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Comment on acp-2021-507

Anonymous Referee #1

Referee comment on "The contribution of coral reef-derived dimethyl sulfide to aerosol burden over the Great Barrier Reef: a modelling study" by Sonya Fiddes et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-507-RC1, 2021

Review of Fiddes et al, The contribution of coral reef-derived dimethyl sulfide to aerosol
burden over the Great Barrier Reef: a modelling study

Short-term regional effects of DMS from coral on aerosol-cloud interactions in October 2016 are studied in the WRF-chem model. While previous observation studies have attributed boundary layer new particle formation events to emissions from the Great Barrier Reef, including coral DMS emissions in WRF-chem made no difference to the aerosol-cloud interactions.

As they point out in their introduction, the authors already published a global modeling study in ACP this year with the message that there are no substantial effects on global climate of coral DMS emissions. In their conclusion, they also commented 'reefs likely play little role in *regional* climate modulation. As coral reefs globally face extinction due to anthropogenic climate change, it is unlikely that the subsequent reduction in precursor aerosols will have a noticeable impact on regional climate.'

This second paper is in some sense a test of the hypothesis quoted above, and the authors find some evidence to support their hypothesis.

Apart from the global scope of the previous study, I note that this previous work was done with a different model (ACCESS-UKCA). The evaluation of DMS concentrations in this current paper uses novel measurements (if my very brief literature search didn't miss anything). The simulations highlight the importance of the updated DMS inventory. The trajectory analysis is also new and valuable. While the end results of the two papers are similar, the methods used are thus very complementary (as expected for global vs short-term regional modeling studies).

I encourage the authors to highlight the differences and new perspectives from the current study when comparing it to their previous work in a revised version.

The paper is well written and the data analysis seems robust and well-explained in my assessment.

I think an appropriately revised version of the paper could readily be suitable for

publication in ACP. I have the following suggestions for improvements.

Major comments

Is this actually the first paper documenting the field campaign, or using measurements from it? If so, the authors could make more of the novelty of these results and put the DMS measurements and field campaign into a broader context.

A fuller description of the meteorology during the case study would be helpful. L250/Figure 5: is the boundary layer height simulated or observed? Were there radiosondes you could use for evaluation of the vertical temperature profile? Do the simulations get the boundary layer height right? Is there a strong inversion that distinguishes the boundary layer from the free troposphere? If so can you explain the lack of clear variation of sulfate aerosol mass across the boundary layer on the left of Figure 6? If not, why split up the boundary layer and free troposphere in Figure 8?

Uncertainties and shortcomings of the study are only briefly discussed in the conclusions of the paper. I recommend a separate section with some more quantitative analysis and sensitivity studies.

In particular, nucleation mechanisms and aerosol microphysics are relevant to the author's comments in the introduction about boundary layer nucleation events from coral, and should be discussed in more detail. Could model shortcomings explain why the authors' simulations did not show up any boundary layer nucleation events?

- Aerosol size distribution.

The authors correctly point out a major shortcoming of the model they use, its crude representation of the aerosol size distribution, at line 341. This could be discussed in more detail. The uncertainties associated with what happens below 40nm (or even much smaller aerosol sizes) in an aerosol model are discussed by Lee et al (2013; https://gmd.copernicus.org/articles/6/1221/2013/gmd-6-1221-2013.html) and Blichner et al (2021 https://acp.copernicus.org/preprints/acp-2021-151/). The configuration of WRF-chem with more size bins, for example as used by Matsui et al (2011, https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2011JD016025) or Zhao et al (2020 https://www.pnas.org/content/117/41/25344) or the regional configuration of the Unified Model with GLOMAP aerosol as per Gordon et al (2018 https://acp.copernicus.org/articles/18/15261/2018/) would have resolved aerosols down to around 3nm in size as in Fiddes et al (2021). I think the authors need to explain in the paper why these models were not used (is the chemistry in UM-UKCA too simple for example? Are the configurations of WRF-chem with more bins too expensive?), and/or quantitatively compare the size distributions from WRF-chem with the size distributions from GLOMAP in Figure 7 of Fiddes et al (2021) in the relevant area.

- Nucleation mechanisms

The section on nucleation pathways did not refer to the substantial corpus of prior work on the mechanisms for, or observations of, atmospheric new particle formation. Mechanisms were also not discussed in detail in the introduction. The brief discussion on nucleation presented at line 346 in the conclusion could be much more comprehensive and presented earlier.

I think the nucleation scheme of Wexler et al (1994) the authors use is the parameterization of Jaeckel-Voirol and Mirabel (1989), which is based on classical nucleation theory. Zaveri et al (2008) points out that no ternary nucleation scheme is included in WRF-chem (the authors' comment on that in this paper could be easily misconstrued). Therefore, there is no participation in nucleation of stabilizing compounds which could substantially increase new particle formation rates, such as ammonia, amines or methanesulfonic acid (e.g. Brean et al 2021 https://www.nature.com/articles/s41561-021-00751-y), all of which are likely present at some level near the Australian coast.

These stabilizing mechanisms are especially important at high temperatures where molecular clusters readily evaporate. Therefore, the importance of boundary layer DMS to marine CCN number concentration may be underestimated. Or, indeed, the concentration of CCN could be overestimated if there are too many nuclei acting as sinks for vapors, and therefore the particles cannot grow large enough to act as CCN- see Sullivan et al (2018; https://www.nature.com/articles/s41612-018-0019-7). I note that Sullivan et al also uses WRF-chem with additional size sections.

Maybe the authors would find the reviews of new particle formation mechanisms by Lee et al (2019 https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2018JD029356) and models by Semeniuk and Dastoor (2018; https://www.sciencedirect.com/science/article/abs/pii/S1352231018300530?via%3Dihub) and papers cited therein helpful to expand their discussion.

I think some sensitivity studies where the nucleation mechanism and/or treatment of nucleation-mode microphysics is varied, and some discussion of the associated uncertainties, may be interesting additions to the paper.

Minor comments

L23 missing citation, perhaps to Merikanto et al (2010)?

L196 correlations between what and what? Presumably observations and model, but please specify.

L246 how DMS has changed because of what? Presumably because of including the DMS source from coral, but please specify. Also should reiterate that these changes are (if I am not mistaken) simulated changes, not observed changes.

L275 a decrease in sulfate compared to what? The area closer to the coast? Please specify.

Appendix: Should explain mechanisms for aerosol-cloud interactions more carefully. How is the second indirect effect parameterized?

Figure 7 the blue diamonds look like circles to me