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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Biogenic influence on cloud microphysics over the global ocean" *by* A. Lana et al.

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Authors: Thank you for your valuable comments. Responses in situ.

Overall: Reviewer: This paper attempts to determine which types of marine biogenic aerosols affect the cloud microphysical properties near the emission source using statistical analysis of satellite data as the main tool. The high negative correlations between liquid cloud effective radii and sulfur/organic secondary aerosol production at mid and high latitude regions lead to the authors to conclude that it is these aerosols, as opposed to primary organic and sea-salt aerosols, that are major drivers of the variability of cloud microphysics. Despite the fact that paper is well written and topic is relevant to ACPD, I recommend major revisions for the paper prior to publication in ACP.

Major Comments: R: The main critique that I have of this paper is the use of spatio-temporal correlations from satellite data to determine potential causality in cloud microphysical variability. The four types of aerosols examined (sulfur, SOA, primary organics, and sea-salt) have different formation mechanisms. Applying the same correlation method for all four inherently favors one type of formation mechanisms over the other (especially primary vs. secondary aerosols). I would suggest describing in more detail how the aerosol formation mechanisms differ in terms of time scales, and adjust the correlation method to suit this time scale. See Woodhouse et al. (2008) for a modeling study of the relationship between DMS emissions in a particular location and sulfur aerosols.

A: Yes, obviously, the different aerosols examined have different formation mechanisms, as we outline in the introduction and will further describe and discuss in the discussion. Therefore, it is true that their formation kinetics varies from type to type, and this affects their distribution and impact on cloud properties. This can only be resolved by coupling emission flux parameterizations to atmospheric chemistry and physics modeling. But note that models are only as good as our knowledge of mechanisms is, and this is not always recognized in modeling papers. In other words, atmospheric models are quite good at simulating transport (horizontal and vertical) processes but importantly limited at simulating complex mechanisms such as heterogeneous reactions or formation of internal mixtures. Our study takes a different path: weekly and monthly correlations at the regional level should constrain the mechanistic potential for an influence of biogenic emissions on CCN production and cloud microphysics. That is, occurrence of strong correlation (in the expected direction) sets a basis for causal relationship (without proving it), while absence of significant correlation prevents causality to occur. In other words, correlation is a necessary yet not sufficient condition for causality. We agree, though, that key to this argument is to allow time space for aerosol production and cloud condensation mechanisms to occur. This is why our analysis uses 7°x7° (lat x long) windows and a minimum time step of one week. This needed better discussion and recognition in the original manuscript. We

also think that a change in manuscript title will better reflect the focus and limitations of the study: “Potential for a biogenic influence on cloud microphysics over the ocean: a correlation study with satellite-derived data”

R: There also needs to be some evidence that the emission mechanisms used in the paper are accurate. This is shown nicely in Fig. 3 for DMSflux, but not found for the other three aerosol types. I would suggest showing something similar to Fig. 3 (if possible) for the other aerosol types. If the seasonality of emissions does not reflect that of the surface concentrations (see Meskhidze et al., 2011; Westervelt et al. 2011), additional emission mechanisms may need to be included. Accurate inspection of Fig. 1 shows that positive correlations between cloud effective radii and POA/sea-salt occur along 40S. Actually, most of the emissions of POA (Vignati et al., 2010) and sea-salt (de Leeuw et al., 2011) are supposed to occur between 40S and 60S.

A: We agree that some sort of validation of the emission and oxidation flux parameterization was needed. It was quite well shown for DMS because aerosol MSA is a common measurement in ground-based field studies, and this compound originates exclusively from DMS and has no major continental source. However, unlike for biogenic sulfur, there are no exclusive markers for secondary and primary organic aerosols to validate the parameterizations against. A common assumption is that water insoluble organic matter (WIOM) mass in aerosols is predominantly associated with POA, and water soluble organic matter (WSOM) is predominantly associated with SOA (Ceburnis et al., 2008; Facchini et al., 2008b). Although it has been claimed to be an oversimplification, this source attribution based on solubility offers a venue for validation of SOA and POA source functions. The manuscript needed a new section entitled ‘Ground validations of aerosol flux seasonalities’, where our flux parameterizations could be compared to aerosol measurements in different oceanic regions. Former Figure 3 will be converted into Figure 4 and used to validate γDMSflux computations at 6 aerosol sampling stations, removing the re, which has a closer examination in other figures. For organics, though, only a few measurements of the seasonality of aerosol WSOM

and WIOM exist: those of Amsterdam Island (Sciare et al., 2009) and Mace Head (Yoon et al., 2007). We have added a third study with ship-based measurements of organic aerosols along a broad latitudinal transect (Miyazaki et al., 2011). We have constructed a new Figure 5 to validate the SOAflux and the POAflux.

The figure shows that the parameterizations do a good job at reproducing organic aerosol production fluxes over space and time, with the exception of WIOC at Mace Head.

Before doing this validation study, we realized there was room for improving the parameterization of the POAflux. We have maintained the approach of quantifying the sea spray particle flux and converting it to a mass flux of organics as a function of chla concentration, but have better described how this conversion is made (by first converting the particle flux into volume and mass flux), and have updated the parameterization of the organic fraction of sea spray (Gantt et al., 2011).

R: None of the time series Figures (2, 4-6) seem particularly helpful. The useful information from the figures is the correlation value, and these can easily be put into a table. If these figures are not removed, they need units and observations (if available) and can be simplified into seasonal averages.

A: We think it is worth keeping the figure of the Southern Ocean (Figure 2) and those of Amsterdam Island and Mace Head (now Figure 3) as token examples. They help to visually assess the temporal covariations that are numerically expressed by correlation coefficients. They also serve to show what is the typical seasonal pattern for each of the aerosol types and the re, something not shown by the correlation coefficient. We suggest keeping these three cases as contrasting case studies. Their correlation coefficients, along with those of all the other case studies, will be presented in a new Table 1.

R: The strong dependence of the results on the Southern Ocean concerns me. This area has a strong seasonality in meteorological factors such as solar radiation, wind

speed, and sea surface temperature among others that may affect cloud microphysical properties unrelated to aerosol interactions. I would suggest discussing and possibly accounting for the influence of meteorology on clouds in this region.

A: With a re-construction of the paper, the results are not that dependent on the Southern Ocean. It is indeed a region with marked patterns where a strong case can be made, but the paper does not focus only on it. Rather, it is treated as a case study, just like other regions (Amsterdam Island, Mace Head, Shemya Island, Cape Hedo). However, we agree that, in any of the regions, meteorological or cloud macrophysical factors may play as big a role (or bigger) in cloud microphysics as the aerosols. This was overlooked in our paper. Actually, our approach was too naïve at attempting a global analysis of aerosol-cloud droplet size correlations across all sorts of meteorological regimes. We have now changed the object of our global study: the global map of correlations is now built with CCN number correlations instead of re. Both are satellite-derived, but the relationship of aerosol production fluxes to CCN numbers is far more direct than to cloud droplet size, and less prone to influences by other factors such as liquid water or cloud height. Satellite-derived CCN are actually numbers of accumulation mode aerosols, which will potentially act as CCN. The comparison is equally relevant for cloud microphysics but cleaner.

R: On the other hand, in the case of the Equatorial and Tropical North Pacific it is argued that positive correlations between DMSflux and effective radii are likely due to high altitude clouds. As correctly noted by Anonymous Referee #1 authors should limit their statistical analysis using clod top pressure and temperature. Moreover, when talking about the Twomey effect it is implicitly assumed that cloud liquid water path is constant. I did not see any discussion of this in the manuscript.

A: This is a very relevant comment. As commented above, we overlooked the effects of cloud macrophysics and meteorology. As for the altitude of clouds, we used MODIS-derived liquid cloud data, assuming this would select for low clouds. But there are regions where liquid clouds can hold at high altitudes. After the reviewers' comments,

we have applied a cloud filter for our statistical analysis: in our case studies, we have computed Spearman's rank correlations with all data, as in the original manuscript, and subsequently repeated the computation by considering only pixels with a LWP beyond a narrow low range (15 g m-2), then again taking only the pixels with cloud top pressures lower than 680 hPa, and still a third time using only the pixels that fulfilled the two conditions: LWP in the lower quartile and CTP $\geq$ 680 hPa. All correlation coefficients are collected in Table 1.

R: Overall, I think in its current form the paper adds very little to the ongoing debate for the ocean biological influence on cloud microphysics. However, the manuscript can be improved significantly if i) spatio-temporal effect of DMS oxidation are included (i.e., DMS may influence accumulation mode aerosol number 10th of thousands of km downwind) and ii) more detailed satellite data processing is carried out for clouds.

A: Spation-temporal effects of DMS oxidation are not included, but the analyses are conducted within 7°x7° regions and one week minimum. A more detailed satellite data processing for clouds has been conducted for a more meaningful statistical analysis of potential aerosol-cloud interactions.

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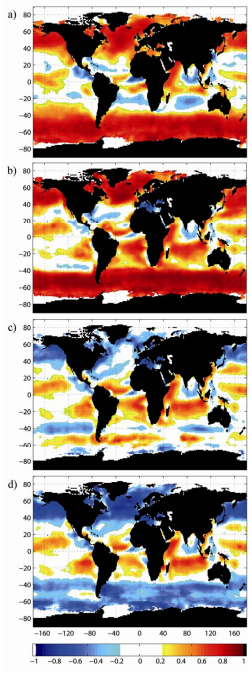

Figure 1. Global maps of Spearman's rank correlation coefficients between monthly series (2001–2009) of MODIS-derived CCN number concentrations and the parameterized fluxes of: **(a)** DMS emission and oxidation in the atmosphere (DMSflux), **(b)** SOA formation in the atmosphere (SOAflux), **(c)** POA emission (POAflux) and **(d)** sea salt emission (SSflux). White areas show non-significant correlations.

Fig. 1. Figure 1

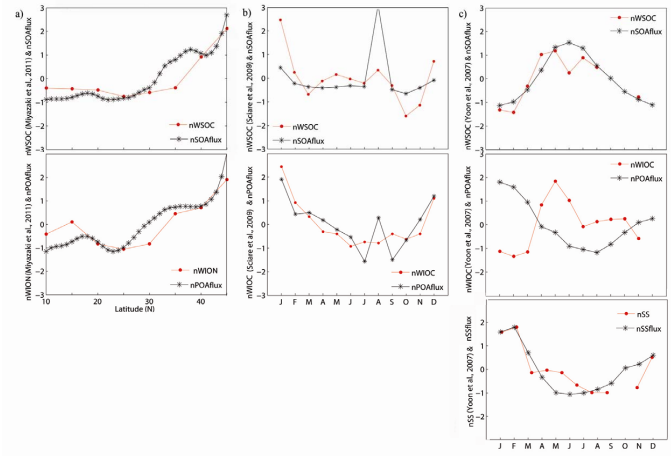

Figure 5. Validation of aerosol flux parameterizations. (a) Standardized latitudinal series of aerosol water soluble organic carbon (WSOC) and water insoluble inorganic nitrogen (WION) measurements reported by Miyazaki et al. (2011), with the corresponding satellite-derived computations of SOAflux and POAflux for September 2008. (b) Satandardized monthly series of aerosol WSOC and WIOC measurements on Amsterdam Island (Sciare et al., 2009), with the corresponding computations of SOAflux and POAflux for years 2005-2007. (c) Standardized monthly series of aerosol WSOC, WIOC and sea salt measurements on Mace Head (Yoon et al., 2009), with the corresponding computations of SOAflux, POAflux and SSflux for years 2002-2004.

Fig. 2. Figure 5

Table 1. Spearman's rank coefficients of correlation between computed weekly marine aerosol production fluxes and satellite-derived cloud droplet radius (r_e) in case study regions. In parantheses, number of weeks with data.

Location	Variable correlated to r_e	All data (2001-2009)	Low-range LWP only[a]	Low clouds only[b]	Low-range LWP & low clouds only
Southern Ocean (40°-60°S circumpolar band)	γDMSflux	-0.93 (409)	-0.90 (408)	-0.90 (409)	-0.87 (408)
	SOAflux	-0.92 (383)	-0.89 (380)	-0.89 (383)	-0.84 (380)
	POAflux	-0.38 (384)	-0.35 (380)	-0.07 (384)	-0.06 (380)
	SSflux	0.63 (384)	0.57 (380)	0.70 (384)	0.64 (380)
Amsterdam Island	γDMSflux	-0.80 (414)	-0.68 (324)	-0.63 (180)	-0.57 (78)
	SOAflux	-0.78 (381)	-0.63 (277)	-0.65 (160)	-0.64 (62)
	POAflux	-0.35 (386)	-0.18 (293)	0.05 (165)	0.11 (63)
	SSflux	0.69 (386)	0.63 (293)	0.59 (165)	0.77 (63)
Shemya Island	γDMSflux	-0.60 (413)	-0.50 (317)	-0.69 (219)	-0.66 (141)
	SOAflux	-0.63 (362)	-0.55 (258)	-0.70 (187)	-0.63 (116)
	POAflux	0.15 (368)	0.12 (280)	0.19 (211)	0.23 (135)
	SSflux	0.41 (368)	0.30 (280)	0.59 (211)	0.60 (135)
Mace Head	γDMSflux	-0.13 (414)	-0.16 (266)	-0.13 (338)	-0.12 (217)
	SOAflux	-0.25 (314)	-0.15 (242)	-0.20 (270)	-0.12 (197)
	POAflux	0.13 (316)	0.08 (246)	0.14 (271)	0.05 (201)
	SSflux	0.34 (316)	0.21 (246)	0.30 (271)	0.16 (201)
Cape Hedo	γDMSflux	0.36 (414)	0.37 (352)	0.21 (336)	0.20 (257)
	SOAflux	-0.54 (392)	-0.47 (334)	-0.53 (313)	-0.57 (238)
	POAflux	-0.24 (393)	-0.14 (335)	-0.31 (314)	-0.27 (239)
	SSflux	-0.06 (393)	0.02 (335)	-0.15 (314)	-0.12 (239)

a: LWP within 15 g m^{-2}, at the lower quartile of the annual variability
b: cloud top pressure >680 hPa

Fig. 3. Table 1