Atmos. Chem. Phys. Discuss., 12, C9689–C9699, 2012
www.atmos-chem-phys-discuss.net/12/C9689/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ice nuclei in marine air: bioparticles or dust?" *by* S. M. Burrows et al.

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Received and published: 21 November 2012

Complete referee comments are included below in italics.

In this work the authors propose a new parameterization of biological ice nuclei (IN) emissions from marine biological sources, and use it to assess the importance of biological IN against dust IN in marine environments. The authors conclude that biological sources would dominate. Although somehow simplified, I find this work interesting and the issues it raises relevant for the scientic community. Therefore I recommend its publication in ACP after some comments have been addressed.

We thank the anonymous referee for the positive overall evaluation and comments on our manuscript. Our responses to specific comments follow.

As a general comment, the remarks of both referees led us to emphasize our aims and underlying assumptions more explicitly.

In the revised manuscript, we will add the following new introductory section 1.4 to clarify our aims:

"Aims and Approach

The aims of this study are 1. to use published observational data to estimate the global emissions of ice nuclei resulting from a hypothesized marine biological source, 2. to compare the near-surface-air concentration of marine biological IN to the simulated concentration of dust IN at the same temperature, and thereby 3. to identify regions in which marine biological IN are most likely to play a role in driving IN concentrations, relative to dust.

The hypothesized source is estimated using the following assumptions:

1. there is a primary source of ice nucleating particles to the atmosphere from sea spray,

2. this source is associated with biologically-derived material,

3. the concentration of marine biological IN in sea spray is proportional to the mass of marine biological particulates in sea spray."

1 General Comments

1- *The authors use retrieved POC and in-situ measurements of IN concentration to develop a parameterization of marine IN emissions as a function of POC. Then the simulated POC is used to estimate the IN concentration, which by design is in agreement with the in situ data. Instead of going throught this circular procedure wouldn't*

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it be easier just to directly compare the estimated dust-IN concentration against the in-situ data for marine IN? If the idea is to simulate the global distribution of marine IN where in situ data is not available then this should be validated using an independent data set. Is the simulation able to reproduce at least qualitatively the distributions of Figure 1? Why the other data sets are not used to validate the simulation?

A direct comparison of the dust IN concentration with the in-situ data for marine IN is shown in Figure 6 (zonal mean). At first glance, the simulated dust IN concentrations appear high enough to explain the observed IN counts, but the in situ IN concentrations are probably underestimated by a factor of ten or more (see text), and simulated dust mass concentrations in this region are typically overestimates by a factor of ten or more (Huneeus et al., 2011). So the dust may not be enough to explain the observed IN concentrations in the MBL.

By introducing a parametrization of hypothesized marine IN emissions, we can make use of spatial information about the generation of sea spray and distribution of biological activity to predict where biological IN are most likely to be important, and put them "on the map". Using this approach, we extrapolate what we infer from observations about marine IN, and estimate the spatial distribution of IN that would result globally.

There are multiple difficulties involved in using the other data sets (shown in Figure 1) for intercomparison. One major difficulty is that these datasets used a variety of different experimental techniques, so that the data from different studies are not intercomparable. A second difficulty is that the B73 dataset is climatological, with data averaged spatially over 5-by-5 degree intervals, and over a period of three years. The other data sets, by contrast, are from individual short-term field campaigns, which makes those data less suitable for direct comparison with any simulation results global model (although the experimental methods used in later studies were much improved).

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Interactive Comment

2- *The fact that marine IN concentration could be as high as dust IN concentration over the Southern ocean does not neccesarily mean that they would be as important for cloud formation. Biological particles can be easily incorporated into cloud droplets and be scavenged out before they can reach low temperature. On the other hand, free tropospheric IN may be more likely to inuence cloud formation at low T. This needs to be more clearly stated in the paper.*

This is an important point, which we do not address in this study. We do not attempt here to assess cloud and climate impacts of marine biological IN, and indeed to do so would be premature. Instead, we have the more limited goal of attempting to assess their likely distribution, which could be an important step towards a future assessment of their climate impact.

We note that we have already addressed this issue in Figure 5, which shows that the relative contribution of the marine biogenic IN would decrease with altitude, relative to dust IN, such that the relevance might be high near the surface, but still very low at 5 km. Since Southern Ocean clouds in particular usually have cloud tops below 3 km, any particles emitted there do not have to get very high up to potentially impact clouds.

However, this point could be emphasized more clearly and sooner in the text. To do so, we will add the following statement to section 1.4 of the revised manuscript (following the summary of aims and approach):

"We deal here with the concentrations of IN in near-surface air. However, we note that particles emitted from the surface are removed more rapidly from the atmosphere than particles transported from distant sources at higher altitudes (Figure 5). As a result, at cloud altitude the relative contribution of marine biogenic particles to the total particle population and the IN population will be reduced. An evaluation of cloud impacts is beyond the scope of this paper, but a potentially interesting topic for future study."

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Also, the variability both in marine emissions and in long-distance transported dust concentrations would be very high; since we examine averages, it is likely that local emissions would dominate the IN population at certain times, while long-distance transport would dominate at other times (in analogy to the observations over the Amazon rainforest by Prenni et al., 2009). We will add a comment to this effect near the end of Section 3.3.

It also seems that the supplemental material is quite relevant for the discussion. The authors may consider including it into the main text.

3- *The simplifications behind Equations 2 and 3 should be clearly stated. I would think that a general expression* α *= f(P OM, P OC, EF, Seasalt,T, RH . . .) is more appropiate as there is no reason to believe that the relations are linear. Then some assumptions may be introduced to arrive to simple expressions like Eqs. 2 and 3, but the procedure must be explicitly shown. I also nd puzzling that the authors didn't consider temperature and relative humidity as determinant factors to calculate* α *. If only immersion freezing is considered then it must be clearly stated (as condesation and deposition would depend on RH). Still the temperature dependency is missing, which is clearly signicant, as shown in Figure A1.*

It is not known in general which marine particles produce IN activity, nor how this activity varies with temperature and relative humidity. We did include an overview of some relevant measurements of the IN activity of marine particulate matter (collected from the source by filtration) as a function of temperature in Figure A1. There are only a small number of independent observations, and the differences are quite large between the magnitudes of IN activity shown in various observations.

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The development of a parametrization for the dependence of marine IN activity on temperature and relative humidity is a challenging and potentially fruitful topic for future study, but goes beyond the scope of this manuscript.

By choosing a single temperature for comparison, we are able to make use of available observations to focus on an estimate of the geographic distribution and number concentrations of marine biological IN, and to compare this to observed average IN concentrations, and to a simulated geographic distribution of dust IN. Our choice of -15°C was guided primarily by the use of -15°C as a reference temperature for presentation of observations by Bigg (1973).

2 Specific comments

Page 4374. Line 13. I think the analysis shows that IN emissions from biological sources can be as high as from dust sources, however whether IN concentrations are actually determined by them is a different issue (See comment 1 above).

We will the phrase **near-surface-air** here since this is what we deal with, rather than concentrations at cloud altitudes:

"Our analysis suggests that marine biological IN are most likely to play a dominant role in determining **near-surface-air** IN concentrations over the Southern Ocean..."

See also our response to general comments 1 and 2 above.

Page 4378. Line 15. Why is this particular temperature selected?

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In a revised version of the manuscript, we will add the following text at the beginning of the methods section to clarify our approach:

"For the purposes of comparing the geographic distribution with observations and with dust, we chose to estimate the distribution of marine biological IN at a single temperature. We chose -15 ℃, the temperature at which the geographic distribution of IN concentrations from B73 is presented. However, the source of marine IN has not yet been unambiguously identified and the relationship between temperature and IN activity in marine surface water samples is not yet clear from currently available data (Table A1 and Figure A1). Given the limited data, we did not feel justified in assuming a temperature dependence of the ice-active fraction, although we note that B73 observed IN concentrations at -10 ℃ to be about one order of magnitude lower than at -15 ℃, which is roughly consistent with the experiments of Schnell and Vali (1975); dust IN activity also is observed to decrease by approximately an order of magnitude over the same temperature range (Niemand et al., 2012). Similarly, the dependence of IN counts on atmospheric relative humidity is beyond the scope of this work."

Page 4378. Line 24. Typically, the IN concentration changes several orders of magnitude with temperature, using T = |15° may overestimate IN at higher T by much more than a factor of 10 (as for example shown in Figure A1).

See above comments.

Page 4380. Line 4. The rationale behind these equations is not clear, and must be clearly stated.

Page 4383. Line 8. I am not sure what T42L90 means, but if it is 4° by 5° it may be too

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coarse. This must be justied.

T42L90 means a horizontal grid spacing of about 2.8° and 90 vertical levels. In view of the limited knowledge, this fairly coarse resolution is sufficiently precise for the current study. By way of comparison, the B73 data were averaged on a 5° by 5° grid.

Page 4385. Lines 14-18. This statement is confusing. Please rephrase for clarity.

Manuscript text:

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, 100nm 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 100nm 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.

Page 4386. Lines 6-9. This is agreement by design. B73 data were used to generate the IN emissions parameterization. Is there an independent data set that can be used for validation?

Please see discussion above.

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Page 4386. Line 13. This is confusing. First, I am not sure what the authors mean by the absolute value of the IN concentration. Second I don't understand how it can be consistent with a large uncertainty. Please rephrase.

Page 4386. Lines 14-16. A plot with the spatial distribution of IN concentration would better support this statement. Also Figure A1 shows a T dependency of IN concentration which the authors largely neglect in this work.

Please see discussion above regarding the T dependency of the IN concentration.

On page 4386, lines 14-16, we state:

"The absolute value of the simulated IN concentrations is consistent within the large uncertainties both with the B73 measurements and with knowledge about the concentrations 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."

Page 4387. Lines 7-8. It must be clearly stated that this refers to the marine contribution to IN total emissions, regardless of whether they are actually incorporated in a cloud. The actual effect can be different since marine IN are emitted from below (as oppose to dust IN which come from above) and therefore more easily scavenged by liquid activation processes.

We will add the following text to Section 3.3 to :

"... so biogenic particles do not have to reach very high altitudes in order to potentially impact clouds. While we consider average concentrations here, since the variability

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both in marine emissions and long-distance transported dust concentrations is very high, local marine emissions could dominate the cloud-height IN population at certain times, while long-distance transport dominates at other times (in analogy to the observations over the Amazon rainforest by Prenni et al., 2009)."

Page 4387. Line 27. Why 1%? Is there an study to support this?

The assumption that 1% of the bacteria and fungal spores carry the gene for IN activity.derives from Hoose et al. 2010b.

Page 4388. Line 9. What do you mean by the main text?

This was a typo, we thank the referee for pointing it out and will correct it in the revised manuscript by deleting the words "of the main text".

Page 4389. Line 9. These statement implies several assumptions that must be clearly stated. The impact of marine IN emissions would be signicant only during the Austral winter when the temperature is low enough to render signicant ice nucleation in the 800 m boundary layer. Also mixing with sulfate would make these particles good CCN and limit their effect as IN. The latter process could also deactivate the IN active sites.

Again, we attempt here only to simulate IN counts as they would be observed at a standard temperature of 15° C, and we compare these with simulated dust IN counts at the same temperature.

We will include a short discussion of these issues at the end of Section 3.3.

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Page 4390. Line 12. Is there a reason to believe that contact nucleation with biological IN may be effective in this case? Actually I would imagine that if the biological IN are hygroscopic (which they tend to be) then at the moment of contact they would be coated with water. Thus, the surface tension difference between the droplet and the IN, which is believed to enhance ice nucleation activity in contact as compared to immersion freezing [e.g., Tabazadeh et al., 2002], would be small and marine IN may not be more effective contact IN than immersion freezing IN.

We concur that biogenic IN will likely be hygroscopic and would likely be coated with water, which is readily available in the marine boundary layer. So, we would expect the immersion nucleation mode to be the most important. We mentioned the exclusion of the contact mode in filter measurements because it was a concern raised in comments on an earlier version of this manuscript.

We will add a sentence noting this issue:

"However, we expect that since marine biogenic particles are likely hygroscopic and exposed to sufficient water vapor in the marine boundary layer, they likely to be coated with water nearly permanently, thus reducing the significance of the contact nucleation mode with respect to the immersion nucleation mode."

3 References

A. Tabazadeh, Y. S. Djikaev, and H. Reiss. Surface crystallization of supercooled water in clouds. 2002. PNAS 99 (25) 15873-15878; doi:10.1073/pnas.252640699

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 4373, 2012.