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Title: Large contribution of organics to condensational growth and formation of cloud condensation nuclei (CCN) in remote marine boundary layer

We thank the anonymous referees for their valuable and constructive comments/suggestions on our manuscript. We have revised the manuscript accordingly and please find our point-to-point responses below.

Comments by Anonymous Referee #1:

General Comments:

Zheng et al present a long time series of CCN measurements in the remote marine boundary layer. Measurements of this kind are rare, especially during nucleation events. The authors systematically characterize a series of nucleation events to calculate the hygroscopicity parameter for the condensing material. Surprisingly, they find that it is much lower than that for sulfate as would be predicted. These measurements provide some of the first direct evidence that condensation of organic material in the marine boundary layer contributes to particle growth with important implications for CCN. The paper is well written and should be published in ACP. I have only a few comments:

Detailed Comments:

1) Are there any direct gas-phase measurements that can be used to support the airmass characterization efforts? Even CO would be helpful!

Responses:

Long-term trace gas measurements at the ENA site include CO and O_3, both of which come mainly from long-range transport (Zheng et al., 2018). Following the reviewer's suggestion, we compared the mixing ratios of CO and O_3 of different air mass origins (Fig. R1). Both CO and O_3 mixing ratios are the highest in continental air masses and lowest for mid-latitude ocean air masses, supporting the airmass classification.

We've added this information into the modified manuscript (Line 229-230 in the revised manuscript):

"Here we classify the origin of air mass during the growth events into four types: (1) continental air masses from North America or Europe, (2) the Arctic, (3) the subtropical, and (4) the mid-latitude Atlantic. … The mixing ratios of CO and O_3 measured at the ENA site exhibit the highest and lower values for the continental and mid-latitude ocean airmasses, respectively (Fig. S3), supporting the effectiveness of the classifications."

And Fig. R1 is added as Fig. S3 in modified SI.

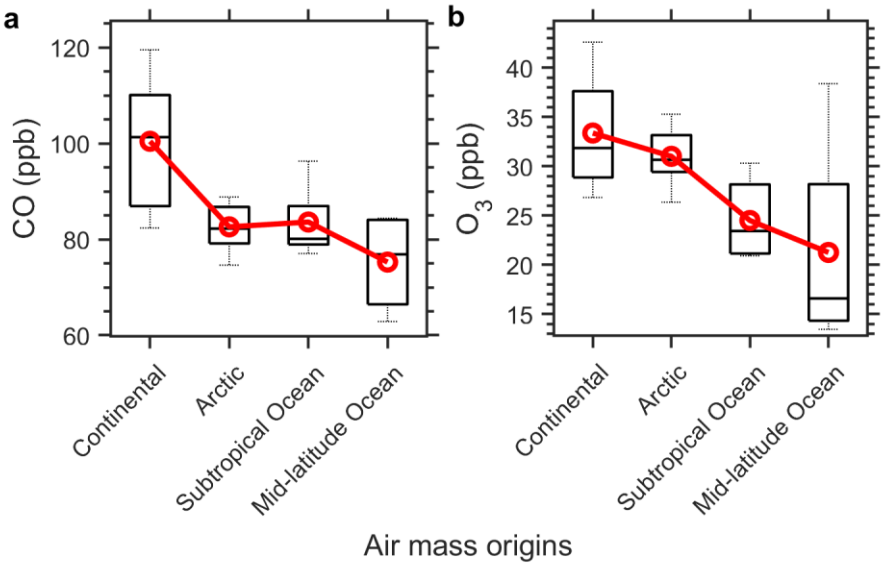

Fig. R1 *(added as Figure S3 in updated SI)* **Trace gas mixing ratios in different air mass origins.** (a) CO and (b) O_3.

2) It would be helpful if the authors could include some context for what the kappa value is for different potential condensing species (e.g., BVOC oxidation products, MSA, carbonyl compounds which may dominate the photo-chemical VOC pathway).

Responses:

Previously we've listed this information in Table S1. To make the relevant information clearer, we've moved Table S1 into the modified manuscript as Table 1.

Table R1 ***(added as Table 1 in updated manuscript)*****: Hygroscopicity parameter κ of potential condensing species over remote oceans**

Compound	κ_{GF}	κ_{CCN}	Reference

H_2SO_4	1.19	0.90	(Petters and Kreidenweis, 2007)
NH_4HSO_4	1.0	0.9	(Schmale et al., 2018)
$(NH_4)_3H(SO_4)_2$	0.51	0.65	(Petters and Kreidenweis, 2007)
$(NH_4)_2SO_4$	0.53	0.61	(Petters and Kreidenweis, 2007)
CH_3SO_3H (MSA)	0.36	<0.44	(Johnson et al., 2004; Tang et al., 2019)
α−pinene/O_3/dark SOA	0.022~0.037	0.1	(Petters and Kreidenweis, 2007)
β−pinene/O_3/dark SOA	0.009~0.022	0.1	(Petters and Kreidenweis, 2007)
SOA particles generated via OH radical oxidation	0~0.3 (20% to 50% lower than corresponding κ_{CCN})	0~0.3 (Generally below the line of: (0.29 ± 0.05)*O:C	(Chang et al., 2010; Massoli et al., 2010)

3) Is the MSA-SO2 yield really constant in MERRA-2 or does it still depend on temperature as that dictates the branching between H-abstraction and OH addition in DMS+OH? This is what yields distinct MSA/SO2 ratios. Is it possible to simply use the concentration of SO2 as an indicator?

Responses:

As pointed out by the reviewer, the DMS branching ratio depends on many factors, including the temperature (Arsene et al., 2001; Hynes et al., 1986; Williams et al., 2001; Yin et al., 1990). However, to the best of our knowledge, a constant DMS branching ratio (SO_2:MSA = 75:25) is employed in MERRA-2 for simplicity (Chin et al., 2000; Randles et al., 2017), as in the standard GEOS-Chem model (Chatfield and Crutzen, 1990; Chin et al., 1996). Besides the branching ratio, several factors also contribute to the variation of the MSA/SO_2 concentration ratio, including other sources of SO_2 and the sinks of MSA and SO_2. We note that the MSA/SO_2 concentration ratio is substantially lower than any reported branching ratio, and one likely explanation is that SO_2 from other sources, including long range transported continental emissions, and emissions from volcanos and shipping, contributes substantially to the variation of MSA/SO_2 concentration ratio.

The average hygroscopicity of the condensed species, κ_c, is expected to strongly correlate with the volume ratios of condensed organics and inorganics (e.g., sulfate), instead of their absolute concentrations. In this study, MSA/SO_2 ratio is used as a surrogate of the relative abundance of condensing biogenic secondary

organics and sulfates, which is expected to anti-correlate with κ_c. We don't expect the concentration of SO_2 as an effective indictor, as SO_2 concentration alone does not correlate well with the volume ratio of condensed organics and inorganics, and thus κ_c.

4) Is it possible to extract from the growth rates any information on the concentration of condensing species? This would help in the comparison with the magnitude of BVOC ocean emissions (e.g., is 10ppt steady-state monoterpene sufficient to sustain this type of growth?)

Responses:

We thank the reviewer for the suggestion. Extracting the concentration of condensing species and precursor BVOC requires a number of parameters, including the concentrations of oxidants, existing aerosol Fuchs surface area, the volatility of the condensing organics, and the activity coefficients of the organics in the growing particles. A systematic study on the concentration of condensing species and precursors is beyond the scope of this manuscript, but is the focus of our future work.

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