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Excess radiocarbon constraints on air-sea gas exchange and the uptake of CO_2 by the oceans

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[1] We re-assess the constraints that estimates of the global ocean excess radiocarbon inventory (I^E) place on air-sea gas exchange. We find that the gas exchange scaling parameter a_q cannot be constrained by I^E alone. Non-negligible biases in different global wind speed data sets require a careful adaptation of a_q to the wind field chosen. Furthermore, a_q depends on the spatial and temporal resolution of the wind fields. We develop a new wind speed- and inventory-normalized gas exchange parameter a_q^N which takes into account these biases and which is easily adaptable to any new estimate of I^E. Our study yields an average estimate of a_q of 0.32 ± 0.05 for monthly mean winds, lower than the previous estimate (0.39) from Wanninkhof (1992). We calculate a global annual average piston velocity for CO_2 of 16.7 ± 2.9 cm/hr and a gross CO_2 flux between atmosphere and ocean of 73 ± 10 PgC/yr, significantly lower than results from previous studies. **Citation:** Naegler, T., P. Ciais, K. B. Rodgers, and I. Levin (2006), Excess radiocarbon constraints on air-sea gas exchange and the uptake of CO_2 by the oceans, *Geophys. Res. Lett.*, *33*, L11802, doi:10.1029/2005GL025408.

1. Introduction

[2] Individual ocean regions are temporally varying sources or sinks of atmospheric CO_2, depending on factors such as sea surface temperature and salinity, biological activity, or the upwelling of CO_2-rich waters to the surface. Full thermodynamic equilibrium is thus never reached for CO_2 at the air-sea interface. This yields a permanent flux (F) of CO_2 to and from the ocean, which is proportional to the CO_2 partial pressure difference between the sea surface and the atmosphere (ΔpCO_2), the temperature-dependent solubility (L), and the transfer- (or piston-) velocity (k). Thus F can be expressed as

$$F(\vec{x},t) = k(\vec{x},t) \cdot L(\vec{x},t) \cdot \Delta pCO_2(\vec{x},t) \quad (1)$$

[3] A global gridded climatology of ΔpCO_2 has been derived using in situ measurements [*Takahashi et al.*, 2002], albeit with some gaps, but there remains a fundamental uncertainty in the determination of the piston velocity k when calculating the air-sea flux of CO_2.

[4] Numerous process studies under both controlled conditions (e.g., wind tunnels) and real-world conditions (e.g., tracer releases at sea) have measured the dependency of k as a function of wind speed u, but no unified theory exists yet. Commonly accepted relationships for k(u) assume in sections linear [*Liss and Merlivat*, 1986], quadratic [*Wanninkhof*, 1992; *Nightingale et al.*, 2000] or cubic functions [*Wanninkhof and McGillis*, 1999] of wind speed. The most widely used gas exchange parameterization is the quadratic k(u) relationship from *Wanninkhof* [1992]:

$$k(\vec{x},t) = a_q \cdot u(\vec{x},t)^2 \cdot (Sc(\vec{x},t)/660)^{-0.5} \quad (2)$$

where Sc refers to the Schmidt number. a_q denotes a global gas exchange scaling parameter, which cannot be measured directly, but has to be calibrated against observations, e.g., the observed ocean excess radiocarbon inventory (I^E) [*Broecker et al.*, 1985; *Wanninkhof*, 1992] (see also section 2). "Excess radiocarbon" refers to human-induced changes in the radiocarbon (^{14}C) inventory of the main carbon reservoirs. These changes are caused by the production of ^{14}C in atmospheric nuclear bomb tests and the release of ^{14}C by the nuclear industry. As a second, yet smaller effect, the combustion of ^{14}C-free fossil carbon dilutes the atmospheric ^{14}C-to-C ratio of CO_2 (the so-called "Suess effect"), causing a ^{14}C flux from ocean (and biosphere) to the atmosphere.

[5] Ocean ^{14}C observations performed during the Geochemical Ocean Section Study (GEOSECS) and the World Ocean Circulation Experiment (WOCE) ocean surveys in the 1970s and 1990s respectively, allowed estimations of I^E for these two periods. From their I^E estimate based on the GEOSECS data ($289 \cdot 10^{26}$ atoms ^{14}C), *Broecker et al.* [1985] inferred a global mean piston velocity of $\langle k \rangle$ = 21.9 ± 3.3 cm/hr. *Wanninkhof* [1992] used this estimate of $\langle k \rangle$ to constrain the parameter a_q for a quadratic k(u) relationship (0.39 cm/hr/(m/s)^2), which has since then been widely used as the standard gas exchange parameterization. (Note that we omit the unit cm/hr/(m/s)^2 for a_q in the remaining text.) However, two more recent estimates of I^E for GEOSECS [*Peacock*, 2004] and WOCE [*Key et al.*, 2004] which are based on improved methods to separate the excess ^{14}C signal from the natural background suggest lower I^E and consequently smaller $\langle k \rangle$ and a_q values. *Naegler and Levin* [2006] demonstrated with the help of a simple global ^{14}C cycle model that these values for I^E from *Peacock* [2004] and [*Key et al.*, 2004] (corrected as described below) are consistent with our current knowledge of the global excess ^{14}C budget, in contrast to the older GEOSECS estimate from *Broecker et al.* [1985], which has served as

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Table 1. Characteristics of the Five Wind Speed Climatologies Used in This Study: Global Annual Mean Wind Speeds Over the Ice-Free Ocean $\langle u \rangle$ and $\langle u^2 \rangle$, Resulting Gas Exchange Parameter a_q for Different Temporal and Spatial Resolutions, the Product $a_q \cdot \langle u^2 \rangle$, and the Global Annual Mean Piston Velocity k (Calculated According to Equation (2))[a]

Parameter	Unit	NCEP	ECMWF	SSMI	QSCAT	ERS12	Mean ± σ	σ/Mean
$\langle u \rangle$	m/s	6.6	7.0	7.8	7.9	7.3	7.3 ± 0.6	0.08
$\langle u^2 \rangle$	$(m/s)^2$	46.9	53.1	65.0	66.4	57.1	57.7 ± 8.2	0.14
a_q (monthly winds, 5° ×4° grid)	$(cm/hr)/(m/s)^2$	0.40	0.35	0.29	0.29	0.34	0.33 ± 0.05	0.14
a_q (monthly winds, 1° ×1° grid)	$(cm/hr)/(m/s)^2$	0.39	0.34	0.28	0.27	0.32	0.32 ± 0.05	0.14
a_q (daily winds, 1° × 1° grid)	$(cm/hr)/(m/s)^2$	0.38	0.33	-	-	-	-	-
$a_q \cdot \langle u^2 \rangle$	cm/hr	18.1	17.9	18.2	18.1	18.4	18.1 ± 0.2	0.01
$\langle k \rangle$	cm/hr	16.6	16.2	16.4	17.3	16.9	16.7 ± 0.4	0.02

[a]Note that for the satellite wind speed data sets (SSM/I, QSCAT and ERS1/2), no daily resolution with full global coverage is available. $\langle u \rangle$, $\langle u^2 \rangle$, $a_q \cdot \langle u^2 \rangle$ and $\langle k \rangle$ are calculated from monthly winds on a 1° × 1° grid.

a reference value for the calibration of the k(u) relationship [*Wanninkhof*, 1992]. Consequently, these findings require a revision of the excess ^{14}C constraints on air-sea gas exchange.

[6] Additional attention has to be paid to biases in available wind speed products which significantly affect the piston velocity k and consequently the fluxes F. *Wanninkhof* [1992] and *Wanninkhof et al.* [2002] already noticed the dependency of the k(u) parameterization on the temporal resolution of the wind fields. Here we additionally focus on biases introduced by the choice of a particular wind speed data set and a particular temporal and spatial resolution and provide a new approach to calculate unbiased values of k.

2. Methods

[7] The net excess ^{14}C flux across the ocean surface, F^E_{14CO2}, depends on the same piston velocity k(u) and solubility L as the flux of CO_2 (equation (1)), but is driven by the effective (i.e., fractionation-corrected) excess $^{14}CO_2$ partial pressure difference $\Delta p^{14}CO_2^E$ between atmosphere and ocean. Following *Wanninkhof* [1992], we assume a quadratic k(u) relationship (equation (2)). Note that our approach is, however, in principle applicable for any k(u) relationship which depends on a single scaling parameter. (For the sake of simplicity, we omit the variability in space and time ($\vec{x}$, t) from our notation in what follows.) $\Delta p^{14}CO_2^E$ is calculated as the difference between the effective total $^{14}CO_2$ partial pressure difference $\Delta p^{14}CO_2$ and the effective prebomb (i.e., pre-1945) $^{14}CO_2$ partial pressure difference $\Delta p^{14}CO_2^{pre}$:

$$\Delta p^{14}CO_2^E = \Delta p^{14}CO_2 - \Delta p^{14}CO_2^{pre} \quad \text{with} \tag{3}$$

$$\Delta p^{14}CO_2 = R_A^{14} \cdot pCO_2^A \cdot \alpha_{14}^{AO} - R_O^{14} \cdot pCO_2^O \cdot \alpha_{14}^{OA} \tag{4}$$

where R_A^{14} and R_O^{14} are the ^{14}C/C isotope ratios in the atmosphere and the surface ocean, respectively. pCO_2^A and pCO_2^O are the atmospheric and oceanic CO_2 partial pressure histories, α_{14}^{AO} (=0.9980^2) and α_{14}^{OA} (=0.9897^2) are the kinetic fractionation factors for the transfer of $^{14}CO_2$ from atmosphere to ocean and vice versa.

[8] As the total excess ^{14}C inventory in the ocean is equal to the (temporally and spatially) integrated flux of excess ^{14}C into the ocean, the gas exchange parameter a_q, crucial in the parameterization for any air-sea gas exchange relationship (equation (2)), can be calculated from the observed excess ^{14}C inventory I^E in the ocean at a given reference date t_1:

$$a_q = \frac{I^E(t_1)}{\int_{t_0}^{t_1} \int_{S_{oce}} u^2 \left(\frac{Sc}{660}\right)^{-0.5} \cdot L \cdot \Delta p^{14}CO_2^E \, dS \, dt} \tag{5}$$

where t_0 indicates pre-bomb times (pre-1945) and S_{oce} is the surface of the ice-free ocean. R_A^{14} and R_O^{14} are calculated from observation-based reconstructions of the zonal mean Δ^{14}C history in the atmosphere and the sea surface from *Hesshaimer* [1997]. The atmospheric pCO_2 history has been reconstructed using available records of the atmospheric CO_2 mixing ratio [*Etheridge et al.*, 1998; *Keeling and Whorf*, 2004; GlobalView (CDIAC, Oak Ridge, Tennessee; anonymous ftp to ftp.cmdl.noaa.gov)]. The global distribution of pCO_2^O for 1995 is taken from *Takahashi et al.* [2002]. The relative increase of pCO_2^O between 1945 and 1995 has been estimated with the IPSL Ocean General Circulation Model (J. Orr and J.-C. Dutay, personal communication, 2004). To take into account the uncertainties of global wind speed products and biases due to spatial and temporal averaging, we used five different climatological wind speed data sets (Table 1) from satellite observations (ERS1/2 [*Bentamy et al.*, 1998], QSCAT [*Lungu*, 2001], SSM/I [*Boutin and Etcheto*, 1996]) or reanalysis products from numerical weather prediction models (ECMWF [*Gibson et al.*, 1997] and NCEP [*Kalnay et al.*, 1996]) and tested different temporal and spatial resolutions of these wind fields.

[9] Our study is based on the new I^E estimates from *Peacock* [2004] for GEOSECS (241 · 10^{26} atoms ^{14}C ± 25%, multitracer correlation method) and from *Key et al.* [2004] for WOCE (313 · 10^{26} atoms ^{14}C ± 15%). Neither the *Peacock* [2004] nor the *Key et al.* [2004] inventory estimates used data with fully representative global coverage. We corrected the original inventory estimates for these missing ocean areas by estimating the relative contribution of the missing areas to I^E, using the OPA Ocean General Circulation Model [*Rodgers et al.*, 2004]. With this correction applied, I^E for the mid-1970s from *Peacock* [2004] increases to 245 · 10^{26} atoms ^{14}C. Furthermore, the *Key et al.* [2004] study uses North Atlantic samples from the mid-1980s, whereas the rest of the world ocean was sampled in the 1990s (with a mid-point in 1995). To correct for this

temporal bias in the WOCE data for the North Atlantic, we estimated the increase of I^E in the North Atlantic between 1985 and 1995 again with the OPA OGCM. The two corrections of the *Key et al.* [2004] inventory estimate result in a total I^E in the mid-1990s of $355 \cdot 10^{26}$ atoms ^{14}C. Due to the consistency of these I^E values with a totally independent ^{14}C modelling study [*Naegler and Levin*, 2006], we assume that the real uncertainties of I^E are smaller than reported by *Peacock* [2004] and *Key et al.* [2004], namely on the order of 10% for both inventories.

3. Results and Discussion

[10] Table 1 summarizes the results for the gas exchange parameter a_q calculated for the different temporally and spatially resolved wind fields. Clearly a_q cannot be unambiguously constrained by I^E alone, but depends on a number of factors.

[11] First, due to the non-linear k(u) relationship and the co-variability of $\Delta p^{14}CO_2$, solubility L, Schmidt-Number Sc and wind speed u, a_q depends on the temporal and spatial resolution of the study, with higher resolution implying lower values for a_q. This effect is more pronounced for interannually varying winds (not shown) than for climatological wind fields (as used in Table 1).

[12] Second, and even more importantly, a_q depends on the wind field used. We found significant differences between the five different wind speed data sets, leading to a significant uncertainty in a_q: The five wind speed data sets used here show a standard deviation of 14% in the global annual mean squared wind speed over the ice-free ocean $\langle u^2 \rangle$ (Table 1), which translates into a spread of similar magnitude in $a_q \pm = 0.32 \pm 0.05$, calculated on a $1° \times 1°$ grid and for monthly average wind fields.

[13] However, in contrast to a_q, the product $a_q \langle u^2 \rangle$ depends only weakly on the wind fields used; it shows a spread of only about 1.0% around the mean for monthly $1° \times 1°$ fields (Table 1) and a spread of 2% for the $5° \times 4°$ resolution (not shown). Thus, the dependence of a_q on the wind speed data set (and the dependence of a_q on I^E) can be reduced greatly by defining a new wind-speed- and inventory-normalized gas exchange parameter a_q^N, namely

$$a_q^N \equiv \frac{a_q \cdot \langle u^2 \rangle}{I^E} \quad \Leftrightarrow \quad a_q = a_q^N \cdot \frac{I^E}{\langle u^2 \rangle} \tag{6}$$

[14] As for the product $a_q \cdot \langle u^2 \rangle$, but in contrast to a_q, the parameter a_q^N depends only weakly on the wind speed product and its resolution. Thus a_q^N allows us to balance biases in the piston velocity k introduced by biases in the wind fields. Table 2 summarizes our results for a_q^N.

[15] Equation (6) now allows us to calculate values of a_q for each wind speed data set whose global annual mean squared wind speed $\langle u^2 \rangle$ is known. Furthermore, I^E in equation (6) can be adapted to future improved estimates of this quantity. In this formulation, uncertainties of each individual estimate of a_q calculated according to equation (6) for a given wind speed data set originate from uncertainties in I^E of ± 10%, which translate into uncertainties in a_q of ± 0.03, and from uncertainties in a_q^N, which are estimated as follows:

Table 2. New Normalized Gas Exchange Parameter a_q^N (Defined in Equation (6)) for Different Spatial Resolutions and for Different I^E Targets[a]

Temporal Resolution	Spatial Resolution	GEOSECS (1975)	WOCE (1995)
Monthly	$5° \times 4°$	0.0760 ± 3%	0.0524 ± 3%
Monthly	$1° \times 1°$	0.0740 ± 1%	0.0511 ± 1%
Daily	$1° \times 1°$	0.0733 ± 1%	0.0506 ± 1%

[a]The values given are averages (± 1 standard deviation) over all five (two in the case of daily winds) wind speed data sets. Units are in (cm/hr)/10^{26} ^{14}C atoms. Note that the values shown here are calculated from climatological wind fields. However, a_q^N for the corresponding interannually varying wind fields is virtually identical.

[16] The uncertainty of a_q^N is dominated by the uncertainty of the integrated $\Delta p^{14}CO_2^E$, which again is dominated by the uncertainty of $p^{14}CO_2^O$. As revealed in a comparison of reconstructed sea surface Δ^{14}C with observations, the uncertainty in the reconstruction is less than 30‰ [*Naegler*, 2005], resulting in a 3% uncertainty in R_O^{14}. The uncertainty of the reconstructed pCO_2^O is difficult to assess, but probably not larger than (globally) 10%. This results in an upper limit of the uncertainty of a_q^N of 15%, resulting in an overall uncertainty of a_q due to uncertainties in I^E and a_q^N of ± 0.06 for a specific wind speed data set.

[17] Our revised set of estimates for a_q for monthly mean winds is systematically lower (up to 40%) than the value from *Wanninkhof* [1992] for long-term averaged winds (a_q = 0.39, averaging period ≥1 month). This is primarily because different wind speed estimates and a lower I^E value were employed in our study, but also because of the low spatial and temporal resolution of *Wanninkhof* [1992] as well as the resulting neglect of correlations between L, Sc, u, and $\Delta^{14}pCO_2$. For daily winds that are available only from Numerical Weather Prediction models, we obtain a_q = 0.38 (NCEP) and a_q = 0.33 (ECMWF) (Table 1). Our values are both higher than the value given by *Wanninkhof* [1992] for short-term winds (a_q = 0.31), although we would expect lower values due to the lower I^E used in our study. When estimating a_q for short term winds, *Wanninkhof* [1992] assumed that the spectrum of short-term wind speeds u is characterized by a Rayleigh distribution. If this is the case, the ratio $R = \langle u^2 \rangle / \langle u \rangle^2$ has a characteristic constant value of 1.25, which allows one to extrapolate a_q for short-term wind speeds from estimates of a_q for long-term averages. However, as already stated by *Wanninkhof et al.* [2002], the assumption of a Rayleigh distribution does not hold over large areas of the oceans. Consequently, R is generally lower than 1.25 [*Wanninkhof et al.*, 2002], yielding a too low gas exchange parameter for short-term winds (a_q = 0.31 [*Wanninkhof*, 1992]) compared to a_q for long-term winds (0.39) and also lower values than our estimates of a_q for daily averaged winds.

[18] From our new estimates of a_q we calculate global average values for the piston velocity k of 16.7 ± 2.9 cm/hr. The uncertainty estimate given here comprises uncertainties in the wind fields, in $\Delta p^{14}CO_2$ and in I^E. Mainly due to the lower ocean excess ^{14}C inventory used, our value is lower than the previous estimates of 21.9 ± 3.3 cm/hr from *Broecker et al.* [1985] and used by *Wanninkhof* [1992].

[19] Our k values (short-term winds) are generally higher than the field-based k(u) relationship from *Nightingale et al.*

[2000], which is adequate for field studies on a local scale. This is probably due to the different scales of the studies, but also due to the neglect of chemical enhancement in our study. However, we believe that results from the excess ^{14}C method and field studies will converge with increasing resolution of global wind fields.

[20] Although our method does not assume a Rayleigh distribution of wind speeds, it implicitly assumes that the same wind speed distribution is valid everywhere over the ocean (i.e., $R = \langle u^2 \rangle / \langle u \rangle^2$ is globally constant). Regional deviations from this assumption cause regional biases in k [*Wanninkhof et al.*, 2002], which increase with decreasing resolution of the wind fields. Our method therefore tends to underestimate k in high latitudes and overestimate k in low latitudes. However, for monthly mean winds, these biases probably do not exceed 5% for the zonal average of k. This was tested using a method similar to that used by *Wanninkhof et al.* [2002].

[21] When applied to ocean pCO_2 data [*Takahashi et al.*, 2002] and smoothed atmospheric CO_2 observations (GlobalView, see above), our new numbers yield a net ocean CO_2 uptake rate of 1.57 ± 0.30 PgC/yr for 1995, lower than previous estimates based on the same method from *Takahashi et al.* [2002] (1.64 PgC/yr, corrected flux estimates), but still consistent within the uncertainties. At first glance, the small difference between these two estimates is surprising, as *Takahashi et al.* [2002] used the high $a_q = 0.39$ from *Wanninkhof* [1992] (and NCEP winds) for their calculations. However, NCEP winds have low $\langle u^2 \rangle$ and therefore require a high a_q (Table 1). Thus our a_q value for NCEP winds is, by chance, close to the value from *Wanninkhof* [1992], resulting in similar values for the net ocean CO_2 flux in our study and in that by *Takahashi et al.* [2002]. In contrast, our estimate of the gross CO_2 exchange between the atmosphere and the ocean (73 ± 10 PgC/yr) is ≈20% lower than previous estimates [*Broecker et al.*, 1985] of 88 PgC/yr.

4. Conclusions

[22] The observed excess ^{14}C inventory in the ocean (I^E) provides important constraints on air-sea gas exchange. However, uncertainties in the global wind fields and the dependence of the "optimal" gas exchange parameterization on the spatial and temporal resolution of the study complicate the simulation of air-sea fluxes. Consequently, in any gas exchange simulation, the gas exchange parameter a_q has to be carefully chosen depending on the wind speed data set used (and its resolution) in order to calculate an unbiased piston velocity. In our study, we introduce a new wind-speed- and inventory-normalized gas exchange parameter a_q^N. Once the value of a_q^N is tabulated for a given spatial and temporal resolution, the gas exchange parameter a_q can easily be adjusted to any wind speed data set and any estimate of the ocean excess ^{14}C inventory to allow the calculation of an unbiased global mean piston velocity. As we rely on recent I^E estimates [*Peacock*, 2004; *Key et al.*, 2004] which are lower than the values previously used to constrain air-sea gas exchange [*Broecker et al.*, 1985], our study yields significantly lower values (20–40%) for the gas exchange parameter a_q for long-term averaged winds, the global annual mean piston velocity k and the gross atmosphere-ocean CO_2 exchange than previous studies [*Broecker et al.*, 1985; *Wanninkhof*, 1992].

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