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## ***Interactive comment on* “Boundary layer new particle formation over East Antarctic sea ice – possible Hg driven nucleation?” by R. S. Humphries et al.**

### **Anonymous Referee #2**

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The manuscript “Boundary layer new particle formation over East Antarctic sea ice – possible Hg driven nucleation?” presents a data set of limited gas and aerosol-phase measurements to support a hypothesis of mercury driven new particle formation. The data set was collected over a 32-day long field campaign and measured aerosol concentrations at 2 size thresholds and atmospheric gaseous composition data from a GC-ECD, MAXDOAS, and O<sub>3</sub> analyzer. During the campaign, a single 4-hour long period of elevated solar radiation occurred approximately 2 hours before a new particle formation event. While no aerosol nanoparticle or cluster compositional data is available, the authors present a well-reasoned hypothesis that gaseous mercury is the

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cause of the new particle formation event. The authors present a novel particle formation mechanism that may occur in remote regions which is well within the scope of Atmospheric Chemistry and Physics.

The authors suggest that mercury, rather than volatile organics, halogens, or sulfur, is responsible for the observed particle formation event. Due to the lack of aerosol speciation ability, there is no proof that the authors' proposed mechanism is the cause of this particular nucleation event. However, the authors attempt to rule out the alternative causes of the event and present generally sound reasoning in doing so. While I believe this manuscript should be accepted for publication, there are a number of extremely important points that need to be further discussed. Generally these points revolve around the authors' dismissal of sulfur and halogens in the particle formation event, two keystones of the authors' proposal of mercury driven particle formation. Addressing these points is critical and publication should be contingent on successful explanation of these major concerns.

Major comments:

1) Sulfur – The reliance on a box model, and an admitted simple model at that, to discount the importance of sulfur in the particle formation event may be acceptable if certain criteria are met. However, it is unacceptable as it currently stands in the manuscript. First, no results of the box models are explicitly stated or presented. There is no indication if the models were orders of magnitude off of reproducing observations nor even which direction the models were off in (over or under prediction). Additionally, even if the box model results are described in better detail, a thorough discussion regarding the limitations of the model used needs to be included. In particular, a thorough literature review regarding the state of the science of DMS/MSA reactions in polar regions and an overview of the reaction schema in TOMAS, would be helpful in determining the plausibility of using the model. Furthermore, the pulse of DMS (p. 19489, Lines 17-20) is not discussed in the results section. The authors admit a pulse followed by zero emissions is unlikely to represent reality and do not then make an attempt to

model a scenario with different DMS output conditions.

2) Halogens – The manuscript brushes off Bromine chemistry by saying that it is “within the noise of the day” and that the Bromine concentration is not as high as previous BrO events. However, the data, specifically the MAX-DOAS, is not presented in a thorough enough manner to convincingly discredit such a well-observed new particle source. At the very least, the MAX-DOAS data needs to be presented consistently and thoroughly throughout the manuscript. For example, column BrO (presented in molec/cm<sup>2</sup> indicating it is not total column BrO as the caption suggests) is presented in Fig. A1 but BrO mixing ratios in ppt are discussed in the manuscript. “The noise of the day” should also be discussed in more depth. Additionally, a discussion on the MAX-DOAS, its limitations, and its sensitivities needs to be included.

The whole paper relies on halogens and sulfur being discounted but the paucity of data and discussion presented is concerning. Iodine is the only convincingly discredited halogen. The dismissal of Chlorine because Bromine is a faster reaction should also be backed up through model results at the least.

#### Minor Comments

1) HYSPLIT – HYSPLIT is an extremely useful tool. However, it is only as good as the data set which feeds it. Unfortunately, the data set for the southern ocean is sparse and HYSPLIT results must be interpreted very carefully. At the very least, there needs to be a discussion about the uncertainties associated with using HYSPLIT near Antarctica. I believe the other metrics for identifying a single air mass are satisfactory for this paper but the discussion on air mass history comes with very large asterisks.

2) P19499, Line 2: should “long” be “longer”?

3) P19504, Line 12: What is the errant “3.6.4”?

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