

Atmospheric deposition of organic and black carbon to the global oceans

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ABSTRACT

Atmospheric deposition of total organic carbon (OC) and black carbon (BC) is lacking or not fully accounted in most current models of the global carbon cycling, specially those fluxes related to gas phase OC. Here, we develop and apply a methodology to estimate wet and dry deposition of total OC to the oceans, based on monthly satellite measurements of aerosol size distributions, wind speed, etc., and estimates of deposition for aerosols and organic compounds. The parameterization of dry deposition velocities account for the dependence of turbulent transport with aerosol diameter, wind speed and the formation of marine aerosol, etc. Gravitational settling is estimated as a function of wet particle diameter, thus including hygroscopic growth due to ambient humidity. Global dry deposition of aerosol OC is estimated to be 11 Tg C y⁻¹ and wet deposition of particle and gaseous OC are estimated as 47 and 187 Tg C y⁻¹, respectively. Due to their pulsing variability, wet deposition fluxes can be important locally and as a temporal source of OC to surface waters. Dry and wet deposition of black carbon to the global ocean are estimated to be 2 and 10 Tg C yr⁻¹, respectively, with higher fluxes in the northern hemisphere and for inter-tropical regions. Finally, considerations on the potential magnitude of the hitherto neglected gross air–sea diffusive exchange fluxes of OC are discussed. Even though the magnitude and direction of these cannot be constrained here, evidence of its important role is given. This study, thus, shows that there is an important spatial and temporal variability in atmosphere–ocean exchanges of OC and BC at different scales, and calls for the need for further research on the important role that these exchanges play in the global carbon cycle.

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1. Introduction

The global carbon budget is a key node of the functioning of the Earth System that affects climate and its change, the global transport of pollutants, and perturbations of biogeochemical cycles (Fasham, 2003; Jurado et al., 2004; Sarmiento et al., 2004; Ramanathan and Garmichael, 2008). However, current regional and global models of the carbon cycle do not account for comprehensible

atmosphere–ocean exchanges of organic carbon. This is not due to the negligible magnitude of these atmospheric deposition processes, but, rather, due to a current paucity of measurements and quantitative knowledge. Nevertheless, the atmosphere is known to play a key role in several aspects of global cycling of organic compounds. The atmosphere is an efficient chemical reactor, responsible for major oxidative losses of organic compounds due to OH and O₃ radicals (Lohmann et al., 2006). Furthermore, since atmospheric transport is fast in comparison to oceanic transport, it is an efficient medium for redistributing chemicals at regional and global scale.

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Currently, the ocean is assumed to be a sink of atmospheric carbon as discerned from CO₂ measurements only, accounting for 30–40% of the anthropogenic perturbation in the carbon cycle (see Houghton, 2003 for example). The continents are also thought to be a sink of the anthropogenic emissions of carbon but there are major uncertainties in the regional variability of their source/sink magnitude (Houghton, 2003). As far organic carbon is concerned, continental ecosystems can be a source of reduced biogenic carbon to the atmosphere (Fuentes et al., 2000), in addition to anthropogenic sources (Schauer et al., 2002), which will in part undergo long range atmospheric transport.

The exchange of carbon in its oxidized form (carbon dioxide) has received major attention by the scientific community, yielding hundreds of thousands of measurements of CO₂ disequilibrium between surface seawaters and the lower atmosphere, which have allowed to estimate the global atmosphere–ocean exchange of CO₂ with uncertainties below 20% (Takahashi et al., 2002). This contrasts with the poor knowledge available on the significance and magnitude of atmosphere–ocean exchange of reduced carbon at regional and global scales, especially in the form of organic carbon. Indeed, estimates of air–sea exchanges are available only for a few organic compounds that are of concern for their toxicity (Jurado et al., 2004, 2005) or of interest for their biogeochemical significance (Simó and Dachs, 2002; Jacob et al., 2005).

Atmospheric deposition of organic carbon can occur by: (i) dry deposition of aerosol-bound organic compounds; (ii) diffusive air–water exchange of gas phase organic compounds; and (iii) wet deposition by precipitation scavenging of gas and aerosol phase organic compounds. Until recently, the only measurements available were those related to wet deposition (Willey et al., 2000; Raymond, 2005). Willey and coworkers reviewed measurements of non-purgeable dissolved organic carbon (DOC) in rain water and reached a global estimation of dissolved organic carbon wet deposition of 400 Tg C y⁻¹, of which 90 Tg C y⁻¹ correspond to inputs to the ocean. This figure is significantly higher than global estimates using models based on emission inventories. However, there is a considerable variability in DOC concentrations in rain-water (Raymond, 2005). Other available, indirect estimates of dry deposition of aerosol-bound OC are reported elsewhere (del Giorgio and Duarte, 2002; de Madron et al., 2003), and aerosol-bound organic carbon inputs have been recently reported for the NE Subtropical Atlantic (Duarte et al., 2006), but none of these account for depositional fluxes associated to gas phase organic compounds.

However, it is well known that dry aerosol deposition of organic compounds, although eventually important locally or regionally, represents a small fraction of the total atmospheric deposition of organic compounds (Gigliotti et al., 2002; Jurado et al., 2005). This is so widely accepted in the literature of organic volatile and semivolatile compounds, that this dry aerosol component of the flux is often assumed to be negligible and not quantified (Dachs et al., 2002; Simó and Dachs, 2002). Therefore, a reliable assessment of the global atmospheric OC deposition to the ocean must extend beyond the flux associated to aerosols to estimate the global wet and eventually diffusive

deposition of OC. Indeed, a recent study by Dachs et al. (2005) suggests that these diffusive exchanges are large in magnitude, very dynamic and important for the regional carbon budget of the north east Atlantic, and could also be significant in other regions and globally.

The objective of this study is to provide a comprehensive estimation of the global dry and wet deposition of organic compounds and black carbon (BC) to the ocean, thus filling a gap in current accounts of the global carbon cycle, and to point out some of the research needs for the coming decade. The estimates presented here are obtained by combining satellite measurements of a number of variables such as aerosol size distribution, temperature, etc, field measurements of aerosol concentrations and deposition, and parameterization of deposition velocities.

2. Model development and data sources

2.1. Parameterization of atmospheric deposition fluxes

Atmospheric depositional fluxes of aerosol OC, namely dry and wet deposition, have been parameterized using the methodology previously developed for individual organic compounds such as persistent organic pollutants (POPs) (Jurado et al., 2004, 2005). Fluxes of dry deposition of aerosol-bound organic and black carbon (F_{DD_OC} and F_{DD_BC} , mg m⁻² d⁻¹, respectively) are estimated as the product of organic/black carbon aerosol phase concentration (C_{P_OC} and C_{P_BC} , mg m⁻³, respectively) and the dry deposition velocity of aerosol (v_D , m d⁻¹):

$$\begin{aligned} F_{DD_OC} &= v_D C_{P_OC} \\ F_{DD_BC} &= v_D C_{P_BC} \end{aligned} \quad (1)$$

The total carbonaceous aerosol dry deposition flux (F_{DD}) is the sum of F_{DD_OC} and F_{DD_BC} . Wet deposition flux of particulate OC and BC ($F_{WD_P_OC}$ and $F_{WD_P_BC}$, mg m⁻² d⁻¹) is given by (Bidleman, 1988):

$$\begin{aligned} F_{WD_P_OC} &= p_0 W_p C_{P_OC} \\ F_{WD_P_BC} &= p_0 W_p C_{P_BC} \end{aligned} \quad (2)$$

where p_0 is the precipitation rate (m d⁻¹) and W_p the particle washout ratio (dimensionless).

Both v_D and p_0 have been derived from climatological monthly mean satellite products at a resolution of 1° × 1°, which cover the global oceans and allow to assess the spatial and temporal variability. The parameter v_D is dependent not only on atmospheric turbulence (as influenced by wind speed), but also on the aerosol size. A single lognormal distribution for the number concentration of aerosols has been assumed, the aerosol population has been grouped into size intervals, and widely used parameterizations applied (Slinn and Slinn, 1980; Williams, 1982) in order to obtain the deposition velocity of each aerosol size interval. A detailed explanation of the methodology is reported elsewhere (Jurado et al., 2004) and Fig. 1 shows the dependence of v_D on wind speed and aerosol size. Briefly, the parameterization used for v_D is an update of the Williams model (1992) that accounts for turbulent transport enhanced at high wind speed, humidity influence on particle size, thus influencing gravitational settling, etc. The

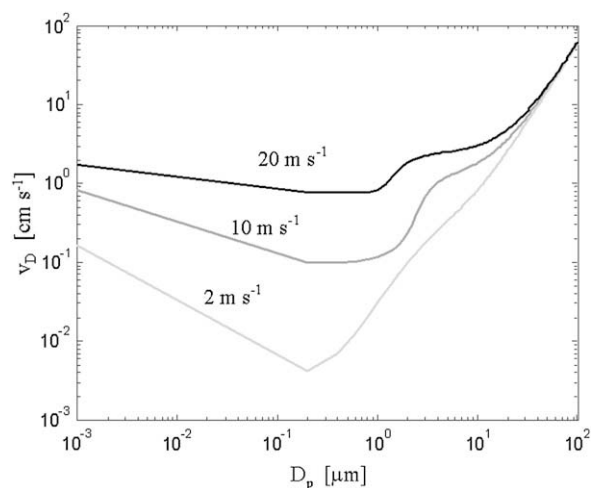


Fig. 1. Dry deposition velocity versus aerosol diameter for different wind speeds. Assumptions for this figure are sea surface temperature of 298K, relative humidity about 80%, and aerosol density of 2 g cm^{-3} .

detailed explanation of the parameterization leading to Fig. 1 and used in this work has been described elsewhere (Jurado et al., 2004, and its supplementary material).

Particle washout (W_p) has been reported to depend on aerosol size but contradictory trends can be found in the literature with highly variable (more than two orders of magnitude) W_p values (see Jurado et al., 2005 for a review). This can be explained by the complexity of the scavenging process. Some studies predict higher W_p for larger aerosols (Radke et al., 1980). Conversely, Mircea et al. (2000) estimated scavenging coefficients with almost no dependence on raindrop size distribution. Furthermore, there is no obvious dependence of the particle scavenging rate on the rainfall intensity (Nicholson et al., 1991). Provided the uncertainty constrains, W_p has been assumed constant and equal to $2 \cdot 10^5$. This value has been adopted in various modeling exercises for semivolatile organic compounds and fits well field measurements (Mackay et al., 1986; Jurado et al., 2005). This assumption does not result in an important uncertainty addition to the wet deposition flux for semivolatile compounds that are mainly found in the gas phase. A detailed discussion of this parameterization is also found elsewhere (Jurado et al., 2005), and the overall uncertainty in wet deposition flux estimates are discussed below.

Table 1

Characteristic organic carbon and black carbon aerosol content for each of the considered aerosol types

Aerosol type	f_{OC} (%)	f_{BC} (%)		Reference
Biomass burning	19.1	15.3	Submicron aerosol	(Brasseur et al., 2003)
	3.8	0.6	Supermicron aerosol	
Industrial emissions	15	5.1	Submicron aerosol	(Dachs and Eisenreich, 2000)
	10	1.4	Supermicron aerosol	
Dust	3.5	0.5	Supermicron aerosol	(Kinne et al., 2003)
Sea-salt	1.2	0.1	Supermicron aerosol	(Quinn et al., 2002) (OC), (Cavalli et al., 2004) (EC)
Continentially influenced	8	1.2	Submicron aerosol	(Brasseur et al., 2003)
	3.2	0.5	Supermicron aerosol	
Background aerosol	8	1.9	Submicron aerosol	(Brasseur et al., 2003)
	4.3	0.9	Supermicron aerosol	

2.2. Estimation of aerosol OC and BC concentrations

The global distribution of aerosol carbon concentration, i.e. $C_{p,OC}$ and $C_{p,BC}$, is an important issue to resolve before deposition estimates are made. Different sampling devices provide measurement of different populations of aerosols which influence the measurements of OC and BC concentrations. In addition, aerosols over the oceans are the result of a complex interplay of sources, processes and sinks (Heintzenberg et al., 2000). Measured concentrations show high variability (see supplementary material) of the aerosol distributions and concentrations, in part could be due to differences in the measurement methods used (Kinne et al., 2003). In addition, there is a significant paucity of measurements in the southern hemisphere. Therefore, a simple extrapolation from the available measured concentrations to obtain a global coverage over the oceans seems inappropriate, even less if temporal variability is to be addressed. Instead we have estimated the carbonaceous concentrations over the oceans from remote sensing data. Concentrations of OC and BC in aerosol are given by:

$$\begin{aligned} C_{p,OC} &= \text{TSP} \cdot f_{OC} \\ C_{p,BC} &= \text{TSP} \cdot f_{BC} \end{aligned} \quad (3)$$

where f_{OC} and f_{BC} ([0–1]) are the unitary mass fraction of organic and black carbon, respectively, in the aerosol (gC gTSP^{-1}), and TSP (mg m^{-3}) is the average total suspended matter in the atmospheric boundary layer (ABL). TSP has been obtained by applying an algorithm recently developed by Gassó and Hegg (2003) to optical properties given by MODIS sensor, assuming that the mass concentration of aerosols in the ABL doubles that in the free troposphere. This approach approximates well the concentration of aerosol in the marine boundary layer (Gassó and Hegg, 2003).

Both f_{OC} and f_{BC} are strongly dependent on the aerosol source and type. Examining various literature sources and accounting for the satellite retrieved parameters, we have considered six main categories of aerosols: biomass burning, urban and suburban aerosol -mainly due to industrial emissions-, dust, sea-salt aerosol, continentally influenced air masses in the distant marine atmosphere and baseline background aerosol. Each one of these aerosol types has characteristic f_{OC} and f_{BC} for their submicron and supermicron aerosol (Table 1). For dust and sea-salt aerosol we have just considered the supermicron fraction of aerosols. Higher fractions of organic and black carbon are found

with the submicron sizes (Table 1), even though these contribute less to depositional fluxes. Maximum percentages of carbonaceous matter are found in industrial emissions and fine biomass burning aerosols, emitted from vegetation fires (Table 1).

An important issue is the role of primary and secondary aerosols that are not defined by a separate category in Table 1 but included into the remaining aerosol groups. For example, Aller et al., 2005 has described the occurrence of virus and bacteria in marine aerosols, but this organic matter is here accounted within the sea-salt aerosol (or marine primary aerosol) or within the background aerosol. Similarly, it is known that secondary organic aerosol can be important from continental systems (Fuentes et al., 2000) but this is implicitly accounted within the continentally derived aerosol type. It is clear that the role of biogenic primary and secondary aerosols will receive much more attention in the coming decade which will allow constraining the importance of this fraction of the exchangeable carbon between the ocean and the atmosphere.

The spatial distribution of the predominant aerosol type has been approximated by determining thresholds to available satellite measures of variables such as: aerosol optical depth (AOD), its variability (σ_{AOD}), the fraction of small aerosols (η), wind speed distributions (u_{10}) and fire-counts (Table A1.1, in the supporting material). This approach is based on published sources and previous knowledge of aerosol spatial and seasonal distributions (Husar et al., 1997; Heintzenberg et al., 2000; Kaufman et al., 2001, 2002; Smirnov et al., 2002; Brasseur et al., 2003; Textor et al., 2005). A detailed explanation of the reasoning and hypotheses used, as well maps of the modeled aerosol distributions at monthly scale are reported in the supporting information.

3. Results and discussion

3.1. Aerosol organic carbon concentrations and validation

Monthly averages of the global distribution of C_{OC} and C_{BC} (Fig. 2) were obtained combining Eq. (3), TSP estimates

and concentrations of OC and BC (see supplementary material). In order to validate these results, we have performed a comparison between a comprehensive number of published measurements from coastal stations, research cruises, aircrafts and sun photometers and our estimated C_{P_OC} and C_{P_BC} (see Table A2 in supplementary material for published measurements). Estimated C_{P_OC} and C_{P_BC} agreed with measurements in the northern hemisphere within a factor of 2–3 in most cases, but tend to overestimate southern hemisphere OC and BC concentrations (Fig. 3), where coarse sea-salt aerosol plays a major role (see Fig. A1.1 in supplementary material). This discrepancy could be associated to the uncertainty associated to the load of organic matter in sea-salt aerosol in the remote marine aerosol (O'Dowd et al., 2004, 2008; Shinozuka et al., 2004; Phinney et al., 2006). The fraction of OC in marine aerosol is estimated to be small in this study (see Table 1), lower than some reports (O'Dowd et al., 2004; Duarte et al., 2006), since only aerosol in the lower part of the atmospheric boundary layer will be relevant for deposition estimates. Still, for recently generated marine aerosol, the composition of the aerosol may be similar to that of seawater (Lewis and Schwartz, 2004), and thus it is possible that in some cases marine aerosol has even lower contributions of organic matter. However, as shown in Fig. 3, there is a lack of measurements in the southern ocean.

The estimates provided here of OC and BC concentrations in aerosol are made for the complete size distribution of aerosols, while most of the various sampling methods used for the measurement of aerosol (see supplementary material) do not sample, in most cases, the complete aerosol population, thus contributing to part of the discrepancy between estimates and measurements. Nevertheless, this comparison is justified because there is not a clear relationship between OC and BC concentrations and sampling method used for the reviewed literature data set.

3.2. Dry deposition of aerosol OC to the global oceans

Dry deposition fluxes of OC aerosol to the global oceans show a large geographical variability (Fig. 4). Estimated

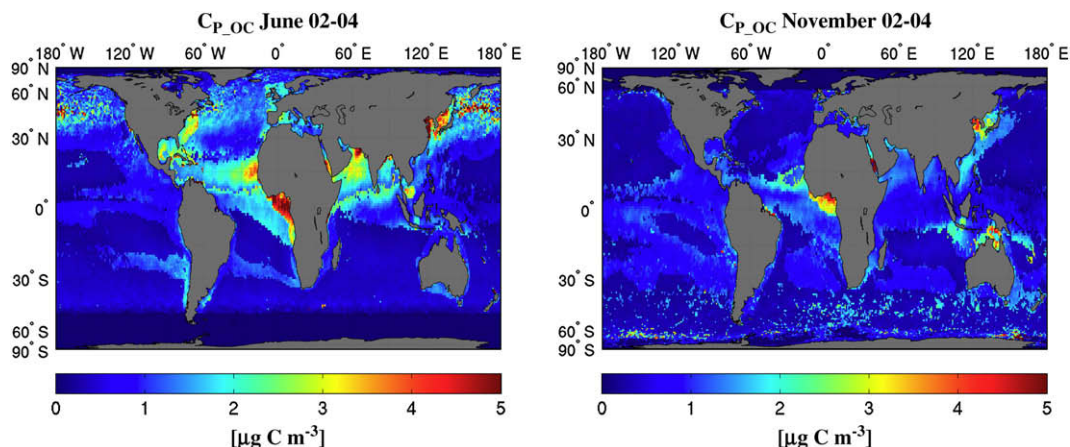


Fig. 2. Global distribution of aerosol organic carbon for June and November 2002–2004.

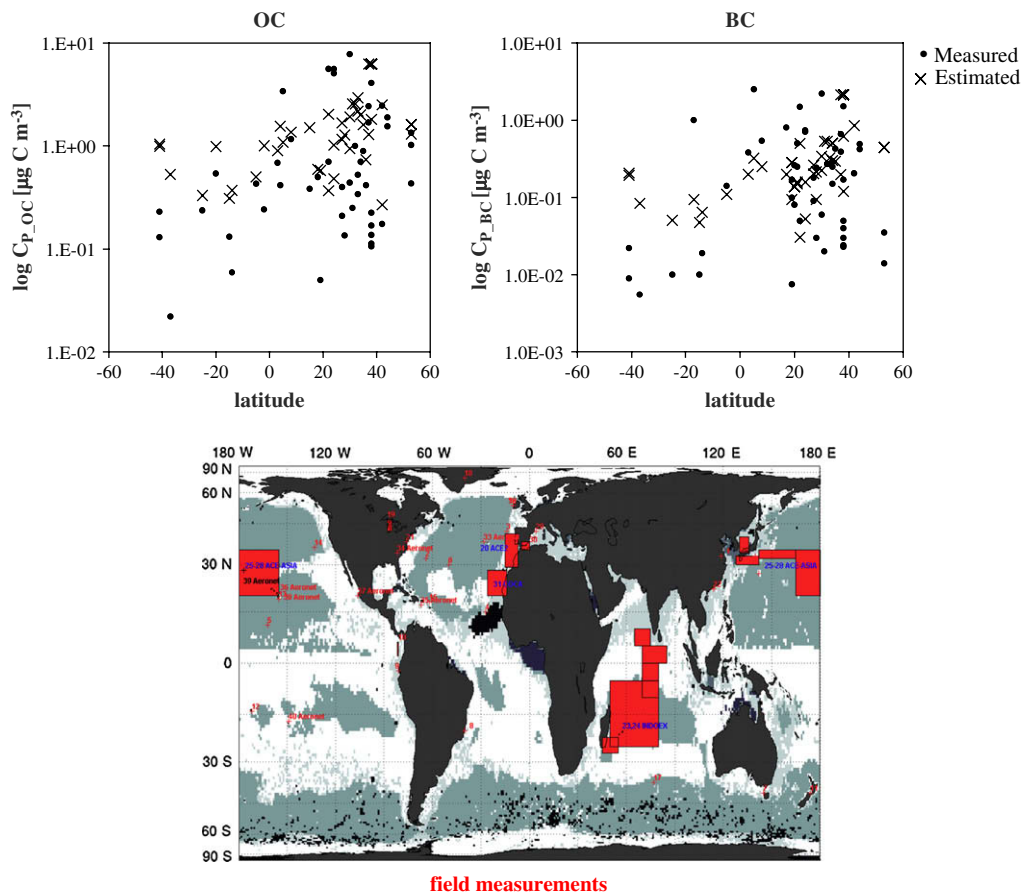


Fig. 3. Comparison of measurements and predictions of aerosol organic carbon and black carbon. Also a map indicating the location of measurements is shown (cross: sampling site, rectangle: spatial coverage of the campaign).

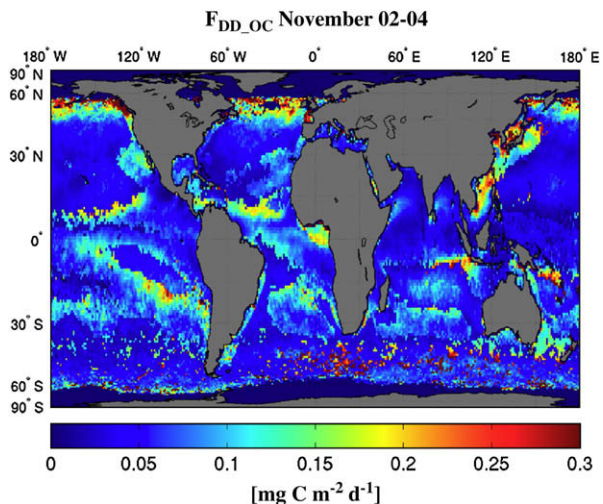


Fig. 4. Global distribution of dry aerosol deposition of organic carbon for November 2002–2004.

fluxes were higher in regions with high aerosol OC concentrations (see Fig. 2), and where environmental conditions, such as larger aerosol size and high wind speed, lead to larger dry deposition velocities. Dry deposition fluxes tend to be high at high latitude regions, where wind speed is higher, and in dust influenced areas. Even though dust has a low OC content (see Table 1), its large aerosol size and the relatively high wind speeds in trade winds regions do result in relatively high depositional fluxes, consistent with recently reported measurements (Dachs et al., 2005; Del Vento and Dachs, 2007). In addition, fluxes are especially important in areas with high concentrations of sea-salt, due to the scavenging role of this on organic compounds near the sea surface, as has been suggested for individual organic compounds (Jurado et al., 2004), or similarly, for fog in other regions (Collet et al., 2008). The results shown here and previous reports of relatively high concentrations of OC in marine aerosols (Monahan and Dam, 2001; Cavalli et al., 2004) point out to the important role that marine aerosols play in the lower atmosphere, an issue that requires further research. Conversely, OC dry aerosol deposition is especially low in the middle of the oceanic gyres.

Table 2

Latitudinal yearly averaged and accumulated fluxes of organic carbon (OC) and black carbon (BC)

	Average [$\text{mg m}^{-2} \text{d}^{-1}$]					Accumulated [Tg y^{-1}]				
	$F_{\text{DD,OC}}$	$F_{\text{DD,BC}}$	$F_{\text{WD,P,OC}}$	$F_{\text{WD,P,BC}}$	$F_{\text{WD,G,OC}}$	$F_{\text{DD,OC}}$	$F_{\text{DD,BC}}$	$F_{\text{WD,P,OC}}$	$F_{\text{WD,P,BC}}$	$F_{\text{WD,G,OC}}$
60N–90N	0.18	0.03	0.17	0.03	0.35	0.19	0.03	0.41	0.07	1.93
30N–60N	0.11	0.02	0.53	0.10	0.98	1.51	0.24	8.81	1.74	25.52
0–30N	0.08	0.02	0.70	0.18	1.83	2.40	0.48	21.96	5.56	72.50
30S–0	0.08	0.01	0.33	0.07	1.48	2.61	0.46	10.46	2.22	60.77
60S–30S	0.13	0.01	0.18	0.02	0.79	3.69	0.41	5.43	0.73	25.87
90S–60S	0.21	0.03	0.05	0.01	0.17	0.51	0.07	0.15	0.02	0.74
TOTAL						11	2	47	10	187

Dry deposition: F_{DD} ; wet deposition of particles: $F_{\text{WD,P}}$; wet deposition of gases: $F_{\text{WD,G}}$.

Table 2 shows the average and yearly accumulated OC dry deposition fluxes to the global oceans for latitudinal bands. The yearly accumulated flux is the sum of each of the monthly climatological means. The global dry deposition flux of OC is 11 Tg C y^{-1} , with a major contribution from the 30°S – 60°S region and inter-tropical latitudinal bands. This flux is less than 1% of the net CO_2 exchange between the atmosphere and the ocean (Takahashi et al., 2002; Fletcher et al., 2006). Even small, an important fraction of aerosol OC appears to be effectively transferred to deep waters and oceanic sediments as it is ubiquitously shown by molecular signals in settling particles and sediments (Gagosian et al., 1987; Wakeham et al., 1997).

3.3. Wet deposition of aerosol OC to the global oceans

Wet deposition does scavenge aerosols and gas phase compounds; therefore it is necessary to consider both contributors to the total wet deposition flux. Wet scavenging of aerosol OC and BC is estimated from Eq. (2). Fig. 5A shows the global distribution of wet deposition of aerosol OC for a representative month and Table 2 shows the average and accumulated fluxes for latitudinal bands. These fluxes follow the variability of aerosol OC concentration and precipitation rates. Therefore, it is clear that higher fluxes are found in the inter-tropical area and at high latitudes where annual precipitation is more than 10

fold that of other regions. Rain water scavenges aerosols very effectively. Thus, a rain event can remove most of the aerosols in the atmospheric boundary layer and contribute with a pulse input of OC, and other elements, to the ocean. It has been reported that these inputs can fuel bacterial productivity (Seitzinger and Sanders, 1999) and thus have an important impact on oceanic processes. This contrasts with dry deposition of aerosol OC, which even though can also have a pulsing nature, rank well below in magnitude to the inputs associated with intense rain events, and it has been shown that do not induce a significant increase of bacterial productivity (Duarte et al., 2006).

Concerning black carbon, Table 2 shows the estimated average and accumulated dry and wet aerosol deposition. Black carbon deposition is about 2 and 10 Tg C y^{-1} for dry and wet aerosol deposition. The total depositional flux is much higher than the values of 0.17 – 0.32 Tg C y^{-1} used in current global transport models of aerosol BC (Cooke and Wilson, 1996; Wang, 2004). The discrepancy between these and present estimates could be caused by the different approaches used, since the models were based on emission inventories and constant dry deposition velocities, thus not accounting for the large temporal and spatial variability in deposition fluxes. However, more measurements of BC over the oceans are needed to constrain these estimations.

The global aerosol OC wet deposition flux is calculated to represent 47 Tg C y^{-1} . This figure is within the range of

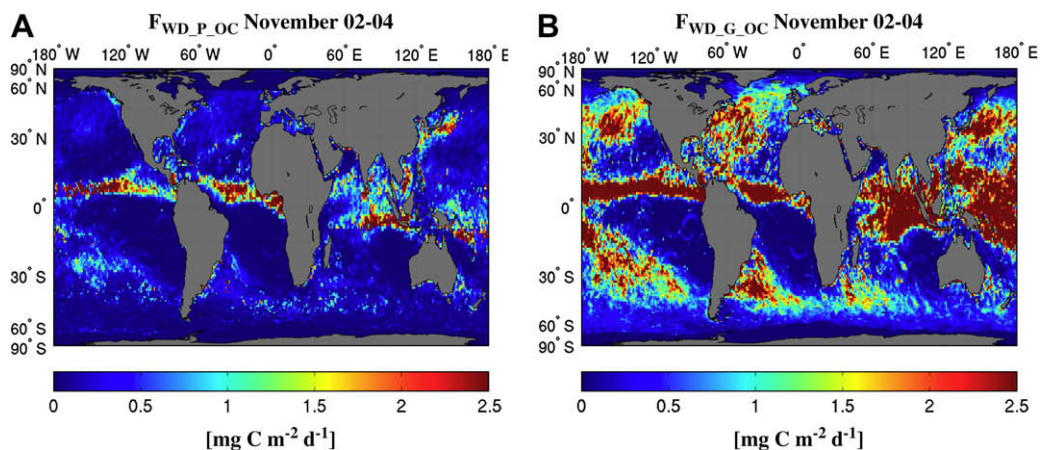


Fig. 5. Global distribution of wet deposition of organic carbon for November 2002–2004. Results for wet deposition of aerosol OC (A) and gas phase OC (B) are shown separately.

the estimate (20–79 Tg C y⁻¹) made by [Duce et al. \(1991\)](#) two decades ago. Together with the dry deposition flux of OC it results in an input of 58 Tg C y⁻¹, about half of the global emission estimate of 106 Tg C y⁻¹ of aerosol OC ([Liou et al., 1996](#)), which was the best available estimate until the recent work by [Willey et al. \(2000\)](#). These authors arrived to an estimate of 90 Tg C y⁻¹ for total wet deposition to the oceans, including dissolved and particulate OC from rainwater. It is difficult to ascertain which fraction of Willey's et al. estimate is due to particulate wet scavenging. Even though particulate OC only made 10–15% of total rainwater OC, aerosol OC could also be transferred to the dissolved phase during scavenging. This leads to the consideration of the magnitude of wet deposition of total gas phase organic compounds; the estimation of which has not yet been attempted for total organic carbon, but has been given for some individual organic compounds ([Gigliotti et al., 2002](#); [Jurado et al., 2005](#)

Table 3

Comparison between rainwater POC and DOC measured in island and coastal sites and those estimated in this study

Source	Location	(lat, lon)	Modeled POC	Modeled POC + DOC (μM)	Measured POC + DOC (μM)
(Raymond, 2005)	New Haven CT, coastal	(41N, 73W)	27	67	52
(Kieber et al., 2002)	New Zealand, marine	(46S, 169E)	23	63	58
(Willey et al., 2000)	New Zealand, marine	(46S, 169E)	23	63	24
(Willey et al., 2000)	Wilmington NC, coastal	(34N, 78W)	26	66	114
(Eklund et al., 1997)	Costa Rica, coastal	(10N, 84W)	18	58	58
(Sempere and Kawamura, 1996)	W. Pacific, marine	(20S, 160E)	7	47	125
(Zafirou et al., 1985)	Enewetak Atoll, marine	(12N, 163E)	5	45	22
(Williams, 1967)	N. of Samoa, marine	(6S, 174W)	7	47	56
(Williams, 1967)	Del Mar CA, marine	(33N, 117W)	21	58	24
Mean \pm SE			17 \pm 9	57 \pm 9	59 \pm 37

wet deposition fluxes (Jurado et al., 2005), and Dachs et al. (2005) reached the same conclusion for total OC. Therefore, it is possible that global fluxes of exchangeable OC are much higher than those reported for wet and dry deposition, and even comparable to those reported for CO₂. This confirms the urgency for a trans-regional assessment of air–water diffusive fluxes of organic carbon, an improvement of the existing methods for such measurements, and the elucidation of their role in the regional and global carbon cycle.

3.6. Research directions on the atmosphere–ocean exchanges of organic matter and black carbon

The present study shows that there is a high spatial and seasonal variability in the dry and wet deposition fluxes of OC and BC. The results have been validated with the available measurements, but there is an urgent need for trans-regional measurements of the different atmospheric deposition fluxes. Especially urgent is the determination of the diffusive fluxes of organic matter between the atmosphere and the ocean, which are suggested to be important, and the elucidation of the major families of organic compounds accounting for these fluxes. Once the regional and global total atmosphere–ocean exchange of OC and BC are constrained, it will be possible to evaluate the implications of these fluxes for the global cycle of carbon.

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Appendix A. Supplemental material

Supplementary information for this manuscript can be downloaded at doi: [10.1016/j.atmosenv.2008.07.029](https://doi.org/10.1016/j.atmosenv.2008.07.029).

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