

Referee #2

The authors thank the anonymous referee #2 for his/her constructive comments and corrections which have helped to improve our original manuscript. **Referee comments which we are responding are given in small italics below.**

First, we bring to the referee attention that due to a recent request from modelers working on the SEP region, we think that it will be useful to provide the SSA mass mixing ratio associated with our clean SSA size distributions along 20 South. Therefore, Fig 11. will updated in the final revision as shown below.

General comments :

This is an interesting study that predicts the contribution of sea-salt aerosol particles to potential CCN in a clean marine region. It concludes that not only source functions, but also sinks (in particular, precipitation), are required to explain the observational data. The paper has a nice progression from background information, to various pieces of evidence, to the final result. I recommend it be published after some minor revisions/corrections as described below.

Reply : Thank you for the positive assessment concerning the general overview of our manuscript.

The introductory material states that the contribution of sea-salt particles to the marine ultrafine number is still debated, and the authors' method does not directly measure sea-salt via composition, but instead as refractory particles. This made me a little uncomfortable with the initial designation of the term SSA (sea-salt aerosol) for data acquired in this way. However, strong evidence is presented that these particles are predominately sea-salt (at least for the VOCALS regions) i.e, the AMS, which includes smaller particles, measured very low organic signature, the refractory signal is correlated with windspeed, etc. A summary or reiteration of this evidence would be beneficial when the SSA designation is directly applied to the thermal measurements .

Reply : We note that we elected not to use the familiar term Sea Salt Aerosol but rather Sea Spray Aerosol in view of the increasing complexity being associated with emissions from sea spray. The initial designation of the term SSA is defined for the first time in the introduction as Sea Spray Aerosol (p3281, line 5). Then, later within the same paragraph a definition for Sea Spray Aerosol is given (line 8-11) : “ In this study, SSA represent all the inorganic materials (sea-salt), organic matter and other surface active materials (exopolymer) that can be incorporated into aerosol from bursting air bubbles at the ocean surface.”

However, it might be unclear that we assume the measured SSA being refractory over the SEP because for clean conditions most of the volatile aerosol are expected to be sulfate species derived from DMS (Clarke, A. D.: A thermo-optic technique for in-situ analysis of size-resolved aerosol physico-chemistry, Atmos. Environ., 25, 635–644, doi:10.1016/0960-1686(91)90061-B, 1991. 3281, 3286, 3287, 3293).

To clarify this issue we add to the text (3294, line 2) :”Since the pollution indicators are well below the thresholds established above and the volatile fraction mostly composed of sulfates from DMS (Clarke, 1991), we argue that the size distribution shown here is representative of clean SSA over the SEP and can be integrated to access SSA CCN number.”

We also do state that most of coarse aerosols are made of sea-salt, as it shown in several studies, but we do not state anything about the submicrometer particles since other non-volatile marine particulate

matter can also incorporate the freshly produced sea droplets. (see Bigg, E. K., and C. Leck (2008), The composition of fragments of bubbles bursting at the ocean surface, *J. Geophys. Res.*, 113, D11209, doi:10.1029/2007JD009078.).

To make it more clear we will add to the text (3286, line 20) : “When heated over 300 °C, most mass associated with species like sulfuric acid, ammonium sulfate/bisulfate are removed (Clarke, 1991) and the non-volatile aerosol components such as BC, dust and sea-salt remain. The latter is not the only non-volatile component in SSA, other marine particulates stable at high temperatures can possibly be transferred to the atmosphere by bursting bubbles (Bigg and Leck, 2008).

Throughout the paper, single or narrow-range supersaturation values are used for calculations based on the location of the Hoppel minimum (for example, middle of p. 3300, elsewhere). In fact stratocumulus clouds include air parcels with a continuum of different updraft and supersaturation histories (e.g., Snider et al., JGR, 2003, Twohy et al, ACP, 2013). Particles smaller than the Hoppel minimum can be activated in strong updrafts; they just have not been through enough condensation/evaporation cycles to grow through the gap. These issues should at least be acknowledged.

Reply : The center of the Hoppel minimum indicates the most common or frequent supersaturation present in the ambient clouds. The width of the Hoppel minimum (range of diameters) also reflects the influence of the range of supersaturation active in the clouds. We will note that cloud supersaturation is not directly measurable and that the Hoppel minimum provides the best constraint but on a cloud scale there is significant variability.

It should be noted that sea-salt particles are important not only for number of CCN/cloud microphysics, but also for reactions with sulfur species and aqueous-phase chemistry (O’Dowd et al., GRL, 1999, Hegg et al., JGR, 1992).

Reply : We agreed that this information is missing in the introduction. It will be added to final revision.

Minor Comments/Typos :

The paper is filled with typos, which are distracting and suggest a lack of attention to detail. The ones I caught are given below.

Abstract: suggest changing “ambient CCN” in last sentence to “potential CCN”, as that seems to be more correct.

Reply : Agreed. It will be corrected in the revised manuscript.

p 3281 line 7: Do you mean dimethyl sulfide?

Reply : Yes, It will be changed to dimethyl sulfide.

Paragraph starting line 8: A useful reference here would also be: Heintzenberg J, Covert D C and Van Dingenen R 2000: Size distribution and chemical composition of marine aerosols: a compilation and review Tellus B 52, 1104–22.

Reply : Agreed. Thank you for the suggestion and the paper will be quoted in the final revision.

p. 3282 line 1: second “their” should be “there”.

Reply : Ok

line 8: insert “previously” before “activated”.

Reply : Agreed.

Line 24: something is wrong with this sentence, do you mean “issued” instead of “issue”?

Reply : Yes. It is the organic matter originated from biogenic activity.

Line 26: “with a very little contribution of the sea-salts mass” also is grammatically awkward.

Reply : Agreed. It will be changed to “ ... , these can seasonally dominate the mass of submicrometer marine aerosols with a very little contribution from the sea-salts.”

p. 3283: Another grammar problem: “are well correlated with surface wind speeds which is considered as the major parameter” Line 25.

Reply : Agreed. It will be changed to “ ... are well correlated with the surface wind speed which is considered as the major parameter for SSA production ... ”

What about advection?

Reply : While advection may transport SSA in (and out) of a region, it is not a parameter linked directly to production. Our model assessment assumes advection is transporting aerosol produced over the past two days to the 20 South line. On any given flight, advection may be weaker or stronger and concentrations will vary with upwind production and removal. Our assumption is that by averaging over these multiple flights, such variability will be minimized. SSA with a diameter of 1 μm can travel in the MBL during more than a week before being removed by dry deposition (see Lewis et Schwartz 2004, p76). However, in many regions, and particularly over the SEP, SSA concentrations are frequently affected by drizzle. Unless Lagrangian Flights are performed, it is very difficult to adequately assess the air mass history. However, the time spent by larger particles ($>5 \mu\text{m}$) in the MBL is reduced to less than half a day to only several minutes for SSA of 20 μm (see also Lewis et Schwartz 2004, p76). Therefore, a better relationship with wind speed has often been found since it is easy to measure and assume to be representative of the local production.

p. 3284: “cover” should be “covered”.

Reply : Ok.

p. 3285 line 7: “mission” should be “missions”. “coast” is missing.

Reply : Ok.

line 15: “grow” should be “growth”.

Reply : Ok.

The comparison with the GNI is interesting and valuable.

Reply : Thank you for this positive comment.

p. 3286 line 13: "particle" should be "particles". Line 22: "organic" should be "organics".

Reply : Ok.

p. 3287: "describe" should be "described". line 3-4:

Reply : Ok.

Doesn't the DMA actually measure mobility size?

Reply : That is correct but the measured mobility distribution was then inverted to an aerosol size distribution . Details are found in Zhou, J., Hygroscopic properties of atmospheric aerosol particles in various environments, Ph.D. thesis, 166 pp., ISBN 91-7874-120-3, LUTFD2/ (TFKF-1025)/1-166/ (2001), Lund University, Lund, Sweden, 2001. This reference will be added in the revised manuscript.

line 14: insert "of" before "the OPC". Refer should be "referred" etc

Reply : Ok.

p. 3288, line 3: "particles" should be "particle" line 11: "exited" should be "excited".

Reply : Ok.

Line 16: "and" should be "who" or some other phrase.

Reply : Agreed. It will be replaced by two sentences : " Data from the same instrument were used in Shank et al. (2012). They found a particle size detection limit of ... "

p 3289 line 9: acquired is misspelled.

Reply : Ok.

p 3290 line 4: insert "to" after "refer". insert comma after ddry. line 17: "value" should be "values".

Reply : Ok.

"in the meanwhile keeping an reasonable amount of data" is very awkward.

Reply : Ok. The whole sentence will be replaced by : "It consists in constraining all the data for BC, angstrom coefficients and organics at values intended to ensure low pollution. However, these thresholds should be high enough to provide a database that is also statistically significant."

p. 3292, line 4-5: criteria should be criterion.

Reply : Ok.

p. 3292, line 7: "affected" should be "affect". line 9: Do you mean organic concentrations? If so, insert "organic" before "concentrations".

Reply : Ok.

p. 3293 line 4: Delete “volatile” ammonium sulfate is not volatile under atmospheric conditions, which is what is discussed here. Same on p. 3300 line 9.

Reply : Ok.

line 5: Specify what diameters you’re using for “associated with”. End of page: Just because there is a Hoppel minimum there doesn’t mean smaller particles aren’t sometimes activated at higher supersaturations (see earlier discussion). Some comparison with measured updraft velocities would be interesting.

Reply : Yes, there are smaller particles activated than the lowest point of the Hoppel minimum but these are also indicated by the shape of the minimum. As noted above, the location and width of the Hoppel minimum constrains the mean ambient cloud supersaturation and its variability.

p 3294 line 11: Insert “see also” before Murphy et al., 1998 since the reference does not seem to be the direct source of your prior statement, just additional support for it.

Reply : Agreed.

line 16: delete “aerosol” (redundant with next line). line 17: change “aerosols” to “aerosol particles”.

Reply : Ok.

p 3299, line 2: Careful with wording. If I understand correctly, these are not activated particles but simply a predicted number of SS aerosol particles in a certain size range, based on the assumptions.

Reply : The sentence will be replaced by : “ Finally, we model SSA larger than 0.060 μm that can be activated at a standard supersaturation of 0.25 % (see Sect. 3.2). “

p. 3299-3300 discussion of precipitation: It’s not clear if you mean the precipitation sink is through nucleation and coalescence scavenging of particles, or impaction scavenging of aerosol by falling drizzle/raindrops. Also, I can think of other reasons why the SSA number concentration would be reduced offshore. For example, the higher supersaturations in offshore clouds (due to lower CCN concentrations and generally stronger updrafts) would activate smaller SSA, subjecting them to all the in-cloud loss mechanisms discussed above. Also, the larger drops offshore will enhance coalescence and reduce particle number even if drizzle drops evaporate below cloud, rather than precipitating. Perhaps these complexities should be mentioned.

Reply : Here we mean that the precipitation sink is through coalescence scavenging of SSA/CCN by falling drizzle/raindrops. This will be added to the final revision. It is correct that other processes influencing the CCN concentrations in cloud should be mentioned. However, we believe that the decrease of SSA (Fig 11.) and the great increase in precipitation (Fig 10d.) deeper clouds for more offshore locations along 20 South is a strong argument for coalescence scavenging.

p. 3300, line 6: “imply” should be “implies”.

Reply : The sentence line 6 will be replaced by “Consequently, these result imply that SSA”

p. 3300 : Here and in Fig. 12 the Hoppel minimum for clean air is specified as $\sim 0.05-0.08$ but an earlier example (p. 3292) uses 0.075-0.11, which seems much higher. A more representative example should be used to avoid confusion.

Reply : Fig 6. will be updated (see below) with a new clean size distribution measured along 20 South. The Hoppel minimum observed for this new figure is within $\sim 0.05\text{-}0.08 \mu\text{m}$ (estimate range of supersaturation $\sim 0.20\text{-}0.30\%$). We will also add in the discussion that most typical supersaturation values are constrained by the narrow grey line shown in that figure but the shape of the minimum (when compared to an original monomodal distribution) is extended down to much smaller sizes. These would have to be calculated by fits to the original distributions (which we do not have here) but it a significant fraction of sizes down to $\sim 0.035 \mu\text{m}$ were also activated.

line 23-24: Suggest changing “depth and width” to “height and width” if that is what you mean.

Reply : Ok.

The differences could also be a result of higher supersaturations due to fewer CCN (less competition for vapor) and stronger updrafts farther offshore.

Reply : Agreed, this comment will be add to the final revision.

p. 3301 line 10: ‘averaged” should be “average”. Line 17: Insert “clean” before MBL CCN.

Reply : Ok.

Fig 4 caption: “Total number of data” data points at specify sample rate.

Reply : Ok.

Fig. 11 :

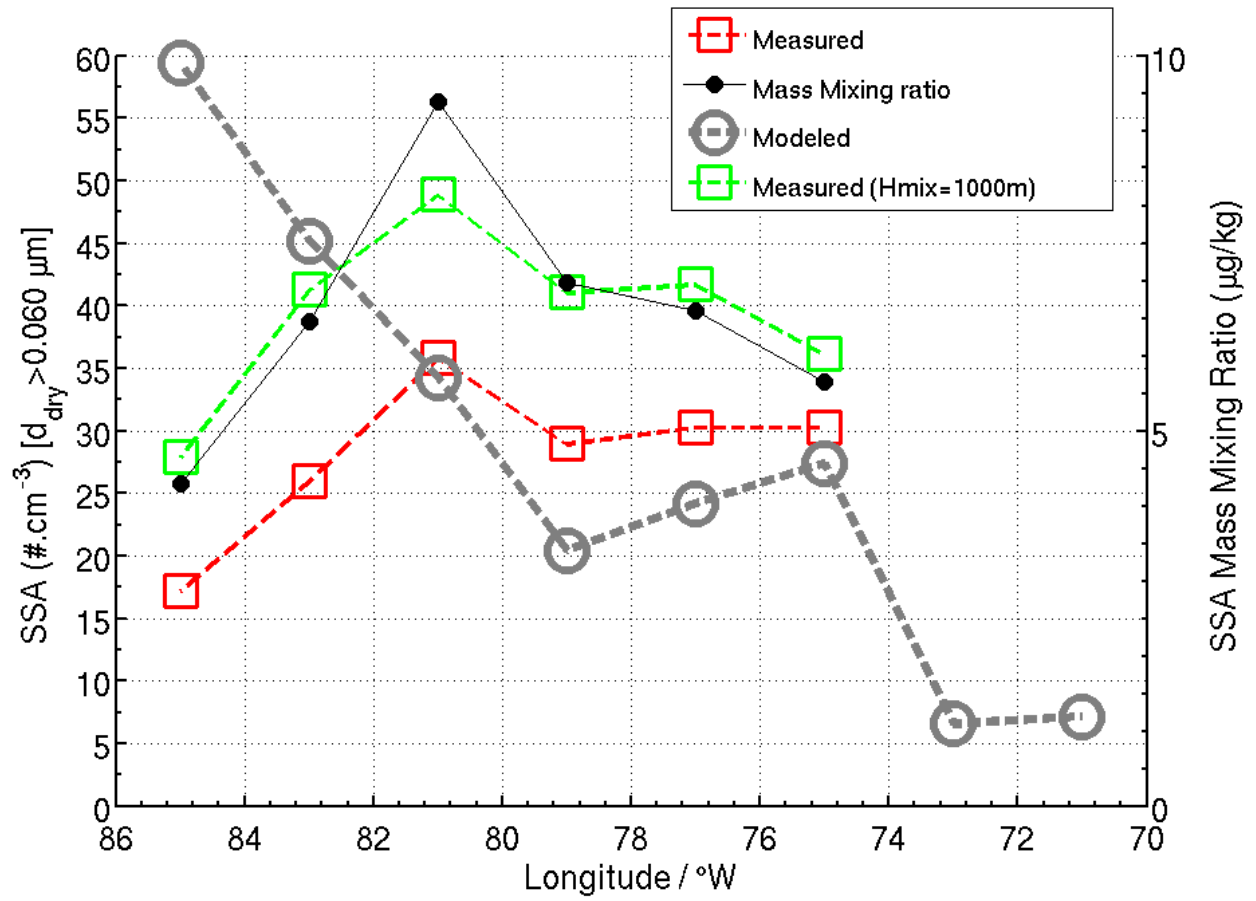


Fig 6a :

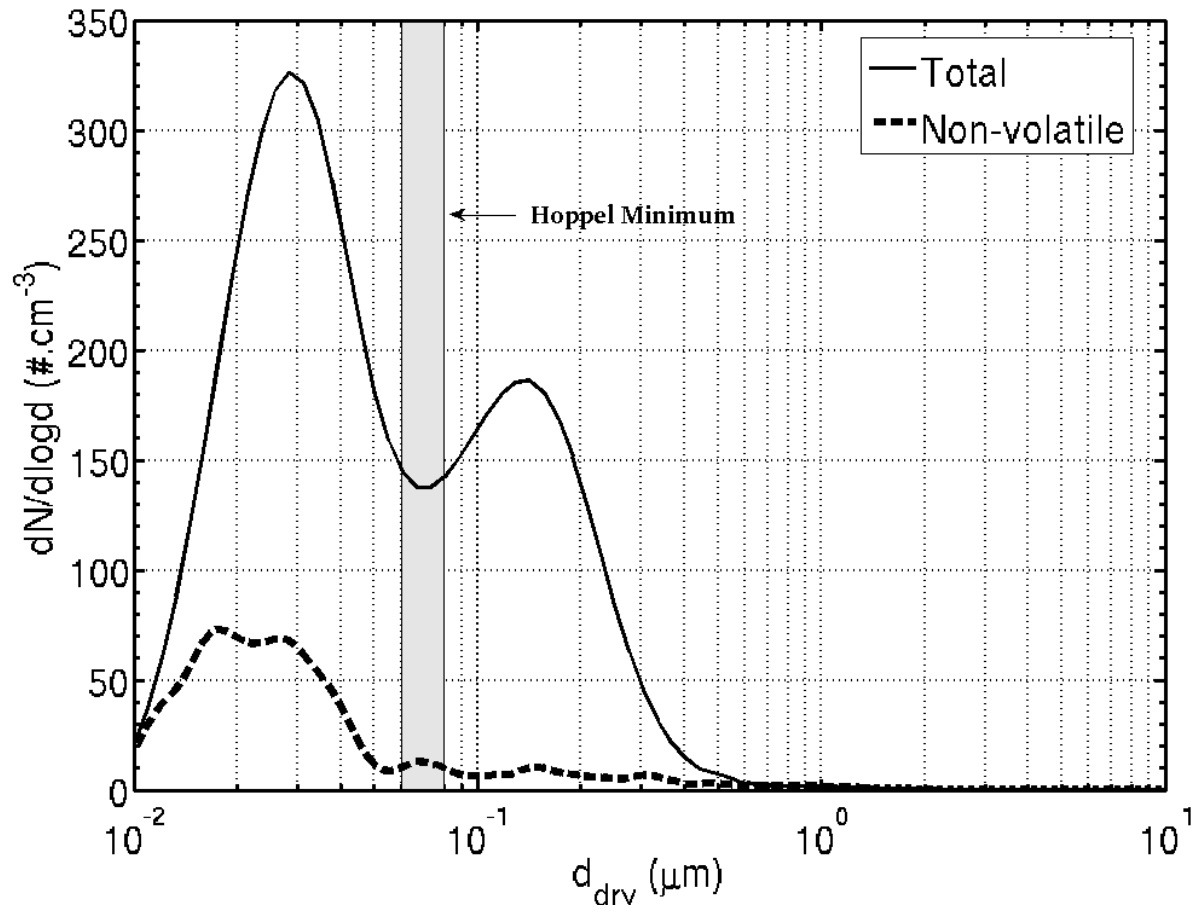


Fig. 6b :

