Atmos. Chem. Phys. Discuss., 11, C2366–C2371, 2011
www.atmos-chem-phys-discuss.net/11/C2366/2011/

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.

Anonymous Referee #1

Received and published: 28 April 2011

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

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

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 it's 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.

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."

"Quantifying" this would require a perfect model of a perfect parameterization and there are reasons to assume neither are the case. Rather, this model estimates an impact by assuming the validity of a relationship that has been called into question regarding its global application and that is shown here to yield concentrations inconsistent with observations. Please rephrase to reflect this.

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).

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

ACPD

11, C2366–C2371, 2011

remote sensing.

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.

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

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.

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?

P5762 section 2.2.2 While reference to combustion/continantal aerosol is made and it is know to be effectively transported across the Atlantic (available to be entrained into marine bundary 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.

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.

ACPD

11, C2366–C2371, 2011

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).

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.

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.

In other words, after presenting this figure, a more extensive discussion and assess-

ACPD

11, C2366–C2371, 2011

ment 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 (eg. chlorphyll 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, 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 helpfull to show Mace Head (Eg. 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 envelop 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 non-oceanic sources. How does that assumption affect model performance globally?

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 —The 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?

ACPD

11, C2366–C2371, 2011

P5772 L11 Authors say — “Since our emission source is almost a factor of two larger than several past studies (Gantt et al., 2009; Spracklen et al., 2008; Vignati et al., 2009), our predicted CCN impacts may be correspondingly high.” Please explain argument. If the number emission is too high, then presumably a lower number concentration would suggest a larger OC fraction per particle (assuming OC flux from Chl is the same) and a somewhat larger relative change – though as you point out, it may still be small.

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