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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.

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 O_3 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 CO_2 measurements only, accounting for 30–40% of the anthropogenic perturbation in the carbon cycle (see Houghton, 2003 for example). The continents are also though 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 CO_2 disequilibrium between surface seawaters and the lower atmosphere, which have allowed to estimate the global atmosphere–ocean exchange of CO_2 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 $1^\circ \times 1^\circ$, 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 un 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