atmospheric mass balance of arsenic. *J. Geophys Res* **85**(C8), 4512-4518.

- Andreae M. O. and Andreae T. W. (1989) Dissolved arsenic species in the Schelde estuary and watershed, Belgium, Estuarine. *Coastal and Shelf Science* **29**, 421-433.
- Andreae M. O., Byrd J. T., and Froelich P. N. (1983) Arsenic, antimony, germanium, and tin in the Tejo Estuary, Portal: modeling a polluted estuary. *Environ. Sci. Technol.* **17**, 731-737.
- Appelo C. A. J., van der Weiden M. J. J., Tournassat C., and Charlet L. (2002) Surface complexation of ferrous iron and carbonate on ferrihydrite and the mobilization of arsenic. *Environ. Sci. Technol.* **36**, 3096-3103.
- Arai Y., Elzinga E. J., and Sparks D. L. (2001) X-ray absorption spectroscopy investigation of arsenite and arsenate adsorption at the aluminum oxide-water interface. *J. Colloid Interf. Sci.* **235**, 80-88.
- Ayotte J. D., Montgomery D. L., Flanagan S. M., and Robinson K. W. (2003) Arsenic in groundwater in eastern New England: occurrence, controls, and human health implications. *Environ. Sci. Technol.* **37**, 2075-2083.
- Azcue J. M., Mudroch A., Rosa F., and Hall G. E. M. (1994) Effects of abandoned gold mine tailings on the Arsenic concentrations in water and sediments of Jack of Clubs Lake, B.C. *Environ. Sci. Technol.* **15**, 669-678.
- Azcue J. M. and Nriagu J. O. (1994) Arsenic: historical perspectives. In *Advances in Environmental Science and Technology*. Wiley.
- Azcue J. M. and Nriagu J. O. (1995) Impact of abandoned mine tailings on the arsenic concentrations in Moira lake, Ontario. *J. Geochem. Explor.* **52**, 81-89.
- Barbaris B. and Bettertin E. A. (1996) Initial snow chemistry survey of the Mogollon rim in Arizona. *Atmos. Environ.* **30**, 3093-3103.
- Barber L. B. I., Thurman E. M., and Runnells D. D. (1992) Geochemical Heterogeneity in a Sand and Gravel Aquifer: Effect of Sediment Mineralogy and Particle Size on the Sorption of Chlorobenzenes. *J. Contam. Hydrol.* **9**, 35-54.

- Belzile N. and Lebel J. (1986) Capture of arsenic by pyrite in near shore sediments. *Chem. Geol.* **54**, 279-281.
- Benning L. G., Wilkin R. T., and Barnes H. L. (2000) Reaction pathways in the Fe-S system below 100 degrees. *C. Chem. Geol.* **167**, 25-51.
- Benson L. V. and Spencer R. J. (1983) A hydrochemical reconnaissance of the Walker River Basin, California and Nevada: U.S., pp. 83-740. Geological Survey Open-File Report.
- Berner R. A. (1981) A new geochemical classification of sedimentary environments. *J. Sediment. Petrol.* **51**, 359-365.
- Bhattacharya P., Chatterjee D., and Jacks G. (1997) Occurrence of arseniccontaminated groundwater in alluvial aquifers from Delta Plains, eastern India. *Int. J. Water Resour. D.* **13**, 79-92.
- Böhlke J. K., Wanty R. B., Tuttle M., Delin G., and Landon M. (2002) Denitrification in the recharge area and discharge area of a transient agricultural nitrate plume in a glacial outwash sand aquifer, Minnesota. *Water Resour. Res.* 38, 1105, doi:10.1029/2001WR000663.
- Boni M., Costabile S., De Vivo B., and Gasparrini M. (1999) Potential environmental hazard in the mining district of Southern Iglesiente (SW Sardinia, Italy). *Journ. Geochem. Expl.* **67**, 417-430.
- Bowell R. J. (1994) Sorption of arsenic by iron oxides and hydroxides in solis. *Appl. Geochem.* **9**, 279-286.
- Bright D. A., Dodd M., and Reimer K. J. (1996) Arsenic in subarctic lakes influenced by gold mine effluent: The occurrence of organoarsenicals and 'hidden' arsenic. *Sci. Total Environ.* **180**, 165-182.
- Bundschuh J., Farias B., Martin R., Storniolo A., Bhattacharya P., Cortes J., Bonorino G., and Albouy R. (2004) Groundwater arsenic in the Chaco-Pampean PLain, Argentina: case study from Robles country, Santiago del Estero Province. *Appl. Geochem.* **19**, 231-243.

- Canfield D. E., Raiswell R., and Bottrell S. (1992) The reactivity of sedimentary iron minerals toward sulfide. *Am. J. Sci.* **292**, 659–683.
- Chapelle F. H. and McMahon P. (1991) Geochemistry of dissolved inorganic carbon in a Coastal-Plain aquifer 1. Sulfate from confining beds as an oxidant in microbial CO<sub>2</sub> production. *Jour. of Hydrology.* **127**, 85-108.
- Chapelle F. H., McMahon P. B., Dubrovsky N. M., Fujii R. F., Oaksford E. T., and Vroblesky D. A. (1995) Deducing the distribution of terminal electron-accepting processes in hydrologically diverse groundwater systems. *Water Resour. Res.* 31, 359-371.
- Chen Y. W., Deng T. L., Filella M., and Belzile N. (2003) Distribution and Early Diagenesis of Antimony Species in Sediments and Porewaters of Freshwater Lakes. *Environ. Sci. Technol.* **37**, 1163-1168.
- Cherry J. A., Shaikh A. U., Tallman D. E., and Nicholson R. V. (1979) Arsenic species as an indicator of redox conditions in groundwater. *J. Hydrol.* **43**, 373-392.
- Chiu V. Q. and Hering J. G. (2000) Arsenic adsorption and oxidation at manganite surfaces. 1- Method for simultaneous determination of adsorbed and dissolved arsenic species. *Environ. Sci. Technol.* **34**(10), 2029-2034.
- Christensen T. H., Bjerg P. L., Banwart S. A., Jakobsen R., Heron G., and Albrechtsen H. J. (2000) Characterization of redox conditions in groundwater contaminant plumes. *J. Contam. Hydrol.* **45**, 165-241.
- Coston J. A., Fuller C. C., and Davis J. A. (1995) Pb<sup>2+</sup> and Zn<sup>2+</sup> adsorption by a natural aluminum- and iron- bearing surface coating on an aquifer sand. *Geochim. Cosmochim. Acta* **59**(17), 3535-3547.
- Courtin-Nomade A., Bril H., Neel C., and Lenain J. F. (2003) Arsenic in iton cements developed within tailings of a former metalliferous mine Enguialès, Aveyron, France. *Appl. Geochem.* **18**, 395-408.
- Cullen W. R. and Reimer K. J. (1989) Arsenic Speciation in the Environment. *Chem. Rev.* **89**, 713-764.

- Cummings D. E., Caccavo F., JR., Fendorff S., and Rosenzweig R. F. (1999) Arsenic mobilization by the dissimilatory Fe(III)-reducing bacterium Shewanella alga BrY. *Environ. Sci. Technol.* **33**, 723-729.
- Darland J. E. and Inskeep W. P. (1997) Effects of pH and phosphate competition on the transport of arsenate. *Journal of Environmental Quality* **26**, 1133-1139.
- Davis J. A., Kent D. B., Rea B. A., Garabedian S. P., and Anderson L. D. (1991) Effect of the geochemical environment on heavy-metal transport in ground water. In U.S. Geological Survey Water-Resources Investigations Report, Vol. 91-4034 (ed. G. E. Mallard and D. A. Aronson), pp. 53-62.
- Dixit S. and Hering J. G. (2003) Comparison of Arsenic(V) and Arsenic(III) Sorption onto Iron Oxide Minerals: Implications for Arsenic Mobility. *Environ. Sci. Technol.* **37**, 4182-4189.
- dos Santos Afonso M. and Stumm W. (1992) Reductive dissolution of iron(III) (hydr)oxides by hydrogen sulfide. *Langmuir* **8**, 1671-1675.
- Dowdle P. R., Laverman A. M., and Oremland R. S. (1996) Bacterial dissimilatory reduction of arsenic (V) to arsenic (III) in anoxic sediments. *Appl. Environ. Microbiol.* 62, 1664-1669.
- DPHE B., and MML. (1999) Groundwater Studies for Arsenic Contamination in Bangladesh. British Geological Survey.
- Driehaus W., Seitl R., and Jekel M. (1995) Oxidation of Arsenate III with manganese oxides in water treatment. *Water Res.*
- Dzombak D. A. and Morel F. M. M. (1990) *Surface complexation modeling: Hydrous ferric oxide*. John Wiley & Sons.
- Eary L. E. (1992) The solubility of amorphous  $As_2S_3$  from 25 to 90°C. *Geochim. Cosmochim. Acta* **56**, 2267-2280.
- Eary L. E. and Rai D. (1988) Chromate removal from aqueous waste by reduction with freeous iron. *Environ. Sci. Technol.* **22**, 972-977.

- Eary L. E. and Schramke J. A. (1990) Rates of inorganic oxidation reactions involving dissolved oxygen. In *Chemical Modeling in Aqueous Systems II*, Vol. Am. Chem. Soc. Symp. Series 416 (ed. D. C. Melchior and R. L. Bassett), pp. 379-396.
- Ford R. G. (2002) Rates of hydrous ferric oxide crystallization and the influence on coprecipitated arsenate. *Environ. Sci. Technol.* **36**, 2459-2463.
- Frau F. and Ardau C. (2003) Geochemical controls on arsenic distribution in the Baccu Locci stream catchment (Sardinia, Italy) affected by past mining. *Appl. Geochem.* 18, 1373-1386.
- Frau F. and Ardau C. (2004) Mineralogical controls on arsenic mobility in the Baccu Locci stream catchment (Sardinia, Italy) affected by past mining. *Mineral Mag* 68, 15-30.
- Froelich P. N., Kaul L. W., Byrd J. T., Andreae M. O., and Roe K. K. (1985) Arsenic, barium, germanium, tin, dimethylsulfide, and nutrient biogeochemistry in Charlotte Harbor, Florida, a phosphorus-enriched estuary, Estuarine. *Coastal* and Shelf Science **20**, 239-264.
- Froelich P. N., Klinkhammer G. P., Bender M. L., Luedtke N. A., Heath G. R., Cullen D., Dauphin P., Hammond D., Hartman B., and Maynard V. (1979) Early oxidation of organic matter in pelagic sediments of the eastern equatorial Atlantic: Suboxic diagenesis. *Geochim. Cosmochim. Acta* 43, 1075-1090.
- Fukushi K., Sasaki M., Sato T., Yanase N., Amano H., and Ikeda H. (2003) A natural attenuation of arsenic in drainage from an abandoned arsenic mine dump. *Appl. Geochem.* 18, 1267-1278.
- Fuller C. C., Davis J. A., and Waychunas G. A. (1993) Surface chemistry of ferrihydrite: Part 2. Kinetics of arsenate adsorption and coprecipitation. *Geochim. Cosmochim. Acta* 57, 2271-2282.
- Galán E., Gómez-Ariza J. L., González I., Fernández-Caliani J. C., Morales E., and Giráldez I. (2003) Heavy netal partitioning in river sediments severely polluted by acid mine drainage in the Iberian Pyrite Belt. *Appl. Geochem.* **13**, 409-421.

- Harrington J. M., Fendorf S. E., and Rosenzweig R. F. (1998) Biotic generation of arsenic(III) in metal(loid)-contaminated lake sediments. *Environ. Sci. Technol.* 32, 2425-2430.
- Harvey C. F., Swartz C. H., Badruzzaman A. B., Keon-Blute N., Yu W., Ali M. A., Jay J., Beckie R., Niedan V., Brabander D., Oates P. M., Ashfaque K. N., Islam S., Hemond H. F., and Ahmed M. F. (2002) Arsenic Mobility and Groundwater Extraction in Bangladesh. *Science. Vol.* 298, 1602-1606.
- Haury V., Jann S., Kofod M., Scholz C., and Isenbeck-Schroter M. (2000) Redoxinduced species distribution of arsenic in a suboxic groundwater environment column experiments. *International conference on groundwater research*, 197-198.
- Hiemstra T. and van Riemsdijk W. H. (1999) Surface structural ion adsorption modeling of competitive binding of oxyanions by metal (hydr)oxides. *J. Colloid Interf. Sci.* **210**, 182-193.
- Hingston F. J., Posner A. M., and Quirk J. P. (1971) Competitive adsorption of negatively charged ligands on oxide surfaces. *Disc. Farad. Soc.* **52**, 334-342.
- Höhn R., Isenbeck-Schröter M., Niedan V., Scholz C., Tretner A., Jann S., Stadler S., Kent D., and Davis J. (2005a) Tracer test with Arsenic (V) to study transport and reaction rates in an iron-reducing environment at the USGS Cape Cod Site (Mass., USA). *Geochim. Cosmochim. Acta* submitted.
- Höhn R., Isenbeck-Schröter M., Niedan V., Scholz C., Tretner A., Jann S., Stadler S., Kent D. B., Davis J. A., and Jakobsen R. (2001) Tracer test with arsenic(V) in an iron-reducing environment at the USGS Cape Cod Site (Mass. USA). *WRI-10*, 1099-1102.
- Höhn R., Isenbeck-Schroter M., and Wallschläger D. (2005b) Arsenic reduction by dissolved sulfide at MnO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub> surfaces. *Water Res.* **submitted**.
- Holleman A. F. and Wiberg E. (1990) *Lehrbuch der Anorganischen Chemie*. WAlter de Gruyter& Co.

- Huerta-Diaz M. A. and Morse J. W. (1992) Pyritisation of trace metals in anoxic marine sediments. *Geochim. Cosmochim. Acta* **56**, 2681-2702.
- Isenbeck-Schröter M., Doring U., Moller A., Schröter J., and Matthes G. (1993) Experimental approach and simulation of the retention processes limiting orthophosphate transport in groundwater. *J. Contam. Hydrol.* **14**, 143-161.
- Jakobsen R., Albrechtsen H. J., Rasmussen M., Bay H., Bjerg P. L., and Christensen T. H. (1998) H<sub>2</sub> Concentrations in a Landfill Leachate Plume (Grinsted, Denmark): In Situ Energetics of Terminal Electron Acceptor Processes. *Environ. Sci. Technol.* 32, 2142-2148.
- Jambor J. L. and Blowes D. W. (1998) Theory and applications of mineralogy in environmental studies of sulfide-bearing mine waste. In *Short Course Handbook* on Ore and Environmental Mineralogy, Vol. 27 (ed. L. J. Cabri and D. J. Vaughan), pp. 367-401.
- Johnson C. A. and Thornton I. (1987) Hydrological and chemical factors controlling the concentrations of Fe, Cu, Zn and As ina river system contaminated by acid mine drainage. *Water Res.* **21**(3), 359-365.

Kabata-Pendias A. (2001) Trace Elements in Soils and Plants. CRC.

- Kent D. B., Davis J. A., Anderson L. C. D., Rea B. A., and Wait T. D. (1994)
   Transport of chromium and selenium in the suboxic zone of a shallow aquifer:
   Influnece of redox and adsorption reactions. *Water Res.* **30**, 1099-1114.
- Kent D. B. and Fox P. M. (2004) The influence of groundwater chemistry on arsenic concentrations and speciation in a quartz sand and gravel aquifer. *Geochemical Transactions* 5(1), 1-12.
- Keon N. E., Swartz C. H., Brabander D. J., Harvey C., and Hemond H. F. (2001) Validation of an arsenic sequential extraction method for evaluating mobility in sediments. *Environ. Sci. Technol.* **35**, 2778-2784.
- LeBlanc D. R., Garabedian S. P., Hess K. M., Gelhar L. W., Quadri R. D., Stollenwerk K. G., and Wood W. W. (1991) Large-scale natural gradient tracer

test in sand and gravel, Cape Cod, Massachusetts, 1, Experimental design and observed tracer movement. *Water Resour. Res.* **27**(5), 895-910.

- Lederer P. (1998) Toxikologie von Arsen im Trinkwasser. In Arsen im Grund und Trinkwasser Bayerns (ed. U. Kleeberger, H. Frisch, and G. Heinrichs), pp. 73-81. Sven von Loga.
- Lin Z. and Puls R. W. (2000) Adsorption, desorption and oxidation of arsenic affected by clay minerals and aging process. *Environ. Geol.* **39**, 753-759.
- Lovley D. R., Chapelle F. H., and Woodward J. C. (1994) Used of dissolved H<sub>2</sub> concentrations to determine distribution of microbially catalyzed redox reactions in anoxic groundwater. *Environ. Sci. Technol.* **28**, 1205-1210.
- Mallick S. and Rajagopal N. R. (1996) Groundwater development in the arsenicaffected alluvial belt of West Bengal - Some Question. *Curr. Sci. India.* **70**, 956-958.
- Mandal B. K., Chowdhury T. R., Samanta G., Mukherjee D. P., Chanda C. R., Saha K. C., and Chakraborti D. (1996) Impact of safe water for drinking and cooking on five arsenic-affected families for 2 years in West Bengal, India. *Sci. Total Environ.* **218**, 185-201.
- Manning B. A., Fendorf S., Bostick B., and Suarez D. L. (2002) Arsenic(III) oxidation and arsenic(V) adsorption reactions on synthetic birnessite. *Environ. Sci. Technol.* 36, 976-981.
- Manning B. A. and Goldberg S. (1996) Modeling arsenate competitive adsorption on kaolinite, montmorillonite, and illite. *Clays Clay Min.* **44**, 609-623.
- Manning B. A. and Goldberg S. (1997) Adsorption and stability of arsenic(III) at the clay mineral-water interface. *Environ. Sci. Technol.* **21**, 2005-2011.
- Masscheleyn P. H., DeLaune R. D., and Patrick J., W. H. (1991) Effect of redox potential and pH on arsenic speciation and solubility in a contaminated soil. *Environ. Geol.* **25**(8), 1414-1419.

- McArthur J. M., Banerjee D. M., Hudson-Edwards K. A., Mishra R., Purohit R., Ravenscroft P., Cronin A., Howarth R. J., Chatterjee A., Talukder T., Lowry D., Houghton S., and Chadha D. K. (2004) Natural organic matter in sedimentary basins and its relation to arsenic in anoxic ground water: the example of West Bengal and its worldwide implications. *Appl. Geochem.* **19**, 1255-1293.
- McArthur J. M., Ravenscroft P., Safiullah S., and Thirlwall M. F. (2001) Arsenic in groundwater: testing pollution mechanisms for aquifers in Bangladesh. *Water Resour. Res.* **37**, 109-117.
- McCobb T. D., LeBlanc D. R., Walter D. A., Hess K. M., Kent D. B., and Smith R. L. (2003) Phosphorus in a ground-water contaminant plume discharging to Ashumet Pond, Cape Cod, Massachusetts. *1999. U. S. Geological Survey Water Resour. Invest.*, Rept. 02-4306, 69 p.
- McLaren S. J. and Kim N. D. (1995) Evidence for a seasonal fluctuation of arsenic in New Zealand's longest river and the effect of treatment on concentrations in drinking water. *Environ. Pollut.* **90**, 67-73.
- Moore J. N., Walker J. R., and Hayes T. H. (1990) Reaction scheme for the oxidation of As(III) to As(V) by birnessite. *Clay Clay Miner* **38**, 549-555.
- Moore N. J., Ficklin W. H., and Johns C. (1988) Partitioning of Arsenic and Metals in Reducing Sulfidic Sediments. *Environ. Sci. Technol.* **22**, 432-437.
- Myers D. J., Heimbrock M. E., Osteryoung J., and Morrison S. M. (1973) Arsenic oxidation state in the presence of microorganisms : examination by differential pulse polarography. *Environ. lett.* **5**(1), 53-61.
- Nesbitt H. W., Canning G. W., and Bancroft G. M. (1998) XPS study of reductive dissolution of birnessite by H3AsO3, with constraints on reaction mechanism. *Geochim. Cosmochim. Acta* **62**, 2097-2110.
- Nicholson R. V., Gillham R. W., and Reardon E. J. (1988) Pyrite oxidation in carbonate-buffered solution: 1. Experimental kinetics. *Geochim. Cosmochim. Acta* 52, 1077-1085.

- Nickson R., McArthur J., Burgess W., Ahmed M., Ravenscroft P., and Rahman M. (1998) Arsenic poisoning of groundwater in Bangladesh. *Nature* **395**, 338.
- Nickson R., McArthur J. M., Ravenscroft P., Burgess W. G., and Ahmed M. (2000) Mechanism of arsenic release to groundwater, Bangladesh and West Bengal. *Appl. Geochem.* **15**, 403-411.
- Niggemeyer A., Spring S., Stackebrandt E., and Rosenzweig R. F. (2001) Isolation and characterization of a novel As(V)-reducing bacterium: implications for arsenic mobilization and the genus Desulfitobacterium. *Appl. Environ. Microb.* 67, 5568-5580.
- Nimick D. A., Moore J. N., Dalby C. E., and Savka M. W. (1998) The fate of geothermal arsenic in the Madison and Missouri Rivers, Montana and Wyoming. *Water Resour. Res.* 34(11), 3051-3067.
- Nordstrom D. K. (2000) Advances in the hydrogeochemistry and microbiology of acid mine waters. *Int. Geol. Rev.* **42**, 499-515.
- Nordstrom D. K. and Alpers C. N. (1999) Geochemistry of acid mine waste. In *The environmental geochemistry of ore deposits*, Vol. 6A, pp. 133-160. Plumlee, G.S.

Logsdon, M.J.

- O'Reilly S. E., Strawn D. G., and Sparks D. L. (2001a) Residence Time effects on Arsenate Adsorption/Desorption Mechanisms on Goethite. *Soil Sci. Soc. Am. J.*65, 67-77.
- O'Reilly S. E., Strawn D. G., and Sparks D. L. (2001b) Residence Time Effects on Arsenate/Desorption Mechanisms on Goethite. *Soil Sci. Soc. Am. J.* **65**, 67-77.
- Oremland R. S., Dowdle P. R., Hoeft S., Sharp J. O., Schaefer J. K., Miller L. G., Switzer Blum J., Smith R. L., Bloom N. S., and Wallschlaeger D. (2000)
  Bacterial dissimilatory reduction of arsenate and sulfate in meromictic Mono Lake, California. *Geochim. Cosmochim. Acta* 64, 3073-3084.
- Oscarson D. W., Huang P. M., and Liaw W. K. (1980) The oxidation of arsenite by aquatic sediments. *J. Environ Qual* **9**, 700-703.

- Oscarson D. W., Huang P. M., and Liaw W. K. (1981) Role of manganese in the oxidation of arsenite by freshwater lake sediments. *Clay Clay Miner* **29**, 219-225.
- Ottley C. J., Davison W., and Edmunds W. M. (1997) Chemical catalysis of nitrate reduction by iron(II). *Geochim. Cosmochim. Acta* **61**, 1819-1828.
- Parkhurst D. L. and Appelo C. A. J. (1999) PHREEQC. US Geological Survey.
- Parkhurst D. L., Stollenwerk K. G., and Colman J. A. (2003) Reactive-transport simulation of phosphorus in the sewage plume at the Massachusetts Military Reservation, Cape Cod, Massachusetts. U. S. Geological Survey Water Resour. Invest. Rept., 03-4017, 33.
- Postma D., Boesen C., Henning K., and Larsen F. (1991) Nitrate reduction in an unconfined sandy aquifer: water chemistry, reduction processes and geochemical modeling. *Water Res.* **27**, 2027-2045.
- Poulton S. W., Krom M. D., and Raiswell R. (2004) A revised scheme for the reactivity of iron (oxyhydr)oxide minerals towards dissolved sulfide. *Geochim. Cosmochim. Acta* 68, 3703-3715.
- Ravenscroft P., McArthur J. M., and Hoque V. (2001) Geochemical and palaeohydrological controls on pollution of groundwater by arsenic. In *Arsenic Exposure and Health Effects IV* (ed. W. R. Chappell, C. O. Abernathy, and R. Calderon), pp. 53-77. Elsevier Science Ltd., Oxford.
- Reynolds J. G., Naylor D. V., and Fendorf S. E. (1999) Arsenate sorption in phosphate amended soils during flooding and aeration cycles. *Soil Sci. Soc. Am. J.* **63**, 1149-1156.
- Riederer J. (1987) Archäologie und Chemie Einblicke in die Vergangenheit.
- Robinson B. H., Brooks R. R., Outred H. A., and Kirkman J. H. (1995) Mercury and Arsenic in Trout from the Taupo Volcanic Zone and Waikato River, North Island, New Zealand. *Chem. Spec Biovaliab.* **7**(1), 27-32.

- Rochette E. A., Bostick B. C., Li G., and Fendorf S. (2000) Kinetics of Arsenate Reduction by Dissolved Sulfide. *Environ. Sci. Technol.* **34**, 4714-4720.
- Rüde T. R. (1996) Beiträge zur Geochemie des Arsens, Karlsruhe. Karlsruher Geochemische Hefte **10**, 1-206.
- Rüde T. R. (2001) Adsorption of arsenic at Fe-oxides. In *Jahresbericht 2001* (ed. R. Gerke, U. Krell, and J. R. Schneider), pp. 869-870. Hasylab.
- Savoie J. and LeBlanc D. R. (1998) Water-quality data for wells and methods of analyses for samples collected in a plume of sewage-contaminated ground water, Ashumet Valley, Cape Cod, Massachusetts, pp. 97-4269. 1993-94: U.S. Geological Survey Water-Resources Investigations Report.
- Schröter J. (1984) Mikro- und Makrodispersivität poröser Grundwasserleiter. *Meyniana* **36**, 1-34.
- Schumann W. (1985) *Der neue BLV Steine und Mineralienführer*. Verlagsgesellschaft mbH München, Wein, Zürich.
- Scott M. J. and Morgan J. J. (1995) Reactions at the oxide surfaces. 1. Oxidation of As(III) by Synthentic Birnessite. *Environ. Sci. Technol.* **29**(8), 1898-1905.
- Scudlark J. R. and Church T. M. (1988) The atmospheric deposition of arsenic and association with acid precipitation. *Atmos. Environ.* **22**, 937-943.
- Senn D. B. and Hemond H. F. (2002) Nitrate controls on iron and arsenic in an urban lake. *Sci.* **296**, 2373-2375.
- Seyler P. and Martin J. M. (1991) Arsenic and selenium in a pristine river-estuarine system: the Krka, Yugoslavia. *Mar. Chem.* **34**, 137–151.
- Simon G., Huang H., Penner-Hahn J. E., Kesler S. E., and Kao L. S. (1999) Oxidation state of gold and arsenic in gold-bearing arsenian pyrite. *Am Mlineral* **84**, 1071-1079.
- Smedley P. L. and Kinniburgh D. G. (2002) A review of the source, behaviour and distribution of arsenic in natural waters. *Appl. Geochem.* **17**, 517-568.

- Smedley P. L., Zhang M., Zhang G., and Luo Z. (2001) Arsenic and other redoxsensitive elements in groundwater from the Huhhot Basin, Inner Mongolia. *Water-Rock Interaction*, 581–584.
- Smith J. V. S., Jankowski J., and Sammut J. (2003) Vertical distribution of As (III) and As (V) in a coastal sandy aquifer: factors controlling the concentration and speciation of arsenic in the Stuarts Point groundwater system, northern New South Wales Australia. *Appl. Geochem.* **18**, 1479-1496.
- Smith R. L., Garabedian S. P., and Brooks M. H. (1996) Comparison of denitrification activity measurements in ground water using cores and natural gradient tracer tests. *Environ. Sci. Technol.* **30**, 3448-3456.
- Smith R. L., Howes B., and Duff J. H. (1991) Denitrification in nitrate contaminated groundwater: Occurance in steep vertical geochemical gradients. *Geochim. Cosmochim. Acta* 55, 1815-1825.
- Stadler S., Isenbeck-Schröter M., Jann S., Höhn R., Kent D., Davis J., Niedan V., Scholz C., and Tretner A. (in prep.) Reduction and transport behavior under natural groundwater conditions; a Cape Cod study. *Geochim. Cosmochim. Acta*.
- Stadler S., Jann S., Höhn R., Isenbeck-Schröter M., Niedan V., Scholz C., Tretner A., Davis J. A., and Kent D. B. (2001) Tracer tests with As(III) in the oxic and suboxic groundwater zones at the USGS Cape Cod site, Mass., USA. *WRI-10*, 1013-1016.
- Stollenwerk K. G. (1996) Modeling phosphate transport in sewage-contaminated groundwater, Cape Cod, Massachusetts. *Appl. Geochem.* **11**, 317-357.
- Straub R. L., Benz M., Schink B., and Widdel F. (1996) Anaerobic, nitrate dependent microbial oxidation of ferrous iron. *Appl. Environ. Microb.* **62**, 1458-1460.
- Sun X. and Doner H. (1998) Adsorption and oxidation of Arsenite on Goethite. *Soil. Sci.* **163**, 278-287.
- Swartz C. H., Blute N. K., Badruzzaman A. B., Ali M. A., Brabander D., Jay J., Besancon J., Islam S., Hemond H. F., and Harvey C. F. (2004) Mobility of

arsenic in a Bangladesh aquifer: Inferences from geochemical profiles, leaching data, and mineral characterization. *Geochim. Cosmochim. Acta* **68**, 4539-4557.

- Tournassat C., Charlet L., Bosbach D., and Manceau A. (2002) Arsenic(III) Oxidation by Birnessite and Precipitation of Manganese(II) Arsenate. *Environ. Sci. Technol.* 36, 493-500.
- Tretner A. (2002) Sorptions- und Redoxprozesse von Arsen an oxidischen Oberflächen - Experimentelle Untersuchungen, University of Heidelberg.
- Tretner A., Isenbeck-Schröter M., and Rüde T. R. (2005) Oxidation of Arsenic III on iron hydroxide mineral surfaces. *Environ. Sci. Technol.* **submitted**.
- Van Cappellen P. and Wang Y. (1996) Cycling of iron and manganese in surface sediments: A general theory for the coupled transport and reaction of carbon, oxygen, nitrogen, sulfur, iron and manganese. *Amer. J. Sci.* 296, 197-243.
- Vink B. W. (1996) Stability relations of antimony and arsenic compounds in the light of revised and extended Eh-pH diagrams. *Chem. Geol.* **130**, 21-30.
- Wagner F., Berner Z. A., and Stüben D. (2005) Arsenic in groundwater of Bengal Delta Plain: Geochemical evidences for small scale redox zonation in the aquifer. **in press**.
- Wallschläger D., Wilkin R. T., and Ford R. G. (2002) Determination of reduced arsenic-thio species in waters by ion chromatography-inductively-coupled plasma-mass spectrometry (IC-ICP-MS). *Winter Conf Plasma Spectrochem. Scottsdale AZ*.
- Walter D. A., Rea B. A., Stollenwerk K. G., and Savoie J. G. (1996) Geochemical and hydrological controls on phosphorus transport in a sewage contaminated sand and gravel aquifer near Ashumet Pond, Cape Cod, MAssachusetts. U.S. Geological Survey Water-Supply Paper 3463, 89.
- Waltham C. A. and Eick M. J. (2002) Kinetics of Arsenic Adsorption on Goethite in the Presence of Sorbed Silicic Acid. *Soil Sci. Soc. Am. J.* **66**, 818-825.

- Welch A. H. and Stollenwerk K. G. (2003) *Arsenic in Groundwater Geochemistry and Occurence*. Kluwer Academic Publishers.
- Welch A. H., Westjohn D. B., Helsel D. R., and Wanty R. B. (2000) Arsenic in ground water of the United States. *Ground Water* **38**(4), 589-604.
- WHO. (1993) Guidelines for Drinking Water Quality. World Health Organization.
- Wilkin R. T., Wallschläger D., and Ford R. G. (2003) Speciation of arsenic in sulfidic waters. *Geochem. T.* **4**, 1-7.
- Winston H. Y., Harvey C. M., and Harvey C. F. (2003) Arsenic in groundwater in Bangladesh: A geostatistical and epidemiological framework for evaluating health effects and potential remedies. *Water Resour Res* **39**, 1-17.
- Wolthers M., Butler I., Rickard D., and van der Weijden C. (2000) Arsenic Incorporation into Pyrite at low Temperature, Experimental Results. *Goldschmidt*, 1101.
- Xiaohua S. and Doner H. E. (1996) An Investigation of arsenate and arsenite bonding structures on goethite by FTIR. *Soil. Sci.* **161**, 865-872.
- Zapico M. M., Vales S., and Cherry J. A. (1987) A wireline piston core barrel for sampling cohesionless sand and gravel below the water table. *Ground Water Monit. Rev.*, 74-82.
- Zheng Y., Stute M., van Geen A., Gavrieli I., Dhar R., Simpson H. J., Schlosser P., and Ahmed K. M. (2004) Redox control of arsenic mobilization in Bangladesh groundwater. *Appl. Geochem.* **19**, 201-214.
- Zobrist J., Dowdle P. R., Davis J. A., and Oremland R. S. (1998) Microbial arsenate reduction vs. arsenate sorption: Experiments with ferrihydrite suspensions. *Mineral. Mag.* **62A**, 1707-1708.
- Zobrist J., Dowdle P. R., Davis J. A., and Oremland R. S. (2000) Mobilisation of Arsenite by Dissimilatory Reduction of Adsorbed Arsenate. *Environ. Sci. Technol.* **34**(22), 4747-4753.

#### Acknowledgment

This project has been impossible without the financial support from the DFG (Deutsche Forschungsgemeinschaft).

Furthermore I thank all, who were involved by the development and the success of this work, by initiation this work, by the support of the analytical setup, the measurements, the discussions, the support by computer problems, the review of the manuscript, the certificate and much more:

Maria Boni, Jim Davis, Franco Frau, Torsten Hoffmann, Renate Höhn, Margot Isenbeck-Schröter, Rasmus Jakobsen, Steffen Jann, Doug Kent, Nils Kunze, Volker Niedan, Sebastian Pfeil, Silvia Pontis, Stefan Rheinberger, Christian Scholz, Hartwig Schröder, Susanne Stadler, Andreas Tretner, Dirk Wallschläger, Laurence Warr and all I forgot mistakenly.

# 11. Appendix

# 11.1 Cape Cod

name	date	As(III) [µg/I]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2 01	25.6.00 19:00	3.45		2.86	50	0.89	142.11
M2 01	25.6.00 23:00	3.23		3.34	50	0.23	166.77
M2 01	26.6.00 7:00	3.4		2.93	50	0.78	145.72
M2 01	26.6.00 11:00	3.9		3.03	50	1.19	150.31
M2 01	26.6.00 15:00	2.46		2.92	50	0.00	146.17
M2 01	27.6.00 11:00	1.99		2.38	50	0.00	119.15
M2 01	15.7.00 9:00	9.37		17.19	25	1.66	428.09
M2 01	21.7.00 9:00	9.9		17.76	25	1.94	442.06
M2 01	24.7.00 9:00	13.88		18.79	25	5.52	464.23
M0.00		2.4		0.57	50	0.04	05 50
M2-02	24.6.00 15:00	3.4		0.57	50	2.94	25.56
M2-02	24.6.00 19:00	2.1		2.19	50	0.13	109.37
M2-02	24.6.00 23:00	8.52		8.68	50	0.72	433.28
M2-02	25.6.00 3:00	8.86		8.66	50	1.09	431.91
M2-02	25.6.00 7:00	9.13		9.48	50	0.61	473.39
M2-02	25.6.00 11:00	8.89		9.37	50	0.47	468.03
M2-02	25.6.00 15:00	8.99		8.67	50	1.21	432.29
M2-02	25.6.00 19:00	8.44		9.06	50	0.29	452.71
M2-02	25.6.00 23:00	8.71		8.96	50	0.66	447.34
M2-02	26.6.00 7:00	8.69		9.3	50	0.33	464.67
M2-02	26.6.00 11:00	9.3		9.38	50	0.87	468.13
M2-02	26.6.00 15:00	8.88		9.72	50	0.13	485.87
M2-02	27.6.00 11:00	8.06		7.44	50	1.39	370.61
M2-02	3.7.00 9:00	5.79		4.94	100	0.00	497.16
M2-02	15.7.00 9:00	10.71		19.57	25	1.94	487.31
M2-02	21.7.00 9:00	10.33		18.43	25	2.07	458.68
M2-02	24.7.00 9:00	14.06		19.16	25	5.54	473.46
M2-02	24.7.00 19:00	14.59	_	19.02	25	6.14	469.36
M2-02	25.7.00 19:00	4.11	5	10.49	50	11.31	513.19
M2-02	26.7.00 9:00	3.93	5	10.18	50	10.68	498.32
M2-02	26.7.00 19:00	3.97	5	10.35	50	10.73	506.77
M2-02	27.7.00 9:00	2.99	5	7.58	50	8.28	370.72
M2-02	27.7.00 19:00	2.84	5	6.42	50	8.58	312.42
M2-02	28.7.00 9:00	2.82	5	5.39	50	9.42	260.08
M2-02	28.7.00 19:00	2.94	5	5.14	50	10.26	246.74
M2-02	29.7.00 9:00	3.11	5	5.15	50	11.12	246.38
M2-02	29.7.00 19:00	3.32	5	4.91	50	12.40	233.10
M2-02	30.7.00 9:00	1.22	5	2.79	50	3.65	135.85
M2-02	30.7.00 19:00	3.32	5	4.51	50	12.77	212.73
M2-02	31.7.00 9:00	4.33	5	4.68	50	17.76	216.24
M2-02	31.7.00 19:00	3.69	5	4.55	50	14.62	212.88
M2-02	1.8.00 9:00	4.23	5	4.53	50	17.39	209.11
M2-02	1.8.00 19:00	2.39	5	4.07	50	8.44	195.06
M2-02	2.8.00 9:00	2.2	5	4.18	50	7.37	201.63

## Tab.: A1 arsenic concentration from M2

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2-02	2.8.00 19:00	2.16	5	4.43	50	6.94	214.56
M2-02	3.8.00 9:00	2.59	5	4.55	50	9.02	218.48
M2-02	3.8.00 19:00	3.26	5	4.51	50	12.47	213.03
M2-02	4.8.00 9:00	3.94	5	4.57	50	15.87	212.63
M2-02	4.8.00 19:00	4.43	5	4.46	50	18.47	204.53
M2-02	5.8.00 9:00	2.24	5	4.12	50	7.63	198.37
M2-02	5.8.00 19:00	4.75	5	4.25	50	20.29	192.21
M2-02	6.8.00 9:00	3.77	5	4.13	50	15.41	191.09
M2-02	6.8.00 19:00	4.04	5	4.03	50	16.88	184.62
M2-02	7.8.00 9:00	11.43	5	4.82	50	53.78	187.22
M2-02	7.8.00 19:00	6.8	5	4.7	50	30.32	204.68
M2-02	8.8.00 9:00	7.43	5	4.57	50	33.64	194.86
M2-02	8.8.00 19:00	3.83	10	3.74	50	35.57	151.43
M2-02	9.8.00 9:00	5.29	10	3.72	50	50.46	135.54
M2-02	9.8.00 19:00	4.42	10	3.5	50	41.80	133.20
M2-02	11.8.00 9:00	4.17	10	3.75	50	39.03	148.47
M2-02	11.8.00 19:00	3.08	10	3.27	50	28.37	135.13
M2-02	12.8.00 9:00	8.24	10	3.59	50	80.62	98.88
M2-02	12.8.00 19:00	6.54	10	3.39	50	63.49	106.01
M2-02	13.8.00 9:00	5.91	10	3.63	50	56.86	124.64
M2-02	13.8.00 19:00	1.64	10	3	50	13.95	136.05
M2-02	14.8.00 9:00	2.9	10	2.67	50	27.08	106.42
M2-02	14.8.00 19:00	2.9	10	3.35	50	26.46	141.04
M2-02	15.8.00 9:00	6.36	10	3.21	50	61.82	98.68
M2-02	15.8.00 19:00	2.83	10	3.23	50	25.86	135.64
M2-02	16.8.00 9:00	4.58	10	3.24	50	43.67	118.33
M2-02	16.8.00 19:00	3.45	10	2.84	50	32.53	109.47
M2-02	17.8.00 9:00	1.36	10	2.48	50	11.58	112.42
M2-02	17.8.00 19:00	4.27	10	2.93	50	40.80	105.70
M2-02	18.8.00 9:00	4.24	10	2.9	50	40.52	104.48
M2-02	18.8.00 19:00	5.09	10	2.71	50	49.35	86.15
M2-02	19.8.00 9:00	3.23	10	2.7	50	30.42	104.58
M2-02	21.8.00 9:00	2.06	10	2.5	50	18.69	106.31
M2-02	22.8.00 9:00	4.25	10	4.68	25	41.13	75.87
M2-02	23.8.00 9:00	4.01	10	4.55	25	38.75	75.00
M2-02	24.8.00 9:00	2.14	10	4	25	19.96	80.04
M2-02	25.8.00 9:00	2.3	10	4.11	25	21.54	81.21
M2-02	26.8.00 9:00	1.05	10	4.23	25	8.75	97.00
M2-02	27.8.00 9:00	0.28	10	2.3	50	0.74	114.26
M2-02	28.8.00 9:00	0.63	5	2.08	50	1.30	102.70
M2-02	29.8.00 9:00	3.62	10	2.08	50	34.96	69.04
M2-02	30.8.00 9:00	3.86	10	2.09	50	37.39	67.11
M2-02	31.8.00 9:00	3.95	10	1.95	50	38.44	59.06
M2-02	1.9.00 9:00	3.98	10	1.96	50	38.73	59.27
M2-02	2.9.00 9:00	5.08	10	1.96	50	49.93	48.07
M2-02	3.9.00 9:00	2.84	10	1.75	50	27.32	60.18
M2-02	4.9.00 9:00	3.14	10	1.84	50	30.29	61.71
M2-02	5.9.00 9:00	3.57	10	2.07	50	34.46	69.04
M2-02	6.9.00 9:00	2.11	10	1.86	50	19.78	73.22
M2-02	7.9.00 9:00	2.75	10	1.58	50	26.56	52.44
M2-02	8.9.00 9:00	2.98	10	1.51	50	28.96	46.54
M2-02	9.9.00 9:00	2.89	10	1.44	50	28.11	43.89
M2-02	10.9.00 9:00	2.09	10	1.5	50	19.91	55.09

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2-02	11.9.00 9:00	2.56	10	1.25	50	24.92	37.58
M2-02	12.9.00 9:00	2.69	10	1.18	50	26.31	32.69
M2-02	13.9.00 9:00	2.77	10	1.19	50	27.12	32.38
M2-02	14.9.00 9:00	2.86	10	1.18	50	28.04	30.96
M2-02	21.9.00 9:00	3.48	10	1.14	50	34.39	22.61
M2-02	30.9.00 9:00	2.62	10	1.11	50	25.66	29.84
M2-02	7.10.00 9:00	1.97	10	0.97	50	19.17	29.33
M2-02	14.10.00 9:00	2.21	10	5.39	10	21.52	32.38
M2-02	21.10.00 9:00	4.42	5	5.39	10	21.52	32.38
M2-02	28.10.00 9:00	4.8	5	4.05	10	23.70	16.80
M2-02	8.11.00 9:00	4.3	5	3.73	10	21.21	16.09
M2-03	25.7.00 19:00	3.85	5	9.76	50	9.68	478.32
M2-03	26.7.00 9:00	3.65	5	10.16	50	8.26	499.74
M2-03	26.7.00 19:00	3.94	5	11.19	50	8.68	550.82
M2-03	27 7 00 9.00	2 77	5	12 42	50	1 46	619 54
M2-03	27 7 00 19:00	2.56	5	87	50	4 18	430.82
M2-03	28 7 00 9.00	2.60	5	5 79	50	7 41	282.09
M2-03	28 7 00 19:00	2.01	5	5.6	50	6.33	273 67
M2-03	29 7 00 9.00	2.53	5	5 39	50	7 41	262.09
M2-03	29.7.00 19:00	2.00	5	4 95	50	6.43	241 07
M2-03	30 7 00 9.00	1.08	5	4.00	50	0.45	237 35
M2-03	30 7 00 19:00	1.00	5	4.76	50	4 28	233 72
M2-03	31 7 00 9.00	1.75	5	4.70	50	4.20	229.29
M2-03	31 7 00 19:00	1.70	5	4.86	50	4.43	238 57
M2-03	1 8 00 9.00	1.04	5	4.65	50	4.45	228.06
M2-03	1 8 00 19:00	1.58	5	4 37	50	3.60	214 90
M2-03	28009.00	1.60	5	4.34	50	3 79	213.00
M2-03	280019.00	1.67	5	4 46	50	3 97	219.03
M2-03	3800900	1.07	5	4 26	50	4 48	208.52
M2-03	38001900	1.78	5	4 45	50	4 54	217 96
M2-03	4 8 00 9.00	1.92	5	4 69	50	5 01	229.49
M2-03	4.8.00 19:00	1.85	5	4.99	50	4.35	245.15
M2-03	58009.00	1.55	5	4 88	50	2.93	241.07
M2-03	5.8.00 19:00	2.62	5	4.97	50	8.30	240.20
M2-03	6.8.00 9:00	2.07	5	4.97	50	5.49	243.01
M2-03	6.8.00 19:00	1.57	5	5.08	50	2.83	251.17
M2-03	7.8.00 9:00	12.97	5	5.75	50	60.31	227.19
M2-03	7.8.00 19:00	14.96	5	5.45	50	70.76	201.74
M2-03	8.8.00 9:00	15.79	5	5.61	50	74.84	205.66
M2-03	8.8.00 19:00	8.91	10	5.32	50	85.49	180.51
M2-03	9.8.00 9:00	7.4	10	4.86	50	70.55	172.45
M2-03	9.8.00 19:00	8.07	10	5.15	50	77.09	180.41
M2-03	11.8.00 9:00	8.95	10	4.88	50	86.35	157.65
M2-03	11.8.00 19:00	9.66	10	4.86	50	93.61	149.39
M2-03	12.8.00 9:00	9.19	10	4.7	50	88.98	146.02
M2-03	12.8.00 19:00	9.48	10	4.82	50	91.82	149.18
M2-03	13.8.00 9:00	3.62	10	4.88	50	31.96	212.04
M2-03	13.8.00 19:00	7.93	10	3.97	50	76.87	121.63
M2-03	14.8.00 9:00	6.83	10	4.39	50	65.21	154.29
M2-03	14.8.00 19:00	8.42	10	4.4	50	81.43	138.57
M2-03	15.8.00 9:00	3.65	10	4.24	50	32.92	179.08

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2-03	15.8.00 19:00	8.73	10	3.81	50	85.19	105.31
M2-03	16.8.00 9:00	7.13	10	4.24	50	68.43	143.57
M2-03	16.8.00 19:00	1.9	10	3.95	50	15.36	182.14
M2-03	17.8.00 9:00	6.19	10	3.58	50	59.51	119.49
M2-03	17.8.00 19:00	7	10	3.76	50	67.59	120.41
M2-03	18.8.00 9:00	4.98	10	3.78	50	46.96	142.04
M2-03	18.8.00 19:00	4.74	10	3.62	50	44.67	136.33
M2-03	19.8.00 9:00	7.94	10	3.63	50	77.32	104.18
M2-03	20.8.00 9:00	9.42	10	3.41	50	92.64	77.86
M2-03	21.8.00 9:00	9	10	3.07	50	88.70	64.80
M2-03	22.8.00 9:00	10.04	10	6.11	25	99.33	53.42
M2-03	23.8.00 9:00	10.44	10	5.89	25	103.52	43.73
M2-03	24.8.00 9:00	8.83	10	5.46	25	87.32	49.18
M2-03	25.8.00 9:00	7.39	10	5.47	25	72.62	64.13
M2-03	29.8.00 9:00	9.03	10	2.71	50	89.38	46.12
M2-03	30.8.00 9:00	7.21	10	2.56	50	70.96	57.04
M2-03	31.8.00 9:00	6.76	10	2.76	50	66.16	71.84
M2-03	1.9.00 9:00	6.43	10	2.53	50	63.03	63.47
M2-03	2.9.00 9:00	6.62	10	2.38	50	65.12	53.88
M2-03	3.9.00 9:00	7	10	2.51	50	68.87	56.63
M2-03	4.9.00 9:00	7.76	10	2.54	50	76.59	50.41
M2-03	5.9.00 9:00	6.94	10	2.42	50	68.35	52.65
M2-03	6.9.00 9:00	6.21	10	2.12	50	61.20	44.80
M2-03	7.9.00 9:00	7.03	10	2.02	50	69.67	31.33
M2-03	8.9.00 9:00	6.35	10	2.02	50	62.73	38.27
M2-03	9.9.00 9:00	6.64	10	2.01	50	65.70	34.80
M2-03	10.9.00 9:00	5.91	10	1.82	50	58.45	32.55
M2-03	11.9.00 9:00	5.9	10	1.86	50	58.31	34.69
M2-03	12.9.00 9:00	6.5	10	1.96	50	64.33	33.67
M2-03	13.9.00 9:00	6.08	10	1.78	50	60.22	28.78
M2-03	14.9.00 9:00	5.49	10	1.55	50	54.44	23.06
M2-03	21.9.00 9:00	5.27	10	1.68	50	52.06	31.94
M2-03	30.9.00 9:00	4.21	10	1.4	50	41.53	28.47
M2-03	7.10.00 9:00	4.1	10	1.29	50	40.52	23.98
M2-03	14.10.00 9:00	3.56	10	7.49	10	34.80	40.10
M2-03	21.10.00 9:00	7.52	5	6.86	10	36.97	31.63
M2-03	28.10.00 9:00	2.59	5	1.03	10	13.00	-2.70
M2-03	8.11.00 9:00	7.22	5	4.98	10	35.82	13.98
						-	
M2-04	24.6.00 15:00	4.4		4.96	50	0.00	248.07
M2-04	24.6.00 19:00	4.33		4.59	50	0.20	229.30
M2-04	24.6.00 23:00	9.76		8.89	50	1.79	442.71
M2-04	25.6.00 3:00	9.92		9.09	50	1.77	452.73
M2-04	25.6.00 7:00	9.5		9.02	50	1.41	449.59
M2-04	25.6.00 11:00	8.89		9.25	50	0.58	461.92
M2-04	25.6.00 15:00	8.69		9.16	50	0.45	457.55
M2-04	25.6.00 19:00	8.74		8.79	50	0.84	438.66
M2-04	25.6.00 23:00	8.51		9.16	50	0.27	457.73
M2-04	26.6.00 7:00	9.75		9.31	50	1.40	464.10
M2-04	26.6.00 11:00	9.56		9.4	50	1.12	468.88
M2-04	26.6.00 15:00	8.66		10.54	50	0.00	527.84
M2-04	3.7.00 9:00	5.76		4.7	100	0.00	472.75
name	date	As(III) [µg/I]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
-------	---------------	-------------------	----------	-------------------	----------	--------------	------------
M2-04	15.7.00 9:00	10.48		11.55	50	0.09	577.41
M2-04	21.7.00 9:00	9.7		19.08	25	1.13	475.87
M2-04	24.7.00 9:00	14.56		18.94	25	6.15	467.35
M2-04	24.7.00 19:00	14.19		18.28	25	6.07	450.93
M2-04	25.7.00 9:00	11.05		18.06	25	2.98	448.52
M2-04	25.7.00 19:00	2.59	5	9.96	50	4.06	493.94
M2-04	26.7.00 9:00	2.92	5	11.59	50	4.25	575.25
M2-04	26.7.00 19:00	2.85	5	10.69	50	4.71	529.79
M2-04	27.7.00 9:00	1.24	5	10.11	50	0.00	508.45
M2-04	27.7.00 19:00	2.6	5	5.1	50	8.56	246.44
M2-04	28.7.00 9:00	1.37	5	5.24	50	2.17	259.83
M2-04	28.7.00 19:00	1.22	5	4.44	50	2.14	219.86
M2-04	29.7.00 9:00	1.44	5	4.43	50	3.27	218.23
M2-04	29.7.00 19:00	1.9	5	4.72	50	5.35	230.65
M2-04	30.7.00 9:00	1.26	5	3.39	50	3.31	166.19
M2-04	30.7.00 19:00	1.54	5	4.9	50	3.35	241.65
M2-04	31.7.00 9:00	3.81	5	4.89	50	14.92	229.58
M2-04	31.7.00 19:00	2.46	5	4.83	50	8.10	233.40
M2-04	1.8.00 9:00	1.34	5	4.94	50	2.30	244.70
M2-04	1.8.00 19:00	1.31	5	5.44	50	1.68	270.32
M2-04	2.8.00 9:00	1.46	5	5.2	50	2.67	257.33
M2-04	2.8.00 19:00	1.48	5	5.37	50	2.61	265.89
M2-04	3.8.00 9:00	1.57	5	5.32	50	3.12	262.88
M2-04	3.8.00 19:00	1.42	5	5.24	50	2.43	259.57
M2-04	4.8.00 9:00	1.64	5	5.47	50	3.34	270.16
M2-04	4.8.00 19:00	1.42	5	5.28	50	2.39	261.61
M2-04	5.8.00 9:00	2.86	5	5.12	50	9.87	246.13
M2-04	5.8.00 19:00	3.19	5	5.31	50	11.38	254.12
M2-04	6.8.00 9:00	10.22	5	4.86	50	47.58	195.42
M2-04	6.8.00 19:00	5.71	5	4.77	50	24.70	213.80
M2-04	7.8.00 9:00	19.4	5	5.27	50	93.95	169.55
M2-04	7.8.00 19:00	10.33	5	5.1	50	47.92	207.08
M2-04	8.8.00 19:00	8.68	10	4.23	50	84.51	126.99
M2-04	9.8.00 9:00	14.83	10	4.16	50	147.20	60.80
M2-04	9.8.00 19:00	10.15	10	3.92	50	99.77	96.23
M2-04	11.8.00 9:00	2.38	10	3.46	50	21.07	151.93
M2-04	11.8.00 19:00	8.21	10	3.55	50	80.35	97.15
M2-04	12.8.00 9:00	8.61	10	3.52	50	84.45	91.55
M2-04	12.8.00 19:00	6.71	10	3.16	50	65.43	92.57
M2-04	13.8.00 9:00	5.73	10	3.27	50	55.35	108.15
M2-04	13.8.00 19:00	2.4	10	3.12	50	21.58	134.42
M2-04	14.8.00 9:00	5.38	10	2.97	50	52.06	96.44
M2-04	14.8.00 19:00	5.09	10	3.02	50	49.06	101.94
M2-04	15.8.00 9:00	8.18	10	2.94	50	80.60	66.40
M2-04	15.8.00 19:00	9.44	10	2.85	50	93.52	48.98
M2-04	16.8.00 9:00	7.85	10	2.93	50	77.25	69.25
M2-04	16.8.00 19:00	6.52	10	2.89	50	63.75	80.75
M2-04	17.8.00 9:00	1.67	10	2.95	50	14.30	133.20
M2-04	17.8.00 19:00	3.99	10	2.43	50	38.40	83.10
M2-04	18.8.00 9:00	4.55	10	2.6	50	43.95	86.05
M2-04	18.8.00 19:00	5.68	10	2.59	50	55.47	74.03
M2-04	19.8.00 9:00	11.39	10	2.64	50	113.57	18.43
M2-04	20.8.00 9:00	10.67	10	5.75	50	103.39	184.11

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2-04	20.8.00 19:00	6.87	10	2.54	50	67.63	59.37
M2-04	21.8.00 9:00	6.07	10	2.56	50	59.47	68.53
M2-04	22.8.00 9:00	4.78	10	4.35	25	46.68	62.07
M2-04	23.8.00 9:00	3.82	10	5.8	25	36.24	108.76
M2-04	24.8.00 9:00	6.13	10	3.94	25	60.62	37.88
M2-04	25.8.00 9:00	3.84	10	3.85	25	37.34	58.91
M2-04	26.8.00 9:00	1.62	10	3.88	25	14.72	82.28
M2-04	29.8.00 9:00	5.8	10	1.9	50	57.32	37.68
M2-04	30.8.00 9:00	5.48	10	1.77	50	54.18	34.32
M2-04	31.8.00 9:00	4.85	10	1.82	50	47.72	43.28
M2-04	1.9.00 9:00	4.77	10	1.78	50	46.94	42.06
M2-04	2.9.00 9:00	5.28	10	1.78	50	52.14	36.86
M2-04	3.9.00 9:00	4.48	10	1.58	50	44.17	34.83
M2-04	4.9.00 9:00	5.15	10	1.62	50	50.96	30.04
M2-04	5.9.00 9:00	4.05	10	1.65	50	39.73	42.77
M2-04	6.9.00 9:00	2.93	10	1.3	50	28.65	36.35
M2-04	7.9.00 9:00	3.95	10	1.2	50	39.12	20.88
M2-04	8.9.00 9:00	4.83	10	1.19	50	48.09	11.41
M2-04	9.9.00 9:00	3.68	10	1.09	50	36.48	18.02
M2-04	10.9.00 9:00	4.27	10	1.07	50	42.50	11.00
M2-04	11 9 00 9.00	3.5	10	1 14	50	34 60	22 40
M2-04	12 9 00 9:00	3.83	10	1 14	50	37.96	19.04
M2-04	13 9 00 9 00	3.94	10	0.88	50	39.32	4 68
M2-04	14 9 00 9:00	3 65	10	1.06	50	36.20	16 80
M2-04	21 9 00 9:00	3 19	10	0.92	50	31.64	14.36
M2-04	30.9.00.9.00	2.24	10	1.02	50	21.87	29.63
M2-04	7 10 00 9.00	2.24	10	0.92	50	21.07	23.03
M2-04	1/ 10.00 0.00	2.41	10	5.24	10	23.70	22.50
M2-04	21 10 00 9:00	2.43 4.43	5	<u> </u>	10	23.55	25.31
M2-04	28.10.00.9:00	4.43 // 13	5	3 73	10	21.03	16.96
M2-04	8 11 00 9:00	5.52	5	3.55	10	20.34	8.04
1012-04	0.11.00 9.00	5.52	5	0.00	10	27.40	0.04
M0.05	24 0 00 45:00	E 4E		0.00	0	5.00	44.00
M2 05	24.6.00 15:00	5.45 0.70		9.63	2	5.20	14.06
M2 05	24.6.00 19:00	0.73		11.04	2	0.34	21.74
IVI2 05	24.6.00 23:00	1.54		1.75	50	0.00	87.54
IVIZ 05	25.6.00 3.00	1.44		1.43	50	0.16	71.34
IVI2 05	25.6.00 7:00	1.53		1.73	50	0.00	80.33
IVIZ 05	25.6.00 11.00	2.00		2.75	50	0.19	137.31
IVI2 05	25.6.00 15:00	2.80		2.98	50	0.18	148.82
IVI2 05	25.6.00 19:00	3.48		3.69	50	0.16	184.34
M2 05	25.6.00 23:00	4.14		4.92	50	0.00	246.29
M2 05	26.6.00 7:00	5.37		6.1	50	0.00	305.12
M2 05	26.6.00 11:00	5.78		6.11	50	0.29	305.21
M2 05	26.6.00 15:00	5.92		6.14	50	0.40	306.60
IVI2 05	3.7.00 9:00	5.87		3.92	100	0.00	393.21
IVI2 05	15.7.00 9:00	8.76		9.32	50	0.38	465.62
IVI2 05	21.7.00 9:00	9.9		1/.//	25	1.94	442.31
M2 05	24.7.00 9:00	14.28		18.08	25	6.26	445./4
IVI2 05	24.7.00 19:00	13.41		17.49	25	5.64	431.61
M2 05	25.7.00 9:00	9.7		16.23	25	2.44	403.31
M2 05	25.7.00 19:00	2.14	5	8.95	50	2.69	444.81
M2 05	26.7.00 9:00	2.24	5	8.52	50	3.60	422.40

name	date	As(III) [µg/I]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2 05	26.7.00 19:00	2	5	8.04	50	2.81	399.19
M2 05	27.7.00 9:00	1.84	5	5.19	50	4.61	254.89
M2 05	27.7.00 19:00	1.94	5	4.75	50	5.52	231.98
M2 05	28.7.00 9:00	2.13	5	4.24	50	6.96	205.04
M2 05	28.7.00 19:00	2.02	5	3.73	50	6.87	179.63
M2 05	29.7.00 9:00	2.24	5	3.76	50	7.96	180.04
M2 05	29.7.00 19:00	1.97	5	3.72	50	6.62	179.38
M2 05	30.7.00 9:00	1.07	5	3.06	50	2.64	150.36
M2 05	30.7.00 19:00	2.51	5	3.54	50	9.54	167.46
M2 05	31.7.00 9:00	1.38	5	3.62	50	3.71	177.29
M2 05	31.7.00 19:00	1.81	5	3.72	50	5.81	180.19
M2 05	1.8.00 9:00	1.95	5	5.6	50	4.80	275.20
M2 05	1.8.00 19:00	1.67	5	3.53	50	5.27	171.23
M2 05	2.8.00 9:00	1.74	5	3.44	50	5.71	166.29
M2 05	2.8.00 19:00	2.26	5	3.49	50	8.31	166.19
M2 05	3.8.00 9:00	3.18	5	3.72	50	12.78	173.22
M2 05	3.8.00 19:00	3.86	5	3.65	50	16.31	166.19
M2 05	4.8.00 9:00	4.75	5	3.58	50	20.90	158.10
M2 05	4.8.00 19:00	4.6	5	3.59	50	20.13	159.37
M2 05	5.8.00 9:00	6.02	5	3.76	50	27.21	160.79
M2 05	5.8.00 19:00	5.76	5	3.84	50	25.81	166.19
M2 05	6.8.00 9:00	5.84	5	3.75	50	26.30	161.20
M2 05	6.8.00 19:00	4.83	5	3.48	50	21.40	152.60
M2 05	7.8.00 9:00	15.83	5	4.14	50	76.81	130.19
M2 05	7.8.00 19:00	10.61	5	3.97	50	50.38	148.12
M2 05	8.8.00 9:00	11.97	5	3.96	50	57.32	140.68
M2 05	8.8.00 19:00	6.76	10	3.54	50	65.59	111.41
M2 05	9.8.00 9:00	7.98	10	3.4	50	78.15	91.85
M2 05	9.8.00 19:00	6.45	10	3.52	50	62.46	113.54
M2 05	11.8.00 9:00	6.52	10	3.3	50	63.37	101.63
M2 05	11.8.00 19:00	4.58	10	3.28	50	43.63	120.37
M2 05	12.8.00 9:00	6.87	10	3.2	50	67.03	92.97
M2 05	12.8.00 19:00	5.77	10	3.05	50	55.96	96.54
M2 05	13.8.00 9:00	8.45	10	3.06	50	83.24	69.76
M2 05	13.8.00 19:00	2.43	10	2.73	50	22.24	114.26
M2 05	14.8.00 9:00	5.55	10	2.89	50	53.87	90.63
M2 05	14.8.00 19:00	6.21	10	2.87	50	60.61	82.89
M2 05	15.8.00 9:00	9.63	10	2.78	50	95.52	43.48
M2 05	15.8.00 19:00	5.94	10	2.65	50	58.06	74.44
M2 05	16.8.00 9:00	7.11	10	2.68	50	69.95	64.05
M2 05	16.8.00 19:00	6.03	10	2.61	50	59.01	71.49
M2 05	17.8.00 9:00	1.47	10	2.42	50	12.75	108.25
M2 05	17.8.00 19:00	4.51	10	2.37	50	43.75	74.75
M2 05	18.8.00 9:00	4.65	10	2.43	50	45.12	76.38
M2 05	18.8.00 19:00	5.48	10	2.27	50	53.72	59.78
M2 05	19.8.00 9:00	7.01	10	2.44	50	69.15	52.85
M2 05	20.8.00 9:00	8.04	10	2.34	50	79.73	37.27
M2 05	21.8.00 9:00	3.89	10	2.25	50	37.55	74.95
M2 05	22.8.00 9:00	5.43	10	3.56	25	53.66	35.34
M2 05	23.8.00 9:00	6.67	10	3.45	25	66.34	19.91
M2 05	24.8.00 9:00	6.34	10	3.31	25	63.04	19.71
M2 05	25.8.00 9:00	4.27	10	3.15	25	42.04	36.71
M2 05	26.8.00 9:00	5.29	10	3.12	25	52.44	25.56

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2 05	29.8.00 9:00	4.66	10	1.88	50	45.73	48.27
M2 05	30.8.00 9:00	5.16	10	1.61	50	51.07	29.43
M2 05	31.8.00 9:00	4.42	10	1.39	50	43.74	25.76
M2 05	1.9.00 9:00	4.67	10	1.35	50	46.32	21.18
M2 05	2.9.00 9:00	4.77	10	1.28	50	47.40	16.60
M2 05	3.9.00 9:00	4.62	10	1.43	50	45.74	25.76
M2 05	4.9.00 9:00	4.36	10	1.26	50	43.24	19.76
M2 05	5.9.00 9:00	4.1	10	1.39	50	40.48	29.02
M2 05	6.9.00 9:00	2.75	10	1.17	50	26.93	31.57
M2 05	7.9.00 9:00	3.71	10	0.95	50	36.91	10.59
M2 05	8.9.00 9:00	3.56	10	1.03	50	35.31	16.19
M2 05	9.9.00 9:00	3.08	10	0.8	50	30.63	9.37
M2 05	10.9.00 9:00	3.67	10	0.95	50	36.50	11.00
M2 05	11.9.00 9:00	4.24	10	0.98	50	42.28	6.72
M2 05	12.9.00 9:00	3.48	10	0.96	50	34.56	13.44
M2 05	13.9.00 9:00	3.7	10	0.87	50	36.88	6.62
M2 05	14.9.00 9:00	3.07	10	0.85	50	30.48	12.02
M2 05	21.9.00 9:00	2.83	10	0.81	50	28.08	12.42
M2 05	30.9.00 9:00	2.54	10	0.83	50	25.10	16.40
M2 05	7.10.00 9:00	1.84	10	0.7	50	18.10	16.90
M2 05	14.10.00 9:00	5.14	5	4.51	10	25.34	19.76
M2 05	21.10.00 9:00	4.38	5	4.18	10	21.54	20.26
M2 05	28.10.00 9:00	4.55	5	3.16	10	22.59	9.01
M2 05	8.11.00 9:00	4.55	5	3.09	10	22.60	8.30
M2.06	25 6 00 10:00	4.46		6 72	2	4 20	0.14
M2 06	25.0.00 19.00	4.40		0.72	<u> </u>	4.30	9.14
M2 06	25.0.00 23.00	0.00		1.17	50	0.00	70.60
M2 06	26.6.00 7.00	0.05		1.41	50	0.00	70.00
M2.06	20.0.00 11.00	0.95		12.16	50	0.00	00.20
M2 06	20.0.00 15.00	1.11		12.10	5	0.00	62.24
M2 06	27.0.00 11.00	1.00		7 79	10	0.00	02.34
M2 06	15 7 00 9:00	2.02		7.70	10	0.22	72.66
M2 06	21 7 00 9:00	2.97		9.50	10	0.75	75.00 95.15
M2 06	1 8 00 9:00	2.20		6.86	10	0.75	50.60
M2 06	1.8.00 9.00	6.88		6.53	10	5.00	59.00
M2 06	2 8 00 9:00	5.42		5.87	10	3.01	54 26
M2 06	2.8.00 9.00	0.72		5.8	10	9.93	J4.20 /0.17
M2 06	2.0.00 19.00	9.72		5.3	10	6.03	49.17
M2 06	3.8.00 10.00	11.06		5.04	10	11.26	40.90
M2 06	4 8 00 9.00	11.50		1.68	10	11.20	35.14
M2 06	4.8.00 9.00	13.12		4.00	10	12.50	20.51
M2 06	5 8 00 9.00	12.12		5/3	10	11.59	12 74
M2 06	5 8 00 10:00	12.55		3.45	10	12.11	26.00
M2.00	6 8 00 0.00	10 12		3.51	10	9.65	20.33
M2 06	6 8 00 10.00	12 28		3.45	10	11 07	23.33
M2 06	7 8 00 0.00	3 50	5	0.76	50	17.58	22.33
M2 06	7 8 00 10.00	3.55	5	0.70	50	18.87	20.42
M2 06	8 8 00 0.00	2.05	5	0.0	50	13.02	20.52
M2 06	8 8 00 10.00	2.07	10	29.25		10.90	Q 12
M2 06	9 8 00 9.00	2.00	10	29.20		22.07	7 00
M2 06	98001900	1 91	10	26.87		18.96	7.00
	0.0.00 10.00			20.07		10.00	1.01

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2 06	11.8.00 9:00	1.51	10	23.63		14.94	8.69
M2 06	11.8.00 19:00	1.61	10	23.56		15.96	7.60
M2 06	12.8.00 9:00	1.57	10	22.76		15.57	7.19
M2 06	12.8.00 19:00	1.72	10	22.92		17.10	5.82
M2 06	13.8.00 9:00	1.52	10	22.73		15.06	7.67
M2 06	13.8.00 19:00	1.26	10	23.08		12.41	10.67
M2 06	14.8.00 9:00	1.28	10	22		12.63	9.37
M2 06	14.8.00 19:00	1.68	10	21.91		16.71	5.20
M2 06	15.8.00 9:00	1.93	10	21.4		19.26	2.14
M2 06	15.8.00 19:00	1.62	10	20.52		16.12	4.40
M2 06	16.8.00 9:00	1.88	10	20.71		18.76	1.95
M2 06	16.8.00 19:00	1.74	10	20.37		17.35	3.02
M2 06	17.8.00 9:00	0.93	10	21.55		9.08	12.47
M2 06	17.8.00 19:00	1.81	10	21.56		18.04	3.52
M2 06	18.8.00 9:00	1.72	10	19.38		17.16	2.22
M2 06	18.8.00 19:00	1.51	10	18.91		15.03	3.88
M2 06	19.8.00 9:00	1.41	10	19.47		14.00	5.47
M2 06	20.8.00 9:00	1.69	10	19.42		16.85	2.57
M2 06	21.8.00 9:00	10.31		2.01	10	10.13	9.97
M2 06	22.8.00 9:00	2.92	5	3.2	25	13.40	66.60
M2 06	23.8.00 9:00	2.97	5	3.08	25	13.71	63.29
M2 06	24.8.00 9:00	3.53	5	3.42	25	16.41	69.09
M2 06	25.8.00 9:00	3.07	5	3.12	25	14.20	63.80
M2 06	26.8.00 9:00	12.42	5	3.04	25	61.84	14.16
M2 06	27.8.00 9:00	0.11	10	16.84		0.81	16.03
M2 06	28.8.00 9:00	0.07	5	17.63		0.03	17.60
M2 06	29.8.00 9:00	2.98	5	15.15		14.90	0.25
M2 06	30.8.00 9:00	2.92	5	15.35		14.59	0.76
M2 06	31.8.00 9:00	2.94	5	15.03		14.69	0.34
M2 06	1.9.00 9:00	2.91	5	15.02		14.54	0.48
M2 06	2.9.00 9:00	2.94	5	15.44		14.69	0.75
M2 06	3.9.00 9:00	2.71	5	15.64		13.51	2.13
M2 06	4.9.00 9:00	2.73	5	1.82	10	13.57	4.63
M2 06	5.9.00 9:00	1.26	10	1.8	10	12.50	5.50
M2 06	6.9.00 9:00	0.8	10	16.36		7.85	8.51
M2 06	7.9.00 9:00	1.14	10	15.82		11.32	4.50
M2 06	8.9.00 9:00	1.22	10	15.7		12.14	3.56
M2 06	9.9.00 9:00	0.94	10	15.6		9.29	6.31
M2 06	10.9.00 9:00	0.99	10	15.36		9.80	5.56
M2 06	11.9.00 9:00	1.25	10	15.93		12.44	3.49
M2 06	12.9.00 9:00	1.22	10	14.52		12.16	2.36
M2 06	13.9.00 9:00	1.25	10	14.89		12.46	2.43
M2 06	14.9.00 9:00	1.17	10	14.43		11.65	2.78
M2 06	21.9.00 9:00	1.25	10	15.22		12.45	2.77
M2 06	30.9.00 9:00	1.19	10	0.28	50	11.86	2.14
M2 06	7.10.00 9:00	13.83		15.82		13.79	2.03
M2 06	14.10.00 9:00	2.55	5	1.8	10	12.65	5.35
M2 06	21.10.00 9:00	3.26	5	2.02	10	16.23	3.97
M2 06	28.10.00 9:00	2.96	5	2.84	5	14.81	0.00
M2 06	8.11.00 9:00	3.2	5	3.04	5	16.01	0.00

M2 07 25.6.00 19:00 8.9 5.68 2 8.85 2.51
--

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2 07	25.6.00 23:00	2.64		8.37	2	2.38	14.36
M2 07	26.6.00 7:00	8.97		8.56	2	8.82	8.30
M2 07	26.6.00 11:00	9.07		7.81	2	8.95	6.67
M2 07	26.6.00 15:00	7.2		3.7	5	6.99	11.51
M2 07	27.6.00 11:00	4.84		2.53	5	4.70	7.95
M2 07	3.7.00 9:00	11.03		7.89	2	10.94	4.84
M2 07	15.7.00 9:00	12.94		1.58	10	12.89	2.91
M2 07	21.7.00 9:00	9.61		1.23	10	9.56	2.74
M2 07	24.7.00 9:00	9.33		1.5	10	9.23	5.77
M2 07	19.8.00 9:00	2.03	5	11.37		10.13	1.24
M2 07	20.8.00 9:00	2.03	5	11.91		10.12	1.79
M2 07	21.8.00 9:00	4.47		2.2	5	4.35	6.65
M2 07	22.8.00 9:00	9.45		10.39		9.43	0.96
M2 07	23.8.00 9:00	11.05		10.73		11.06	0.00
M2 07	24.8.00 9:00	9.75		10.51		9.74	0.77
M2 07	25.8.00 9:00	9.57		10.8		9.55	1.25
M2 07	26.8.00 9:00			10.45			
M2 07	27.8.00 9:00	0.13	10	11.74		1.11	10.63
M2 07	28.8.00 9:00	0.03	5	11.92		0.00	11.99
M2 07	29.8.00 9:00	0.6	5	4.67		2.97	1.70
M2 07	30.8.00 9:00	2.18	5	10.82		10.90	0.00
M2 07	31.8.00 9:00	2.22	5	10.82		11.11	0.00
M2 07	1.9.00 9:00	2.15	5	10.59		10.75	0.00
M2 07	2.9.00 9:00	2.29	5	11.64		11.45	0.19
M2 07	3.9.00 9:00	2.19	5	0.97	10	10.97	0.00
M2 07	4.9.00 9:00	2.01	5	0.98	10	10.05	0.00
M2 07	5.9.00 9:00	2.06	5	1.18	10	10.27	1.53
M2 07	6.9.00 9:00	1.42	5	11.13		7.03	4.10
M2 07	7.9.00 9:00	1.76	5	11.22		8.76	2.46
M2 07	8.9.00 9:00	1.88	5	10.98		9.37	1.61
M2 07	9.9.00 9:00	1.65	5	10.96		8.20	2.76
M2 07	10.9.00 9:00	1.78	5	11.01		8.86	2.15
M2 07	21.9.00 9:00	2.01	5	11.02		10.03	0.99
M2 07	7.10.00 9:00	10.57		11.16		10.56	0.60
M2 07	28.10.00 9:00	10.96		11.03		10.96	0.07
M2 07	8.11.00 9:00	11.48		10.49		11.50	0.00
M2 09	26 6 00 15:00	10.01		2 66	5	0.05	2.25
IVIZ UO	20.00.15:00	10.01		∠.60	Э	9.95	3.35

IVIZ 08	26.6.00 15:00	10.01		2.66	5	9.95	3.35
M2 08	19.8.00 9:00	2.15	5	11.74		10.73	1.01
M2 08	20.8.00 9:00	2.19	5	11.6		10.94	0.66
M2 08	21.8.00 9:00	4.11		2.29	5	3.98	7.47
M2 08	22.8.00 9:00	9.64		10.81		9.62	1.19
M2 08	23.8.00 9:00	10.84		10.85		10.84	0.01
M2 08	24.8.00 9:00	10.05		10.81		10.04	0.77
M2 08	25.8.00 9:00	9.49		10.55		9.47	1.08
M2 08	26.8.00 9:00			10.75			
M2 08	27.8.00 9:00			11.69			
M2 08	28.8.00 9:00	9.85		11.28		9.82	1.46
M2 08	29.8.00 9:00	2.16	5	10.74		10.80	0.00
M2 08	30.8.00 9:00	2.25	5	10.77		11.26	0.00
M2 08	31.8.00 9:00	2.18	5	10.65		10.90	0.00
M2 08	1.9.00 9:00	2.1	5	10.65		10.50	0.15

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2 08	2.9.00 9:00	2.19	5	11.01		10.95	0.06
M2 08	3.9.00 9:00	2.17	5	10.71		10.85	0.00
M2 08	4.9.00 9:00	2.12	5	10.79		10.60	0.19
M2 08	5.9.00 9:00	1.98	5	11.87		9.86	2.01
M2 08	6.9.00 9:00	1.27	5	11.01		6.26	4.75
M2 08	7.9.00 9:00	1.32	5	10.31		6.53	3.78
M2 08	8.9.00 9:00	1.82	5	10.9		9.07	1.83
M2 08	9.9.00 9:00	1.81	5	10.63		9.02	1.61
M2 08	10.9.00 9:00	1.82	5	10.94		9.07	1.87
M2 08	21.9.00 9:00	1.97	5	10.62		9.84	0.78
M2 08	7.10.00 9:00	10.03		11.01		10.01	1.00
M2 08	28.10.00 9:00	11.25		10.61		11.26	0.00
M2 08	8.11.00 9:00	11.31		10.68		11.32	0.00

	M2-09	24.7.00 9:00	14.47		3.04	10	14.18	16.22
--	-------	--------------	-------	--	------	----	-------	-------

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-01	27.6.00 19:00	11.81		12.15		11.80	0.35
M3-01	29.6.00 15:00	3.18		12.62		3.01	9.61
M3-01	29.6.00 23:00	0.14		13.05		0.00	13.15
M3-01	30.6.00 7:00	0.05		12.88		0.00	13.07
M3-01	30.6.00 23:00	5.65		6.75	2	5.51	7.99
M3-01	1.7.00 7:00	8.91		5.63	2	8.87	2.39
M3-01	1.7.00 15:00	8.27		5.48	2	8.22	2.74
M3-01	1.7.00 23:00	9.52		5.2	2	9.50	0.90
M3-01	2.7.00 9:00	9.19		6.68	2	9.11	4.25
M3-01	2.7.00 19:00	8.63		6.55	2	8.55	4.55
M3-01	3.7.00 9:00	9.09		6.67	2	9.01	4.33
M3-01	3.7.00 19:00	8.26		6.86	2	8.16	5.56
M3-01	6.7.00 9:00	9.01		5.87	2	8.96	2.78
M3-01	9.7.00 9:00	11		6.96	2	10.95	2.97
M3-01	12.7.00 9:00	12.19		1.36	10	12.16	1.44
M3-01	15.7.00 9:00	11.55		1.35	10	11.51	1.99
M3-01	18.7.00 9:00	10.3		1.15	10	10.28	1.22
M3-01	21.7.00 9:00	10.18		12.48		10.14	2.34
M3-01	24.7.00 9:00	8.34		10.15		8.31	1.84
M3-02	27.6.00 19:00	0.2		13.44		0.00	13.48
M3-02	28.6.00 19:00	0.17		13.3		0.00	13.37
M3-02	29.6.00 7:00	0.22		13.07		0.00	13.09
M3-02	29.6.00 15:00	0.16		12.47		0.00	12.54
M3-02	29.6.00 23:00	3.59		5.92	2	3.44	8.40
M3-02	30.6.00 7:00	9.93		5.81	2	9.90	1.72
M3-02	30.6.00 23:00	8.82		6.3	2	8.75	3.85
M3-02	1.7.00 7:00	8.28		5.72	2	8.22	3.22
M3-02	1.7.00 15:00	6.9		5.61	2	6.82	4.40
M3-02	1.7.00 23:00	8.16		5.67	2	8.10	3.24
M3-02	2.7.00 9:00	7.99		6.58	2	7.90	5.26
M3-02	2.7.00 19:00	7.16		6.02	2	7.07	4.97
M3-02	3.7.00 9:00	6.11		5.35	2	6.03	4.67
M3-02	3.7.00 19:00	5.37		4.94	2	5.29	4.59
M3-02	6.7.00 9:00	1.72		1.02	2	1.71	0.33
M3-02	9.7.00 9:00	4.22		2.45	2	4.21	0.69
M3-02	12.7.00 9:00	1.55		3.17		1.52	1.65
M3-02	15.7.00 9:00	0.71		1.98		0.69	1.29
M3-02	18.7.00 9:00	0.89		2.02		0.87	1.15
M3-02	21.7.00 9:00	0.76		8.19		0.62	7.57
M3-02	24.7.00 9:00	0.74		17.73		0.43	17.30
M3-02	24.7.00 19:00	0.93		17.47		0.63	16.84
M3-02	25.7.00 9:00	0.92		16.22		0.64	15.58
M3-02	25.7.00 19:00	0.88		1.48	10	0.62	14.18
M3-02	26.7.00 9:00	1.05		1.89	10	0.72	18.18
M3-02	26.7.00 19:00	0.86		1.52	10	0.60	14.60
M3-02	27.7.00 9:00	0.76		0.58	25	0.51	13.99
M3-02	27.7.00 19:00	1.06		1.9	10	0.73	18.27
M3-02	28.7.00 9:00	1.07		1.6	10	0.80	15.20

Tab.: A2 arsenic concentration from M3

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-02	28.7.00 19:00	0.87		1.83	10	0.55	17.75
M3-02	29.7.00 9:00	1.1		2.15	10	0.73	20.77
M3-02	29.7.00 19:00	1.07		2.18	10	0.69	21.11
M3-02	30.7.00 9:00	1.08		2.35	10	0.67	22.83
M3-02	30.7.00 19:00	1.09		2.16	10	0.71	20.89
M3-02	31.7.00 9:00	1.08		2.22	10	0.69	21.51
M3-02	31.7.00 19:00	0.76		1.6	10	0.48	15.52
M3-02	1.8.00 9:00	1.12		2.29	10	0.72	22.18
M3-02	1.8.00 19:00	1.26		2.26	10	0.87	21.73
M3-02	2.8.00 9:00	1.19		2.38	10	0.78	23.02
M3-02	2.8.00 19:00	1.23		2.58	10	0.78	25.02
M3-02	3.8.00 9:00	1.52		2.81	10	1.03	27.07
M3-02	3.8.00 19:00	1.41		2.79	10	0.92	26.98
M3-02	4.8.00 9:00	1.7		2.82	10	1.21	26.99
M3-02	4.8.00 19:00	1.58		3.05	10	1.05	29.45
M3-02	5.8.00 9:00	1.71		2.81	10	1.23	26.87
M3-02	5.8.00 19:00	2.06		2.83	10	1.58	26.72
M3-02	6.8.00 9:00	4.25		2.93	10	3.79	25.51
M3-02	6.8.00 19:00	7.57		3.23	10	7.12	25.18
M3-02	7.8.00 9:00	2.24	5	1.53	25	10.70	27.55
M3-02	7.8.00 19:00	2.61	5	1.34	25	12.68	20.82
M3-02	8.8.00 9:00	1.62	5	1.72	25	7.46	35.54
M3-02	8.8.00 19:00	16.78		3.75	10	16.40	21.10
M3-02	9.8.00 9:00	10.56		5.89	10	9.67	49.23
M3-02	9.8.00 19:00	4.87		3.81	10	4.26	33.84
M3-02	11.8.00 9:00	18.23		4.18	10	17.80	24.00
M3-02	11.8.00 19:00	14.53		4.12	10	14.04	27.16
M3-02	12.8.00 9:00	14.27		4.43	10	13.72	30.58
M3-02	12.8.00 19:00	18.2		4	10	17.80	22.20
M3-02	13.8.00 9:00	20.16		4.17	10	19.77	21.93
M3-02	13.8.00 19:00	10.77		4.27	10	10.18	32.52
M3-02	14.8.00 9:00	18.03		4.59	10	17.52	28.38
M3-02	14.8.00 19:00	19.96		4.07	10	19.58	21.12
M3-02	15.8.00 9:00	2.87	10	4.05	10	28.48	12.02
M3-02	15.8.00 19:00	2.34	10	3.15	10	23.25	8.25
M3-02	16.8.00 9:00	2.64	10	3.59	10	26.23	9.67
M3-02	16.8.00 19:00	1.86	10	3.24	10	18.35	14.05
M3-02	17.8.00 9:00	0.82	10	4.25	10	7.57	34.93
M3-02	17.8.00 19:00	3.03	10	3.95	10	30.13	9.37
M3-02	18.8.00 9:00	1.21	10	3.48	10	11.68	23.12
M3-02	18.8.00 19:00	2.65	10	3.28	10	26.38	6.42
M3-02	19.8.00 9:00	1.85	10	3.1	10	18.27	12.73
M3-02	21.8.00 9:00	2.52	5	2.87	10	12.30	16.40
M3-02	22.8.00 9:00	1.64	10	5.11	5	16.23	9.32
M3-02	23.8.00 9:00	1.22	10	5.16	5	11.95	13.85
M3-02	24.8.00 9:00	1.83	10	5.18	5	18.16	7.74
M3-02	25.8.00 9:00	1.19	10	4.93	5	11.67	12.98
M3-02	26.8.00 9:00	1.73	10	4.79	5	17.18	6.77
M3-02	29.8.00 9:00	1.34	10	0.83	25	13.27	7.48
M3-02	30.8.00 9:00	1.35	10	0.91	25	13.33	9.42
M3-02	31.8.00 9:00	1.23	10	0.89	25	12.12	10.13
M3-02	1.9.00 9:00	1.39	10	0.98	25	13.71	10.79
M3-02	2.9.00 9:00	1.21	10	0.84	25	11.94	9.06

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-02	3.9.00 9:00	1.41	10	0.74	25	14.02	4.48
M3-02	4.9.00 9:00	1.08	10	0.97	25	10.55	13.70
M3-02	5.9.00 9:00	1.23	10	0.99	25	12.07	12.68
M3-02	6.9.00 9:00	0.8	10	21.93		7.74	14.19
M3-02	7.9.00 9:00	1.12	10	22.7		10.99	11.71
M3-02	8.9.00 9:00	0.87	10	22.76		8.44	14.32
M3-02	9.9.00 9:00	1.03	10	23.1		10.07	13.03
M3-02	10.9.00 9:00	0.77	10	25.22		7.38	17.84
M3-02	11.9.00 9:00	1.15	10	23.51		11.28	12.23
M3-02	12.9.00 9:00	1.19	10	22.44		11.71	10.73
M3-02	13.9.00 9:00	1.35	10	0.86	25	13.35	8.15
M3-02	14.9.00 9:00	1.15	10	0.75	25	11.37	7.38
M3-02	21.9.00 9:00	0.96	10	0.77	25	9.42	9.83
M3-02	7.10.00 9:00	11.42		0.88	25	11.23	10.77
M3-02	14.10.00 9:00	9.36		2.76	10	9.03	18.57
M3-02	21.10.00 9:00	7.78		3.44	10	7.29	27.11
M3-02	28.10.00 9:00	11.88		2.31	10	11.67	11.43
M3-02	8.11.00 9:00	9.61		3.65	10	9.12	27.38
M3-03	27.6.00 19:00	0.06		12.48		0.00	12.65
M3-03	28.6.00 19:00	0.14		11.23		0.00	11.29
M3-03	29.6.00 7:00	0.14		11.78		0.00	11.85
M3-03	29.6.00 15:00	8.31		5.8	2	8.25	3.35
M3-03	29.6.00 23:00	6.62		5.34	2	6.55	4.13
M3-03	30.6.00 7:00	8.84		5.68	2	8.79	2.57
M3-03	30.6.00 23:00	6.3		8.58	2	6.10	11.06
M3-03	1.7.00 7:00	6.96		8.9	2	6.76	11.04
M3-03	1.7.00 15:00	5.24		9.61	2	4.98	14.24
M3-03	1.7.00 23:00	7.37		2.32	10	7.08	16.12
M3-03	2.7.00 9:00	7.45		11.3	2	7.17	15.43
M3-03	2.7.00 19:00	5.99		9.9	2	5.74	14.06
M3-03	3.7.00 9:00	5.49		9.48	2	5.24	13.72
M3-03	3.7.00 19:00	5.62		8.51	2	5.41	11.61
M3-03	4.7.00 9:00	5.65		4.68	2	5.58	3.78
M3-03	5.7.00 9:00	4.14		2.5	2	4.12	0.88
M3-03	5.7.00 19:00	4.48		2.86	2	4.46	1.26
M3-03	6.7.00 9:00	1.06		0.19	2	1.07	0.00
M3-03	6.7.00 19:00	3.79		2.26	2	3.78	0.74
M3-03	7.7.00 9:00	3.3		2.02	2	3.29	0.75
M3-03	7.7.00 19:00	2.59		1.6	2	2.58	0.62
M3-03	8.7.00 9:00	2.4		1.3	2	2.40	0.20
M3-03	8.7.00 19:00	2.41		1.31	2	2.41	0.21
M3-03	9.7.00 9:00	1.91		1.1	2	1.90	0.30
M3-03	9.7.00 19:00	1.7		1.01	2	1.69	0.33
M3-03	10.7.00 9:00	1.88		1.14	2	1.87	0.41
M3-03	10.7.00 19:00	1.92		1.2	2	1.91	0.49
M3-03	11.7.00 9:00	1.94		1.22	2	1.93	0.51
M3-03	11.7.00 19:00	1.41		2.13		1.40	0.73
M3-03	12.7.00 9:00	1.55		2.6		1.53	1.07
M3-03	12.7.00 19:00	1.81		3.16	ļ	1.79	1.37
M3-03	13.7.00 9:00	1.83		2.55		1.82	0.73
M3-03	13.7.00 19:00	1.22		1.98		1.21	0.77

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-03	14.7.00 9:00	2.06		2.93		2.04	0.89
M3-03	14.7.00 19:00	1.57		2.46		1.55	0.91
M3-03	15.7.00 9:00	1.81		3.45		1.78	1.67
M3-03	15.7.00 19:00	1.68		3.75		1.64	2.11
M3-03	16.7.00 9:00	1.54		4.33		1.49	2.84
M3-03	16.7.00 19:00	1.65		5.41		1.58	3.83
M3-03	17.7.00 9:00	2.02		9.09		1.89	7.20
M3-03	17.7.00 19:00	1.97		1.25	10	1.78	10.72
M3-03	18.7.00 9:00	2.9		3.51	10	2.31	32.79
M3-03	19.7.00 9:00	2.61		3.83	10	1.96	36.34
M3-03	19.7.00 19:00	3.02		5.07	10	2.15	48.55
M3-03	20.7.00 9:00	3.51		6.22	10	2.43	59.77
M3-03	20.7.00 19:00	3.89		7.6	10	2.57	73.43
M3-03	21.7.00 9:00	5.79		6.08	25	3.11	148.89
M3-03	21.7.00 19:00	4.78		5.11	25	2.53	125.22
M3-03	22.7.00 9:00	4.69		5.58	25	2.22	137.28
M3-03	22.7.00 19:00	5.62		6.16	25	2.90	151.10
M3-03	23.7.00 9:00	5.43		6.1	25	2.73	149.77
M3-03	23.7.00 19:00	5.65		6.48	25	2.78	159.22
M3-03	24.7.00 9:00	6.41		6.88	25	3.37	168.63
M3-03	24.7.00 19:00	6.36		7.11	25	3.22	174.53
M3-03	25.7.00 9:00	6.05		7.7	25	2.63	189.87
M3-03	25.7.00 19:00	1.08	5	3.83	50	1.99	189.51
M3-03	26.7.00 9:00	1.1	5	3.5	50	2.39	172.61
M3-03	26.7.00 19:00	1.13	5	3.93	50	2.15	194.35
M3-03	27.7.00 9:00	1.07	5	3.54	50	2.20	174.80
M3-03	27.7.00 19:00	1.23	5	4.1	50	2.51	202.49
M3-03	28.7.00 9:00	1.71	5	5.39	50	3.77	265.73
M3-03	28.7.00 19:00	1.84	5	5.73	50	4.12	282.38
M3-03	29.7.00 9:00	1.39	5	4.75	50	2.72	234.78
M3-03	29.7.00 19:00	1.15	5	3.68	50	2.48	181.52
M3-03	30.7.00 9:00	1.08	5	3.66	50	2.14	180.86
M3-03	30.7.00 19:00	1.11	5	3.82	50	2.15	188.85
M3-03	31.7.00 9:00	1.21	5	4.05	50	2.45	200.05
M3-03	31.7.00 19:00	1.43	5	4.27	50	3.37	210.13
M3-03	1.8.00 9:00	1.51	5	4.52	50	3.55	222.45
M3-03	1.8.00 19:00	1.72	5	4.76	50	4.40	233.60
M3-03	2.8.00 9:00	1.8	5	4.56	50	4.99	223.01
M3-03	2.8.00 19:00	1.49	5	4.54	50	3.43	223.57
M3-03	3.8.00 9:00	1.28	5	4.04	50	2.81	199.19
M3-03	3.8.00 19:00	1.27	5	4.17	50	2.64	205.86
M3-03	4.8.00 9:00	1.27	5	4.24	50	2.58	209.42
M3-03	4.8.00 19:00	1.29	5	4.2	50	2.72	207.28
M3-03	5.8.00 9:00	1.33	5	3.91	50	3.19	192.31
M3-03	5.8.00 19:00	1.54	5	4.23	50	3.96	207.54
M3-03	6.8.00 9:00	1.7	5	4.11	50	4.89	200.61
M3-03	6.8.00 19:00	1.47	5	4.19	50	3.64	205.86
M3-03	7.8.00 9:00	8.69	5	4.78	50	39.87	199.13
M3-03	7.8.00 19:00	5.83	5	4.2	50	25.83	184.17
M3-03	8.8.00 9:00	6.86	5	4.28	50	31.01	182.99
M3-03	11.8.00 19:00	3.72	10	4.07	50	34.15	169.35
M3-03	12.8.00 9:00	4.55	10	4.34	50	42.36	174.64
M3-03	12.8.00 19:00	3.36	10	4.27	50	30.30	183.20

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-03	13.8.00 9:00	4.26	10	4.42	50	39.33	181.67
M3-03	13.8.00 19:00	1.93	10	4.66	50	15.38	217.62
M3-03	14.8.00 9:00	3.82	10	4.42	50	34.85	186.15
M3-03	14.8.00 19:00	3.97	10	4.37	50	36.42	182.08
M3-03	15.8.00 9:00	4.98	10	4.62	50	46.48	184.52
M3-03	15.8.00 19:00	4.67	10	4.6	50	43.34	186.66
M3-03	16.8.00 9:00	5.42	10	4.37	50	51.19	167.31
M3-03	16.8.00 19:00	5.23	10	4.48	50	49.15	174.85
M3-03	17.8.00 9:00	1.86	10	4.31	50	14.99	200.51
M3-03	17.8.00 19:00	4.12	10	4.26	50	38.05	174.95
M3-03	18.8.00 9:00	2.76	10	4.21	50	24.25	186.25
M3-03	18.8.00 19:00	5.1	10	4.16	50	48.12	159.88
M3-03	19.8.00 9:00	7.92	10	4.23	50	76.77	134.73
M3-03	21.8.00 9:00	9.72	5	3.95	50	45.87	151.63
M3-03	22.8.00 9:00	6.59	10	3.94	50	63.50	133.50
M3-03	23.8.00 9:00	5.57	10	3.85	50	53.19	139.31
M3-03	24.8.00 9:00	7.66	10	3.47	50	74.82	98.68
M3-03	25.8.00 9:00	6.12	10	3.85	50	58.79	133.71
M3-03	26.8.00 9:00	7.41	10	3.57	50	72.19	106.31
M3-03	27.8.00 9:00	2.53	10	4.81	40	22.24	170.16
M3-03	28.8.00 9:00	5.03	10	4.99	40	47.56	152.04
M3-03	29.8.00 9:00	7.55	10	3.87	50	73.34	120.16
M3-03	30.8.00 9:00	6.15	10	3.65	50	59.28	123.22
M3-03	31.8.00 9:00	5.21	10	3.57	50	49.78	128.72
M3-03	1.9.00 9:00	3.89	10	3.73	50	36.19	150.31
M3-03	2.9.00 9:00	6.12	10	3.57	50	59.05	119.45
M3-03	3.9.00 9:00	5.78	10	3.33	50	55.81	110.69
M3-03	4.9.00 9:00	5.87	10	3.31	50	56.74	108.76
M3-03	5.9.00 9:00	4.98	10	3.68	50	47.34	136.66
M3-03	6.9.00 9:00	3.22	10	3	50	30.04	119.96
M3-03	7.9.00 9:00	5.49	10	3.04	50	53.12	98.88
M3-03	8.9.00 9:00	4.96	10	3.08	50	47.69	106.31
M3-03	9.9.00 9:00	4.73	10	3.02	50	45.40	105.60
M3-03	10.9.00 9:00	3.45	10	2.7	50	32.66	102.34
M3-03	11.9.00 9:00	2.54	10	3.06	50	23.06	129.94
M3-03	12.9.00 9:00	4.25	10	2.79	50	40.72	98.78
M3-03	13.9.00 9:00	5.27	10	2.58	50	51.30	77.70
M3-03	14.9.00 9:00	5.74	10	2.66	50	56.01	76.99
M3-03	21.9.00 9:00	5.62	10	2.57	50	54.87	73.63
M3-03	7.10.00 9:00	5.63	10	2.41	50	55.12	65.38
M3-03	14.10.00 9:00	3.18	10	5.04	25	30.07	95.93
M3-03	21.10.00 9:00	2.92	10	4.84	25	27.52	93.48
M3-03	28.10.00 9:00	6.84	5	3.54	25	0.00	90.12
M3-03	8.11.00 9:00	4.89	5	2.99	25	0.00	76.12
M0.04		0.00		7.00		0.00	0.04
1013-04	I ∠7.6.00 19:00 I	0.02		1.89		0.00	8.01

M3-04	27.6.00 19:00	0.02	7.89		0.00	8.01
M3-04	28.6.00 19:00	0.45	24.49		0.01	24.48
M3-04	29.6.00 7:00	0.89	37.74		0.21	37.53
M3-04	29.6.00 15:00	1.21	12.62	5	0.08	63.02
M3-04	29.6.00 23:00	1.68	15.18	5	0.32	75.58
M3-04	30.6.00 7:00	7.17	9.26	10	5.60	87.00
M3-04	30.6.00 23:00	7.15	10.88	10	5.29	103.51

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-04	1.7.00 7:00	7.11		9.6	10	5.48	90.52
M3-04	1.7.00 15:00	6.66		10.91	10	4.78	104.32
M3-04	1.7.00 23:00	7.26		9.48	10	5.66	89.14
M3-04	2.7.00 9:00	6.66		10.45	10	4.87	99.63
M3-04	2.7.00 19:00	6.16		9.54	10	4.52	90.88
M3-04	3.7.00 9:00	6.2		8.53	10	4.75	80.55
M3-04	3.7.00 19:00	5.33		7.21	10	4.11	67.99
M3-04	4.7.00 9:00	4.53		3.55	10	3.96	31.54
M3-04	5.7.00 9:00	3.85		4	2	3.77	4.23
M3-04	5.7.00 19:00	2.73		2.31	2	2.70	1.92
M3-04	6.7.00 9:00	2.55		1.88	2	2.53	1.23
M3-04	6.7.00 19:00	3.05		2.2	2	3.03	1.37
M3-04	7.7.00 9:00	2.58		1.88	2	2.56	1.20
M3-04	7.7.00 19:00	2.79		2.57	2	2.75	2.39
M3-04	8.7.00 9:00	1.54		2.36	2	1.48	3.24
M3-04	8.7.00 19:00	2.88		1.09	10	2.73	8.17
M3-04	9.7.00 9:00	2.8		2.53	10	2.39	22.91
M3-04	9.7.00 19:00	2.61		2.83	10	2.14	26.16
M3-04	10.7.00 9:00	0.41		2.6	10	0.00	26.06
M3-04	10.7.00 19:00	3.78		5.72	10	2.80	54.40
M3-04	11.7.00 9:00	3.88		8.04	10	2.48	77.92
M3-04	11.7.00 19:00	3.96		9.55	10	2.28	93.22
M3-04	12.7.00 9:00	4.28		10.93	10	2.35	106.95
M3-04	12.7.00 19:00	4.14		12.22	10	1.98	120.22
M3-04	13.7.00 9:00	4.66		13.11	10	2.34	128.76
M3-04	13.7.00 19:00	4.34		14.22	10	1.81	140.39
M3-04	14.7.00 9:00	5.44		15.81	10	2.64	155.46
M3-04	14.7.00 19:00	4.32		14.81	10	1.68	146.42
M3-04	15.7.00 9:00	5.81		18.96	10	2.44	187.16
M3-04	15.7.00 19:00	6.81		20.06	10	3.26	197.34
M3-04	16.7.00 9:00	6.85		20.34	10	3.25	200.15
M3-04	16.7.00 19:00	7.12		22.05	10	3.21	217.29
M3-04	17.7.00 9:00	8.16		21.63	10	4.34	211.96
M3-04	17.7.00 19:00	7.78		21.88	10	3.91	214.89
M3-04	18.7.00 9:00	7.92		20.85	10	4.24	204.26
M3-04	18.7.00 19:00	8.72		23.23	10	4.62	227.68
M3-04	19.7.00 9:00	8.93		24.82	10	4.54	243.66
M3-04	19.7.00 19:00	9.02		24.6	10	4.68	241.32
M3-04	20.7.00 9:00	9.37		12.55	25	3.79	309.96
M3-04	20.7.00 19:00	9.96		12.81	25	4.27	315.98
M3-04	21.7.00 9:00	9.91		14.49	25	3.45	358.80
M3-04	21.7.00 19:00	10.18		14.49	25	3.73	358.52
M3-04	22.7.00 9:00	9.98		14.38	25	3.57	355.93
M3-04	22.7.00 19:00	10.33		15.47	25	3.43	383.32
M3-04	23.7.00 9:00	10.18		13.51	25	4.18	333.57
M3-04	23.7.00 19:00	10.93		13.99	25	4.72	345.03
M3-04	24.7.00 9:00	10.89		13.77	25	4.78	339.47
M3-04	24.7.00 19:00	10.45		13.81	25	4.31	340.94
M3-04	25.7.00 9:00	8.72		13.76	25	2.57	341.43
M3-04	25.7.00 19:00	2.13	5	6.66	50	4.74	328.26
M3-04	26.7.00 9:00	2.05	5	6.38	50	4.59	314.41
M3-04	26.7.00 19:00	2.04	5	6.67	50	4.27	329.23
M3-04	27.7.00 9:00	2.1	5	6.21	50	5.00	305.50

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-04	27.7.00 19:00	2	5	6.55	50	4.18	323.32
M3-04	28.7.00 9:00	1.95	5	6.33	50	4.13	312.37
M3-04	28.7.00 19:00	1.96	5	6.69	50	3.85	330.65
M3-04	29.7.00 9:00	2.08	5	6.35	50	4.77	312.73
M3-04	29.7.00 19:00	2.18	5	7.06	50	4.63	348.37
M3-04	30.7.00 9:00	2.47	5	8.1	50	5.15	399.85
M3-04	30.7.00 19:00	3.22	5	9.7	50	7.51	477.49
M3-04	31.7.00 9:00	1.75	5	5.06	50	4.27	248.73
M3-04	31.7.00 19:00	3.49	5	11.29	50	7.42	557.08
M3-04	1.8.00 9:00	3.26	5	9.67	50	7.74	475.76
M3-04	1.8.00 19:00	2.83	5	8.56	50	6.56	421.44
M3-04	2.8.00 9:00	2.19	5	7.04	50	4.70	347.30
M3-04	2.8.00 19:00	1.99	5	4.5	50	6.01	218.99
M3-04	3.8.00 9:00	1.76	5	5.55	50	3.87	273.63
M3-04	3.8.00 19:00	1.7	5	4.95	50	4.12	243.38
M3-04	4.8.00 9:00	1.61	5	4.53	50	4.05	222.45
M3-04	4.8.00 19:00	1.64	5	5.02	50	3.75	247.25
M3-04	5.8.00 9:00	1.53	5	5	50	3.21	246.79
M3-04	5.8.00 19:00	1.66	5	5.24	50	3.65	258.35
M3-04	6.8.00 9:00	1.65	5	5.37	50	3.48	265.02
M3-04	6.8.00 19:00	1.57	5	4.65	50	3.73	228.77
M3-04	7.8.00 9:00	3	5	4.56	50	11.10	216.90
M3-04	7.8.00 19:00	7.9	5	5.02	50	35.62	215.38
M3-04	8.8.00 9:00	4.99	5	5.15	50	20.69	236.81
M3-04	8.8.00 19:00	0.66	10	5.08	50	2.07	251.93
M3-04	9.8.00 9:00	0.69	10	5.21	50	2.25	258.25
M3-04	9.8.00 19:00	3.09	10	5.03	50	26.86	224.64
M3-04	11.8.00 9:00	2.61	10	5.01	50	21.99	228.51
M3-04	11.8.00 19:00	4.81	10	5.17	50	44.24	214.26
M3-04	12.8.00 9:00	5.74	10	5.14	50	53.74	203.26
M3-04	12.8.00 19:00	4.53	10	5.3	50	41.27	223.73
M3-04	13.8.00 9:00	3.87	10	5.03	50	34.80	216.70
M3-04	13.8.00 19:00	2.61	10	5.04	50	21.96	230.04
M3-04	14.8.00 9:00	3.5	10	5.14	50	30.93	226.07
M3-04	14.8.00 19:00	5.57	10	5.19	50	51.96	207.54
M3-04	15.8.00 9:00	7.22	10	5.23	50	68.73	192.77
M3-04	15.8.00 19:00	7.56	10	5.27	50	72.16	191.34
M3-04	16.8.00 9:00	9.87	10	5.44	50	95.52	176.48
M3-04	16.8.00 19:00	8.71	10	5.62	50	83.55	197.45
M3-04	17.8.00 9:00	1.85	10	5.1	50	14.16	240.84
M3-04	17.8.00 19:00	5.46	10	5.36	50	50.69	217.31
M3-04	18.8.00 9:00	6.69	10	5.13	50	63.42	193.08
M3-04	18.8.00 19:00	7.75	10	5.54	50	73.84	203.16
M3-04	19.8.00 9:00	11.64	10	5.51	50	113.48	162.02
M3-04	21.8.00 9:00	14.39	5	4.66	50	69.00	164.00
M3-04	22.8.00 9:00	7.02	10	4.91	50	66.99	178.51
M3-04	23.8.00 9:00	7.52	10	4.88	50	72.11	171.89
M3-04	24.8.00 9:00	10.43	10	4.94	50	101.68	145.32
M3-04	25.8.00 9:00	11.62	10	4.84	50	113.89	128.11
M3-04	26.8.00 9:00	13.1	10	4.68	50	129.11	104.89
M3-04	29.8.00 9:00	12.12	10	5.12	50	118.73	137.27
M3-04	30.8.00 9:00	9.41	10	5.13	50	91.12	165.38
M3-04	31.8.00 9:00	8.79	10	4.98	50	84.95	164.05

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-04	1.9.00 9:00	11.02	10	5.2	50	107.45	152.55
M3-04	2.9.00 9:00	9.84	10	4.82	50	95.79	145.21
M3-04	3.9.00 9:00	10.03	10	4.71	50	97.82	137.68
M3-04	4.9.00 9:00	8.75	10	4.55	50	84.93	142.57
M3-04	5.9.00 9:00	7.86	10	5.18	50	75.29	183.71
M3-04	6.9.00 9:00	3.29	10	4.34	50	29.53	187.47
M3-04	7.9.00 9:00	7.27	10	4.44	50	69.96	152.04
M3-04	8.9.00 9:00	8.32	10	4.34	50	80.75	136.25
M3-04	9.9.00 9:00	5.09	10	4.28	50	47.91	166.09
M3-04	10.9.00 9:00	5.03	10	4.47	50	47.12	176.38
M3-04	11.9.00 9:00	7.69	10	4.51	50	74.18	151.32
M3-04	12.9.00 9:00	8.73	10	4.35	50	84.91	132.59
M3-04	13.9.00 9:00	7.99	10	4.36	50	77.37	140.63
M3-04	14.9.00 9:00	6.11	10	4.12	50	58.44	147.56
M3-04	21.9.00 9:00	4	10	3.8	50	37.25	152.75
M3-04	30.9.00 9:00	3.93	10	3.72	50	36.61	149.39
M3-04	7.10.00 9:00	5.38	10	3.73	50	51.37	135.13
M3-04	14.10.00 9:00	2.96	10	8.17	25	26.40	177.85
M3-04	21.10.00 9:00	4.41	10	7.52	25	41.46	146.54
M3-04	28.10.00 9:00	6.01	5	5.83	25	27.93	117.82
M3-04	8.11.00 9:00	10.09	5	4.82	25	49.17	71.33
M3-05	27.6.00 19:00	0.23		12.44		0.01	12.43
M3-05	28.6.00 19:00	0.1		9.9		0.00	9.98
M3-05	29.6.00 7:00	0.13		9.19		0.00	9.23
M3-05	29.6.00 15:00	0.16		8.79		0.00	8.79
M3-05	29.6.00 23:00	6.97		8.59		6.94	1.65
M3-05	30.6.00 7:00	7.93		8.78		7.91	0.87
M3-05	30.6.00 23:00	5.92		8.53		5.87	2.66
M3-05	1.7.00 7:00	5.05		4.12	2	4.99	3.25
M3-05	1.7.00 15:00	3.67		4.14	2	3.59	4.69
M3-05	1.7.00 23:00	5.02		4.08	2	4.96	3.20
M3-05	2.7.00 9:00	5.38		3.59	2	5.35	1.83
M3-05	2.7.00 19:00	4.59		3.61	2	4.54	2.68
M3-05	3.7.00 9:00	4.77		3.24	2	4.74	1.74
M3-05	3.7.00 19:00	4.43		3.19	2	4.39	1.99
M3-05	4.7.00 9:00	4.59		2.7	2	4.58	0.82
M3-05	5.7.00 9:00	2.69		1.57	2	2.68	0.46
M3-05	5.7.00 19:00	3.73		2.17	2	3.72	0.62
M3-05	6.7.00 9:00	0.76		1.68	2	0.71	2.65
M3-05	6.7.00 19:00	3.51		2.12	2	3.50	0.74
M3-05	7.7.00 9:00	3.32		1.85	2	3.31	0.39
M3-05	7.7.00 19:00	2.24		1.51	2	2.23	0.79
M3-05	8.7.00 9:00	1.68		0.94	2	1.68	0.20
M3-05	8.7.00 19:00	1.98		1.02	2	1.98	0.06
M3-05	9.7.00 9:00	1.66		0.97	2	1.65	0.29
M3-05	9.7.00 19:00	1.82		1.14	2	1.81	0.47
M3-05	10.7.00 9:00	1.66		3.79	2	1.55	6.03
M3-05	10.7.00 19:00	0.66		1.16	2	0.63	1.69
M3-05	11.7.00 9:00	2.05		1.03	2	2.05	0.01
M3-05	11.7.00 19:00	1.53		2.65		1.51	1.14
M3-05	12.7.00 9:00	1.02		1.65		1.01	0.64

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-05	12.7.00 19:00	0.25		1.38		0.23	1.15
M3-05	13.7.00 9:00	1.28		2.74		1.25	1.49
M3-05	13.7.00 19:00	0.61		2.19		0.58	1.61
M3-05	14.7.00 9:00	1.19		2.98		1.16	1.82
M3-05	14.7.00 19:00	0.81		7.06		0.70	6.36
M3-05	15.7.00 9:00	1.54		2.6		1.52	1.08
M3-05	15.7.00 19:00	1.15		2.96		1.12	1.84
M3-05	16.7.00 9:00	1.13		3.46		1.09	2.37
M3-05	16.7.00 19:00	1.28		3.14		1.25	1.89
M3-05	17.7.00 9:00	1.41		1.01		1.42	-0.41
M3-05	17.7.00 19:00	1.3		2.99		1.27	1.72
M3-05	18.7.00 9:00	1.6		3.05		1.57	1.48
M3-05	18.7.00 19:00	1.45		3.16		1.42	1.74
M3-05	19.7.00 9:00	1.83		2.95		1.81	1.14
M3-05	19.7.00 19:00	1.43		3.48		1.39	2.09
M3-05	20.7.00 9:00	2		4.74		1.95	2.79
M3-05	20.7.00 19:00	2.01		8.35		1.89	6.46
M3-05	21.7.00 9:00	2.3		23.35		1.91	21.44
M3-05	22.7.00 9:00	2.39		10.51	5	1.47	51.08
M3-05	22.7.00 19:00	3.43		13.56	5	2.25	65.55
M3-05	23.7.00 9:00	3.59		8.64	10	2.07	84.33
M3-05	23.7.00 19:00	4.6		10.72	10	2.72	104.48
M3-05	24.7.00 9:00	5.39		12.56	10	3.19	122.41
M3-05	24.7.00 19:00	5.41		12.02	10	3.31	116.89
M3-05	25.7.00 9:00	4.85		13.75	10	2.42	135.08
M3-05	25.7.00 19:00	1.16	5	6.91	25	2.74	170.01
M3-05	26.7.00 9:00	1.21	5	7.31	25	2.81	179.94
M3-05	26.7.00 19:00	1.23	5	7.46	25	2.84	183.66
M3-05	27.7.00 9:00	1.24	5	7.81	25	2.73	192.52
M3-05	27.7.00 19:00	1.25	5	7.95	25	2.72	196.03
M3-05	28.7.00 9:00	1.36	5	8.6	25	2.98	212.02
M3-05	28.7.00 19:00	1.41	5	8.77	25	3.16	216.09
M3-05	29.7.00 9:00	1.45	5	9.1	25	3.21	224.29
M3-05	29.7.00 19:00	1.43	5	9	25	3.16	221.84
M3-05	30.7.00 9:00	1.62	5	9.89	25	3.72	243.53
M3-05	30.7.00 19:00	2.06	5	12.26	25	4.87	301.63
M3-05	31.7.00 9:00	3.7	5	16.82	25	11.13	409.37
M3-05	31.7.00 19:00	4.69	5	22.25	25	13.68	542.57
M3-05	1.8.00 9:00	4.99	5	21.12	25	15.73	512.27
M3-05	1.8.00 19:00	3.87	5	15.9	25	12.42	385.08
M3-05	2.8.00 9:00	3.73	5	10.4	25	14.23	245.77
M3-05	2.8.00 19:00	3.02	5	8.72	25	11.38	206.62
M3-05	3.8.00 19:00	3.95	5	9.08	25	15.95	211.05
M3-05	4.8.00 9:00	6.1	5	9.27	25	26.81	204.94
M3-05	4.8.00 19:00	5.19	5	9.37	25	22.13	212.12
M3-05	5.8.00 9:00	6.12	5	9.84	25	26.65	219.35
M3-05	5.8.00 19:00	5.59	5	9.36	25	24.17	209.83
M3-05	6.8.00 9:00	5.37	5	9.49	25	22.99	214.26
M3-05	6.8.00 19:00	5.82	5	9.29	25	25.38	206.87
M3-05	7.8.00 9:00	6.85	5	9.11	25	30.70	197.05
M3-05	7.8.00 19:00	4.95	5	9.05	25	21.06	205.19
M3-05	8.8.00 9:00	6.48	5	8.87	25	28.93	192.82
M3-05	8.8.00 19:00	3.41	10	9.92	25	30.18	217.82

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-05	9.8.00 9:00	3.32	10	9.71	25	29.36	213.39
M3-05	9.8.00 19:00	2.43	10	10.14	25	20.10	233.40
M3-05	11.8.00 9:00	3.27	10	10.64	25	28.42	237.58
M3-05	11.8.00 19:00	4.06	10	10.45	25	36.56	224.69
M3-05	12.8.00 9:00	3.73	10	11.32	25	32.80	250.20
M3-05	12.8.00 19:00	2.54	10	10.61	25	21.00	244.25
M3-05	13.8.00 9:00	0.78	10	10.59	25	3.09	261.66
M3-05	13.8.00 19:00	1.73	10	9.81	25	13.12	232.13
M3-05	14.8.00 9:00	3.62	10	10.34	25	32.13	226.37
M3-05	14.8.00 19:00	4.22	10	10.49	25	38.17	224.08
M3-05	15.8.00 9:00	12.32	10	10.04	25	120.86	130.14
M3-05	15.8.00 19:00	5.38	10	10.59	25	49.93	214.82
M3-05	16.8.00 9:00	7.44	10	11.18	25	70.64	208.86
M3-05	16.8.00 19:00	6.99	10	10.54	25	66.35	197.15
M3-05	17.8.00 9:00	1.59	10	10.42	25	11.42	249.08
M3-05	17.8.00 19:00	3.69	10	10.53	25	32.75	230.50
M3-05	18.8.00 9:00	6.28	10	10.94	25	58.94	214.56
M3-05	18.8.00 19:00	3.96	10	10.56	25	35.49	228.51
M3-05	19.8.00 9:00	4.13	10	10.82	25	37.10	233.40
M3-05	21.8.00 9:00	14.42	5	5.08	50	68.77	185.23
M3-05	22.8.00 9:00	6.92	10	4.79	50	66.08	173.42
M3-05	23.8.00 9:00	5.64	10	4.7	50	53.13	181.87
M3-05	24.8.00 9:00	9.07	10	4.62	50	88.13	142.87
M3-05	25.8.00 9:00	6.51	10	4.57	50	62.10	166.40
M3-05	29.8.00 9:00	7.66	10	9.97	25	73.43	175.82
M3-05	30.8.00 9:00	6.66	10	10.11	25	63.19	189.56
M3-05	31.8.00 9:00	7.1	10	9.81	25	67.81	177.44
M3-05	1.9.00 9:00	8.08	10	9.73	25	77.82	165.43
M3-05	2.9.00 9:00	6.06	10	9.47	25	57.37	179.38
M3-05	3.9.00 9:00	8.02	10	9.32	25	77.40	155.60
M3-05	4.9.00 9:00	8.86	10	9.46	25	85.89	150.61
M3-05	5.9.00 9:00	6.6	10	9.81	25	62.71	182.54
M3-05	6.9.00 9:00	4.68	10	9.08	25	43.50	183.50
M3-05	7.9.00 9:00	6.55	10	9.35	25	62.42	171.33
M3-05	8.9.00 9:00	7.62	10	9.34	25	73.32	160.18
M3-05	9.9.00 9:00	7.52	10	9.13	25	72.39	155.86
M3-05	10.9.00 9:00	4.74	10	8.76	25	44.25	174.75
M3-05	11.9.00 9:00	2.69	10	8.38	25	23.55	185.95
M3-05	12.9.00 9:00	7.34	10	8.35	25	70.92	137.83
M3-05	13.9.00 9:00	7.57	10	8.82	25	73.05	147.45
M3-05	14.9.00 9:00	4.72	10	8.52	25	44.16	168.84
M3-05	21.9.00 9:00	5.26	10	8.49	25	49.67	162.58
M3-05	30.9.00 9:00	6.02	10	7.63	25	57.81	132.94
M3-05	7.10.00 9:00	3.78	10	6.92	25	35.32	137.68
M3-05	14.10.00 9:00	4.47	10	7.54	25	42.06	146.44
M3-05	21.10.00 9:00	3.93	10	7.02	25	36.80	138.70
M3-05	28.10.00 9:00	6.04	5	4.86	25	28.53	92.97
M3-05	8.11.00 9:00	6.97	5	4.25	25	33.54	72.71

M3-06	27.6.00 19:00	0.19	13.25	0.00	13.30
M3-06	28.6.00 19:00	0.16	10.98	0.00	11.02
M3-06	29.6.00 7:00	0.04	10.08	0.00	10.22

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-06	29.6.00 15:00	0.15		9.76		0.00	9.79
M3-06	29.6.00 23:00	0.13		9.38		0.00	9.42
M3-06	30.6.00 7:00	5.14		9.03		5.07	3.96
M3-06	30.6.00 23:00	4.04		8.56		3.96	4.60
M3-06	1.7.00 7:00	6.49		3.92	2	6.47	1.37
M3-06	1.7.00 15:00	5.99		4.22	2	5.95	2.49
M3-06	1.7.00 23:00	6.14		4	2	6.11	1.89
M3-06	2.7.00 19:00	5.85		3.88	2	5.81	1.95
M3-06	3.7.00 9:00	4.87		3.48	2	4.83	2.13
M3-06	3.7.00 19:00	4.59		3.16	2	4.56	1.76
M3-06	6.7.00 9:00	1.27		0.76	2	1.27	0.25
M3-06	9.7.00 9:00	1.15		0.76	2	1.14	0.38
M3-06	12.7.00 9:00	1.38		2.21		1.36	0.85
M3-06	15.7.00 9:00	1.79		2.9		1.77	1.13
M3-06	18.7.00 9:00	1.36		3.99		1.31	2.68
M3-06	21.7.00 9:00	1.37		2.67		1.35	1.32
M3-06	24.7.00 9:00	2.61		5.84	5	2.12	27.08
M3-06	4.8.00 9:00	3.35	5	13.88	10	14.51	124.29
M3-06	4.8.00 19:00	3.29	5	14.12	10	14.16	127.04
M3-06	5.8.00 9:00	3.41	5	14.38	10	14.73	129.07
M3-06	5.8.00 19:00	3.42	5	14.67	10	14.72	131.98
M3-06	7.8.00 9:00	4.28	5	13.72	10	19.28	117.92
M3-06	7.8.00 19:00	4.01	5	13.93	10	17.86	121.44
M3-06	8.8.00 9:00	4.94	5	13.92	10	22.60	116.60
M3-06	8.8.00 19:00	2.46	10	16.18	10	22.09	139.71
M3-06	9.8.00 9:00	0.68	10	16.38	10	3.92	159.88
M3-06	9.8.00 19:00	2.03	10	16.04	10	17.73	142.67
M3-06	11.8.00 9:00	2.96	10	15.79	10	27.25	130.65
M3-06	11.8.00 19:00	2.54	10	15.54	10	23.02	132.38
M3-06	12.8.00 9:00	3.05	10	15.96	10	28.13	131.47
M3-06	12.8.00 19:00	2.22	10	15.46	10	19.77	134.83
M3-06	13.8.00 9:00	2.78	10	15.99	10	25.38	134.52
M3-06	13.8.00 19:00	1.4	10	15.43	10	11.43	142.87
M3-06	14.8.00 9:00	4.43	10	15.89	10	42.20	116.70
M3-06	14.8.00 19:00	3.01	10	15.83	10	27.75	130.55
M3-06	15.8.00 9:00	5.05	10	16	10	48.49	111.51
M3-06	15.8.00 19:00	3.33	10	15.94	10	30.99	128.41
M3-06	16.8.00 9:00	4.61	10	16.91	10	43.85	125.25
M3-06	16.8.00 19:00	3.28	10	16.23	10	30.43	131.87
M3-06	17.8.00 9:00	0.9	10	15.61	10	6.30	149.80
M3-06	17.8.00 19:00	0.61	10	16.2	10	3.24	158.76
M3-06	18.8.00 9:00	0.45	10	16.82	10	1.50	166.70
M3-06	18.8.00 19:00	3.68	10	16.87	10	34.38	134.32
M3-06	19.8.00 9:00	4.6	10	16.68	10	43.79	123.01
M3-06	20.8.00 19:00	6.15	10			62.63	-62.63
M3-06	21.8.00 9:00	9.38	5	3.17	50	44.85	113.65
M3-06	22.8.00 9:00	5.93	10	6.48	25	57.42	104.58
M3-06	23.8.00 9:00	4.4	10	6.41	25	41.87	118.38
M3-06	24.8.00 9:00	6.37	10	6.52	25	61.88	101.12
M3-06	25.8.00 9:00	6.38	10	6.31	25	62.08	95.67
M3-06	26.8.00 9:00	5.99	10	6.78	25	57.89	111.61
M3-06	27.8.00 9:00	0.97	10	4.17	40	6.82	159.98
M3-06	28.8.00 9:00	3.18	10	5.09	40	28.65	174.95

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-06	29.8.00 9:00	6.07	10	17.74	10	58.56	118.84
M3-06	30.8.00 9:00	5.91	10	17.69	10	56.94	119.96
M3-06	31.8.00 9:00	6.41	10	17.51	10	62.07	113.03
M3-06	1.9.00 9:00	8.96	10	17.72	10	87.99	89.21
M3-06	2.9.00 9:00	6.8	10	17.47	10	66.04	108.66
M3-06	3.9.00 9:00	3.93	10	16.17	10	37.06	124.64
M3-06	4.9.00 9:00	5.31	10	16.1	10	51.12	109.88
M3-06	5.9.00 9:00	4.96	10	18.04	10	47.20	133.20
M3-06	6.9.00 9:00	3	10	17.53	10	27.34	147.96
M3-06	7.9.00 9:00	3.17	10	17.67	10	29.04	147.66
M3-06	8.9.00 9:00	4.24	10	17.47	10	39.97	134.73
M3-06	9.9.00 9:00	3.45	10	16.87	10	32.04	136.66
M3-06	10.9.00 9:00	2.08	10	17.04	10	18.06	152.34
M3-06	11.9.00 9:00	5.45	10	16.51	10	52.47	112.63
M3-06	12.9.00 9:00	5.04	10	16.09	10	48.37	112.53
M3-06	13.9.00 9:00	4.63	10	15.98	10	44.22	115.58
M3-06	14.9.00 9:00	3.74	10	15.77	10	35.19	122.51
M3-06	21.9.00 9:00	4.41	10	14.97	10	42.16	107.54
M3-06	7.10.00 9:00	7.87		4.17	10	7.25	34.45
M3-06	14.10.00 9:00	5.96	5	5.69	25	27.74	114.51
M3-06	21.10.00 9:00	5.91	5	5.69	25	27.48	114.77
M3-06	28.10.00 9:00	7.38	5	3.55	25	35.95	52.80
M3-06	8.11.00 9:00	3.73	5	2.89	25	17.67	54.58
M3-07	27.6.00 19:00	0.14		12.67		0.00	12.76
M3-07	28.6.00 19:00	0.13		12.98		0.00	13.09
M3-07	29.6.00 7:00	0.09		12.16		0.00	12.29
M3-07	29.6.00 15:00	0.13		11.79		0.00	11.87
M3-07	29.6.00 23:00	0.13		11.46		0.00	11.54
M3-07	30.6.00 7:00	0.13		11.32		0.00	11.40
M3-07	30.6.00 23:00	7.86		10.45		7.81	2.64
M3-07	1.7.00 7:00	7.74		4.7	2	7.71	1.69
M3-07	1.7.00 15:00	6.1		4.67	2	6.04	3.30
M3-07	1.7.00 23:00	6.36		4.39	2	6.32	2.46
M3-07	2.7.00 9:00	5.75		4.51	2	5.69	3.33
M3-07	2.7.00 19:00	5.66		4.28	2	5.61	2.95
M3-07	3.7.00 9:00	5.07		4.36	2	5.00	3.72
M3-07	3.7.00 19:00	4.52		3.86	2	4.46	3.26
M3-07	6.7.00 9:00	0.59		0.36	2	0.59	0.13
M3-07	9.7.00 9:00	2.61		1.66	2	2.60	0.72
M3-07	12.7.00 9:00	1.35		1.56		1.35	0.21
M3-07	15.7.00 9:00	0.66		1.36		0.65	0.71
M3-07	18.7.00 9:00	0.75		1.61		0.73	0.88
M3-07	21.7.00 9:00	0.81		1.96		0.79	1.17
M3-07	24.7.00 9:00	1.37		2.69		1.35	1.34
M3-07	19.8.00 9:00	12.74		1.56	10	12.69	2.91
M3-07	20.8.00 9:00	2.23	5	2.04	10	10.98	9.42
M3-07	22.8.00 9:00	9.82		1.9	10	9.65	9.35
M3-07	23.8.00 9:00	10.3		1.85	10	10.15	8.35
M3-07	24.8.00 9:00	11.69		2	10	11.54	8.46
M3-07	25.8.00 9:00	13.23		2.01	10	13.10	7.00
M3-07	26.8.00 9:00	14.06		2.18	10	13.92	7.88

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M3-07	29.8.00 9:00	13.73		3.2	10	13.40	18.60
M3-07	30.8.00 9:00	16.17		3.27	10	15.87	16.83
M3-07	31.8.00 9:00	16.72		3.14	10	16.45	14.95
M3-07	1.9.00 9:00	12.73		3.62	10	12.30	23.90
M3-07	2.9.00 9:00	2.11	10	3.65	10	20.82	15.68
M3-07	3.9.00 9:00	16.03		3.94	10	15.60	23.80
M3-07	4.9.00 9:00	2.3	10	4.39	10	22.62	21.28
M3-07	6.9.00 9:00	2.53	5	4.43	10	12.07	32.23
M3-07	7.9.00 9:00	3.53	5	4.64	10	17.12	29.28
M3-07	8.9.00 9:00	3.76	5	4.97	10	18.23	31.47
M3-07	9.9.00 9:00	2.47	5	5.16	10	11.63	39.97
M3-07	10.9.00 9:00	2.67	5	5.37	10	12.61	41.09
M3-07	11.9.00 9:00	3.93	5	5.92	10	18.92	40.28
M3-07	12.9.00 9:00	4.29	5	6.12	10	20.72	40.48
M3-07	13.9.00 9:00	5.23	5	6.61	10	25.42	40.68
M3-07	14.9.00 9:00	5.09	5	6.54	10	24.72	40.68
M3-07	21.9.00 9:00	3.76	5	7.59	10	17.75	58.15
M3-07	7.10.00 9:00	2.06	10	8.59	10	19.40	66.50
M3-07	14.10.00 9:00	5.3	5	10.24	10	25.11	77.29
M3-07	21.10.00 9:00	5.1	5	9.49	10	24.23	70.67
M3-07	28.10.00 9:00	5.88	5	7.28	10	28.60	44.20
M3-07	8.11.00 9:00	5.36	5	6.24	10	26.15	36.25
	-			-	-		
M3-08	27.6.00 19:00	0.16		11.13		0.00	11.17
M3-08	19.8.00 9:00	10.49		10.49		10.49	0.00
M3-08	20.8.00 9:00	1.69	5	9.69		8.43	1.26
M3-08	21.8.00 9:00	5.12		9.29		5.04	4.25
M3-08	22.8.00 9:00	5.74		15.89		5.55	10.34
M3-08	23.8.00 9:00	8.33		8.8		8.32	0.48
M3-08	24.8.00 9:00	8.29		8.95		8.28	0.67
M3-08	25.8.00 9:00	9.35		8.91		9.36	0.00
M3-08	26.8.00 9:00	9.33		9.08		9.33	0.00
M3-08	27.8.00 9:00	0.03	10	5.05	2	0.12	9.98
M3-08	28.8.00 9:00	0.17	10	5.31	2	1.54	9.08
M3-08	29.8.00 9:00	8.97		10.07		8.95	1.12
M3-08	30.8.00 9:00	9.83		10.15		9.82	0.33
M3-08	31.8.00 9:00	9.44		10.54		9.42	1.12
M3-08	1.9.00 9:00	9.01		9.91		8.99	0.92
M3-08	2.9.00 9:00	9.39		9.97		9.38	0.59
M3-08	3.9.00 9:00	9.28		9.97		9.27	0.70
M3-08	4.9.00 9:00	2.97		9.79		2.84	6.95
M3-08	5.9.00 9:00	9.43		10.63		9.41	1.22
M3-08	6.9.00 9:00	1.3	5	9.94		6.44	3.50
M3-08	7.9.00 9:00	1.69	5	10.77		8.41	2.36
M3-08	8.9.00 9:00	1.5	5	9.81		7.46	2.35
M3-08	9.9.00 9:00	1.51	5	10.53		7.50	3.03
M3-08	10.9.00 9:00	1.51	5	10.13		7.50	2.63
M3-08	21.9.00 9:00	1.87	5	10.14		9.34	0.80
M3-08	7.10.00 9:00	9.21		0.9	10	9.21	0.00
M3-08	28.10.00 9:00	10.63		10.98		10.62	0.36
M3-08	8.11.00 9:00	10.59		10.34		10.59	0.00

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.
M4 02	24.7.00 9:00	11.34		12.06		11.33
M4 02	19.8.00 9:00	11.48		11.45		11.48
M4 02	20.8.00 9:00	12.32		11.97		12.33
M4 02	21.8.00 9:00	9.28		5.88	2	9.23
M4 02	22.8.00 9:00	9.96		12.48		9.91
M4 02	23.8.00 9:00	11.06		12.69		11.03
M4 02	24.8.00 9:00	9.93		12.87		9.87
M4 02	25.8.00 9:00	10.65		12.68		10.61
M4 02	26.8.00 9:00	11.14		12.96		11.10
M4 02	27.8.00 9:00	9.61		6.02	2	9.56
M4 02	28.8.00 9:00	10.94		6.01	2	10.92
M4 02	8.11.00 9:00	12.09		11.31		12.11
M4 03	24.7.00 9:00	10.68		10.68		10.68
M4 03	19.8.00 9:00	10.39		10.33		10.39
M4 03	20.8.00 9:00	10.81		10.9		10.81
M4 03	21.8.00 9:00	7.32		5.11	2	7.26
M4 03	22.8.00 9:00	9.33		11.39		9.29
M4 03	23.8.00 9:00	10.1		11.34		10.07
M4 03	24.8.00 9:00	9.4		11.24		9.36
M4 03	25.8.00 9:00	10.08		11.13		10.06
M4 03	26.8.00 9:00	10.21		11.57		10.18
M4 03	27.8.00 9:00	7.99		5.35	2	7.93
M4 03	28.8.00 9:00	9.58		5.13	2	9.57

As(V) cal.

0.73 0.00 2.53 2.57 1.66 3.00 2.07 1.86 2.48 1.10 0.00

0.00 0.09 2.96 2.10 1.27 1.88 1.07 1.39 2.77 0.69

0.00

0.62

10.31

10.80

## Tab.: A3 arsenic concentration from M4

M4 03

M4 03

14.9.00 9:00

8.11.00 9:00

10.3

10.81

M4 04	24.7.00 9:00	10.41	10.82		10.40	0.42
M4 04	19.8.00 9:00	10.46	10.66		10.46	0.20
M4 04	20.8.00 9:00	11.87	11.05		11.89	0.00
M4 04	21.8.00 9:00	7.11	4.71	2	7.06	2.36
M4 04	22.8.00 9:00	9.28	11		9.24	1.76
M4 04	23.8.00 9:00	9.89	10.94		9.87	1.07
M4 04	24.8.00 9:00	9.47	11.19		9.43	1.76
M4 04	25.8.00 9:00	9.93	10.88		9.91	0.97
M4 04	26.8.00 9:00	9.96	10.77		9.94	0.83
M4 04	27.8.00 9:00	8.27	5.13	2	8.23	2.03
M4 04	28.8.00 9:00	9.98	5.54	2	9.96	1.12
M4 04	14.9.00 9:00	10.6	10.42		10.60	0.00
M4 04	8.11.00 9:00	10.73	10.63		10.73	0.00

9.91

11.42

M4 05	19.8.00 9:00	10.6	10.65		10.60	0.05
M4 05	20.8.00 9:00	10.25	10.67		10.24	0.43
M4 05	21.8.00 9:00	7.31	5.44	2	7.24	3.64
M4 05	22.8.00 9:00	9.23	11.22		9.19	2.03
M4 05	23.8.00 9:00	9.67	12.33		9.62	2.71

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M4 05	24.8.00 9:00	9.14		10.86		9.10	1.76
M4 05	25.8.00 9:00	9.41		11		9.38	1.62
M4 05	26.8.00 9:00	9.81		10.79		9.79	1.00
M4 05	27.8.00 9:00	8.38		5.48	2	8.33	2.63
M4 05	28.8.00 9:00	10.02		5.55	2	10.00	1.10
M4 05	14.9.00 9:00	9.95		10.2		9.94	0.26
M4 05	8.11.00 9:00	11.33		10.28		11.35	0.00

M4 06	19.8.00 9:00	10.67	11.11		10.66	0.45
M4 06	20.8.00 9:00	11.09	10.78		11.10	0.00
M4 06	21.8.00 9:00	6.78	6.07	2	6.67	5.47
M4 06	22.8.00 9:00	8.8	11.14		8.75	2.39
M4 06	23.8.00 9:00	9.44	10.89		9.41	1.48
M4 06	24.8.00 9:00	8.68	10.75		8.64	2.11
M4 06	25.8.00 9:00	9.25	11.12		9.21	1.91
M4 06	26.8.00 9:00	9.84	10.78		9.82	0.96
M4 06	27.8.00 9:00	8.06	5.45	2	8.00	2.90
M4 06	28.8.00 9:00	9.34	5.78	2	9.29	2.27
M4 06	14.9.00 9:00	10.18	10.56		10.17	0.39

M4 07	19.8.00 9:00	11.42	11.12		11.40	0.00
M4 07	20.8.00 9:00	11.89	11.38		11.92	0.00
M4 07	21.8.00 9:00	7.56	5.53	2	6.63	4.43
M4 07	22.8.00 9:00	9.27	11.5		8.64	2.86
M4 07	23.8.00 9:00	10.16	11.49		9.75	1.74
M4 07	24.8.00 9:00	9.4	11.14		8.89	2.25
M4 07	25.8.00 9:00	9.52	11.1		9.05	2.05
M4 07	26.8.00 9:00	9.94	11.2		9.55	1.65
M4 07	27.8.00 9:00	9.13	5.87	2	8.41	3.33
M4 07	28.8.00 9:00	9.99	5.98	2	9.42	2.54

M4 08	19.8.00 9:00	10.37	10.59		10.23	0.36
M4 08	20.8.00 9:00	11.1	11.15		11.00	0.15
M4 08	21.8.00 9:00	7.66	5.23	2	6.90	3.56
M4 08	22.8.00 9:00	7.79	10.71		7.00	3.71
M4 08	23.8.00 9:00	9.8	11.14		9.39	1.75
M4 08	24.8.00 9:00	9.19	11		8.67	2.33
M4 08	25.8.00 9:00	9.59	10.7		9.24	1.46
M4 08	26.8.00 9:00	9.89	10.86		9.57	1.29
M4 08	27.8.00 9:00	8.32	5.57	2	7.55	3.59
M4 08	28.8.00 9:00	10.01	5.07	2	9.90	0.24

Tab.: A	A4 arsei	nic conc	entration	from	M5
10011			ontration		

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M5-01	1.7.00 15:00	11.33		5.86	2	11.32	0.40
M5-01	1.7.00 23:00	10.9		5.74	2	10.89	0.59
M5-01	2.7.00 9:00	7.09		3.79	2	7.08	0.50
M5-01	2.7.00 19:00	12.41		6.01	2	12.42	0.00
M5-01	3.7.00 9:00	11.93		6.07	2	11.93	0.21
M5-02	1.7.00 7:00	9.27		6.07	2	9.22	2.92
M5-02	1.7.00 15:00	4.86		6.28	2	4.72	7.84
M5-02	1.7.00 23:00	9.67		6.15	2	9.62	2.68
M5-02	2.7.00 9:00	5.06		3.55	2	5.02	2.08
M5-02	2.7.00 19:00	9.17		6.89	2	9.09	4.69
M5-02	3.7.00 9:00	8.06		5.46	2	8.01	2.91
M5-02	4.7.00 9:00	10.49		5.93	2	10.46	1.40
M5-02	5.7.00 9:00	8.13		5.15	2	8.09	2.21
M5-02	6.7.00 9:00	7.26		4.78	2	7.22	2.34
M5-02	7.7.00 9:00	9.03		5.07	2	9.01	1.13
M5-02	8.7.00 9:00	8.53		4.99	2	8.50	1.48
M5-02	9.7.00 9:00	8.79		4.91	2	8.77	1.05
M5-02	10.7.00 9:00	8.49		4.37	2	8.49	0.25
M5-02	11.7.00 9:00	9.87		4.29	2	9.89	0.00
M5-02	12.7.00 9:00	9.12		9.49		9.11	0.38
M5-02	13.7.00 9:00	9.22		9.75		9.21	0.54
M5-02	14.7.00 9:00	9.02		9.58		9.01	0.57
M5-02	15.7.00 9:00	9.11		8.77		9.12	0.00
M5-02	16.7.00 9:00	8.57		8.95		8.56	0.39
M5-02	17.7.00 9:00	7.87		8.1		7.87	0.23
M5-02	18.7.00 9:00	7.46		7.55		7.46	0.09
M5-02	19.7.00 9:00	6.94		6.95		6.94	0.01
M5-02	20.7.00 9:00	6.37		6.16		6.37	0.00
M5-02	21.7.00 9:00	5.68		5.46		5.68	0.00
M5-02	22.7.00 9:00	4.72		4.98		4.72	0.26
M5-02	23.7.00 9:00	3.63		3.85		3.63	0.22
M5-02	24.7.00 9:00	3.67		3.64		3.67	0.00
M5-02	24.7.00 19:00	2.95		3.53		2.94	0.59
M5-02	25.7.00 9:00	2.51		3.87		2.49	1.38
M5-02	25.7.00 19:00	1.43		2.62		1.41	1.21
M5-02	26.7.00 9:00	0.96		2.38		0.93	1.45
M5-02	26.7.00 19:00	1.41		1.9		1.40	0.50
M5-02	27.7.00 9:00	1.96		2.41		1.95	0.46
M5-02	27.7.00 19:00	1.8		2.51		1.79	0.72
M5-02	28.7.00 9:00	1.31		2.16		1.29	0.87
M5-02	28.7.00 19:00	0.93		1.58		0.92	0.66
M5-02	29.7.00 9:00	1.66		2.25		1.65	0.60
M5-02	29.7.00 19:00	1.39		1.8		1.38	0.42
M5-02	30.7.00 9:00	1.29		1.73		1.28	0.45
M5-02	30.7.00 19:00	1.35		1.95		1.34	0.61
M5-02	31.7.00 9:00	1.44		2.05		1.43	0.62
M5-02	31 7 00 19 00	1 43		1 99		1 42	0.57

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M5-02	1.8.00 9:00	1.6		1.94		1.59	0.35
M5-02	1.8.00 19:00	1.72		1.8		1.72	0.08
M5-02	2.8.00 9:00	1.78		2.07		1.77	0.30
M5-02	2.8.00 19:00	2.27		2.53		2.27	0.26
M5-02	3.8.00 9:00	1.51		1.66		1.51	0.15
M5-02	3.8.00 19:00	2.23		2.35		2.23	0.12
M5-02	4.8.00 9:00	1.85		1.98		1.85	0.13
M5-02	4.8.00 19:00	1.94		1.99		1.94	0.05
M5-02	5.8.00 9:00	1.8		1.87		1.80	0.07
M5-02	5.8.00 19:00	1.8		2.2		1.79	0.41
M5-02	6.8.00 9:00	1.88		2.02		1.88	0.14
M5-02	6.8.00 19:00	1.89		2.07		1.89	0.18
M5-02	7.8.00 9:00	0.28	5	2.16		1.39	0.77
M5-02	7.8.00 19:00	0.35	5	2.37		1.74	0.63
M5-02	8.8.00 9:00	0.28	5	2.05		1.39	0.66
M5-02	8.8.00 19:00	1.85		2.12		1.85	0.27
M5-02	9.8.00 9:00	1.79		2.36		1.78	0.58
M5-02	9.8.00 19:00	1.78		2.03		1.78	0.25
M5-02	11.8.00 9:00	1.94		2.19		1.94	0.25
M5-02	11.8.00 19:00	1.8		2.04		1.80	0.24
M5-02	12.8.00 9:00	1.85		2		1.85	0.15
M5-02	12.8.00 19:00	1.8		1.93		1.80	0.13
M5-02	13.8.00 9:00	2.09		2.08		2.09	0.00
M5-02	13.8.00 19:00	1.86		1.9		1.86	0.04
M5-02	14.8.00 9:00	1.81		2.12		1.80	0.32
M5-02	14.8.00 19:00	2.38		2.62		2.38	0.24
M5-02	15.8.00 9:00	2.06		2.09		2.06	0.03
M5-02	15.8.00 19:00	2.57		2.93		2.56	0.37
M5-02	16.8.00 9:00	2.19		2.46		2.19	0.27
M5-02	16.8.00 19:00	1.94		2.35		1.93	0.42
M5-02	17.8.00 9:00	1.99		2.15		1.99	0.16
M5-02	17.8.00 19:00	2.09		2.18		2.09	0.09
M5-02	18.8.00 9:00	2.06		2.06		2.06	0.00
M5-02	18.8.00 19:00	2.1		2.06		2.10	0.00
M5-02	19.8.00 9:00	1.67		2.03		1.66	0.37
M5-02	20.8.00 9:00	1.92		2		1.92	0.08
M5-02	21.8.00 9:00	1.15		0.98	2	1.14	0.82
M5-02	22.8.00 9:00	1.68		2.18		1.67	0.51
M5-02	23.8.00 9:00	2.1		2.4		2.09	0.31
M5-02	24.8.00 9:00	1.44		2.41		1.42	0.99
M5-02	25.8.00 9:00	2.13		2.43		2.12	0.31
M5-02	26.8.00 9:00	2.25		2.72		2.24	0.48
M5-02	27.8.00 9:00	2.08		1.24	2	2.07	0.41
M5-02	28.8.00 9:00	2.17		1.19	2	2.17	0.21
M5-02	29.8.00 9:00	2.7		2.84		2.70	0.14
M5-02	30.8.00 9:00	2.57		3.18		2.56	0.62
M5-02	31.8.00 9:00	2.68		3.05		2.67	0.38
M5-02	1.9.00 9:00	2.97		3.3		2.96	0.34
M5-02	2.9.00 9:00	3.17		4.78		3.14	1.64
M5-02	3.9.00 9:00	3.1		3.6		3.09	0.51
M5-02	4.9.00 9:00	3.68		3.95		3.68	0.27
M5-02	5.9.00 9:00	3.36		4.59		3.34	1.25
M5-02	6.9.00 9:00	2.75		4.28		2.72	1.56

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M5-02	7.9.00 9:00	3.5		4.54		3.48	1.06
M5-02	8.9.00 9:00	3.58		4.91		3.56	1.35
M5-02	9.9.00 9:00	3.69		5.1		3.66	1.44
M5-02	10.9.00 9:00	3.89		5.32		3.86	1.46
M5-02	11.9.00 9:00	3.99		5.34		3.97	1.37
M5-02	12.9.00 9:00	4.44		5.48		4.42	1.06
M5-02	13.9.00 9:00	5.09		5.8		5.08	0.72
M5-02	14.9.00 9:00	4.82		5.57		4.81	0.76
M5-02	21.9.00 9:00	6.79		9.65		6.74	2.91
M5-02	30.9.00 9:00	9		24.14		8.72	15.42
M5-02	7.10.00 9:00	12.81		3.78	10	12.35	25.45
M5-02	14.10.00 9:00	2.96	5	6.09	10	13.95	46.95
M5-02	21.10.00 9:00	5.07	5	7.28	10	24.48	48.32
M5-02	28.10.00 9:00	4.19	5	6.44	10	20.15	44.25
M5-02	8.11.00 9:00	4.34	5	6.75	10	20.86	46.64
M5-03	1.7.00 7:00	11.57		6.44	2	11.55	1.33
M5-03	1.7.00 15:00	10.43		6.6	2	10.38	2.82
M5-03	1.7.00 23:00	10.43		6.49	2	10.38	2.60
M5-03	2.7.00 9:00	8.57		6.69	2	8.48	4.90
M5-03	2.7.00 19:00	11.33		6.34	2	11.31	1.37
M5-03	3.7.00 9:00	11.75		5.99	2	11.75	0.23
M5-03	4.7.00 9:00	9.21		5.28	2	9.19	1.37
M5-03	5.7.00 9:00	6.6		4.85	2	6.54	3.16
M5-03	6.7.00 9:00	6.27		4.13	2	6.23	2.03
M5-03	7.7.00 9:00	9.05		4.19	2	9.06	0.00
M5-03	8.7.00 9:00	5.25		3.95	2	5.20	2.70
M5-03	9.7.00 9:00	7.21		3.63	2	7.21	0.05
M5-03	10.7.00 9:00	6.8		3.37	2	6.80	0.00
M5-03	11.7.00 9:00	7.56		3.65	2	7.56	0.00
M5-03	12.7.00 9:00	7.05		7.54		7.04	0.50
M5-03	13.7.00 9:00	6.36		7.08		6.35	0.73
M5-03	14.7.00 9:00	6.89		7.24		6.88	0.36
M5-03	15.7.00 9:00	5.96		5.8		5.96	0.00
M5-03	16.7.00 9:00	4.35		4.41		4.35	0.06
M5-03	17.7.00 9:00	2.26		2.42		2.26	0.16
M5-03	18.7.00 9:00	1.4		1.54		1.40	0.14
M5-03	19.7.00 9:00	1.47		1.72		1.47	0.25
M5-03	20.7.00 9:00	1.63		1.88		1.63	0.25
M5-03	21.7.00 9:00	1.12		1.49		1.11	0.38
M5-03	22.7.00 9:00	0.53		1.28		0.52	0.76
M5-03	23.7.00 9:00	0.98		1.7		0.97	0.73
M5-03	24.7.00 9:00	1.46		1.86		1.45	0.41
M5-03	24.7.00 19:00	1.08		1.71		1.07	0.64
M5-03	25.7.00 9:00	1.08		1.64		1.07	0.57
M5-03	25.7.00 19:00	0.8		2.23		0.77	1.46
M5-03	26.7.00 9:00	0.64		1.46		0.62	0.84
M5-03	26.7.00 19:00	0.93		1.58		0.92	0.66
M5-03	27.7.00 9:00	1.02		1.65		1.01	0.64
M5-03	27.7.00 19:00	0.99		1./5		0.98	0.77
M5-03	28.7.00 9:00	0.91		1.71		0.90	0.81
M5-03	28.7.00 19:00	0.85		1.57		0.84	0.73

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M5-03	29.7.00 9:00	1.3		1.85		1.29	0.56
M5-03	29.7.00 19:00	0.92		1.57		0.91	0.66
M5-03	30.7.00 9:00	1.02		1.69		1.01	0.68
M5-03	30.7.00 19:00	1.06		1.59		1.05	0.54
M5-03	31.7.00 9:00	1.4		1.78		1.39	0.39
M5-03	31.7.00 19:00	1.07		1.69		1.06	0.63
M5-03	1.8.00 9:00	1.53		1.84		1.52	0.32
M5-03	1.8.00 19:00	1.5		1.77		1.50	0.27
M5-03	2.8.00 9:00	1.51		1.71		1.51	0.20
M5-03	2.8.00 19:00	1.57		1.79		1.57	0.22
M5-03	3.8.00 9:00	1.77		1.82		1.77	0.05
M5-03	3.8.00 19:00	1.49		1.88		1.48	0.40
M5-03	4.8.00 9:00	1.58		1.85		1.58	0.27
M5-03	4.8.00 19:00	1.55		1.76		1.55	0.21
M5-03	5.8.00 9:00	1.65		1.89		1.65	0.24
M5-03	5.8.00 19:00	1.51		1.84		1.50	0.34
M5-03	6.8.00 9:00	1.57		1.9		1.56	0.34
M5-03	6.8.00 19:00	1.52		1.81		1.51	0.30
M5-03	7.8.00 9:00	0.3	5	2.44		1.48	0.96
M5-03	7.8.00 19:00	0.26	5	1.82		1.29	0.53
M5-03	8.8.00 9:00	0.27	5	1.93		1.34	0.59
M5-03	8.8.00 19:00	1.74		0.14	10	1.75	0.00
M5-03	9.8.00 9:00	1.68		0.14	10	1.69	0.00
M5-03	9.8.00 19:00	1.77		0.2	10	1.77	0.23
M5-03	11.8.00 9:00	1.81		0.11	10	1.82	0.00
M5-03	11.8.00 19:00	1.96		0.2	10	1.96	0.04
M5-03	12.8.00 9:00	1.97		0.16	10	1.98	0.00
M5-03	12.8.00 19:00	2.42		0.14	10	2.44	0.00
M5-03	13.8.00 9:00	2.15		0.18	10	2.16	0.00
M5-03	13.8.00 19:00	2.26		0.17	10	2.27	0.00
M5-03	14.8.00 9:00	2.51		0.18	10	2.52	0.00
M5-03	14.8.00 19:00	2.83		0.22	10	2.84	0.00
M5-03	15.8.00 9:00	2.52		0.19	10	2.53	0.00
M5-03	15.8.00 19:00	2.91		0.34	10	2.90	0.50
M5-03	16.8.00 9:00	2.96		0.25	10	2.97	0.00
M5-03	16.8.00 19:00	2.97		0.22	10	2.98	0.00
M5-03	17.8.00 9:00	3.21		0.28	10	3.22	0.00
M5-03	17.8.00 19:00	3.34		0.28	10	3.35	0.00
M5-03	18.8.00 9:00	3.67		0.21	10	3.70	0.00
M5-03	18.8.00 19:00	3.79		0.44	10	3.78	0.62
M5-03	19.8.00 9:00	3.26		0.4	10	3.25	0.75
M5-03	20.8.00 9:00	4.1		0.97	5	4.09	0.76
M5-03	21.8.00 9:00	3.5		2.43	2	3.48	1.38
M5-03	22.8.00 9:00	4.14		1.07	5	4.12	1.23
M5-03	23.8.00 9:00	4.57		1.21	5	4.54	1.51
M5-03	24.8.00 9:00	4.24		2.37	5	4.10	7.75
M5-03	25.8.00 9:00	5.7		2.39	5	5.59	6.36
M5-03	26.8.00 9:00	5.03		3.93	5	4.76	14.89
M5-03	27.8.00 9:00	8.34		10.41	2	8.11	12.71
M5-03	28.8.00 9:00	8.12		12.48	2	7.81	17.15
M5-03	29.8.00 9:00	10.87		3.02	10	10.52	19.68
M5-03	30.8.00 9:00	13.98		4.02	10	13.50	26.70
M5-03	31.8.00 9:00	13.05		4.43	10	12.48	31.82

$\begin{array}{c c c c c c c c c c c c c c c c c c c $	name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	1.9.00 9:00	15.67		5.02	10	15.04	35.16
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	2.9.00 9:00	13.6		5.6	10	12.82	43.18
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	3.9.00 9:00	2.99	10	6.47	10	29.26	35.44
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	5.9.00 9:00	2.18	10	7.61	10	20.80	55.30
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	6.9.00 9:00	3.89	5	7.46	10	18.44	56.16
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	M5-03	7.9.00 9:00	3.31	5	7.94	10	15.40	64.00
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	8.9.00 9:00	4.69	5	8.52	10	22.32	62.88
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	9.9.00 9:00	6.53	5	8.92	10	31.61	57.59
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	10.9.00 9:00	2.77	5	9.2	10	12.42	79.58
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	11.9.00 9:00	4.51	5	9.32	10	21.25	71.95
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	12.9.00 9:00	2.29	10	9.75	10	21.53	75.97
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	13.9.00 9:00	5.13	5	10.83	10	24.13	84.17
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	14.9.00 9:00	9.84	5	10.67	10	48.15	58.55
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	21.9.00 9:00	5.16	5	12.69	10	23.95	102.95
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	30.9.00 9:00	2.07	5	14.9	10	7.81	141.19
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	7.10.00 9:00	8.22	5	16.07	10	38.91	121.79
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	M5-03	14.10.00 9:00	4.35	10	8.14	25	40.57	162.93
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	M5-03	21.10.00 9:00	2.5	10	8.23	25	21.69	184.06
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	M5-03	28.10.00 9:00	10.42	5	6.48	25	50.09	111.91
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-03	8.11.00 9:00	8.11	5	5.87	25	38.60	108.15
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	M5.04	4 7 00 7:00	5.00		E 44	0	5.40	5.00
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	M5-04	1.7.00 7:00	5.20		5.41	2	5.16	5.66
M5-04   1.7.00 2300   7.6   5.59   2   7.33   3.65     M5-04   2.7.00 9:00   8.03   5.13   2   7.99   2.27     M5-04   2.7.00 19:00   9.36   5.95   2   9.31   2.59     M5-04   3.7.00 9:00   10.46   5.31   2   10.46   0.16     M5-04   4.7.00 9:00   10.01   5.57   2   9.99   1.15     M5-04   5.7.00 9:00   7.34   4.04   2   7.33   0.75     M5-04   6.7.00 9:00   7.5   4.2   2   7.48   0.92     M5-04   8.7.00 9:00   7.5   4.2   2   7.48   0.92     M5-04   10.7.00 9:00   9.46   4.53   2   9.47   0.00     M5-04   10.7.00 9:00   9.46   4.53   2   9.47   0.00     M5-04   11.7.00 9:00   9.87   10.38   9.86   0.52     M5-04   13.7.00 9:00   9.87   10.38<	M5-04	1.7.00 15:00	0.56		5.8	2	0.36	11.24
M5-04   2.7.00   9:00   8.03   5.13   2   7.99   2.27     M5-04   2.7.00   19:00   9.36   5.95   2   9.31   2.59     M5-04   3.7.00   9:00   10.46   5.31   2   10.46   0.16     M5-04   4.7.00   9:00   10.01   5.57   2   9.99   1.15     M5-04   5.7.00   9:00   6.22   4.69   2   6.16   3.22     M5-04   6.7.00   9:00   7.5   4.2   2   7.48   0.92     M5-04   8.7.00   9:00   7.5   4.2   2   7.48   0.92     M5-04   9.7.00   9:00   4.98   4.16   2   4.92   3.40     M5-04   10.7.00   9:00   6.05   2.88   2   6.06   0.00     M5-04   11.7.00   9:00   9.87   10.38   9.86   0.52     M5-04   13.7.00   9:00   9.87   <	M5-04	1.7.00 23:00	7.6		5.59	2	7.53	3.65
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M5-04	2.7.00 9:00	8.03		5.13	2	7.99	2.27
M5-04   3.7.00 9:00   10.46   5.31   2   10.46   0.16     M5-04   4.7.00 9:00   10.01   5.57   2   9.99   1.15     M5-04   5.7.00 9:00   6.22   4.69   2   6.16   3.22     M5-04   6.7.00 9:00   7.34   4.04   2   7.33   0.75     M5-04   7.7.00 9:00   7.5   4.2   2   7.48   0.92     M5-04   8.7.00 9:00   9.46   4.53   2   9.47   0.00     M5-04   10.7.00 9:00   6.05   2.88   2   6.06   0.00     M5-04   11.7.00 9:00   10.12   4.84   2   10.13   0.00     M5-04   12.7.00 9:00   9.87   10.38   9.86   0.52     M5-04   13.7.00 9:00   9.83   10.31   9.82   0.49     M5-04   15.7.00 9:00   9.26   9.38   9.26   0.12     M5-04   16.7.00 9:00   8.95   8.86   8.95	NI5-04	2.7.00 19:00	9.30		5.95	2	9.31	2.59
M3-04   4.7.00 9:00   6.21   4.69   2   6.16   3.22     M5-04   6.7.00 9:00   7.34   4.04   2   7.33   0.75     M5-04   6.7.00 9:00   7.5   4.2   2   7.48   0.92     M5-04   7.7.00 9:00   7.5   4.2   2   7.48   0.92     M5-04   8.7.00 9:00   4.98   4.16   2   4.92   3.40     M5-04   9.7.00 9:00   9.46   4.53   2   9.47   0.00     M5-04   10.7.00 9:00   6.05   2.88   2   6.06   0.00     M5-04   11.7.00 9:00   9.87   10.38   9.86   0.52     M5-04   13.7.00 9:00   9.87   10.2   9.97   0.23     M5-04   15.7.00 9:00   9.26   9.38   9.26   0.12     M5-04   16.7.00 9:00   9.26   8.91   9.27   0.00     M5-04   16.7.00 9:00   8.41   8.93   8.40   0.53	IVID-04	3.7.00 9:00	10.46		5.31	2	10.46	0.16
M3-04   3.7.00   9.00   7.32   4.09   2   6.16   3.22     M5-04   6.7.00   9:00   7.34   4.04   2   7.33   0.75     M5-04   7.7.00   9:00   7.5   4.2   2   7.48   0.92     M5-04   8.7.00   9:00   4.98   4.16   2   4.92   3.40     M5-04   9.7.00   9:00   6.05   2.88   2   6.06   0.00     M5-04   10.7.00   9:00   6.05   2.88   2   6.06   0.00     M5-04   11.7.00   9:00   10.12   4.84   2   10.13   0.00     M5-04   12.7.00   9:00   9.87   10.2   9.97   0.23     M5-04   13.7.00   9:00   9.26   9.38   9.26   0.12     M5-04   16.7.00   9:00   9.26   8.91   9.27   0.00     M5-04   16.7.00   9:00   8.79   8.86   8.79	IVI3-04	4.7.00 9.00	6.22		0.07	2	9.99	1.15
M6-04   6.7.00 9.00   7.34   4.04   2   7.33   0.73     M5-04   7.7.00 9:00   7.5   4.2   2   7.48   0.92     M5-04   8.7.00 9:00   4.98   4.16   2   4.92   3.40     M5-04   9.7.00 9:00   9.46   4.53   2   9.47   0.00     M5-04   10.7.00 9:00   6.05   2.88   2   6.06   0.00     M5-04   11.7.00 9:00   10.12   4.84   2   10.13   0.00     M5-04   13.7.00 9:00   9.87   10.38   9.86   0.52     M5-04   13.7.00 9:00   9.87   10.2   9.97   0.23     M5-04   14.7.00 9:00   9.83   10.31   9.82   0.49     M5-04   15.7.00 9:00   9.26   9.38   9.26   0.12     M5-04   16.7.00 9:00   8.95   8.86   8.95   0.00     M5-04   17.7.00 9:00   8.79   8.86   0.53   M5-04   0.53	IVI3-04	5.7.00 9.00	0.22		4.09	2	0.10	3.22
M5-04   1.7.00 9.00   1.3   4.2   2   1.46   0.92     M5-04   8.7.00 9:00   4.98   4.16   2   4.92   3.40     M5-04   9.7.00 9:00   9.46   4.53   2   9.47   0.00     M5-04   10.7.00 9:00   6.05   2.88   2   6.06   0.00     M5-04   11.7.00 9:00   10.12   4.84   2   10.13   0.00     M5-04   12.7.00 9:00   9.87   10.38   9.86   0.52     M5-04   13.7.00 9:00   9.87   10.2   9.97   0.23     M5-04   14.7.00 9:00   9.83   10.31   9.82   0.49     M5-04   15.7.00 9:00   9.26   9.38   9.26   0.12     M5-04   16.7.00 9:00   8.95   8.86   8.95   0.00     M5-04   16.7.00 9:00   8.95   8.86   8.79   0.07     M5-04   19.7.00 9:00   8.79   8.86   8.79   0.00     M5-04 <td>M5 04</td> <td>6.7.00 9.00 7 7 00 0:00</td> <td>7.34</td> <td></td> <td>4.04</td> <td>2</td> <td>7.33</td> <td>0.75</td>	M5 04	6.7.00 9.00 7 7 00 0:00	7.34		4.04	2	7.33	0.75
M5-04   8.7.00 9:00   9.46   4.16   2   4.92   3.40     M5-04   9.7.00 9:00   9.46   4.53   2   9.47   0.00     M5-04   10.7.00 9:00   6.05   2.88   2   6.06   0.00     M5-04   117.00 9:00   10.12   4.84   2   10.13   0.00     M5-04   12.7.00 9:00   9.87   10.38   9.86   0.52     M5-04   13.7.00 9:00   9.97   10.2   9.97   0.23     M5-04   14.7.00 9:00   9.83   10.31   9.82   0.49     M5-04   15.7.00 9:00   9.26   9.38   9.26   0.12     M5-04   16.7.00 9:00   9.26   8.91   9.27   0.00     M5-04   16.7.00 9:00   8.95   8.86   8.95   0.00     M5-04   18.7.00 9:00   8.25   7.74   8.26   0.00     M5-04   19.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   21.	IVI3-04	7.7.00 9.00 9.7.00 0:00	7.5 4.09		4.2	2	7.48	0.92
MS-04   9.7.00 9.00   9.46   4.33   2   9.47   0.00     M5-04   10.7.00 9:00   6.05   2.88   2   6.06   0.00     M5-04   11.7.00 9:00   10.12   4.84   2   10.13   0.00     M5-04   12.7.00 9:00   9.87   10.38   9.86   0.52     M5-04   13.7.00 9:00   9.97   10.2   9.97   0.23     M5-04   14.7.00 9:00   9.83   10.31   9.82   0.49     M5-04   14.7.00 9:00   9.26   9.38   9.26   0.12     M5-04   16.7.00 9:00   9.26   8.91   9.27   0.00     M5-04   16.7.00 9:00   8.95   8.86   8.95   0.07     M5-04   18.7.00 9:00   8.79   8.86   0.53   0.53     M5-04   19.7.00 9:00   8.25   7.74   8.26   0.00     M5-04   20.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   21.7.00 9:00	M5 04	0.7.00 9.00	4.90		4.10	2	4.92	3.40
MS-04   10.7.00 9.00   0.03   2.88   2   0.06   0.00     M5-04   11.7.00 9:00   10.12   4.84   2   10.13   0.00     M5-04   12.7.00 9:00   9.87   10.38   9.86   0.52     M5-04   13.7.00 9:00   9.97   10.2   9.97   0.23     M5-04   14.7.00 9:00   9.83   10.31   9.82   0.49     M5-04   15.7.00 9:00   9.26   9.38   9.26   0.12     M5-04   16.7.00 9:00   9.26   8.91   9.27   0.00     M5-04   16.7.00 9:00   8.95   8.86   8.95   0.00     M5-04   17.7.00 9:00   8.79   8.86   8.79   0.07     M5-04   19.7.00 9:00   8.25   7.74   8.26   0.00     M5-04   20.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   21.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   23.7.00 9:00   5.8	M5-04	9.7.00 9.00	9.40		4.00	2	9.47	0.00
MS-04 11.7.00 9.00 9.87 10.38 9.86 0.52   M5-04 12.7.00 9:00 9.87 10.2 9.97 0.23   M5-04 13.7.00 9:00 9.83 10.31 9.82 0.49   M5-04 14.7.00 9:00 9.83 10.31 9.82 0.49   M5-04 15.7.00 9:00 9.26 9.38 9.26 0.12   M5-04 16.7.00 9:00 9.26 8.91 9.27 0.00   M5-04 16.7.00 9:00 8.95 8.86 8.95 0.00   M5-04 17.7.00 9:00 8.95 8.86 8.95 0.00   M5-04 19.7.00 9:00 8.79 8.86 8.79 0.07   M5-04 19.7.00 9:00 8.25 7.74 8.26 0.00   M5-04 21.7.00 9:00 7.99 7.46 8.00 0.00   M5-04 23.7.00 9:00 7.09 6.93 7.09 0.00   M5-04 24.7.00 9:00 5.8 5.65 5.80 0.00   M5-04 24.7.00 9:00 5.03 5.46	M5 04	11.7.00 9.00	0.00		2.00	2	0.00	0.00
M3-04 12.7.00 9.00 9.97 10.30 9.97 0.32   M5-04 13.7.00 9:00 9.97 10.2 9.97 0.23   M5-04 14.7.00 9:00 9.83 10.31 9.82 0.49   M5-04 15.7.00 9:00 9.26 9.38 9.26 0.12   M5-04 16.7.00 9:00 9.26 8.91 9.27 0.00   M5-04 16.7.00 9:00 8.95 8.86 8.95 0.00   M5-04 17.7.00 9:00 8.95 8.86 8.95 0.00   M5-04 19.7.00 9:00 8.79 8.86 8.79 0.07   M5-04 19.7.00 9:00 8.41 8.93 8.40 0.53   M5-04 20.7.00 9:00 8.25 7.74 8.26 0.00   M5-04 21.7.00 9:00 7.99 7.46 8.00 0.00   M5-04 23.7.00 9:00 7.09 6.93 7.09 0.00   M5-04 23.7.00 9:00 5.8 5.65 5.80 0.00   M5-04 24.7.00 19:00 5.03 5.46	M5-04	12 7 00 9:00	0.87		4.04	~	0.13	0.00
M3-04   13.7.00 9.00   9.87   10.2   9.87   0.23     M5-04   14.7.00 9:00   9.83   10.31   9.82   0.49     M5-04   15.7.00 9:00   9.26   9.38   9.26   0.12     M5-04   16.7.00 9:00   9.26   8.91   9.27   0.00     M5-04   16.7.00 9:00   8.95   8.86   8.95   0.00     M5-04   17.7.00 9:00   8.79   8.86   8.95   0.00     M5-04   19.7.00 9:00   8.79   8.86   8.79   0.07     M5-04   19.7.00 9:00   8.41   8.93   8.40   0.53     M5-04   20.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   21.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   23.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   23.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 19:00   5.03   5.46   5.02<	M5 04	12.7.00 9.00	9.07		10.30		9.00	0.32
M0-04 14.7.00 9:00 9.33 10.31 3.62 0.49   M5-04 15.7.00 9:00 9.26 9.38 9.26 0.12   M5-04 16.7.00 9:00 9.26 8.91 9.27 0.00   M5-04 17.7.00 9:00 8.95 8.86 8.95 0.00   M5-04 17.7.00 9:00 8.79 8.86 8.79 0.07   M5-04 18.7.00 9:00 8.79 8.86 8.79 0.07   M5-04 19.7.00 9:00 8.25 7.74 8.26 0.00   M5-04 20.7.00 9:00 7.99 7.46 8.00 0.00   M5-04 21.7.00 9:00 7.99 7.46 8.00 0.00   M5-04 22.7.00 9:00 7.09 6.93 7.09 0.00   M5-04 23.7.00 9:00 5.8 5.65 5.80 0.00   M5-04 24.7.00 9:00 5.8 5.65 5.80 0.00   M5-04 24.7.00 9:00 5.8 5.65 5.80 0.00   M5-04 25.7.00 9:00 4.25 4.71 4.24	M5-04	14.7.00 9.00	9.97		10.2		9.97	0.23
M3-04   13.7.00 9.00   9.20   9.30   9.20   0.12     M5-04   16.7.00 9:00   9.26   8.91   9.27   0.00     M5-04   17.7.00 9:00   8.95   8.86   8.95   0.00     M5-04   17.7.00 9:00   8.79   8.86   8.95   0.00     M5-04   18.7.00 9:00   8.79   8.86   8.79   0.07     M5-04   19.7.00 9:00   8.25   7.74   8.26   0.00     M5-04   20.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   21.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   23.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   23.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 9:00   5.03   5.46   5.02   0.44     M5-04   25.7.00 9:00   3.7   4.73   3.68	M5-04	15.7.00.9:00	9.00		0.38		9.02	0.49
MS-04   10.1.00 3.00   3.20   0.31   3.21   0.00     M5-04   17.7.00 9:00   8.95   8.86   8.95   0.00     M5-04   18.7.00 9:00   8.79   8.86   8.79   0.07     M5-04   19.7.00 9:00   8.41   8.93   8.40   0.53     M5-04   20.7.00 9:00   8.25   7.74   8.26   0.00     M5-04   21.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   22.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   23.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 19:00   5.03   5.46   5.02   0.44     M5-04   25.7.00 9:00   4.25   4.71   4.24   0.47     M5-04   25.7.00 19:00   3.7   4.73   3.68   1.05     M5-04   25.7.00 19:00   3.7   4.73   3.68 <td>M5-04</td> <td>16.7.00 9.00</td> <td>9.20</td> <td></td> <td>9.50 8.01</td> <td></td> <td>9.20</td> <td>0.12</td>	M5-04	16.7.00 9.00	9.20		9.50 8.01		9.20	0.12
M3-04   17.1.00 9.00   8.93   6.00   6.93   6.00     M5-04   18.7.00 9:00   8.79   8.86   8.79   0.07     M5-04   19.7.00 9:00   8.41   8.93   8.40   0.53     M5-04   20.7.00 9:00   8.25   7.74   8.26   0.00     M5-04   21.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   22.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   23.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 9:00   5.03   5.46   5.02   0.44     M5-04   25.7.00 9:00   4.25   4.71   4.24   0.47     M5-04   25.7.00 9:00   3.7   4.73   3.68   1.05     M5-04   25.7.00 9:00   3.09   3.66   3.08   0.58	M5-04	17.7.00 9:00	8.05		8.86		9.27	0.00
MS-04   10.1.00 3.00   0.13   0.00     M5-04   19.7.00 9:00   8.41   8.93   8.40   0.53     M5-04   20.7.00 9:00   8.25   7.74   8.26   0.00     M5-04   21.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   22.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   23.7.00 9:00   6.68   6.27   6.69   0.00     M5-04   24.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 19:00   5.03   5.46   5.02   0.44     M5-04   25.7.00 9:00   4.25   4.71   4.24   0.47     M5-04   25.7.00 19:00   3.7   4.73   3.68   1.05     M5-04   25.7.00 9:00   3.09   3.66   3.08   0.58	M5-04	18 7 00 9:00	8 70		8.86		8 70	0.00
MS-04   13.7.00 3.00   0.41   0.33   0.40   0.33     M5-04   20.7.00 9:00   8.25   7.74   8.26   0.00     M5-04   21.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   22.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   23.7.00 9:00   6.68   6.27   6.69   0.00     M5-04   24.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 9:00   5.03   5.46   5.02   0.44     M5-04   25.7.00 9:00   4.25   4.71   4.24   0.47     M5-04   25.7.00 19:00   3.7   4.73   3.68   1.05     M5-04   26.7.00 9:00   3.09   3.66   3.08   0.58	M5-04	19.7.00 9.00	8/1		8.00		8.40	0.07
MS-04   20.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   21.7.00 9:00   7.99   7.46   8.00   0.00     M5-04   22.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   23.7.00 9:00   6.68   6.27   6.69   0.00     M5-04   24.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 19:00   5.03   5.46   5.02   0.44     M5-04   25.7.00 9:00   4.25   4.71   4.24   0.47     M5-04   25.7.00 19:00   3.7   4.73   3.68   1.05     M5-04   26.7.00 9:00   3.09   3.66   3.08   0.58	M5-04	20.7.00.9.00	8.25		7.74		8.26	0.00
M5-04   22.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   22.7.00 9:00   7.09   6.93   7.09   0.00     M5-04   23.7.00 9:00   6.68   6.27   6.69   0.00     M5-04   24.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 19:00   5.03   5.46   5.02   0.44     M5-04   25.7.00 9:00   4.25   4.71   4.24   0.47     M5-04   25.7.00 19:00   3.7   4.73   3.68   1.05     M5-04   26.7.00 9:00   3.09   3.66   3.08   0.58	M5-04	21.7.00 9.00	7 90		7.46		8 00	0.00
MS 04   22.1.00 5.00   1.00   0.00     M5-04   23.7.00 9:00   6.68   6.27   6.69   0.00     M5-04   24.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 19:00   5.03   5.46   5.02   0.44     M5-04   25.7.00 9:00   4.25   4.71   4.24   0.47     M5-04   25.7.00 19:00   3.7   4.73   3.68   1.05     M5-04   26.7.00 9:00   3.09   3.66   3.08   0.58	M5-04	22.7.00.9.00	7.09		6.93		7.09	0.00
M5-04   24.7.00 9:00   5.8   5.65   5.80   0.00     M5-04   24.7.00 19:00   5.03   5.46   5.02   0.44     M5-04   25.7.00 9:00   4.25   4.71   4.24   0.47     M5-04   25.7.00 19:00   3.7   4.73   3.68   1.05     M5-04   26.7.00 9:00   3.09   3.66   3.08   0.58	M5-04	23 7 00 0.00	6.68		6.00		6 60	0.00
M5-04   24.7.00 19:00   5.03   5.46   5.02   0.44     M5-04   25.7.00 9:00   4.25   4.71   4.24   0.47     M5-04   25.7.00 19:00   3.7   4.73   3.68   1.05     M5-04   26.7.00 9:00   3.09   3.66   3.08   0.58	M5-04	24 7 00 9.00	5.00		5.65		5.03	0.00
M5-04   25.7.00 9:00   4.25   4.71   4.24   0.47     M5-04   25.7.00 19:00   3.7   4.73   3.68   1.05     M5-04   26.7.00 9:00   3.09   3.66   3.08   0.58	M5-04	24.7.00 3.00	5.03		5.05		5.00	0.00
Mo or   Zorrio 0.00   4.20   4.71   4.24   0.47     M5-04   25.7.00 19:00   3.7   4.73   3.68   1.05     M5-04   26.7.00 9:00   3.09   3.66   3.08   0.58	M5-04	25 7 00 0.00	<i>1</i> 25		<u>⊿</u> 71		1.02	0.44
M5-04 26.7.00 9:00 3.09 3.66 3.08 0.58	M5-04	25.7.00 9.00	3.7		4.73		3.62	1.47
	M5-04	26.7.00 9:00	3.09	<u> </u>	3.66		3.08	0.58

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M5-04	26.7.00 19:00	3.25		3.76		3.24	0.52
M5-04	27.7.00 9:00	4.56		5.31		4.55	0.76
M5-04	27.7.00 19:00	3.87		4.59		3.86	0.73
M5-04	28.7.00 9:00	3.81		4.35		3.80	0.55
M5-04	28.7.00 19:00	3.95		4.86		3.93	0.93
M5-04	29.7.00 9:00	4.04		5.17		4.02	1.15
M5-04	29.7.00 19:00	3.71		4.52		3.70	0.82
M5-04	30.7.00 9:00	3.53		4.64		3.51	1.13
M5-04	30.7.00 19:00	3.37		4.15		3.36	0.79
M5-04	31.7.00 9:00	3.21		4.27		3.19	1.08
M5-04	31.7.00 19:00	2.71		3.61		2.69	0.92
M5-04	1.8.00 9:00	3.27		4.06		3.26	0.80
M5-04	1.8.00 19:00	3.07		4.07		3.05	1.02
M5-04	2.8.00 9:00	3.11		3.86		3.10	0.76
M5-04	2.8.00 19:00	3.38		4.22		3.36	0.86
M5-04	3.8.00 9:00	2.62		2.96		2.61	0.35
M5-04	3.8.00 19:00	2.11		2.75		2.10	0.65
M5-04	4.8.00 9:00	2.81		3.22		2.80	0.42
M5-04	4.8.00 19:00	2.24		2.44		2.24	0.20
M5-04	5.8.00 9:00	2.69		3.29		2.68	0.61
M5-04	5.8.00 19:00	2.64		3.02		2.63	0.39
M5-04	6.8.00 9:00	2.57		2.7		2.57	0.13
M5-04	6.8.00 19:00	2.15		2.56		2.14	0.42
M5-04	7.8.00 9:00	0.43	5	2.61		2.14	0.47
M5-04	7.8.00 19:00	0.42	5	2.51		2.09	0.42
M5-04	8.8.00 9:00	0.37	5	2.54		1.84	0.70
M5-04	8.8.00 19:00	2.21		2.6		2.20	0.40
M5-04	9.8.00 9:00	2.18		2.28		2.18	0.10
M5-04	9.8.00 19:00	2.12		2.25		2.12	0.13
M5-04	11.8.00 9:00	2.19		2.52		2.18	0.34
M5-04	11.8.00 19:00	1.95		2.28		1.94	0.34
M5-04	12.8.00 9:00	2.06		2.2		2.06	0.14
M5-04	12.8.00 19:00	2.21		2.27		2.21	0.06
M5-04	13.8.00 9:00	1.79		2.34		1.78	0.56
M5-04	13.8.00 19:00	1.99		2.23		1.99	0.24
M5-04	14.8.00 9:00	1.91		2.24		1.90	0.34
M5-04	14.8.00 19:00	2.06		2.12		2.06	0.06
M5-04	15.8.00 9:00	2.12		2.53		2.11	0.42
M5-04	15.8.00 19:00	1.96		1.93		1.96	0.00
M5-04	16.8.00 9:00	2.07		1.95		2.07	0.00
M5-04	16.8.00 19:00	2.07		1.85		2.07	0.00
M5-04	17.8.00 9:00	2.01		2.58		2.00	0.58
M5-04	17.8.00 19:00	2.01		1.82		2.01	0.00
M5-04	18.8.00 9:00	1.91		2.08		1.91	0.17
M5-04	18.8.00 19:00	1.91		2.12		1.91	0.21
M5-04	19.8.00 9:00	1.43		1.99		1.42	0.57
M5-04	20.8.00 9:00	2.11		2.17		2.11	0.06
M5-04	21.8.00 9:00	1.07		2.38		1.05	1.33
M5-04	22.8.00 9:00	1.75		2.41		1.74	0.67
M5-04	23.8.00 9:00	2.13		2.39		2.13	0.26
M5-04	24.8.00 9:00	1.75		2.37		1.74	0.63
M5-04	25.8.00 9:00	2.11		2.37		2.11	0.26
M5-04	26.8.00 9:00	2.38		2.65		2.38	0.27

name	date	As(III) [µg/I]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M5-04	27.8.00 9:00	2.47		1.02	2	2.48	0.00
M5-04	28.8.00 9:00	2.2		1.19	2	2.20	0.18
M5-04	29.8.00 9:00	2.34		3.16		2.32	0.84
M5-04	30.8.00 9:00	2.38		3.21		2.36	0.85
M5-04	31.8.00 9:00	1.92		3.26		1.90	1.36
M5-04	1.9.00 9:00	2.42		2.87		2.41	0.46
M5-04	2.9.00 9:00	2.37		2.59		2.37	0.22
M5-04	3.9.00 9:00	2.38		2.54		2.38	0.16
M5-04	4.9.00 9:00	2.65		2.9		2.65	0.25
M5-04	5.9.00 9:00	2.09		2.86		2.08	0.78
M5-04	6.9.00 9:00	1.78		2.81		1.76	1.05
M5-04	7.9.00 9:00	1.92		2.51		1.91	0.60
M5-04	8.9.00 9:00	2		2.91		1.98	0.93
M5-04	9.9.00 9:00	2.1		2.89		2.09	0.80
M5-04	10.9.00 9:00	1.95		3.66		1.92	1.74
M5-04	11.9.00 9:00	2.49		3.92		2.46	1.46
M5-04	12.9.00 9:00	2.65		3.49		2.63	0.86
M5-04	13.9.00 9:00	2.95		3.48		2.94	0.54
M5-04	14.9.00 9:00	2.99		3.33		2.98	0.35
M5-04	21.9.00 9:00	3.74		4.98		3.72	1.26
M5-04	30.9.00 9:00	6.43		11.77		6.33	5.44
M5-04	7.10.00 9:00	7.35		17.07		7.17	9.90
M5-04	14.10.00 9:00	2.08	5	2.21	10	10.19	11.91
M5-04	21.10.00 9:00	4.98	5	2.28	10	24.94	0.00
M5-04	28.10.00 9:00	2.84	5	2.58	10	13.99	11.81
M5-04	8.11.00 9:00	4.14	5	4.02	10	20.34	19.86
	4 7 00 7 00	0.50		544	0	0.57	0.05
M5-05	1.7.00 7:00	9.58		5.11	2	9.57	0.65
IVID-05	1.7.00 15:00	10.42		5.4	2	10.41	0.39
IVI5-05	1.7.00 23:00	9.1		5.08	2	9.08	1.08
IVID-05	2.7.00 9:00	1.12		4.71	2	0.97	8.45
1VID-05	2.7.00 19.00	0.1		4.00	2	0.00	9.39
IVID-05	3.7.00 9:00	0.09		2.07	2	0.00	5.35
IVID-05	15.8.00 9:00	3.59		3.53		3.59	0.00
IVID-05	15.8.00 19:00	3.34		3.44		3.34	0.10
IVI5-05	16.8.00 9:00	3.4		3.47		3.40	0.07
M5-05	16.8.00 19:00	2.93		3.27		2.92	0.35
IVI5-05	17.8.00 9:00	3.14		3.45		3.13	0.32
IVI5-05	17.8.00 19:00	3.28		<u>৩.//</u>		3.27	0.50
		3.37		3.63		3.37	0.26
IVI5-05	18.8.00 19:00	3.2		3.99		3.19	0.80
IVID-UD	19.8.00 9:00	2.40		3.79		2.44	1.35
CU-CIVI	20.8.00 9:00	3.59		3.64		3.59	0.05

3.94

3.93

3.84

3.87

4.01

3.81

1.69

1.84

3.85

2

2

21.8.00 9:00

22.8.00 9:00

23.8.00 9:00

24.8.00 9:00

25.8.00 9:00

26.8.00 9:00

27.8.00 9:00

28.8.00 9:00

29.8.00 9:00

M5-05

M5-05

M5-05

M5-05

M5-05

M5-05

M5-05

M5-05

M5-05

2.47

3.17

3.54

3.25

3.53

3.62

3.16

3.49

4.03

2.44

3.16

3.53

3.24

3.52

3.62

3.16

3.49

4.03

1.50

0.77

0.31

0.63

0.49

0.19

0.22

0.19

0.00

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M5-05	30.8.00 9:00	4.16		4.15		4.16	0.00
M5-05	31.8.00 9:00	3.95		3.72		3.95	0.00
M5-05	1.9.00 9:00	4.14		3.81		4.15	0.00
M5-05	2.9.00 9:00	3.98		3.92		3.98	0.00
M5-05	3.9.00 9:00	3.23		3.83		3.22	0.61
M5-05	4.9.00 9:00	3.41		0.72	5	3.41	0.19
M5-05	5.9.00 9:00	3.05		1.16	5	3.00	2.80
M5-05	6.9.00 9:00	2.49		3.24		2.48	0.76
M5-05	7.9.00 9:00	2.61		3.56		2.59	0.97
M5-05	8.9.00 9:00	2.84		3.43		2.83	0.60
M5-05	9.9.00 9:00	2.68		3.29		2.67	0.62
M5-05	10.9.00 9:00	2.98		3.68		2.97	0.71
M5-05	11.9.00 9:00	2.67		3.48		2.66	0.82
M5-05	12.9.00 9:00	2.78		3.55		2.77	0.78
M5-05	13.9.00 9:00	3.1		3.26		3.10	0.16
M5-05	14.9.00 9:00	3.17		3.4		3.17	0.23
M5-05	21.9.00 9:00	3.03		3.38		3.02	0.36
M5-05	30.9.00 9:00	3.43		3.93		3.42	0.51
M5-05	7.10.00 9:00	5.92		6.22		5.91	0.31
M5-05	14.10.00 9:00	9.93		11.85		9.89	1.96
M5-05	21.10.00 9:00	10.47		13.81		10.41	3.40
M5-05	28.10.00 9:00	2.44	5	2.32	5	12.21	0.00
M5-05	8.11.00 9:00	2.66	5	2.37	5	13.33	0.00
M5-06	1.7.00 7:00	0.12		5.04	2	0.00	10.14
M5-06	1.7.00 15:00	0.13		5.22	2	0.00	10.50
M5-06	1.7.00 23:00	3.21		4.97	2	3.09	6.85
M5-06	2.7.00 9:00	-0.01		4.75	2	0.00	9.68
M5-06	2.7.00 19:00	9.22		5.38	2	9.19	1.57
M5-06	3.7.00 9:00	6.05		2.63	2	6.06	0.00
M5-06	15.8.00 9:00	3.43		4.2		3.42	0.78
M5-06	15.8.00 19:00	3.25		4.22		3.23	0.99
M5-06	16.8.00 9:00	3.16		4.08		3.14	0.94
M5-06	16.8.00 19:00	3.32		3.88		3.31	0.57
M5-06	17.8.00 9:00	3.47		4.14		3.46	0.68
M5-06	17.8.00 19:00	3.19		3.7		3.18	0.52
M5-06	18.8.00 9:00	3.49		4.22		3.48	0.74
M5-06	18.8.00 19:00	3.5		4		3.49	0.51
M5-06	19.8.00 9:00	2.66		4.12		2.63	1.49
M5-06	20.8.00 9:00	3.85		4.02		3.85	0.17
M5-06	21.8.00 9:00	2.38		4.15		2.35	1.80
M5-06	22.8.00 9:00	3.13		4.19		3.11	1.08
M5-06	23.8.00 9:00	3.78		4.24		3.77	0.47
M5-06	24.8.00 9:00	3.1		4.18		3.08	1.10
M5-06	25.8.00 9:00	3.67		4.25		3.66	0.59
M5-06	26.8.00 9:00	3.64		4.18		3.63	0.55
M5-06	27.8.00 9:00	3.29		1.75	2	3.29	0.21
M5-06	28.8.00 9:00	3.52		1.78	2	3.52	0.04
M5-06	29.8.00 9:00	4.2		4.47		4.20	0.27
M5-06	30.8.00 9:00	4.49		3.95		4.50	0.00
M5-06	31.8.00 9:00	4.57		4.42		4.57	0.00
M5-06	1.9.00 9:00	4.5		4.4		4.50	0.00

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M5-06	2.9.00 9:00	4.43		3.95		4.44	0.00
M5-06	3.9.00 9:00	3.49		4.19		3.48	0.71
M5-06	4.9.00 9:00	3.61		0.73	5	3.61	0.04
M5-06	5.9.00 9:00	3.36		4.09		3.35	0.74
M5-06	6.9.00 9:00	2.67		3.75		2.65	1.10
M5-06	7.9.00 9:00	2.86		3.7		2.84	0.86
M5-06	8.9.00 9:00	2.98		3.64		2.97	0.67
M5-06	9.9.00 9:00	2.9		3.59		2.89	0.70
M5-06	10.9.00 9:00	2.56		3.24		2.55	0.69
M5-06	11.9.00 9:00	2.98		3.73		2.97	0.76
M5-06	12.9.00 9:00	3.14		3.8		3.13	0.67
M5-06	13.9.00 9:00	3.49		3.96		3.48	0.48
M5-06	14.9.00 9:00	3.32		3.72		3.31	0.41
M5-06	21.9.00 9:00	3.52		3.84		3.51	0.33
M5-06	30.9.00 9:00	4.15		4.93		4.14	0.79
M5-06	7.10.00 9:00	5.05		5.66		5.04	0.62
M5-06	14.10.00 9:00	6.65		7.99		6.63	1.36
M5-06	21.10.00 9:00	7.27		9.32		7.23	2.09
M5-06	28.10.00 9:00	9.11		8.62		9.12	0.00
M5-06	8.11.00 9:00	10.51		9.73		10.52	0.00
M5-07	1.7.00 7:00	0.6		5.21	2	0.42	10.00
M5-07	1.7.00 15:00	0.13		5.43	2	0.00	10.93
M5-07	1.7.00 23:00	5.25		5.38	2	5.15	5.61
M5-07	2.7.00 9:00	0.15		5.87	2	0.00	11.80
M5-07	2.7.00 19:00	9.94		4.64	2	9.95	0.00
M5-07	3.7.00 9:00	6.77		6.25	2	6.66	5.84
M5-07	19.8.00 9:00	1.98		3.02		1.96	1.06
M5-07	20.8.00 9:00	2.35		2.54		2.35	0.19
M5-07	21.8.00 9:00	1.25		2.7		1.22	1.48
M5-07	22.8.00 9:00	2		2.79		1.99	0.80
M5-07	23.8.00 9:00	2.38		2.8		2.37	0.43
M5-07	24.8.00 9:00	2.03		2.72		2.02	0.70
M5-07	25.8.00 9:00	2.49		2.77		2.48	0.29
M5-07	26.8.00 9:00	2.46		2.76	0	2.45	0.31
M5-07	27.8.00 9:00	2.12		1.28	2	2.11	0.45
M5-07	28.8.00 9:00	2.34		1.28	2	2.34	0.22
M5-07	29.8.00 9:00	2.95		3.08		2.95	0.13
M5-07	30.8.00 9:00	2.98		2.91		2.98	0.00
M5-07	31.8.00 9:00	3.01		3.07		3.01	0.06
M5-07	1.9.00 9:00	3.22		3.04		3.22	0.00
IVI5-07	2.9.00 9:00	3.25		3.35		3.25	0.10
M5-07	3.9.00 9.00	2.97		4.48	~	2.94	1.54
	4.9.00 9:00	2.87		0.41	C	2.89	0.00
IVI5-07	5.9.00 9:00	2.87		3.56		2.86	0.70
	0.9.00 9:00	2.47		3.31		2.45	0.00
IVI5-07	7.9.00 9:00	2.87		3.74		2.85	0.89
	0.9.00 9:00	3.10		4.03		3.14	0.03
	9.9.00 9:00	3.44		4.39		3.42	0.9/
IVID-U7		3.11		5.U3		3./3	1.20
	12.0.00.0:00	4.4		5.54 6.04		4.50	1.10
	12.9.00 9:00	4.99		0.04		4.97	1.07

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M5-07	13.9.00 9:00	5.5		6.69		5.48	1.21
M5-07	14.9.00 9:00	5.4		6.81		5.37	1.44
M5-07	21.9.00 9:00	6.68		8.27		6.65	1.62
M5-07	30.9.00 9:00	8.57		9.24		8.56	0.68
M5-07	7.10.00 9:00	8.8		9.61		8.79	0.82
M5-07	14.10.00 9:00	9.72		11.61		9.69	1.92
M5-07	21.10.00 9:00	9.25		12.09		9.20	2.89
M5-07	28.10.00 9:00	10.2		10.17		10.20	0.00
M5-07	8.11.00 9:00	11.94		10.82		11.96	0.00
145.00		0.00		5.07			
M5-08	1.7.00 15:00	9.39		5.37	2	9.37	1.37
M5-08	1.7.00 23:00	7.19		5.12	2	7.13	3.11
M5-08	2.7.00 9:00	0.02		5.1	2	0.00	10.37
M5-08	2.7.00 19:00	0.12		5.39	2	0.00	10.86
M5-08	3.7.00 9:00	0.16		5.57	2	0.00	11.18
M5-08	19.8.00 9:00	10.09		12.22		10.05	2.17
M5-08	20.8.00 9:00	11.23		11		11.23	0.00
M5-08	21.8.00 9:00	8.87		12.07		8.81	3.26
M5-08	22.8.00 9:00	9.1		11.93		9.05	2.88
M5-08	23.8.00 9:00	10.28		12.01		10.25	1.76
M5-08	24.8.00 9:00	8.19		12.23		8.12	4.11
M5-08	25.8.00 9:00	10.01		12.33		9.97	2.36
M5-08	26.8.00 9:00	10.42		12.28	-	10.39	1.89
M5-08	27.8.00 9:00	10.36		6.07	2	10.33	1.81
M5-08	28.8.00 9:00	11.39		6.32	2	11.37	1.27
M5-08	29.8.00 9:00	14.05		13.05		14.07	0.00
M5-08	30.8.00 9:00	13.84		13.31		13.85	0.00
M5-08	31.8.00 9:00	14.2		13.59		14.21	0.00
M5-08	1.9.00 9:00	14.41		14.15		14.41	0.00
M5-08	2.9.00 9:00	14.12		14.01	_	14.12	0.00
M5-08	3.9.00 9:00	12.56		2.68	5	12.54	0.86
M5-08	4.9.00 9:00	13.23		2.71	5	13.22	0.33
M5-08	5.9.00 9:00	12.15	_	2.99	5	12.10	2.85
M5-08	6.9.00 9:00	1.9	5	13.93		9.42	4.51
M5-08	7.9.00 9:00	1.9	5	13.76		9.42	4.34
M5-08	8.9.00 9:00	1.98	5	13.61		9.83	3.78
M5-08	9.9.00 9:00	1.88	5	14.08		9.31	4.77
M5-08	10.9.00 9:00	1.93	5	14.26		9.57	4.69
M5-08	11.9.00 9:00	1.98	5	13.95		9.83	4.12
M5-08	12.9.00 9:00	2.07	5	14.39		10.28	4.11
M5-08	13.9.00 9:00	2.59	5	13.9		12.93	0.97
M5-08	14.9.00 9:00	2.55	5	14.42		12.72	1.70
M5-08	21.9.00 9:00	2.6	5	15.16		12.96	2.20
M5-08	30.9.00 9:00	1.3	10	14.77		12.97	1.80
M5-08	/.10.00 9:00	13.08		14.06		13.06	1.00
M5-08	14.10.00 9:00	2.61	5	15.64		13.00	2.64
M5-08	21.10.00 9:00	2.32	5	14.91		11.54	3.37
M5-08	28.10.00 9:00	12.14		11.91		12.14	0.00
M5-08	8.11.00 9:00	10.29		9.99		10.30	0.00

M3-09 1.7.00 13.00 11.24 3.30 2 11.24 0.00
--

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M5-09	1.7.00 23:00	10.31		5.18	2	10.31	0.05
M5-09	2.7.00 9:00	8.99		5.85	2	8.94	2.76
M5-09	2.7.00 19:00	10.97		5.54	2	10.97	0.11

Tab.: A5 arsenic concentration from M6

name	date	As(III) [µg/I]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M6-02	1.7.00 7:00	6.58		6.26	2	6.47	6.05
M6-02	1.7.00 15:00	0.15		6.17	2	0.00	12.41
M6-02	1.7.00 23:00	9.51		6.28	2	9.45	3.11
M6-02	2.7.00 9:00	7.44		6.31	2	7.35	5.27
M6-02	2.7.00 19:00	8.98		6.09	2	8.92	3.26
M6-02	3.7.00 9:00	9.12		5.83	2	9.07	2.59
M6-02	4.7.00 9:00	9.33		5.47	2	9.30	1.64
M6-02	5.7.00 9:00	7.78		4.91	2	7.74	2.08
M6-02	6.7.00 9:00	5.12		3.53	2	5.08	1.98
M6-02	7.7.00 9:00	8.73		4.48	2	8.73	0.23
M6-02	8.7.00 9:00	6.9		4.68	2	6.85	2.51
M6-02	9.7.00 9:00	7.84		4.12	2	7.83	0.41
M6-02	10.7.00 9:00	7.62		4	2	7.61	0.39
M6-02	11.7.00 9:00	6.86		3.76	2	6.85	0.67
M6-02	12.7.00 9:00	5.83		8.23		5.79	2.44
M6-02	13.7.00 9:00	7.29		8.47		7.27	1.20
M6-02	14.7.00 9:00	6.72		7.73		6.70	1.03
M6-02	15.7.00 9:00	5.99		7.1		5.97	1.13
M6-02	16.7.00 9:00	5.5		6.32		5.48	0.84
M6-02	17.7.00 9:00	4.78		5.36		4.77	0.59
M6-02	18.7.00 9:00	4.02		4.46		4.01	0.45
M6-02	19.7.00 9:00	2.92		3.1		2.92	0.18
M6-02	20.7.00 9:00	2.27		2.56		2.26	0.30
M6-02	21.7.00 9:00	1.22		1.96		1.21	0.75
M6-02	22.7.00 9:00	0.51		1.21		0.50	0.71
M6-02	23.7.00 9:00	0.13		0.36		0.13	0.23
M6-02	24.7.00 9:00	0.67		1.11		0.66	0.45
M6-02	25.7.00 9:00	0.51		1.07		0.50	0.57
M6-02	26.7.00 9:00	0.37		0.97		0.36	0.61
M6-02	27.7.00 9:00	0.76		1.34		0.75	0.59
M6-02	28.7.00 9:00	0.76		1.27		0.75	0.52
M6-02	29.7.00 9:00	0.69		1.16		0.68	0.48
M6-02	30.7.00 9:00	0.92		1.29		0.91	0.38
M6-02	31.7.00 9:00	0.97		1.29		0.96	0.33
M6-02	1.8.00 9:00	1.03		1.29		1.03	0.26
M6-02	2.8.00 9:00	1.3		1.51		1.30	0.21
M6-02	3.8.00 9:00	1.31		1.62		1.30	0.32
M6-02	4.8.00 9:00	1.29		1.55		1.29	0.26
M6-02	5.8.00 9:00	1.25		1.57		1.24	0.33
M6-02	6.8.00 9:00	2.69		3.2		2.68	0.52
M6-02	7.8.00 9:00	0.2	5	1.91		0.98	0.93
M6-02	8.8.00 9:00	0.23	5	1.63		1.14	0.49
IVI6-02	9.8.00 9:00	1.47		1.66		1.47	0.19
M6-02	10.8.00 9:00	4 47		4.07		4 47	0.00
IVI6-02	11.8.00 9:00	1.47		1.67		1.4/	0.20
M6-02	12.8.00 9:00	1.54		1.64		1.54	0.10
	13.8.00 9:00	1.62		1./1		1.62	0.09
IVI0-02	14.0.00 9:00	1.04		1.03		1.04	0.19
IVI6-02	15.8.00 9:00	1.8		2.08		1./9	0.29

name	date	As(III)	dilution	As(tot)	dilution	As(III) cal	$A_{S}(V)$ cal
namo	duto	[µg/l]	dilation	[µg/l]	difation		
M6-02	16.8.00 9:00	1.85		2.06		1.85	0.21
M6-02	17.8.00 9:00	1.9		1.99		1.90	0.09
M6-02	18.8.00 9:00	1.8		2.48		1.79	0.69
M6-02	19.8.00 9:00	1.64		2.33		1.63	0.70
M6-02	20.8.00 9:00	2.17		2.72		2.16	0.56
M6-02	21.8.00 9:00	1.47		2.92		1.44	1.48
M6-02	22.8.00.9.00	2 54		3.11		2 53	0.58
M6-02	23.8.00.9.00	3.02		3 58		3.01	0.57
M6-02	24.8.00 9:00	3.01		4		2 00	1 01
M6-02	25.8.00.9:00	3.47		4 24		3.46	0.78
M6-02	26.8.00 9:00	3.8/		1.65		3.40	0.70
M6-02	20.0.00 9.00	3.88		2.00	2	3.03	0.02
M6 02	27.0.00 9.00	3.00		2.24	2	3.07	1.06
NIC-02	20.0.00 9.00	4.40		2.75	∠	4.44	1.00
NI6-02	30.8.00 9:00	0.00		8.24		0.52	1.72
NI6-02	1.9.00 9:00	0.09		11.03		6.61	4.42
IVI6-02	3.9.00 9:00	7.05		13.11		6.94	6.17
M6-02	5.9.00 9:00	5.18		17.42		4.96	12.46
M6-02	7.9.00 9:00	8.79		23.28		8.52	14.76
M6-02	9.9.00 9:00	18.67		22.85		18.59	4.26
M6-02	11.9.00 9:00	12.4		30.6	1.0	12.07	18.53
M6-02	13.9.00 9:00	11.79		4.02	10	11.27	28.93
M6-02	14.9.00 9:00	15.45		3.95	10	15.01	24.49
M6-02	21.9.00 9:00	11.16		5.04	10	10.44	39.96
M6-02	30.9.00 9:00	2.01	10	5.98	10	19.37	40.43
M6-02	7.10.00 9:00	1.96	10	6.3	10	18.80	44.20
M6-02	14.10.00 9:00	2.1	10	7.78	10	19.96	57.84
M6-02	21.10.00 9:00	3.78	5	8.1	10	17.76	63.24
M6-02	28.10.00 9:00	5.03	5	6.25	10	24.47	38.03
M6-02	8.11.00 9:00	4.28	5	6.24	10	20.65	41.75
	1					1	
M6-03	1.7.00 7:00	3.83		5.58	2	3.70	7.46
M6-03	1.7.00 15:00	0.08		5.52	2	0.00	11.16
M6-03	1.7.00 23:00	6.42		5.89	2	6.32	5.46
M6-03	2.7.00 9:00	0.07		5.99	2	0.00	12.13
M6-03	2.7.00 19:00	6.63		6.11	2	6.53	5.69
M6-03	3.7.00 9:00	6.33		6.15	2	6.22	6.08
M6-03	4.7.00 9:00	9.16		5.99	2	9.11	2.87
M6-03	5.7.00 9:00	8.9		5.74	2	8.85	2.63
M6-03	6.7.00 9:00	5.71		5.45	2	5.61	5.29
M6-03	7.7.00 9:00	9.08		5.33	2	9.05	1.61
M6-03	8.7.00 9:00	5.43		4.59	2	5.36	3.82
M6-03	9.7.00 9:00	7.52		4.97	2	7.48	2.46
M6-03	10.7.00 9:00	8.61		5.42	2	8.57	2.27
M6-03	11.7.00 9:00	7.72		4.68	2	7.69	1.67
M6-03	12.7.00 9:00	7.71		10.28		7.66	2.62
M6-03	13.7.00 9:00	8.89		10.9		8.85	2.05
M6-03	14.7.00 9:00	9.25		10.9		9.22	1.68
M6-03	15.7.00 9:00	8.71		10.37		8.68	1.69
M6-03	16.7.00 9:00	8.69		9.67		8.67	1.00
M6-03	17.7.00 9:00	8.27		9.32		8.25	1.07
M6-03	18.7.00 9:00	8.07		9.05		8.05	1.00
M6-03	19.7.00 9:00	7.9		8.61		7.89	0.72

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M6-03	20.7.00 9:00	7.63		8.04		7.62	0.42
M6-03	21.7.00 9:00	7.17		7.97		7.16	0.81
M6-03	22.7.00 9:00	6.39		7.96		6.36	1.60
M6-03	23.7.00 9:00	5.35		6.53		5.33	1.20
M6-03	24.7.00 9:00	4.79		5.67		4.77	0.90
M6-03	25.7.00 9:00	4.63		5.52		4.61	0.91
M6-03	26.7.00 9:00	3.79		4.56		3.78	0.78
M6-03	27.7.00 9:00	4.65		5.03		4.64	0.39
M6-03	28.7.00 9:00	2.92		5.13		2.88	2.25
M6-03	29.7.00 9:00	4.53		4.97		4.52	0.45
M6-03	30.7.00 9:00	2.75		4.24		2.72	1.52
M6-03	31.7.00 9:00	3.9		4.73		3.88	0.85
M6-03	1.8.00 9:00	2.8		4.17		2.77	1.40
M6-03	2.8.00 9:00	3.42		4.01		3.41	0.60
M6-03	3.8.00 9:00	3.66		4.1		3.65	0.45
M6-03	4.8.00 9:00	3.35		3.47		3.35	0.12
M6-03	5.8.00 9:00	2.9		3.26		2.89	0.37
M6-03	6.8.00 9:00	1.28		1.46		1.28	0.18
M6-03	7.8.00 9:00	0.42	5	3.2		2.08	1.12
M6-03	8.8.00 9:00	0.38	5	2.92		1.88	1.04
M6-03	9.8.00 9:00	2.86		2.92		2.86	0.06
M6-03	11.8.00 9:00	2.64		2.66		2.64	0.02
M6-03	12.8.00 9:00	2.69		2.93		2.69	0.24
M6-03	13.8.00 9:00	2.51		3.21		2.50	0.71
M6-03	14.8.00 9:00	2.44		2.68		2.44	0.24
M6-03	15.8.00 9:00	2.75		2.91		2.75	0.16
M6-03	16.8.00 9:00	2.66		2.8		2.66	0.14
M6-03	17.8.00 9:00	2.72		2.97		2.72	0.25
M6-03	18.8.00 9:00	2.38		2.83		2.37	0.46
M6-03	19.8.00 9:00	2.16		2.78		2.15	0.63
M6-03	20.8.00 9:00	2.69		3.02		2.68	0.34
M6-03	21.8.00 9:00	1.5		2.84		1.48	1.36
M6-03	22.8.00 9:00	2.25		2.66		2.24	0.42
M6-03	23.8.00 9:00	2.53		3.03		2.52	0.51
M6-03	24.8.00 9:00	2.46		2.88		2.45	0.43
M6-03	25.8.00 9:00	2.62		3.03		2.61	0.42
M6-03	26.8.00 9:00	2.59		2.98		2.58	0.40
M6-03	27.8.00 9:00	2.38		1.31	2	2.38	0.24
M6-03	28.8.00 9:00	2.43		1.26	2	2.43	0.09
M6-03	30.8.00 9:00	3.66		3.77		3.66	0.11
M6-03	1.9.00 9:00	3.6		3.41		3.60	0.00
M6-03	3.9.00 9:00	3.44		3.32		3.44	0.00
M6-03	5.9.00 9:00	2.59		3.78		2.57	1.21
M6-03	7.9.00 9:00	2.17		3.35		2.15	1.20
M6-03	9.9.00 9:00	3.09		4.11		3.07	1.04
M6-03	11.9.00 9:00	2.49		3.36		2.47	0.89
M6-03	13.9.00 9:00	2.79		3.31		2.78	0.53
M6-03	14.9.00 9:00	3.06		3.43		3.05	0.38
M6-03	21.9.00 9:00	3.83		4.32		3.82	0.50
M6-03	30.9.00 9:00	6.37		6.87		6.36	0.51
M6-03	7.10.00 9:00	7.29		8.17		7.27	0.90
M6-03	14.10.00 9:00	1.7	5	2.65	5	8.41	4.84
M6-03	21.10.00 9:00	1.81	5	3.02	5	8.94	6.16
name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
-------	---------------	-------------------	----------	-------------------	----------	--------------	------------
M6-03	28.10.00 9:00	2.12	5	2.49	5	10.57	1.88
M6-03	8.11.00 9:00	2.49	5	3.03	5	12.40	2.75
M6-04	1.7.00 7:00	9.56		5.29	2	9.54	1.04
M6-04	1.7.00 15:00	10.14		5.38	2	10.13	0.63
M6-04	1.7.00 23:00	10.35		5.31	2	10.34	0.28
M6-04	2.7.00 9:00	0.13		5.65	2	0.00	11.37
M6-04	2.7.00 19:00	0.11		5.73	2	0.00	11.56
M6-04	3.7.00 9:00	0.04		5.39	2	0.00	10.94
M6-04	4.7.00 9:00	0.14		5.27	2	0.00	10.59
M6-04	5.7.00 9:00	1.94		5.69	2	1.77	9.61
M6-04	6.7.00 9:00	7.02		6.04	2	6.93	5.15
M6-04	7.7.00 9:00	10.98		6.66	2	10.94	2.38
M6-04	8.7.00 9:00	3.58		5.48	2	3.44	7.52
M6-04	9.7.00 9:00	12.06		6.35	2	12.05	0.65
M6-04	10.7.00 9:00	9.75		6.91	2	9.68	4.14
M6-04	11.7.00 9:00	9.47		5.83	2	9.43	2.23
M6-04	12.7.00 9:00	9.59		12.64		9.53	3.11
M6-04	13.7.00 9:00	10.47		12.2		10.44	1.76
M6-04	14.7.00 9:00	10.63		12.69		10.59	2.10
M6-04	15.7.00 9:00	10.09		11.45		10.07	1.38
M6-04	16.7.00 9:00	9.49		10.92		9.46	1.46
M6-04	17.7.00 9:00	9.35		10.4		9.33	1.07
M6-04	18.7.00 9:00	9.56		10.49		9.54	0.95
M6-04	19.7.00 9:00	9.59		10.65		9.57	1.08
M6-04	20.7.00 9:00	9.68		10.59		9.66	0.93
M6-04	21.7.00 9:00	8.72		10.47		8.69	1.78
M6-04	22.7.00 9:00	8.86		10.52		8.83	1.69
M6-04	23.7.00 9:00	8.5		9.94		8.47	1.47
M6-04	24.7.00 9:00	8.33		9.91		8.30	1.61
M6-04	25.7.00 9:00	8		9.73		7.97	1.76
M6-04	26.7.00 9:00	8.19		8.98		8.18	0.80
M6-04	27.7.00 9:00	7.41		7.31		7.41	0.00
M6-04	28.7.00 9:00	6.74		8.21		6.71	1.50
M6-04	29.7.00 9:00	6.02		6.43		6.01	0.42
M6-04	30.7.00 9:00	5.3		5.78		5.29	0.49
M6-04	31.7.00 9:00	4.37		4.93		4.36	0.57
M6-04	1.8.00 9:00	3.74		4.13		3.73	0.40
M6-04	2.8.00 9:00	3.15		3.4		3.15	0.25
M6-04	3.8.00 9:00	2.32		2.56		2.32	0.24
M6-04	4.8.00 9:00	1.68		1.84		1.68	0.16
M6-04	5.8.00 9:00	1.48		1.55		1.48	0.07
M6-04	6.8.00 9:00	1.26		1.4		1.26	0.14
M6-04	7.8.00 9:00	0.19	5	1.71		0.94	0.77
M6-04	8.8.00 9:00	0.28	5	1.76		1.39	0.37
M6-04	9.8.00 9:00	1.69		1.57		1.69	0.00
M6-04	11.8.00 9:00	1.7		1.75		1.70	0.05
M6-04	12.8.00 9:00	1.72		1.83		1.72	0.11
M6-04	13.8.00 9:00	1.8		1.78		1.80	0.00
M6-04	14.8.00 9:00	1.77		1.77		1.77	0.00
M6-04	15.8.00 9:00	1.73		1.84		1.73	0.11
M6-04	16.8.00 9:00	1.81		1.8		1.81	0.00

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M6-04	17.8.00 9:00	1.78		1.95		1.78	0.17
M6-04	18.8.00 9:00	1.75		1.84		1.75	0.09
M6-04	19.8.00 9:00	1.49		1.9		1.48	0.42
M6-04	20.8.00 9:00	1.92		1.99		1.92	0.07
M6-04	21.8.00 9:00	1.12		2.2		1.10	1.10
M6-04	22.8.00 9:00	1.78		2.09		1.77	0.32
M6-04	23.8.00 9:00	1.97		2.21		1.97	0.24
M6-04	24.8.00 9:00	1.67		2.03		1.66	0.37
M6-04	25.8.00 9:00	1.84		2.02		1.84	0.18
M6-04	26.8.00 9:00	1.85		2.07		1.85	0.22
M6-04	27.8.00 9:00	1.7		0.91	2	1.70	0.12
M6-04	28.8.00 9:00	1.8		0.94	2	1.80	0.08
M6-04	30.8.00 9:00	2.28		2.26		2.28	0.00
M6-04	3.9.00 9:00	1.91		1.96		1.91	0.05
M6-04	5.9.00 9:00	1.71		2.24		1.70	0.54
M6-04	7.9.00 9:00	1.36		1.97		1.35	0.62
M6-04	9.9.00 9:00	1.46		2.08		1.45	0.63
M6-04	11.9.00 9:00	1.4		2.1		1.39	0.71
M6-04	13.9.00 9:00	1.86		2.16		1.85	0.31
M6-04	14.9.00 9:00	1.77		2.03		1.77	0.26
M6-04	21.9.00 9:00	1.85		2.15		1.84	0.31
M6-04	30.9.00 9:00	2.08		4.13		2.04	2.09
M6-04	7.10.00 9:00	6.06		6.56		6.05	0.51
M6-04	14.10.00 9:00	7		1.74	5	6.97	1.73
M6-04	21.10.00 9:00	6.95		2.01	5	6.89	3.16
M6-04	28.10.00 9:00	8.05		1.52	5	8.06	0.00
M6-04	8.11.00 9:00	9.1		1.73	5	9.11	0.00

M6 05	19.8.00 9:00	2.2	2.7		2.19	0.51
M6 05	20.8.00 9:00	2.65	2.64		2.65	0.00
M6 05	21.8.00 9:00	1.56	2.84		1.54	1.30
M6 05	22.8.00 9:00	2.37	2.78		2.36	0.42
M6 05	23.8.00 9:00	2.48	2.92		2.47	0.45
M6 05	24.8.00 9:00	2.27	2.81		2.26	0.55
M6 05	25.8.00 9:00	2.51	2.76		2.51	0.25
M6 05	26.8.00 9:00	2.44	2.93		2.43	0.50
M6 05	27.8.00 9:00	2.29	1.11	2	2.29	0.00
M6 05	28.8.00 9:00	2.46	1.74	2	2.44	1.04
M6 05	30.8.00 9:00	2.97	3.34		2.96	0.38
M6 05	1.9.00 9:00	2.99	3.11		2.99	0.12
M6 05	3.9.00 9:00	2.54	2.52		2.54	0.00
M6 05	5.9.00 9:00	2.17	2.85		2.16	0.69
M6 05	7.9.00 9:00	1.79	2.65		1.77	0.88
M6 05	9.9.00 9:00	8.93	10.47		8.90	1.57
M6 05	11.9.00 9:00	1.91	2.7		1.90	0.80
M6 05	13.9.00 9:00	2.23	2.56		2.22	0.34
M6 05	14.9.00 9:00	2.26	2.51		2.26	0.25
M6 05	21.9.00 9:00	2.33	2.7		2.32	0.38
M6 05	30.9.00 9:00	2.53	2.89		2.52	0.37
M6 05	7.10.00 9:00	3.51	5.66		3.47	2.19
M6 05	14.10.00 9:00	4.82	1.14	5	4.80	0.90
M6 05	21.10.00 9:00	6.43	1.77	5	6.39	2.46

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M6 05	28.10.00 9:00	7.4		7.74		7.39	0.35
M6 05	8.11.00 9:00	8.21		8.53		8.20	0.33

-						
M6 06	19.8.00 9:00	5.05	5.57		5.04	0.53
M6 06	20.8.00 9:00	5.46	5.69		5.46	0.23
M6 06	21.8.00 9:00	4.65	5.93		4.63	1.30
M6 06	22.8.00 9:00	4.63	6.04		4.60	1.44
M6 06	23.8.00 9:00	5.43	6.24		5.42	0.82
M6 06	24.8.00 9:00	5.38	5.95		5.37	0.58
M6 06	25.8.00 9:00	5.31	6.06		5.30	0.76
M6 06	26.8.00 9:00	5.49	6.14		5.48	0.66
M6 06	27.8.00 9:00	5	2.78	2	4.99	0.57
M6 06	28.8.00 9:00	5.52	2.84	2	5.52	0.16
M6 06	30.8.00 9:00	6.29	6.46		6.29	0.17
M6 06	1.9.00 9:00	6.38	6.85		6.37	0.48
M6 06	3.9.00 9:00	5.7	5.53		5.70	0.00
M6 06	5.9.00 9:00	4.78	6.22		4.75	1.47
M6 06	7.9.00 9:00	4.45	5.78		4.43	1.35
M6 06	9.9.00 9:00	9	10.82		8.97	1.85
M6 06	11.9.00 9:00	4.7	6		4.68	1.32
M6 06	13.9.00 9:00	5.34	5.86		5.33	0.53
M6 06	14.9.00 9:00	5.29	5.82		5.28	0.54
M6 06	21.9.00 9:00	5.63	6.09		5.62	0.47
M6 06	30.9.00 9:00	6.35	6.99		6.34	0.65
M6 06	7.10.00 9:00	7.14	7.28		7.14	0.14
M6 06	14.10.00 9:00	7.89	1.96	5	7.85	1.95
M6 06	21.10.00 9:00	7.55	2.07	5	7.50	2.85
M6 06	28.10.00 9:00	8.21	8.48		8.21	0.27
M6 06	8.11.00 9:00	9.03	8.97		9.03	0.00

M6 07	19.8.00 9:00	9.53	10.74		9.51	1.23
M6 07	20.8.00 9:00	10.05	10.95		10.03	0.92
M6 07	21.8.00 9:00	10.28	12		10.25	1.75
M6 07	22.8.00 9:00	7.08	11.85		6.99	4.86
M6 07	23.8.00 9:00	9.99	11.49		9.96	1.53
M6 07	24.8.00 9:00	9.78	11.76		9.74	2.02
M6 07	25.8.00 9:00	10.2	11.72		10.17	1.55
M6 07	26.8.00 9:00	9.98	11.45		9.95	1.50
M6 07	27.8.00 9:00	10.51	5.45	2	10.50	0.40
M6 07	28.8.00 9:00	10.28	5.57	2	10.26	0.88
M6 07	30.8.00 9:00	11.88	12.06		11.88	0.18
M6 07	1.9.00 9:00	11.85	16.83		11.76	5.07
M6 07	3.9.00 9:00	10	10.22		10.00	0.22
M6 07	5.9.00 9:00	9.16	11		9.13	1.87
M6 07	7.9.00 9:00	9.16	10.84		9.13	1.71
M6 07	9.9.00 9:00	4.4	5.66		4.38	1.28
M6 07	11.9.00 9:00	8.91	10.93		8.87	2.06
M6 07	13.9.00 9:00	9.06	10.01		9.04	0.97
M6 07	14.9.00 9:00	9.13	9.91		9.12	0.79
M6 07	21.9.00 9:00	8.78	9.74		8.76	0.98
M6 07	30.9.00 9:00	10.08	10.69		10.07	0.62

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M6 07	7.10.00 9:00	9.78		10.52		9.77	0.75
M6 07	14.10.00 9:00	10.26		2.55	5	10.21	2.54
M6 07	21.10.00 9:00	9.62		3.45	5	9.48	7.77
M6 07	28.10.00 9:00	9.74		10.33		9.73	0.60
M6 07	8.11.00 9:00	10.23		10.36		10.23	0.13
M6 08	19.8.00 9:00	8.17		9.28		8.15	1.13
M6 08	20.8.00 9:00	9.34		10.48		9.32	1.16
M6 08	21.8.00 9:00	9.56		10.54		9.54	1.00
M6 08	22.8.00 9:00	5.87		11.04		5.78	5.26
M6 08	23.8.00 9:00	9.58		11.09		9.55	1.54
M6 08	24.8.00 9:00	9.72		11.31		9.69	1.62
M6 08	25.8.00 9:00	9.71		11.45		9.68	1.77
M6 08	26.8.00 9:00	9.61		11.08		9.58	1.50
M6 08	27.8.00 9:00	10.3		5.51	2	10.29	0.73
M6 08	28.8.00 9:00	10.23		5.48	2	10.22	0.74
M6 08	30.8.00 9:00	11.55		11.94		11.54	0.40
M6 08	1.9.00 9:00	11.75		11.89		11.75	0.14
M6 08	3.9.00 9:00	10.31		10.02		10.32	0.00
M6 08	5.9.00 9:00	8.63		10.84		8.59	2.25
M6 08	7.9.00 9:00	9.05		10.7		9.02	1.68
M6 08	9.9.00 9:00	1.18		2.52		1.16	1.36
M6 08	11.9.00 9:00	9.57		10.82		9.55	1.27
M6 08	13.9.00 9:00	9.32		10.18		9.30	0.88
M6 08	14.9.00 9:00	9.25		10.23		9.23	1.00
M6 08	21.9.00 9:00	9.36		10.19		9.34	0.85
M6 08	30.9.00 9:00	10.76		11.3		10.75	0.55
M6 08	7.10.00 9:00	10.79		10.64		10.79	0.00
M6 08	14.10.00 9:00	10.69		2.86	5	10.62	3.68
M6 08	21.10.00 9:00	10.04		2.72	5	9.97	3.63
M6 08	28.10.00 9:00	10.58		10.73		10.58	0.15
M6 08	8.11.00 9:00	10.86		13.15		10.82	2.33

#### Tab.: A6 arsenic concentration from M8

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M8 02	19.8.00 9:00	9.29		9.9		9.28	0.62
M8 02	21.8.00 9:00	9.97		10.57		9.96	0.61
M8 02	23.8.00 9:00	9.63		10.06		9.63	0.43
M8 02	25.8.00 9:00	8.83		9.89		8.82	1.07
M8 02	27.8.00 9:00	8.97		4.87	2	8.96	0.78
M8 03	19.8.00 9:00	10.19		10.92		10.18	0.74
M8 03	21.8.00 9:00	11.26		12.07		11.25	0.82
M8 03	23.8.00 9:00	11.21		11.31		11.21	0.10
M8 03	25.8.00 9:00	10.57		11.21		10.56	0.65
M8 03	27.8.00 9:00	11.09		5.75	2	11.09	0.41
M8 03	14.9.00 9:00	10.82		11.28		10.82	0.46
M8 04	19 8 00 9.00	9.14		9.62		9.14	0.48
M8 04	21.8.00 9:00	11.11		11.87		11.10	0.77
M8 04	23.8.00 9:00	10.49		10.98		10.49	0.49
M8 04	25.8.00 9:00	10.13		10.91		10.12	0.79
M8 04	27.8.00 9:00	9.83		5.61	2	9.82	1.40
M8 04	14.9.00 9:00	9.13		9.63		9.12	0.51
				1 1			
M8 05	19.8.00 9:00	10.32		10.73		10.32	0.41
M8 05	21.8.00 9:00	10.89		11.34		10.89	0.45
M8 05	23.8.00 9:00	10.54		11.01		10.54	0.47
M8 05	25.8.00 9:00	9.93		10.43		9.92	0.51
M8 05	27.8.00 9:00	10.54		5.41	2	10.54	0.28
M8 05	14.9.00 9:00	10		10.26		10.00	0.26
M8 06	19.8.00 9:00	10.98		11.67		10.97	0.70
M8 06	21.8.00 9:00	10.93		11.36		10.93	0.43
M8 06	23.8.00 9:00	10.78		10.97		10.78	0.19
M8 06	25.8.00 9:00	10.3		11.1		10.29	0.81
M8 06	27.8.00 9:00	10.5		5.21	2	10.50	0.00
M8 06	14.9.00 9:00	11.05		11.37	_	11.05	0.32
M8 07	19 8 00 9.00	10.85		11 18		10.85	0.33
M8 07	21 8 00 9.00	11.57		12.38		11 56	0.33
M8 07	23 8 00 9.00	10.99		11 53		10.98	0.52
M8 07	25.8 00 9.00	10.63		11 54		10.62	0.92
M8 07	27.8.00 9.00	10.44	<u> </u>	5.61	2	10.43	0.79
					-		
M8 08	19 8 00 9.00	10 34		10 79		10 34	0.45
M8 08	21 8 00 9.00	11 25		12 13		11 24	0.89
M8 08	23.8.00 9:00	10.74		10.87		10.74	0.13

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M8 08	25.8.00 9:00	10.34		10.95		10.33	0.62
M8 08	27.8.00 9:00	9.88		5.7	2	9.86	1.54

Tab.: A7 arsenic concentration from M9

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M9 02	15.8.00 9:00	7.14		6.54		7.15	0.00
M9 02	15.8.00 19:00	7.21		6.62		7.22	0.00
M9 02	16.8.00 9:00	7.06		6.55		7.07	0.00
M9 02	16.8.00 19:00	6.97		6.59		6.98	0.00
M9 02	17.8.00 9:00	7.08		6.45		7.09	0.00
M9 02	17.8.00 19:00	7.07		6.49		7.08	0.00
M9 02	18.8.00 9:00	7.01		6.54		7.02	0.00
M9 02	18.8.00 19:00	7.09		6.46		7.10	0.00
M9 02	19.8.00 9:00	7.08		6.61		7.09	0.00
M9 02	20.8.00 9:00	6.87		5.82		6.89	0.00
M9 02	21.8.00 9:00	5.79		5.71		5.79	0.00
M9 02	22.8.00 9:00	6.04		6.13		6.04	0.09
M9 02	23.8.00 9:00	6.2		5.8		6.21	0.00
M9 02	24.8.00 9:00	6.04		5.6		6.05	0.00
M9 02	25.8.00 9:00	6.15		5.36		6.16	0.00
M9 02	26.8.00 9:00	6.16		5.52		6.17	0.00
M9 02	27.8.00 9:00	6.03		2.93	2	6.03	0.00
M9 02	28.8.00 9:00	6.08		2.76	2	6.09	0.00
M9 02	30.8.00 9:00	6.4		6.85		6.39	0.46
M9 02	1.9.00 9:00	6.62		6.53		6.62	0.00
M9 02	3.9.00 9:00	5.92		5.45		5.93	0.00
M9 02	5.9.00 9:00	5.11		5.85		5.10	0.75
M9 02	7.9.00 9:00	4.91		5.91		4.89	1.02
M9 02	9.9.00 9:00	5.29		5.88		5.28	0.60
M9 02	11.9.00 9:00	5.06		5.93		5.04	0.89
M9 02	13.9.00 9:00	5.55		5.85		5.54	0.31
M9 02	14.9.00 9:00	5.78		6.13		5.77	0.36
M9 02	21.9.00 9:00	5.58		6.02		5.57	0.45
M9 02	30.9.00 9:00	6.04		6.36		6.03	0.33
M9 02	7.10.00 9:00	6.46		6.42		6.46	0.00
M9 02	14.10.00 9:00	6.91		8.47		6.88	1.59
M9 02	21.10.00 9:00	6.49		6.91		6.48	0.43
M9 02	28.10.00 9:00	8.34		8.41		8.34	0.07
M9 02	8.11.00 9:00	9.85		9.83		9.85	0.00
M9 03	15.8.00 9:00	4,97		4.37		4,98	0.00
M9 03	15.8.00 19:00	4.78		4.32		4.79	0.00
M9 03	16.8.00 9:00	4.8		4.31		4.81	0.00
M9 03	16.8.00 19:00	4.74		4.27		4.75	0.00
M9 03	17.8.00 9:00	4.73		4.34		4.74	0.00
M9 03	17.8.00 19:00	4.55		4.05		4.56	0.00
M9 03	18.8.00 9:00	4.08		4.16		4.08	0.08
M9 03	18.8.00 19:00	4.18		4.2		4.18	0.02
M9 03	19.8.00 9:00	4.17		4.66		4.16	0.50
M9 03	20.8.00 9:00	4.1		4.34		4.10	0.24
M9 03	21.8.00 9:00	3.5		4.21		3.49	0.72
M9 03	22.8.00 9:00	4.34		4.05		4.35	0.00
M9 03	23.8.00 9:00	4.39		3.96		4.40	0.00

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M9 03	24.8.00 9:00	4.08		4.02		4.08	0.00
M9 03	25.8.00 9:00	4.19		3.89		4.20	0.00
M9 03	26.8.00 9:00	4.43		3.98		4.44	0.00
M9 03	27.8.00 9:00	3.82		1.93	2	3.82	0.04
M9 03	28.8.00 9:00	3.77		1.97	2	3.77	0.17
M9 03	30.8.00 9:00	4.57		4.49		4.57	0.00
M9 03	1.9.00 9:00	4.55		4.39		4.55	0.00
M9 03	3.9.00 9:00	3.97		3.88		3.97	0.00
M9 03	5.9.00 9:00	3.57		4.22		3.56	0.66
M9 03	7.9.00 9:00	2.97		3.68		2.96	0.72
M9 03	9.9.00 9:00	2.5		3.77		2.48	1.29
M9 03	11.9.00 9:00	2.99		3.52		2.98	0.54
M9 03	13.9.00 9:00	3.33		3.62		3.32	0.30
M9 03	14.9.00 9:00	3.3		3.73		3.29	0.44
M9 03	21.9.00 9:00	3.61		4.07		3.60	0.47
M9 03	30.9.00 9:00	5.69		6.26		5.68	0.58
M9 03	7.10.00 9:00	7.25		7.68		7.24	0.44
M9 03	14.10.00 9:00	9.55		2.27	5	9.52	1.83
M9 03	21.10.00 9:00	10.13		3.14	5	10.03	5.67
M9 03	28.10.00 9:00	2.55	5	2.58	5	12.75	0.15
M9 03	8.11.00 9:00	2.97	5	2.72	5	14.87	0.00
M9 04	15.8.00 9:00	3.21		3.17		3.21	0.00
M9 04	15.8.00 19:00	3.25		3.1		3.25	0.00
M9 04	16.8.00 9:00	3.02		3.05		3.02	0.03
M9 04	16.8.00 19:00	3.1		3.1		3.10	0.00
M9 04	17.8.00 9:00	2.96		3.07		2.96	0.11
M9 04	17.8.00 19:00	3.19		3.07		3.19	0.00
M9 04	18.8.00 9:00	3.06		3.07		3.06	0.01
M9 04	18.8.00 19:00	3.02		3.01		3.02	0.00
M9 04	19.8.00 9:00	3.23		3.37		3.23	0.14
M9 04	20.8.00 9:00	3.23		2.78		3.24	0.00
M9 04	21.8.00 9:00	2.34		3.02		2.33	0.69
M9 04	22.8.00 9:00	3.11		2.87		3.11	0.00
M9 04	23.8.00 9:00	3.49		3.07		3.50	0.00
M9 04	24.8.00 9:00	3.1		2.84		3.10	0.00
M9 04	25.8.00 9:00	3		2.63		3.01	0.00
M9 04	26.8.00 9:00	3.83		3.47		3.84	0.00
M9 04	27.8.00 9:00	2.79		1.38	2	2.79	0.00
M9 04	28.8.00 9:00	2.9		1.38	2	2.90	0.00
M9 04	30.8.00 9:00	3.53		3.21		3.54	0.00
M9 04	1.9.00 9:00	3.63		3.63		3.63	0.00
M9 04	3.9.00 9:00	2.96		2.95		2.96	0.00
M9 04	5.9.00 9:00	2.87		3.2		2.86	0.34
M9 04	7.9.00 9:00	2.28		2.93		2.27	0.66
M9 04	9.9.00 9:00	2.93		3.03		2.93	0.10
M9 04	11.9.00 9:00	2.59		3.24		2.58	0.66
M9 04	13.9.00 9:00	3.41		3.75		3.40	0.35
M9 04	14.9.00 9:00	3.4		3.82		3.39	0.43
M9 04	21.9.00 9:00	4.72		5.22		4.71	0.51
M9 04	30.9.00 9:00	6.43		7.39		6.41	0.98
M9 04	7.10.00 9:00	7.98		8.39		7.97	0.42

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M9 04	14.10.00 9:00	10.77		2.78	5	10.71	3.19
M9 04	21.10.00 9:00	10.59		2.92	5	10.52	4.08
M9 04	28.10.00 9:00	2.74	5	3.04	5	13.67	1.53
M9 04	8.11.00 9:00	2.95	5	5.68	5	14.50	13.90
M9 05	15.8.00 9:00	8.8		9.99		8.78	1.21
M9 05	15.8.00 19:00	8.72		10.28		8.69	1.59
M9 05	16.8.00 9:00	8.83		10		8.81	1.19
M9 05	16.8.00 19:00	8.74		9.5		8.73	0.77
M9 05	17.8.00 9:00	8.6		9.46		8.58	0.88
M9 05	17.8.00 19:00	8.42		9.67		8.40	1.27
M9 05	18.8.00 9:00	8.56		9.77		8.54	1.23
M9 05	18.8.00 19:00	8.26		10.94		8.21	2.73
M9 05	19.8.00 9:00	8.13		9.41		8.11	1.30
M9 05	20.8.00 9:00	8.28		9.12		8.26	0.86
M9 05	21.8.00 9:00	8.12		8.89		8.11	0.78
M9 05	22.8.00 9:00	8.02		8.07		8.02	0.05
M9 05	23.8.00 9:00	8.11		7.79		8.12	0.00
M9 05	24.8.00 9:00	7.84		7.81		7.84	0.00
M9 05	25.8.00 9:00	7.64		7.62		7.64	0.00
M9 05	26.8.00 9:00	7.59		7.37		7.59	0.00
M9 05	27.8.00 9:00	7.12		3.62	2	7.12	0.12
M9 05	28.8.00 9:00	6.81		3.63	2	6.80	0.46
M9 05	30.8.00 9:00	8.01		7.94		8.01	0.00
M9 05	1.9.00 9:00	7.51		8.1		7.50	0.60
M9 05	3.9.00 9:00	6.88		6.45		6.89	0.00
M9 05	5.9.00 9:00	6.29		6.74		6.28	0.46
M9 05	7.9.00 9:00	5.37		6.49		5.35	1.14
M9 05	9.9.00 9:00	4.79		5.85		4.77	1.08
M9 05	11.9.00 9:00	5.15		6.11		5.13	0.98
M9 05	13.9.00 9:00	5.64		5.77		5.64	0.13
M9 05	14.9.00 9:00	5.41		5.88		5.40	0.48
M9 05	21.9.00 9:00	5.41		5.67		5.41	0.26
M9 05	30.9.00 9:00	5.11		5.65		5.10	0.55
M9 05	7.10.00 9:00	5.56		5.69		5.56	0.13
M9 05	14.10.00 9:00	5.76		6.75		5.74	1.01
M9 05	21.10.00 9:00	5.13		9.29		5.05	4.24
M9 05	28.10.00 9:00	5.98		5.59		5.99	0.00
M9 05	8.11.00 9:00	7.6		6.56		7.62	0.00

name	date	As(III)	dilution	As(tot)	dilution		$A_{S}(V)$ cal
name	dato	[µg/l]	Gildtoll	[µg/l]	dildtori		/10(1) 0uii
P						1	-
M10 02	19.8.00 9:00	5.82		5.8		5.82	0.00
M10 02	21.8.00 9:00	4		4.83		3.98	0.85
M10 02	23.8.00 9:00	4.6		4.09		4.61	0.00
M10 02	25.8.00 9:00	3.88		3.96		3.88	0.08
M10 02	27.8.00 9:00	3.96		2.09	2	3.96	0.22
M10 02	7.10.00 9:00	7.27		7.51		7.27	0.24
F						1	
M10 03	19.8.00 9:00	7.98		7.64		7.99	0.00
M10 03	21.8.00 9:00	6.2		6.5		6.19	0.31
M10 03	23.8.00 9:00	5.98		5.8		5.98	0.00
M10 03	25.8.00 9:00	5.39		5.24		5.39	0.00
M10 03	27.8.00 9:00	5.22		2.7	2	5.22	0.18
M10 03	14.9.00 9:00	4.39		4.35		4.39	0.00
M10 03	30.9.00 9:00	4.18		4.65		4.17	0.48
M10 03	7.10.00 9:00	4.45		4.66		4.45	0.21
M10 03	14.10.00 9:00	5		5.66		4.99	0.67
M10 03	21.10.00 9:00	4.78		11.92		4.63	7.29
M10 03	28.10.00 9:00	5.77		5.63		5.77	0.00
M10 03	8.11.00 9:00	6.72		7.77		6.70	1.07
M10 04	19.8.00 9:00	7.37		7.15		7.37	0.00
M10 04	21.8.00 9:00	6.14		6.18		6.14	0.04
M10 04	23.8.00 9:00	5.67		6.02		5.66	0.36
M10 04	25.8.00 9:00	5.23		5.09		5.23	-0.14
M10 04	27.8.00 9:00	5.15		2.62	2	5.15	0.09
M10 04	14.9.00 9:00	3.58		3.71		3.58	0.13
M10 04	30.9.00 9:00	3.83		3.99		3.83	0.16
M10 04	7.10.00 9:00	4.08		4.36		4.07	0.29
M10 04	14.10.00 9:00	4.85		5.54		4.84	0.70
M10 04	21.10.00 9:00	4.72		6.29		4.69	1.60
M10 04	28.10.00 9:00	5.76		5.88		5.76	0.12
M10 04	8.11.00 9:00	7.94		7.22		7.95	0.00
M10 05	19.8.00 9:00	11.24		10.29		11.26	0.00
M10 05	21.8.00 9:00	9.94		0.11		10.14	0.00

#### Tab.: A8 arsenic concentration from M10

M10 05	19.8.00 9:00	11.24	10.29		11.26	0.00
M10 05	21.8.00 9:00	9.94	0.11		10.14	0.00
M10 05	23.8.00 9:00	9.18	8.53		9.19	0.00
M10 05	25.8.00 9:00	8.58	8.62		8.58	0.04
M10 05	27.8.00 9:00	7.97	4.05	2	7.97	0.13
M10 05	14.9.00 9:00	6.15	6.09		6.15	0.00
M10 05	30.9.00 9:00	4.69	4.99		4.68	0.31
M10 05	7.10.00 9:00	4.66	5.02		4.65	0.37
M10 05	14.10.00 9:00	5.28	5.89		5.27	0.62
M10 05	21.10.00 9:00	4.88	6.51		4.85	1.66
M10 05	28.10.00 9:00	4.81	4.73		4.81	0.00
M10 05	8.11.00 9:00	5.24	5.14		5.24	0.00

name	date	As(III) [µg/l]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
-							
M10 06	19.8.00 9:00	13.92		12.78		13.94	0.00
M10 06	21.8.00 9:00	12.59		12.81		12.59	0.22
M10 06	23.8.00 9:00	11.66		11.68		11.66	0.02
M10 06	25.8.00 9:00	11.14		10.91		11.14	0.00
M10 06	27.8.00 9:00	11.28		5.98	2	11.27	0.69
M10 06	14.9.00 0:00	9.76		10.14		9.75	0.39
M10 06	30.9.00 9:00	10.53		10.82		10.52	0.30
M10 06	14.10.00 9:00	10.48		11.95		10.45	1.50
M10 06	21.10.00 9:00	9.89		6.36		9.96	0.00
M10 06	28.10.00 9:00	9.83		9.59		9.83	0.00
M10 06	8.11.00 9:00	10.46		10.12		10.47	0.00

M10 07	19.8.00 9:00	12.3	11.94		12.31	0.00
M10 07	21.8.00 9:00	11.08	11.25		11.08	0.17
M10 07	23.8.00 9:00	10.9	10.63		10.91	0.00
M10 07	25.8.00 9:00	10.77	10.3		10.78	0.00
M10 07	27.8.00 9:00	11.23	5.87	2	11.22	0.52

M10 08	19.8.00 9:00	12.62	12.22		12.63	0.00
M10 08	21.8.00 9:00	11.28	11.11		11.28	0.00
M10 08	23.8.00 9:00	10.52	10.71		10.52	0.19
M10 08	25.8.00 9:00	10.21	10.52		10.20	0.32
M10 08	27.8.00 9:00	11.14	5.59	2	11.14	0.04

name	As(III) [µg/I]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
M2-01	10.32		5.99	2	10.29	1.69
M2-02	10.05		5.93	2	10.02	1.84
M2-03	9.26		5.4	2	9.23	1.57
M2-04	8.82		5.13	2	8.79	1.47
M2-05	10.02		5.5	2	10.00	1.00
M2-06	9.94		5.67	2	9.91	1.43
M2-07	10.56		6.01	2	10.53	1.49
		-	-	-		
M3-01	7.43		4.78	2	7.39	2.17
M3-02	8.42		4.98	2	8.39	1.57
M3-03	9.57		5.67	2	9.54	1.80
M3-04	7.98		4.74	2	7.95	1.53
M3-05	9.08		5.22	2	9.06	1.38
M3-06	7.66		5.16	2	7.61	2.71
M3-07	8.05		5.57	2	7.99	3.15
M5-01	2.3	5	7.16	2	11.45	2.87
M5-02	3.58	5	10.08	2	17.86	2.30
M5-03	9.62		5.9	2	9.58	2.22
M5-04	3.75	5	9.93	2	18.73	1.13
M5-05	3.08	5	8.6	2	15.37	1.83
M5-06	2.5	5	6.26	2	12.50	0.02
M5-07	2.52	5	6.76	2	12.58	0.94
M8-01	7.12		4.36	2	7.09	1.63
M8-02	8.01		5.15	2	7.97	2.33
M8-03	2.31	5	5.65	2	11.55	-0.25
M8-04	2.58	5	7.57	2	12.86	2.28
M8-05	2.39	5	6.96	2	11.91	2.01
M8-06	9.46		5.45	2	9.43	1.47
M8-07	10.55		6.38	2	10.51	2.25
M9-01	3.36		1.97	2	3.35	0.59
M9-02	3.17		1.91	2	3.16	0.66
M9-03	5.69		3.47	2	5.67	1.27

3.1

3.04

2

2

M9-04 M9-05

4.99

6.03

Tab.: A9 arsenic background concentration

4.97

6.03

1.23 0.05

#### date Fe [mg/l] Mn [mg/l] 24.7.00 9:00 0.00 0.24 24.7.00 19:00 0.26 0.00 25.7.00 9:00 0.01 0.24 25.7.00 19:00 0.00 0.24 0.22 26.7.00 9:00 0.00 26.7.00 19:00 0.00 0.19 27.7.00 9:00 0.00 0.13 27.7.00 19:00 0.00 0.14 28.7.00 9:00 0.00 0.17 28.7.00 19:00 0.00 0.21 29.7.00 9:00 0.00 0.22 29.7.00 19:00 0.22 0.00 30.7.00 9:00 0.01 0.22 30.7.00 19:00 0.02 0.22 0.22 31.7.00 9:00 0.03 31.7.00 19:00 0.03 0.23 1.8.00 9:00 0.02 0.24 1.8.00 19:00 0.01 0.23 2.8.00 9:00 0.03 0.23 2.8.00 19:00 0.01 0.23 3.8.00 9:00 0.02 0.23 0.24 3.8.00 19:00 0.02 4.8.00 9:00 0.04 0.24 4.8.00 19:00 0.02 0.24 0.25 5.8.00 9:00 0.04 5.8.00 19:00 0.03 0.25 6.8.00 9:00 0.05 0.25 6.8.00 19:00 0.04 0.25 7.8.00 9:00 0.05 0.20 7.8.00 19:00 0.08 0.11 8.8.00 9:00 0.09 0.12 8.8.00 19:00 0.06 0.11 9.8.00 9:00 0.07 0.11 0.06 9.8.00 19:00 0.11 10.8.00 9:00 0.08 0.12 11.8.00 9:00 0.10 0.12 11.8.00 19:00 0.07 0.12 12.8.00 9:00 0.07 0.13 12.8.00 19:00 0.06 0.13 13.8.00 9:00 0.08 0.13 13.8.00 19:00 0.05 0.14 14.8.00 9:00 0.07 0.14 0.05 0.15 14.8.00 19:00 15.8.00 9:00 0.07 0.15 15.8.00 19:00 0.06 0.15 16.8.00 9:00 0.09 0.16 16.8.00 19:00 0.07 0.16 17.8.00 9:00 0.06 0.18 17.8.00 19:00 0.06 0.18 18.8.00 9:00 0.09 0.19 18.8.00 19:00 0.08 0.19

#### Tab.: A10 cations from M2-03

date	Fe [mg/l]	Mn [mg/l]
19.8.00 9:00	0.11	0.19
20.8.00 9:00	0.11	0.22
21.8.00 9:00	0.18	0.21
22.8.00 9:00	0.13	0.22
23.8.00 9:00	0.15	0.23
24.8.00 9:00	0.23	0.29
25.8.00 9:00	0.25	0.31
26.8.00 9:00	0.24	0.33
27.8.00 9:00	0.34	0.43
28.8.00 9:00	0.31	0.47
29.8.00 9:00	0.25	0.51
30.8.00 9:00	0.52	0.59
31.8.00 9:00	0.45	0.62
1.9.00 9:00	0.48	0.63
2.9.00 9:00	0.88	0.62
3.9.00 9:00	1.32	0.64
4.9.00 9:00	1.62	0.60
5.9.00 9:00	2.44	0.52
6.9.00 9:00	2.61	0.48
7.9.00 9:00	3.03	0.44
8.9.00 9:00	3.65	0.39
9.9.00 9:00	3.84	0.36
10.9.00 9:00	4.28	0.33
11.9.00 9:00	4.91	0.29
12.9.00 9:00	4.96	0.27
13.9.00 9:00	5.29	0.26
14.9.00 9:00	5.76	0.25
21.9.00 9:00	6.55	0.22
30.9.00 9:00	7.75	0.18
7.10.00 9:00	7.87	0.18
14.10.00 9:00	8.17	0.18
21.10.00 9:00	8.13	0.18
28.10.00 9:00	9.04	0.14
8.11.00 9:00	8.74	0.18

# Tab.: A11 cations from M3-03

date	Fe [mg/l]	Mn [mg/l]
26.6.00 19:00	14.34	0.26
26.6.00 23:00	13.96	0.24
27.6.00 7:00	12.92	0.23
27.6.00 19:00	12.26	0.22
28.6.00 7:00	12.4	0.21
28.6.00 19:00	12.09	0.21
29.6.00 7:00	11.81	0.2
29.6.00 15:00	12.08	0.2
29.6.00 23:00	12.35	0.21
30.6.00 7:00	12.46	0.21
30.6.00 15:00	11.75	0.23
30.6.00 23:00	12.95	0.22
1.7.00 7:00	13.24	0.23
1.7.00 15:00	12.94	0.22
1.7.00 23:00	10.63	0.19
2.7.00 9:00	9.91	0.18

date	Fe [mg/l]	Mn [mg/l]
2.7.00 19:00	14.12	0.26
3.7.00 9:00	11.96	0.22
3.7.00 19:00	9.62	0.19
4.7.00 9:00	7.41	0.16
5.7.00 9:00	5.68	0.15
5.7.00 19:00	5.74	0.14
6.7.00 19:00	4.23	0.12
7.7.00 9:00	3.25	0.11
7.7.00 19:00	1.71	0.1
8.7.00 9:00	1.19	0.1
8.7.00 19:00	1	0.09
9.7.00 9:00	0.4	0.09
9.7.00 19:00	0.19	0.09
10.7.00 9:00	0.1	0.08
10.7.00 19:00	0.09	0.09
11.7.00 9:00	0.07	0.09
11.7.00 19:00	0.08	0.09
12.7.00 9:00	0.05	0.09
12.7.00 19:00	0.05	0.09
13.7.00 9:00	0.04	0.09
13.7.00 19:00	0.03	0.09
14.7.00 9:00	0.05	0.09
14.7.00 19:00	0.03	0.09
15.7.00 9:00	0.02	0.09
15.7.00 19:00	0.04	0.09
16.7.00 9:00	0.02	0.09
16.7.00 19:00	0.05	0.09
17.7.00 9:00	0.03	0.09
17.7.00 19:00	0.04	0.09
18.7.00 9:00	0.06	0.09
18.7.00 19:00	0.04	0.09
19.7.00 9:00	0.03	0.09
19.7.00 19:00	0.03	0.09
20.7.00 9:00	0.02	0.1
20.7.00 19:00	0.03	0.1
21.7.00 9:00	0.02	0.1
21.7.00 19:00	0.02	0.1
22.7.00 9:00	0.01	0.11
	0.03	0.11
23.7.00 9:00	0.01	0.11
23.7.00 19.00	0.03	0.11
24.7.00 9.00	0.03	0.12
24.7.00 19.00	0.03	0.12
26.7.00 19.00	0.01	0.12
26.7.00 9.00	0.01	0.12
27 7 00 0.00	0.02	0.13
27.7.00 3.00	0.00	0.13
28 7 00 9.00	0.02	0.12
28.7.00 3.00	0.01	0.09
29.7.00 19.00	0.00	0.05
29.7.00.19.00	0	0.12
30 7 00 9 00	0.02	0.12
30.7.00 19:00	0.01	0.13
31,7,00 9:00	0.02	0.13
· · ····		

date	Fe [mg/l]	Mn [mg/l]
31.7.00 19:00	0.03	0.14
1.8.00 9:00	0.05	0.14
1.8.00 19:00	0.06	0.14
2.8.00 9:00	0.06	0.15
2.8.00 19:00	0.06	0.15
3.8.00 9:00	0.09	0.16
3.8.00 19:00	0.08	0.17
4.8.00 9:00	0.09	0.17
4.8.00 19:00	0.1	0.17
5.8.00 9:00	0.12	0.17
5.8.00 19:00	0.15	0.18
6.8.00 9:00	0.16	0.18
6.8.00 19:00	0.16	0.18
7.8.00 9:00	0.19	0.19
7.8.00 19:00	0.18	0.19
8.8.00 9:00	0.2	0.2
8.8.00 19:00	0.19	0.2
9.8.00 9:00	0.21	0.19
9.8.00 19:00	0.2	0.2
10.8.00 9:00	0.22	0.2
11.8.00 9:00	0.23	0.21
11.8.00 19:00	0.25	0.21
12.8.00 9:00	0.24	0.22
12.8.00 19:00	0.26	0.21
13.8.00 9:00	0.26	0.22
13.8.00 19:00	0.32	0.22
14.8.00 9:00	0.28	0.23
14.8.00 19:00	0.28	0.23
15.8.00 9:00	0.27	0.23
15.8.00 19:00	0.3	0.23
16.8.00 9:00	0.3	0.23
16.8.00 19:00	0.3	0.23
17.8.00 9:00	0.4	0.23
17.8.00 19:00	0.36	0.23
18.8.00 9:00	0.35	0.24
18.8.00 19:00	0.35	0.23
19.8.00 9:00	0.39	0.24
20.8.00 9:00	0.39	0.23
21.8.00 9:00	0.38	0.24
22.8.00 9:00	0.39	0.25
23.8.00 9:00	0.42	0.25
24.8.00 9:00	0.44	0.26
25.8.00 9:00	0.44	0.27
26.8.00 9:00	0.47	0.27
27.8.00 9:00	0.48	0.29
28.8.00 9:00	0.47	0.31
29.8.00 9:00	0.49	0.33
30.8.00 9:00	0.54	0.36
31.8.00 9:00	0.58	0.39
1.9.00 9:00	0.65	0.43
2.9.00 9:00	0.71	0.44
3.9.00 9:00	0.78	0.47
4.9.00 9:00	0.89	0.48
5.9.00 9:00	1	0.5
6.9.00 9:00	1.09	0.52

date	Fe [mg/l]	Mn [mg/l]
7.9.00 9:00	1.19	0.5
8.9.00 9:00	1.51	0.51
9.9.00 9:00	1.76	0.51
10.9.00 9:00	2.03	0.49
11.9.00 9:00	2.29	0.48
12.9.00 9:00	2.48	0.47
13.9.00 9:00	2.53	0.45
14.9.00 9:00	2.82	0.45
21.9.00 9:00	4.43	0.4
30.9.00 9:00	5.63	0.33
7.10.00 9:00	6.45	0.31
14.10.00 9:00	6.64	0.29
21.10.00 9:00	7.01	0.29
28.10.00 9:00	7.57	0.29
8.11.00 9:00	7.67	0.28

## Tab.: A12 cations from M5-03

date	Fe [mg/l]	Mn [mg/l]
30.6.00 15:00	19.4	0.25
1.7.00 7:00	20.03	0.26
1.7.00 15:00	18.21	0.24
1.7.00 23:00	17.93	0.24
2.7.00 9:00	16.27	0.22
2.7.00 19:00	15.71	0.2
3.7.00 9:00	15.58	0.2
4.7.00 9:00	15	0.2
5.7.00 9:00	12.92	0.18
6.7.00 9:00	14.76	0.21
7.7.00 9:00	12.48	0.15
8.7.00 9:00	12.66	0.17
9.7.00 9:00	12.57	0.17
10.7.00 9:00	12.35	0.17
11.7.00 9:00	12.59	0.16
12.7.00 9:00	12.22	0.15
13.7.00 9:00	10.56	0.14
14.7.00 9:00	11.13	0.13
15.7.00 9:00	9.68	0.11
16.7.00 9:00	8.78	0.1
17.7.00 9:00	4.08	0.05
18.7.00 9:00	0.95	0.07
19.7.00 9:00	0.34	0.08
20.7.00 9:00	0.16	0.08
21.7.00 9:00	0.1	0.07
22.7.00 9:00	0.06	0.07
23.7.00 9:00	0.05	0.07
24.7.00 9:00	0.25	0.06
24.7.00 19:00	0.07	0.07
25.7.00 9:00	0.06	0.07
26.7.00 19:00	0.03	0.06
27.7.00 9:00	0.16	0.07
27.7.00 19:00	0.13	0.06
28.7.00 9:00	0.11	0.07
28.7.00 19:00	0.1	0.07

date	Fe [mg/l]	Mn [mg/l]
29.7.00 9:00	0.24	0.07
29.7.00 19:00	0.12	0.07
30.7.00 9:00	0.11	0.06
30.7.00 19:00	0.15	0.06
31.7.00 9:00	0.27	0.06
1.8.00 9:00	0.27	0.04
1.8.00 19:00	0.24	0.03
2.8.00 9:00	0.31	0.02
2.8.00 19:00	0.33	0.02
3.8.00 9:00	0.8	0.06
3.8.00 19:00	0.33	0.02
4.8.00 9:00	0.52	0.04
4.8.00 19:00	0.35	0.05
5.8.00 9:00	0.5	0.05
5.8.00 19:00	0.39	0.06
6.8.00 9:00	0.51	0.06
6.8.00 19:00	0.36	0.06
7.8.00 9:00	0.44	0.07
7.8.00 19:00	0.39	0.06
8.8.00 9:00	0.45	0.06
8.8.00 19:00	0.4	0.07
9.8.00 9:00	0.41	0.06
9.8.00 19:00	0.37	0.06
10.8.00 15:00	0.42	0.06
11.8.00 9:00	0.46	0.07
11.8.00 19:00	0.49	0.07
12.8.00 9.00	0.44	0.06
12.8.00 19.00	0.40	0.07
13.8.00 9.00	0.45	0.07
14 8 00 0.00	0.42	0.07
14.8.00 9.00	0.30	0.07
15.8.00.9.00	0.53	0.07
15.8.00 19:00	0.00	0.07
16.8.00.9.00	0.38	0.07
16.8.00 19:00	0.36	0.06
17.8.00 9.00	0.44	0.06
17,8.00 19:00	0.4	0.06
18.8.00 9:00	0.37	0.05
18.8.00 19:00	0.35	0.06
19.8.00 9:00	0.33	0.05
20.8.00 9:00	0.35	0.05
21.8.00 9:00	0.32	0.05
22.8.00 9:00	0.32	0.05
23.8.00 9:00	0.34	0.05
24.8.00 9:00	0.4	0.05
25.8.00 9:00	0.4	0.05
26.8.00 9:00	0.42	0.05
27.8.00 9:00	0.44	0.05
28.8.00 9:00	0.4	0.06
29.8.00 9:00	0.38	0.06
30.8.00 9:00	0.43	0.06
31.8.00 9:00	0.39	0.05
1.9.00 9:00	0.43	0.06
2.9.00 9:00	0.44	0.07

date	Fe [mg/l]	Mn [mg/l]
3.9.00 9:00	0.47	0.07
5.9.00 9:00	0.46	0.08
6.9.00 9:00	0.42	0.08
7.9.00 9:00	0.41	0.09
8.9.00 9:00	0.46	0.1
9.9.00 9:00	0.5	0.11
10.9.00 9:00	0.45	0.11
11.9.00 9:00	0.47	0.12
12.9.00 9:00	0.45	0.12
13.9.00 9:00	0.44	0.13
14.9.00 9:00	0.56	0.14
21.9.00 9:00	0.63	0.18
30.9.00 9:00	0.61	0.22
7.10.00 9:00	0.61	0.23
14.10.00 9:00	0.61	0.23
21.10.00 9:00	0.61	0.23
28.10.00 9:00	0.63	0.24
8.11.00 9:00	0.7	0.32

#### Tab.: A 13 cations from M9-03

date	Fe [mg/l]	Mn [mg/l]
30.6.00 9:00	14.49	0.27
10.8.00 9:00	12.31	0.16
15.8.00 9:00	12.05	0.19
15.8.00 19:00	12.2	0.2
16.8.00 9:00	12.44	0.2
16.8.00 19:00	12.28	0.2
17.8.00 9:00	12.14	0.2
17.8.00 19:00	12.16	0.2
18.8.00 9:00	12.02	0.19
18.8.00 19:00	12.07	0.2
19.8.00 9:00	11.72	0.19
20.8.00 9:00	11.64	0.18
22.8.00 9:00	10.78	0.16
23.8.00 9:00	9.99	0.15
24.8.00 9:00	9.41	0.14
25.8.00 9:00	8.74	0.13
26.8.00 9:00	8.57	0.12
27.8.00 9:00	7.88	0.11
28.8.00 9:00	7.34	0.1
30.8.00 9:00	6.38	0.08
1.9.00 9:00	5.73	0.08
3.9.00 9:00	5.24	0.07
5.9.00 9:00	5.26	0.07
7.9.00 9:00	3.38	0.09
9.9.00 9:00	4.77	0.07
11.9.00 9:00	4.49	0.07
13.9.00 9:00	4.43	0.07
14.9.00 9:00	4.25	0.07
21.9.00 9:00	4.17	0.08
30.9.00 9:00	4.16	0.09
7.10.00 9:00	4.13	0.1

date	Fe [mg/l]	Mn [mg/l]
14.10.00 9:00	4.07	0.1
21.10.00 9:00	4.12	0.09
28.10.00 9:00	4.05	0.09
8.11.00 9:00	3.76	0.09

sample	Mg	Ca	K	Na	Fe	Mn
	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]	[mg/l]
	-				-	
M2,PT01 20.06.00	3.19	9.17	12.79	12.9	16.53	0.28
M2,GNT02 20.06.00	2.22	7.39	10.08	12.74	13.65	0.211
M2,RT03 20.06.00	1.72	7.53	9.14	12.93	11.7	0.181
M2,BUT04 20.06.00	1.68	7.44	9.72	13.54	10.88	0.183
M2,BKT05 20.06.00	1.68	7.53	9.53	13.87	12	0.178
M2,WT06 20.06.00	1.56	6.95	9.25	14.21	11.01	0.17
M2,007 20.06.00	1.52	7.24	8.32	15.76	11.73	0.162
M3,PT01 20.06.00	2.36	9.38	12.11	12.6	13.99	0.292
M3,GNT02 20.06.00	2.75	9.63	11.99	12.58	17.81	0.32
M3,RT03 20.06.00	2.83	8.45	12.16	13.59	18.12	0.287
M3,BUT04 20.06.00	1.68	7.26	9.45	12.23	13	0.206
M3,BKT05 20.06.00	1.71	7.67	10.09	13.51	14.36	0.219
M3,WT06 20.06.00	1.7	7.38	9.84	13.61	12.58	0.199
M3,007 20.06.00	1.68	7.26	9.81	14.2	13.68	0.204
M5,PT01 20.06.00	2.66	9.81	12.52	12.48	18.06	0.31
M5,GNT02 20.06.00	3.28	9.43	12.58	13.15	25.8	0.317
M5,RT03 20.06.00	1.79	7.37	9.56	12.6	12.16	0.208
M5,BUT04 20.06.00	1.84	7.94	10.35	13.12	20.02	0.226
M5,BKT05 20.06.00	1.74	7.62	9.89	13.22	17.9	0.213
M5,WT06 20.06.00	1.76	7.45	10.06	14.04	13.08	0.214
M5,007 20.06.00	1.62	7.32	9.25	15.09	13.42	0.2
M9,PT01 20.06.00	2.71	9.71	13.01	12.89	9.01	0.304
M9,GNT02 20.06.00	3.33	8.59	11.27	12.64	10.22	0.279
M9,RT03 20.06.00	2.32	7.92	10.59	13.42	10.7	0.239
M9,BUT04 20.06.00	1.75	7.38	10.14	13.12	7.49	0.205
M9,BKT05 20.06.00	1.84	7.76	10.39	13.49	6.72	0.213
8-12,W15 20.06.00 BK14mix	2.23	11.44	7.68	16.78	nn	0.193

# Tab.: A14 cations background concentrations

## Tab.: A15 field and fieldlaboratory measurements

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
23.05.2000	8-12	1			1.89								
23.05.2000	8-12	9			0.47								
23.05.2000	8-12	10			0.42								
23.05.2000	8-12	11			0.38								
07.06.2000	8-12	11									4		
10.06.2000	8-12	11	6.49									0.77	
14.06.2000	8-12	11	5.91										
23.05.2000	8-12	12			0.08	0							
07.06.2000	8-12	12									3.7		
10.06.2000	8-12	12	6.53									0.69	
14.06.2000	8-12	12	5.93										
23.05.2000	8-12	13			0.07	0							
07.06.2000	8-12	13									3.7		
10.06.2000	8-12	13	6.58									0.63	
14.06.2000	8-12	13	5.93										
23.05.2000	8-12	14			0.05	0							
07.06.2000	8-12	14									3.8		
10.06.2000	8-12	14	6.54									0.6	
14.06.2000	8-12	14	6.04										
23.05.2000	8-12	15			0.07	0							
07.06.2000	8-12	15									4.05		
10.06.2000	8-12	15	6.51									0.6	
14.06.2000	8-12	15	6.12										
20.06.2000	8-12		5.9									7.4	
23.06.2000	8-12		6.08									7.35	
09.07.2000	Influer	nt	6.25									7.2	
15.07.2000	Influer	nt	6.15									7.2	
21.07.2000	Influer	nt	6.03									7.3	
25.06.2000	Injecta	ite	6.19									11.2	

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
26.06.2000	Injecta	te	6.29									11.2	
28.06.2000	Injecta	te	6.35									10.8	
09.07.2000	Injecta	te	6.43									10.8	
15.07.2000	Injecta	te	6.41									11.6	
21.07.2000	Injecta	te	6.32									11.1	
30.06.2000	M10	2	6.45									7.4	
04.07.2000	M10	2	6.62									13.6	
30.06.2000	M10	3	6.38									5.4	
09.07.2000	M10	4	6.66									16.1	
12.07.2000	M10	4	5.92									15	
15.07.2000	M10	4	6.58									14.7	
18.07.2000	M10	4	6.51									14.6	
24.07.2000	M10	4	6.2									13.6	
23.05.2000	M2	1	6.47	217	0.03								
30.05.2000	M2	1	6.33	269	0.03								
07.06.2000	M2	1									3.25		
10.06.2000	M2	1	6.32									0.79	
14.06.2000	M2	1	6.55										
20.06.2000	M2	1	6.29									7.8	
23.05.2000	M2	2	6.46	251	0.11								
30.05.2000	M2	2	6.38	244	0.04								
07.06.2000	M2	2									3.15		
10.06.2000	M2	2	6.41									0.64	
14.06.2000	M2	2	6.61										
20.06.2000	M2	2	6.59									6.3	
23.06.2000	M2	2	6.35									6.1	
07. Jul	M2	2	5.78									11.8	
09.07.2000	M2	2	6.39									11.1	
12.07.2000	M2	2	6.04									11.2	
15.07.2000	M2	2	6.4									11.4	
18.07.2000	M2	2	6.38									11.3	
21.07.2000	M2	2	6.3									11.2	

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
24.07.2000	M2	2	6.02									11.8	
17.08.2000	M2	2	6.22										
23.05.2000	M2	3	6.49	221	0.06								
30.05.2000	M2	3	6.43	218	0								
07.06.2000	M2	3									3.8		
10.06.2000	M2	3	6.44									0.51	
14.06.2000	M2	3	6.64										
20.06.2000	M2	3	6.2									5.1	
23.06.2000	M2	3	6.3									6.1	
24.07.2000	M2	3	5.97		0.15							8.6	
25.07.2000	M2	3	6.08		0.11							11.8	
26.07.2000	M2	3	6.12		0.065							11	
27.07.2000	M2	3	5.68		0.05							4.7	
28.07.2000	M2	3	5.98		0.03							3.4	
29.07.2000	M2	3	5.82		0.04							3.5	
30.07.2000	M2	3	5.96		0.07							4.1	
31.07.2000	M2	3	5.93		0.08							3.8	
01.08.2000	M2	3	6.2		0.15							3.7	
02.08.2000	M2	3	5.87		0.1							3.95	
03.08.2000	M2	3	6.25		0.05							3.7	
04.08.2000	M2	3	5.05		0.045							3.8	
05.08.2000	M2	3	5.85		0.035							4	
06.08.2000	M2	3	6.4		0.04							3.8	
07.08.2000	M2	3	6.43		0.02							4	
08.08.2000	M2	3	5.73		0.045							4	
11.08.2000	M2	3	6.67		0.005							4.1	
14.08.2000	M2	3	5.92		0.04							3.9	
17.08.2000	M2	3	6.18		0								
17.08.2000	M2	3	6.03									4	
20.08.2000	M2	3	6.2									3.8	72
23.08.2000	M2	3	6.13									4.15	84
26.08.2000	M2	3	6.14									4.4	78
29.08.2000	M2	3	6.22										52

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
01.09.2000	M2	3	6.19										45
04.09.2000	M2	3	6.31									4.9	16
07.09.2000	M2	3	6.3										-2
10.09.2000	M2	3	6.38										-19
13.09.2000	M2	3	5.72									4.8	-23
30.09.2000	M2	3	6.44									5.2	
23.05.2000	M2	4	6.51	215	0.15								
30.05.2000	M2	4	6.44	217	0								
07.06.2000	M2	4									3.55		
10.06.2000	M2	4	6.5									0.5	
14.06.2000	M2	4	6.65										
20.06.2000	M2	4	6.12									4.8	
23.06.2000	M2	4	6.45									4.8	
25.06.2000	M2	4	6.35									11.3	
30.06.2000	M2	4	5.84									10.6	
04.07.2000	M2	4	6.38									11.2	
07. Jul	M2	4	5.83									12	
09.07.2000	M2	4	6.39									11	
12.07.2000	M2	4	6.08									11.3	
15.07.2000	M2	4	6.39									11.3	
18.07.2000	M2	4	6.3									11.3	
21.07.2000	M2	4	6.31									11.4	
24.07.2000	M2	4	5.98									11.5	
25.07.2000	M2	4	5.83									11.6	
26.07.2000	M2	4	5.98									8.2	
27.07.2000	M2	4	5.75									3.4	
28.07.2000	M2	4	5.65									3.4	
29.07.2000	M2	4	6.39									3.4	
30.07.2000	M2	4	6.12									3.7	
31.07.2000	M2	4	5.5									3.6	
01.08.2000	M2	4	5.7									3.7	
02.08.2000	M2	4	6									3.6	

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
03.08.2000	M2	4	6.26									3.7	
04.08.2000	M2	4	5.92									3.6	
05.08.2000	M2	4	5.78									3.6	
06.08.2000	M2	4	6.44									3.5	
07.08.2000	M2	4	6.03									3.55	
08.08.2000	M2	4	5.97									3.5	
11.08.2000	M2	4	5.55									3.5	
14.08.2000	M2	4	5.87									3.4	
17.08.2000	M2	4	6.15										
17.08.2000	M2	4	6.19									3.55	
23.08.2000	M2	4	6.18									3.85	33
26.08.2000	M2	4	6.29									4	-5
29.08.2000	M2	4	6.34										2
01.09.2000	M2	4	6.37										-16
04.09.2000	M2	4	6.37									4.7	-22
07.09.2000	M2	4	6.34										-34
10.09.2000	M2	4	6.42										-50
13.09.2000	M2	4	5.56									4.8	-45
30.09.2000	M2	4	6.38									5.1	
23.05.2000	M2	5	6.5	218	0.15								
30.05.2000	M2	5	6.46	217	0								
07.06.2000	M2	5									3.2		
10.06.2000	M2	5	6.52									0.49	
14.06.2000	M2	5	6.7										
17.08.2000	M2	5	6.3										
23.05.2000	M2	6	6.5	218	0.1								
30.05.2000	M2	6	6.47	218									
07.06.2000	M2	6									3.9		
10.06.2000	M2	6	6.54									0.47	
14.06.2000	M2	6	6.7										
17.08.2000	M2	6	6.51										
23.05.2000	M2	7	6.51	215	0.06								

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
30.05.2000	M2	7	6.48	216									
07.06.2000	M2	7									3.9		
10.06.2000	M2	7	6.54									0.46	
14.06.2000	M2	7	6.72										
26.06.2000	M2	7	6.22									7.6	
23.05.2000	M2	8	6.52	211	0.1								
30.05.2000	M2	8	6.49	213									
07.06.2000	M2	8									4.45		
10.06.2000	M2	8	6.56									0.46	
14.06.2000	M2	8	6.74										
23.05.2000	M2	9	6.5	212	0.09								
30.05.2000	M2	9	6.5	212									
07.06.2000	M2	9									4.35		
10.06.2000	M2	9	6.54									0.46	
14.06.2000	M2	9	6.74										
23.05.2000	M2	10	6.4	267	0.01								
30.05.2000	M2	10	6.5	217									
07.06.2000	M2	10									4.5		
10.06.2000	M2	10	6.55									0.45	
14.06.2000	M2	10	6.75										
23.05.2000	M2	11	6.52	219	0.11								
30.05.2000	M2	11	6.49	220									
07.06.2000	M2	11									4		
10.06.2000	M2	11	6.54									0.45	
14.06.2000	M2	11	6.73										
23.05.2000	M2	12	6.5	220	0.09								
30.05.2000	M2	12	6.51	219									
07.06.2000	M2	12									4.35		
10.06.2000	M2	12	6.55									0.48	
14.06.2000	M2	12	6.76										
23.05.2000	M2	13	6.5	214	0.03								
30.05.2000	M2	13	6.52	213									

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
07.06.2000	M2	13									3.7		
10.06.2000	M2	13	6.56									0.46	
14.06.2000	M2	13	6.77										
23.05.2000	M2	14	6.51	222	0.05								
30.05.2000	M2	14	6.52	222									
07.06.2000	M2	14									4.05		
10.06.2000	M2	14	6.57									0.48	
14.06.2000	M2	14	6.78										
23.05.2000	M2	15	6.51	222	0.04								
30.05.2000	M2	15	6.53	220									
07.06.2000	M2	15									2.95		
10.06.2000	M2	15	6.56									0.47	
14.06.2000	M2	15	6.79										
20.08.2000	M2	4	6.2									3.5	25
23.05.2000	М3	1	6.71	265	0.06								
30.05.2000	M3	1	6.38	254									
26.06.2000	М3	1	6.42									6.8	
23.05.2000	М3	2	6.64	271	0.1								
30.05.2000	М3	2	6.38	264									
20.06.2000	М3	2	6.21									7.4	
26.06.2000	M3	2	6.43									11.6	
17.08.2000	М3	2	6.32										
23.05.2000	М3	3	6.63	265	0.1								
30.05.2000	M3	3	6.4	256									
25.06.2000	M3	3	6.35									8.6	
26.06.2000	M3	3	6.53									15.8	
28.06.2000	M3	3	6.15									13.5	
30.06.2000	M3	3	6.11									12.8	
06.07.2000	M3	3	6.11										
07. Jul	M3	3	5.91									10.8	
09.07.2000	M3	3	6.29									11.1	
12.07.2000	M3	3	6.17									11.4	

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
15.07.2000	M3	3	6.42									11.7	
18.07.2000	M3	3	6.46									12.1	
21.07.2000	M3	3	6.52									12.2	
17.08.2000	M3	3	6.2										
30.09.2000	M3	3	6.2									4.1	
23.05.2000	M3	4	6.66	224	0.11								
30.05.2000	M3	4	6.46	217									
25.06.2000	M3	4	6.48									14.5	
26.06.2000	M3	4	6.39									14.2	
28.06.2000	M3	4	6.2									13	
30.06.2000	M3	4	6.15									12.4	
04.07.2000	M3	4	6.33									11.5	
12.07.2000	M3	4	6.22									11.7	
15.07.2000	M3	4	6.39									11.8	
18.07.2000	M3	4	6.34									11.9	
21.07.2000	M3	4	6.32									11.6	
24.07.2000	M3	4	5.94									11.7	
25.07.2000	M3	4	6.15									11.8	
26.07.2000	M3	4	6.12									11.9	
27.07.2000	M3	4	5.73									12.1	
28.07.2000	M3	4	6.55									12.1	
29.07.2000	M3	4	5.85									11.1	
30.07.2000	M3	4	5.82									9	
31.07.2000	M3	4	6.3									5.9	
01.08.2000	M3	4	5.81									4.2	
02.08.2000	M3	4	6.03									3.65	
03.08.2000	M3	4	6.26								3.5		
04.08.2000	M3	4	6.04								3.45		
05.08.2000	M3	4	5.84								3.45		
06.08.2000	M3	4	6.34								3.55		
07.08.2000	M3	4	6.14								3.55		
08.08.2000	M3	4	6.2									3.55	

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
11.08.2000	M3	4	5.8									3.7	
14.08.2000	M3	4	5.78									4.6	
17.08.2000	M3	4	6.26										
17.08.2000	M3	4	6.29									4	
20.08.2000	M3	4	6.2									4	53
23.08.2000	M3	4	6.22									4	62
26.08.2000	M3	4	6.21									3.9	65
29.08.2000	M3	4	6.26										74
01.09.2000	M3	4	6.27										56
04.09.2000	M3	4	6.2									3.8	61
07.09.2000	M3	4	6.2										60
10.09.2000	M3	4	6.31										40
13.09.2000	M3	4	5.5									3.6	49
30.09.2000	M3	4	6.18									3.6	
23.05.2000	M3	5	6.61	217	0.06								
30.05.2000	M3	5	6.47	216									
26.06.2000	M3	5	6.25									15.7	
17.08.2000	M3	5	6.37										
23.05.2000	M3	6	6.61	222	0.05								
30.05.2000	M3	6	6.5	220									
26.06.2000	M3	6	6.2									13.4	
17.08.2000	M3	6	6.25										
23.05.2000	M3	7	6.6	228	0.11								
30.05.2000	M3	7	6.5	219									
23.05.2000	M3	8	6.6	222	0.05								
30.05.2000	M3	8	6.51	217									
23.05.2000	M3	9	6.59	218	0.04								
30.05.2000	M3	9	6.51	215									
23.05.2000	M3	10	6.58	215	0.1								
30.05.2000	M3	10	6.52	212									
23.05.2000	M3	11	6.58	213	0.06								
30.05.2000	M3	11	6.52	216									

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
23.05.2000	M3	12	6.56	218	0.13								
30.05.2000	M3	12	6.52	218									
23.05.2000	M3	13	6.57	219	0.08								
30.05.2000	M3	13	6.53	220									
23.05.2000	M3	14	6.57	215	0.09								
30.05.2000	M3	14	6.54	215									
23.05.2000	M3	15	6.57	218	def.								
30.05.2000	M3	15	6.54	220									
23.05.2000	M5	1	6.64	262	0.1								
30.05.2000	M5	1	6.4	264									
23.05.2000	M5	2	6.58	264	0.06								
30.05.2000	M5	2	6.4	260									
20.06.2000	M5	2	6.12									7.2	
28.06.2000	M5	2	6.3									8.7	
23.05.2000	M5	3	6.61	232	0.07								
30.05.2000	M5	3	6.45	233									
26.06.2000	M5	3	6.56									5	
28.06.2000	M5	3	6.46									6.3	
30.06.2000	M5	3	6.12									6.5	
04.07.2000	M5	3	6.62									13.2	
06.07.2000	M5	3	6.28									12.7	
09.07.2000	M5	3	6.28									11.4	
12.07.2000	M5	3	6.12									10.7	
15.07.2000	M5	3	6.31									9.2	
18.07.2000	M5	3	6.33									10.3	
21.07.2000	M5	3	6.41								12		
24.07.2000	M5	3	6.18								12.5		
25.07.2000	M5	3	6.24								11.9		
26.07.2000	M5	3	5.98								12.2		
27.07.2000	M5	3	5.81								12.3		
28.07.2000	M5	3	5.85								12		
29.07.2000	M5	3	5.9									13.2	

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
30.07.2000	M5	3	5.7									12.1	
31.07.2000	M5	3	5.94									11.7	
01.08.2000	M5	3	5.93									8.1	
02.08.2000	M5	3	5.07									5.1	
03.08.2000	M5	3	6.49									4.65	
04.08.2000	M5	3	6.06									3.9	
05.08.2000	M5	3	5.79									3.65	
06.08.2000	M5	3	6.47									3.6	
07.08.2000	M5	3	5.82									3.4	
08.08.2000	M5	3	6.41									3.6	
11.08.2000	M5	3	5.95									3.9	
14.08.2000	M5	3	6.27									4	
17.08.2000	M5	3	5.81									4.1	
20.08.2000	M5	3	6.25									6.5	-12
23.08.2000	M5	3	6.27									4.3	6
26.08.2000	M5	3	6.31									4.45	-15
29.08.2000	M5	3	6.29										44
01.09.2000	M5	3	6.3										7
04.09.2000	M5	3	6.22									4.1	35
07.09.2000	M5	3	6.2										46
10.09.2000	M5	3	6.34										24
13.09.2000	M5	3	6.2									4.1	2
30.09.2000	M5	3	6.25									4	
23.05.2000	M5	4	6.62	217	0.02								
30.05.2000	M5	4	6.48	218									
28.06.2000	M5	4	6.35									8.1	
30.06.2000	M5	4	6.15									12	
30.09.2000	M5	4	6.25									4.1	
23.05.2000	M5	5	6.59	222	0.24								
30.05.2000	M5	5	6.5	222							↓		
28.06.2000	M5	5	6.2								4.8		
23.05.2000	M5	6	6.58	224	0.05								

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
30.05.2000	M5	6	6.5	222									
23.05.2000	M5	7	6.59	218	0.15								
30.05.2000	M5	7	6.52	218									
23.05.2000	M5	8	6.57	218	0.27								
30.05.2000	M5	8	6.53	220									
23.05.2000	M5	9	6.58	213	0.05								
30.05.2000	M5	9	6.54	215									
23.05.2000	M5	10	6.58	213	0.07								
30.05.2000	M5	10	6.54	216									
23.05.2000	M5	11	6.58	206	0.2								
30.05.2000	M5	11	6.53	220									
23.05.2000	M5	12	6.57	214	0.02								
30.05.2000	M5	12	6.54	218									
23.05.2000	M5	13	6.57	215	0.02								
30.05.2000	M5	13	6.55	217									
23.05.2000	M5	14	6.58	214	0.14								
30.05.2000	M5	14	6.56	221									
23.05.2000	M5	15	6.58	215	0.08								
30.05.2000	M5	15	6.58	221									
23.05.2000	M8	1	6.6	233	0.09								
30.05.2000	M8	1	6.34	232									
23.05.2000	M8	2	6.54	218	0.07								
30.05.2000	M8	2	6.36	216									
23.05.2000	M8	3	6.54	223	0								
30.05.2000	M8	3	6.41	218									
23.05.2000	M8	4	6.55	214	0.03								
30.05.2000	M8	4	6.42	210									
23.05.2000	M8	5	6.58	208	0.07								
30.05.2000	M8	5	6.44	211									
23.05.2000	M8	6	6.57	205	0.08								
30.05.2000	M8	6	6.47	211									
23.05.2000	M8	7	6.57	206	0								

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
30.05.2000	M8	7	6.48	212									
23.05.2000	M8	8	6.57	211	0								
30.05.2000	M8	8	6.49	218									
23.05.2000	M8	9	6.56	210	0.07								
30.05.2000	M8	9	6.5	218									
23.05.2000	M8	10	6.57	213	0.01								
30.05.2000	M8	10	6.52	219									
23.05.2000	M8	11	6.58	204	0.02								
30.05.2000	M8	11	6.55	221									
23.05.2000	M8	12	6.56	213	0.07								
30.05.2000	M8	12	6.54	220									
23.05.2000	M8	13	6.57	212	0								
30.05.2000	M8	13	6.55	218									
23.05.2000	M8	14	6.58	209	0.05								
30.05.2000	M8	14	6.56	214									
23.05.2000	M8	15	6.58	207	0.01								
30.05.2000	M8	15	6.57	211									
20.06.2000	M9	2	6.07									7.1	
06.07.2000	M9	2	6.1									14.5	
06.07.2000	M9	3	6.08									15.2	
12.07.2000	M9	4	6.16									12.8	
15.07.2000	M9	4	6.54									12.9	
18.07.2000	M9	4	6.36									12.1	
24.07.2000	M9	4	6.19									11.2	
27.07.2000	M9	4	5.71									11.2	
30.07.2000	M9	4	5.7									10.9	
02.08.2000	M9	4	5.97									7.4	
03.08.2000	M9	4	6.41									6.3	
04.08.2000	M9	4	6.04									6.3	
05.08.2000	M9	4	5.76								6		
06.08.2000	M9	4	6.49									5.7	
07.08.2000	M9	4	6.24									5.7	

date	MLS	Level	рН	conductivity (µS/cm)	oxygen (ppm)	Fe (ppm)	Br (mg/l)	As (mg/l)	altitude( Fuß)	рН	phosphate (ppm)	alkalinity (0,1m)(ml)	eH (mV)
08.08.2000	M9	4	5.45									5.7	
11.08.2000	M9	4	5.65									5.4	
14.08.2000	M9	4	6.26									5.5	
17.08.2000	M9	4	6.19									5.5	
20.08.2000	M9	4	6.33									5.5	-42
23.08.2000	M9	4	6.35									5.4	-15
26.08.2000	M9	4	6.38									5.2	-46
29.08.2000	M9	4	6.35										7
01.09.2000	M9	4	6.36										-24
04.09.2000	M9	4	6.21									4.95	9
07.09.2000	M9	4	6.19										18
10.09.2000	M9	4	6.35										-7
13.09.2000	M9	4	6.09									4.9	-11
30.09.2000	M9	4	6.36									4.7	

#### Tab.: A16 anions from M2

				a and until vitry data at a r				
	wavelength de	etector				conductivity	/ detector	
sample	CI	Br	NO3	PO4	SO4	Br	NO3	
PT01								
3.7. 9.00	13.48	126.77	5.65	2.56	9.21	129.39	5.14	
GNT02								
3.7. 9.00	11.57	118.43	4.81	2.20	8.14	120.16	5.86	
BUT04				-				
3.7. 9.00	7.98	50.57	1.86	0.00	4.07	53.10	1.95	
BKT05								
3.7. 9.00	3.85	23.63	0.00	0.00	2.86	30.16	0.50	
WT06								
3.7. 9.00	9.03	102.00	0.00	0.00	6.71	115.77	3.26	
O07								
3.7. 9.00	13.13	0.00	0.00	0.00	9.36	0.98	0.69	
### Tab.: A17 anions from M3

	conductivity	detector					
sample	CI	Br	NO3	PO4	SO4	Br	NO3
PT 01							
standard1	15.73	4.34	0.95	2.44	4.52	5.30	1.07
26.6. 15.00	48.73	0.00	0.00	0.00	11.42	0.00	0.00
26.6. 19.00	6.56	0.00	0.00	0.00	2.31	0.00	0.00
26.6.23.00	29.63	34.28	0.00	0.00	8.38	43.53	2.16
27.6. 7.00	4.29	0.00	0.00	0.00	0.00	0.00	0.45
27.6. 19.00	46.57	0.00	0.00	0.00	10.35	0.00	0.26
28.6. 7.00	7.98	0.00	0.00	0.00	2.09	0.00	0.00
29.06. 23.00	21.16	26.33	2.08	0.00	7.63	33.81	2.41
GNT 02	22.04	0.00	4.00	E 4 E	0.24	10.00	2.04
	33.01	9.02	1.86	5.15	9.34	10.29	2.04
25.6. 3.00	71.58	0.00	0.00	0.00	13.05	0.00	0.04
25.6.7.00	17.64	1.69	0.00	0.00	4.42	2.28	0.16
25.6. 11.00	14.72	0.53	0.00	0.00	3.35	0.83	0.06
25.6. 15.00	51.55	6.90	0.00	0.00	11.99	8.99	0.44
25.6. 19.00	19.57	3.72	0.00	0.00	6.35	6.74	1.48
25.6. 23.00	29.71	14.53	0.00	0.00	10.29	20.07	1.05
26.6. 7.00	11.30	12.91	0.00	0.00	3.48	18.84	0.92
26.6. 11.00	32.79	48.88	0.00	0.00	13.43	60.75	3.35
26.6. 15.00	16.80	29.09	0.00	0.00	6.28	37.83	2.35
26.6. 19.00	6.29	8.94	0.00	0.00	2.83	11.97	1.79
27.6. 7.00	19.55	49.09	0.00	0.00	8.07	61.65	2.41
27.6. 19.00	39.34	106.63	5.74	0.00	16.98	120.67	6.98
28.6. 7.00	12.99	35.60	3.29	0.00	6.88	41.59	3.92
29.6. 7.00	15.82	51.70	0.00	0.00	6.83	66.25	2.75
29.6. 15.00	28.18	112.14	2.98	0.00	11.77	124.74	4.74
29.6. 23.00	26.88	88.78	4.31	0.00	12.46	103.05	5.11
30.6. 7.00	9.01	37.16	0.00	0.00	3.99	48.31	1.78
24.7.9.00	14.80	86.36	4.83	0.00	7.55	93.74	6.00
25.7.9.00	18.67	124.49	5.36	0.00	9.44	139.77	6.38
26.7. 9.00	10.89	67.53	0.00	0.00	6.86	81.28	2.89
27.7.9.00	4.43	30.18	0.00	0.00	3.06	37.01	0.61
28.7. 9.00	105.61	136.43	0.00	0.00	17.90	154.29	5.79
29.7. 9.00	3.19	22.72	0.00	0.00	2.49	32.55	0.76
30.7. 9.00	15.41	101.80	0.00	0.00	9.57	118.25	2.38
31.7. 9.00	19.59	125.50	0.00	0.00	10.62	138.15	4.53
1.8. 9.00	25.25	75.69	0.00	0.21	9.31	55.43	0.98
2.8. 9.00	27.40	179.94	0.00	0.00	12.99	104.54	1.17
3.8. 9.00	29.59	155.38	0.00	0.00	13.95	107.72	0.00
4.8. 9.00	31.17	112.01	0.00	0.00	14.68	85.29	0.00
5.8. 9.00	7.88	18.44	0.00	0.00	4.20	20.23	0.00
6.8. 9.00	25.19	41.43	0.00	0.18	11.14	38.81	0.00
7.8. 9.00	11.20	10.18	0.00	0.00	4.93	11.66	0.00
8.8. 19.00	6.46	2.83	0.00	0.00	4.20	3.41	0.68
09.8. 9.00	6.88	1.99	0.00	0.00	4.03	1.79	0.55
09.8. 19.00	22.84	0.88	6.24	1.26	13.94	1.42	8.81
10.8. (sp3)	6.87	1.29	0.00	0.00	4.68	1.03	1.12
11.8.9.00	3.94	0.00	0.00	0.00	2.94	0.00	1.49
11.8. 19.00	8.74	0.00	0.00	0.00	5.11	0.00	0.32

	wavelength de	etector	conductivity detector				
sample	CI	Br	NO3	PO4	SO4	Br	NO3
12.8. 9.00	26.68	1.43	0.00	0.00	14.73	1.42	0.70
12.8. 19.00	19.08	0.58	0.00	0.00	10.51	0.57	0.41
13.8. 9.00	16.29	0.00	0.00	0.00	9.43	0.00	0.54
13.8. 19.00	6.01	0.00	0.65	0.00	3.81	0.00	0.79
14.8. 9.00	19.01	0.00	0.00	0.00	10.62	0.73	0.30
14.8. 19.00	5.75	0.00	0.00	0.00	3.61	0.00	0.87
RT03	92 23	19.96	5 14	9 37	21 20	19.94	5 18
25.6. 3.00	23.99	6.84	0.32	0.00	5.45	8.25	0.47
25.6.7.00	37.31	18.66	0.63	0.00	9 47	19.16	0.76
25.6 11.00	35.43	2 50	0.00	0.00	8 49	3 36	0.21
25.6 15.00	6 66	0.00	0.00	0.00	2.98	2 67	0.40
25.6 19.00	18 47	0.00	1 50	0.00	7.95	1.93	2 37
25.6.23.00	12 10	12 54	2.33	0.00	5.92	17 69	3.06
26.6.7.00	18.36	52 49	0.00	0.00	9.02	65 43	2 74
26.6 11.00	28.20	94 77	4 46	0.00	11.38	106.31	5.67
26.6.15.00	7 77	27.62	0.00	0.00	4 88	36 21	1 45
26.6 19.00	13.08	51.24	0.00	0.00	6.61	64.52	2 48
26.6.23.00	11 16	22.26	3.27	0.00	6.86	28.66	4.38
27.6.7.00	13 13	44 52	0.00	0.00	5 46	56.91	2 12
27.6 19.00	29.42	109.81	5.83	0.00	13.09	122 73	6.94
28.6.7.00	28.99	91 54	3.95	0.00	11.38	106.90	5 25
29.6.7.00	17 41	63.02	3.06	0.00	7.91	74 27	3.96
29.6. 15.00	18.57	8.20	0.00	0.00	4.52	12.48	1.27
29.6. 23.00	16.54	63.46	2.53	0.00	7.51	74.57	3.10
30.6.7.00	26.54	100.74	5.26	0.00	11.14	114.95	6.33
3.7. 9.00	7.46	88.35	0.00	0.00	5.30	97.06	2.77
3.7. 19.00	10.19	107.61	0.00	0.00	7.05	119.15	3.21
4.7. 9.00	14.54	134.85	0.00	0.00	9.15	136.78	4.54
5.7.9.00	12.79	149.44	0.00	0.00	9.05	162.27	0.00
5.7. 19.00	11.61	123.82	0.00	0.00	7.96	140.26	0.00
6.7. 9.00	13.66	149.69	0.00	0.00	9.65	167.12	0.00
6.7. 19.00	7.99	90.22	0.00	0.00	5.44	105.34	0.00
7.7.9.00	13.95	151.13	0.00	0.00	9.44	165.68	0.00
8.7. 9.00	13.18	171.07	0.00	0.00	8.97	188.71	0.00
8.7. 19.00	14.73	176.51	0.00	0.00	9.65	166.22	0.00
10.7. 19.00	15.80	135.12	0.00	0.00	9.36	147.70	4.34
11.7. 9.00	15.11	149.42	0.00	0.00	10.01	158.98	4.15
12.7. 9.00	11.42	86.37	3.32	0.00	6.39	118.91	6.21
13.7. 9.00	14.31	109.29	4.22	0.00	8.02	140.12	6.13
14.7. 9.00	13.27	96.75	4.15	0.00	7.59	124.25	6.55
15.7.9.00	12.07	89.96	4.17	0.00	7.75	114.39	5.82
16.7.9.00	12.26	96.62	4.43	0.00	7.94	119.69	6.16
17.7.9.00	14.81	109.87	4.81	0.00	8.48	133.11	6.70
18.7.9.00	4.81	33.18	0.00	0.00	2.69	45.07	0.00
19.7. 9.00	15.49	107.95	4.94	0.00	9.12	128.34	6.21
20.7.9.00	16.49	117.56	5.17	0.00	8.73	137.10	7.02
21.7.9.00	15.96	103.41	5.52	0.00	9.02	120.85	7.52
22.7.9.00	16.26	105.28	5.62	0.00	9.33	120.76	6.94
23.7.9.00	15.67	107.57	5.51	0.00	9.10	124.83	7.68
24.7.9.00	16.08	113.45	6.03	0.00	9.28	128.05	7.58

	wavelength de		conductivity detector				
sample	CI	Br	NO3	PO4	SO4	Br	NO3
25.7. 9.00	17.04	116.34	5.80	0.00	9.19	131.01	7.45
26.7. 9.00	9.83	65.65	2.28	1.48	5.44	54.50	2.33
27.7.9.00	23.55	57.41	1.89	1.29	5.30	48.50	1.95
28.7. 9.00	9.98	22.55	0.00	1.83	5.64	25.90	0.00
29.7. 9.00	72.73	45.33	1.81	2.03	14.45	39.47	1.69
30.7. 9.00	10.90	28.16	0.94	0.39	5.71	27.18	1.01
31.7. 9.00	32.12	10.72	0.00	1.76	16.80	13.04	0.00
1.8. 9.00	31.15	1.53	0.00	1.98	16.86	2.04	0.00
2.8. 9.00	31.22	0.23	0.00	1.93	16.79	0.35	0.00
3.8. 9.00	31.16	0.14	0.00	1.81	16.85	0.16	0.00
4.8. 9.00	31.32	0.10	0.00	1.89	17.08	0.16	0.00
5.8. 9.00	7.65	0.00	0.00	0.87	5.27	0.08	0.00
6.8. 9.00	31.64	0.10	0.00	1.74	17.72	0.14	0.00
7.8. 9.00	9.30	0.00	0.00	1.05	5.86	0.10	0.00
8.8. 19.00	4.15	0.00	0.10	0.78	2.82	0.00	0.08
09.8. 9.00	5.09	0.00	0.00	0.97	3.81	0.00	0.02
09.8.019.00	6.60	0.00	0.00	1.45	5.28	0.00	0.02
10.8. (sp3)	4.36	0.00	0.00	0.00	4.02	0.00	0.74
11.8. 9.00	25.35	0.48	0.00	1.46	14.49	1.13	0.10
11.8. 19.00	18.75	0.00	0.00	1.76	12.23	0.21	0.03
12.8. 9.00	16.48	0.00	0.00	2.12	12.59	0.00	0.04
12.8. 19.00	26.59	0.00	0.13	1.93	16.37	0.00	0.17
13.8. 9.00	29.96	0.62	0.00	2.13	18.44	0.96	0.19
13.8. 19.00	6.69	0.00	0.00	1.16	4.37	0.08	0.07
14.8. 9.00	19.63	0.26	0.00	1.96	12.64	0.30	0.04
14.8. 19.00	5.95	0.00	0.00	1.57	5.37	0.00	0.02

standard1	15.72	4.22	1.12	2.22	4.51	5.19	1.21
25.6. 3.00	14.57	0.00	0.48	0.00	5.58	0.00	0.62
25.6. 7.00	34.60	9.38	0.32	0.00	10.59	10.71	0.43
25.6. 11.00	17.72	13.07	0.88	0.00	6.46	14.09	0.98
25.6. 15.00	18.49	24.44	1.86	0.00	8.70	31.84	3.22
25.6. 19.00	20.85	39.73	2.89	0.00	8.34	50.31	3.32
25.6. 23.00	28.76	98.23	3.82	0.00	13.05	110.10	5.22
26.6. 7.00	17.77	24.87	2.35	0.00	7.08	33.06	3.89
26.6. 11.00	26.20	71.66	6.77	0.00	12.67	85.41	8.15
26.6. 15.00	31.66	91.70	7.52	0.00	13.24	104.38	8.00
26.6. 19.00	20.72	87.75	2.79	0.00	9.25	100.72	3.76
26.6. 23.00	18.62	77.22	2.36	0.00	8.04	89.69	3.25
27.6. 7.00	26.95	108.78	4.60	0.00	12.31	122.01	5.40
27.6. 19.00	28.89	116.18	4.96	0.00	12.47	127.23	6.10
28.6. 7.00	12.33	50.16	0.00	0.00	6.76	63.01	2.51
29.6. 7.00	25.40	100.37	4.59	0.00	10.58	113.66	5.27
29.6. 15.00	17.26	56.96	1.72	0.00	8.61	68.51	3.02
29.6. 23.00	16.97	57.42	2.77	0.00	7.23	69.31	3.79
30.6. 7.00	21.14	86.70	2.66	0.00	9.10	100.22	3.60
2.7. 9.00	12.59	88.63	0.00	0.00	6.95	101.44	2.39
2.7. 19.00	13.86	174.98	0.00	0.00	9.41	169.32	3.09
3.7. 9.00	13.44	154.62	0.00	0.00	9.64	154.59	2.68
3.7. 19.00	13.35	152.89	0.00	1.70	9.77	151.30	2.41

wavelength detector						conductivity detector	
sample	CI	Br	NO3	PO4	SO4	Br	NO3
4.7. 9.00	14.47	134.87	0.00	0.00	9.59	144.83	3.52
5.7. 9.00	15.37	150.86	0.00	0.00	9.64	149.95	0.00
5.7. 19.00	14.79	150.59	0.00	0.00	9.37	149.27	0.00
6.7. 9.00	14.52	148.30	0.00	0.00	9.06	149.18	0.00
6.7. 19.00	12.45	131.47	0.00	0.00	9.11	134.81	0.00
7.7.9.00	14.32	152.82	0.00	0.76	9.26	157.43	1.81
7.7. 19.00	15.45	154.57	0.00	0.00	9.36	151.43	3.65
8.7. 9.00	14.39	162.73	0.00	0.00	8.81	152.75	3.65
8.7. 19.00	14.79	136.00	0.00	0.00	8.83	150.99	3.57
10.7. 9.00	15.38	130.77	0.00	0.00	9.41	147.21	4.43
10.7. 19.00	633.45	156.16	4.05	0.00	11.82	160.91	5.44
11.7. 9.00	15.26	138.65	0.00	0.00	9.23	155.28	4.71
12.7. 9.00	20.19	125.61	4.68	1.37	9.98	132.23	6.73
13.7. 9.00	19.86	143.48	5.02	0.00	10.14	146.65	7.09
14.7. 9.00	18.63	125.38	5.51	0.00	9.79	129.47	6.52
15.7. 9.00	17.79	118.26	5.02	0.00	9.62	125.98	7.60
16.7. 9.00	15.75	95.27	5.00	0.00	8.80	102.13	6.97
17.7. 9.00	14.10	94.57	5.00	0.00	8.32	101.37	6.51
18.7. 9.00	19.61	128.62	6.33	0.00	10.89	132.57	8.26
19.7. 9.00	18.76	125.31	5.78	0.00	9.79	130.79	8.35
20.7. 9.00	19.31	145.42	6.76	0.00	10.35	147.06	8.50
21.7. 9.00	14.17	89.98	5.05	0.91	8.26	95.82	6.41
22.7. 9.00	18.92	121.19	6.51	2.13	11.14	125.92	8.47
23.7. 9.00	18.67	123.91	6.15	1.49	10.06	127.66	7.79
24.7. 9.00	19.15	127.77	5.50	3.40	9.37	132.61	8.00
25.7.9.00	18.62	129.52	6.20	1.88	10.79	134.36	7.68
26.7. 9.00	11.82	79.72	2.61	2.65	6.24	58.57	2.62
27.7.9.00	6.18	41.39	1.67	2.24	3.39	34.90	1.68
28.7. 9.00	3.39	20.58	0.00	1.74	1.75	22.08	0.00
29.7.9.00	11.89	60.67	2.19	2.34	6.45	47.22	2.21
30.7. 9.00	3.55	11.93	0.00	2.19	2.46	13.61	0.00
31.7.9.00	31.16	11.65	0.00	4.44	19.53	12.27	0.16
1.8. 9.00	11.59	0.20	0.00	2.42	8.40	0.13	0.00
2.8. 9.00	32.02	0.14	0.18	3.14	20.47	0.15	0.31
3.8. 9.00	37.52	2.29	0.00	2.46	21.35	2.93	0.00

BKT (	05
-------	----

standard3	50.38	20.25	4.91	9.83	20.25	20.02	5.04
25.6. 3.00	18.55	0.53	0.21	0.00	9.15	0.74	0.31
25.6. 7.00	8.09	0.49	0.00	0.00	4.52	0.72	0.05
25.6. 11.00	31.81	0.00	0.00	0.00	15.64	0.00	0.08
25.6. 15.00	8.07	0.00	0.00	0.00	6.75	0.59	1.94
25.6. 19.00	11.25	3.76	2.37	0.00	8.36	5.36	3.08
25.6. 23.00	24.64	0.00	1.55	0.00	14.56	0.00	1.94
26.6. 11.00	10.90	60.61	0.00	0.00	8.97	70.60	5.63
26.6. 15.00	49.12	161.29	5.77	0.00	24.89	164.64	6.20
26.6. 19.00	10.71	86.33	0.00	0.00	9.39	101.05	3.53
26.6. 23.00	13.42	90.12	4.71	0.00	11.17	103.29	5.68
27.6. 7.00	17.07	115.56	7.61	0.00	13.46	125.78	8.17
27.6. 19.00	29.18	155.50	5.22	0.00	18.04	159.88	7.32
28.6. 7.00	7.01	44.07	0.00	0.00	5.41	56.15	2.72
29.6. 7.00	15.40	96.39	5.67	0.00	12.71	109.00	6.48
29.6. 15.00	10.68	50.61	4.99	0.00	8.18	61.53	6.18

	wavelength detector						
sample	CI	Br	NO3	PO4	SO4	Br	NO3
29.6. 23.00	13.20	83.58	4.44	0.00	9.89	95.25	5.68
3.7. 9.00	13.33	128.79	0.00	0.00	8.79	133.36	1.91
3.7. 19.00	10.62	124.82	0.00	0.00	7.55	133.49	2.11
4.7. 9.00	14.68	149.73	0.00	0.00	9.58	158.25	1.24
5.7. 9.00	12.47	129.99	0.00	0.00	8.54	148.63	0.00
5.7. 19.00	13.97	142.34	0.00	0.00	9.22	162.19	0.00
6.7. 9.00	14.34	142.64	0.00	0.00	8.53	163.31	0.00
7.7. 9.00	14.33	148.37	0.00	0.00	9.21	166.35	0.00
7.7. 19.00	14.02	146.44	0.00	0.00	9.94	167.83	0.00
8.7. 9.00	14.69	148.89	0.00	0.00	9.65	168.79	0.00
8.7. 19.00	15.26	167.08	0.00	0.00	9.15	186.05	0.00
24.7. 9.00	20.12	152.04	7.26	0.00	10.84	150.16	7.55
25.7.9.00	13.81	92.03	3.64	0.00	7.67	100.90	4.17
26.7. 9.00	14.89	66.40	2.51	0.00	6.52	74.65	4.62
27.7.9.00	8.29	53.97	0.00	0.00	5.49	67.86	2.87
28.7. 9.00	11.86	72.71	0.00	0.00	6.69	85.51	3.92
29.7. 9.00	19.34	123.10	5.33	0.00	10.09	134.06	6.39
30.7. 9.00	12.67	66.30	0.00	0.00	7.50	84.39	3.91
31.7. 9.00	29.01	11.45	0.00	4.06	19.28	16.08	0.55
1.8. 9.00	41.26	1.73	0.00	5.76	23.98	2.56	0.16
2.8. 9.00	15.86	1.35	0.00	1.79	11.18	1.64	0.07
3.8. 9.00	37.53	0.68	0.00	2.29	23.83	0.79	0.12

WT 06							
standard1	8.71	4.11	1.23	2.32	4.52	5.10	1.31
25.6. 23.00	24.36	7.53	1.38	0.00	13.75	10.30	2.80
26.6. 7.00	15.93	13.05	0.00	0.00	9.86	18.37	1.05
26.6. 11.00	5.13	30.47	0.00	0.00	3.77	39.16	1.65
26.6. 15.00	10.59	49.95	2.67	0.00	8.42	60.75	4.03
26.6. 19.00	6.50	37.69	0.00	0.00	5.90	48.03	2.01
26.6. 23.00	5.74	39.81	0.00	0.00	4.79	49.33	2.34
27.6. 7.00	9.62	64.24	2.87	0.00	7.77	74.21	4.35
27.6. 19.00	12.26	78.00	5.54	0.00	10.36	89.52	5.48
28.6. 7.00	6.64	33.81	2.71	0.00	6.34	41.94	3.40
29.6. 7.00	11.24	84.53	3.26	0.00	8.69	95.94	4.69
29.6. 15.00	9.56	66.72	3.09	0.00	6.88	75.89	3.48
29.6. 23.00	9.70	67.44	2.94	0.00	8.15	77.84	3.49
30.6. 7.00	11.31	75.80	3.98	0.00	8.12	87.30	4.19

O 07							
standard2	18.44	9.07	1.82	5.40	9.13	10.29	2.01
26.6. 15.00	9.99	19.59	0.00	0.00	7.13	25.05	2.07
26.6. 19.00	10.91	43.53	0.00	0.00	7.28	54.41	1.93
26.6. 23.00	12.47	32.94	5.47	0.00	11.82	41.52	7.00
27.6. 7.00	8.61	58.27	2.89	0.00	6.75	69.23	3.98
27.6. 19.00	19.60	114.82	8.05	0.00	16.35	125.18	9.70
28.6. 7.00	5.21	37.28	0.00	0.00	4.66	46.68	2.25
29.6. 7.00	12.94	96.48	3.75	0.00	9.46	108.58	4.80
29.6. 15.00	14.45	99.41	4.69	0.00	11.88	110.88	6.33
29.6. 23.00	9.28	57.44	3.27	0.00	7.04	68.26	4.12

	wavelength de	conductivity detector					
sample	CI Br NO3 PO4 SO4				Br	NO3	
GY 08							

26.6. 15.00	7.71	0.00	0.00	0.00	4.16	2.14	0.33
26.6. 19.00	4.34	0.00	0.00	0.00	2.47	1.45	0.40
26.6. 23.00	7.80	3.74	0.00	0.00	4.49	5.22	0.24
27.6.7.00	23.62	45.20	5.45	0.00	15.92	54.30	5.86
27.6. 19.00	16.17	94.25	3.99	0.00	12.41	107.02	4.36
28.6. 7.00	6.55	33.59	3.35	0.00	5.88	42.19	4.28

### Tab.: A18 anions from M4

	wavelength d		conductivity detector				
sample	CI	Br	NO3	PO4	SO4	Br	NO3
GNT02							
24.7. 9.00	39.72	103.04	0.00	0.52	18.94	74.23	0.61
			•			•	•
RT03		•		1		•	
24.7. 9.00	27.97	6.05	0.00	0.16	17.49	6.80	0.00
BUT04	0.10			0.11	1.00		
standard1	8.19	5.16	1.04	2.41	4.22	5.98	0.97
27.6. 11.00	12.00	0.00	0.00	0.00	8.02	0.00	0.00
27.6. 15.00	30.39	0.00	0.00	0.00	17.66	0.00	0.00
27.6. 19.00	30.50	0.00	0.00	0.00	18.62	0.00	0.00
27.6. 23.00	22.95	0.00	0.00	0.00	13.20	0.00	0.03
28.6. 7.00	29.64	0.00	0.00	0.00	18.46	0.00	0.00
28.6. 11.00	29.61	0.00	0.00	0.00	18.35	0.00	0.00
24.7. 9.00	30.37	30.03	0.00	0.28	16.74	28.01	0.00
BKT05							
standard1	8.19	5.16	1.04	2.41	4.22	5.98	0.97
27.6. 7.00	29.97	0.00	0.00	0.00	18.11	0.00	0.00
27.6. 11.00	12.92	0.00	0.00	0.00	8.91	0.00	0.00
27.6. 15.00	14.55	0.00	0.00	0.00	11.92	0.00	0.00
27.6. 19.00	25.77	0.00	0.00	0.00	15.75	0.00	0.00
27.6. 23.00	23.92	0.00	0.00	0.00	14.21	0.00	0.00
28.6. 7.00	20.60	0.00	0.00	0.00	9.35	0.00	0.00
28.6. 11.00	29.72	0.00	0.00	0.00	18.43	0.00	0.00
WT06							
27.6. 7.00	33.30	0.00	0.00	0.00	18.96	0.00	0.00
27.6. 11.00	22.52	0.00	0.00	0.00	13.59	0.00	0.05
27.6. 15.00	26.97	0.00	0.00	0.00	16.78	0.00	0.00
27.6. 19.00	31.17	0.00	0.00	0.00	18.08	0.00	0.00
27.6. 23.00	11.38	0.00	0.00	0.00	7.01	0.00	0.05
28.6. 7.00	26.69	0.00	0.00	0.00	15.08	0.00	0.00
28.6. 11.00	28.77	0.00	0.00	0.00	16.67	0.00	0.04

	wavelengt det	ector				/ detector	
sample	CI	Br	NO3	PO4	SO4	Br	NO3
PT 01							
standard1	8.07	4.13	0.83	2.17	4.20	4.85	0.98
27.6.7.00	26.94	0.00	0.36	0.00	8.40	0.00	0.18
27.6. 11.00	15.08	0.00	0.00	0.00	4.98	0.00	0.03
27.6. 15.00	9.35	0.00	0.00	0.00	3.49	0.00	0.10
27.6. 19.00	28.79	3.81	0.00	0.00	11.09	4.81	0.21
27.6. 23.00	7.87	0.00	0.00	0.00	2.92	0.00	0.03
28.6. 7.00	37.39	0.00	0.00	0.00	12.02	0.00	0.08
28.6. 11.00	16.54	0.00	0.00	0.00	5.02	0.06	0.16
28.6. 15.00	37.18	0.16	0.00	2.41	12.13	0.30	0.05
28.6. 23.00	3.77	0.53	0.00	0.00	1.46	0.75	0.20
29.6. 7.00	29.87	0.35	0.00	0.00	9.55	0.62	0.06
29.6. 15.00	37.19	0.22	0.00	0.00	12.01	0.34	0.04
29.6. 23.00	23.27	0.39	0.00	0.00	7.53	0.59	0.11
30.6. 7.00	33.76	0.70	0.00	0.00	11.53	1.01	0.09
30.6. 23.00	28.63	0.62	0.39	0.00	13.01	0.87	0.53
1.7. 15.00	36.41	0.85	0.00	0.00	12.25	1.25	0.05
1.7.23.00	36.71	0.40	0.00	0.00	12.15	0.56	0.05
2.7. 9.00	37.63	0.35	0.00	0.00	12.68	0.30	0.00
2.7. 19.00	39.22	0.42	0.00	0.00	12.58	0.45	0.05
GNT 02 standard3	47 99	19.32	4 91	9.42	20.13	19 70	5.08
27.6.7.00	26.63	0.00	1.98	0.75	13 21	0.00	2.22
27.6.11.00	7.61	0.00	0.92	0.00	8 31	0.00	1 90
27.6.15.00	7.01	0.00	0.02	0.00	5.05	0.61	0.32
27.6.19.00	25 50	8.04	1 33	0.00	12 75	10.76	2.12
27.6.23.00	23.43	14.38	1.00	0.00	11.93	17.59	1 91
28.6.7.00	20.80	18.73	0.78	0.00	10.07	24 53	1.01
28.6.11.00	21.00	15.96	1 97	0.00	12 74	21.80	2 65
28.6.15.00	21.46	28.53	1.67	0.00	11 29	36.98	1 79
28.6 19.00	8.88	10.02	0.00	0.96	5.63	14 75	1.33
28.6.23.00	17.30	13 19	5.66	0.00	14.30	16.98	6.78
29.6.7.00	18.47	36.82	1.91	0.00	10.31	46.00	2 12
29.6 15.00	17.92	56.63	2 27	0.00	11 59	69 44	2.97
29.6. 23.00	12.86	63.13	1.67	0.00	8.68	76.71	2.58
30.6.7.00	12.64	41.38	4.97	0.00	12.56	53.09	5.97
30.6. 23.00	16.18	98.05	4.28	0.00	11.70	111.13	4.75
1.7.7.00	15.09	98.93	2.80	0.00	13.04	113.80	4.23
1.7. 15.00	19.21	99.82	7.44	0.00	14.09	114.56	8.52
1.7. 23.00	34.08	106.02	5.98	0.00	69.13	117.88	11.51
2.7. 9.00	9.70	75.44	0.00	0.00	8.33	88.15	2.37
2.7. 19.00	15.33	123.14	0.00	0.00	12.02	137.70	4.22
3.7. 9.00	16.05	121.24	0.00	0.00	11.94	133.32	3.49
4.7. 9.00	14.92	115.27	0.00	0.00	11.19	125.10	2.75
5.7.9.00	14.85	110.88	0.00	0.00	12.86	119.77	0.00
6.7. 9.00	13.65	117.67	0.00	0.00	9.63	139.25	0.00
7.7.9.00	17.1461	128.495	0	0	12.5323	142.981	2.62922
24.7.9.00	10.44	57.98	0.00	0.00	5.64	68.18	2.09
25.7.9.00	19.03	121.23	0.00	0.00	10.09	135.48	2.28

# Tab.: A19 anions from M5

	wavelengt det	ector				conductivity detector	
sample	CI	Br	NO3	PO4	SO4	Br	NO3
26.7. 9.00	8.15	51.93	0.00	0.00	4.87	62.53	1.11
27.7.9.00	13.24	96.54	0.00	0.00	8.44	110.23	1.57
28.7. 9.00	19.16	129.36	0.00	0.00	11.24	144.62	2.29
29.7.9.00	6.77	45.78	0.00	0.00	4.53	54.62	2.20
30.7. 9.00	16.96	119.29	0.00	0.00	10.08	130.46	1.52
31.7. 9.00	13.46	45.43	0.00	0.00	8.30	57.24	2.13
1.8. 9.00	8.08	16.37	1.62	0.00	4.22	20.05	2.35
2.8. 9.00	24.78	47.44	0.00	0.00	16.37	55.33	1.25
3.8. 9.00	26.14	36.82	0.00	0.00	16.10	47.81	1.41
4.8. 9.00	20.84	29.16	0.00	0.00	12.64	36.30	1.79
5.8. 9.00	5.58	6.03	0.00	0.00	4.13	8.09	1.42
6.8. 9.00	46.69	66.32	0.00	0.00	25.50	55.25	0.00
7.8. 9.00	8.78	7.93	0.00	0.00	5.75	9.44	0.00
8.8. 19.00	15.18	10.66	0.00	0.00	9.28	13.03	0.00
09.8. 9.00	49.55	23.75	0.00	0.00	27.53	23.98	0.00
09.8.019.00	10.36	8.23	0.00	0.00	5.98	10.00	0.13
10.8. (sp3)	19.63	6.92	3.13	0.00	12.04	10.21	5.68
11.8. 9.00	12.92	5.91	0.00	0.00	7.11	7.08	0.00
11.8. 19.00	57.31	15.67	0.00	0.00	30.55	17.05	0.25
12.8. 9.00	12.17	3.50	0.00	0.00	6.66	4.85	0.11
12.8. 19.00	27.27	3.62	0.00	0.00	15.15	5.04	0.11
13.8. 9.00	26.76	6.11	0.00	0.00	13.61	7.32	0.00
13.8. 19.00	25.54	5.08	0.00	0.00	13.31	6.15	0.00
14.8. 9.00	14.92	2.01	0.00	0.00	7.92	2.85	0.00
14.8. 19.00	28.72	6.67	0.00	0.00	14.53	8.05	0.12

RT 03

standard2	17.62	8.61	1.89	5.65	9.38	9.98	2.09
27.6. 7.00	22.15	0.00	0.58	0.00	13.92	0.00	0.95
27.6. 11.00	5.78	0.00	0.00	0.00	5.24	0.00	0.50
27.6. 15.00	5.28	0.00	0.95	0.00	3.85	0.00	1.61
27.6. 19.00	23.67	1.53	0.00	0.80	12.70	3.09	0.49
27.6. 23.00	11.28	1.47	0.00	0.00	7.26	3.01	0.93
28.6. 7.00	22.00	0.00	0.00	0.00	13.54	0.00	0.32
28.6. 11.00	21.39	2.41	0.00	0.00	13.10	3.79	1.20
28.6. 15.00	14.12	14.55	1.74	0.00	10.90	19.20	2.60
28.6. 19.00	5.18	8.09	1.66	0.00	4.21	10.38	2.76
28.6. 23.00	14.09	32.53	3.27	0.00	11.61	41.83	4.64
29.6. 7.00	11.24	54.08	1.90	0.00	9.42	63.54	3.13
29.6. 15.00	16.24	104.46	4.37	0.00	11.77	115.37	5.93
29.6. 23.00	10.74	92.49	2.01	0.00	9.04	105.29	3.35
30.6. 7.00	11.36	93.99	4.23	0.00	9.74	105.36	3.45
30.6. 23.00	14.30	113.07	3.62	0.00	10.01	124.43	3.88
1.7. 7.00	14.34	117.68	2.74	0.00	10.00	128.75	4.27
1.7. 15.00	13.72	120.71	4.24	0.00	10.65	130.17	3.91
1.7. 23.00	14.91	115.10	3.83	0.00	10.91	125.59	4.62
2.7. 9.00	10.68	67.57	4.37	0.00	9.36	73.86	5.70
2.7. 19.00	16.44	130.46	4.42	1.12	13.04	137.01	5.52
3.7. 9.00	19.02	127.83	4.70	0.00	18.03	131.84	5.62
4.7. 9.00	20.45	129.08	0.00	0.00	11.76	145.34	4.57
5.7. 9.00	12.14	85.61	0.00	0.00	9.43	102.03	1.52
6.7. 9.00	15.47	155.10	0.00	0.00	13.62	168.88	1.53

	wavelengt det	ector				conductivity	/ detector
sample	CI	Br	NO3	PO4	SO4	Br	NO3
7.7.9.00	14.33	132.72	0.00	0.00	10.21	146.39	0.00
24.7. 9.00	16.14	109.13	5.37	0.00	9.89	120.32	8.18
25.7.9.00	22.54	156.07	8.73	0.00	13.26	164.83	9.94
26.7. 9.00	15.91	110.13	4.06	0.00	9.83	122.38	6.75
27.7.9.00	3.42	27.00	2.37	0.00	3.32	35.84	4.07
28.7. 9.00	12.62	94.39	3.60	0.00	7.84	108.30	5.58
29.7. 9.00	21.88	154.72	7.15	0.00	12.74	168.49	10.19
30.7. 9.00	14.87	109.08	5.42	0.00	8.53	121.76	7.10
31.7. 9.00	10.40	61.90	2.53	0.00	6.65	76.81	4.35
1.8. 9.00	24.54	34.68	0.00	0.00	17.33	47.47	2.77
2.8. 9.00	28.37	5.14	1.31	0.00	19.72	7.47	2.09
3.8. 9.00	27.77	1.90	0.00	0.00	21.13	1.26	1.83
4.8. 9.00	25.48	36.24	0.00	0.00	17.02	47.88	2.26
5.8. 9.00	11.48	1.06	4.65	0.00	7.85	1.16	6.13
6.8. 9.00	20.03	9.66	0.00	0.00	13.99	11.15	0.07
7.8. 9.00	18.87	0.79	0.00	0.00	13.19	1.04	0.06
8.8. 19.00	24.93	0.12	0.00	0.00	17.62	0.19	0.04
09.8. 9.00	19.09	0.00	0.00	0.00	14.77	0.15	0.00
09.8.019.00	8.64	0.00	0.00	0.00	7.46	0.00	0.00
10.8. (sp3)	23.09	0.00	0.00	0.00	17.36	0.75	0.51
11.8. 9.00	4.91	0.00	0.00	0.00	3.76	0.00	0.00
11.8. 19.00	38.50	1.57	0.00	0.00	25.92	2.34	0.12
12.8. 9.00	9.15	0.00	0.00	0.00	7.98	0.00	0.00
12.8. 19.00	12.36	0.00	0.00	0.00	9.34	0.00	0.05
13.8. 9.00	18.76	0.11	0.00	0.00	14.42	0.12	0.00
13.8. 19.00	51.31	2.57	0.00	0.00	32.51	3.84	0.21
14.8. 9.00	24.51	0.26	0.00	0.00	18.34	0.27	0.00
14.8. 19.00	6.52	0.00	0.00	0.00	5.59	0.06	0.00

standard1	8.51	4.26	1.03	2.34	4.40	5.22	1.06
27.6.7.00	18.12	0.00	0.00	0.00	10.43	0.00	0.38
27.6. 11.00	47.89	3.99	0.00	0.00	22.90	7.52	0.28
27.6. 19.00	26.55	0.00	0.00	0.00	13.90	0.00	0.57
27.6. 23.00	9.07	1.78	0.00	0.00	5.40	2.19	0.42
28.6. 7.00	7.04	0.00	0.00	0.00	6.07	1.57	0.40
28.6. 19.00	7.92	9.91	0.00	0.00	5.37	13.45	1.29
28.6. 23.00	6.71	8.94	0.00	0.00	3.83	13.36	1.28
29.6. 7.00	21.74	41.75	0.00	0.00	12.75	54.45	3.28
29.6. 15.00	19.57	43.71	0.00	0.00	13.15	54.05	2.17
29.6. 23.00	19.24	62.19	0.00	0.00	12.99	73.14	3.17
30.6. 7.00	15.05	47.97	0.00	0.00	9.68	59.96	3.19
30.6. 23.00	17.19	76.73	0.00	0.00	11.37	92.38	2.93
1.7. 7.00	18.88	81.23	0.00	0.00	12.71	97.13	3.29
1.7. 15.00	22.76	82.41	0.00	0.00	11.56	98.26	3.32
1.7. 23.00	16.06	90.34	0.00	0.00	10.46	106.10	3.32
2.7. 9.00	7.15	35.22	0.00	0.00	7.11	46.13	1.74
2.7. 19.00	11.67	86.49	0.00	0.00	8.70	103.87	3.09
4.7. 9.00	15.04	121.60	0.00	0.00	12.05	140.63	3.75
5.7.9.00	13.25	108.95	0.00	0.00	11.28	127.11	2.15

	wavelengt det	ector				conductivity	/ detector
sample	CI	Br	NO3	PO4	SO4	Br	NO3
6.7. 9.00	14.55	132.88	0.00	0.00	11.37	145.16	0.00
7.7.9.00	15.43	135.71	0.00	0.00	11.17	147.52	1.62
24.7.9.00	6.54	41.58	0.00	0.00	3.95	52.63	2.94
25.7.9.00	21.97	150.64	0.00	0.00	12.36	167.50	2.61
26.7.9.00	7.67	48.46	0.00	0.00	4.61	59.79	2.31
27.7.9.00	22.31	151.02	0.00	0.00	12.30	166.11	2.33
28.7.9.00	13.66	94.02	0.00	0.00	7.92	110.61	2.35
29.7.9.00	20.55	147.15	0.00	0.00	12.77	161.94	2.16
30.7.9.00	7.96	54.17	2.13	0.00	4.53	62.00	3.34
31.7. 9.00	22.25	161.61	3.00	0.00	12.19	169.91	5.22
1.8. 9.00	13.82	84.52	0.00	0.00	8.23	99.72	2.40
2.8. 9.00	10.31	60.56	0.00	0.00	5.99	74.44	2.22
3.8. 9.00	21.39	147.67	0.00	0.00	12.52	160.99	2.08
4.8.9.00	15.77	54.99	0.00	0.00	9.27	66.91	2.03
5.8. 9.00	10.63	18.82	0.00	0.00	6.87	24.08	2.33
6.8. 9.00	37.84	19.87	0.00	0.00	24.52	20.42	0.00
7.8. 9.00	12.29	3.03	0.00	0.00	8.32	3.84	0.00
8.8. 19.00	13.23	0.15	0.00	0.09	8.91	0.22	0.00
09.8. 9.00	22.63	1.01	0.00	0.00	14.61	1.24	0.00
09.8.019.00	11.51	0.00	0.00	0.00	7.91	0.00	0.00
10.8. (sp3)	9.83	0.00	0.00	0.00	8.73	0.00	1.68
11.8. 9.00	4.73	0.00	0.00	0.00	3.37	0.00	0.00
11.8. 19.00	31.13	0.00	0.00	0.00	20.80	0.08	0.00
12.8. 9.00	14.35	0.00	0.00	0.00	12.28	0.00	0.00
12.8. 19.00	9.12	0.00	0.00	0.00	7.17	0.00	0.00
13.8. 9.00	22.18	0.14	0.00	0.00	16.63	0.18	0.00
13.8. 19.00	16.79	0.00	0.00	0.00	13.75	0.00	0.00
14.8. 9.00	7.33	0.00	0.00	0.00	7.39	0.00	0.00
14.8. 19.00	17.85	0.00	0.00	0.00	13.57	0.00	0.00

### **BKT 05**

standard2	18.29	8.98	1.71	5.19	9.07	10.52	2.02
27.6.7.00	18.95	0.00	0.00	0.00	10.22	0.00	0.28
27.6. 11.00	8.17	0.00	0.00	0.00	4.70	1.33	0.36
27.6. 19.00	25.51	0.00	0.00	0.00	13.40	0.00	0.00
27.6. 23.00	11.59	0.00	0.00	0.00	6.09	0.00	0.34
28.6. 7.00	25.22	0.00	0.00	0.00	13.91	1.56	0.60
28.6. 11.00	8.51	0.00	0.00	0.00	5.58	0.00	1.04
28.6. 15.00	12.16	0.00	0.00	0.00	8.10	0.00	0.00
28.6. 19.00	47.83	7.57	0.00	0.00	20.84	9.07	0.86
28.6. 23.00	16.02	0.00	0.00	0.00	9.01	2.91	0.38
29.6. 7.00	23.00	0.00	0.00	0.00	11.69	0.91	0.00
29.6. 15.00	16.41	0.00	0.00	0.00	10.43	0.00	1.10
29.6. 23.00	17.53	0.00	0.00	0.00	9.90	0.00	0.00
30.6. 7.00	25.40	0.00	0.00	0.00	13.87	0.00	0.48
30.6. 23.00	24.84	0.00	0.00	0.00	12.65	0.00	0.00
1.7. 7.00	27.39	0.00	1.35	0.00	18.00	0.00	2.03
1.7. 15.00	25.02	0.00	0.00	0.00	13.42	1.65	0.39
1.7. 23.00	24.21	3.09	0.00	0.00	14.35	4.30	0.41
2.7. 9.00	16.55	9.44	2.56	3.98	15.22	13.64	4.58
2.7. 19.00	0.58	0.00	0.00	0.00	0.64	0.00	0.00

wavelengt de	tector				conductivity	v detector
CI	Br	NO3	PO4	SO4	Br	NO3
49.56	19.85	4.71	9.36	19.86	20.29	5.08
24.37	0.00	0.00	0.00	12.43	0.00	0.27
7.81	0.00	0.00	0.00	5.25	0.00	0.60
6.21	0.00	0.00	0.00	4.37	0.00	0.00
24.74	0.00	0.00	0.00	13.47	0.00	0.00
5.68	0.00	0.00	0.00	3.29	0.00	0.00
15.82	0.00	0.00	0.00	9.68	0.00	0.82
18.79	0.00	0.00	0.00	9.91	0.73	0.00
12.97	0.00	0.00	0.00	7.68	0.62	0.36
21.66	3.33	0.00	0.00	11.66	6.21	0.64
47.91	7.05	0.00	0.00	23.57	9.54	0.60
23.71	0.00	0.00	0.00	13.05	0.00	0.00
23.63	21.90	0.00	0.00	13.28	30.35	1.30
17.33	9.20	0.00	0.00	10.06	14.20	0.55
23.37	0.00	0.00	0.00	12.41	1.69	0.42
17.79	74.36	0.00	0.00	11.89	90.82	2.92
18.28	84.99	2.83	0.00	12.63	99.23	4.12
17.82	88.35	0.00	0.00	12.10	105.96	4.12
18.01	98.49	0.00	0.00	11.59	116.60	4.18
10.72	87.81	0.00	0.00	8.49	98.96	4.75
14.51	115.93	0.00	0.00	11.29	134.50	3.15
	-	-				
	wavelengt dei CI 49.56 24.37 7.81 6.21 24.74 5.68 15.82 18.79 12.97 21.66 47.91 23.71 23.63 17.33 23.37 17.79 18.28 17.82 18.01 10.72 14.51	Variable         GI         Br           49.56         19.85           24.37         0.00           7.81         0.00           6.21         0.00           24.74         0.00           24.74         0.00           15.82         0.00           15.82         0.00           12.97         0.00           21.66         3.33           47.91         7.05           23.71         0.00           23.63         21.90           17.33         9.20           23.37         0.00           17.79         74.36           18.28         84.99           17.82         88.35           18.01         98.49           10.72         87.81           14.51         115.93	Cl         Br         NO3           49.56         19.85         4.71           24.37         0.00         0.00           7.81         0.00         0.00           6.21         0.00         0.00           24.74         0.00         0.00           15.82         0.00         0.00           15.82         0.00         0.00           12.97         0.00         0.00           12.97         0.00         0.00           23.63         21.90         0.00           23.63         21.90         0.00           17.33         9.20         0.00           17.82         88.35         0.00           17.82         88.35         0.00           17.82         88.35         0.00           14.51         115.93         0.00	Vavelengt detector           Cl         Br         NO3         PO4           49.56         19.85         4.71         9.36           24.37         0.00         0.00         0.00           7.81         0.00         0.00         0.00           6.21         0.00         0.00         0.00           24.74         0.00         0.00         0.00           5.68         0.00         0.00         0.00           15.82         0.00         0.00         0.00           12.97         0.00         0.00         0.00           12.97         0.00         0.00         0.00           23.63         21.90         0.00         0.00           23.63         21.90         0.00         0.00           17.33         9.20         0.00         0.00           17.79         74.36         0.00         0.00           17.82         88.35         0.00         0.00           18.01         98.49         0.00         0.00           14.51         115.93         0.00         0.00	Wavelengt detector         Cl         Br         NO3         PO4         SO4           49.56         19.85         4.71         9.36         19.86           24.37         0.00         0.00         0.00         12.43           7.81         0.00         0.00         0.00         5.25           6.21         0.00         0.00         0.00         4.37           24.74         0.00         0.00         0.00         3.29           15.82         0.00         0.00         0.00         9.68           18.79         0.00         0.00         0.00         9.91           12.97         0.00         0.00         0.00         7.68           21.66         3.33         0.00         0.00         11.66           47.91         7.05         0.00         0.00         13.05           23.63         21.90         0.00         0.00         13.28           17.33         9.20         0.00         0.00         13.28           17.33         9.20         0.00         0.00         12.63           17.82         88.35         0.00         0.00         12.63           17.82         88.35	wavelengt detector         conductivity           Cl         Br         NO3         PO4         SO4         Br           49.56         19.85         4.71         9.36         19.86         20.29           24.37         0.00         0.00         0.00         12.43         0.00           7.81         0.00         0.00         0.00         4.37         0.00           6.21         0.00         0.00         0.00         13.47         0.00           24.74         0.00         0.00         0.00         3.29         0.00           15.82         0.00         0.00         0.00         9.68         0.00           15.82         0.00         0.00         0.00         9.91         0.73           12.97         0.00         0.00         0.00         7.68         0.62           21.66         3.33         0.00         0.00         11.66         6.21           47.91         7.05         0.00         0.00         13.05         0.00           23.63         21.90         0.00         0.00         13.05         0.00           23.37         0.00         0.00         0.00         12.41         1.69<

standard2	18.65	9.11	1.84	5.39	9.26	10.52	2.06
27.6.7.00	24.65	0.00	0.00	0.00	13.35	0.00	0.26
27.6. 11.00	8.94	0.00	0.00	0.00	6.95	0.00	0.25
27.6. 15.00	4.86	0.00	0.00	0.00	2.39	0.49	0.00
27.6. 19.00	15.58	0.00	0.00	0.00	9.73	0.00	0.00
27.6. 23.00	10.82	0.00	0.00	0.00	7.64	0.00	0.67
28.6. 7.00	36.66	0.00	2.63	0.00	19.38	0.00	2.98
28.6. 11.00	11.51	0.00	0.00	0.00	6.98	0.00	0.53
28.6. 19.00	13.39	0.00	0.00	0.00	7.31	0.00	0.24
28.6. 23.00	1.88	0.00	0.00	0.00	0.82	0.00	0.24
29.6. 7.00	30.00	2.17	0.00	0.00	15.28	3.47	0.29
29.6. 15.00	17.52	12.93	0.00	0.00	10.24	18.33	1.38
29.6. 23.00	14.07	38.99	0.00	0.00	9.24	49.21	1.60
30.6. 7.00	11.39	47.25	0.00	0.00	8.51	59.79	1.83
30.6. 23.00	15.46	94.26	0.00	0.00	11.11	110.39	3.52
1.7. 7.00	15.37	95.92	0.00	0.00	10.76	113.53	4.05
1.7. 15.00	15.75	99.42	2.62	0.00	11.71	112.07	3.90
1.7. 23.00	15.22	97.93	0.00	0.00	10.59	114.22	4.41
2.7. 9.00	14.55	76.36	3.74	0.00	18.01	83.78	4.99
2.7. 19.00	12.82	103.18	0.00	0.00	9.52	118.28	3.13

GY 08							
27.6.7.00	14.76	0.00	0.00	0.00	6.83	0.00	0.33
27.6. 11.00	11.50	0.00	0.00	0.00	7.21	0.00	1.08
27.6. 15.00	8.68	0.00	0.00	0.00	5.28	3.32	0.46
27.6. 19.00	26.37	0.00	0.00	0.00	12.39	0.00	0.54
27.6. 23.00	8.77	0.00	0.00	0.00	5.09	0.00	0.00

	wavelengt det	vavelengt detector					
sample	CI	Br	NO3	PO4	SO4	Br	NO3
28.6. 7.00	16.64	0.00	0.00	0.00	8.06	0.00	0.00
28.6. 11.00	21.36	0.00	0.00	0.00	10.44	0.00	0.54
30.6. 23.00	25.10	0.00	0.00	0.00	13.14	0.00	0.00
1.7. 15.00	25.98	6.61	0.00	0.00	13.77	9.25	0.69
1.7.23.00	22.71	30.78	0.00	0.00	12.46	38.91	1.21
2.7. 9.00	6.12	22.66	0.00	0.00	6.27	29.52	0.56
2.7. 19.00	15.98	64.55	0.00	0.00	11.07	79.28	2.52

#### Y 09

1 05							
standard1	8.46	4.37	0.83	2.58	4.53	5.28	0.97
27.6.7.00	26.81	0.00	0.00	0.00	10.48	0.00	0.00
27.6. 11.00	7.61	0.00	0.00	0.00	5.46	1.00	0.87
27.6. 15.00	10.77	0.00	0.00	0.00	5.53	0.00	0.31
27.6. 19.00	27.40	0.00	0.00	0.00	12.14	0.00	0.18
27.6. 23.00	50.43	9.17	0.00	0.00	22.18	12.21	0.78
28.6. 7.00	46.37	2.67	0.00	0.00	21.03	4.04	1.01
28.6. 11.00	25.03	0.00	0.00	0.00	10.85	0.00	0.22
30.6. 23.00	26.36	0.00	0.00	0.00	12.84	0.00	0.00
1.7. 15.00	27.37	0.00	0.00	0.00	12.77	0.00	0.52
1.7. 23.00	26.85	0.00	0.00	0.00	14.00	0.00	0.00
2.7. 9.00	3.60	0.00	0.00	0.00	3.17	1.76	0.46
2.7. 19.00	21.25	0.00	0.00	0.00	12.92	0.00	2.39

## Tab.: A20 anions from M6

	wavelength o	letector				conductivity detector		
sample	CI	Br	NO3	PO4	SO4	Br	NO3	
GNT 02					4	l contraction of the second seco		
standard2	18.34	9.15	1.79	5.32	9.04	10.26	1.94	
28.6. 15.00	8.19	24.17	0.00	0.00	5.19	30.60	1.15	
28.6. 19.00	6.92	17.53	0.00	0.00	4.86	23.96	0.94	
28.6. 23.00	8.57	30.19	0.00	0.00	7.01	39.09	1.43	
29.6. 7.00	9.05	30.53	0.00	0.00	6.31	39.39	1.24	
29.6. 15.00	7.93	34.75	0.00	0.00	5.61	43.54	1.44	
29.6. 23.00	7.38	34.06	0.00	0.00	5.69	43.35	1.42	
30.6. 7.00	13.72	86.01	0.00	0.00	9.08	99.49	3.40	
30.6. 23.00	13.63	118.43	3.15	0.00	10.04	127.26	4.01	
1.7.7.00	15.19	115.73	3.20	0.00	11.35	125.93	5.00	
1.7. 15.00	20.97	112.49	2.34	0.00	10.85	122.52	4.54	
1.7. 23.00	14.88	114.83	3.97	0.00	10.81	124.75	4.85	
2.7. 9.00	13.34	90.69	3.58	1.78	11.55	99.32	4.99	
2.7. 19.00	12.06	82.53	3.97	0.00	9.58	91.29	5.30	
3.7. 9.00	6.18	46.82	0.00	0.78	6.21	58.80	2.31	
7.7.	15.80	149.04	0.00	0.00	11.05	178.79	0.00	
24.7. 9.00	12.13	75.31	0.00	0.00	7.16	92.62	1.41	
25.7.9.00	22.27	136.49	0.00	0.00	11.67	155.34	3.91	
26.7. 9.00	12.51	66.09	0.00	0.00	8.28	82.83	1.50	
27.7.9.00	18.40	128.99	0.00	0.00	9.63	150.58	2.90	
28.7. 9.00	6.34	40.41	0.00	0.00	3.95	53.38	0.37	
29.7. 9.00	9.03	49.28	0.00	0.00	6.14	63.81	0.80	
30.7. 9.00	27.49	46.82	0.00	0.00	16.50	59.71	1.48	
31.7. 9.00	14.20	16.29	0.00	0.00	9.29	21.41	0.32	
1.8. 9.00	30.06	20.53	0.00	0.00	16.94	27.42	0.30	
2.8. 9.00	27.39	10.51	0.00	0.00	16.88	14.24	0.00	
3.8. 9.00	31.84	1.90	0.00	0.00	18.33	2.40	0.10	
4.8. 9.00	38.66	0.43	0.00	0.00	22.31	0.41	0.00	
5.8. 9.00	35.84	30.52	0.00	0.00	19.95	31.34	0.00	
6.8. 9.00	21.50	2.15	0.00	0.00	15.40	2.83	0.00	
7.8. 9.00	35.35	8.88	0.00	0.00	19.09	10.60	0.00	
09.08. 9.00	21.13	0.37	0.24	0.00	12.72	0.35	0.43	
10.08. (sp3)	25.54	2.56	1.21	0.00	17.56	4.10	1.53	
11.08. 9.00	29.85	0.00	0.00	0.00	17.39	0.00	0.00	
12.08. 9.00	10.63	0.00	0.00	0.00	6.49	0.00	0.00	
13.08. 9.00	14.14	0.00	0.00	0.00	9.36	0.00	0.00	
14.08. 9.00	20.39	0.51	1.05	0.00	7.02	0.64	1.45	

RT 03							
standard3	49.67	20.17	4.87	10.00	20.13	19.68	4.79
28.6. 15.00	21.79	0.00	0.00	0.00	12.35	0.00	0.35
28.6. 19.00	14.01	0.00	0.00	0.00	7.77	0.00	0.30
28.6. 23.00	8.58	0.00	0.00	0.00	6.42	0.00	0.28
29.6. 7.00	20.16	4.35	0.00	0.00	13.91	5.95	0.41
29.6. 15.00	18.15	15.65	0.00	0.00	11.05	21.48	0.64
29.6. 23.00	20.33	39.86	0.00	0.00	12.21	50.75	1.67
30.6. 7.00	18.30	35.30	0.00	0.00	11.95	46.38	1.58
30.6. 23.00	19.77	52.81	0.00	0.00	12.85	67.19	2.20
1.7.7.00	19.28	56.37	0.00	0.00	11.94	70.68	2.49

	wavelength of	detector		conductivity detector				
sample	CI	Br	NO3	PO4	SO4	Br	NO3	
1.7. 15.00	17.47	61.52	0.00	0.00	11.41	75.70	2.51	
1.7.23.00	17.80	72.17	0.00	0.00	12.17	87.20	2.82	
2.7. 9.00	7.80	37.11	0.00	0.00	8.16	47.36	2.60	
2.7. 19.00	13.90	121.98	4.24	0.00	12.02	128.60	4.78	
3.7. 9.00	4.76	39.98	0.00	0.00	4.74	49.47	2.60	
7.7.	11.34	55.43	0.00	0.00	9.93	68.50	1.92	
24.7. 9.00	8.30	53.23	0.00	0.00	5.20	68.73	0.00	
25.7.9.00	21.59	149.67	0.00	0.00	11.19	168.99	0.62	
26.7. 9.00	23.39	153.71	0.00	0.00	12.59	171.80	0.00	
27.7.9.00	21.22	145.41	0.00	0.00	11.78	164.96	0.77	
28.7. 9.00	20.38	141.87	0.00	0.00	11.81	160.74	0.00	
29.7. 9.00	10.47	70.76	0.00	0.00	6.20	87.38	0.00	
30.7. 9.00	20.50	149.68	0.00	0.00	12.09	168.04	0.00	
31.7. 9.00	13.18	89.19	0.00	0.00	7.70	106.57	0.00	
1.8. 9.00	22.86	159.95	0.00	0.00	13.56	175.23	0.00	
2.8. 9.00	20.18	118.60	0.00	0.00	12.04	137.96	0.00	
3.8. 9.00	30.32	123.52	0.00	0.26	16.09	92.22	0.00	
4.8. 9.00	13.71	7.68	0.00	0.00	9.71	9.60	0.00	
5.8. 9.00	20.78	6.40	0.00	0.00	13.18	8.00	0.00	
6.8. 9.00	37.87	0.00	0.16	0.00	23.45	0.09	0.22	
7.8. 9.00	24.09	0.51	0.00	0.00	15.12	0.69	0.00	
09.08. 9.00	213.93	0.00	0.32	0.23	24.46	0.28	0.42	
10.08. (sp3)	22.65	2.27	1.23	0.00	15.90	3.46	1.26	
11.08. 9.00	32.76	0.40	0.51	0.00	20.46	0.58	0.72	
12.08. 9.00	8.13	0.00	0.00	0.00	5.91	0.00	0.00	
13.08. 9.00	20.89	0.00	0.00	0.00	14.86	0.00	0.00	
14.08. 9.00	23.69	0.30	0.52	0.00	11.83	0.41	0.75	

standard1	8.16	4.11	0.79	2.40	4.10	5.19	0.98
27.6. 7.00	32.55	0.00	0.00	0.00	15.34	0.00	0.00
27.6. 11.00	20.62	0.00	0.00	0.00	10.39	0.00	0.00
27.6. 15.00	21.68	1.43	0.00	0.00	9.22	1.89	0.07
27.6. 19.00	32.29	0.00	0.00	0.00	15.28	0.00	0.00
27.6. 23.00	15.70	0.00	0.00	0.00	7.67	0.00	0.00
28.6. 7.00	21.80	3.87	0.00	0.00	12.20	5.95	0.42
28.6. 11.00	2.33	0.00	0.00	0.00	1.16	0.00	0.00
28.6. 15.00	25.61	0.00	0.00	0.00	14.39	0.00	0.55
28.6. 19.00	12.53	0.00	0.00	0.00	7.90	0.49	0.35
28.6. 23.00	28.08	9.22	0.00	0.00	14.32	12.98	0.58
29.6. 7.00	22.33	0.00	0.00	0.00	12.87	0.00	0.35
29.6. 15.00	17.21	0.00	0.00	0.00	10.72	0.00	0.00
29.6. 23.00	17.08	6.63	0.00	0.00	9.83	9.33	0.75
30.6.7.00	18.72	0.00	0.00	0.00	11.14	0.72	1.53
30.6. 23.00	26.07	0.00	0.00	0.00	17.06	0.00	1.15
1.7. 7.00	24.61	0.00	0.00	0.00	14.40	0.00	1.52
1.7. 15.00	25.57	0.00	0.00	0.00	13.66	0.00	0.28
1.7. 23.00	22.69	5.88	0.00	0.00	13.42	8.98	0.51
2.7. 9.00	6.08	42.83	0.00	0.00	5.62	54.41	1.94
2.7. 19.00	22.03	137.85	5.17	0.00	15.34	139.88	5.64
3.7. 9.00	15.41	94.55	6.64	0.00	13.23	100.61	7.97
7.7.	11.81	102.11	0.00	0.00	10.66	117.44	3.07
24.7. 9.00	20.97	159.02	0.00	0.00	10.50	176.77	0.00

	wavelength of	detector		conductivity detector			
sample	CI	Br	NO3	PO4	SO4	Br	NO3
25.7.9.00	21.25	158.32	0.00	0.00	11.38	174.49	0.00
26.7. 9.00	11.31	77.41	0.00	0.00	6.47	92.62	0.00
27.7.9.00	23.27	140.64	0.00	0.00	12.87	157.51	0.57
28.7. 9.00	22.56	143.85	0.00	0.00	12.48	160.72	0.00
29.7. 9.00	9.69	52.19	0.00	0.00	6.36	65.52	0.98
30.7. 9.00	23.93	139.90	0.00	0.00	13.74	160.51	2.18
31.7. 9.00	23.64	145.03	0.00	0.00	12.80	164.71	1.72
1.8. 9.00	30.41	152.74	7.43	0.00	19.13	164.91	8.20
2.8. 9.00	23.24	156.13	0.00	0.00	11.94	174.81	0.52
3.8. 9.00	16.65	124.72	0.00	0.00	8.56	91.49	0.00
4.8. 9.00	17.63	91.40	0.00	0.00	9.18	72.25	0.00
5.8. 9.00	19.05	1.57	0.00	0.00	11.85	1.99	0.00
6.8. 9.00	39.37	0.33	0.18	0.00	22.60	0.49	0.21
7.8. 9.00	17.10	0.00	0.10	0.00	10.51	0.00	0.10
8.8. 9.00	39.13	0.00	0.00	0.00	22.57	0.00	0.00
09.08. 9.00	11.87	0.12	0.00	0.00	9.00	0.16	0.00
10.08. (sp3)	14.44	0.00	1.26	0.00	10.63	0.00	2.95
11.08. 9.00	9.22	0.00	0.10	0.00	6.89	0.00	0.16
12.08. 9.00	3.99	0.00	0.00	0.00	3.23	0.00	0.00
13.08. 9.00	19.74	0.32	0.32	0.00	11.53	0.48	0.50
14.08. 9.00	9.49	0.00	0.00	0.00	6.90	0.00	0.00

#### **BKT 05**

standard3	49.35	20.03	4.92	9.54	20.11	19.84	4.90
27.6.7.00	30.00	0.00	0.00	0.00	14.23	0.00	0.07
27.6. 11.00	15.35	0.00	0.00	0.00	6.80	0.00	0.02
27.6. 15.00	19.05	0.00	0.00	0.00	9.44	0.00	0.10
27.6. 19.00	33.35	0.00	0.00	0.00	15.22	0.00	0.02
27.6. 23.00	45.19	0.00	0.00	0.00	9.30	0.00	0.16
28.6. 7.00	25.56	0.00	0.00	0.00	12.29	0.00	0.09
28.6. 11.00	13.21	1.62	0.00	0.00	6.28	2.25	0.15

#### WT 06

standard1	8.19	4.13	0.82	2.58	4.17	5.11	0.97
27.6. 7.00	346.47	48.61	0.00	1.99	11.45	0.00	0.00
27.6. 11.00	34.90	0.00	0.00	0.00	13.55	0.00	0.03
27.6. 15.00	11.24	0.00	0.00	0.00	5.52	0.00	0.05
27.6. 19.00	33.46	0.00	0.00	0.00	14.54	0.00	0.04
27.6. 23.00	21.10	0.00	0.00	0.00	9.69	0.00	0.04
28.6. 7.00	35.45	0.00	0.31	0.00	15.31	0.00	0.34
28.6. 11.00	32.01	0.00	0.00	0.00	13.64	0.00	0.14

# Tab.: A 21 anions from M9

	wavelength	detector			conductivity detector		
sample	CI	Br	NO3	PO4	SO4	Br	NO3
 GNT 02							
29.6. 7.00	45.37	0.00	0.00	0.00	15.21	0.00	0.07
29.6. 15.00	7.37	0.00	0.00	0.00	2.73	0.00	0.13
29.6. 23.00	37.31	0.00	0.00	0.00	12.32	0.00	0.08
30.6. 7.00	61.62	0.00	0.00	0.00	20.47	0.07	0.21
30.6. 23.00	37.69	0.00	0.00	0.00	12.91	0.00	0.00
1.7.7.00	37.78	0.00	0.00	0.00	13.73	0.12	0.12
1.7. 15.00	18.71	4.06	0.00	0.00	7.95	5.09	0.27
1.7. 23.00	36.75	1.85	0.00	0.00	13.95	2.45	0.15
2.7. 9.00	16.61	5.75	0.00	0.00	10.36	6.25	0.79
2.7. 19.00	23.72	13.88	0.00	0.00	12.71	18.58	1.04
3.7. 9.00	21.26	30.80	0.00	0.00	11.96	39.46	1.43
4.7. 9.00	22.83	49.11	0.00	0.00	15.14	62.99	2.83
5.7. 9.00	23.12	65.05	3.05	0.00	17.40	72.77	3.76
6.7. 9.00	18.32	68.76	0.00	0.00	10.17	81.96	2.40
7.7. 9.00	18.70	75.20	0.00	0.00	11.79	90.35	3.20
9.7. 9.00	16.57	95.35	0.00	0.00	10.28	115.83	0.00
12.7.9.00	16.45	97.73	0.00	0.00	9.74	117.22	1.94
15.7. 9.00	16.56	124.46	0.00	0.00	11.58	143.19	1.86
24.7. 9.00	15.42	143.94	0.00	0.00	7.75	151.44	0.00
8.8. 19.00	17.43	36.10	0.00	0.00	11.70	43.55	0.00
09.8. 9.00	20.67	18.14	0.00	0.00	13.38	23.50	0.00
09.8.019.00	12.23	16.89	0.00	0.00	7.57	23.51	0.62
10.8. (sp3)	21.73	41.86	0.00	0.00	13.50	53.79	1.64
11.8. 9.00	14.60	28.80	0.00	0.00	9.17	37.54	0.00
11.8. 19.00	11.45	18.97	0.00	0.00	7.07	24.88	0.47
12.8. 9.00	17.60	25.54	0.00	0.00	8.41	35.75	1.01
12.8. 19.00	38.32	37.56	0.00	0.00	16.37	50.10	1.20
13.8. 9.00	11.64	20.66	0.00	0.00	6.54	27.27	0.71
13.8. 19.00	16.67	36.64	0.00	0.00	10.49	45.46	0.00
14.8. 9.00	47.38	26.99	0.00	0.00	21.52	38.04	0.00
14.8. 19.00	25.03	37.64	0.00	0.00	12.72	45.49	0.00
15.8. 9.00	25.36	21.44	0.00	0.00	13.20	28.99	0.85
15.8. 19.00	14.34	19.89	0.00	0.00	8.85	25.39	0.00
16.8. 9.00	224.45	12.95	0.00	0.00	10.81	18.49	1.49
16.8. 19.00	14.04	11.07	0.00	0.00	8.47	14.72	0.88
17.8. 9.00	17.41	11.94	0.00	0.00	9.28	15.59	0.48
17.8. 19.00	13.05	8.99	0.00	0.00	6.88	11.96	0.90
18.8. 9.00	17.98	15.03	0.00	0.00	10.58	20.50	0.00
18.8. 19.00	25.94	10.81	0.00	0.00	12.72	16.44	0.79
19.8. 9.00	2.44	1.03	0.00	0.00	3.02	2.09	0.94
20.8. 9.00	13.51	4.83	0.00	0.00	8.24	5.91	0.26
21.8.9.00	32.90	7.35	0.00	0.00	16.30	8.27	0.00
22.8. 9.00	5.44	0.78	0.00	0.00	3.34	1.06	0.00
23.8.9.00	36.86	2.10	0.25	0.00	21.51	2.84	0.42
24.8.9.00	20.18	0.56	0.00	0.00	11.28	1.03	0.00
25.8.9.00	28.94	0.74	0.00	0.00	14.67	1.29	0.00
26.8.9.00	15.27	0.32	0.21	0.00	9.13	0.39	0.38
21.8.9.00	13.89	0.00	0.00	0.00	8.01	0.40	0.02
28.8.9.00	34.66	0.00	0.00	0.00	16.60	0.36	0.00
30.8. 9.00	33.08	0.14	0.00	0.00	16.23	0.14	0.00

	wavelength	detector				conductivit	y detector
sample	CI	Br	NO3	PO4	SO4	Br	NO3
· ·							
RT 03							
standard1	8.55	4.34	1.06	2.45	4.46	5.19	1.09
29.6. 7.00	28.08	0.00	0.00	0.00	12.92	0.00	0.00
29.6. 15.00	10.87	0.00	0.00	0.00	8.18	0.00	0.02
29.6. 23.00	11.17	0.00	0.00	0.00	6.02	0.00	0.12
30.6. 7.00	26.67	0.00	0.00	0.00	13.92	0.00	0.08
30.6. 23.00	32.31	0.00	0.00	0.00	16.37	0.00	0.06
1.7. 7.00	32.18	0.00	0.00	0.00	16.54	0.00	0.07
1.7. 15.00	14.47	1.47	0.00	0.00	7.44	2.11	0.07
1.7. 23.00	32.41	0.00	0.00	0.00	16.48	0.00	0.03
2.7. 9.00	22.33	0.00	1.89	0.00	17.88	0.86	1.89
2.7. 19.00	10.60	0.77	0.00	0.00	8.15	1.54	0.56
3.7. 9.00	11.06	11.47	0.98	0.00	7.89	13.68	1.19
4.7. 9.00	17.42	77.42	0.00	0.00	12.75	93.15	3.33
5.7. 9.00	6.04	40.37	0.00	0.00	7.21	49.86	2.04
6.7. 9.00	12.71	110.38	0.00	0.00	11.06	125.41	3.73
7.7. 9.00	15.40	116.13	0.00	0.00	14.18	132.43	4.26
9.7. 9.00	16.91	113.83	0.00	0.00	14.62	129.62	3.45
12.7.9.00	14.11	127.01	0.00	0.00	11.27	145.10	1.81
15.7. 9.00	13.34	144.00	0.00	0.00	8.91	151.28	0.00
24.7. 9.00	16.18	136.64	0.00	0.00	8.87	155.38	2.14
8.8. 19.00	29.78	0.00	0.00	0.00	19.89	0.13	0.00
09.8. 9.00	11.34	1.40	0.00	0.00	8.41	1.94	0.00
09.8. 19.00	16.12	0.00	0.00	0.00	11.42	0.09	0.02
10.8. (sp3)	44.08	7.03	0.00	0.00	29.36	7.72	1.59
11.8. 9.00	12.69	0.55	0.53	0.00	7.81	0.74	0.69
11.8. 19.00	53.04	9.73	0.00	0.00	18.35	10.85	0.22
12.8. 9.00	10.09	0.28	0.00	0.00	5.76	0.38	0.00
12.8. 19.00	7.77	0.00	0.00	0.00	6.78	0.00	0.00
13.8. 9.00	23.99	0.69	0.49	0.00	15.83	0.88	0.55
13.8. 19.00	68.39	8.58	0.63	0.00	17.22	9.49	0.79
14.8. 9.00	47.69	3.08	0.96	0.00	24.94	3.79	0.99
14.8. 19.00	39.36	3.77	0.00	0.00	11.22	4.48	0.08
15.8. 9.00	17.98	0.00	0.00	0.00	13.61	0.00	0.02
15.8. 19.00	28.00	0.00	0.00	0.00	19.92	0.00	0.00
16.8. 9.00	90.11	0.00	0.00	0.00	9.36	0.00	0.05
16.8. 19.00	29.04	0.67	0.00	0.00	14.61	0.99	0.00
17.8.9.00	19.03	0.00	0.00	0.00	13.87	0.00	0.00
17.8. 19.00	20.28	1.48	1.30	0.00	12.85	1.95	1.69
18.8. 9.00	22.07	1.96	1.56	0.00	12.86	2.59	1.94
18.8. 19.00	23.19	1.19	0.96	0.00	15.03	1.55	1.24
19.8. 9.00	29.97	0.07	0.00	0.00	20.76	0.09	0.04
20.8. 9.00	9.65	0.00	0.00	0.00	8.40	0.00	0.00
21.8.9.00	29.68	0.00	0.00	0.00	20.80	0.00	0.04
22.8.9.00	6.4/	0.00	0.00	0.00	5.45	0.00	0.00
23.8. 9.00	22.80	0.00	0.00	0.00	17.93	0.00	0.09
24.8.9.00	44.64	0.56	0.07	0.00	20.45	0.62	0.18
<u>25.8. 9.00</u>	17.35	0.00	0.00	0.00	14.15	0.00	0.00
20.8. 9.00	32.28	0.00	0.25	0.00	22.07	0.00	0.31
21.0. 9.00	10.14	0.00	0.00	0.00	21 50	0.00	0.00
20.0. 9.00	35.43	0.00	0.00	0.00	Z1.3Z	0.00	0.00

	wavelength	detector	conductivity de		y detector		
sample	CI	Br	NO3	PO4	SO4	Br	NO3
BUT 04							
standard2	18.43	9.14	1.93	5.26	9.18	10.51	2.05
29.6. 7.00	33.66	0.00	0.60	0.00	14.99	0.00	0.75
29.6. 15.00	26.41	0.00	0.00	0.00	15.46	0.00	0.00
29.6. 23.00	31.20	0.00	0.00	0.00	17.30	0.00	0.09
30.6. 7.00	31.24	0.00	0.00	0.00	17.94	0.00	0.10
30.6. 23.00	30.21	16.75	0.41	0.00	16.66	17.63	0.59
1.7. 7.00	28.45	45.79	1.31	0.00	16.04	39.30	1.53
1.7. 15.00	16.90	46.96	1.06	0.00	10.84	39.92	1.24
1.7. 23.00	24.17	102.44	2.50	0.00	15.00	71.59	2.68
2.7. 9.00	12.65	76.59	0.00	1.10	10.48	90.28	2.53
2.7. 19.00	13.98	81.08	0.00	0.00	10.65	96.15	3.92
3.7. 9.00	16.00	106.86	0.00	0.00	13.55	123.23	4.28
4.7. 9.00	15.15	114.56	0.00	0.00	13.41	128.30	3.73

$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.53           .92           .28           .73           .32           .66           .95           .00           .44           .00           .30           .64
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.92           .28           .73           .32           .66           .95           .00           .44           .00           .30           .64
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.28 .73 .32 .66 .95 .00 .44 .00 .30 .64
4.7.9.00 $15.15$ $114.56$ $0.00$ $0.00$ $13.41$ $128.30$ $128.30$ $5.7.9.00$ $9.04$ $57.62$ $0.00$ $0.00$ $7.92$ $71.02$ $128.55$ $6.7.9.00$ $14.00$ $110.60$ $0.00$ $0.00$ $11.35$ $125.55$ $125.55$ $7.7.9.00$ $17.64$ $128.06$ $0.00$ $0.00$ $11.35$ $125.55$ $125.55$ $9.7.9.00$ $17.66$ $155.12$ $0.00$ $0.00$ $11.32$ $172.84$ $012.22$ $15.7.9.00$ $16.29$ $137.32$ $0.00$ $0.00$ $12.22$ $155.92$ $157.9.00$ $14.94$ $142.36$ $0.00$ $0.00$ $9.40$ $148.20$ $012.22$ $24.7.9.00$ $16.89$ $132.20$ $0.00$ $0.00$ $8.55$ $151.57$ $8.8.19.00$ $32.03$ $6.38$ $0.45$ $0.00$ $18.37$ $7.08$ $009.8.900$ $18.33$ $3.21$ $0.00$ $0.00$ $12.97$ $4.23$ $009.8.900$ $18.33$ $3.21$ $0.00$ $0.00$ $4.30$ $1.22$ $012.97$ $11.8.9.00$ $4.39$ $0.50$ $0.00$ $0.00$ $3.71$ $0.68$ $012.84$ $0.00$ $11.8.9.00$ $4.39$ $0.50$ $0.00$ $0.00$ $12.24$ $1.26$ $002$ $12.8.9.00$ $14.87$ $0.64$ $0.00$ $0.00$ $12.24$ $1.26$ $002$ $12.8.9.00$ $17.57$ $1.82$ $0.62$ $0.00$ $8.06$ $2.36$ $002$ $14.8.9.00$ <td>3.73 3.32 5.66 5.95 5.00 5.00 5.00 5.00 5.00 5.00 5.00</td>	3.73 3.32 5.66 5.95 5.00 5.00 5.00 5.00 5.00 5.00 5.00
5.7.9.00 $9.04$ $57.62$ $0.00$ $0.00$ $7.92$ $71.02$ $2.5$ $6.7.9.00$ $14.00$ $110.60$ $0.00$ $0.00$ $11.35$ $125.55$ $3.7.9.00$ $9.7.9.00$ $17.64$ $128.06$ $0.00$ $0.00$ $11.32$ $172.84$ $(12.7.9.00)$ $16.29$ $137.32$ $0.00$ $0.00$ $11.32$ $172.84$ $(12.7.9.00)$ $16.29$ $137.32$ $0.00$ $0.00$ $12.22$ $155.92$ $2.5.92$ $15.7.9.00$ $14.94$ $142.36$ $0.00$ $0.00$ $9.40$ $148.20$ $(12.7.9.00)$ $24.7.9.00$ $16.89$ $132.20$ $0.00$ $0.00$ $8.55$ $151.57$ $2.8.8.19.00$ $32.03$ $6.38$ $0.45$ $0.00$ $18.37$ $7.08$ $(0.9.8.9.00)$ $9.8.9.00$ $18.33$ $3.21$ $0.00$ $0.00$ $12.97$ $4.23$ $(0.9.8.19.00)$ $15.79$ $1.82$ $0.77$ $0.00$ $9.33$ $2.40$ $(0.9.8.19.00)$ $11.8.9.00$ $4.39$ $0.50$ $0.00$ $0.00$ $4.30$ $1.22$ $(11.8.9.00)$ $4.39$ $0.50$ $0.00$ $0.00$ $13.00$ $4.53$ $12.8.9.00$ $14.87$ $0.64$ $0.00$ $0.00$ $12.24$ $1.26$ $(11.8.9.00)$ $27.33$ $0.60$ $0.00$ $10.20$ $0.30$ $(13.8.9.00)$ $27.33$ $0.60$ $0.00$ $10.20$ $0.30$ $(14.8.9.00)$ $17.57$ $1.82$ $0.62$ $0.00$ $8.06$ <	2.32 5.66 5.95 5.00 .44 5.00 .30 5.64
6.7.9.00 $14.00$ $110.60$ $0.00$ $0.00$ $11.35$ $125.55$ $125.55$ $7.7.9.00$ $17.64$ $128.06$ $0.00$ $0.00$ $12.69$ $139.22$ $139.22$ $9.7.9.00$ $17.66$ $155.12$ $0.00$ $0.00$ $11.32$ $172.84$ $0.01$ $12.7.9.00$ $16.29$ $137.32$ $0.00$ $0.00$ $12.22$ $155.92$ $15.7.9.00$ $14.94$ $142.36$ $0.00$ $0.00$ $9.40$ $148.20$ $0.02$ $24.7.9.00$ $16.89$ $132.20$ $0.00$ $0.00$ $8.55$ $151.57$ $12.32$ $8.8.19.00$ $32.03$ $6.38$ $0.45$ $0.00$ $18.37$ $7.08$ $0.09.8.900$ $09.8.9.00$ $18.33$ $3.21$ $0.00$ $0.00$ $12.97$ $4.23$ $0.00$ $09.8.19.00$ $15.79$ $1.82$ $0.77$ $0.00$ $9.33$ $2.40$ $0.00$ $11.8.9.00$ $4.39$ $0.50$ $0.00$ $0.00$ $4.30$ $1.22$ $0.00$ $11.8.9.00$ $4.39$ $0.50$ $0.00$ $0.00$ $3.71$ $0.68$ $0.00$ $12.8.9.00$ $14.87$ $0.64$ $0.00$ $0.00$ $12.24$ $1.26$ $0.00$ $12.8.9.00$ $14.87$ $0.64$ $0.00$ $0.00$ $18.58$ $1.16$ $0.00$ $14.8.9.00$ $27.33$ $0.60$ $0.00$ $0.00$ $14.62$ $2.13$ $0.00$ $14.8.9.00$ $17.57$ $1.82$ $0.62$ $0.00$ $8.06$ $2.36$ $0.$	
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.95 .00 .44 .00 .30 .64
9.7.9.00 $17.66$ $155.12$ $0.00$ $0.00$ $11.32$ $172.84$ $(12.7.9.00)$ $12.7.9.00$ $16.29$ $137.32$ $0.00$ $0.00$ $12.22$ $155.92$ $(12.7.9.00)$ $15.7.9.00$ $14.94$ $142.36$ $0.00$ $0.00$ $9.40$ $148.20$ $(12.7.9.00)$ $24.7.9.00$ $16.89$ $132.20$ $0.00$ $0.00$ $8.55$ $151.57$ $(12.7.9.00)$ $8.8.19.00$ $32.03$ $6.38$ $0.45$ $0.00$ $18.37$ $7.08$ $(12.97)$ $0.98.9.00$ $18.33$ $3.21$ $0.00$ $0.00$ $12.97$ $4.23$ $(12.97)$ $0.98.9.00$ $15.79$ $1.82$ $0.77$ $0.00$ $9.33$ $2.40$ $(11.8.9.0)$ $0.98.19.00$ $15.79$ $1.82$ $0.77$ $0.00$ $9.33$ $2.40$ $(11.8.9.0)$ $11.8.9.00$ $4.39$ $0.50$ $0.00$ $0.00$ $4.30$ $1.22$ $(11.8.9.0)$ $11.8.9.00$ $4.39$ $0.50$ $0.00$ $0.00$ $13.00$ $4.53$ $(12.8.9.0)$ $12.8.9.00$ $14.87$ $0.64$ $0.00$ $0.00$ $12.24$ $1.26$ $(12.8.19.0)$ $22.42$ $0.93$ $0.00$ $0.00$ $19.84$ $0.81$ $(13.8.9.0)$ $(14.8.9.0)$ $17.57$ $1.82$ $0.62$ $0.00$ $8.06$ $2.36$ $(14.8.9.0)$ $(14.8.9.0)$ $13.01$ $0.21$ $0.00$ $0.00$ $14.62$ $2.13$ $(14.8.9.0)$ $13.01$ $0.21$ $0.00$ $0.00$ $14.62$ $2.13$	.00 .44 .00 .30 .64
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.44 .00 .30 .64
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.00 .30 .64
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	.30 .64
8.8.19.00 $32.03$ $6.38$ $0.45$ $0.00$ $18.37$ $7.08$ $(0)$ $09.8.9.00$ $18.33$ $3.21$ $0.00$ $0.00$ $12.97$ $4.23$ $(0)$ $09.8.19.00$ $15.79$ $1.82$ $0.77$ $0.00$ $9.33$ $2.40$ $(0)$ $10.8.(sp3)$ $6.03$ $1.34$ $0.00$ $0.00$ $4.30$ $1.22$ $(0)$ $11.8.9.00$ $4.39$ $0.50$ $0.00$ $0.00$ $3.71$ $0.68$ $(0)$ $11.8.9.00$ $4.39$ $0.50$ $0.00$ $0.00$ $3.71$ $0.68$ $(0)$ $11.8.9.00$ $29.86$ $3.62$ $0.84$ $0.00$ $13.00$ $4.53$ $(0)$ $12.8.9.00$ $14.87$ $0.64$ $0.00$ $0.00$ $12.24$ $1.26$ $(0)$ $12.8.19.00$ $22.42$ $0.93$ $0.00$ $0.00$ $16.58$ $1.16$ $(0)$ $13.8.9.00$ $27.33$ $0.60$ $0.00$ $0.00$ $19.84$ $0.81$ $(0)$ $14.8.9.00$ $17.57$ $1.82$ $0.62$ $0.00$ $8.06$ $2.36$ $(0)$ $14.8.9.00$ $13.01$ $0.21$ $0.00$ $0.00$ $14.62$ $2.13$ $(0)$ $15.8.9.00$ $25.37$ $1.56$ $0.83$ $0.00$ $14.62$ $2.13$ $(0)$ $16.8.9.00$ $251.90$ $122.85$ $0.00$ $0.30$ $15.04$ $0.00$ $(0)$ $16.8.19.00$ $21.13$ $0.95$ $0.72$ $0.00$ $12.62$ $1.29$ $(0)$ $17.8$	.64
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	.20
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.96
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.63
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.00
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.06
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.00
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.00
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.00
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	.44
14.8. 19.00         13.01         0.21         0.00         0.00         10.20         0.30         0           15.8. 9.00         25.37         1.56         0.83         0.00         14.62         2.13         1           15.8. 19.00         25.33         0.18         0.00         0.00         18.50         0.19         0           16.8. 9.00         251.90         122.85         0.00         0.30         15.04         0.00         0           16.8. 19.00         21.13         0.95         0.72         0.00         12.62         1.29         0           17.8. 9.00         13.98         0.00         0.00         0.00         17.92         0.07         0	.69
15.8.9.00         25.37         1.56         0.83         0.00         14.62         2.13           15.8.19.00         25.33         0.18         0.00         0.00         18.50         0.19         0           16.8.9.00         251.90         122.85         0.00         0.30         15.04         0.00         0           16.8.19.00         21.13         0.95         0.72         0.00         12.62         1.29         0           17.8.9.00         13.98         0.00         0.00         0.00         17.92         0.07         0	.00
15.8.         19.00         25.33         0.18         0.00         0.00         18.50         0.19         0           16.8.         9.00         251.90         122.85         0.00         0.30         15.04         0.00         0           16.8.         19.00         21.13         0.95         0.72         0.00         12.62         1.29         0           17.8.         9.00         13.98         0.00         0.00         11.10         0.00         0           17.8.         19.00         23.91         0.00         0.00         17.92         0.07         0	.09
16.8. 9.00         251.90         122.85         0.00         0.30         15.04         0.00         0           16.8. 19.00         21.13         0.95         0.72         0.00         12.62         1.29         0           17.8. 9.00         13.98         0.00         0.00         0.00         11.10         0.00         0           17.8. 19.00         23.91         0.00         0.00         0.00         17.92         0.07         0	.00
16.8. 19.00         21.13         0.95         0.72         0.00         12.62         1.29         0           17.8. 9.00         13.98         0.00         0.00         0.00         11.10         0.00         0           17.8. 19.00         23.91         0.00         0.00         0.00         17.92         0.07         0	.05
17.8. 9.00         13.98         0.00         0.00         0.00         11.10         0.00         0           17.8. 19.00         23.91         0.00         0.00         0.00         17.92         0.07         0	.91
	.00
	.05
18.8. 9.00         22.80         1.03         1.26         0.00         14.73         1.34	.54
18.8. 19.00         40.11         4.41         0.97         0.00         17.13         5.31	.06
<u>19.8. 9.00</u> <u>20.61</u> <u>0.00</u> <u>0.00</u> <u>16.05</u> <u>0.00</u> (	.00
20.8. 9.00 17.50 0.20 0.00 0.00 12.73 0.26 (	.02
21.8. 9.00 29.29 0.00 0.00 0.00 21.31 0.00 (	.11
22.8. 9.00 9.14 0.00 0.00 0.22 7.57 0.00 (	.00
23.8. 9.00 11.82 0.00 0.32 0.00 9.38 0.16 (	.52
<u>24.8. 9.00</u> <u>21.83</u> <u>0.00</u> <u>0.00</u> <u>16.77</u> <u>0.12</u> (	.00
25.8.9.00 20.58 0.00 0.10 0.00 16.27 0.00 (	.16
<u>26.8. 9.00</u> <u>31.53</u> <u>0.31</u> <u>0.30</u> <u>0.00</u> <u>20.92</u> <u>0.53</u> (	.46
27.8.9.00 18.29 0.00 0.00 0.00 14.76 0.00 (	
28.8. 9.00         32.38         0.00         0.00         22.08         0.00         (	.00

	wavelength		conductivity detector				
sample	CI	Br	NO3	PO4	SO4	Br	NO3

<b>BKT 05</b>							
standard3	50.07	20.08	5.00	9.46	20.19	20.26	5.06
29.6. 7.00	31.76	0.00	0.00	0.00	11.72	0.00	0.16
29.6. 15.00	15.37	0.00	0.13	0.00	9.10	0.00	0.21
29.6. 23.00	14.32	0.00	0.00	0.00	7.97	0.00	0.16
30.6. 7.00	32.01	0.00	0.00	0.00	16.52	0.00	0.09
30.6. 23.00	32.32	0.00	0.00	0.00	16.73	0.00	0.09
1.7. 7.00	32.12	0.00	0.00	0.00	16.57	0.00	0.06
1.7. 15.00	13.61	0.00	0.00	0.00	7.96	0.21	0.19
1.7. 23.00	31.90	0.00	0.00	0.00	16.67	0.00	0.10
2.7. 9.00	22.73	0.00	2.58	0.00	14.08	0.00	2.00
2.7. 19.00	28.41	0.00	0.82	0.00	13.56	0.00	1.05
3.7. 9.00	22.81	1.30	0.00	0.00	12.56	1.29	1.67
4.7. 9.00	22.71	23.21	0.93	0.00	12.92	30.50	2.08
5.7.9.00	23.61	42.85	3.18	0.00	14.93	50.82	4.09
6.7. 9.00	21.24	60.16	0.00	0.00	14.27	72.96	2.44
7.7. 9.00	14.00	82.73	3.84	0.00	9.59	89.68	2.81
9.7. 9.00	14.04	124.62	0.00	0.00	10.43	137.05	2.38
12.7.9.00	13.55	132.51	0.00	0.00	9.64	138.65	0.00
15.7. 9.00	15.36	128.89	0.00	0.00	10.51	148.48	1.62
8.8. 19.00	31.50	23.30	0.89	0.09	13.72	22.12	0.82
09.8. 9.00	25.01	37.07	0.00	0.00	15.61	31.87	0.00
09.8. 19.00	17.20	9.83	0.97	0.00	9.77	10.67	1.13
10.8. (sp3)	5.54	2.76	0.00	0.00	3.91	4.73	0.46
11.8. 9.00	31.75	13.20	0.00	0.12	20.30	16.01	0.00
11.8. 19.00	17.33	6.27	0.00	0.10	11.32	7.26	0.00
12.8. 9.00	56.17	6.96	0.40	0.00	16.98	8.05	0.55
12.8. 19.00	23.58	5.02	0.00	0.00	15.62	5.98	0.00
13.8. 9.00	28.98	4.64	0.00	0.00	19.17	6.26	0.00
13.8. 19.00	10.00	0.94	0.00	0.00	7.10	1.46	0.00
14.8. 9.00	33.68	4.16	0.94	0.00	16.07	5.15	1.07
14.8. 19.00	14.11	0.62	0.00	0.14	9.61	0.83	0.00
15.8. 9.00	24.41	1.99	0.86	0.00	10.63	2.66	1.14
15.8. 19.00	25.07	2.10	0.90	0.00	12.40	2.72	1.10
16.8. 9.00	250.92	5.04	0.00	1.26	22.58	7.50	0.02
16.8. 19.00	29.95	0.18	0.00	0.00	20.34	0.24	0.00
17.8. 9.00	17.21	0.23	0.22	0.00	11.62	0.38	0.36
17.8. 19.00	22.37	0.61	0.49	0.00	15.63	0.71	0.65
18.8. 9.00	32.38	0.67	0.00	0.00	22.14	0.89	0.07
18.8. 19.00	28.75	0.00	0.00	0.00	19.75	0.00	0.02
19.8. 9.00	16.20	0.00	0.00	0.00	13.15	0.00	0.09
20.8. 9.00	19.67	0.00	0.00	0.00	14.97	0.00	0.00
21.8. 9.00	29.73	0.00	0.00	0.00	20.75	0.00	0.03
22.8. 9.00	17.37	0.00	0.09	0.00	13.70	0.00	0.20
23.8. 9.00	30.01	0.00	0.16	0.00	21.66	0.00	0.32
24.8. 9.00	12.75	0.00	0.00	0.00	11.36	0.00	0.03
25.8.9.00	24.99	0.21	0.50	0.00	18.55	0.39	0.74
26.8. 9.00	24.40	0.20	0.30	0.00	17.54	0.31	0.44
27.8.9.00	19.40	0.00	0.17	0.00	15.22	0.00	0.30
28.8. 9.00	31.49	0.00	0.00	0.00	22.03	0.00	0.00

	wavelength d	etector				conductivity	detector
sample	CI	Br	NO3	PO4	SO4	Br	NO3
GNT 02	-	-		-		-	
standard1	8.21	4.12	0.92	2.37	4.18	5.26	1.00
29.6.7.00	38.29	0.00	0.15	0.00	12.60	0.00	0.25
29.6. 15.00	36.76	0.00	0.15	0.00	13.97	0.09	0.28
29.6. 23.00	7.33	0.00	0.00	0.00	3.73	0.16	0.06
30.6.7.00	22.59	3.10	0.20	0.00	9.11	4.18	0.31
30.6. 23.00	30.64	42.19	0.90	0.00	13.70	36.91	1.14
1.7.7.00	28.79	59.56	1.46	0.00	13.69	47.97	1.72
1.7. 15.00	20.76	44.51	1.09	0.00	10.32	38.24	1.30
1.7.23.00	26.34	77.51	1.75	0.00	13.37	59.14	2.06
2.7.9.00	7.41	25.04	0.00	0.00	5.44	36.13	1.06
2.7. 19.00	17.52	87.03	2.44	0.00	10.77	99.27	3.23
3.7. 9.00	16.79	99.98	3.93	0.00	12.23	111.70	4.40
4.7. 9.00	15.98	110.88	3.79	0.00	10.47	121.92	3.81
5.7.9.00	9.07	41.05	1.26	0.00	5.07	47.64	1.95
6.7. 9.00	15.76	107.03	0.00	0.00	9.47	124.79	3.34
7.7.9.00	10.67	54.30	0.00	0.00	7.72	68.29	2.02
9.7. 9.00	15.63	127.32	0.00	0.00	9.56	144.02	1.59
12.7.9.00	17.64	124.84	0.00	0.00	13.49	146.41	2.52
15.7. 9.00	5.36	46.03	0.00	0.00	4.18	56.35	0.00

## Tab.: A22 anions from M10

RT 03

standard2	18.18	9.12	1.75	5.22	8.96	10.65	1.98
29.6. 7.00	17.48	0.00	0.00	0.00	9.03	0.00	0.09
29.6. 15.00	16.34	0.00	0.00	0.00	8.97	0.00	0.12
29.6. 23.00	14.23	0.00	0.00	0.00	7.14	0.00	0.00
30.6. 7.00	11.59	0.00	0.00	0.00	5.81	0.00	0.03
30.6. 23.00	32.74	0.00	0.20	0.00	15.69	0.08	0.35
1.7. 15.00	31.54	0.00	0.00	0.00	14.95	0.00	0.11
1.7. 23.00	32.33	0.00	0.00	0.00	15.69	0.00	0.05
2.7. 9.00	6.91	0.00	0.00	0.00	4.84	0.00	0.00
2.7. 19.00	23.07	0.00	0.00	0.00	11.88	0.00	0.29
3.7. 9.00	29.38	0.00	2.67	0.75	14.40	1.20	3.09
4.7. 9.00	22.07	15.28	0.00	0.00	13.17	20.58	0.95
5.7.9.00	18.50	51.23	3.18	0.00	11.91	61.44	3.74
6.7. 9.00	21.38	97.52	4.93	0.00	13.09	109.01	5.36
7.7.9.00	16.10	122.81	4.04	0.00	12.04	135.60	4.62
9.7. 9.00	17.22	122.78	0.00	0.00	9.83	145.60	3.76
12.7.9.00	14.15	137.07	0.00	0.00	9.99	145.96	1.59
15.7. 9.00	14.75	131.09	0.00	0.00	10.97	149.66	1.89

standard3	49.11	19.92	4.80	9.48	19.77	20.63	5.00
29.6. 7.00	28.15	0.00	0.15	0.00	16.74	0.00	0.25
29.6. 15.00	33.56	0.00	0.30	0.00	15.43	0.00	0.38

	wavelength d	etector				conductivity	detector
sample	CI	Br	NO3	PO4	SO4	Br	NO3
29.6. 23.00	30.54	0.00	0.00	0.00	14.63	0.00	0.13
30.6. 7.00	22.82	0.00	0.00	0.00	9.85	0.20	0.08
30.6. 23.00	33.23	0.00	0.11	0.00	15.51	0.00	0.13
1.7. 15.00	42.22	4.92	0.00	0.00	18.83	6.54	0.21
1.7. 23.00	33.09	0.00	0.13	0.00	15.61	0.00	0.22
2.7. 9.00	3.52	0.00	0.00	0.00	2.45	0.00	0.78
2.7. 19.00	23.84	0.00	0.00	0.00	13.50	0.00	0.56
3.7. 9.00	25.33	0.00	1.88	0.00	20.08	0.00	2.82
4.7. 9.00	22.51	0.00	0.00	0.00	12.62	0.00	0.66
5.7.9.00	10.21	9.28	2.28	0.00	5.69	11.23	3.21
6.7. 9.00	18.38	47.70	0.00	0.00	11.36	63.11	1.62
7.7.9.00	15.59	132.65	4.59	0.00	12.42	145.39	4.85
9.7. 9.00	13.84	126.54	3.71	0.00	9.26	139.55	3.68
12.7.9.00	13.94	130.20	0.00	0.00	9.81	146.89	1.37
15.7. 9.00	13.34	85.78	0.00	0.00	9.86	104.81	2.33

BKT 05							
standard1	8.38	4.11	1.05	2.32	4.46	5.35	1.21
29.6. 7.00	10.39	0.00	0.78	0.00	6.80	0.00	0.87
29.6. 15.00	26.26	0.00	0.00	0.00	11.80	0.00	0.00
29.6. 23.00	61.47	1.02	0.63	0.00	27.67	1.47	0.76
30.6. 7.00	55.62	0.19	0.14	0.00	25.09	0.30	0.22
30.6. 23.00	34.17	0.00	0.00	0.00	15.19	0.06	0.11
1.7. 15.00	31.92	0.00	0.00	0.00	15.18	0.00	0.08
1.7.23.00	33.74	0.00	0.00	0.00	15.14	0.00	0.10
2.7. 9.00	22.42	0.00	1.73	0.00	14.05	0.66	2.85
2.7. 19.00	24.49	0.00	0.00	0.00	11.25	0.00	0.58
3.7. 9.00	27.07	0.00	0.94	0.00	17.10	0.00	1.31
4.7. 9.00	24.13	0.00	0.00	0.00	12.92	0.00	0.62
5.7. 9.00	13.20	0.00	0.50	0.00	7.43	0.00	0.74
6.7. 9.00	22.72	0.00	0.00	0.00	12.65	0.00	0.25
7.7.9.00	10.38	0.00	0.81	0.00	6.70	0.00	1.37
9.7. 9.00	20.09	16.98	2.92	0.00	11.15	21.25	2.82
12.7.9.00	19.37	55.14	0.00	0.00	11.62	71.04	1.45
15.7.9.00	5.63	55.38	0.00	0.00	5.11	63.43	0.00

	sample	date	As(III)	dilution	As(tot)	dilution	As(III) cal.	As(V) cal.
			[µg/I]		[µg/I]			. /
1	11000						<u> </u>	
	M303x1	15.09.2000	7.42	5	15.12	10	34.77	116.43
	M303x1	19.09.2000	3.67	10	15.12	10	34.36	116.84
	M303x1	21.09.2000	3.35	10	15.12	10	31.10	120.10
	M303x1	25.09.2000	4.03	10	15.12	10	38.04	113.16
	M303x1	26.09.2000	3.54	10	15.12	10	33.04	118.16
	M303x1	03.10.2000	3.46	10	15.12	10	32.22	118.98
	M303x1	10.10.2000	3.47	10	14.92	10	32.36	116.84
	M303x1	13.10.2000	3.50	10	14.92	10	32.67	116.53
	M303x1	23.10.2000	3.98	10	14.92	10	37.57	111.63
no fridge	M303x1	15.01.2001	2.45	10	14.75	10	21.99	125.51
	M303x2	15.09.2000	6.90	5	14.96	10	32.15	117.45
	M303x2	19.09.2000	3.20	10	14.96	10	29.60	120.00
	M303x2	21.09.2000	2.93	10	14.96	10	26.84	122.76
	M303x2	25.09.2000	3.78	10	14.96	10	35.52	114.08
	M303x2	26.09.2000	3.14	10	14.96	10	28.99	120.61
	M303x2	03.10.2000	3.21	10	14.96	10	29.70	119.90
	M303x2	10.10.2000	3.34	10	15.95	10	30.83	128.67
	M303x2	13.10.2000	3.40	10	15.95	10	31.44	128.06
	M303x2	23.10.2000	3.76	10	15.95	10	35.11	124.39
no fridge	M303x2	15.01.2001	2.25	10	14.58	10	19.98	125.82
	-	-			-			
	M303x3	15.09.2000	5.87	5	15.06	10	26.88	123.72
	M303x3	19.09.2000	2.66	10	15.06	10	24.07	126.53
	M303x3	21.09.2000	2.40	10	15.06	10	21.42	129.18
	M303x3	25.09.2000	3.07	10	15.06	10	28.25	122.35
	M303x3	26.09.2000	2.44	10	15.06	10	21.82	128.78
	M303x3	03.10.2000	2.63	10	15.06	10	23.76	126.84
	M303x3	10.10.2000	2.70	10	15.16	10	24.46	127.14
	M303x3	13.10.2000	2.65	10	15.16	10	23.95	127.65
	M303x3	23.10.2000	2.91	10	15.16	10	26.60	125.00
no fridge	M303x3	15.01.2001	1.74	10	14.59	10	14.78	131.12
	M303x4	15.09.2000	6.14	5	15.31	10	28.20	124.90
	M303x4	19.09.2000	2.78	10	15.31	10	25.24	127.86
	M303x4	21.09.2000	2.68	10	15.31	10	24.22	128.88
	M303x4	25.09.2000	3.33	10	15.31	10	30.85	122.25
	M303x4	26.09.2000	2.73	10	15.31	10	24.73	128.37
	M303x4	03.10.2000	2.88	10	15.31	10	26.26	126.84
	M303x4	10.10.2000	2.87	10	15.16	10	26.19	125.41
	M303x4	13.10.2000	3.02	10	15.16	10	27.72	123.88
	M303x4	23.10.2000	3.39	10	15.16	10	31.50	120.10
no fridge	M303x4	15.01.2001	2.24	10	14.92	10	19.81	129.39

Tab.: A23 storage test with samples from 12.09.00

	sample	date	As(III) [µg/I]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
	M303x5	15.09.2000	5.78	5	15.12	10	26.40	124.80
	M303x5	19.09.2000	2.68	10	15.12	10	24.26	126.94
	M303x5	21.09.2000	2.47	10	15.12	10	22.12	129.08
	M303x5	25.09.2000	3.12	10	15.12	10	28.75	122.45
	M303x5	26.09.2000	2.54	10	15.12	10	22.83	128.37
	M303x5	03.10.2000	2.73	10	15.12	10	24.77	126.43
	M303x5	10.10.2000	2.85	10	15.12	10	26.00	125.20
	M303x5	13.10.2000	2.83	10	15.12	10	25.79	125.41
	M303x5	23.10.2000	3.24	10	15.12	10	29.98	121.22
no fridge	M303x5	15.01.2001	1.93	10	14.94	10	16.64	132.76

	sample	date	As(III) [µg/I]	dilution	As(tot) [µg/l]	dilution	As(III) cal.	As(V) cal.
	-				-	-		
	1	15.09.2000	10.99	10	12.41	10	109.61	14.49
	1	19.09.2000	9.66	10	12.41	10	96.04	28.06
	1	21.09.2000	10.29	10	12.41	10	102.47	21.63
	1	25.09.2000	11.19	10	12.41	10	111.65	12.45
	1	26.09.2000	10.89	10	12.41	10	108.59	15.51
	1	10.10.2000	10.44	10	12.15	10	104.05	17.45
	1	13.10.2000	10.89	10	12.15	10	108.64	12.86
	1	23.10.2000	10.91	10	12.15	10	108.85	12.65
	1	20.11.2000	11.65	10	12.15	10	116.40	5.10
no fridge	1	15.01.2001	5.34	10	11.88	10	52.06	66.74
	2	15.09.2000	10.94	10	12.23	10	109.14	13.16
	2	19.09.2000	9.87	10	12.23	10	98.22	24.08
	2	21.09.2000	10.53	10	12.23	10	104.95	17.35
	2	25.09.2000	11.34	10	12.23	10	113.22	9.08
	2	26.09.2000	11.06	10	12.23	10	110.36	11.94
	2	10.10.2000	10.30	10	12.43	10	102.56	21.74
	2	13.10.2000	10.84	10	12.43	10	108.07	16.23
	2	23.10.2000	10.96	10	12.43	10	109.30	15.00
	2	20.11.2000	11.51	10	12.43	10	114.91	9.39
no fridge	2	15.01.2001	5.93	10	12.04	10	58.05	62.35
	3	15.09.2000	10.99	10	12.47	10	109.60	15.10
	3	19.09.2000	9.94	10	12.47	10	98.88	25.82
	3	21.09.2000	10.76	10	12.47	10	107.25	17.45
	3	25.09.2000	11.24	10	12.47	10	112.15	12.55
	3	26.09.2000	10.65	10	12.47	10	106.13	18.57
	3	10.10.2000	10.34	10	12.30	10	103.00	20.00
	3	13.10.2000	10.79	10	12.30	10	107.59	15.41
	3	23.10.2000	10.95	10	12.30	10	109.22	13.78
	3	20.11.2000	11.24	10	12.30	10	112.18	10.82
no fridge	3	15.01.2001	5.74	10	11.91	10	56.14	62.96

Tab.: A24 storage test with anoxic water + 100µg/I As(III)

date	time	hours	total hours	discarge	total	total corrected	average	tracer addition	total tracer	average tracer	Lf1	Lf2	comment
24.06.2000	12:00	0	0	420	0	0		3	3	0.000	405	192	
24.06.2000	19:00	7	7	420	140	177	25.29	0	3	0.000			
25.06.2000	07:00	12	19	410	435	472	24.84	2	5	0.105			
25.06.2000	12:00	5	24	435	568	605	25.21	0	5	0.083	406		
25.06.2000	15:00	3	27	430				0	5	0.074			
25.06.2000	21:00	6	33	440	800	837	25.36	1	6	0.091			
25.06.2000	23:00	2	35	440				0	6	0.086	406		
26.06.2000	07:00	8	43	425	1050	1087	25.28	0	6	0.070			
26.06.2000	11:00	4	47	425	62	1149	24.45	0	6	0.064	414		
26.06.2000	16:45	5.75	52.75		225	1312	24.87	3	9	0.114			
26.06.2000	23:00	6.25	59					0	9	0.102	425		
27.06.2000	11:30	12.5	71.5	390	657	1744	24.39	2	11	0.112	431	191	
27.06.2000	15:15	3.75	75.25	435	750	1837	24.41	2	13	0.133	410	192	
27.06.2000	20:00	4.75	80		861	1948	24.35	0	13	0.125			
27.06.2000	23:00	3	83	460	955	2042	24.60	0	13	0.120			
28.06.2000	08:00	9	92	450	1190	2277	24.75	0	13	0.109			
28.06.2000	15:00	7	99	450	1383	2470	24.95	2	15	0.121			
28.06.2000	19:00	4	103	440	1499	2586	25.11	0	15	0.117	403		
28.06.2000	23:00	4	107	455	1588	2675	25.00	0	15	0.112			
29.06.2000	08:00	9	116	450	1837	2924	25.21	1	16	0.112			
29.06.2000	14:45	6.75	122.75	430	2009	3096	25.22	2	18	0.122			
29.06.2000	23:00	8.25	131	425	2234	3321	25.35	0	18	0.115	413		
30.06.2000	07:00	8	139	420	2444	3531	25.40	1	19	0.115			
30.06.2000	15:30	8.5	147.5	450	2635	3722	25.23	1	20	0.115	413		HPLC ausgef
01.07.2000	09:00	17.5	165	450	2693	3780	22.91	0	20	0.103	400		GEO ausgef
01.07.2000	15:45	6.75	171.75	420	2865	3952	23.01	1	21	0.105	413	190	
01.07.2000	23:00	7.25	179	400	3019	4106	22.94	0	21	0.101	435	191	
02.07.2000	10:00	11	190	430	3266	4353	22.91	2	23	0.105	444	196	
02.07.2000	19:15	9.25	199.25	430	3500	4587	23.02	1	24	0.105	435	200	
03.07.2000	09:30	14.25	213.5	440	3885	4972	23.29	1	25	0.103	412	192	
03.07.2000	18:00	8.5	222	440	4108	5195	23.40	1	26	0.104	420	194	

# Tab.: A25 flow rates of the tracer- and goundwater pumps

dato	timo	hours	total	discargo	total	total corrected	avorado	tracer	total	average	I f1	1 f2	commont
uate	ume	nours	hours	uiscarge	lotai	total confected	average	addition	tracer	tracer			comment
04.07.2000	11:30	17.5	239.5	430	4569	5656	23.62	2	28	0.104	422	194	
05.07.2000	09:45	22.25	261.75	430	5140	6227	23.79	3	31	0.107	430	195	
05.07.2000	19:30	9.75	271.5	420	5381	6468	23.82	0	31	0.103	429	197	
06.07.2000	09:45	14.25	285.75	370	5736	6823	23.88	1	32	0.101			
06.07.2000	19:45	10	295.75	440	5991	7078	23.93	2	34	0.105			
07.07.2000	10:30	14.75	310.5	320	6307	7394	23.81	2	36	0.106	434	199	
07.07.2000	18:45	8.25	318.75	450	6526	7613	23.88	0	36	0.104			
08.07.2000	09:45	15	333.75	343	6902	7989	23.94	1	37	0.102			
08.07.2000	18:30	8.75	342.5	470	7125	8212	23.98	1	38	0.102	407		
09.07.2000	10:00	15.5	358	470	7565	8652	24.17	3	41	0.106	428	205	
09.07.2000	18:45	8.45	366.45	430	7796	8883	24.24	1	42	0.106			
10.07.2000	09:00	14.25	380.7	420	8173	9260	24.32	1	43	0.105	457	205	
10.07.2000	19:00	10	390.7	450	8424	9511	24.34	0	43	0.102	446	206	
11.07.2000	10:30	15.5	406.2	470	8850	9937	24.46	3	46	0.106	436	205	
12.07.2000	09:30	23	429.2	430	9408	10495	24.45	1	47	0.103	454	207	
12.07.2000	19:00	9.5	438.7	430	9650	10737	24.47	2	49	0.105			
13.07.2000	09:00	14	452.7	430	10005	11092	24.50	0	49	0.102	437	208	
13.07.2000	19:30	10.5	463.2	440	10276	11363	24.53	3	52	0.106	437	211	
14.07.2000	09:00	13.5	476.7	440	10629	11716	24.58	1	53	0.105	430	210	
14.07.2000	18:30	9.5	486.2	425	10881	11968	24.62	0	53	0.103	445	211	
15.07.2000	10:30	16	502.2	425	11275	12362	24.62	2	55	0.104			Airtrap leer!
15.07.2000	18:45	8.25	510.45	465	11505	12592	24.67	0	55	0.102			
16.07.2000	09:30	14.75	525.2	365	11830	12917	24.59	2	57	0.103	449	221	
16.07.2000	19:00	11.5	536.7	450	12084	13171	24.54	1	58	0.102			
17.07.2000	09:45	14.75	551.45	340	12430	13517	24.51	2	60	0.103	453	212	
17.07.2000	18:45	9	560.45	470	12655	13742	24.52	0	60	0.102			
18.07.2000	10:00	15.25	575.7	410	13018	14105	24.50	3	63	0.104	449	210	
18.07.2000	18:30	8.5	584.2	460	13256	14343	24.55	0	63	0.103			
19.07.2000	09:30	15	599.2	240	13571	14658	24.46	2	65	0.103	588	210	
19.07.2000	19:30	10	609.2	460	13826	14913	24.48	1	66	0.103			
20.07.2000	09:30	14.5	623.7	445	14202	15289	24.51	1	67	0.103	436	211	
20.07.2000	19:00	9.5	633.2	450	14459	15546	24.55	1	68	0.103			
21.07.2000	10:00	15	648.2	460	14854	15941	24.59	2	70	0.103	439	213	
21.07.2000	19:00	9	657.2	435	15094	16181	24.62	1	71	0.103			

date	time	hours	total	discarge	total	total corrected	average	tracer	total	average	Lf1	Lf2	comment
uaro		neuro	hours	ulocal go	lotai		aronago	addition	tracer	tracer			
22.07.2000	09:30	14.5	671.7	460	15480	16567	24.66	0	71	0.101	423	210	
22.07.2000	19:00	8.5	680.2	400	15704	16791	24.69	3	74	0.104	461	213	
23.07.2000	19:45	24.75	704.95	430	16339	17426	24.72	2	76	0.104	447	210	
24.07.2000	10:45	15	719.95	420	16715	17802	24.73	Rest 1,5					

# 11.2 Arsenic reduction with dissolved sulfide

	75 μmol/L		150 µmol/L		300 µmol/L		150 µmol/L bottle	
days	As(III) [µmol/I]	As(V) [µmol/l]	As(III) [µmol/I]	As(V) [µmol/l]	As(III) [µmol/I]	As(V) [µmol/l]	As(III) [µmol/I]	As(V) [µmol/l]
1	19.5	796	24.2	752	30.2	724		
2	18.2	835	33.3	809	77.2	741		
3	21.4	751	34.9	688	106.3	590		
4	23.3	749	42.6	736	144.6	603	48.5	794
5	23.7	688	52.3	726	136.5	574	62.3	727
6	22.8	675	57.9	626	177.1	544	73.8	650
7	28.2	732	61.6	702	178.7	565	73.3	713
8	13.8	738	51.3	770	128.8	627	68.0	747
9	33.6	694	78.8	701	219.9	537	91.2	694
10	26.1	707	68.7	679	201.9	539	83.5	643
11	23.1	753	70.7	667	166.9	520	87.5	651
12	19.5	696	62.7	583	159.6	522	86.9	669
13	13.6	752	68.2	642	179.2	505	98.3	644
14	15.8	691	68.8	661	105.4	629	101.9	665
15	32.6	658	78.4	667	200.7	631	106.4	719
16	30.6	837	87.7	828	119.4	762	113.0	779
18	24.6	723	82.7	687	181.5	627	115.6	715
21	44.6	822	103.8	775	356.0	501	131.8	768
30	59.3	723	104.8	689	307.0	449		
49	48.5	934	110.4	758	297.6	621		

Tab. A26 Long term test with As(V) and different concentrations of  $H_2S$ 

Tab. A27 Long term test with As(V), Na<sub>2</sub>S, H<sub>2</sub>S, buffer, MnO<sub>2</sub> or  $Fe_2O_3$ 

comple	dovo	As(III)	As(V)	ъЦ	Mn2+	Fe2+	Sulfid
Sample	uays	[µmol/L]	[µmol/L]	μц	[mmol/L]	[µmol/L]	[mmol/L]
As(V)	1	1.37	99.53				
	2			4.21			
	8	0.08	105.25	4.23			
As(V)+ Buffer	1	0.42	82.78				
	2			6.88			
	8	0.46	84.87	6.76			
As(V)+buffer+H2S	1	0.13	81.81				
	2			6.85			0.25
	3	0.00	85.74				

	6	0.02	90.90			
	8	0.67	107.24	6.83		19.44
	10	1.04	83.95			
	13	1.30	76.24			
	17	1.58	87.63			
	20	1.54	83.24			
	22	1.43	83.65			
	29	1.74	84.41			
	36	1.72	74.15			
	38	1.82	84.06			
	41	1.69	74.86			
	43	1.57	78.95			
	45	1.57	93.76			
	48	1.50	95.90			
	51	1.41	93.25			
	55	1.40	104.28			
	59	0.75	89.57			
	62	0.42	95.55			
	64	0.27	86.00			
	69	0.10	73.18			
As(V)+buffer+Na2S	1	0.19	82.57			
						0.74
	2			6.85		0.74
	2 3	0.00	86.05	6.85		0.74
	2 3 6	0.00	86.05 75.32	6.85		0.74
	2 3 6 8	0.00 0.17 0.16	86.05 75.32 95.70	6.85		0.74
	2 3 6 8 10	0.00 0.17 0.16 0.41	86.05 75.32 95.70 71.39	6.85		0.74
	2 3 6 8 10 13	0.00 0.17 0.16 0.41 0.87	86.05 75.32 95.70 71.39 71.24	6.85		0.74
	2 3 6 8 10 13 17	0.00 0.17 0.16 0.41 0.87 1.26	86.05 75.32 95.70 71.39 71.24 82.37	6.85		0.74
	2 3 6 8 10 13 17 20	0.00 0.17 0.16 0.41 0.87 1.26 1.62	86.05 75.32 95.70 71.39 71.24 82.37 80.43	6.85		0.74
	2 3 6 8 10 13 17 20 22	0.00 0.17 0.16 0.41 0.87 1.26 1.62 1.65	86.05 75.32 95.70 71.39 71.24 82.37 80.43 82.27	6.85		0.29
	2 3 6 8 10 13 17 20 22 29	0.00 0.17 0.16 0.41 0.87 1.26 1.62 1.65 2.75	86.05 75.32 95.70 71.39 71.24 82.37 80.43 82.27 77.31	6.85		0.29
	2 3 6 8 10 13 17 20 22 29 36	0.00 0.17 0.16 0.41 0.87 1.26 1.62 1.65 2.75 3.82	86.05 75.32 95.70 71.39 71.24 82.37 80.43 82.27 77.31 75.12	6.92		0.29
	2 3 6 8 10 13 17 20 22 29 36 38	0.00 0.17 0.16 0.41 0.87 1.26 1.62 1.65 2.75 3.82 3.68	86.05 75.32 95.70 71.39 71.24 82.37 80.43 82.27 77.31 75.12 70.93	6.85		0.74
	2 3 6 8 10 13 17 20 22 29 36 38 43	0.00 0.17 0.16 0.41 0.87 1.26 1.62 1.65 2.75 3.82 3.68 3.62	86.05 75.32 95.70 71.39 71.24 82.37 80.43 82.27 77.31 75.12 70.93 73.94	6.92		0.29
	2 3 6 8 10 13 17 20 22 29 36 38 43 45	0.00 0.17 0.16 0.41 0.87 1.26 1.62 1.65 2.75 3.82 3.68 3.62 4.64	86.05 75.32 95.70 71.39 71.24 82.37 80.43 82.27 77.31 75.12 70.93 73.94 95.70	6.92		0.29
	2 3 6 8 10 13 17 20 22 29 36 38 43 45 48	0.00 0.17 0.16 0.41 0.87 1.26 1.62 1.65 2.75 3.82 3.68 3.62 4.64 4.56	86.05 75.32 95.70 71.39 71.24 82.37 80.43 82.27 77.31 75.12 70.93 73.94 95.70 89.47	6.92		0.29
	2 3 6 8 10 13 17 20 22 29 36 38 43 45 48 51	0.00 0.17 0.16 0.41 0.87 1.26 1.62 1.65 2.75 3.82 3.68 3.62 4.64 4.56 4.57	86.05 75.32 95.70 71.39 71.24 82.37 80.43 82.27 77.31 75.12 70.93 73.94 95.70 89.47 98.15	6.92		0.74

	59	4.95	89.57			
	62	5.16	87.17			
	64	4.54	100.60			
	69	4.36	77.47			
	79	0.00	0.00			
As(V)+buffer+Na2S+MnO2	1	0.25	7.59		0.44	
	2			6.75		0.00
	3	0.00	1.74		0.58	
	6	0.00	0.69		1.00	
	8	0.04	0.31	6.78		0.00
	10	0.00	0.31			
	13	0.00	0.00			
	17	0.00	0.98			
	20	0.00	0.33			
	22	0.00	0.94			
	29	0.00	0.17			
	34				1.10	
	36	0.00	0.00		1.52	
	38	0.00	0.00		2.21	
	41	0.00	0.43		2.34	
	43	0.06	0.14		2.18	
	45	0.01	0.18		2.16	
	48	0.03	1.06		2.13	
	51	0.00	0.00		2.02	
	55	0.06	0.10		1.98	
	59	0.02	0.06		1.87	
	62	0.01	0.02		1.81	
	64	0.06	0.04		1.79	
	69	0.02	0.11		1.65	
	79	0.00	0.00		1.67	
As(V)+buffer+H2S+MnO2	1	0.08	0.68		0.46	
	2			6.79		0.00
	3	0.00	1.39		0.59	
	6	0.00	1.00		0.96	
	8	0.10	0.43	6.77		0.00
	10	0.00	0.31			
	13	0.00	0.34			

	17	0.01	0.20				
	20	0.00	0.38				
	22	0.00	0.23				
	29	0.00	0.07				
	34				1.10		
	36	0.00	0.37		1.53		
	38	0.00	2.05		2.45		
	41	0.00	0.00		3.14		
	43	0.00	0.62		2.92		
	45	0.03	0.93		2.82		
	48	0.01	2.34		2.81		
	51	0.00	1.81		2.82		
	55	0.05	0.38		2.81		
	59	0.01	0.63		2.82		
	62	0.05	1.93		2.73		
	64	0.03	0.03		2.75		
	69	0.01	0.04		2.68		
	79	0.00	0.00		2.74		
As(V)+buffer+H2S+Fe2O3	1	0.21	0.43			0 54	
		0.21	0.40			0.04	
	2	0.21	0.40	6.84		0.00	0.36
	2	0.00	0.00	6.84		0.00	0.36
	2 3 6	0.00	0.00	6.84		0.00 8.95 0.00	0.36
	2 3 6 8	0.00 0.00 0.00	0.00 0.00 0.00	6.84 6.82		0.00 0.00 8.95 0.00	0.36
	2 3 6 8 10	0.00 0.00 0.00 0.01	0.00 0.00 0.00 0.35	6.84		0.00 0.00 8.95 0.00	0.36
	2 3 6 8 10 13	0.00 0.00 0.00 0.01 0.00	0.00 0.00 0.00 0.35 0.47	6.84		0.00 8.95 0.00	0.36
	2 3 6 8 10 13 17	0.00 0.00 0.00 0.01 0.00 0.19	0.00 0.00 0.00 0.35 0.47 0.59	6.84		0.00 8.95 0.00	0.36
	2 3 6 8 10 13 17 20	0.00 0.00 0.00 0.01 0.00 0.19 0.00	0.00 0.00 0.00 0.35 0.47 0.59 2.00	6.84		0.00 8.95 0.00	0.36
	2 3 6 8 10 13 17 20 22	0.00 0.00 0.00 0.01 0.00 0.19 0.00 0.00	0.40 0.00 0.00 0.35 0.47 0.59 2.00 0.59	6.84		0.00 8.95 0.00	0.36
	2 3 6 8 10 13 17 20 22 29	0.00 0.00 0.00 0.01 0.00 0.19 0.00 0.00	0.40 0.00 0.00 0.35 0.47 0.59 2.00 0.59 0.65	6.84		0.00 8.95 0.00	0.36
	2 3 6 8 10 13 17 20 22 29 36	0.21 0.00 0.00 0.00 0.01 0.00 0.00 0.00 0.00 0.00	0.40 0.00 0.00 0.35 0.47 0.59 2.00 0.59 0.65 0.80	6.84		0.04 0.00 8.95 0.00	0.36
	2 3 6 8 10 13 17 20 22 29 36 38	0.21 0.00 0.00 0.00 0.01 0.00 0.00 0.00 0.00 0.00 0.02	0.40 0.00 0.00 0.35 0.47 0.59 2.00 0.59 0.65 0.80 0.15	6.84		0.04 0.00 8.95 0.00 4.05 3.01	0.36
	2 3 6 8 10 13 17 20 22 29 36 38 41	0.21 0.00 0.00 0.01 0.00 0.19 0.00 0.00 0.00 0.00 0.00 0.02 0.25	0.40 0.00 0.00 0.35 0.47 0.59 2.00 0.59 0.65 0.80 0.15 0.00	6.84		0.04 0.00 8.95 0.00 4.05 3.01 5.03	0.36
	2 3 6 8 10 13 17 20 22 29 36 38 41 43	0.21 0.00 0.00 0.01 0.00 0.19 0.00 0.00 0.00 0.00 0.00 0.02 0.25 0.27	0.00 0.00 0.00 0.35 0.47 0.59 2.00 0.59 0.65 0.80 0.15 0.00 0.23	6.84		0.00 8.95 0.00 4.05 3.01 5.03 6.61	0.36
	2 3 6 8 10 13 17 20 22 29 36 38 41 43 45	0.21 0.00 0.00 0.00 0.01 0.00 0.00 0.00 0.00 0.00 0.00 0.02 0.25 0.27 0.46	0.40 0.00 0.00 0.35 0.47 0.59 2.00 0.59 0.65 0.80 0.15 0.00 0.23 0.10	6.84		0.00 8.95 0.00 4.05 3.01 5.03 6.61 9.11	0.36
	2 3 6 8 10 13 17 20 22 29 36 38 41 43 45 48	0.21 0.00 0.00 0.00 0.01 0.00 0.19 0.00 0.00 0.00 0.00 0.00 0.00 0.02 0.25 0.27 0.46 0.36	0.00 0.00 0.00 0.35 0.47 0.59 2.00 0.59 0.65 0.80 0.15 0.00 0.23 0.10 0.11	6.84		0.00 8.95 0.00 4.05 3.01 5.03 6.61 9.11 6.52	0.36
	2 3 6 8 10 13 17 20 22 29 36 38 41 43 45 48 51	0.21 0.00 0.00 0.00 0.01 0.00 0.19 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.02 0.25 0.27 0.46 0.36 0.33	0.40 0.00 0.00 0.35 0.47 0.59 2.00 0.59 0.65 0.80 0.15 0.00 0.23 0.10 0.11 0.05	6.84		0.00 8.95 0.00 4.05 3.01 5.03 6.61 9.11 6.52 7.75	0.36
	2 3 6 8 10 13 17 20 22 29 36 38 41 43 45 48 51 55	0.21 0.00 0.00 0.00 0.01 0.00 0.19 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.02 0.25 0.27 0.46 0.36 0.33 0.28	0.40 0.00 0.00 0.35 0.47 0.59 2.00 0.59 0.65 0.80 0.15 0.00 0.23 0.10 0.11 0.05 0.08	6.84		0.04 0.00 8.95 0.00 4.05 3.01 5.03 6.61 9.11 6.52 7.75 10.03	0.36

	62	0.15	0.10			6.73	
	64	0.25	0.08			9.69	
	69	0.16	0.07			6.37	
	79	0.00	0.00			7.14	
As(V)+buffer+Na2S+Fe2O3	1	0.09	0.27			0.79	
	2			6.86			0.59
	3	0.00	0.00			1.58	
	6	0.00	0.00			0.32	
	8	0.06	1.38	6.87			0.00
	10	0.00	0.00				
	13	0.00	0.04				
	17	0.01	1.40				
	20	0.00	0.39				
	22	0.00	0.23				
	29	0.00	0.02				
	36	0.00	0.40			0.68	
	38	0.06	0.19			1.19	
	41	0.53	0.22			1.39	
	43	0.44	0.59			2.52	
	45	1.56	0.25			3.24	
	48	0.99	0.18			2.26	
	51	1.08	0.08			1.74	
	55	0.80	0.17			1.51	
	59	0.56	0.10			0.00	
	62	0.53	0.07			1.41	
	64	0.47	0.13			2.15	
	69	0.46	0.08			1.57	
	79	0.00	0.00			2.01	
As(V)+buffer+MnO2	1	0.34	18.23		0.36		
	2			6.78			
	3	0.00	5.52		0.54		
	6	0.06	2.09		0.95		
	8	0.00	1.33	6.75			
	10	0.02	0.82				
	10	0.04	0.77				
	13	0.01					
	13	0.02	0.56				

	38	0.00	0.00		1.84		
	41	0.00	0.00		2.05		
	43	0.00	0.00		2.10		
	45	0.00	0.00		2.17		
	48	0.00	0.00		2.25		
	51	0.00	0.00		2.31		
	55	0.00	0.00		2.43		
	59	0.00	0.00		2.48		
	62	0.00	0.00		2.51		
	64	0.00	0.00		2.56		
	69	0.00	0.00		2.59		
	79	0.00	0.00		2.69		
As(V)+buffer+Fe2O3	1	0.16	7.28			0.48	
	3	0.01	0.68			1.06	
	6	2.05	0.18			0.05	
	8	0.08	0.00	6.8			
	10	0.35	0.58				
	13	0.25	0.08				
	17	0.83	0.60				
	36	0.00	0.00			0.26	
	38	0.00	0.00			0.52	
	41	0.00	0.00			0.25	
	43	0.00	0.00			1.25	
	45	0.00	0.00			0.74	
	48	0.00	0.00			0.48	
	51	0.07	1.89			0.00	
	55	0.00	0.00			0.00	
	59	0.00	0.00			0.00	
	62	0.00	0.00			0.00	
	64	0.00	0.00			0.52	
	69	0.00	0.00			0.21	
	79	0.00	0.00			0.49	
buffer+Na2S+MnO2	1				0.45		
	2			6.81			0.00
	3				0.59		
	6				1.00		
	8			6.81			0.00

	34			1.11		
	36			1.52		
	38			2.23		
	41			2.38		
	43			2.28		
	45			2.25		
	48			2.12		
	51			2.05		
	55			1.94		
	59			1.89		
	62			1.87		
	64			1.82		
	69			1.66		
	79			1.67		
Tris+H2S+Fe2O3	1				0.82	
	2		6.82		0.00	0.02
	3				0.72	
	6				0.13	
	8		6.82			0.00
	36				4.83	
	38				7.50	
	41				11.19	
	43				9.72	
	45				11.84	
	48				7.79	
	51				8.56	
	55				8.15	
	59				5.37	
	62				6.12	
	64				7.02	
	69				5.71	
	70				6 9 9	

Tab. A28 As-S complex determination measured by Dirk Wallschläger

Sample ID	As(III) [ppb]	As(V) [ppb]	As-S [ppb]	sum all species [ppb]
As H2S	0	5.800	0	5.800
As Na2S	0	6.300	64	6.364
AsFe H2S	2	50	1	54
AsFe Na2S	3	39	3	46
MnOAs H2S	2	11	0	13
MnOAs Na2S	1	3	0	4

# 11.3 Sardinia

### 11.3.1 Baccu Locci Stream

# Tab. A29 Supernatant

sample	Cl Conc [mg/l]	NO3 Conc [mg/l]	SO4 Conc [mg/l]	As(III) [µg/I]	As(V) [µg/l]	Fe [mg/l]	Mn [mg/l]	Ca [mg/l]	Mg [mg/l]	K [mg/l]	Na [mg/l]	Zn [mg/l]	pH-Wert
1	109.53	4.65	371.19	3.73	327.27	2.78	1.57	160.4	58	2.3	51.3	0.66	7.53
2	41.18	10.38	369.07	2.01	182.99	n.n.	n.n.	136.3	27.5	2.06	25.1	1.12	7.55
3	39.81	7.35	263.87	1.45	112.55	n.n.	n.n.	89.7	31.3	3.86	24.7	0.68	7.36
4	44.42	4.73	291.92	6.92	390.08	n.n.	0.13	114.1	29.8	2.35	27.9	4.4	7.19
5	43.77	2.10	319.54	10.73	591.27	n.n.	n.n.	111.2	21.2	2.09	34.4	0.16	7.86
6	55.76	5.51	347.70	13.20	1057.80	n.n.	n.n.	110.4	33	2.03	35	0.15	7.8
7	62.17	2.89	300.91	15.29	1029.71	n.n.	n.n.	117.8	31.7	2.22	41.3	0.32	8.18
8	69.20	4.42	245.99	11.27	1034.73	n.n.	n.n.	114.9	27.3	1.93	39.1	0.18	7.95
9	68.40	6.61	177.55	10.02	562.98	n.n.	n.n.	74.7	24.1	2.34	38.9	0.11	7.8
10	66.97	12.21	159.62	9.04	535.96	0.1	n.n.	76.9	22.5	2.49	36.7	0.1	7.73
11	78.92	4.54	205.13	11.67	724.33	0.15	0.06	102.7	31.3	5.29	46.5	0.13	7.94
12	81.84	8.49	184.15	7.42	585.08	0.1	n.n.	83.1	31.1	2.15	46.8	0.08	7.84

### Tab. A30 Total Content

sample	As [mg/kg]	Mn [mg/kg]	Fe %									
1	1127	428	2.20									
2	2877	702	3.61									
3	2626	643	3.15									
4	3442	639	3.06									
5	3768	676	3.33									
6	3316	526	3.02									
7	2623	534	2.82									
8	1988	486	2.65									
9	2030	474	2.66									
10	2074	563	2.61									
11	1613	534	2.61									
12	855	453	2.36									
а	20042	459	3.67									
b	24535	163	4.88									
		AsV				AsIII			Fe			Mn
----	----------------	----------------	----------------	----------------	----------------	----------------	----------------	----------------	---------------	---------------	---------------	---------------
	H2O [ma/ka]	HCI [ma/ka]	PO4 [ma/ka]	NH3 [ma/ka]	H2O [ma/ka]	HCI [ma/ka]	PO4 [ma/ka]	NH3 [ma/ka]	HCI [ma/L]	PO4 [ma/L]	NH3 [ma/L]	NH3 [ma/L]
1	3.27	442	220	23	0.05	15.2	7.0	0.00	235	11.5	5.8	14.7
2	2.53	943	150	46	0.06	23.7	2.4	0.51	447	2.6	14.1	32.0
3	1.33	1052	119	44	0.03	14.7	3.1	0.00	438	1.7	12.2	23.7
4	4.23	1615	215	78	0.02	47.3	5.6	0.33	435	2.4	18.3	25.0
5	6.04	1677	284	67	0.08	48.3	5.6	0.43	526	4.1	17.2	27.2
6	7.51	1419	223	44	0.14	32.2	5.0	0.00	656	2.7	12.9	15.4
7	6.53	1008	199	53	0.13	21.8	5.2	0.00	551	3.6	10.7	19.0
8	5.59	1146	209	50	0.08	15.9	4.7	0.00	679	2.3	12.0	22.8
9	3.64	844	173	68	0.04	14.5	5.1	0.00	308	4.6	18.7	20.9
10	5.23	1145	176	87	0.06	25.1	4.5	0.00	398	4.7	23.9	15.9
11	5.20	747	165	93	0.08	12.1	3.5	0.00	779	4.9	26.5	23.9
12	4.15	581	128	69	0.07	17.8	3.2	0.00	205	3.2	24.1	24.8
а	1.22	14834	2365	1346	0.59	134.8	40.0	0.00	1370	15.3	429.0	0.5
b	1.48	9311	925	609	0.08	333.4	40.0	0.00	1780	16.4	131.0	0.3

Tab. A 31 24 Hour Elution

# Tab. A 32 Long Term Elution

sample	Α	Α	В	В	4	4	6	6
	As(III)	As(V)	As(III)	As(V)	As(III)	As(V)	As(III)	As(V)
	[mg/kg]							
days								
0	0.59	1.22	0.08	1.48	0.02	4.33		
1	0.19	1.54	0.02	1.49	0.04	4.16		
2			0.02	1.80	0.04	3.51	0.09	7.69
3	0.22	0.75	0.02	0.66	0.05	3.07	0.06	5.30
4	0.19	0.78	0.01	0.66	0.04	3.19	0.05	4.08
5	0.11	0.75	0.01	0.55	0.03	1.93	0.05	4.34
7	0.24	0.58	0.01	0.67	0.03	3.02	0.03	2.65
8	0.12	0.79	0.01	0.49	0.02	1.94	0.04	4.04
9	0.11	0.78	0.01	0.51	0.02	1.80	0.02	2.70
10	0.07	0.89	0.01	0.48	0.01	2.00	0.02	2.58
11	0.03	0.72	0.01	0.41	0.01	1.45	0.02	2.56
13	0.02	0.96	0.01	0.63	0.02	1.75	0.02	2.20
14	0.01	0.88	0.01	0.52	0.01	1.57	0.02	3.40
15	0.04	1.12	0.01	0.51	0.01	1.98	0.02	2.64
16	0.01	0.87	0.01	0.41	0.01	1.63	0.03	3.37
17	0.01	0.89	0.01	0.46	0.01	1.46	0.02	3.05
18	0.02	0.91	0.00	0.35	0.01	1.28	0.02	3.08
20	0.01	0.97	0.00	0.39	0.01	1.46	0.02	2.21
21	0.01	1.02	0.02	0.40	0.00	2.23	0.02	3.08
22	0.02	0.83	0.03	0.41	0.02	1.33	0.01	3.61
23	0.01	0.80	0.04	0.42	0.01	1.18	0.03	2.75
24	0.01	0.81	0.05	0.43	0.01	1.07	0.03	2.47
25	0.01	0.83	0.06	0.44	0.01	1.07	0.03	2.26
26	0.01	0.98	0.07	0.45	0.01	1.19	0.02	2.34
27	0.01	0.94	0.00	0.34	0.01	1.03	0.02	2.44
28	0.01	0.99	0.01	0.36	0.01	1.05	0.02	2.12
29	0.01	1.15	0.00	0.32	0.01	0.99	0.02	2.55
30	0.01	1.06	0.01	0.32	0.01	0.90	0.02	2.50
31	0.01	1.04	0.01	0.31	0.01	0.93	0.02	2.52
32	0.01	1.08			0.01	0.87	0.02	2.73
33	0.01	1.05	0.01	0.26	0.01	0.91		
34	0.01	1.01	0.01	0.30	0.01	0.80	0.03	2.52
35	0.01		0.01	0.30	0.01	0.71	0.02	2.42
36	0.01	1.03	0.01	0.31	0.02	1.05	0.02	2.10

## 11.3.2 Palude Sa Masa

Tab. A33 Supernatant

sample	Fe [mg/l]	Mn [mg/l]	pH-Wert
13	0.00	1.44	7.83
14	0.62	0.74	7.64
12	0.00	2.51	7.63
15	105.80	65.55	3.80

sample	As(III) [µg/I]	As(V)[µg/I]
13	0.12	0.62
14	2.75	3.56
12	3.61	3.61
15	0.15	0.00

# Tab. A34 Total content

sample	Ca [mg/kg]	K [mg/kg]	Mg [mg/kg]	Mn [mg/kg]	Fe [mg/kg]	Pb [mg/kg]	SO4 [mg/kg]	Zn [mg/kg]	Ca [%]	K [%]	Mg [%]	Fe [%]	water fraction [%]
1	40301	14498	13346	1196	32071	11133	58146	104120	4.03	1.45	1.33	3.21	30.54
2	109348	11063	21406	1618	31209	19534	37626	27710	10.93	1.11	2.14	3.12	20.95
3	161477	4158	23861	2607	15135	10461	18590	78513	16.15	0.42	2.39	1.51	10.31
4	75344	3372	51163	1889	77206	9671	38601	51290	7.53	0.34	5.12	7.72	13.31
5	133581	1678	42647	1622	13612	7700	20830	28300	13.36	0.17	4.26	1.36	9.00
6	69454	18420	10150	765	30771	122	1748	295	6.95	1.84	1.02	3.08	12.43
7	144060	2487	48158	2217	50910	9409	41510	33279	14.41	0.25	4.82	5.09	8.94
8	145805	2825	43503	1256	37114	5850	22312	31649	14.58	0.28	4.35	3.71	13.70
9	21056	22599	11136	1422	47332	286	2101	981	2.11	2.26	1.11	4.73	14.40
10	21617	22342	11573	1033	47731	415	3183	2413	2.16	2.23	1.16	4.77	13.05
11	49746	15593	5868	276	11126	2380	3780	6143	4.97	1.56	0.59	1.11	4.76
12	84134	6443	37753	522	14050	1299	2472	19030	8.41	0.64	3.78	1.40	9.12
13	80568	12156	25314	1538	46658	12195	30637	23771	8.06	1.22	2.53	4.67	5.86
14	21143	18121	2725	211	9857	710	12104	634	1.31	1.81	0.27	0.99	8.45
15	27651	22117	3280	242	37845	709	30751	1187	0.36	2.21	0.33	3.78	13.73
16	64156	16923	29605	1060	26642	2761	7117	2149	6.42	1.69	2.96	2.66	7.24
17	62933	14068	11970	15674	36862	2169	30999	8990	6.29	1.41	1.20	3.69	27.78

## Tab. A35 24 Hour Elution Test

	H2O		HCI		PO4		NH3		
	As(III) [mg/kg]	As(V) [mg/kg]	As(III) [mg/kg]	As(V) [mg/kg]	As(III) [mg/kg]	As(V) [mg/kg]	As(III) [mg/kg]	As(V) [mg/kg]	Mn in NH3 elution
1	0.0000	0.0067	0.05	2.00	4.63	18.41	1.12	0.79	0.15
2	0.0000	0.0151	11.89	0.42	1.26	11.27	0.14	0.26	0.13
3	0.0000	0.0000	0.06	1.93	0.45	15.44	0.12	0.33	1.01
4	0.0000	0.0094	0.77	11.93	0.49	9.56	0.02	0.38	0.14
5	0.0000	0.0000			0.05	1.47	0.00	0.05	0.31
6			0.01	1.25	0.04	3.68	0.00	0.13	
7	0.0000	0.0163	0.02	1.50	0.03	2.00	0.00	0.01	0.53
8	0.0000	0.0003	0.08	0.81	0.05	2.05	0.00	0.08	0.19
9	0.0000	0.0437	0.00	2.48	0.02	6.09	0.00	0.08	0.87
10	0.0000	0.0388	0.00	2.38	0.12	7.43	0.00	0.13	
11	0.0000	0.0162	1.21	0.09	0.07	1.07	0.01	0.21	0.03
12	0.0260	0.0739	1.62	0.38	0.07	1.04	0.15	0.21	0.02
13	0.0007	0.0029	2.98	0.09	0.22	2.53	0.29	0.14	0.21
14	0.0248	0.0701	0.02	0.00	0.01	0.52	0.50	0.30	0.05
15	0.0022	0.0062	2.76	0.00	0.13	5.58	0.02	0.01	0.10
16	0.0000	0.0055	0.05	0.88	0.02	0.96	0.00	0.09	0.42
17	0.0113	0.0000	0.36	0.00	0.10	0.56	0.12	0.14	4.56

#### Erklärung:

Hiermit erkläre ich, Rouven Höhn (geb. 24.09.73), an Eides statt, dass ich die vorgelegte Dissertation selbst verfasst und mich keiner anderen als der von mir ausdrücklich bezeichneten Quellen und Hilfen bedient habe.

Ebenfalls erkläre ich an Eides statt, dass ich an keiner anderen Stelle ein Prüfungsverfahren beantragt habe, dass ich die Dissertation nicht in dieser oder anderer Form bereits anderweitig als Prüfungsarbeit verwendet habe und dass ich sie an keiner anderen Fakultät als Dissertation vorgelegt habe.

Heidelberg, den 30.05.2005