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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.

**A. Lana et al.**

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

Reviewer: Manuscript deals mostly around the so called CLAW hypothesis, which have been under investigation for few decades, and been recently also challenged. The manuscript is well written and in organized logically. The manuscript deals with relationships between satellite retrieved cloud droplet effective radius and various possible sources of CCN; from DMS oxidation, primary organic aerosol (POA), secondary organic aerosol (SOA) precursors and sea salt aerosol. The manuscript output is correlation coefficients between possible and potential sources and cloud droplet effective radius. There is a strong negative correlation between "potential source for CCN-forming DMS oxidation" and cloud droplet effective radius above part of the oceans, most clearly in southern hemisphere between 40° and 60° S. There is also correlation between SOA and cloud droplet radius. For POA and sea salt there seems not to be correlation or it is positive. Though for the science the contribution of the manuscript is not major, the results might be quite interesting after some data processing. This could be published in ACP after revisions.

Major concerns and suggestions:

R: Even though there is a clear negative correlation between "potential source for CCN-forming DMS oxidation" and cloud effective radius it does not proof that between "potential source for CCN-forming DMS oxidation" is responsible for seasonal behavior of cloud effective radius. Authors also point this out, though it should come out stronger in the text. There is a long way for " potential source for CCN-forming DMS oxidation" or "SOA flux" to affect cloud properties, though natural biological cycle is similar than annual cycle of effective radius, which can be affected by many factors.

A: Yes, we know there is a long way from aerosol or aerosol precursor emission from the surface ocean and cloud properties. In our approach, 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.

In our global analysis, we have shortened this long way by changing the cloud droplet size (re) to CCN number concentration as the correlation counterpart of aerosol fluxes. 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. This will constitute the renewed Figure 1.

The comparison to cloud droplet size (re) is left for regional case studies where a detailed analysis of cloud macrophysical conditions is conducted before applying the correlation analyses (see below).

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: When talking about the indirect effect the liquid water content in clouds should be same in all cases, there is no mentioning about if has this kind of classification has been done for the effective radius data. Also it would be out of interest how cloud optical thickness (which is actually closer to climate effect) is affected by these parameters. There is very little information if there has been any screening of satellite cloud retrievals. Are authors discussing on MBL clouds or all clouds? I think one should make a division between the cases, aerosol having influence on clouds in MBL are most likely different than in free troposphere. More emphasis should be put on the cloud properties, cloud top height and temperature, cloud cover etc..not just effective radius. This might be difficult and require lot of work since one should look into more detailed satellite retrieval data than just monthly or weekly averaged data. However, just to use the current data set is not very satisfying. How would relationships look like if one take for example every January over the period under investigation and compare “ potential source for CCN-forming DMS oxidation” or “SOA flux” against cloud droplet, I suppose the other properties affecting to either one would be closer to each other than taking all data into consideration.

A: This is a very relevant comment. We overlooked the effects of cloud macrophysics and meteorology, which may play as big a role (or bigger) in cloud microphysics as the aerosols. 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. Actually, our approach was too naïve at attempting a global analysis of aerosol-cloud droplet size correlations across all sorts of meteorological regimes. 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 all pixels with a LWP beyond a narrow low range (15 g m-2), then again by 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: The SSflux and POA fluxes do not correlate with cloud droplet effective radius or there is a positive correlation. Very little discussion is given to this positive correlation.

A: Positive correlations were totally unexpected from the mechanistic point of view. The fact that SSflux (generally) and POAflux (occasionally) correlated positively to droplet size must be attributed that primary aerosol emission fluxes are parameterized from wind speed as the main factor, and wind speed tends to be higher in winter, at the season with larger cloud droplets but also higher cloud liquid water (LWP). Positive correlations seem to indicate that sea spray (and particularly the associated sea salt), despite contributing a large share of aerosol mass, does not drive the variability of cloud microphysics. This was also the conclusion of Vallina et al. (2006).

R: I wonder what are the results if one limits the clouds only to MBL clouds.

A: See response above. Results given in Table 1.

R: I wonder what is the main driver in DMS and SOA fluxes. Is it OH, SST, DMS in surface water, wind speed. . . so is one could make this kind of examination against these and cloud properties what would be the correlation coefficients.

A: As requested, we analyzed the drivers of DMS and SOA fluxes in isolation. To do so, we computed the fluxes in the Southern Ocean several times by maintaining one of the involved factors constant throughout the year, and letting all other factors follow their seasonal patterns. Results are shown in Figure Review1. We can observe that the DMS concentration is the main driver of the DMS-oxidation flux seasonality, and the OH radical is the most influential driver for the seasonality of the SOAflux. This is information for the reviewer that will not appear in the revised manuscript.

R: For authors the absolute values are not important. At least I would like to see how much cloud droplet effective radius changes with fluxes or with season, at least for the case studies. For case studies I miss also simple flux (MSA) vs. cloud effective radius plots.

A: Our study is based upon the statistical comparison between time series, seeking to constrain causal relationships. We are not interested in absolute values; note that the parameterizations contain a unit conversion constant. We are interested in the timing and amplitude of the seasonality, which will determine whether or not a variable will have the potential for influencing the variability of another variable as hypothesized. Note that satellite based retrievals tend to systematically deviate from ground (or ship- or aircraft-based) measurements, but both agree in the temporal variability. In particularly, MODIS generally gives larger CCN number concentrations and bigger re.

Minor comments:

R: page 3657; line 3; concentration number -> number concentration A: Corrected.

R: page 3659; Line 22, reference von Glassow, 2007; I did not find from reference list A: Added.

R: page 3661; line 8; definition for SST A: Done.

R: page 3662; line 2; raion ? should it be ratio A: Corrected.

R: page 3661; line 3; definition for ks: A: Added.

R: page 3663; line 25; definition for CHL A: Defined at first appearance.

R: page 3665; line 16; Lana et al. 2011c, I do not think one can refer to manuscript “in preparation” A: We have removed any reference to this paper. Actually, we have removed the use of a mask of continental influence to the marine atmosphere. It did not add much to the story and arguments of this paper, and belongs to another study.

R: what is the y-axis in figs 2, 3, 4 and 5. A: It is standardized values (subtraction of the mean and division by the standard deviation).

R: Fig 2., in inset there is CHL and in text box SOAf. A: Corrected.

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 3655, 2012.

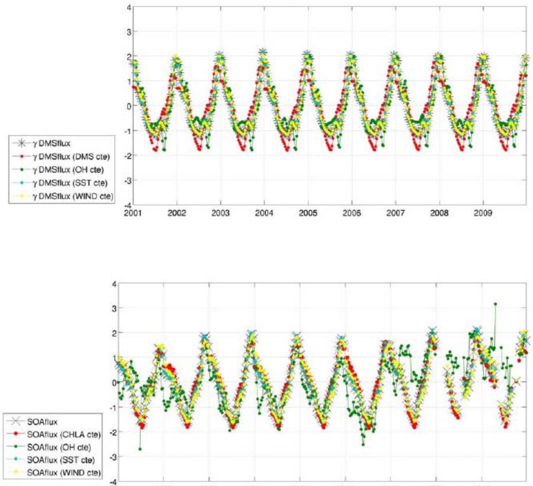

Figure review1 (for reviewer only). Weekly computations of the γDMSflux (top) and the SOAflux (bottom) in the Southern Ocean, repeated keeping constant one of the variables involved in the calculations. Cte = constant.

**Fig. 1.** Figure review1

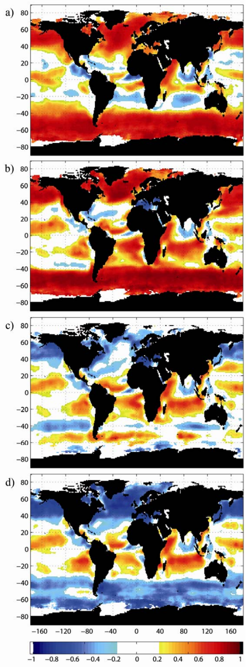

Figure 1. Global maps of Spearman's rank correlation coefficients between monthly series (2001–2009) of MODIS-derived CCN number concentrations and the parameterized fluxes of: **(a)** DMS emission and oxidation in the atmosphere (DMSflux), **(b)** SOA formation in the atmosphere (SOAflux), **(c)** POA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

**Fig. 2.** Figure 1

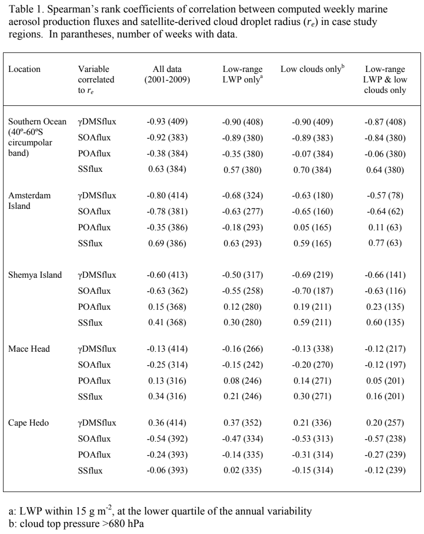

Table 1. Spearman's rank coefficients of correlation between computed weekly marine aerosol production fluxes and satellite-derived cloud droplet radius ($r_e$) in case study regions. In parantheses, number of weeks with data.

| 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 (40º-60ºS 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