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Interactive comment on "Coral reef origins of atmospheric dimethylsulfide at Heron Island, southern Great Barrier Reef, Australia" *by* Hilton B. Swan et al.

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Referee #1

Comment 1: Figure 1b is really a great figure and compelling for the point of the paper, but not discussed at all.

Reply: Fig. 1b is mentioned on P6, L11 to pictorially describe the Capricorn Bunker Group of coral reefs to the SE of Heron Island. Fig. 1b was not discussed in the context of low-level cloud formation over these coral reefs because it is a contentious image among atmospheric scientists. While there may be broad agreement that convective processes have contributed to cloud alignment over the Capricorn Bunker Group of

coral reefs shown in the MODIS image, we provide insufficient evidence to conclude that atmospheric DMS (DMSa) derived from those reefs is the source of those clouds. In our manuscript we provide evidence that the Heron Island reef flat and the Capricorn Bunker Group of reefs can at times be significant sources of DMSa. Those spikes of DMSa can potentially contribute to development of clouds over the GBR; however, as stated in the final sentences of the conclusion "the extent to which DMSa contributes to aerosol production and its role in CCN formation over the GBR is currently unknown. Aerosol formation and evolution studies are, therefore, required to determine if the GBR is a climatically important source of marine aerosol." This is the crux of the matter now requiring further investigation. After reading our manuscript we would like the reader to draw their own conclusion regarding the cloud cover over the reefs shown in Fig. 1b.

Another referee of the manuscript has recommended deleting Fig 1, stating the figure is not necessary to understand the information in the paper, and that it does not provide important new information. This contrasting comment possibly alludes to the contentious nature of the images shown in Fig 1. We wish to promote discussion by presenting this figure, but do not want to draw conclusions beyond what the data we collected allows. The conclusions we have made are in line with the objective of the study stated in the final sentence of the introduction.

Comment 2: The wind compass in Figure 2 is a bit confusing. It would be easier to read and comprehend if it was in a wind rose format.

Reply: The radar compass plot was used in Fig 2 because it provided the clearest representation of the frequency of wind directions for the two seasons when plotted together. When the wind direction data for both seasons is plotted on a wind rose style plot, as shown in Response Fig1, it is difficult to distinguish the seasonal differences because they were minor. What we want to convey by including the compass plot in Fig 2 is to show that there was little difference in the directional frequency of the winds at Heron Island in both the wet and dry seasons.

Comment 3: The description of the GC sampling could be more detailed. I see that the authors cited a previous methods paper, I would still like to know more about this particular experiment. What size tubing? How fast was the sample pumped? How was it pumped? Why was the air trapped 14 mins?

Reply: In order to minimise the length of Section 2: 'Methods', a reference is provided to a 2015 publication that gives a complete description of the instrumentation and its measurement uncertainty. We appreciate that the referee would like more methodological information provided in this results focussed paper, so it will be included in the revision. Answers to the particular questions posed are as follows: Marine air was drawn through 6 mm internal diameter Teflon tubing via a high-capacity oxidant scrubber composed of 1% w/v sodium ascorbate and glycerol impregnated into a 47 mm diameter glass-fibre filter. Surface-level marine air was drawn through the sampling system at a flow rate of approximately 260 mL min-1 using a single-stage diaphragm vacuum pump (Vacuubrand, model ME2, Germany). The air was drawn into a cryogenically cooled trap (cryotrap) that was constructed by passing 1.6 mm diameter Teflon tubing through $\sim$50 cm of copper tubing of 2.0 mm internal diameter, and bending it into a loop. A sample collection time of 14.4 min was used to deliver 3.72 L of air into the cyrotrap. This volume of air was required to concentrate sufficient DMSa for chromatographic analysis to provide a 0.1 nmol m-3 (2 ppt) reporting limit. Calibration was achieved using permeated ethyl methyl sulfide.

Comment 4: I don't like the units of nmol/m3 and appreciate when ppt units also mentioned. Can this be done throughout?

Reply: The photochemical ambient mass balance Eq. 1 used to determine seasonal DMS emission flux requires that DMSa molar concentrations are entered to determine flux in the usual units of μmol m-2 d-1. This is the main reason why DMSa molar concentrations are reported throughout the manuscript. We are aware that DMSa concentrations are dependent on pressure and temperature according to the ideal gas law, and that mixing ratios are more suitable than concentrations to describe the abun-

dance of species in air, particularly when samples are collected in aircraft over various altitudes. All the DMSa measurements we made were at sea level under relatively consistent temperature and pressure. Additionally, the DMSa concentration of nmol m-3 used throughout the manuscript is a SI unit. In contrast, ppt is not an SI unit, and it is recommended by IUPAC that the SI unit of pmol mol-1 is used in place of ppt notation. However, we understand that it is customary for trace species in air to be reported in ppt because familiarity with this unit allows many readers to readily compare abundances of trace gaseous species. We would like to present SI concentrations throughout the manuscript by reporting our surface DMSa measurements in nmol m-3 to maintain consistency with emission fluxes reported in μmol m-2 d-1. Nevertheless, Table 1 will be edited to include the corresponding ppt mixing ratios for the DMSa concentrations given in the table. The limit of reporting of 2 ppt will also be given in brackets next to the concentration in Section 2.1 (P3, L17); however, we think it will clutter the manuscript text to report ppt mixing ratios in brackets next to every concentration mentioned in Section 3: 'Results and Discussion'.

Comment 5: Style of the discussion sections seems a bit off - I miss a general description of the results. Instead, the manuscript seems to delve right into the spikes and the effect of tides.

Reply: An introductory paragraph that provides a general description and summary of the results will be included at the beginning of Section 3: 'Results and Discussion' in the revised manuscript. This introductory paragraph will not repeat information given in Section 4: 'Conclusions'.

Comment 6: How do the authors know that the large spike found during the dry season (shown in figure 6a) is not an instrumental problem? Why should there only be biological shock during the dry season (it seems that there are periods during the wet season also without rainfall and with low tide)?

Reply: The intense DMSa spike detected during the dry season campaign was not

an instrumental problem. As stated in the manuscript, there was an intense odour of DMS at the time the spike was detected, which was evident to a number of people who remarked about the unusually strong “marine odour” coming from the platform reef surrounding the island. Although the DMSa spike was relatively sharp it was not derived from a single measurement. The oceanic background DMSa concentration on 25 July 2013 ranged from 1.0-1.8 nmol m-3 over the entire day prior to the intense DMSa spike in the early evening at 17:50 when the DMSa rapidly rose to 45.9 nmol m-3. The following DMSa measurement was 9.7 nmol m-3 at 18:14, which tapered off from 2.7 nmol m-3 at 18:38 to the preceding background concentration of 1.5 nmol m-3 by 21:24. As mentioned in the discussion, this DMSa spike was brief due to rapid dilution by strong horizontal advection under a wind speed of 9.5 m s-1 at the time of the spike. These conditions require higher resolution sampling to adequately capture this sort of DMSa reef emission, which is one of the reasons why it is recommended in the conclusion that chemical ionisation mass spectrometry could assist further studies of DMSa emissions from the GBR.

We are not saying in the manuscript that biological shock to the reef will only occur during the dry season. It just happened to be during the 2013 dry season campaign that we detected the particularly intense DMS emission that appeared to be brought about by coincidence of environmental conditions that were unfavourable to the coral reef. These conditions included spring low tides which aerially exposes more of the reef for longer periods than neap low tides, coinciding with a brief shower of rain onto the reef flat at the time when the reef was most exposed. Such conditions could occur at other times of the year. Spring low tides occur during December-January in the wet season, so it is possible that intense emissions of DMS could occur at that time if the exposed reef was showered by rainfall. In a previous study on the GBR in February (austral summer) elevated seawater DMS concentrations (54 nM) were measured during low tide when there was 20 minutes of rainfall, which reduced the seawater salinity by 0.75 PSU (Jones et al., 2007, Environmental Chemistry, doi:10.1071/EN06065). This situation is likely to have released a wet season DMSa spike. As is evident from the entire

winter dataset we show here, the intensity of the DMSa spike detected in the early evening of 25 July 2013 was a unique event, and it was good fortune to be on-site at that time with equipment to detect and quantify it.

The referee comments that there are periods during the wet season also without rainfall and with low tide. Fig. 3a shows a period between 8 March and 17 March 2012 when there was only a few convective derived short showers. Low tide during periods of dry weather resulted in few detectable DMSa spikes from the coral reef. In contrast, the convective shower that coincided with low tide in the evening of 14 March produced the second largest DMSa spike (10.6 nmol m 3) recorded during the late summer wet season campaign. This indicates that rain on the reef in the summer season can result in a DMSa spike similar to the spike detected during the winter campaign. The reason that the DMSa spike during the winter was much more intense than the one detected during the summer is not clear, but it may have to do with factors such as the level of the low tide, the time that the coral had been aerially exposed, the temperature of rain that fell on the reef and the resulting surface seawater temperature. It was apparent that it was not the amount of rainfall but when the rainfall occurred that led to detectable DMSa spikes from the coral reef.

Comment 7: Why didn’t the authors play a bit with their results, for example looking at forward trajectories to see where the DMS ends or scaling up to the entire GBR?

Reply: A few forward trajectories were made to observe air mass transport away from Heron Island. The reason these forward trajectories were prepared was to examine where Wedge-tailed Shearwaters (Mutton Birds) that nest on Heron Island might be going to find food. The forward trajectories indicated that if they flew with the wind they would often travel to the Swain Reefs, an extensive reef system to the north of Heron Island (seen in Fig. 2). If this forward trajectory analysis is extended to transport of DMSa spikes from Heron reef it is possible that the DMSa oxidation products might contribute to CCN formation over the Swain Reefs. We do not present evidence in the manuscript to support this possibility and the lead author is reluctant to extrapolate or

scale up our results obtained at Heron Island on the southern GBR to the entire 2,300 km length of GBR. The GBR stretches 13 degrees of latitude along the Queensland coastline and it is risky to assume that processes operating in the northern GBR are the same as those on the southern GBR. The stark contrast in the extent of coral bleaching on the northern GBR compared to the minimal bleaching on the southern GBR during the summer of 2015-16 is a recent pertinent example of differences that can occur along the length of the GBR. Our manuscript was previously submitted to another journal and it was criticised by each referee for the suggestion that the coral reef DMSa spikes observed at Heron Island on the southern GBR may occur over the entire GBR leading to formation of low-level marine clouds that possibly constitutes a regional climate feedback. At the present time that hypothesis remains speculative and much more research is required to determine if the GBR is a climatically influential source of marine aerosol, as stated in the last sentence of the conclusion.

Comment 8: How was H (or MLD??) actually determined? And why do the authors call it MLD? This is terminology used more for water mixed layers. Why not always say H? Or are these values somehow different?

Reply: Atmospheric MLDs were obtained using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) transport and dispersion model developed by NOAA's Air Resources Laboratory, as stated in Section 2.2 of the manuscript. The model, which is described by Stein et al (2015), uses sophisticated computations of atmospheric transport and mixing.

The referee may be more familiar with the MLD when used in the marine context; however, meteorologists and atmospheric scientists also use this terminology to refer to the region of the lower troposphere immediately above the surface where there is nearly constant potential temperature and specific humidity with height. As in the ocean, this atmospheric zone is characterised by turbulence resulting in a stable vertical temperature profile. Given that the MLD terminology may present confusion for marine scientists, the atmospheric MLD will be referred to as the mixed layer height (MLH)

in the revised manuscript, representing the height above the surface of the convective mixed layer or the convective boundary layer.

The MLH is the major part of the marine boundary layer (MBL), which is the height of the atmospheric mixed layer from the ocean surface to a capping inversion, referred to in the manuscript as the entrainment zone. The boundary between the convective mixed layer below and the warmer layer above is marked by the base of the clouds. In Eqn. 1, H describes the mean height of the MBL during each campaign, where as the MLH refers to the height of the MBL at noon for each day during each campaign. Thus, H and MLH are not interchangeable descriptors in the manuscript.

The authors thank the referee for commenting on the manuscript to improve its content.

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Response Fig.1