

## Globally significant oceanic source of organic carbon aerosol

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[1] Significant concentrations of organic carbon (OC) aerosol are observed at three oceanic surface sites (Amsterdam Island, Azores and Mace Head). Two global chemical transport models (CTMs) underpredict OC concentrations at these sites (normalised mean bias of  $-67\%$  and  $-58\%$ ). During periods of high biological activity monthly mean concentrations are underpredicted by a factor of 5–20. At Amsterdam Island and Mace Head, observed OC correlates well ( $R^2 = 0.61\text{--}0.77$ ) with back-trajectory weighted chlorophyll-a, suggesting an oceanic OC source driven by biological activity. We use a combination of remote sensed chlorophyll-a, back trajectories and observed OC to derive an empirical relation between chlorophyll-a and the total oceanic OC emission flux. Using the GEOS-chem CTM we show a global oceanic OC emission, from primary and secondary sources, of  $\sim 8$  Tg/year matches observations. This emission is comparable in magnitude to the fossil fuel OC source and increases the simulated global OC burden by 20%.  
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[2] The possibility of an oceanic organic carbon (OC) aerosol source has been recognised for many years [Blanchard, 1964] with significant concentrations of OC observed at oceanic sites [Novakov *et al.*, 1997; Putaud *et al.*, 2000; Cavalli *et al.*, 2004; O'Dowd *et al.*, 2004; Yoon *et al.*, 2007; Pio *et al.*, 2007]. Postulated source mechanisms include primary emissions of organic-enriched sea-spray aerosol through the bubble bursting mechanism [O'Dowd *et al.*, 2004] and secondary organic aerosol (SOA) formation from oceanic emissions of gas-phase volatile organic compounds (VOCs) [O'Dowd and de Leeuw, 2007].

[3] A regional mechanistic analysis of the primary oceanic OC emission has recently been completed for the North East Atlantic [O'Dowd *et al.*, 2008]. This study implements a combined organic-inorganic sub-micron sea-spray source function into a regional climate model. The source function can simulate the OC fraction in sea-spray observed at Mace Head (on west coast of Ireland).

[4] The global emission of oceanic OC is still highly uncertain. In this study we use observations of OC from 3 oceanic sites, combined with remote-sensed products, back-trajectory analysis and a global chemical transport model (CTM) to produce a top-down estimate of the global oceanic source of OC aerosol (from both primary and secondary sources) to the atmosphere.

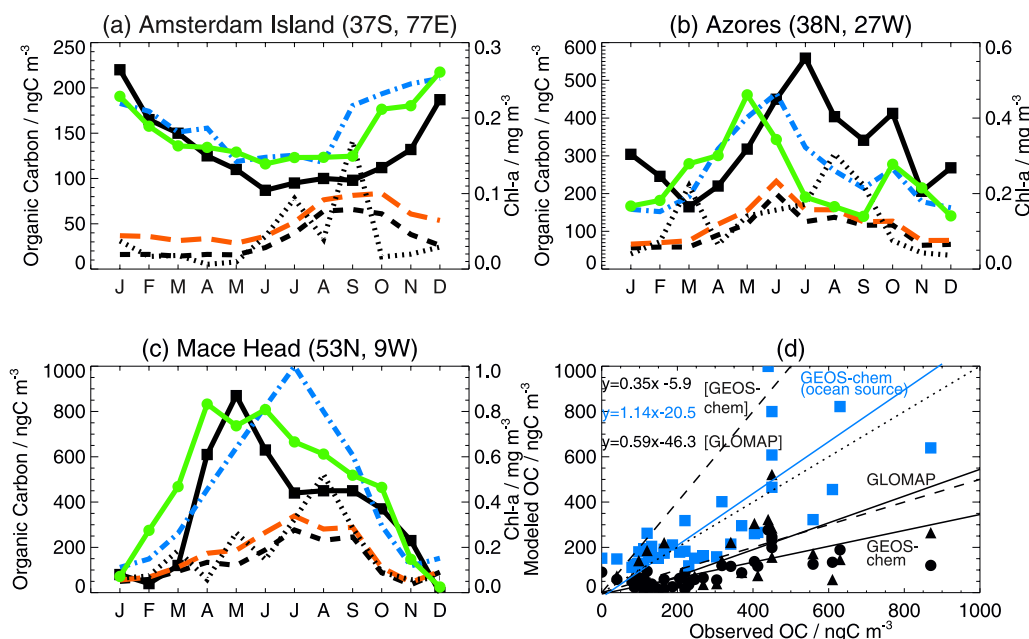
[5] Figure 1 shows the observed seasonal cycle of OC aerosol at 3 marine locations. Also shown are the modeled seasonal cycle of OC from the GEOS-chem [Park *et al.*, 2003] and GLOMAP [Spracklen *et al.*, 2005, 2008] global CTMs. Observations are from Amsterdam Island ( $37^{\circ}31'S$ ,  $77^{\circ}19'E$ ) for June 2003 to July 2005 (J. Sciare *et al.*, manuscript in preparation, 2008), the Azores ( $38^{\circ}41'N$ ,  $27^{\circ}21'W$ ) for July 2002 to June 2004 [Pio *et al.*, 2007] and Mace Head ( $53^{\circ}20'N$ ,  $9^{\circ}54'W$ ) for January 2002 to June 2004 [Yoon *et al.*, 2007]. Measurements are determined from thermal-optical analysis of weekly bulk samples on pre-fired quartz filters. The observations at Mace Head have been filtered using back trajectory analysis and a  $40\text{ ng m}^{-3}$  threshold for black carbon (BC) concentrations to remove continental influence. For comparison with Mace Head, model values for the domain  $27.5^{\circ}\text{--}12.5^{\circ}W$ ,  $44^{\circ}\text{--}56^{\circ}N$  are used. Simulated average BC concentrations in this domain are below the threshold value of  $40\text{ ng m}^{-3}$ . At Amsterdam Island, negligible local contamination and remote marine conditions were assessed from BC measurements performed on these filter samples which showed values systematically below  $50\text{ ngC m}^{-3}$ . Ion determination (including non-sea-salt sulfate and methane sulfonic acid (MSA)) was also performed on these filters following the method reported by Sciare *et al.* [2007].

[6] We use GEOS-chem v7.04 (see <http://www-as.harvard.edu/chemistry/trop/geos>) driven by assimilated meteorological data from the NASA Goddard Earth Observing System (GEOS)-4 at  $4^{\circ} \times 5^{\circ}$  horizontal resolution for the years 2001–2005. Anthropogenic emissions of BC and OC are as described by Park *et al.* [2003] and biomass burning emissions are from the Global Fire Emissions Database version 2 (GFEDv2) [van der Werf *et al.*, 2006]. We assume that 80% of BC and 50% of OC emitted from primary sources are hydrophobic with a 1.2 day e-folding conversion from hydrophobic to hydrophilic [Chin *et al.*, 2002]. We assume a 10% hydrophilic carbon yield of OC from terpenes [Chin *et al.*, 2002] with emissions calculated according to Guenther *et al.* [1995]. We also compare with the GLOMAP size-resolved global aerosol microphysics model. We run this model only for the year 2000 at  $2.8^{\circ} \times 2.8^{\circ}$  horizontal resolution. The model is forced by ECMWF analyses. Aerosol emissions are as described by Spracklen *et al.* [2006].

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**Figure 1.** Seasonal cycle of observed OC aerosol (solid black squares) and back-trajectory weighted SeaWiFS chlorophyll-a concentrations (solid green circles) at (a) Amsterdam Island, (b) the Azores, and (c) Mace Head. Simulated OC is shown without an oceanic OC source (GEOS-chem, black dashed; GLOMAP, black dotted) and for GEOS-chem with a global oceanic emission of  $\sim 1$  Tg OC/year (red long dashed) and  $\sim 8$  Tg OC/year (blue dash-dot). (d) Scatterplot of simulated versus observed monthly mean OC concentrations at the 3 sites. Models with no oceanic OC source (circles, GEOS-chem; triangles, GLOMAP) and GEOS-chem with an  $\sim 8$  Tg OC/year oceanic source (blue squares). The labeled solid lines show reduced major axis regression. Dotted line shows the 1:1 and dashed line the 2:1 relations.

[7] The two CTMs underpredict the observed OC concentrations and do not simulate well the seasonal cycle (Figures 1a–1c). At all three sites the greatest concentrations of OC are observed during summer. Both models best predict OC concentrations during winter months where underprediction is generally less than a factor of 2. Outside of the winter months, the models underpredict concentrations by a factor of 5–20. Figure 1d shows a scatter plot of the monthly mean model and observed concentrations, and demonstrates the low bias in both models (GEOS-chem regression line has slope of  $0.35 \pm 0.04$ , mean bias (MB) of  $-180 \text{ ngC m}^{-3}$ , normalised mean bias (NMB) of  $-67\%$  (mean bias (MB) =  $1/N \sum_{i=1}^n P_i - O_i$  and normalised mean bias (NMB) =  $100\% \times \sum_{i=1}^n (P_i - O_i) / \sum_{i=1}^n O_i$  where  $P_i$  is the model value,  $O_i$  is the observed value and  $N$  is the number of monthly mean model-observation pairs), correlation coefficient  $R^2 = 0.43$ ; GLOMAP regression line has slope  $0.59 \pm 0.08$ , MB of  $-160 \text{ ngC m}^{-3}$ , NMB of  $-58\%$ ,  $R^2 = 0.28$ ).

[8] In contrast, the seasonal variability and magnitude of BC (GEOS-chem: Amsterdam Island NMB  $-2\%$ ,  $R^2 = 0.62$ , Azores NMB  $-24\%$ ,  $R^2 = 0.38$ ; GLOMAP: NMB Amsterdam Island  $-28\%$ ,  $R^2 = 0.45$ , Azores NMB  $-50\%$ ,  $R^2 = 0.19$ ), MSA (GEOS-chem: Amsterdam Island NMB  $32\%$ ,  $R^2 = 0.83$ ) and sulfate aerosol (GEOS-chem: Azores NMB  $-9\%$ ,  $R^2 = 0.47$ ; GLOMAP: NMB  $-9\%$ ,  $R^2 = 0.29$ ) are generally better reproduced, indicating that aerosol transport from the continents, the oceanic sulfur source and aerosol sinks are reasonably represented by the models.

[9] This comparison suggests a missing source of OC aerosol at 3 geographically diverse marine sites. Underprediction of SOA from terrestrial sources may contribute somewhat to the low model bias. The budget of SOA is very uncertain [Goldstein and Galbally, 2007] but the contribution of terrestrial isoprene to OC at these remote sites is currently predicted to be small [Henze and Seinfeld, 2006]. Below we show that the observed OC is correlated to oceanic biology suggesting an oceanic source is a more likely explanation for low model bias.

[10] To explore the possibility of a missing oceanic source of OC, we analysed relationships between the seasonal cycle of OC at surface sites and oceanic chlorophyll-a observed from satellite. Chlorophyll-a is weighted according to the calculated ‘fetch’ of the site from back-trajectory analysis. Kinematic three-day atmospheric back trajectories, arriving hourly at each surface site over a full annual cycle, are calculated using ECMWF ERA-40 reanalysis winds using the OFFLINE trajectory model [Methven, 1997]. Remote-sensed monthly-mean oceanic chlorophyll-a concentrations at  $0.25^\circ$  resolution are derived from SeaWiFS Level 3 daily products provided by the NASA/GSFC/DAAC [O’Reilly et al., 1998]. For each month, weighted chlorophyll-a concentrations for each site are calculated according to overlap of each back-trajectory time-step (where the pressure of the back trajectory is greater than 850 hPa) to each  $0.25^\circ$  surface chlorophyll-a gridbox. We assume chlorophyll-a can impact OC concentrations over the three days prior to air mass arrival. This produces a chlorophyll-a ‘footprint’ for each month and site, which is

the mean chlorophyll-a exposure for air masses arriving in a given month, during their last three days of advection. This technique is an improvement over previous attempts to correlate chlorophyll-a and OC which have used mean chlorophyll-a values over a fixed oceanic region [O'Dowd *et al.*, 2008].

[11] Figure 1 shows that our trajectory-weighted chlorophyll-a product correlates well with observed OC at Amsterdam Island ( $R^2 = 0.60$ ) and Mace Head ( $R^2 = 0.75$ ) but poorly at the Azores ( $R^2 = 0.01$ ). Observed OC at the Azores lag the springtime maximum in chlorophyll-a by about 2 months. Similar behavior has been reported previously for dimethyl sulfide (DMS), with strong correlations between DMS and chlorophyll-a at high latitudes [Vallina *et al.*, 2006] and a lag between the two variables in sub-tropical oceans, referred to as the 'DMS summer paradox' [Simó and Pedrós-Alió, 1999].

[12] Our observed correlations between atmospheric OC and oceanic chlorophyll-a, as well as those from previous studies [O'Dowd *et al.*, 2004, 2008], support a biologically driven oceanic OC source. We scale SeaWiFS chlorophyll-a concentrations, [Chl-a], by an emission factor,  $A$ , to give a total oceanic OC emission:  $OC_{emis} = A$  [Chl-a]. We treat these emissions as water-insoluble (hydrophobic) and implement them in the GEOS-chem CTM. Treating a fraction of the emissions as hydrophilic would increase the atmospheric rate of removal and hence the emission required to match the observations. Our top-down method calculates the total oceanic OC flux (from primary and secondary oceanic sources) by matching modeled and observed OC concentrations, using an empirically based relationship between observed OC and chlorophyll-a. The low time resolution ( $\sim 1$  week) OC filter measurements are sufficient to derive a monthly-mean OC emission dependent on chlorophyll-a but do not allow us to deduce the dependence of the OC emission on wind-speed variability across a given month. The lack of wind speed in our empirical relation  $OC_{emis} = A$  [Chl-a] does not imply a lack of wind speed dependence in the mechanism, only that the effect is averaged out.

[13] We varied the emission factor,  $A$ , until the best match between modeled and observed OC concentrations was obtained. Figures 1a–1c shows the modeled concentrations of OC for two different oceanic OC emission scenarios. An emission factor of  $0.4 \text{ ngC m}^{-2} \text{ s}^{-1}/[\text{mg [Chl-a] m}^{-3}]$  results in a regression line with slope  $0.41 \pm 0.05$ , model MB of  $-160 \text{ ngC m}^{-3}$ , and NMB of  $-58\%$ . The best match requires an emission factor of  $3.2 \text{ ngC m}^{-2} \text{ s}^{-1}/[\text{mg [Chl-a] m}^{-3}]$  resulting in a regression line with slope  $1.14 \pm 0.13$ , model MB of  $18 \text{ ngC m}^{-3}$  and NMB of  $7\%$ .

[14] The conversion timescale from hydrophobic to hydrophilic OC will impact the oceanic emission required to match observations. Park *et al.* [2005] used the GEOS-chem model to show that a conversion timescale of  $1 \pm 1$  day best matched export efficiency of BC from Asia observed during the TRACE-P experiment. Atmospheric conversion timescales are likely to vary both spatially and temporally. Increasing the simulated conversion timescale by a factor of 2 (from 1.2 to 2.4 days) decreases the oceanic OC emission required to match observations by 12%.

[15] For the years 2001–2005, our best match emission factor, corresponds to a global oceanic source of  $7.8 \pm 0.1$

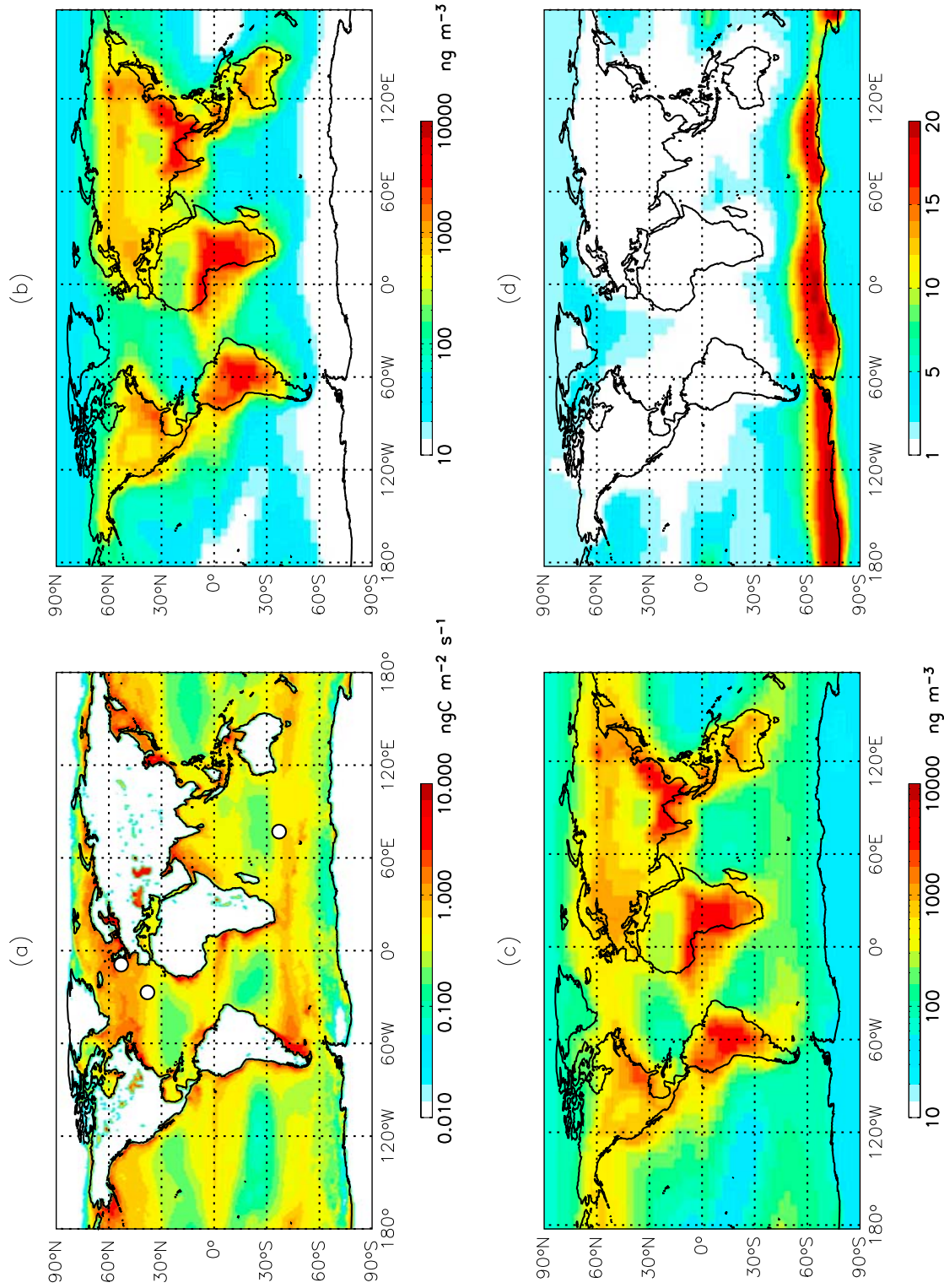
Tg OC  $\text{yr}^{-1}$ . The geographical distribution of our estimated OC source is shown in Figure 2a. This oceanic source increases the simulated annual average (2001–2005) OC burden by 20%, from  $0.68 \text{ Tg C}$  to  $0.82 \text{ Tg C}$ . Figures 2b and 2c show GEOS-chem model annual average surface concentrations of OC without and with an oceanic OC source. Our oceanic OC emissions increase modeled surface OC concentrations by up to  $0.7 \mu\text{g m}^{-3}$ ; with the largest increases in the North Atlantic, North Pacific and the Southern Ocean (between  $30^\circ$  and  $60^\circ\text{S}$ ). Outside of ocean upwelling regions (off the coast of Africa and South America), where chlorophyll-a concentrations are large, tropical oceans show relatively small increases in OC. Regions of continental outflow show less than 30% increase in OC concentrations, whereas the Southern Ocean shows up to a factor of 20 or more enhancement in OC (Figure 2d).

[16] The Southern Ocean source of OC can be further evaluated using observations of OC made during a North to South cruise of the Atlantic Ocean during November to December 1999 [Virkkula *et al.*, 2006]. At latitudes North of  $35^\circ\text{S}$  OC concentrations during this cruise are dominated by continental emissions. South of  $40^\circ\text{S}$  observations of OC are between  $100\text{--}300 \text{ ngC m}^{-3}$ . Without an oceanic OC source the model, sampled along the cruise track, predicts mean OC concentrations of  $13 \text{ ngC m}^{-3}$ . With an ocean OC source the model much better reproduces OC concentrations with a mean of  $240 \text{ ngC m}^{-3}$ .

[17] In this paper, we have produced a global estimate of the total (primary and secondary) oceanic OC source using recent observations of OC and remote sensed chlorophyll-a interpreted using a global CTM. Our global, top-down analysis complements regional, bottom-up studies which have been made of the primary OC source in the North East Atlantic [O'Dowd *et al.*, 2008]. We find that at 3 geographically diverse sites the modeled seasonal cycle of OC is improved by including an oceanic OC source driven by oceanic biological material. Our result implies a large source of OC aerosol ( $\sim 8 \text{ Tg OC/yr}$ ) throughout the global marine atmosphere linked to oceanic biological activity and comparable in magnitude to the anthropogenic fossil and biofuel OC source ( $5\text{--}30 \text{ Tg/yr}$ ) [Liousse *et al.*, 1996; Cooke *et al.*, 1999; Scholes and Andreae, 2000; Bond *et al.*, 2004; Ito and Penner, 2005]. Our emission is much smaller than the recent oceanic emission estimate of  $75 \text{ Tg OC/yr}$  made primarily by matching cloud drop effective radii simulated by a general circulation model with satellite observations [Roelofs, 2008].

[18] The climate impact of the oceanic OC source depends on the fraction that is emitted at submicron sizes. Observations of size-resolved aerosol in the North East Atlantic suggest that  $\sim 70\%$  of water-insoluble organic carbon (WIOC) resides in the submicron mode. If this figure is applicable to the global oceans our estimate would suggest a global submicron OC source of  $\sim 5.5 \text{ Tg/yr}$ , which is likely to have important implications for climate.

[19] Our technique does not allow us to establish the mechanism for this OC emission. However, the large fraction of WIOC observed at the 3 sites during periods of high biological productivity ( $50\text{--}80\%$  of total OC) suggests that primary production of aerosol may dominate the global marine OC source as has been observed in the North Atlantic [O'Dowd *et al.*, 2004; Cavalli *et al.*, 2004].



**Figure 2.** Global distributions as annual average for the years 2001–2005 of (a) estimated oceanic emission of OC scaled by SeaWiFS chlorophyll-a (empty circles show location of observation sites used in this analysis; see Figure 1) and simulated surface concentrations of OC (b) without an oceanic OC source, (c) with an oceanic OC source of ~8 Tg/year, and (d) simulated ratio of surface OC with divided without the oceanic source.

Such a primary particle OC source is an alternative explanation for the observed correlation between cloud drop number and biological activity which has been attributed to production of aerosol from gaseous emissions of oceanic isoprene [Meskhidze and Nenes, 2006]. Assuming an isoprene SOA yield of 3% [Henze and Seinfeld, 2006] our global OC emission estimate would imply an oceanic isoprene emission of  $\sim 250$  Tg, several orders of magnitude larger than current estimates [Palmer and Shaw, 2005].

[20] Further observations of OC in remote marine locations are essential to refine our oceanic emission estimate. Regions of continental outflow which are heavily influenced by primary emissions of OC and terrestrial emissions of biogenic volatile organic carbon make attribution of marine OC difficult. In contrast, large regions of the Southern Ocean experience very little OC from continental sources and are an ideal location to study oceanic emissions.

[21] This primary biological source of OC may result in a similar negative climate feedback to the long-established role of dimethylsulfide in the CLAW mechanism [Charlson et al., 1987]. Model studies of the climate implications of this source through modification of cloud condensation nuclei and cloud droplet number concentrations are now needed.

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