Referee #1

The authors thanks the anonymous referee #1 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 paper addresses the issue of the source of CCN over the "clean" marine boundary layer and how to parameterize this source in global models. The paper is very thorough and methodically takes the reader through the steps needed to show their conclusions (data quality, data stratification, data comparisons, processes controlling concentrations). I recommend publication with minor revisions to address the questions below:

We thanks the referee for his positive general overview of the manuscript.

Specific Comments:

The paper assumes two sources of CCN and that the particles in the MBL are an external mixture of these two sources. Are there any data from the C-130 that show this external mixture? What can be said about the particles smaller than CCN size? Are these mostly SSA? Are these cores on which CCN particles grow?

Reply : In Fig 7b. we found that refractory aerosols with a minimum size of 0.040 μm vary with light scattering at 700 nm for clean air . Given our clean air criteria, this argues strongly for these particles being SSA. We do not say anything about smaller particles since we cannot be sure that non-volatile sizes below 0.040 μm (p 3294) are SSA even under clean conditions. Figure 6a shows most particles are volatile upon heating. We do not show heated and unheated mass distributions in this paper but if we did, the sizes that dominate the volume distribution in Figure 6b do not change their sizes upon heating while those that dominate the number (Fig. 6a) do. Certainly most of the larger sizes are externally mixed. Smaller SSA will be emitted as externally mixed but will age to become internally mixed over time and some will go on to become CCN before being removed. However, as shown in Fig.12b, most CCN in this region are volatile and not SSA. We have tandem DMA data that tends to show a larger fraction of the mass is volatile for the smaller selected sizes but we do not discuss this in this paper. [also see Fig. 2 in Clarke and Porter (1993) "Pacific marine Aerosol 2…" for details Jour. Geophys. Res., 98, pg 16,997].

We estimate CCN sizes based on ambient supersaturation inferred from the Hoppel minimum from a given cloudy region. For example, far offshore (85 ºW) activation of SSA sizes as small as 0.040 μm imply a supersaturation near 0.42%. However, these are only estimated from averaged size distributions that had undergone enough cloud processing cycles to develop a well resolved Hoppel Minimum. In reality, on a cloud scale, both higher/lower supersaturations and higher/lower updrafts will be present and account for the smoothly varying width of the Hoppel minimum present in the distributions. It is also likely that SSA will become increasingly mixed with other species (eg. sulfate) depending upon their age since formation (residence time) and the strength of processes and sources of

other material (coagulation, condensations, collision-coalescence, etc...).

Page 3280 Line 12: what if you don't limit the org concentration? What if this org is part of the SSA?

Reply : One key result of our study is the well established correlation (R^2=0.87) between ultrafine DMA non-volatile aerosol and light scattering coefficients for clean air (Fig 7b.). If organic concentrations are not constrained for very low values, this correlation coefficient become 0.48. We do not exclude that some organics are part of the SSA population over the SEP. However, as also found in Shank et al. 2012, we did not manage to identify a background value for marine organics that would be independent of combustion indicators.

Page 3282 Line 22: what do you mean here by sea-salts. Isn't the controversy whether the sub 0.1 um aerosols are inorganic or organic?

Reply : That is correct. The fraction of inorganic sea-salts and the fraction of organics that composed the submicrometer SSA are still uncertain.

Page 3288 Line 17: Do you mean >.087 and <0.4?

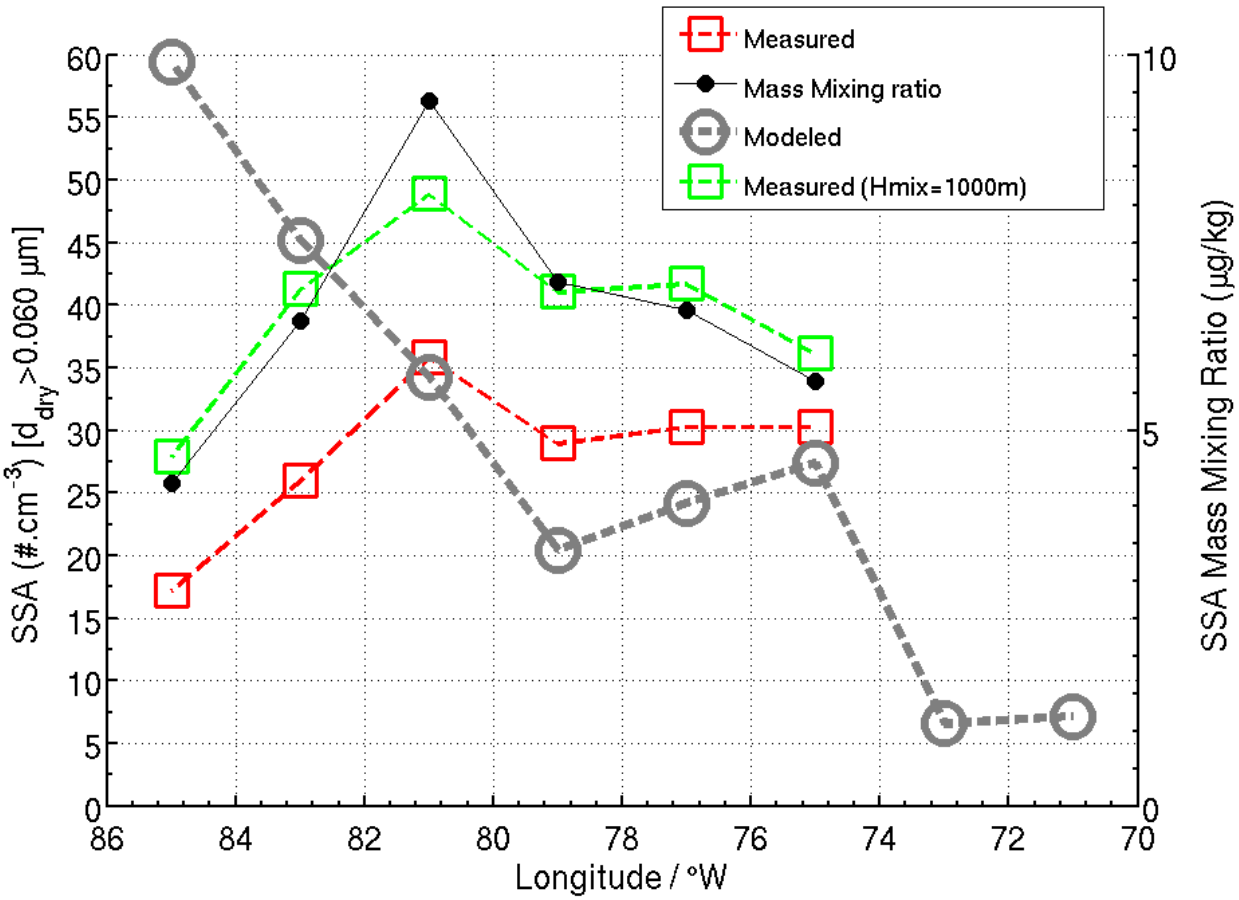
Reply : That is correct. The sentence will be updated with :" … found a minimum and a maximum particle size detection limit of 0.087 µm and 0.4 µm.

<u>Technical corrections</u> :

We thanks the referee for all the comments made below. All corrections will be done in the final manuscript.

Page 3280 Line 11: organic aerosol
Page 3281 Line 8: dimethyl sulfide
Page 3282 Line 2: there instead of their
Page 3283 Line 23: remain instead of remains
Page 3285 Line 6: two pollution survey missions along the coast of .....
Page 3285 Line 7: The non-POC missions ....
Page 3288 Line 16: Data were
Page 3288 Line 20: Flight not flights
Page 3295 Line 4: becomes
Page 3295 Line 12: these DMS data
Page 3300 Line 6: these results imply
Page 3301 Line 6: that there is no obvious
Figure 4: total number of data points
Figure 11: there are no triangles

Fig. 11 :


Measured
Mass Mixing ratio
Modeled
Measured (Hmix=1000m)
SSA (#.cm-3) [ddry>0.060 µm]
SSA Mass Mixing Ratio (µg/kg)
Longitude / °W
60
55
50
45
40
35
30
25
20
15
10
5
0
10
5
0
86
84
82
80
78
76
74
72
70