Atmos. Chem. Phys. Discuss., 11, C9879–C9898, 2011
www.atmos-chem-phys-discuss.net/11/C9879/2011/

Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Effect of primary organic sea spray emissions on cloud condensation nuclei concentrations" *by* D. M. Westervelt et al.

D. M. Westervelt et al.

dwesterv@andrew.cmu.edu

Received and published: 7 October 2011

Reviewer comment:

This paper employs a respected model approach to assessing the potential impact of possible marine organic (OC) emissions on resulting marine CCN. The objectives are of clear significance to the fields of aerosol, clouds and climate and should be of broad interest. However, my comments below speak to my perceived shortcomings of the paper that I feel limit its potential value. The key concerns are: 1) in using a prescribed dependency of OC on Chl as if it were fully established and accepted. 2) assuming primary OC is dominant in all cases (based upon one reference) instead of allowing SOA to play a role and (3) prescribing primary OC as interchangeable with sea-salt in flux formulations and as internally mixed aerosol. These constraints then yield results that the authors demonstrate are inconsistent with observations (by a factor of 5). However, although the authors indicate a possible shortcoming of the prescribed dependency, the flexibility of the model is not explored to try to understand these differences or examine the sensitivity to underlying assumptions. Even so, the authors in their opening line claim the model to “quantify” the marine OC source and its impact on CCN. While their conclusion that marine OC has little influence on global CCN may be true, I feel more should be done to address the issues raised before this quantification can be claimed.

Response:

We appreciate the reviewer’s positive feeback and criticisms. We agree with many of the perceived shortcomings mentioned, and outline our additions and responses to them below.

1) We agree that our discussion of the O’Dowd source function should have focused more on key uncertainties surrounding its applicability. We do not mean to imply that the source function employed is fully established. The dependence of OC on Chl (O’Dowd et al. 2008, updated by Vignati et al. 2009), although problematic, was state of the art at the time this work was begun. The original manuscript outlines the Roelofs (2008), O’Dowd et al. (2008), Gantt et al. (2009), Vignati et al. (2010), and Myriokefalitakis et al (2010) parameterizations in paragraph 2 of the introduction (pg 5759-5760 of the ACPD document). In addition to that text, we have added some more discussion of various source functions and their shortcomings just prior to the aforementioned section:

O’Dowd et al. (2008) developed a source function for primary organic sea spray based on measurements at Mace Head, Ireland. Their empirical relationship connects organic mass fraction in the sea spray aerosol to chlorophyll-a concentrations retrieved by Sea-WIFS (http://oceancolor.gsfc.nasa.gov/SeaWiFS/). The correlation coefficient between organic fraction and chlorophyll-a concentrations is equal to 0.3. A similar correlation has not been observed for data in the Southern Ocean at Amsterdam Island (Claeys et al. 2008). Despite these shortcomings, the O'Dowd et al. (2008) source function remains one of a few viable options for emissions of marine organic carbon aerosol, and has been implemented in at least two large scale models of the atmosphere (Lapina et al., 2011; Vignati et al., 2010). Recently, Gantt et al. (2011) developed a multi-parameter source function and tested it in Meskhidze et al. (2011). In this function, marine organic aerosol emissions are dependent on chlorophyll-a concentrations, wind speed (U10), and particle diameter and are fit to a logistic function. The authors tested three ocean biological activity proxies: chlorophyll-a, dissolved organic carbon (DOC), and particulate organic carbon (POC). Of the three, chlorophyll-a performed the best. Although the use of chlorophyll-a as a biological activity proxy may be problematic, it appears to be one of the best options currently available. The present study and others like it serve as a preliminary test for the O'Dowd et al. (2008) source function. Although we require more ambient datasets for a full assessment, by implementing the source function into a global model and comparing to observations, we test the applicability of the Mace Head source function for predicting OC elsewhere.

This new discussion in the revised manuscript acknowledges the emergent nature of the various source functions

2) We agree that more evidence in the form of citations is needed with our claim that primary OC is more important than secondary. We also cite and acknowledge one study that did find isoprene SOA to be an important source of marine organic aerosol and cloud condensation nuclei. To further establish our comment on the dominance of primary OC, we add several sources: Arnold et al. (2009); Ovadnevaite et al. (2011); Myriokefalitakis et al. (2010); Claeys et al. (2010). This can be found in the first paragraph of the introduction (page 5758 line 24 through page 5759 line 5 of the ACPD manuscript) and is pasted below as well:

Meskhidze and Nenes (2006) found that cloud effective radius was reduced by 30% over a phytoplankton bloom in the Southern Ocean and attributed this decrease to

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

marine isoprene emission and SOA formation. Although oceanic phytoplankton emit several types of volatile organic compounds (VOCs) with isoprene being the major constituent, secondary organic aerosol (SOA) derived from marine sources of isoprene is now thought to comprise less than 10% of the observed total marine organic aerosol (Arnold et al., 2009, Claeys et al. (2010), Myriokefalitakis et. al (2010), Ovadnevaite et al., 2011).

3) The reviewer correctly points out that our original manuscript assumed that marine POA displaces an equal mass of submicron sea salt emission. Although this is physically plausible, it is also plausible that the marine organic emissions occur in addition to the sea salt. Unfortunately, the current measurements, based on the ratio of organics to sea salt, do not help distinguish between these possibilities. The OC replacement method was chosen for our work and we did not provide enough justification or explanation of that choice. We also have now performed extra simulations where we use the "addition of OC" approach. Those results are now in the revised manuscript results (Sects. 3.2 and 3.4) and conclusions (Sect. 4) sections, not quoted in detail here. The abstract of the revised manuscript has been updated to reflect the new results as follows:

To test the sensitivity to uncertainty in the sea spray emissions process, we relax the assumption that sea spray aerosol number and mass remain fixed and instead can add to sea spray emissions rather than replace existing sea salt. In these simulations, we find that marine organic aerosol can increase CCN by up to 50% in the Southern Ocean and 3.7% globally in the austral summer. This vast difference in CCN impact highlights the need for further observational exploration of the sea spray aerosol emission process as well as evaluation and development of model parameterizations.

Also, we have added discussion (Section 2.3.1 regarding the source function) to highlight these uncertainties:

It is currently unclear whether marine OC emissions displace sea salt, completely or

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

partially, or occur in addition to sea salt. Physically, it depends on how organics affect the volume and composition of material ejected by wind-generated bubble bursting. We assume that OC replacement is more likely to happen, but test both methods as mentioned previously. Because the organics are likely sitting at the air-surface interface as either a microphase or a surface active monolayer and are not fully dissolved into bulk seawater, the volume mobilized by wind-generated bubble bursting will include a portion consisting of OC with little or no sea salt. In this scenario, the oceanic organic matter displaces some of what current sea salt parameterizations consider to be only sea salt and water, and thus the combined organic-inorganic source function should subtract sea salt and add OC. However, because seawater is dilute, OC probably does not completely replace sea salt on a 1:1 basis. Oppo et al (1999) gives a theoretical analysis of what the organic monolayer enrichment might look like.

Other comments:

We would like to point out that, although the reviewer is correct that our model-measurement discrepancy is sometimes as large as a factor of 5, it most often is not as large. Except for January and February (months in which biological activity is low anyway), the model is within a factor of 2 to 3 of the observations at Mace Head year round. This is a vast improvement over the factor of 10 underprediction that the model shows for the case without marine OC for all months except January and February. For Amsterdam Island, the comparison is less accurate, which is not surprising since the correlation between chlorophyll and organic aerosol was not found in that location, as the reviewer mentions. However, for January, February, March, and December, the model over predicts by a factor of 2 or less. Only in September, October, and November does the model-measurement disagreement become as large as a factor of 5. Compared to a more across the board underprediction of a factor of 10 in the original model, we have some improvement. The model does somewhat better during the biologically active periods and less well during the inactive periods.

Given that both reviewers perceived poor model-measurement agreement, we have

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

now added analysis that quantifies improvements and remaining discrepancies. This has been added to the manuscript in Sect. 3.3 and briefly summarized here. For Mace Head, the log mean normalized bias (LMNB) averaged over the full year is 0.36 with marine organics, compared to -1.26 in the base case (no marine organics). At Amsterdam Island, the LMNB is 0.51 with marine organics, compared to -0.79 without marine organics. The correlation coefficient values show improvement at Mace Head (-0.115 to 0.248 without and with marine organics, respectively) but not at Amsterdam Island, indicating that the model is just beginning to capture the seasonal trends at Mace Head and not capturing it at all at Amsterdam Island. Although the LMNB and correlation values do not indicate the strongest agreement, when compared to concentrations in the model without the marine organic source function, there is significant improvement.

Regarding the reviewer comment about model sensitivity and flexibility, we have taken some measures to address this request. First, we have added a set of simulations testing the “addition” of OC method instead of the replacement, as mentioned previously. Even within the original manuscript, the flexibility of the model is tested with the different aging scenarios. It is also tested with respect to organic aerosol surfactant effects. Other reasons for performing sensitivity studies, such as newer source functions, were not available at the time this work was begun.

Regarding the opening sentence of the abstract, we agree that “quantify” was too strong of a word. We will replace with “estimate”. The manuscript now reads:

This work estimates the primary marine organic aerosol global emission source and its impact on cloud condensation nuclei (CCN) concentrations by implementing an organic sea spray source function into a series of global aerosol simulations.

Reviewer comment:

Abstract P1 L2 Authors say: “This work quantifies the primary marine organic aerosol global emission source and its impact on cloud condensation nuclei (CCN) concentrations by implementing an organic sea spray source function into a series of global aerosol simulations."

Response:

Addressed above. All instances of "quantify" are replaced with "estimate".

Reviewer comment:

Pg 5762 section 2.2.2 Although carbonaceous sources are mentioned here they are not discussed or compared to marine sources in the model or discussion. The Mace Head combustion criteria for clean air used to be black carbon below about 40ng/g. This is not very clean by some standards. Transport of continental aerosol in the FT and entrainment into the MBL is also well recognized. Some discussion of model observations is merited in this context given apparent poor agreement using the marine OC source function (see comments below).

Response:

We agree that transport of continental aerosol in the free troposphere can influence organic aerosol concentrations at marine locations. Our model shows about 20% organic accumulation-mode aerosol composition by mass at Mace Head in the absence of a marine organic aerosol source. We can get up to 35% organic composition with a marine organic source. For comparisons of carbonaceous aerosol mass to observations (both black carbon and organic carbon) in GISS-TOMAS, we refer the reader to Pierce et al. (2007), who performed this analysis and found good agreement between modeled and measured BC including at marine sites. That work reports a -0.19 log mean normalized bias (LMNB) between simulation and observed black carbon.

In order to elucidate issues of transport and anthropogenic influence, we have modified Figure 1 such that it now shows: sea salt mass concentrations, OC mass concentrations from non-marine sources, and OC mass concentrations from marine sources. Previously, we did not show the OC mass concentrations from non-marine sources. We feel that having both marine and non-marine OC side by side will help readers

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

appreciate the relative contributions of these sources.

Reviewer comment:

P5759 L5 Comment on the apparent contradiction between – “Primary marine organic aerosol exists predominantly in the Aitken and accumulation mode with less than 5% of their mass existing at sizes greater than 1 μm (O’Dowd et al., 2004).” and the referenced result also based upon (O.Dowd): “Gantt et al. (2009) estimated a submicron and supermicron source of 2.9 and 19.4 TgCyr-1, respectively, again using the O’Dowd et al. (2008) source function and chlorophyll-a concentrations retrieved via remote sensing.

Response:

The sentence immediately following Gantt et al. (2009) already mentions potential bias with using the O’Dowd (or slightly modified version) source function for coarse mode emissions. In addition, the following sentence will be added:

Because sea spray exists primarily in the coarse mode, even small fractions of organic mass can result in large absolute quantities of organics in the coarse mode.

Reviewer comment:

P5760 L13 Authors say (part 1) ” Assuming that aerosol size and composition do not change, higher amounts of aerosol lead to higher cloud condensation nuclei (CCN)concentrations, (part 2) perturbing climate by brightening clouds, which is known as the first aerosol indirect effect (Twomey, 15 1977)”. Part 1 is true if “higher amounts” means “greater mass” although it is seldom justified as adding aerosol mass via gas to particle conversion commonly leads to growth of internal mixtures and not directly into new number. Part 2 is true – but as stated but the sentence implies Twomey also claimed part 1 as true – which he did not.

Response:

The citation is now separated to only apply to part 2 as defined by the reviewer above. The manuscript now reads:

Assuming that aerosol size and composition do not change, enhancements in aerosol number and mass lead to higher cloud condensation nuclei (CCN) concentrations. Higher amounts of CCN perturb climate by brightening clouds, which is known as the first aerosol indirect effect (Twomey, 1977).

Reviewer comment:

P5763 L17 How reliable and how sensitive are results to the scavenging assumptions used?

Response:

The reviewer brings up a point here that is applicable to any global aerosol modeling study. CCN results are always very sensitive to wet scavenging treatment in global models. Wet scavenging in GISS-TOMAS has been implemented by past studies and is outside the scope of the present study. See Adams and Seinfeld (2002), Adams and Seinfeld (2003), Pierce and Adams (2006), Lee and Adams (2009).

The percent changes in CCN presented in the paper (Figure 3 in original manuscript) are probably insensitive to scavenging because both the marine organic enrichment and sea salt only simulations being subject to the same scavenging scheme. Additionally, wet scavenging rates can vary by as much as a factor of two across different models and measurements (Textor et al., 2006). As the reviewer points out, the model-measurement comparison presented in this work is sometimes worse than that. In those cases, accuracy of the marine organic source function (and not choice of wet scavenging scheme) is likely the primary factor contributing to the discrepancy.

Reviewer comment:

P5763 Eqn1 - Comment on the justification for the intercept of 10 for zero Chl? Some recent observations indicate a slope through zero would not be unexpected.

Response: We agree that a slope of zero might not be unexpected. However, this is how the O'Dowd correlation is formulated, which was state of the art at the time this work was underway. In response to the reviewer comment, we have performed additional simulations where we use a zero intercept. This simulation has CCN impacts that are virtually identical to those shown in our original base case. Thus, the zero intercept modeling results are not shown.

Reviewer comment:

L11 The source function was applied globally to the existing sea-spray emissions parameterization in the GISS-TOMAS model, described by Clarke et al. (2006) and reviewed in Sect. 2.2.1 (Clarke et al., 2006; Gong, 2003; Martensson et al., 2003; Monahan, 1968, 1986). Confusing as to what the references are referring to?

Response:

We apologize for the excess references. Manuscript has been updated to include only the Clarke reference.

Reviewer comment:

P5762 section 2.2.2 While reference to combustion/continental aerosol is made and it is know to be effectively transported across the Atlantic (available to be entrained into marine boundary layer en route) it is hard to determine how well this source of OC is being included and tracked in the model. If it is being carried by the model, greater attention to the relative role of this source should be included in the discussion.

Response:

A very similar comment was made earlier and is addressed above. See, especially, that Figure 1 now shows a breakdown of OC mass concentrations attributable to non-marine and marine sources.

Reviewer comment:

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

P5769 Model evaluation-Fig.2 Although an apparent relationship between biological production (chlorophyll a concentrations) and organic aerosol was established at Mace Head, Ireland (O'Dowd et al., 2008), it has not been clearly evident at other sites. Claeys et al., 2010 found organic aerosol concentrations on the order of 0.1-0.2 ug m-3 at Amsterdam Island, despite chlorophyll a concentrations similar to those found during periods of high biological production in the North Atlantic, where organic aerosol was measured at 2-4 times higher concentrations (0.4-0.8 ug m-3). Even the Mace Head organic aerosol concentrations are variable at given chlorophyll a concentrations: i.e., during periods of low biological production (chlorophyll concentrations between 0.2-0.3 mg m-3), % organic mass ranges from 10-60% (O'Dowd et al., 2008).

Response:

This comment is very similar to an earlier comment regarding how well established or widely applicable the Mace Head relationship is. The issues the reviewer brings up are addressed previously in this response and in revisions to the manuscript. We agree with the reviewer that there are shortcomings and issues associated with the observed correlation and source function developed from Mace Head data. The correlation was not found to be particularly strong to begin with even at Mace Head, and it has not always been observed at other sites. A comparison of the different source functions can be found in the introduction section of the original manuscript, and a new discussion has been added as is mentioned above. In short, we have updated our manuscript to lessen the implication that the O'Dowd et al. (2008) correlation is fully established and acknowledge that is does not hold up at other sites. We agree that there is not currently a completely robust source function that is without certain shortcomings. However, lack of a robust and established parameterization should not restrict the need to use and test the parameterization in a model.

Reviewer comment:

Global modeling based upon an empirical relationship, such as identified for Mace

Head, is only useful if it has bounded uncertainties that can challenge or be challenged by observations such that a predictive capability can be assessed. These need to estimated and identified here. Figure 2 shows that even for the relationship "tuned" for Mace Head, the model is only close to expected values about one third of the time and the trends are often out of phase. The latter suggests other possible sources are contributing to MACE Head OC for other parts of the year.

Response:

Ultimately, the model inaccuracies are at least partially tied to the problems with marine organic source function. See previous responses regarding the applicability of the O'Dowd et al. (2008) source function. However, inclusion of marine organics does measurably improve model performance. See our response to the reviewer's initial comment for details on changes we have made to the manuscript to quantify the model skill and improvements resulting from adding marine organics.

Reviewer comment:

The situation is worse for Amsterdam Island, where dependencies are expected similar to Mace Head, but there is neither agreement in value (except maybe Jan-Feb) nor trend and the annual means differ by a factor of 5, as mentioned in text.

This suggests either: 1) The model is very wrong and not very useful - so why publish. 2) The relationships employed in the model are incorrect or not implemented appropriately (what are possibilities? - discuss) 3) Seasonal variations in production or removal or entrainment etc. may not be captured in model and skew the seasonal variability - discuss? 4) However, if the authors think that the model is not reasonably expected to be this much in error at these locations (magnitude and phase) then presumably it is telling us something about the applicability of the relationship assumed for Mace Head? As the authors say "The large difference is argued to be due to the "simplicity of the marine aerosol source function" it assumes that they have this view.

Response:

1) For quantitative comparison of model to observation, see above in this document as well as Sect. 3.2 in the updated manuscript. The annual mean error at Amsterdam Island is 273%, not a factor of 5. The reviewer is probably thinking of the following statement in the original manuscript:

At Amsterdam Island, the model consistently predicts higher concentrations than the 2 observations, but still within a factor of 5.

By this we meant, that usually the model-measurement agreement was almost always better than a factor of 5. The revised manuscript now reads:

Here, the LMNB is 0.51 with marine organics, compared to -0.79 without them, indicating that the simulations with marine organics over-predict by a factor of 3 on average.

2) The reviewer states that because the model is not accurate enough, the work is not publication quality. We take an alternative philosophyâĂŤthat measurements and models must constantly give feedback to each other.

3) We have modified the manuscript slightly and introduced a new objective and conclusions. One objective is to test how well a source function developed at Mace Head applies at other locations in terms of OC concentrations. We have found that the O'Dowd source function only modestly improves OC concentrations at Amsterdam Island, the only other site we tested; however, additional locations and model predictions would be useful before generalizing this conclusion. Other factors are probably necessary to include in developing a marine organic source function, such as wind speed, which is only indirectly accounted for in the O'Dowd formulation. Please refer to the Conclusions (Sect. 4) for the updated conclusion, also pasted below:

Intro:

This study and others like it serve as a preliminary test for the O'Dowd et al. (2008) source function. Although we require more ambient datasets for a full assessment, by

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

implementing the source function into a global model and comparing to observations, we test the applicability of the Mace Head source function for predicting OC elsewhere.

Conclusions:

Our errors are in the same range that global aerosols models have previously reported for sea salt. Thus, we cannot unambiguously attribute the errors to the marine organic source function alone since predictions of marine OC concentrations will be affected by the same errors as sea salt, including errors in model wind speed. Nevertheless, we expect that improvements in the marine organic source function are possible.

4) Besides the marine source function per se, there are many likely factors that can help explain the model inaccuracy, a few of which the reviewer lists. A good point of comparison for the model accuracy of a marine emissions source is sea salt aerosol, which we know to be purely of marine origin. Pierce and Adams (2006) compare ambient observations of sea salt concentrations to GISS-TOMAS simulations using the Clarke (2006) parameterization. The model is off by as much as a factor of 4 and as little as a factor of 2, and generally does not fully capture seasonal variability. Global model predictions of wind speed are a major cause of the discrepancy. Of course, marine organic aerosols are subject to the same error as they are generated in the same way. As is mentioned above and in the revised manuscript conclusions, our errors are in the same range that global aerosols models have previously reported for sea salt.

Reviewer comment:

In other words, after presenting this figure, a more extensive discussion and assessment is warranted of these differences and model performance. For example; there may be greater organic aerosol from pollution than expected in original data and/or other fundamental issues are unresolved (e.g. chlorophyll itself is a poor predictor of Org both at Mace Head and globally over the seasons.) So why not use the model to explore whether better agreement can be found for other formulations. For example,

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

the above mentioned non-zero intercept (eq. 1) could bias values quite high for the regions of low Chl. that dominate over most of the global ocean. It would be helpful to show Mace Head (E.g. O'Dowd], 2008, Fig2) and other relevant data and consider any simple alternative formulations that might be justified and improve the capture of the annual trends. In that figure, a line through the lower envelope of data passes through the origin and one might hypothesize that line to be representative and higher deviations from that line might reflect the influence of nonoceanic sources. How does that assumption affect model performance globally?

Response:

We agree that the discrepancy in the OC mass concentrations between the models and measurements, especially at Amsterdam Island, is an ongoing challenge. The accuracy of the model is quantified now in the updated manuscript in Sect 3.3 and in this document. As mentioned above, the zero-intercept approach was attempted. As noted above, it did not change CCN predictions substantially. Moveover, it did not result in significant enough differences to improve the comparison in monthly averaged surface mass concentrations, so it is not included in the revised manuscript. We have also used the model to test another bounding case that assumes that marine organic aerosol mass and number are added on to existing sea salt rather than displacing that sea salt. In this scenario, shown in Fig. 2 of the revised manuscript, we find that predictions of mass concentration monthly averages get worse than they were previously.

As the reviewer suggests, the assumptions used to derive the source function from the Mace Head data may not completely remove continental influence. Particularly, the methods do not allow for much transport or mixing, since the marine OC is correlated to chlorophyll in a fairly specific nearby location. We have used our model to make a preliminary assessment of the validity of these assumptions by examining the correlation between ambient OC fraction and marine emissions just upwind. Specifically, we compared a time series of organic aerosol fraction in marine emissions at a presumed

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

source region just west of our model Mace Head grid cell and the model-predicted OC fraction in the ambient aerosol at the Mace Head grid cell itself. The source function assumes there is a 1:1 correlation between these values, and we can use the model "data" to test whether this is true. The emissions location is a 10 by 10 degree area ranging from 58° N to 48° N in latitude and 22°W to 12°W in longitude. As is done in O'Dowd et al. (2008), we choose a source region slightly upwind of our model location for Mace Head. The model "Mace Head location", or the ambient measurement location is itself is upwind of the coastline in order to avoid continental influences. Since the true Mace Head grid cell in the model always includes continental emissions, shifting westward is one necessary step to mimic how the data is filtered by wind direction. Marine organic mass fraction data at the ambient measurements location were output in 1-hour time resolution and filtered for wind direction and black carbon concentration to match as closely as possible the sampling method described in Cavalli et al. (2004). Model predictions of organic mass fraction at the emissions location were determined from 8-day chlorophyll-a values taken from satellite observations.

We find that there is not a simple 1:1 agreement between composition of emissions and composition of ambient aerosol just downwind. This is evidenced by Fig. 1 below, which shows a scatterplot of marine organic mass fraction in emissions source region (x-axis) and downwind ambient receptor region (y-axis). If composition at the time of emissions of marine organic aerosol matched the composition at the sampling point, the model and measurements should line up closely on a 1:1 line on a scatter plot. Instead, we find that the slope of the model-measurement comparison of organic mass fraction is much less than one (0.15). This disagreement suggests that mixing and dilution play a significant role in determining the fractional mass composition at a site such as Mace Head. Transport of marine organic aerosol generated upwind and mixing with less biologically active areas can suppress the marine organic influence on the receptor region in our model. In addition to explaining the low marine organic mass fractions at Mace Head and other locations in our model, our finding suggests that contrary to the assumptions made in developing the O'Dowd et al. (2008) source

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

function, emissions composition from a source region may not necessarily correspond on a 1:1 basis to ambient composition downwind.

These results are interesting and tentatively support the idea that the ambient Mace Head data may need a more refined analysis to infer quantitatively the marine organic emissions. However, we choose not to include these results in the manuscript itself. This analysis is at or beyond the limits of what can be expected of a global model given its horizontal resolution since the western edge of the source region to the Mace Head grid cell is only 2-3 grid cells. Therefore, we don't consider this analysis to be of publication quality.

Reviewer comment:

P5770 L19 Here the authors mention an internally mixed aerosol that contradicts impression given in P5760 L13 mentioned above. This distinction should receive more attention earlier when this point is being made. Important âĂŤThe authors have assumed an internal mix and assumed OC mass simply replaces some of the sea-salt mass in the model for sea-salt emission. If so, then the small negative effect of organics on CCN, the major conclusion of the paper, appears rather predictable simply from the relative impact these assumptions would have upon the typical size distributions used in prior model analyses. What more do we gain from this global model under the constraints of these assumptions? What might we learn from the model without this constraint?

The O'Dowd 2004 paper shows nearly a factor two difference in accumulation mode diameter from winter to spring (factor of 8 in mass per particle) and organics are argued to be the reason. What does the model see and is an internal mix of primary OC and sea-salt consistent with this enhancement in the accumulation mode?

Response:

The reviewer refers to P5760 L13, which reads, "Assuming that aerosol size and composition do not change, higher amounts of aerosol lead to higher cloud condensation nuclei (CCN) concentrations, perturbing climate by brightening clouds, which is known as the first aerosol indirect effect (Twomey, 15 1977)." This statement does not imply or intend to imply anything about the mixing state between primary marine organics and sea salt. Also, it appears that the reviewer has missed the important qualifier, "assuming that aerosol size and composition do not change". If this is true, then increasing the amount of aerosol means changing aerosol number and mass by the same proportion. It follows that CCN concentrations increase.

Regarding the validity of the "replacement" assumption of marine OC, see previous discussion and text added to the manuscript (quoted above). In particular, we have added discussion of the limitations of the "replacement" assumption, and we have run simulations with the "addition" assumption and now include them in the manuscript discussion. Please refer to Sect. 2.3.1 for details of the simulations and Sect 3.4 for the results. The addition to the abstract regarding this new result was pasted above in this response document. Also a few lines of Sect. 4 have been adjusted to reflect the new conclusions. Discussion also pasted below:

Figure 4 shows the percent change of CCN(0.2%) concentrations assuming that marine organics do not replace sea salt aerosol (Sσ-ORG-ADD simulation). In this plot, surfactant effects are included. We find that the sign and the magnitude of the CCN effect changes significantly when compared with the replacement assumption (S-ORG simulation). The increases in CCN(0.2%) concentrations can be explained by the manner that the source function was applied in these simulations. For these simulations, number and mass of marine organic aerosol is an additional source that does not displace any sea salt. More aerosol number, despite the low hygroscopicity, will lead to more CCN. Similar to the replacement of sea salt simulations, the strongest changes are predicted in the Southern Ocean in both DJF (panel A) and JJA (panel B). The CCN(0.2%) concentrations now increase when compared to a base case simulation by as much as 25-50% in the Southern Ocean during the austral summer. In con-

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

trast, northern hemisphere summer increases in CCN(0.2%) are in the 10-25% range. Globally averaged the increases are 3.7% for the DJF months and 2.9% for JJA.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 5757, 2011.

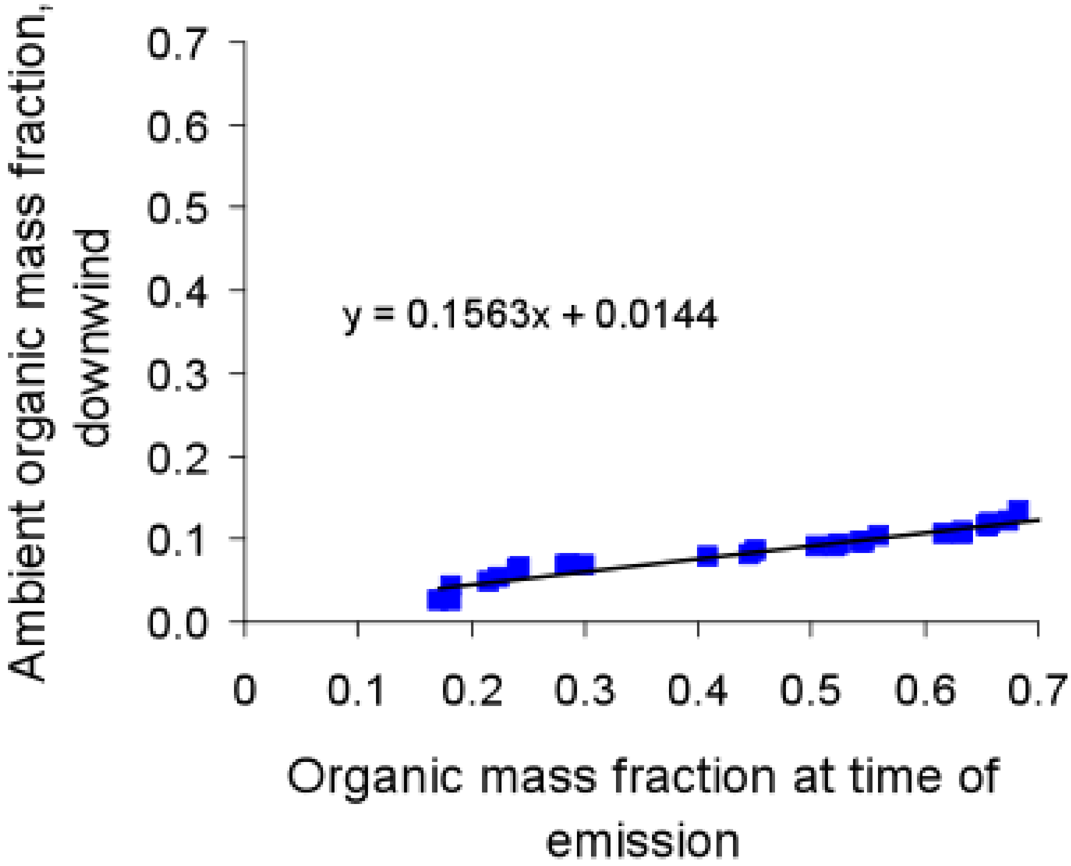

Fig. 1. Scatter plot of modeled marine organic mass fraction at the location for Mace Head ("ambient mass fraction") versus the modeled mass fraction at an adjacent source region ("emission mass fraction")

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper