**Atmospheric
Chemistry
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Discussions**

# Ice nuclei in marine air: bioparticles or dust?

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## Abstract

Ice nuclei can influence the properties of clouds and precipitation, but their sources and distribution in the atmosphere are still not well known. Particularly little attention has been paid to IN sources in marine environments, although anecdotal evidence suggests that IN populations in remote marine regions may be dominated by biological particles associated with sea spray. In this exploratory model study, we aim to bring attention to this long-neglected topic and identify promising target regions for future field campaigns. We assess the likely global distribution of marine biological ice nuclei using a combination of historical observations, satellite data and model output. By comparing simulated marine biological IN distributions and dust IN distributions, we predict strong regional differences in the importance of marine biological IN relative to dust IN. Our analysis suggests that marine biological IN are most likely to play a dominant role in determining IN concentrations over the Southern Ocean, so future field campaigns aimed at investigating marine biological IN should target that region. Climate-related changes in the abundance and emission of biological marine IN could affect marine cloud properties, thereby introducing previously unconsidered feedbacks that influence the hydrological cycle and the Earth's energy balance. Furthermore, marine biological IN may be an important aspect to consider in proposals for marine cloud brightening by artificial sea spray production.

## 1　Introduction

The formation of ice is a crucial process in cloud development, with most precipitation globally originating in clouds containing ice. Because pure water droplets cannot freeze at temperatures above about −40 °C, ice formation in warmer clouds is initiated by airborne particles that serve as ice nuclei (IN). Particles that are effective IN are rare, often fewer than one out of one million airborne particles (Rosinski et al., 1988). Current approaches to understanding global IN distributions consider only desert dust

and other continental sources (DeMott et al., 2010). However, there is anecdotal evidence that a marine source of biological particles determines IN abundance in air over remote, biologically active regions of the ocean.

## 1.1 Evidence for a possible marine biological IN source

Evidence from in situ observations reported in a number of studies scattered over four decades suggests that in remote, biologically active areas of the ocean, the background atmospheric IN concentrations measured on ships are highly influenced by local marine biological activity and sea spray production. Early evidence was provided by Bigg in 1973 (hereafter B73), who presented long-term measurements of IN concentrations in the marine boundary layer south of and around Australia and New Zealand (Bigg, 1973). Since that time, only a handful of further observations of marine IN concentrations have been published (Fig. 1). In the B73 observations, mean concentrations of IN were generally highest around 40° S, a region in which rough seas are common, marine biological activity is strong, and large amounts of sea spray containing organic matter are produced. It was later suggested that the source of IN might be associated with the plankton growing in this nutrient-rich upwelling region of the ocean (Schnell and Vali, 1976). Further evidence for a connection between marine biological activity and marine IN concentrations was uncovered in later studies: Ship-based measurements in remote marine regions found that atmospheric IN concentrations were higher in ocean upwelling regions, or were associated with high concentrations of biological materials in samples of ocean water. Physical and chemical analyses of IN collected in the remote marine boundary layer of the equatorial Pacific Ocean showed that they were submicron-sized, probably carbonaceous particles (Rosinski et al., 1987).

## 1.2  Candidates for marine biological IN

Types of marine biological particles that are found in the atmosphere and could potentially act as IN include:

1. Marine microorganisms: Only a minor fraction of organic particles in the marine aerosol (up to a few percent on average by number) are marine microorganisms, consistent with the composition of the particulate organic matter in the marine surface layer (Leck and Bigg, 2005b). Cultivated samples of certain marine plankton species are reported to be efficient IN, those that have been identified thus far include the phytoplankton species *Cachonina niei*, *Ochromonus danica* and *Porphyridium aerugineum* (Schnell, 1975), the marine dinoflagellate *Heterocapsa niei* (Fall and Schnell, 1985), although in these cases, the cultures included a variety of other species and ice nucleation activity could not be unambiguously attributed to one of these. More recently, IN activity was unambiguously identified in the cosmopolitan marine diatom species *Thalassiosira pseudonana* (Knopf et al., 2011; Alpert et al., 2011a), as well as the marine phytoplankton species *Nannochloris atomus* (Alpert et al., 2011b), but it should be noted that many other plankton cultures did not exhibit ice nucleation activity in laboratory tests (Schnell, 1975), and some samples from high-latitude waters have also been observed to inhibit, rather than promote, ice formation (Junge and Swanson, 2008). IN collected over the remote Pacific Ocean were found to evaporate completely in a vacuum, suggesting that they were not microorganisms (Rosinski et al., 1987).

2. Exopolymer secretions/colloidal aggregates: Another candidate for marine biological IN is aggregates of microcolloids and their components or building blocks, which likely originate from exopolymer secretions (EPS) of marine microbiota. Analysis of TEM photographs of hundreds of particles collected at each of five observation sites has shown that these have equivalent spherical radii that are usually in the range of about 10–40 nm, and can supply as many as 20 % of the particles in that size range (Bigg and Leck, 2008).

Further experiments with modern methods would be needed to characterize the IN activity of various particles in marine organic matter. A recent series of papers report results from such experiments (Knopf et al., 2011; Alpert et al., 2011a,b). These show that biological particles with different biogenic surfaces can have both comparable and contrasting effects on ice nucleation behavior, depending on the temperature, water vapor supersaturation and mode of freezing (deposition or immersion).

## 1.3 Enrichment of marine organic matter in sea spray

Laboratory experiments have shown that organic matter can be strongly enriched during sea spray aerosol formation by bubble bursting. Due to their surface-active properties, organic substances are preferentially collected on the surface of rising bubbles and subsequently transferred to the aerosol with concentrations that are commonly a factor of 10–100 or more higher than in the bulk seawater. The ratio of the concentration of a substance in the aerosol phase to its concentration in the bulk is termed the "enrichment factor" (EF) (see tabulated values in Table B1).

Bubble bursting experiments with ocean water sampled during a phytoplankton bloom show a dominant contribution of water-insoluble organic compounds to the submicron fraction of the marine aerosol, consistent with results from ship and coastal station measurements during bloom periods (O'Dowd et al., 2004; Facchini et al., 2008). In ship-based measurements in the North Atlantic and Arctic Oceans, the observed ratio of organic mass to Na in the submicron aerosol was enriched by a factor of $10^2$–$10^3$ relative to reported sea surface concentration ratios (Russell et al., 2010). This ratio is higher than many other reported enrichment factors, at least in part because the enrichment is greatest in the submicron aerosol. The global marine emissions of submicron primary organic aerosol particles by sea spray have been estimated to be $8.2\,\mathrm{Tg\,yr^{-1}}$ (Vignati et al., 2010).

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## 2   Methods

### 2.1   General equation for marine IN emissions estimate

In order to simulate the spatial and temporal distribution of the marine organic contribution to the atmospheric IN, we parameterized emissions to be consistent with knowledge of biologically-associated sea spray and observed IN concentrations in planktonic matter (Schnell and Vali, 1975). The emissions of biological IN are assumed to be proportional to the wind-speed-dependent emissions of accumulation mode sea spray (with dry radius range $0.05 < r_{dry} < 0.5\,\mu m$), calculated using the LSCE sea spray parameterization (Guelle et al., 2001; Kerkweg et al., 2006b). Sea spray emissions are then multiplied by an ocean biological variable, either chlorophyll or particulate organic carbon (POC), and by a scaling factor $\alpha$, i.e.:

$\alpha\ [(\text{kg sea spray})^{-1}(\text{mg POC or Chl-}a \times \text{m}^{-3})^{-1}]$

$\times$ POC or Chl-$a$ in seawater [mg m$^{-3}$]

$\times$ Accumulation mode sea spray emissions [kg m$^{-2}$ s$^{-1}$]

$= $ Emissions of biological IN active at $-15\,°C$ [m$^{-2}$ s$^{-1}$]                    (1)

The value of $\alpha$ is chosen to be consistent with observations that provide information about the relationship between POC and chlorophyll-$a$ concentrations and the concentration of IN in ocean water: $\alpha_{POC} = (75–900) \times 10^6$ [(kg sea spray)$^{-1}$ (mg POC m$^{-3}$)$^{-1}$] and $\alpha_{Chl} = (15–250) \times 10^9$ [(kg sea spray)$^{-1}$ (mg Chl-$a$ m$^{-3}$)$^{-1}$]. Details about the derivation of these factors follow (also see Table 1, as well as Tables 2 and A1). The values of $\alpha$ are each calculated as the product of five factors that can be estimated based on field or laboratory studies. However, each of these factors has a large uncertainty range, and any errors in the same direction will combine multiplicatively, so the uncertainty in $\alpha$ is at least a factor of 10 in each direction. For the purpose of demonstration, we have chosen values that lead to good agreement with IN concentrations as reported by B73, while remaining consistent with other observations; however,

as discussed further below, we emphasize that new observations will be needed to constrain the uncertainties.

## 2.2 Estimate of scaling factor $\alpha$

The value of $\alpha$ is chosen to be consistent with observations that provide information about the relationship between POC and chlorophyll-$a$ concentrations and the concentration of IN in ocean water. For POC-based emissions, it is given by:

$$\alpha_{POC} =$$

$$\times \ \frac{POM}{POC} \ (= 1.8)$$

$$\times \ \frac{IN}{POM} \ (= 10\text{–}20 \times 10^4 \, \text{mg}^{-1})$$

$$\times \ \text{Enrichment Factor (EF)} \ (= 500\text{–}1000)$$

$$\times \ \text{Particle size correction} \ (= 30\text{–}90)$$

$$/ \ \text{Sea salt in water} \ (= 36 \, \text{kg} \, \text{m}^{-3})$$

$$= 75 \times 10^6 \text{–} 900 \times 10^6 \ [(\text{kg sea spray})^{-1} \ (\text{mg POC} \, \text{m}^{-3})^{-1}] \tag{2}$$

and for chlorophyll-$a$-based emissions, it is given by:

$$\alpha_{Chl} =$$

$$\times \ \text{Bacterial cells per mg Chl-}a \ (= 4\text{–}7 \times 10^{12} \, \text{mg}^{-1})$$

$$\times \ \text{Mass of bacterial cell} \ (= 75\text{–}100 \times 10^{-12} \, \text{mg})$$

$$\times \ \frac{POM}{\text{bacterial mass}} \ (= 35\text{–}64)$$

$$\times \ \frac{IN}{POM} \ (= 10\text{–}20 \times 10^4 \, \text{mg}^{-1})$$

$$\times \ \text{Enrichment Factor (EF)} \ (= 500\text{–}1000)$$

$$/ \ \text{Sea salt in water} \ (= 36 \, \text{kg} \, \text{m}^{-3})$$

$$= 15 \times 10^9 – 250 \times 10^9 \ [(\text{kg sea spray})^{-1}(\text{mg Chl-}a \ \text{m}^{-3})^{-1}] \tag{3}$$

The ranges of values supported by the observations for the scaling factors relating biological proxies to IN concentrations are provided in Table 1. While a range of values are possible, the upper values chosen here for the scaling factors result in approximate agreement between simulated mean IN concentrations and the three-year mean concentrations reported by B73, while the lower values chosen for the scaling factors represent approximately the middles of the ranges of possible values found in the literature. Where not otherwise stated, results shown here use the higher estimate. A further constraint is applied that POM mass emissions not exceed 76 % of accumulation mode sea spray emissions, and bacterial mass emissions (an intermediate result in the Chl-$a$ scaling) not exceed 10 % of accumulation mode sea spray emissions. The emitted particles are assumed to be spherical, with a radius of 0.1 µm for POC-driven emissions and 0.2 µm for chlorophyll-driven emissions. Note that the mass ratio of POC to Chl varies greatly in ocean water, with highest values in chlorophyll-poor regions: one analysis of satellite data found this ratio ranged from 33 to 1736, averaging 575 for Chl $< 0.1 \ \text{mg m}^{-3}$ and 79 for Chl $> 1 \ \text{mg m}^{-3}$ (Stramski et al., 1998).

The greatest uncertainty in the scaling parameters $\alpha_{\text{POC}}$ and $\alpha_{\text{Chl}}$ is the enrichment factor, observed values of which vary over more than two orders of magnitude under different experimental conditions (Table B1). However, it has been consistently observed that enrichment of organics is greatest in accumulation mode sea spray aerosol, justifying our choice of a value at the upper end of this range for our estimate, which is derived from simulation of accumulation mode sea spray.

The second greatest uncertainty is in the fraction of marine organic particles that act as IN. We express this in terms of the number of IN per unit of particulate organic mass (POM). Experiments that measured the concentration of IN in particulate matter sieved from a plankton bloom (Schnell and Vali, 1975) are assumed to be approximately representative of this value, observed concentrations of IN in marine surface waters and marine particulates are shown in Table A1 and Fig. A1. In addition, scaling IN emissions with POM mass allows us to cross-check our simulation results with independent

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observations and models of marine POM concentrations and emissions (Fig. 2).

The overall scaling factor was chosen to obtain approximate agreement between simulated mean IN concentrations and the three-year mean concentrations reported by B73. For each parameter, we list the range of values allowed by observations together with the value chosen for the analysis presented here in Table 1 (supporting information in Tables 2 and B1).

We use satellite-derived proxies to scale the biological fraction of sea spray with ocean biological activity. For ocean surface particulate organic carbon (POC) and chlorophyll-*a* concentrations, we use seasonal climatologies from MODIS-Aqua (July 2002 to June 2010, Esaias et al., 2002).

Finally, the size distribution of the organic sea spray is not well known. The impact this may have on our analysis is discussed Sect. 2.6.

## 2.3 Enrichment factors in sea-to-air-transfer of organic aerosols

Gas bubbles bursting at the water surface eject two types of drops: jet drops and film drops. The jet drops emerge from the bottom of the collapsing bubble cavity, and each drop produces up to ten jet drops, the size of which is on the order of some tenths of a millimetre. Jet drops probably dominate the sea spray aerosol mass. In addition, each bubble produces a large number of film drops, up to about 75 per bubble, which arise from the thin film of water that separates the air in the bubble from the atmosphere. Most of the aerosols smaller than 1–20 µm are believed to be film drops, and film drops dominate the sea spray aerosol production in terms of total number. (Blanchard, 1989).

The studies summarised in Table B1 show that measured enrichment factors can be affected by a variety of variables, including bubble size, bubble rise distance, and chemical properties of the substance in question. For bacteria, increased cell surface hydrophobicity is believed to enhance the attachment to bubbles and therefore the transfer to aerosol. Cell surface hydrophobicity can be affected by cell pigmentation among other variables, this is hypothesised to explain the higher enrichment factors measured

for pigmented as compared to non-pigmented strains of *S. marcescens* (Burger and Bennett, 1985).

The measurement of enrichment factors is not entirely standardised, but two general approaches can be distinguished. The first approach is to artificially generate bubbles to form aerosol from an aqueous solution or marine water sample. This approach has the advantage of allowing controlled, reproducible experiments on the process level. The disadvantage is that the results can depend strongly on the method used to generate the aerosol, and this method may not precisely mimic the natural sea spray formation mechanism. Enrichment factors measured with bubble-bursting experiments refer to the ratio of the concentration of a substance in the aerosol droplets upon production to its concentration in the bulk water. A recent brief review of methods of bubble generation can be found in (Keene et al., 2007).

The second approach is to measure the concentration of a substance in the naturally occurring marine aerosol and the surface sea waters. Often, the submicron surface layer is distinguished from the bulk seawater near the surface. However, because freshly emitted aerosol mixes with a preexisting aerosol population and is rapidly chemically modified after entering the atmosphere, the interpretation of field measurements is ambiguous. To negate the concentrating effect of evaporation, enrichment factors are typically determined relative to the salt content of the aerosol, since sea salt shows little or no enrichment during sea-to-air transfer.

In Table B1, we present enrichment factors for various organic compounds, derived using both of the general approaches described above, and with other variations in methodology, which we summarise in the table. Some recent results on primary and secondary organic marine aerosol are reviewed in Rinaldi et al. (2010).

## 2.4 Sea spray parameterization and model simulations to estimate biological IN emissions

The emission parameterization for biological ice nuclei was implemented in a slightly modified version of the global chemistry-climate model ECHAM5/MESSy-Atmospheric

Chemistry (EMAC) (Jöckel et al., 2006), version 1.9. The following MESSy submodels were utilised for simulation of aerosol emission and deposition processes: online emissions via ONLEM (Kerkweg et al., 2006b), wet deposition (impaction and nucleation scavenging) via SCAV (Tost et al., 2006), and sedimentation and dry deposition via
5    SEDI and DRYDEP, respectively (Kerkweg et al., 2006a). Modifications to the model comprised the addition of emission functions for marine organic particles as described in the main text of this paper, and updates to the SCAV submodel (Tost et al., 2010).

All simulations were conducted in T42L90 resolution, for four simulated years (plus one year of start-up) with climatological sea surface temperatures and online simula-
10   tion of atmospheric dynamics. For ocean surface particulate organic carbon (POC) and chlorophyll-$a$ concentrations, we use seasonal climatologies from MODIS-Aqua (July 2002 to June 2010; Esaias et al., 2002). We emit a monodisperse, passively transported aerosol with a prescribed aerodynamic diameter of 0.1 µm for POC-driven emissions, or 0.26 µm for Chl-$a$-driven emissions, which gave similar results. This is
15   consistent with observations showing marine IN to have diameters between 0.1–0.3 µm (Rosinski et al., 1987, Sect. 2.6). Regions covered by sea ice were excluded, and points where satellite data were not available were set to zero (these largely overlap with sea ice regions).

## 2.5   Estimation of dust IN concentrations

20   State-of-the-art dust treatments in global atmospheric models can simulate dust deposition, surface mass concentrations and satellite-observable indicators of dust concentrations to within approximately a factor of ten. The largest errors generally appear in remote areas; in particular, many models underestimate surface mass concentrations at Antarctic coastal sites, and simulate more accurate surface concentrations
25   at measurement stations affected by Saharan dust than those affected by Asian dust (Huneeus et al., 2011). Unfortunately, no comprehensive evaluation of model dust distributions has yet been published for dust number concentrations. Observations of surface dust concentrations typically report only the mass density of dust in the air,

however, for ice nucleation either the number density or surface concentration density is more relevant. The relationship between these variables depends on the size distribution of dust particles, adding an additional source of model uncertainty.

The dust IN concentration at $-15\,^{\circ}$C is calculated from the dust surface area simulated in CAM-Oslo (Hoose et al., 2010a) and a temperature-dependent active site density (Niemand et al., 2012). This parameterization is obtained from a large number of immersion freezing experiments in the AIDA cloud chamber with natural dust samples, extending the study by Connolly et al. (2009). The uncertainty in the dust IN concentration is estimated to be about a factor of 10 in each direction.

## 2.6 Considerations with regard to particle size

Field campaigns in the equatorial Pacific and the Gulf of Mexico found that marine IN were submicron particles with equivalent radii in the range 50–250 nm (Rosinski et al., 1986, 1987, 1988).

If IN are microorganisms, then we can examine typical sizes of marine microorganisms to determine an approximate size. Numerous studies have estimated the mean size of marine bacteria. The results of some of these studies are presented in Table 2. Bacteria grow larger in the presence of greater nutrient availability, and bacteria in coastal waters are on average larger than bacteria in open ocean. We take a cell volume of $0.075\,\mu\text{m}^3$, or a spherical equivalent radius of 260 nm, as representative of typical marine bacteria sizes. The marine diatoms identified by Knopf et al. (2011) as ice nucleation active were an order of magnitude larger, with a typical diameter of ca. $5\,\mu\text{m}$ (corresponding to a geometric mean surface area of $1.2 \times 10^{-6}\,\text{cm}^2$), however, there are no field measurements available indicating the that such large ice nuclei are present in marine air in significant numbers. We further assume that the bacteria are externally mixed (not coated or attached to other particles), which is supported by single-particle observations (Pósfai et al., 2003; Bigg and Leck, 2001; Leck and Bigg, 2005a,b, 2008). We neglect any possible aerodynamic effects of microorganism shape, i.e. we assume the aerodynamic diameter of the microorganism is equal to its

spherical equivalent diameter.

Turning to studies of organic sea spray, these suggest that the organic fraction of the sea spray aerosol is largest for particle radii smaller than about $r = 100$ nm (maximum relative OM mass contribution and corresponding size fraction: 80 %, $r = 65$ nm (Keene et al., 2007); 77 ± 5 %, $r = 62.5$–125 nm (Facchini et al., 2008); 4 %, 35–38 nm (Modini et al., 2010)). During biologically active periods (algal bloom episodes), observations have found the aerosol mass below 200 nm diameter to be dominated by organic matter (O'Dowd et al., 2004). However, note that the particle size ranges with the greatest organic contribution represent the lower detection limits of the impactor samples in those studies (Facchini et al., 2008; Keene et al., 2007), so they do not rule out a large organic contribution to even smaller particle size ranges.

To test the effect of particle size on our analysis, we simulated the emissions and transport of particles with the following equivalent spherical radii: 50 nm, 100 nm and 260 nm. The change in simulated mean particle number mixing ratios was less than 5 % in all cases. There is, however, a large difference in the associated particle mass mixing ratios, which are implicitly used in converting simulated particle concentrations into ice nucleus concentrations (Table 1). In artificially-generated sea spray, the mass-weighted mean radius for the submicron aerosol is likely in the range 0.125–0.25 μm (Facchini et al., 2008). Relative to the 100 nm particles used in the simulations, particles in this size range have volumes ca. 2–16 times greater, and thus would contain 2–16 times more mass. This must be accounted for in converting between model-simulated aerosol number mixing ratios and aerosol mass densities, as reflected in Table 1.

# 3   Results and discussion

## 3.1   Simulated POM distribution

Simulated distributions of primary organic mass (POM) are broadly comparable with the results of a recent, state-of-the-art model study of marine primary organic aerosol that used chlorophyll maps to partition sea spray production into organic and inorganic parts (Vignati et al., 2010). The results of the simulation are qualitatively consistent with the climatological mean IN concentrations measured by B73, particularly in terms of the zonal distribution (Fig. 6; comparisons of results using different satellite data and the zonal profile of emissions can be found in Figs. S-1 and S-2, respectively). The simulated IN source and concentrations are largest in the mid-latitudes, especially the "roaring forties" of the Southern Hemisphere, where high wind speeds combine with seasonally strong biological activity to produce a strong emission to the atmosphere. The absolute value of the simulated IN concentrations is consistent within the large uncertainties both with the B73 measurements and with knowledge about the con- centrations of ice-nucleating particles in the biologically active surface waters of the oceans (Table A1 and Fig. A1), as well as the inclusion of particulates and especially water-insoluble organics from the marine surface layer into the marine aerosol.

## 3.2   Comparison with latitudinal distribution of other aerosol types

In Fig. 3, we show the zonal distribution measured by B73 together with modern data sources describing the year 2000 latitudinal distributions of various types of natural and man-made aerosols in the marine boundary layer, and our simulated distribution of biological sea spray IN. We note that a continental biological source is unlikely to contribute significantly to simulated biological IN abundance in marine air, except in continental outflow regions, where continental IN may outnumber marine IN (Fig. 7), but are in turn far outnumbered by dust IN.

### 3.3 Comparison of simulated marine biological IN and dust IN

Globally, desert dust is the primary and best-understood source of atmospheric ice nuclei, and has been shown to contribute to ice nuclei populations at great distances from the source regions (DeMott et al., 2003), although the uncertainty in both modelled
5 concentrations of desert dust and the ice nucleating fraction remains large. Comparing our simulated distribution of marine biological IN to simulated distributions of dust IN (Hoose et al., 2010a), we show (Figs. 3 and 4) that marine biological IN are most likely to play an important role in driving ice nuclei concentrations in remote marine regions that are less affected by the long-distance transport of continental dust and more af-
10 fected by sea spray generation due to strong surface winds, especially in the Southern Ocean, where rough seas are common and the biological content of seawater is high. The simulated vertical distribution at 60° S shows a decrease in the relative contribution of marine biological IN with altitude (Fig. 5). This is because marine biological IN, emitted from a local source, are present in highest concentrations in the boundary layer,
15 and decrease in concentration with increasing altitude. Dust IN, by contrast, originate from a distant source, and are this more well-mixed in the remote marine troposphere. For comparison, CLOUDSAT satellite data show that the majority of Southern Ocean clouds have cloud tops below 3 km (Mace et al., 2007).

We suggest that the Southern Ocean should be targeted by future field campaigns
20 aiming to investigate the possible role of marine biological particles as IN.

### 3.4 Estimated relative contributions to biological IN concentrations from marine and continental sources

To estimate the relative contribution to the biological IN population that can be expected from marine sources versus continental sources, we build on work by Hoose et al.
25 (2010b). We use concentrations of bacteria, fungal spores and pollen in CAM-Oslo from that work, and we follow the assumption that all pollen are potentially active as IN, while 1 % of the bacteria and fungal spores carry the gene for IN activity. We

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further assume that on average 10 % of these potentially IN-active bacteria and fungal spores can actually be activated (implicitly, at −15 °C), however, the small number of laboratory studies available indicate that this might range from 0.1 % to 100 % (Després et al., 2011).

5    The estimated relative contribution of marine biological IN is shown in Fig. 7. Because the concentrations of continental biological IN are lower than those of dust IN, their contribution to the global IN distribution is largely masked by dust. Including a continental biological IN source as part of the global total IN would not significantly impact the relative contribution of marine biological IN, as shown in Fig. 4 of the main text. As

10   a result, we do not consider the highly uncertain but very minor contribution of continental biological IN in our main line of analysis. The relative contribution of continental and marine biological IN could be constrained by a field campaign in an area where their contributions might be of similar order of magnitude; promising locations for such a campaign can be identified from Fig. 7.

15   **3.5   Potential implications for marine cloud brightening proposals**

Marine biological IN may also have previously unconsidered consequences for proposals to cool the climate by creating artificial sea spray that would increase the albedo of marine clouds (marine cloud brightening) (Latham et al., 2008), one of the leading proposed schemes for engineering a cooler climate. In one proposed implementation

20   of such a scheme (Salter et al., 2008), a fleet of wind-driven vessels would generate aerosol droplets from surface water at a mean rate of $1.45 \times 10^6 \, \mathrm{m}^{-2} \, \mathrm{s}^{-1}$ over a targeted ocean area of $7.72 \times 10^{10} \, \mathrm{m}^2$. Depending on the technical implementation of spray generation, organic materials might be more or less enriched in concentration during aerosolization (Fuentes et al., 2010). The system proposed by Salter et al.

25   (2008) would include an ultrafiltration system that presumably would remove all or most biological particulates from the generated sea spray. If biological particles were present in the generated sea spray, some fraction of generated particles could be biological IN.

The hardware design proposed by Salter et al. (2008) aims to generate a monodisperse spray of 0.8 µm, with a mean spherical volume of $0.27\,\mu m^3$. The mean volume emission flux of the injected spray in the targeted ocean regions would thus be $3.9 \times 10^5\,\mu m^3\,m^{-2}\,s^{-1}$ of surface seawater. The concentrations of biological IN in that seawater are roughly $(2.7\text{–}32) \times 10^6$ per mg POC or $(1.1\text{–}8.9) \times 10^9$ per mg Chl-$a$ (Table 1). The expected IN flux would thus be $(1\text{–}12) \times 10^{-6}\,m^{-2}\,s^{-1}$ per (mg POC $m^{-3}$ in surface water); or $(0.4\text{–}3.5) \times 10^{-3}\,m^{-2}\,s^{-1}$ per (mg Chl-$a$ $m^{-3}$ in surface water). Assuming a 5-day residence time in an 800 m boundary layer, an an enrichment factor of 1000 during the spray production process, this could result in concentrations between about 100 and 500 IN $m^{-3}$ over large areas of the ocean, which could be be competitive with naturally occuring concentrations in regions not influenced by mineral dust. This would depend greatly, however, on the engineering of a spray generating mechanism that allows organic particulates to enter the artificially produced sea spray at highly enriched concentrations.

## 3.6  Errors in membrane filter measurements and discussion of uncertainties

A complication in interpreting the data reported by B73 arises from the analysis method used in that early study. Particles were collected in the field on a membrane filter, stored, and later analyzed for IN content in the laboratory by exposing the filters to air of controlled humidity and temperature. Early membrane filter methods were subsequently shown to underestimate IN concentrations by a factor of about 10–100 compared to both a continuous flow diffusion chamber (Hussain and Kayani, 1988) and a recently developed method using particle sampling by electrostatic precipitation onto silicon discs (Klein et al., 2010). The major known reason for this underestimation is that IN and other hygroscopic particles compete for and rapidly remove the available water vapor, termed the "vapor depletion effect" (Lala and Jiusto, 1972; Huffman and Vali, 1973). However, underestimates are much smaller for low particle concentrations such as those found in remote marine air (Hussain and Kayani, 1988; Bigg, 1990), so we expect the underestimation to be roughly a factor of 10 for this data set, as indicated

in Fig. 6 by the grey shaded region.

Another source of undercounting in some measurements of IN counts on filter samples is a layer of Vaseline or oil applied to the filter in order to seal the filter pores and improve the thermal contact of the filter to its underlying cold base. The Vaseline or oil can evaporate if the sample is exposed to a vacuum during analysis, and subsequently coat and deactivate IN (Klein et al., 2010). However, the B73 samples were not exposed to a vacuum during processing, thus they were probably not affected by deactivation due to coating, and later experiments by Bigg did not find evidence for this effect (Bigg, 1990).

In addition to these issues, it should be noted that the filter method used in B73 can only capture deposition and immersion nucleation, and not the contact nucleation mode. Similarly, drop freezing measurements of IN concentrations, such as those we use as an estimate of IN concentrations in samples of particulates from a plankton bloom (Schnell and Vali, 1975), measure only the immersion mode of freezing. In spite of the limitations of the membrane filter measurements, the B73 dataset remains uniquely relevant as one of only a very small number of existing IN climatologies and to our knowledge the only such climatology in a marine region.

Given the likely large underestimate of IN concentrations in the B73 filter measurements, the question presents itself whether "true" concentrations a factor of ten greater than those reported by B73 can be explained at all with current models of dust and marine aerosol, since even the sum of these components would then not reproduce the peak zonal mean values in B73 (Fig. 6). Considering the large uncertainties discussed above, we conclude that the location and magnitude of the peak could be consistently explained either by dust or by a marine biological source, or by a combination of the two. A marine biological source is very likely relevant in a latitude band near the Antarctic coast, even if we have overestimated the role of marine biological IN relative to dust IN by an order of magnitude (Fig. 4). Only if our estimate of the proportion of biological to dust IN were incorrect by two orders of magnitude would we expect either dust or marine biological IN to entirely dominate marine IN populations everywhere.

In the absence of better information, we have implicitly assumed that marine IN (active at −15 °C) are equally distributed in marine biological matter around the world. It seems likely that marine IN activity is tied to particular plankton species or their exudates and its geographic distribution would be related to the distribution of that species. As a result, the relative IN activity of marine biological particulates might depend, for example, on sea surface temperature or light availability. This could be determined by new field experiments characterizing the IN activity of environmental samples of marine particulates from different locations. A challenge in such experiments is that IN testing must be performed rapidly following environmental sampling to avoid any biases arising from changes in biologically active samples during storage. The characterization of individual cultivates in the laboratory might also prove useful, especially if important candidates are previously identified in field experiments.

## 4 Conclusions

A commonly held and useful conception of global IN distributions is that IN are more numerous in continental than in marine air (Castro et al., 1998), and the IN population is mostly dominated by large mineral and dust particles (Chen et al., 1998). However, this conception is likely to be refined as new experiments reveal a more nuanced picture of the sources and nature of atmospheric IN. For example, biological particles contribute significantly to the IN concentrations in the clean air over the Amazon rain forest (Pöschl et al., 2010), a "green ocean" regime that shows many characteristics more commonly associated with the marine atmosphere (Williams et al., 2002). As we have shown, there is reason to expect that biological IN play a similar role in determining background IN concentrations in remote marine air, but likely with strong regional differences in importance, just as continental IN populations differ between regions dominated by dust and those dominated by biological particles.

The presence of ice in clouds is very important for the formation of precipitation and cloud lifetime, as well as for cloud radiative forcing and thus the Earth's radiation budget

(Choi et al., 2010), which is poorly predicted in the Southern Ocean by current models (Trenberth and Fasullo, 2010). IN concentrations are especially likely to impact mixed-phase clouds containing both liquid droplets and ice crystals, which are responsible for a large fraction of precipitation globally (Rasmussen et al., 2002). Cloud model simulations of marine mixed-phase boundary-layer clouds in the Arctic indicate that IN concentrations may strongly affect the formation of cloud ice and precipitation, thus affecting cloud lifetime, radiative forcing, and boundary-layer dynamics (Morrison et al., 2005; Prenni et al., 2007; Harrington et al., 1999; Harrington and Olsson, 2001). Model simulations suggest that higher numbers of IN could result in increased freezing of cloud droplets and thus optically thinner ice clouds, and affecting the boundary layer radiation budget (Prenni et al., 2007). Cloud ice plays a crucial role in the formation of precipitation, so a change in biological IN concentrations could affect precipitation and cloud cover in remote marine regions, modifying the hydrological cycle and the energy balance of the Earth. It is significant that the region in which biological sea spray is most likely to play a role in driving IN concentrations is the Southern Ocean. The Southern Ocean has a cloud cover fraction of about 80 %, and Southern Ocean clouds contain an unusually large proportion of supercooled liquid water droplets (Morrison et al., 2011). This likely indicates that droplet freezing is limited by the low availability of IN and these clouds are most likely to be sensitive to the concentrations of ice nuclei. Indeed, one of the few analyses that has found a statistically significant increase in precipitation due to cloud seeding with IN focuses on long-term experiments in Tasmania (Morrison et al., 2009).

Through their influence on the glaciation and microphysics of marine clouds, biological IN may affect climate in multiple ways that are relevant for our understanding of climate change and climate feedback mechanisms. Marine ecosystems are being significantly impacted by human activities and rising ocean temperatures, which could result in a decline in biological productivity and IN concentrations in seawater. On the other hand, in a warming climate wind speeds are expected to increase, resulting in greater emissions of sea spray, including biological IN. A targeted field campaign in the

Southern Ocean will be necessary to clarify the quantity and role of marine biological IN. Such a campaign should combine ship-based and, ideally, airborne measurements of air and water in the Southern Ocean, using recent state-of-the art instruments to measure IN concentrations and composition, as well as the overall dust, biological and organic aerosol concentrations. We suggest and intend to pursue further laboratory, field and model investigations of this challenging and rarely-studied topic.

**Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/12/4373/2012/ acpd-12-4373-2012-supplement.pdf**.

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**Table 1.** Scaling factors applied in estimation of sea spray IN concentrations.

| Factor | Range from literature | "Middle" value | "High" value | References |
|---|---|---|---|---|
| | | **Scaling of POC-driven emissions** | | |
| POM/POC | | **1.8** | **1.8** | (Facchini et al., 2008), (Russell et al., 2010) |
| IN[a] per mg marine POM | $0.1$–$20 \times 10^4$ | $\mathbf{10 \times 10^4}$ | $\mathbf{20 \times 10^4}$ | (Schnell and Vali, 1975) |
| Assumed ratio of total POM to submicron POM mass[b] | ca. 3.4 (range unknown) | **3.4** | **5.6** | (Facchini et al., 2008) |
| Correction for size of submicron POM[c] | ca. 2–16 | **9** | **16** | (Facchini et al., 2008) |
| IN per mg POC in water (product of above factors) | | $\mathbf{5.5 \times 10^6}$ | $\mathbf{32 \times 10^6}$ | |
| Enrichment factor | ca. 10 – 1000 | **500** | **1000** | (Hoffman and Duce, 1976), (Russell et al., 2010), Table B1 |
| IN per mg emitted POC (product with enrichment factor) | | $\mathbf{2.7 \times 10^9}$ | $\mathbf{32 \times 10^9}$ | |
| Maximum POM emissions as percent of sea spray[d] | | 76% | | (Russell et al., 2010) |
| | | **Scaling of chlorophyll-driven emissions (microorganism assumption)** | | |
| Bacterial cells per mg Chl-$a$ | $1 \times 10^{12}$–$7 \times 10^{12}$ | $\mathbf{4 \times 10^{12}}$ | $\mathbf{7 \times 10^{12}}$ | (Li et al., 2004) |
| Mass of marine bacterial cell (mg) | $50$–$100 \times 10^{-12}$ | $\mathbf{75 \times 10^{-12}}$ | $\mathbf{100 \times 10^{-12}}$ | (Ducklow, 2000), Table 2 |
| Ratio of POM to bacteria mass in accumulation mode aerosol | at least ca. 20–50 | **35** | **64** | (Bigg, 2007) |
| IN[a] per mg marine POM | $0.1$–$20 \times 10^4$ | $\mathbf{10 \times 10^4}$ | $\mathbf{20 \times 10^4}$ | (Schnell and Vali, 1975) |
| IN per mg Chl-$a$ in water (product of above factors) | | $\mathbf{1.1 \times 10^9}$ | $\mathbf{8.9 \times 10^9}$ | |
| Enrichment factor | ca. 10–1000 | **1000** | **500** | (Blanchard and Syzdek, 1982), (Aller et al., 2005), (Kuznetsova et al., 2005), Table B1 |
| IN per mg emitted Chl-$a$ (product with enrichment factor) | | $\mathbf{0.53 \times 10^{12}}$ | $\mathbf{8.9 \times 10^{12}}$ | |
| Maximum bacteria emissions as percent of sea spray[d] | | 10% | | |

[a] IN active at $-15\,^\circ$C. Lower value represents drop freezing measurements with concentrated plankton seivings in seawater; upper value represents the same using distilled water. Tests with seawater result in lower IN number counts due to freezing point depression from the salts in the seawater. In marine aerosol samples, certain biological particles such as bacteria are found to be separate from salt particles, while other marine particles contain a mixture of salt and organic matter (Bigg, 2007).

[b] Mass of OM in sea spray is dominated by coarse mode.

[c] Particles are modelled with assumed radius of 0.1 µm for accumulation mode, but mass-weighted mean radius for submicron mode is likely in the range $r = 0.125$–$0.25$ µm, this must be corrected for in calculating the mass density of particles from their number density.

[d] Maximum percent of sea spray mass in accumulation mode.

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**Table 2.** Size of marine bacterial cells. Unless otherwise stated, values are reported as mean ± standard deviation and/or as range. Values in bold are derived from the published values.

| Where | Cell volume $(\mu m^3)$[a] | Total mass $(fg\,cell^{-1})$[b] | Carbon mass $(fg\,cell^{-1})$ | Spherical equivalent radius $(\mu m)$ | Method/notes | Reference |
|---|---|---|---|---|---|---|
| Crane Neck, Long Island | 0.05 | **50** | 20 | **0.23 (0.20–0.26)** | Microscopic volume estimates of cultured cells from natural assemblages | (Lee and Fuhrman, 1987) |
| | (0.036–0.077) | | | **0.27–0.43** | As cited in (Christian and Karl, 1994) | (Bjørnsen, 1986) |
| Open ocean Coastal waters | | | 12.4 ± 6.3 30.2 ± 12.3 | | High temperature catalytic oxidation, used natural assemblages | (Fukuda et al., 1998) |
| Hawaii | | | 10 | | Solved a multiple linear regression using Chl-*a* and ATP data | (Christian and Karl, 1994) |
| Open ocean south of Bermuda | **0.08**[c] | **80** | 15 | **0.27** | Constrained by ancillary measurements, e.g. of PC, Chl, C : Chl ratios etc. | (Caron et al., 1995) |
| Arctic Antarctica Delaware Bay | 0.100 ± 0.04 0.106 ± 0.04 0.087 ± 0.06 | **100 ± 40** **106 ± 40** **87 ± 60** | | **0.29** **0.29** **0.27** | Protein + DAPI staining, automated microscopic counting | (Straza et al., 2009) |
| Ross Sea, Antarctica | 0.06–0.09 | **60–90** | 7–13 | **0.24–0.28** | Mass balance approach using high-precision analyses of DOC and TCO2 | (Carlson et al., 1999, as cited in Ducklow, 2000) |
| 80 lakes in the Pyrenees | 0.16[d] (0.09–0.45) | **160 (90–450)** | | **0.34 (0.28–0.48)** | Epifluorescence microscopy | (Felip et al., 2007) |
| Exponential growth C-limited N-limited P-limited | 1.78 ± 0.17 0.45 ± 0.06 1.40 ± 0.12 2.38 ± 0.38 | **1780 ± 170** **450 ± 60** **1400 ± 120** **2380 ± 380** | 149 ± 8 39 ± 3 92 ± 5 106 ± 23 | **0.75** **0.48** **0.69** **0.83** | Marine bacteria isolated and grown in cultures until nutrient-limited. X-ray microanalysis. | (Vrede et al., 2002) |

[a] Calculated values in bold are spherical equivalent volumes, except as noted.

[b] Calculated masses are based on the assumption that bacterial cell density equals 1 g per $cm^{-3}$.

[c] Calculated using the authors' conversion factor of 183 fg C per $\mu m^{-3}$.

[d] Median.

**Table A1.** Concentrations of IN measured in marine surface waters, selected values. tab:INinseawater

| Description | Value | Location | Reference |
|---|---|---|---|
| Active at $-7\,^\circ$C | | | |
| IN per $cm^3$ seawater | 2–20 | San Diego, California, USA | (Fall and Schnell, 1985) |
| Active at $-10\,^\circ$C | | | |
| Number of IN per µg of particulate matter sieved from a phytoplankton bloom | ca. 10–20 for concentrate in distilled water | Coastal waters near Bedford, Nova Scotia | (Schnell and Vali, 1975) |
| | ca. 0.1 for concentrate in seawater | | |
| IN per $cm^3$ of biologically active seawater | ca. 100–200 | Bedford, Nova Scotia | (Schnell and Vali, 1975) |
| IN per $cm^3$ seawater | 0.1–0.7 | Gulf of Mexico | (Rosinski et al., 1988) |
| Active at $-15\,^\circ$C | | | |
| IN per $cm^3$ seawater | 0.2–2 | Gulf of Mexico | (Rosinski et al., 1988) |
| Active at $-20\,^\circ$C | | | |
| IN per $cm^3$ seawater | 0.3–3 | Gulf of Mexico | (Rosinski et al., 1988) |

**Table B1.** Enrichment factors reported from field measurements and laboratory experiments for aerosolized marine bacteria and organic compounds. tab:enrichment

| Enrichment factor | Substance measured | Notes | Reference |
|---|---|---|---|
| **Enrichment factors for microorganisms** | | | |
| 1200<br>8 | *S. marcescens* suspended in distilled water | 0.38 mm bubbles, top jet drop<br>Fifth or lowest drop in the jet set | (Blanchard and Syzdek, 1978) |
| ca. 10–100<br>ca. 500–600<br>ca. 150–220<br>ca. 70–200 | *S. marcescens* suspended in pond water | 380 µm bubble rising < 1 cm, top jet drop<br>380 µm bubble rising > 5 cm, top jet drop<br>380 µm bubble rising ca. 100 cm, full jet set<br>380 µm bubble rising ca. 30 cm, full jet set | (Blanchard et al., 1981) |
| 10–20 | Suspension of *S. marcescens* in either pond water or distilled water | 1.7 mm bubbles rising 2 cm, film drops | (Blanchard and Syzdek, 1982) |
| 50–100 | Seawater suspension of *S. marinorubra* | Measured on drops < 10 µm | (Cipriano, 1979, as cited in Blanchard, 1989) |
| Mean: 10 (max: 22)<br><br>Mean: 6 (max: 10) | DAPI-stained bacteria in aerosol relative to bulk<br>DAPI-stained in sea-surface microlayer relative to bulk | Ambient samples collected near Long Island, New York, above plankton bloom | (Aller et al., 2005) |
| 37–2545<br>14–585 | Culturable mesophile bacteria<br>Culturable psychrophile bacteria | Bubble measurements with coastal seawater samples from the Gulf of Gdansk. | (Marks et al., 1996) |
| ca. 6–140<br>ca. 4–40<br>ca. 0.6–6<br>ca. 1–3<br>ca. 1–4 | *S. marinorubra*<br>*M. euryhalis*<br>*E. coli*<br>*P. bathycetes*<br>*B. subtilis* | Bubble-bursting with suspensions of bacterial cultures in 0.85 % or 3 % NaCl at 23 °C. EF is for top jet drop, and is larger for smaller drop diameters. For *E. coli*, EF also shown to depend on the culture age. | (Hejkal et al., 1980) |
| 2–27 | DAPI-stained bacteria | Bubble-bursting with samples in NW Mediterranean Sea and Long Island Sound | (Kuznetsova et al., 2005) |
| 2–14<br>0.2–0.6 | Pigmented *S. marcescens*<br>Non-pigmented *S. marcescens* | Bubble-bursting with suspensions in 0.7 % saline solution, bubble diameter 0.66–0.89 mm, top jet drop | (Burger and Bennett, 1985) |
| **Enrichment factors for organic compounds** | | | |
| $10^2$–$10^3$ | Organic mass in submicron aerosol | Enrichment relative to reported Na-to-OM ratios (field measurements). | (Russell et al., 2010) |
| 20–40<br><br><br>10 (max: 15) | Organic particles (polysaccharides, proteinaceous particles and amorphous DAPI-stained particles), 3–40 µm<br>Virus-like particles stained with SYBR gold | Ambient samples collected near Long Island, New York, above plankton bloom | (Aller et al., 2005) |
| $250 \pm 145$ (130–640); $N = 11$<br><br>$74 \pm 27$ (26–105); $N = 7$<br>$1460 \pm 1350$ (320–6080); $N = 7$ | Total organic carbon | Bubble generation, seawater from Narragansett Bay, Rhode Island, USA.<br>Bubble generation, seawater from Sargasso Sea<br>Ambient sampling, Bermuda | (Hoffman and Duce, 1976) |
| mean: 6810<br>median: 387; $N = 26$ | Soluble organic carbon | Bubble generation, seawater from Bermuda. | (Turekian et al., 2003)<br>(Keene et al., 2007) |
| 2–300 | Synthetic silica particles in seawater | Bubble-bursting experiments; EF increased with higher surfactant content | (Cloke et al., 1991) |
| 1.2–20<br><br>5–50<br>$57 \pm 29$<br><br>$44 \pm 22$<br><br>1–20 | Dissolved free amino acids and dissolved combined amino acids<br>Particulate amino acids<br>Proteinaceous particles stained by Coomassie Blue<br>Transparent exopolymer particles stained by Alcian Blue<br>Virus-like particles stained with SYBR gold | Bubble-bursting with samples in NW Mediterranean Sea and Long Island Sound | (Kuznetsova et al., 2005) |

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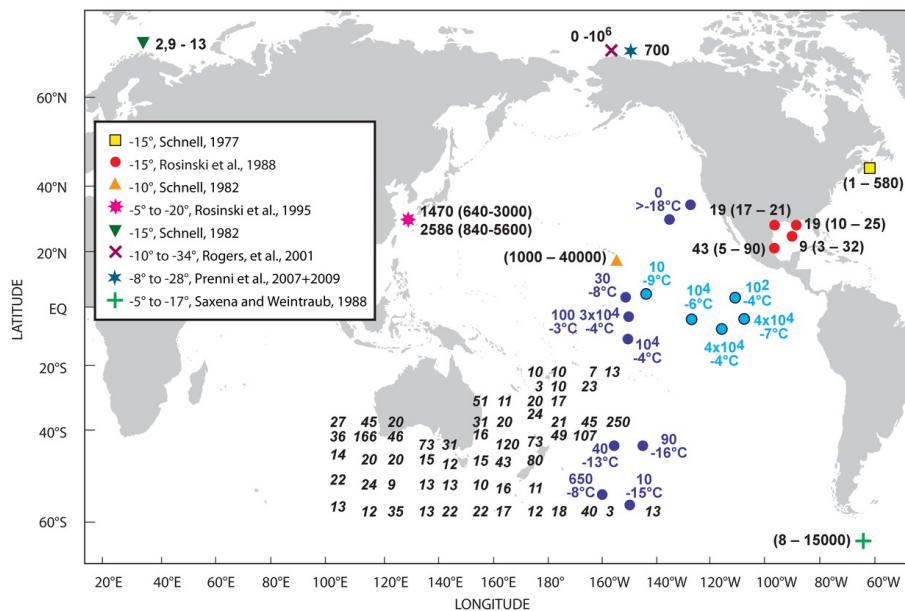Printer-friendly Version

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**Fig. 1.** Number densities of atmospheric ice nuclei (m$^{-3}$) in the marine boundary layer. *Measurements by condensation-followed-by-freezing, ship-based and coastal*: Australia (italic text): −15 °C and water saturation (Bigg, 1973); Pacific Ocean: 29 May–16 June 1985 (Rosinski et al., 1987). 17 February–29 March 1984 (Rosinski et al., 1987). Gulf of Mexico: (Rosinski et al., 1988), East China Sea (Rosinski et al., 1995). Arctic: (Bigg, 1996). Antarctica: (Saxena, 1983). Nova Scotia: (Schnell, 1977). *Airborne measurements by drop freezing:* Hawaii: (Schnell, 1982). *Airborne measurements by continuous flow diffusion chamber:* Arctic: (Rogers et al., 2001), (Prenni et al., 2009).
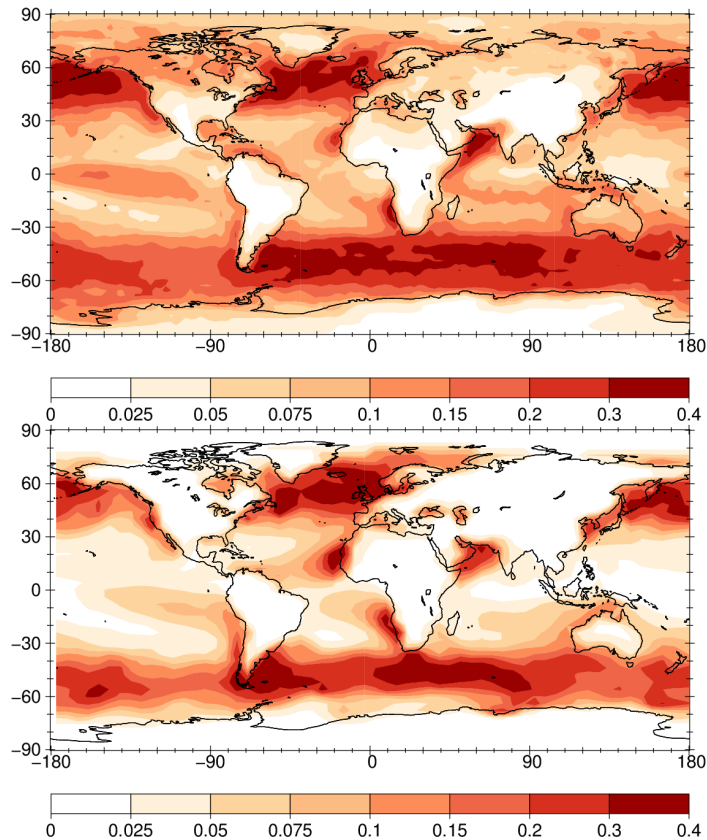
**Fig. 2.** Annual mean mass density of accumulation mode sea spray particulate organic mass (POM = 1.8*POC, $\mu$g m$^{-3}$) in the model surface layer. A: simulation based on MODIS-Aqua POC seasonal climatology (this work). B: modelled water-insoluble organic mass from primary sea spray emissions in the TM5 model (Vignati et al., 2010). Differences in the distributions result from differences in emissions, but also in loss processes and other model components.
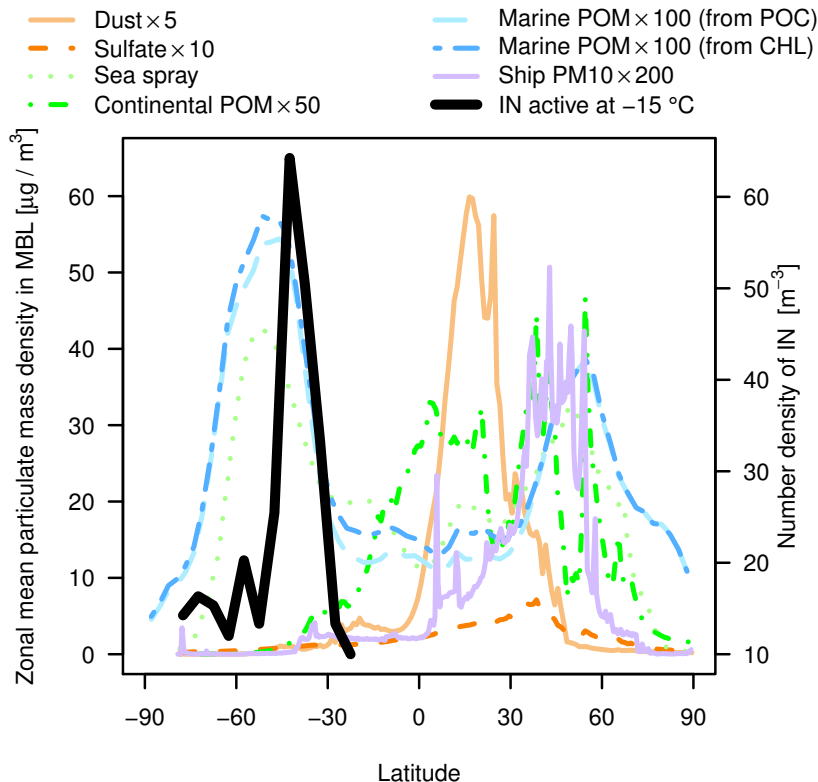
**Fig. 3.** Latitudinal dependence of aerosol surface concentrations over oceans. Left axis: mean surface mass densities, latitudinal average over oceans. Shown here are dust, sulfate, sea spray, and (continental) POM from the AEROCOM median model (year 2000) (Textor et al., 2006; Kinne et al., 2006; Schulz et al., 2006), simulated marine POM (this work) and estimated particulate matter from ship emissions (year 1970, $PM_{10}$, assuming 5-day residence time in 800 m boundary layer) (Lamarque et al., 2010). Right axis: observed number densities of IN active at $T = -15\,°C$, as reported by B73, latitudinal average over sampled region.

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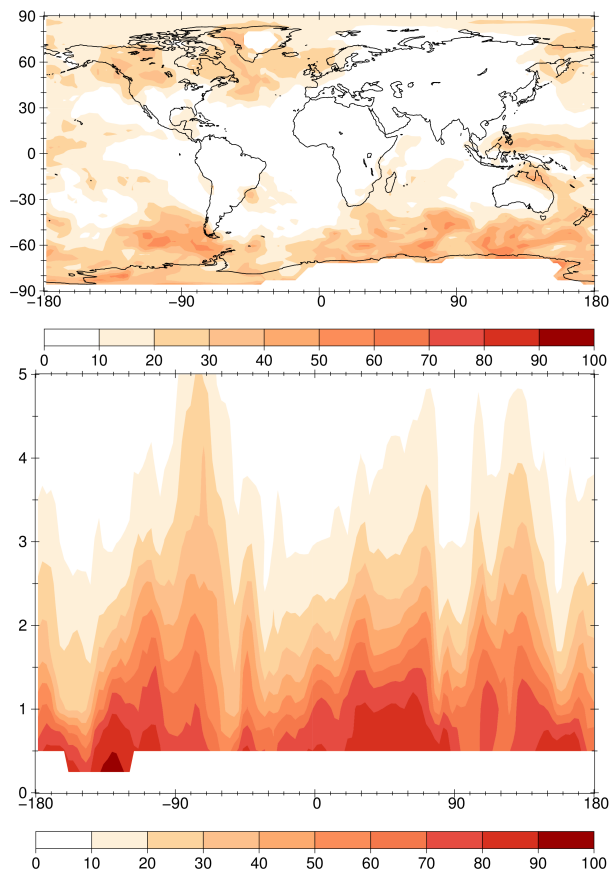
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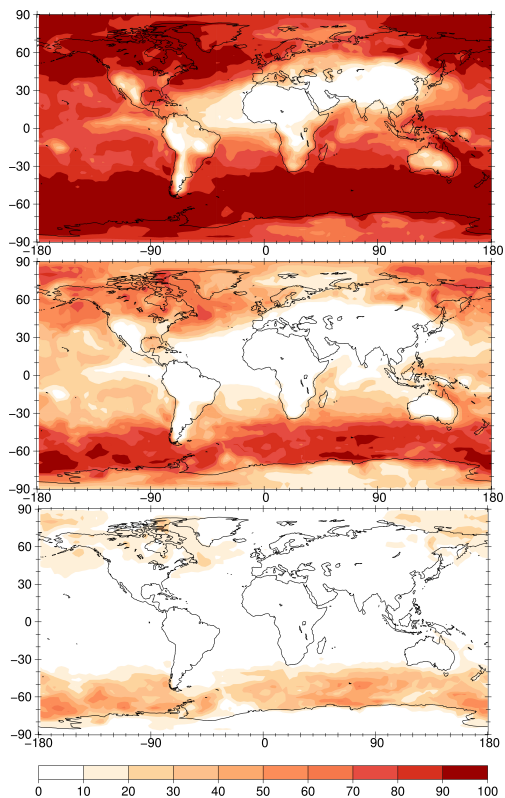
**Fig. 4.** Simulated relative contribution of marine biological IN at −15 °C, as a percentage of the sum of annual mean dust IN and marine biological IN. Middle: estimate using "control" dust IN concentrations and "high" biological IN concentrations; Top: with dust IN reduced (or, equivalently, marine biological IN concentrations increased) by a factor of 10; Bottom: with dust IN increased (or, equivalently, marine biological IN concentrations reduced) by a factor of 10. For zonal mean comparison of simulated marine IN and dust IN, see Fig. 6.

**Fig. 5.** Simulated relative contribution of marine biological IN at −15 °C, as a percentage of the sum of annual mean dust IN and marine biological IN. Top: horizontal cross-section at 2 km altitude. Bottom: Vertical cross-section at 60° S latitude.

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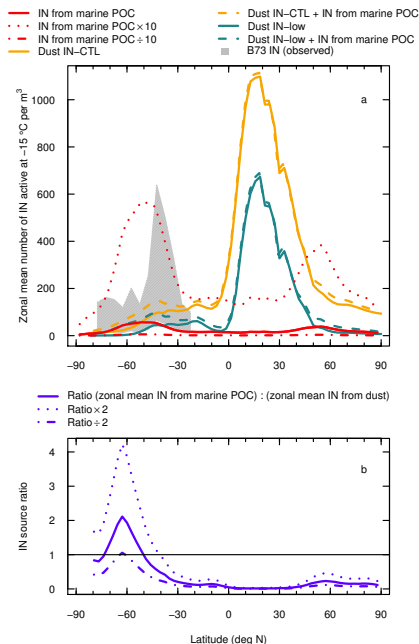
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**Fig. 6. (a)** Latitudinal dependence over oceans of observed IN, simulated marine biological IN, and estimated dust IN concentrations. Red lines: Results of simulations performed on the basis of POC climatological distribution from MODIS-Aqua (alternative results using chlorophyll climatology are shown in the supplementary material). Observed zonal mean IN number densities are from B73; the values as reported in B73 form the lower boundary of the shaded region, the "true" IN values are expected to be about a factor of 10 higher, indicated by the upper boundary of the shaded region. Dust IN concentrations are estimated from two cases of simulated dust number concentrations, a control simulation (CTL; orange solid line) and a simulation with lower dust concentrations (low; turquoise solid line) (Hoose et al., 2010a). The sum of predicted dust IN and marine biological IN is shown for the control dust and low dust cases by dashed lines. **(b)** Ratio of zonal mean marine biological IN to zonal mean dust IN, over oceans.
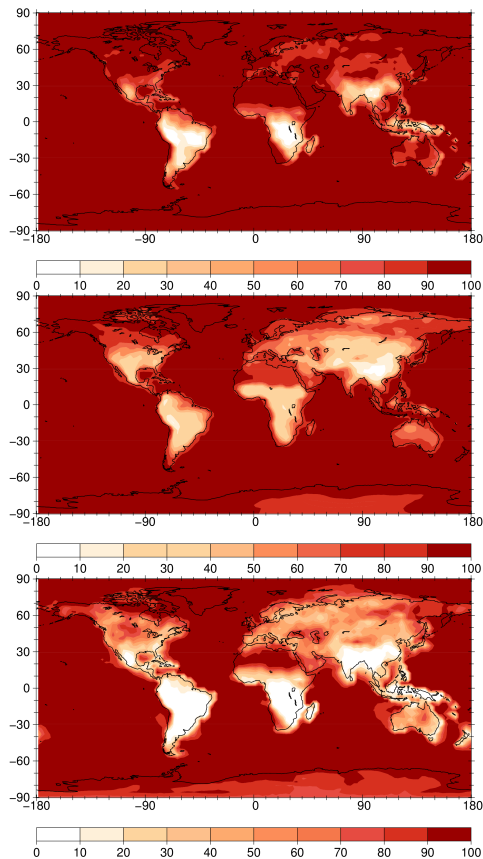
**Fig. 7.** Top: estimated percent of biological IN, active at $-15\,°C$, from a marine source, relative to the total population of biological IN (from continental and marine sources). Bottom: as above, but with marine source increased (or, equivalently, continental source reduced) by a factor of 10 (left), or with marine source decreased (or, equivalently, continental source increased) by a factor of 10 (right).

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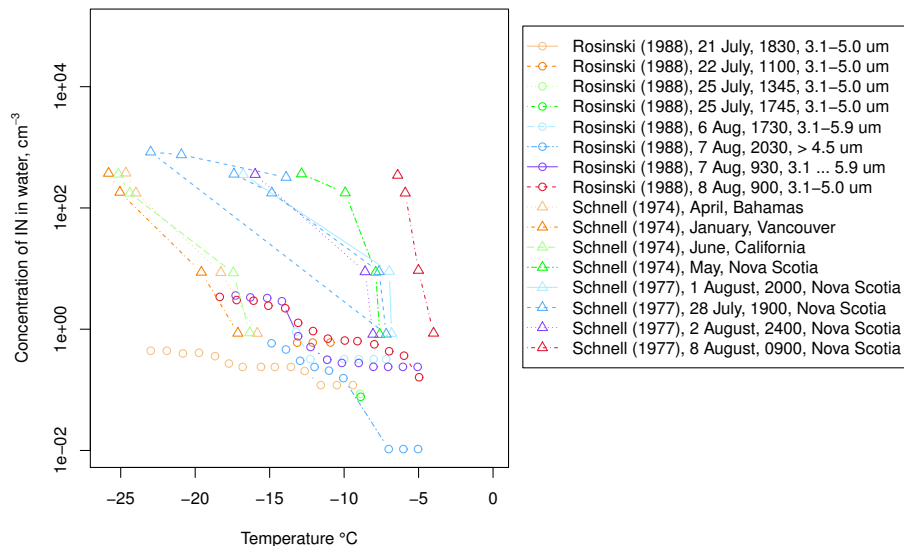
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**Fig. A1.** Ice nucleation spectra of particles separated from sea water by filtration. Rosinski et al. (1988) data (circles) are from the Gulf of Mexico, and IN concentrations were determined by condensation-followed-by-freezing using the dynamic developing chamber of Langer and Rodgers (1975). Results from (Schnell and Vali, 1975) (triangles) are from various locations as indicated, IN concentrations were determined using a drop freezing assay (immersion freezing).