Supplementary information to: Ice nuclei in marine air : bioparticles or dust?

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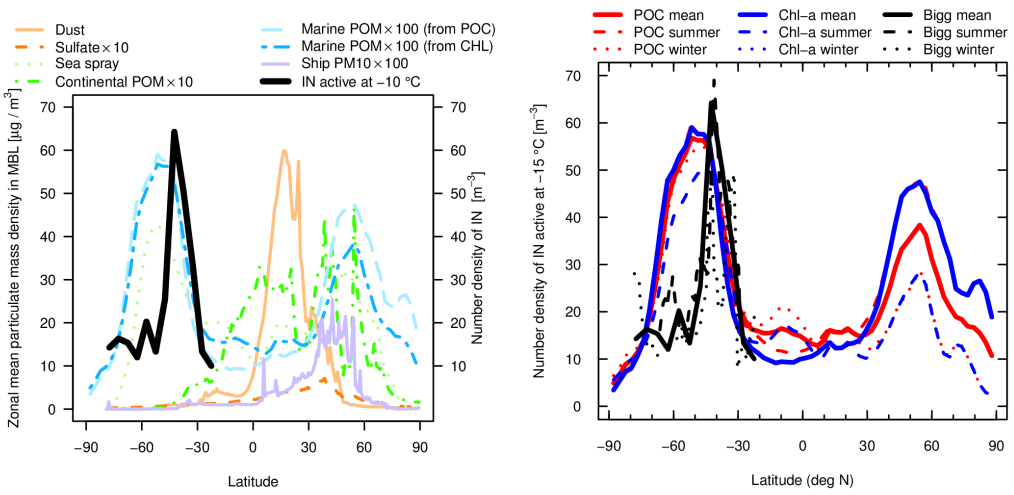
Zonal mean of emissions

Using the distribution of chlorophyll rather than POC to drive the emissions results in a closer match to the zonal mean distribution. Among the possible explanations are that ice nuclei concentrations in water might be more closely associated with phytoplankton than with organic debris, or that the distribution of ice nuclei might be latitudinally dependent.

Interestingly, the peak in the B73 data appears to be more closely matched in shape by the simulated emissions than by the simulated concentrations S-1, and the magnitude of the peak can be approximated by multiplying with a residence time of 2.3 days and dividing by a boundary layer height of 600 m, and assuming 10 IN per g POM and an enrichment factor of 100, all other factors as in Table 1 of the main paper.

We can only speculate about whether this apparent better correspondence is spurious or real. We do note that a better correspondence with the emissions would be expected if the IN decayed close to their source in a way that deactivated them as IN, perhaps due to photochemical processing upon exposure to the atmosphere. For example, it has been proposed that a large part of the marine organic aerosol is made up of exopolymer secretions (EPS) of marine microorganisms. EPS compounds are known to form gels that may disintegrate upon exposure to ultraviolet light or acidification [*Leck and Bigg*, 2005].

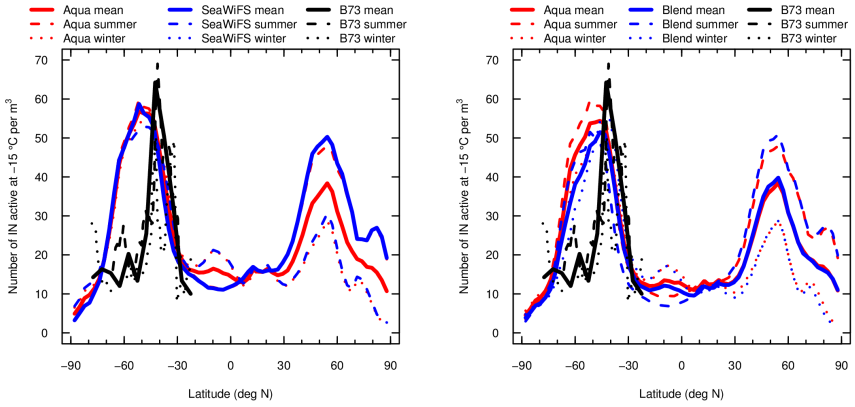


(a) Aerosol type comparison

(b) MODIS-Aqua POC and CHL emissions

Figure S-1: Longitudinal dependence of marine IN number densities from simulated emissions and longitudinally averaged number densities over the Southern Ocean [*Bigg*, 1973]. Number densities from emissions are derived assuming a residence time of 2.3 days, a boundary layer height of 200 m, 10 IN per mg marine POM an enrichment factor of 100 for POC and 10 for CHL, POM / POC = 1.8, POM / bacteria mass = 50, 1 $\times 10^{12}$ bacteria per mg Chl-a, and a mean bacteria mass of 100 $\times 10^{-15}$ g. Simulations using ocean climatologies of POC and chlorophyll from MODIS-Aqua (July 2002 – June 2010, [*Esaias et al.*, 2002]). (b): Comparison to AEROCOM zonal mean MBL aerosol mass densities and ship particulate emissions, in analogy to Figure 3 of main paper. (a): Comparison of emissions driven by MODIS-Aqua POC and CHL, in analogy to Figure 6 of main paper, and Figure S-2.

Comparison of simulations with alternative ocean satellite data



(a) POC-based simulation

(b) CHL-a-based simulation

Figure S-2: Zonal mean simulated marine IN number densities vs. zonal mean observed number densities over the Southern Ocean [*Bigg*, 1973]. (a): Simulations using ocean POC seasonal climatologies from MODIS-Aqua (July 2002 – June 2010, [*Esaias et al.*, 2002]) and SeaWIFS [*Hooker and McClain*, 2000; *Stramski et al.*, 2008]. (b): Simulations using ocean chlorophyll seasonal climatologies from MODIS-Aqua (July 2002 – June 2010), and a climatology constructed from CZCS satellite data and *in situ* data (1978 – 1986 [*Gregg and Conkright*, 2001]).

Comparison of IN concentrations with ship emissions and chlorophyll

Ship emissions are a significant source of particulate matter in remote marine regions and so might be considered a possible local source of IN. However, we consider it unlikely that ship emissions explain the observed IN

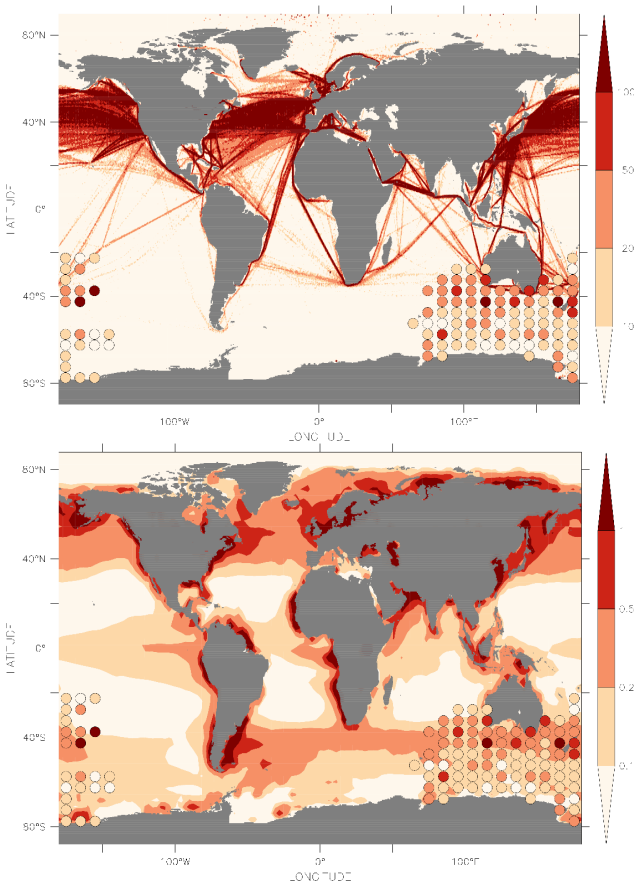


Figure S-3: Geographic distribution of B73 observed IN concentrations, compared to ship emissions and chlorophyll. Top: Natural logarithm of estimated particulates emitted from ships, assuming an 800-m boundary layer height and 5-day residence time in the boundary layer ($\mu g\,m^{-3}$), calculated from estimated year 1970 emissions [*Endresen et al.*, 2003; *Eyring et al.*, 2005, 2010; *Wang et al.*, 2007; *Lamarque et al.*, 2010], with natural logarithm of B73 IN mean number density (m^{-3}) superimposed as coloured dots (different colour scale). Bottom: Mean ocean chlorophyll-a concentrations from MODIS-Aqua (mg m^{-3}, July 2002 – June 2008), with B73 IN number densities (IN active at -15 °C per m^3, × 10).

distribution, because: 1. observations by [*Rosinski et al.*, 1987] found no IN while on the shipping lane from California to Hawaii, but high concentrations of IN in the Equatorial Pacific, far from major shipping lanes. 2. the spatial pattern of ship emissions does not match the spatial pattern of concentrations; closer inspection of the horizontal distribution confirms that the B73 IN concentrations are very high in several regions where there is no ship traffic (Figure S-3).

By contrast, there is a strong correspondence between the IN and chlorophyll spatial distributions (Figure S-3); this spatial correlation between marine primary productivity and B73 IN concentrations led *Schnell and Vali* [1976] to propose a marine biological source of ice nuclei.

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