



OPEN ACCESS

EDITED BY

Rafel Simó,
Institut de Ciències del Mar
(ICM-CSIC), Spain

REVIEWED BY

Michael Kingsford,
James Cook University, Australia
Zhenming Ji,
Sun Yat-sen University, China

*CORRESPONDENCE

Rebecca L. Jackson
rebecca.jackson@csiro.au

†PRESENT ADDRESS

Rebecca L. Jackson,
Coasts and Ocean Research, Oceans
and Atmosphere, Commonwealth
Scientific and Industrial Research
Organisation, Canberra, ACT, Australia

SPECIALTY SECTION

This article was submitted to
Coral Reef Research,
a section of the journal
Frontiers in Marine Science

RECEIVED 01 April 2022

ACCEPTED 19 July 2022

PUBLISHED 12 August 2022

CITATION

Jackson RL, Woodhouse MT,
Gabric AJ, Cropp RA, Swan HB,
Deschaseaux ESM and Trounce H
(2022) Modelling the influence of
coral-reef-derived dimethylsulfide on
the atmosphere of the Great
Barrier Reef, Australia.
Front. Mar. Sci. 9:910423.
doi: 10.3389/fmars.2022.910423

COPYRIGHT

© 2022 Jackson, Woodhouse, Gabric,
Cropp, Swan, Deschaseaux and
Trounce. This is an open-access article
distributed under the terms of the
[Creative Commons Attribution License
\(CC BY\)](https://creativecommons.org/licenses/by/4.0/). The use, distribution or
reproduction in other forums is
permitted, provided the original
author(s) and the copyright owner(s)
are credited and that the original
publication in this journal is cited, in
accordance with accepted academic
practice. No use, distribution or
reproduction is permitted which does
not comply with these terms.

Modelling the influence of coral-reef-derived dimethylsulfide on the atmosphere of the Great Barrier Reef, Australia

Rebecca L. Jackson^{1,2*†}, Matthew T. Woodhouse³,
Albert J. Gabric², Roger A. Cropp², Hilton B. Swan⁴,
Elisabeth S. M. Deschaseaux⁴ and Haydn Trounce⁵

¹Coasts and Ocean Research, Oceans and Atmosphere, Commonwealth Scientific and Industrial Research Organisation, Canberra, ACT, Australia, ²School of Environment and Science, Griffith University, Nathan, QLD, Australia, ³Climate Science Centre, Oceans and Atmosphere, Commonwealth Scientific and Industrial Research Organisation, Aspendale, VIC, Australia, ⁴Centre for Coastal Biogeochemistry, Faculty of Science and Engineering, Southern Cross University, Lismore, NSW, Australia, ⁵International Laboratory for Air Quality and Health, School of Earth and Atmospheric Sciences, Queensland University of Technology, Brisbane, QLD, Australia

Marine dimethylsulfide (DMS) is an important source of natural sulfur to the atmosphere, with potential implications for the Earth's radiative balance. Coral reefs are important regional sources of DMS, yet their contribution is not accounted for in global DMS climatologies or in model simulations. This study accounts for coral-reef-derived DMS and investigates its influence on the atmosphere of the Great Barrier Reef (GBR), Australia, using the Australian Community Climate and Earth System Simulator Atmospheric Model version 2 (ACCESS-AM2). A climatology of seawater surface DMS (DMS_w) concentration in the GBR and an estimate of direct coral-to-air DMS flux during coral exposure to air at low tide are incorporated into the model, increasing DMS emissions from the GBR region by 0.02 Tg yr^{-1} . Inclusion of coral-reef-derived DMS increased annual mean atmospheric DMS concentration over north-eastern Australia by 29%, contributing to an increase in gas-phase sulfate aerosol precursors of up to 18% over the GBR. The findings suggest that the GBR is an important regional source of atmospheric sulfur, with the potential to influence local-scale aerosol-cloud processes. However, no influence on sulfate aerosol mass or number concentration was detected, even with a reduction in anthropogenic sulfur dioxide emissions, indicating that DMS may not significantly influence the regional atmosphere at monthly, annual or large spatial scales. Further research is needed to improve the representation of coral-reef-derived DMS in climate models and determine its influence on local, sub-daily aerosol-cloud processes, for which observational studies suggest that DMS may play a more important role.

KEYWORDS

coral reef, dimethylsulfide (DMS), sulfate, aerosol, ACCESS

1 Introduction

Aerosols and clouds play a key role in the Earth's radiative budget and climate by governing the amount of incoming and outgoing solar radiation and heat (Ramanathan et al., 1989; Andreae, 1995). Since the industrial revolution, anthropogenic greenhouse gas (GHG) emissions have exerted an average global radiative forcing of $+2.72$ (1.96 to 3.84) W m^{-2} (IPCC, 2021). Anthropogenic and natural aerosols have partially offset this warming effect through scattering of incoming short-wave radiation (aerosol-radiation interactions) (McCormick and Ludwig, 1967) and influencing the albedo, lifetime and cover of clouds (aerosol-cloud interactions) (Twomey et al., 1984).

In comparison to GHG emissions, the influence of aerosols on the climate is complex and less well quantified due to uncertainties in aerosol source strengths and their radiative effects (Twomey, 1974; Andreae and Rosenfeld, 2008). Consequently, estimates of aerosol radiative forcing range from -1.9 to -0.1 W m^{-2} (IPCC, 2013; IPCC, 2014). Approximately 45% of the variance in aerosol radiative forcing since the pre-industrial period is derived from uncertainties in natural aerosol sources, including marine dimethylsulfide (DMS) (Carslaw et al., 2013).

The precursor of DMS, dimethylsulfoniopropionate (DMSP), is produced throughout the ocean by a number of marine organisms including planktonic and benthic algae (Stefels, 2000; Sunda et al., 2002), corals and their endosymbiotic dinoflagellates (Raina et al., 2013), and marine bacteria (Curson et al., 2017). DMSP is produced for a range of physiological and ecological functions (Stefels, 2000; McParland and Levine, 2019). Catabolism of DMSP is mediated by the demethylation and cleavage pathways, with the latter producing DMS (Bullock et al., 2017). Seawater surface DMS (DMS_w) is ventilated to the marine boundary layer (MBL) where it is rapidly oxidized, primarily by hydroxyl radicals (OH), to sulfate aerosol precursor gases including sulfur dioxide (SO_2), methanesulfonic acid (MSA), hydroperoxymethyl thioformate (HPMTF) and sulfuric acid (H_2SO_4) (Andreae and Crutzen, 1997; Berndt et al., 2019; Hodshire et al., 2019; Veres et al., 2020).

Sulfuric acid, an aerosol precursor, may condense onto existing particles, facilitating growth to cloud condensation nuclei (CCN), or nucleate to form new non-sea salt sulfate (nss- SO_4) aerosol (Andreae and Crutzen, 1997; Woodhouse et al., 2013). The latter primarily occurs in the free troposphere, where background aerosol concentration and temperature are relatively low and conditions are optimal for the nucleation of trace gases (Korhonen et al., 2008; Sanchez et al., 2018). New nss- SO_4 particles may be entrained from the free troposphere to the MBL and increase the number of CCN and cloud droplets.

When high concentrations of fine aerosol grow rapidly to CCN, cloud droplet number increases and cloud droplet diameter decreases (assuming constant cloud liquid water

content), enhancing the albedo and lifetime of clouds and suppressing precipitation (Rosenfeld et al., 2007; Dave et al., 2019). Conversely, when CCN activation size is not met, water vapor may remain in the atmosphere, suppressing local precipitation and increasing precipitation downwind if conditions for particle growth are more favorable (Andreae and Rosenfeld, 2008; Fan et al., 2018).

The influence of DMS on Earth's radiative balance is dependent on the efficiency of DMS oxidation and nucleation to secondary nss- SO_4 , which ranges from 0.14-0.95 in the MBL (Von Glasow and Crutzen, 2004). The high range in efficiency is due to regional variability in pre-existing aerosols and the aerosol condensational sink (Woodhouse et al., 2013; Hoffmann et al., 2016). The annual mean contribution of DMS to CCN is estimated to be 3.3% in the Northern Hemisphere and 9.9% in the relatively cleaner Southern Hemisphere (Woodhouse et al., 2010). This response varies with season and location, with the greatest response occurring in pristine regions of high biological activity (Korhonen et al., 2008; Lana et al., 2012; Gabric et al., 2013; Woodhouse et al., 2013; Sanchez et al., 2018; Zavarisky et al., 2018). For example, when phytoplankton productivity (and DMS production) increases, DMS-derived sulfates contribute up to 46% of CCN over the Southern Ocean (30 - 45°S) in summer (Korhonen et al., 2008), and 33% of CCN over the North Atlantic in spring (Sanchez et al., 2018) *via* new particle nucleation and particle growth to CCN.

Coral reefs are identified as important regional sources of DMS (Broadbent and Jones, 2004; Swan et al., 2017; Jones et al., 2018). In the coral holobiont, DMS(P) production is upregulated in response to physiological stress caused by exposure to high sea surface temperature (SST), solar irradiance and changes in salinity (Raina et al., 2013; Deschaseaux et al., 2014; Gardner et al., 2016; Hopkins et al., 2016), all of which can be exacerbated during exposure to air at low tide (Buckee et al., 2020). This biogenic sulfur source could facilitate nss- SO_4 aerosol nucleation and growth to CCN, increasing the lifetime and albedo of low-level clouds (LLC) over coral reefs (similarly to Charlson et al., 1987). Evaporation over shallow, warm coral reef waters contributes to the formation of a convective boundary layer (~ 65 - 130 m), with relatively high humidity and temperature that is favourable for low-level cloud formation (McGowan et al., 2019). It has been hypothesized that DMS emissions facilitate the formation of a local or regional negative feedback within the coral reef boundary layer, shading and cooling the coral reef below (Fischer and Jones, 2012; Jones, 2015; Jones et al., 2017).

Global modelling studies have found that marine DMS is an important source of nss- SO_4 aerosol, influencing regional aerosol and cloud microphysical properties (Thomas et al., 2010; Woodhouse et al., 2010; Gabric et al., 2013; Mahajan et al., 2015) and providing a cooling effect of up to 0.45°C (Fiddes et al., 2018). However, with the exception of only two model studies (Fiddes et al., 2021, 2022), the coral reef source of

DMS is not currently accounted for in DMS climatologies or in climate models.

Global climate models typically prescribe DMS_w concentrations using the Kettle et al. (1999) or Lana et al. (2011) monthly mean climatologies. These climatologies were derived from more than three decades of DMS_w observations over most of the global ocean (Kettle et al., 1999). However, very few observations were included for coral reef regions and extrapolation did not account for seasonal or spatial variability across coral reef flats and lagoon waters.

Only two model studies have investigated the importance of DMS flux from coral reefs on the climate (Fiddes et al., 2021; Fiddes et al., 2022). Both of these studies accounted for the coral reef source of DMS_w by adding a scaled laboratory-derived estimate of 50 nmol L^{-1} to the Lana et al. (2011) DMS_w climatology for coral reef regions. This method added 0.03 nmol L^{-1} to the global average seawater concentration and increased global sea-air emissions by 0.3 Tg yr^{-1} of DMS, representing 1.7% of global DMS emission estimates (Fiddes et al., 2021). This is higher than the estimate of $0.06\text{--}0.08 \text{ Tg yr}^{-1}$ of DMS estimated to be released from tropical coral reefs based on field observations (Jones et al., 2018; Jackson et al., 2021).

Despite the relatively high coral reef DMS_w and DMS sea-air flux source, Fiddes et al. (2021) report only a small response in nucleation and Aitken mode aerosol number concentration and mass when the coral reef source of DMS was removed from the global ACCESS-UKCA (Australian Community Climate and Earth System Simulator-United Kingdom Chemistry and Aerosol) model. This suggests that tropical coral reef DMS emissions have no robust impact on contemporary global or regional climate, possibly due to confounding impacts of industrial sulfate emissions and other anthropogenic pollutants dominating the aerosol signal. Fiddes et al. (2022) demonstrated similar findings in the GBR region using the regional WRF-Chem (Weather Research and Forecasting model coupled with chemistry) model. However, observational studies suggest that the coral reef source of DMS may influence sulfate aerosol and LLC properties, at spatial or temporal scales that are perhaps not captured by global or regional climate models (Modini et al., 2009; Swan et al., 2016).

For example, positive correlations have been identified between the number concentration of atmospheric particles ranging from $0.5\text{--}2.5 \mu\text{m}$ (wet diameter) and both DMS sea-air flux (Deschaseaux et al., 2022) and DMS_a concentration (Jackson et al., 2020) at Heron Island in the southern GBR. The magnitude of the correlation between DMS_a and particle number concentration increased during daylight hours, when DMS oxidation rates over relatively clean oceans are highest (Gabric et al., 2008; Gali et al., 2013), and when wind speed was low ($< 2 \text{ m s}^{-1}$) (Jackson et al., 2020). Although the observed size range was larger than newly nucleated particles, the positive correlation during calm, daylight hours may have reflected local condensational growth of existing fine particles ($< 0.5 \mu\text{m}$) to the larger, detectable size range (Jackson et al., 2020). Therefore,

DMS-derived sulfates may play an important role in aerosol composition over the GBR.

A parameterization of DMS_w in the Great Barrier Reef (GBR), Australia, was recently derived from observational data obtained during Marine National Facility RV *Investigator* voyage IN2016_V05 (RVI). This parameterization was used to calculate a 19-year climatology of DMS_w and DMS flux from the GBR using remotely sensed observations (Jackson et al., 2021). The calculated DMS_w climatology reproduced observed seasonal and spatial variability well in the GBR (summarized in Jones et al., 2018) and estimates that the $347,000 \text{ km}^2$ of coral reefs and lagoon waters in the GBR release $0.03\text{--}0.05 \text{ Tg yr}^{-1}$ DMS ($0.015\text{--}0.026 \text{ Tg yr}^{-1} \text{ S}$) (Jackson et al., 2021).

Here, we build on previous modelling work by investigating the influence of coral-reef-derived DMS on the contemporary and a relatively clean regional atmosphere. Coral-reef-derived DMS is accounted for by incorporating a climatology of DMS_w calculated from observational data in the GBR, and an estimate of direct coral-air DMS flux, into the Australian Community Climate and Earth-System Simulator Atmospheric Model version 2 (ACCESS-AM2). The accuracy of DMS sources in the model is evaluated by comparing model output with observations, and the influence of coral-reef-derived DMS on the regional atmosphere over north-eastern Australia is quantified.

2 Materials and methods

2.1 ACCESS-AM2 description

ACCESS-AM2 is a global physical climate model, which contributed to the World Climate Research Programme's Coupled Model Intercomparison Project Phase 6 (CMIP6). The physical atmospheric model in ACCESS is the Met Office Unified Model (UM) version 10.6 (Walters et al., 2019). This version uses the Global Atmosphere 7.1 (GA7.1) configuration and includes the Global Model of Aerosol Processes (GLOMAP) aerosol scheme (Mann et al., 2010; Mann et al., 2012).

GLOMAP-mode resolves aerosol mass, number concentration, size distribution, composition, and optical properties. Prognostic aerosol species included in GLOMAP-mode are sulfate, black carbon, organic carbon and sea salt, which are internally mixed within five modes corresponding to soluble nucleation (dry diameter $< 5 \text{ nm}$), Aitken (dry diameter $5\text{--}50 \text{ nm}$), accumulation (dry diameter $50\text{--}500 \text{ nm}$), coarse (dry diameter $> 500 \text{ nm}$) and insoluble Aitken mode. Processes simulated within GLOMAP-mode include primary emissions, new particle nucleation, particle growth *via* coagulation, condensation and cloud processes, particle removal by dry deposition, and in-cloud and below-cloud scavenging (Mann et al., 2010; Mann et al., 2012). Oxidant concentrations necessary for the sulfur cycle are prescribed from monthly varying inputs.

Atmospheric variables are resolved at a horizontal resolution of 1.25° latitude \times 1.875° longitude (~ 135 km \times ~ 200 km resolution in the low-latitudes), with 85 vertical levels (50 below 18 km and 35 above) reaching a model top of 85 km. Further details on the physical parameterizations in UM GA7.1 are described in detail in [Walters et al. \(2019\)](#). ACCESS-AM2 uses an identical implementation, of which a detailed description is provided in [Bi et al. \(2020\)](#). In this work, an atmosphere-only (ACCESS-AM2) configuration is used ([Bodman et al., 2020](#)).

2.2 Experimental design

2.2.1 Seawater surface DMS concentration

In ACCESS-AM2, DMS_w is prescribed from the global [Kettle et al. \(1999\)](#) monthly mean climatology, henceforth K99 ([Figure 1A](#)). As discussed above, the K99 climatology was derived from DMS_w observations over most of the global ocean ([Kettle et al., 1999](#)). However, very few observations were included for coral reef regions and extrapolation did not account for seasonal or spatial variability across coral reef flats and lagoon waters. For this analysis, K99 is modified to include a climatology of DMS_w derived from an empirical relationship in the GBR ([Jackson et al., 2021](#)), henceforth referred to as the GBR climatology. The GBR climatology calculated in [Jackson et al. \(2021\)](#) (0.25 -degree grid) was scaled up to the model grid by binning the values to a resolution of 1.25×1.875 degrees. K99 was

then modified by substituting pixels within the GBR region (10.5 - 25° S; 142 - 154° E) with the GBR climatology, which added an average of 0.5 nmol to the GBR seawater surface concentration ([Figure 1C](#)).

The GBR climatology was derived from an empirical relationship between DMS_w , SST and PAR, measured during the RVI surveys from 28 September to 24 October 2016. The RVI surveys were undertaken as part of the ‘Great Barrier Reef as a significant source of climatically relevant aerosol particles’ project, known as the ‘Reef to Rainforest’ (R2R) campaign. The RVI path is shown in [Figure 1A](#). The calculated GBR DMS climatology agrees with the range of observed seasonal and spatial (lagoon versus reef flat) seawater DMS concentrations at several locations in the GBR (summarized in [Jones et al., 2018](#)) and is considered to be a reasonable representation of DMS_w produced by corals and other marine organisms in GBR waters.

2.2.2 DMS sea-air and coral-air flux

DMS sea-air flux is calculated using the [Liss and Merlivat \(1986\)](#) parameterisation ([Figure 1B](#)). Current DMS sea-air flux parameterisations do not account for direct release of DMS from coral mucous when the reef is exposed to air during low tide. Coral aerial exposure is an important, albeit intermittent, source of atmospheric DMS (DMS_a) ([Andreae et al., 1983](#)), that can lead to rapidly released plumes of DMS_a over coral reefs ([Jones et al., 2007](#); [Swan et al., 2017](#)). DMS_a plumes over aerially

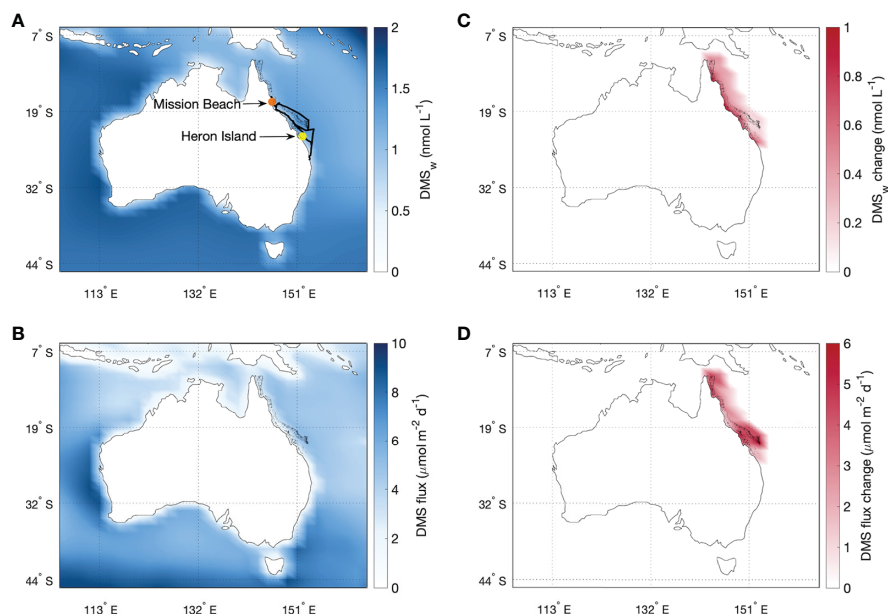


FIGURE 1

Annual mean (A) DMS_w and (B) DMS sea-air flux based on the K99 climatology, and the change in each variable due to the inclusion of (C) the GBR DMS climatology and (D) the GBR DMS climatology and coral-air DMS flux. The RVI path (black), Mission Beach (orange) and Heron Island (yellow) survey locations are shown in (A).

exposed coral reefs are significantly more concentrated than the background DMS_a signal, for which the seasonal average ranges from approximately 1 nmol m^{-3} (~25 ppt) in winter to 4 nmol m^{-3} (~100 ppt) in summer over the GBR (Swan et al., 2017).

It has been estimated that *Acropora* corals exposed to air release $9\text{--}35 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$ (mean $22 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$), according to measurements of gas-phase DMS_a in chamber experiments containing *Acropora* corals exposed to air (Hopkins et al., 2016). To account for direct coral-air DMS flux, the mean of the Hopkins et al. (2016) laboratory-derived estimate was added to the modelled DMS sea-air flux, scaled by the percentage of coral reef cover (i.e. coral-air flux = reef fraction \times 22). The fraction of reef cover was calculated as the number of reef pixels within a 0.25 -degree grid (as determined in Jackson et al., 2021), using a database of coral reef locations obtained from ReefBase (<https://www.refbase.org>) in MATLAB R2020a. The estimate of coral cover across the GBR ranged from $0.01\text{--}34\%$ within the 0.25 degree-resolution grid.

The estimate of coral-air DMS flux used in Jackson et al. (2021) was scaled up to the model grid by binning the mean values to a resolution of 1.25×1.875 degrees. The resulting coral-air DMS flux estimate ranged from $0.2\text{--}2.6 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$ and added an average of $1.4 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$ of DMS to the regional sea-air flux. Our estimate may be a conservative representation of coral-air DMS flux, given that it is two orders of magnitude lower than the $0.01\text{--}0.02 \text{ Tg yr}^{-1}$ estimate of Hopkins et al. (2016).

The approach used to estimate coral-air DMS flux is limited as it assumes that *Acropora* spp. are the sole source of DMS and it does not account for variability in the extent of coral exposure or the complexity of the reef environment (Hopkins et al., 2016). Further research is needed to reduce the uncertainty in coral-air DMS flux, to accurately scale laboratory-derived fluxes to the natural coral reef environment, and to account for diurnal variability in DMS coral-air flux and DMS_a concentration with variability in tidal cycles and photo-oxidation rates, respectively. Nevertheless, inclusion of the coral-air DMS flux estimate provides a more accurate estimate of total DMS flux from the GBR.

Incorporating both the GBR climatology and coral-air DMS flux into the model increased DMS flux from the GBR by an average of $3 \text{ } \mu\text{mol m}^{-2} \text{ d}^{-1}$ (Figure 1D); equivalent to 0.02 Tg yr^{-1} of DMS. In this study, coral-reef-derived DMS refers to the total contribution of DMS from corals and other organisms within the GBR, and includes the GBR DMS_w climatology and coral-air DMS flux estimates described above.

2.3 Analysis

Simulations were run for a 12-month period from January to December 2016, to coincide with three surveys undertaken

as part of the R2R campaign in February and September–October 2016. Meteorology in the model was nudged to the ERA-5 data set (Hersbach et al., 2020). Although nudging dampens meteorological responses, this method allows simulated responses to be attributed to a change in an independent variable (e.g. DMS_w and coral-air DMS flux), while eliminating the need to run computationally expensive, long-term simulations to account for internal model variability. Nudging also allows the best comparison with field observations, by minimising uncertainty associated with meteorology.

As part of the R2R campaign, measurements were taken on board the RVI from 28 September to 24 October 2016. During the same period, shore-based measurements were made using the Atmospheric Integrated Research Facility for Boundaries and Oxidative Experiments (AIRBOX) mobile atmospheric chemistry laboratory at Mission Beach (17.82°S ; 146.12°E) from 20 September to 16 October 2016. An overview paper describing these two field campaigns and datasets is currently in preparation (Trounce et al.). A prior survey was undertaken at Heron Island (23.44°S ; 151.91°E) on the southern GBR from 5 to 18 February 2016 (Swan and Jones, 2017; see also Deschaseaux et al., 2022). The RVI path, Mission Beach and Heron Island survey locations are shown in Figure 1A. A brief description of these surveys undertaken for the R2R campaign and methods used are provided in the Supplementary Information (SI).

To investigate whether the model output represents observations, modelled DMS_a (parts per trillion or pmol mol^{-1}) and wind speed (m s^{-1}) are compared with hourly mean observations for the RVI, Mission Beach and Heron Island survey periods and coordinates (± 0.05 degrees). Given the coarse model resolution, modelled DMS_a and wind speed were first interpolated to a 0.1 -degree ($\sim 10 \text{ km}$) grid using a linear interpolation in MATLAB v2020a.

2.3.1 Experiment 1

For the control simulation, DMS_w was prescribed from the oceanic K99 climatology (Figure 1A). For the experimental simulation, DMS_w in the GBR region is prescribed from the GBR climatology and includes the estimate of coral-air DMS flux (henceforth GBR DMS). The change in annual mean DMS_a , gas-phase SO_2 and H_2SO_4 (ppt), sulfate aerosol mass (ppt), the number concentration of aerosol in the four soluble GLOMAP size modes (m^{-3}), aerosol with dry diameter greater than 3 nm (N3) (cm^{-3}), CCN with dry diameter greater than 70 nm (CCN70) (cm^{-3}) and cloud droplets (Nd) (m^{-3}), and surface downwelling short-wave radiation (SWR) (W m^{-2}) is then quantified between the control and experimental simulations over north-eastern Australia ($9.5\text{--}26^\circ\text{S}$; $135\text{--}155^\circ\text{E}$). The analysis focused on this region to capture changes due to the inclusion of coral-reef-derived DMS.

2.3.2 Experiment 2

Given the global and Australian Government initiatives to shift towards renewable energy, anthropogenic sulfur emissions may decline in future. Therefore, it is important to understand the relative importance of biogenic and anthropogenic sulfate aerosol sources, and the effects of a change in emissions on the aerosol system. The influence of coral-reef-derived DMS in a relatively clean atmosphere is investigated by repeating the control and experimental simulations described above, with high level anthropogenic SO₂ emissions from north-eastern Australia removed from the model. ‘High level’ anthropogenic SO₂ in ACCESS-AM2 includes 100% of energy sector emissions, and 50% of industrial emissions. Given that the focus of this study is on atmospheric sulfate, other anthropogenic emissions (e.g. black carbon) were not modified, but could also have an impact.

3 Results

3.1 Comparison of observed and modelled data

Simulations using K99 and GBR DMS performed similarly in predicting observations of DMS_a at the local, hourly scale (Figures 2A–C). Both K99 and GBR DMS sources overestimated observed DMS_a during the R2R surveys (Table 1). Given that the nudged model simulations predicted observed wind speed moderately well ($r > 0.5$, $p < 0.001$) (Figures 2D–F), the differences between observed and modelled DMS_a are largely due to DMS_w and coral-air DMS flux sources in the model. Nevertheless, modelled DMS_a was within the range of concentrations previously reported for the GBR, which range from approximately 50–200 ppt (mean ~100 ppt) in summer

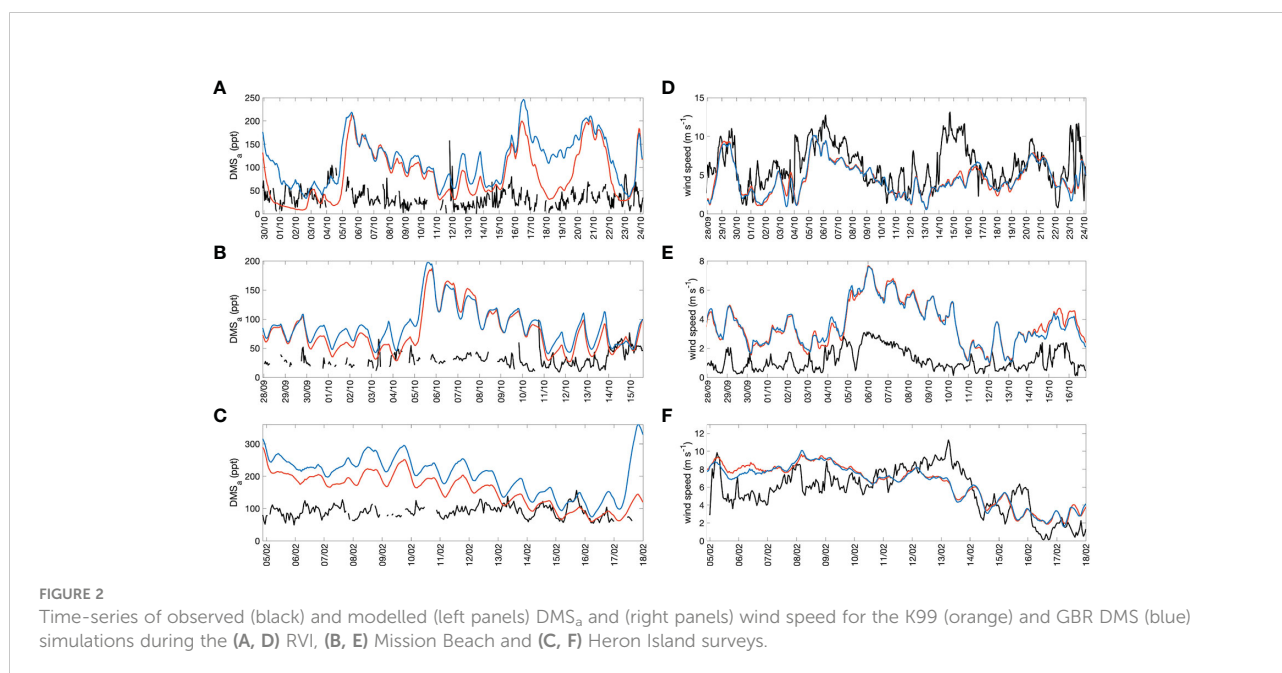


TABLE 1 Mean and range of observed and modelled DMS_a (ppt or pmol mol⁻¹) during the RVI, Mission Beach and Heron Island surveys of the R2R campaign [concentration (deviation from observed mean)].

		Mean	Range
RVI (n = 456)	Observed	33	<1 – 158
	K99	86 (+ 53)	8 – 213
	GBR DMS	114 (+ 81)	33 – 245
Mission Beach (n = 296)	Observed	31	9 – 99
	K99	80 (+ 49)	27 – 187
	GBR DMS	89 (+ 58)	41 – 195
Heron Island (n = 274)	Observed	90	49 – 156
	K99	157 (+ 67)	60 – 289
	GBR DMS	199 (+ 109)	74 – 316

(Jones et al., 2007; Swan et al., 2017; Swan and Jones, 2020; 2017) and can exceed 500 ppt over aerially exposed coral reefs (Jones et al., 2007; Swan et al., 2017). Therefore, the DMS_w climatologies in the model are assumed to represent the upper range of observed DMS in the GBR.

DMS_a concentration varies with the rate of DMS sea-air flux (which is primarily dependent on seawater surface DMS concentration, SST and wind speed) (Yang et al., 2011), coral-air DMS flux (Swan et al., 2017) and photochemical processes such as production of OH and subsequent oxidation of DMS_a to sulfate aerosol precursor compounds (Ayers and Gillett, 2000; Barnes et al., 2006). Consequently, observed and modelled DMS_a display a diel cycle (Figures 2A–C), decreasing around midday when solar radiation and the abundance of oxidative free radicals are highest (Gabric et al., 2008; Galí et al., 2013; Jackson et al., 2020). Increases in modelled DMS_a often aligned with modelled wind speed, reflecting wind-driven sea-air flux (Figure 2). Occasional pulses in modelled DMS_a occurred for the GBR DMS simulation during low wind speeds, reflecting the addition of coral-air flux (e.g. on 18 February 2016; Figure 2C). Conversely, observations of DMS_a and wind speed did not align well, possibly reflecting advection to and from the measurement platform.

Observations of DMS_a at Heron Island were approximately two-times higher than the RVI and Mission Beach observations, highlighting the variability in DMS concentrations in the GBR. It was expected that observed DMS_a would be highest at Heron Island, given that measurements were taken less than 100 m from the coral reef flat and were more likely to capture the high end of atmospheric concentrations over the reef. Further, the RVI and Mission Beach surveys took place six months after the March–April 2016 mass coral bleaching event on the GBR, which led to the loss of up to 40% of hard coral cover by November 2016 (Hughes et al., 2018). Observed DMS_w (not shown here) and DMS_a were lower during the RVI and Mission Beach surveys than previously reported for the GBR region (Swan and Jones, 2017; Jones et al., 2018, 2020), which may have been in part due to the loss of DMS-producing hard corals. Prior to the 2016 mass coral bleaching event, DMS_a measured at Heron Island ranged from 49–156 ppt (Table 1). However, with the exception of a pulse of DMS_a on 12 October 2016, DMS_a was below 100 ppt during the RVI and Mission Beach surveys (Figures 2A, B).

Considering that the K99 and GBR DMS sources are monthly mean climatologies (and fixed coral-air DMS flux) with no diurnal variation, a degree of variability was expected between the observed and coarse resolution modelled DMS_a . While modelled DMS_a represents the upper range of observations previously reported for the GBR region, simulations using K99 and GBR DMS sources still provide valuable insight into the influence of coral-reef-derived DMS on the regional atmosphere. Further research will improve our understanding of the complex coral reef sulfur cycle and will

further improve the representation of coral-reef-derived DMS in climate models.

3.2 Influence of coral-reef-derived DMS over north-eastern Australia

Inclusion of coral-reef-derived DMS increased annual mean DMS_a concentration over the GBR by approximately 150% (Figures 3A, D), resulting in an area-averaged increase of 29% (29.4 ppt) over north-eastern Australia (Table 2). Similarly, annual mean gas-phase SO_2 and H_2SO_4 increased by a respective 1% and 3% over north-eastern Australia, in part due to the increase in DMS_a over the GBR, where the strongest increase in SO_2 (up to 17.6%) and H_2SO_4 (up to 14.9%) occurred (Figure 3). Regardless of the increase in SO_2 and H_2SO_4 (Figures 3E, F), concentrations over marine areas are negligible in comparison to industrial regions of coastal and inland Australia (Figures 3B, C).

Changes for annual mean sulfate aerosol mass and number concentration over north-eastern Australia were small ($\leq 2.4\%$), yet revealed some interesting results. Nucleation mode sulfate mass and number concentration decreased by a respective -1% and -0.6% over north-eastern Australia, while sulfate mass increased by similar magnitude for the Aitken mode (+0.8%) and to a lesser extent the accumulation mode (+0.01%) (Table 2). A small increase in sulfate mass (+2.4%) and number concentration (+0.5%) also occurred for coarse mode aerosols. The small increase in Aitken and accumulation mode mass without a corresponding increase in number suggests that additional sulfate contributed to the growth of these size modes *via* condensation or coagulation on existing particles.

Negligible changes occurred for N3, CCN70, Nd and surface SWR ($\leq 0.1\%$) that were likely driven by internal model and meteorological variability. While meteorology in the model was nudged to the ERA-5 dataset, minor differences in meteorological fields can still occur between model runs (as occurred in this study for wind speed; see Table 2). A time-series of each variable area-averaged over north-eastern Australia is provided in SI Figure 1. Mapped changes for sulfate aerosol mass, particle number concentrations and SWR are provided in SI Figures 2–4.

Similarly to previous model studies (Fiddes et al., 2021; Fiddes et al., 2022), the weak response to the addition of coral-reef-derived DMS may be due to the dominant influence of anthropogenic sulfur and other emissions from north-eastern Australia (Chen et al., 2019). Anthropogenic sources can dominate aerosol emissions, particularly over coastal and inland regions. Consequently, the dominant pathway for biogenic trace gases in the marine boundary layer is condensation or coagulation onto existing particles, often resulting in aerosol growth rather than new particle nucleation (Woodhouse et al., 2013; Hoffmann et al., 2016).

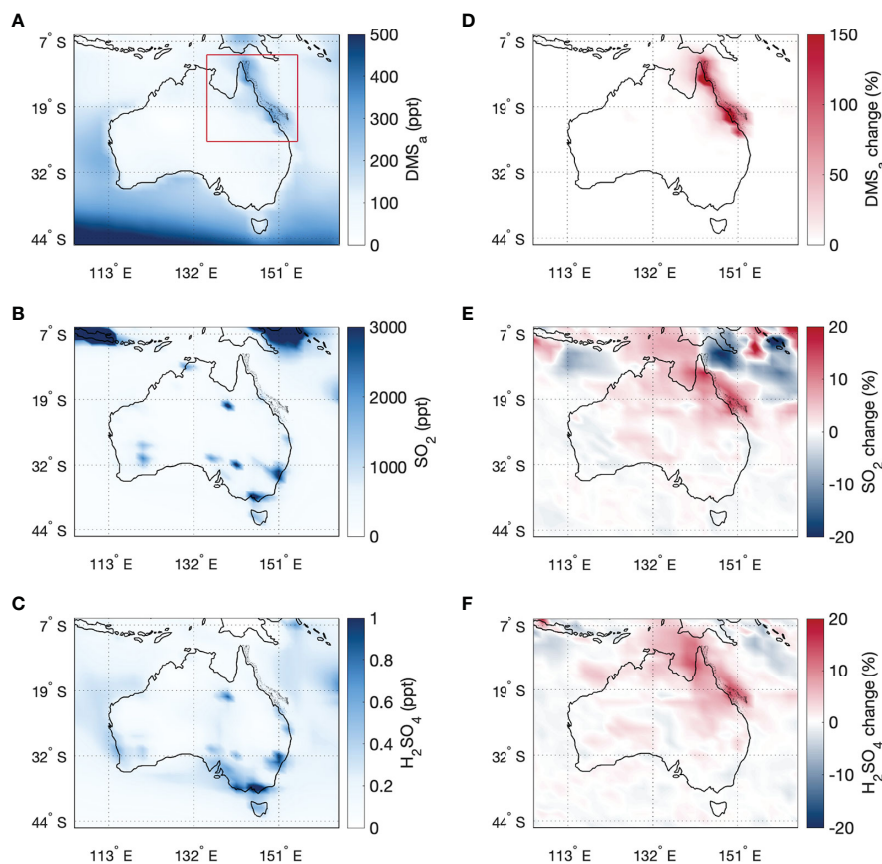


FIGURE 3

Annual mean (A) DMS_a , (B) SO_2 and (C) H_2SO_4 for the GBR DMS simulation, and (D–F) the percentage change in each variable between the GBR DMS and K99 simulations. The north-eastern Australian region ($9.5\text{--}26^\circ\text{S}$; $135\text{--}155^\circ\text{E}$) for which the area-averaged changes are calculated for Table 2 is shown in red in panel (A).

Given the response detected for DMS_a and gas-phase sulfate aerosol precursor compounds (SO_2 and H_2SO_4) at the annual mean time-scale, a stronger response in aerosol mass and number concentration may occur at local or sub-daily time scales as observational studies have suggested (Modini et al., 2009; Swan et al., 2016; Cropp et al., 2018), or with a reduction in anthropogenic pollutants. The latter is investigated below, where the influence of coral-reef-derived DMS is quantified when high-level anthropogenic SO_2 emissions (e.g. from industrial chimneys, shown as hotspots exceeding 2000 ppt in Figure 3B) are removed from north-eastern Australia.

3.3 Influence of coral-reef-derived DMS in a relatively clean atmosphere

After removing high level anthropogenic SO_2 emissions, the inclusion of coral-reef-derived DMS again increased DMS_a , SO_2 and H_2SO_4 concentrations over north-eastern Australia

(Table 2). The increases are similar in space and magnitude to the results presented in section 3.2 above, however are stronger over the GBR (Figure 4). The stronger response is likely due to the addition of DMS-derived sulfur to a reduced pool of atmospheric SO_2 and H_2SO_4 concentrations.

The change in sulfate aerosol mass and particle number concentration between the K99 and GBR DMS simulations were small ($\leq 3.6\%$) (Table 2; SI Figure 5). Sulfate aerosol mass and number concentration decreased for the nucleation and accumulation mode, yet increased for the Aitken mode. For coarse mode aerosols, there was an increase in sulfate mass, without a corresponding increase in number concentration (Table 2). As for section 3.2, the inconsistent changes in sulfate mass and number concentration between aerosol size modes suggests that DMS-derived sulfate may be a more important contributor to aerosol mass and growth of existing aerosols, rather than nucleation of new particles at the monthly and annual mean time-scale, even with a reduction in anthropogenic sulfur emissions.

TABLE 2 Annual mean change [actual (percentage)] between the GBR DMS and K99 simulations area-averaged over north-eastern Australia.

	Experiment 1	Experiment 2
DMS flux ($\mu\text{mol m}^{-2} \text{d}^{-1}$)	+0.6 (25.1%)	+0.6 (25.9%)
DMS _a (ppt)	+29.4 (29.2%)	+29.5 (29.3%)
SO ₂ (ppt)	+5.6 (1.4%)	+3.0 (0.9%)
H ₂ SO ₄ (ppt)	+5.4 × 10 ⁻³ (3.0%)	+6.2 × 10 ⁻³ (4.0%)
Nuc. sul. mass (ppt)	-5.2 × 10 ⁻⁵ (1.0%)	-4.9 × 10 ⁻⁶ (0.1%)
Ait. sul. mass (ppt)	+0.3 (0.8%)	+0.4 (1.2%)
Acc. sul. mass (ppt)	+0.04 (0.01%)	-14.5 (3.6%)
Crs. sul. mass (ppt)	+0.2 (2.4%)	+0.05 (0.7%)
Nuc. no. (m ⁻³)	-1.1 × 10 ⁻²¹ (0.6%)	-1.1 × 10 ⁻²¹ (0.6%)
Ait. no. (m ⁻³)	-1.5 × 10 ⁻²⁰ (0.2%)	+1.1 × 10 ⁻²⁰ (0.2%)
Acc. no. (m ⁻³)	-8.4 × 10 ⁻²¹ (0.1%)	-1.1 × 10 ⁻²⁰ (0.1%)
Crs. no. (m ⁻³)	+2.3 × 10 ⁻²² (0.5%)	-8.6 × 10 ⁻²³ (0.2%)
N3 (cm ⁻³)	-0.3 (0.1%)	+0.3 (0.1%)
CCN70 (cm ⁻³)	-0.03 (0.01%)	-0.3 (0.1%)
Nd (m ⁻³)	+0.07 (0.04%)	-0.3 (0.2%)
SWR (W m ⁻²)	+0.07 (0.03%)	-0.2 (0.1%)
Wind (m s ⁻¹)	-7.3 × 10 ⁻³ (0.1%)	+3.9 × 10 ⁻³ (0.1%)

* Nuc., nucleation; Ait., Aitken; Acc., accumulation; Crs., coarse; sul., sulfate; no., number concentration; SWR, surface downwelling short-wave radiation; Wind, wind speed at 10 m.

4 Discussion

Inclusion of coral-reef-derived DMS added an average of 0.5 nmol L⁻¹ to the sea surface DMS climatology and 3 $\mu\text{mol m}^{-2} \text{d}^{-1}$ (0.02 Tg yr⁻¹) to the DMS sea-air flux for the GBR region. The additional DMS increased modelled annual mean DMS_a concentration by approximately 150% over the GBR and by 29% over north-eastern Australia. The concentration of gas-phase sulfate aerosol precursor compounds (SO₂ and H₂SO₄) increased by up to 17.6% over the GBR, supporting previous findings that the GBR is an important regional source of atmospheric sulfur (Broadbent and Jones, 2004; Jones et al., 2018; Jackson et al., 2021).

The increase in DMS_a and subsequent increase in sulfate aerosol precursors is primarily attributed to the inclusion of

coral-to-air DMS flux. The GBR DMS_w climatology did not substantially increase seawater surface DMS concentration (< 1 nmol L⁻¹; Figure 1C), and likely had a minor contribution to the ~150% increase in DMS_a (Figure 3D). Previous field work has suggested that coral-air DMS flux is a stronger, although intermittent, contributor to DMS_a than sea-air flux in the GBR (Swan et al., 2017), leading to plumes of DMS_a above coral reefs that can exceed 500 ppt (Jones et al., 2007; Swan et al., 2017). This study supports this hypothesis, where an increase in DMS_a, SO₂ and H₂SO₄ was detected over the GBR (Figure 3), primarily due to the inclusion of coral-air DMS flux.

Gas-phase SO₂ may be further oxidised to H₂SO₄, which may condense onto existing particles as the dominant pathway within the marine boundary layer (Woodhouse et al., 2013;

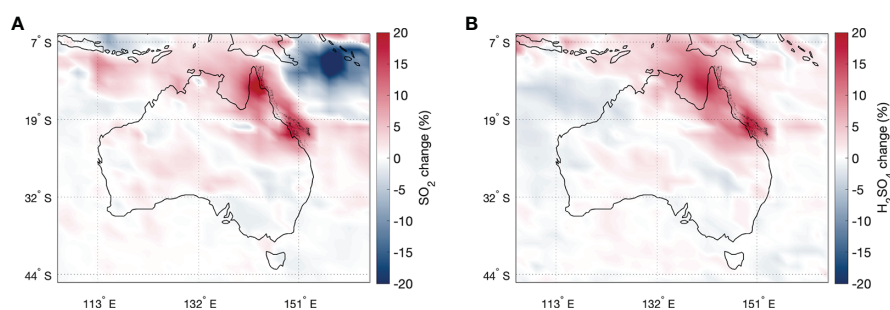


FIGURE 4

Change in annual mean (A) SO₂ and (B) H₂SO₄ between the GBR DMS and K99 simulations, when high anthropogenic SO₂ emissions from north-eastern Australia have been removed.

Hoffmann et al., 2016), or nucleate to form new nss-SO₄ particles (Andreae and Crutzen, 1997). Despite the increase in gas-phase sulfate aerosol precursors, coral-reef-derived DMS had no clear influence on modelled sulfate aerosol mass or number concentration at the monthly or annual mean time-scale, supporting previous model studies (Fiddes et al., 2021; Fiddes et al., 2022) which demonstrate that DMS-derived sulfates do not play an important role in aerosol formation or growth over large temporal and spatial scales. Fiddes et al. (2021) estimated that DMS_w derived from global tropical coral reefs contributed 0.3 Tg yr⁻¹ S to the global sea-air flux in the global ACCESS-UKCA atmospheric model. Despite modelled DMS flux being an order of magnitude higher than the current study, Fiddes et al. (2021) found that coral-reef-derived DMS had only a minor influence on sulfate aerosol mass and number concentration for nucleation and Aitken mode aerosols, and no significant influence on cloud-relevant particles or the Earth's radiative balance. Fiddes et al. (2022) reported similar findings in the GBR region using the regional WRF-Chem model.

Small ($\leq 2.4\%$) and inconsistent changes in sulfate aerosol mass and number concentration over north-eastern Australia in the present study are more likely due to internal model variability and minor changes in meteorology, than to coral-reef-derived DMS. While meteorology was nudged to the ERA-5 dataset, minor differences between model runs can occur. In this analysis, annual mean wind speed at 10 m differed by $\pm 0.1\%$ between the control and experimental simulations (Table 2).

In comparison to the increase in gas-phase sulfate aerosol precursors, the negligible change in aerosol mass and number concentration may be due to the dominant influence of anthropogenic and other natural emissions (e.g. dust) from north-eastern Australia (Chen et al., 2019). To investigate whether the sensitivity of aerosol to DMS increased in a relatively less polluted atmosphere, we repeated the simulations with reduced anthropogenic SO₂ emissions from north-eastern Australia; representing an optimistic future scenario where a shift to renewable energy may reduce anthropogenic pollutants. It was hypothesised that by reducing emissions from industrial hotspots, such as Gladstone in south-east Queensland, a stronger response in aerosol mass and number concentration to DMS might be detected. However, the results presented in section 3.3 highlight the complexity and non-linearity of the aerosol system, where a change in aerosol sources does not necessarily result in a change in aerosol composition or concentration of the same direction or magnitude.

Addition of coral-reef-derived DMS to a simulated cleaner regional atmosphere did increase DMS_a, SO₂ and H₂SO₄. However, no clear influence was detected for sulfate aerosol mass or particle number concentration, suggesting that even with a reduction in anthropogenic sulfur emissions, coral-reef-derived DMS may not play a significant role in aerosol production and processing over the GBR. Again, negligible

($\leq 3.6\%$) changes occurred that were not restricted to the GBR region. These changes are more likely due to model and meteorological variability than coral-reef-derived DMS and so are not discussed further here.

While this analysis provides further insight into the influence of coral-reef-derived DMS on the contemporary and relatively clean regional atmosphere, further research is needed to improve DMS sources in the model. Simulations using both K99 and GBR DMS overestimated observed DMS_a during the 2016 R2R campaign, yet modelled concentrations were still within the range previously reported for the GBR (Swan and Jones, 2017; Jones et al., 2018; Swan and Jones, 2020). As discussed above, the R2R surveys were taken approximately two weeks prior, and six months after the 2016 mass coral bleaching event, which led to the loss of up to 40% of hard coral cover in the GBR (Hughes et al., 2018). The loss of DMS-producing corals may be one reason why observed DMS_w and DMS_a during the RVI and Mission Beach surveys were low compared to previous surveys and modelled concentrations, highlighting the complexity in predicting coral-reef-derived DMS.

Further research is also required to understand the aerosol chemistry system and how it responds to changes in anthropogenic and natural emissions sources. While DMS emissions from the GBR did not influence aerosol mass or number concentration at the scales modelled in this study, observational studies suggest that DMS may be more important in local or sub-daily feedbacks. Nucleation events forming new particles that primarily consist of sulfates have been observed over the GBR (Modini et al., 2009), and coincided with elevated DMS_a concentrations on one occasion in the southern GBR (Swan et al., 2016). Further, DMS emissions and DMS_a concentration are positively correlated with atmospheric particle number concentration ($>0.5 \mu\text{m}$ wet diameter) at Heron Island, likely reflecting condensational growth of smaller aerosols to the detectable size range (Jackson et al., 2020; Deschaseaux et al., 2022). Given that DMS emissions from the GBR are comparable to other highly productive regions such as the Southern Ocean or North Atlantic, where DMS-derived sulfates account for a significant portion of CCN (Korhonen et al., 2008; Sanchez et al., 2018), it is possible that DMS influences aerosol processes in the GBR at scales that are not captured by the coarse resolution global model.

5 Conclusions

This modelling study supports previous findings that the GBR is an important regional source of atmospheric sulfur. Inclusion of coral-reef-derived DMS increased the concentration of atmospheric DMS by 150% and sulfate aerosol precursor compounds by up to 18% over the GBR, with potential implications for local or sub-daily aerosol processes. However, no influence on modelled sulfate aerosol mass or particle

number concentration was detected in this study. While reducing anthropogenic SO₂ emissions did not strengthen the aerosol response to coral-reef-derived DMS, other anthropogenic and natural (e.g. dust) emissions from north-eastern Australia may confound the biogenic sulfur signal, particularly over large temporal and spatial scales. A stronger response to coral-reef-derived DMS may occur at local, sub-daily time-scales, that are perhaps not captured by global or regional climate models, or in the absence of anthropogenic emissions. However, further research is needed to disentangle the relative influence of natural and anthropogenic aerosol sources. Given the predicted rates of coral reef degradation and the global initiatives to shift towards renewable energy, it is important to understand how the complex aerosol-cloud system will respond to changes in natural and anthropogenic emissions.

Data availability statement

The ACCESS-AM2 atmospheric model used for this study is a licensed product of the UK Met Office and is available to specific users under a license agreement. The model output can be provided on request. The Heron Island DMS_a field data (Swan and Jones, 2017) are available from the Southern Cross University online repository (<https://researchportal.scu.edu.au/>). The RVI underway data (in2016_v05uwy5min_csv.zip) is available from the CSIRO Marine National Facility repository (<https://marlin.csiro.au/>) under a Creative Commons Attribution 4.0 License. The Heron Island wind speed data is available from H. B. Swan and E. S. M. Deschaseaux upon request. The RVI DMS_a data, and Mission Beach DMS_a and wind speed data are available from the relevant institutions upon request.

Author contributions

RJ completed the model simulations, data analysis and prepared the manuscript. MW provided guidance on the model setup, experimental design and analysis. HS, ED and HT provided field data. All authors contributed to the review and finalisation of the manuscript.

Funding

Research undertaken on Marine National Facility RV *Investigator* voyage IN2016_V05 (RVI) and the work of G. B. Jones, ED and HS at Heron Island was supported by Australian Research Council Discovery Project grant DP150101649 ‘The Great Barrier Reef as a significant source of climatically relevant aerosol particles’. RJ was supported by an Australian Government Research Training Program Scholarship,

Commonwealth Scientific and Industrial Research Organisation post-graduate scholarship, and the Griffith University School of Environment and Science.

Acknowledgments

We acknowledge the use of the CSIRO Marine National Facility in undertaking this research. The authors gratefully acknowledge Professor Zoran Ristovski, Chief Scientist on the voyage IN2016_V05 ‘The Great Barrier Reef as a significant source of climatically relevant aerosol particles’, the crew of the Marine National Facility RV *Investigator*, and all Mission Beach onsite personnel from CSIRO, Southern Cross University and Queensland University of Technology for the provision of DMS_a and wind speed measurements during the September-October 2016 field campaign. We also acknowledge the Australian Research Council for funding under the Discovery Project DP150101649. HS, ED and G. B. Jones are gratefully acknowledged for making available their Heron Island DMS_a and wind speed datasets. ED and HS would also like to thank the staff of the Heron Island Research Station for support during their field work. The modelling was undertaken using the National Computational Infrastructure facilities, supported by the Australian Government. The work forms part of RJ’s PhD thesis, entitled ‘Coral reefs as a source of dimethylsulfide (DMS) and the influence on the atmosphere of the Great Barrier Reef, which will be available on the Griffith University online repository (<https://research-repository.griffith.edu.au>) from 14 June 2023.

Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Publisher’s note

All claims expressed in this article are solely those of the authors and do not necessarily represent those of their affiliated organizations, or those of the publisher, the editors and the reviewers. Any product that may be evaluated in this article, or claim that may be made by its manufacturer, is not guaranteed or endorsed by the publisher.

Supplementary material

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fmars.2022.910423/full#supplementary-material>

References

- Andreae, M. O. (1995). "Climatic effects of changing atmospheric aerosol levels," in *In world survey of climatology* (Netherlands: Elsevier), vol. 16, 347–398. doi: 10.1016/S0168-6321(06)80033-7
- Andreae, M. O., Barnard, W. R., and Ammons, J. M. (1983). The biological production of dimethylsulfide in the ocean and its role in the global atmospheric sulfur budget. *Ecol. Bulletins*, 35, 167–177.
- Andreae, M. O., and Crutzen, P. J. (1997). Atmospheric aerosols: Biogeochemical sources and role in atmospheric chemistry. *Science* 276 (5315), 1052–1058. doi: 10.1126/science.276.5315.1052
- Andreae, M. O., and Rosenfeld, D. (2008). Aerosol–cloud–precipitation interactions. part 1. the nature and sources of cloud-active aerosols. *Earth-Science. Rev.* 89 (1–2), 13–41. doi: 10.1016/j.earscirev.2008.03.001
- Ayers, G. P., and Gillett, R. W. (2000). DMS and its oxidation products in the remote marine atmosphere: Implications for climate and atmospheric chemistry. *J. Sea. Res.* 43 (3–4), 275–286. doi: 10.1016/S1385-1101(00)00022-8
- Barnes, I., Hjorth, J., and Mihalopoulos, N. (2006). Dimethyl sulfide and dimethyl sulfoxide and their oxidation in the atmosphere. *Chem. Rev.* 106 (3), 940–975. doi: 10.1021/cr020529+
- Berndt, T., Scholz, W., Mentler, B., Fischer, L., Hoffmann, E. H., Tilgner, A., et al. (2019). Fast peroxy radical isomerization and OH recycling in the reaction of OH radicals with dimethyl sulfide. *J. Phys. Chem. Lett.* 10 (21), 6478–6483. doi: 10.1021/acs.jpcclett.9b02567
- Bi, D., Dix, M., Marsland, S., O'Farrell, S., Sullivan, A., Bodman, R., et al. (2020). Configuration and spin-up of ACCESS-CM2, the new generation Australian Community Climate and Earth System Simulator Coupled Model. *J. South. Hemisphere. Earth Syst. Sci.* 70 (1), 225–251. doi: 10.1071/ES19040
- Bodman, R. W., Karoly, D. J., Dix, M. R., Harman, I. N., Srinovsky, J., Dobrohotoff, P. B., et al. (2020). Evaluation of CMIP6 AMIP climate simulations with the ACCESS-AM2 model. *J. South. Hemisphere. Earth Syst. Sci.* 70 (1), 166–179. doi: 10.1071/ES19033
- Broadbent, A. D., and Jones, G. B. (2004). DMS and DMSP in mucus ropes, coral mucus, surface films and sediment pore waters from coral reefs in the Great Barrier Reef. *Mar. Freshw. Res.* 55 (8), 849–855. doi: 10.1071/MF04114
- Buckee, J., Pattiaratchi, C., and Verduin, J. (2020). Partial mortality of intertidal corals due to seasonal daytime low water levels at the Houtman Abrolhos Islands. *Coral. Reefs*. 39 (3), 537–543. doi: 10.1007/s00338-019-01887-5
- Bullock, H. A., Luo, H., and Whitman, W. B. (2017). Evolution of dimethylsulfoniopropionate metabolism in marine phytoplankton and bacteria. *Front. Microbiol.* 8. doi: 10.3389/fmicb.2017.00637
- Carslaw, K. S., Lee, L. A., Reddington, C. L., Pringle, K. J., Rap, A., Forster, P. M., et al. (2013). Large Contribution of natural aerosols to uncertainty in indirect forcing. *Nature* 503 (7474), 67–71. doi: 10.1038/nature12674
- Charlson, R. J., Lovelock, J. E., Andreae, M. O., and Warren, S. G. (1987). Oceanic phytoplankton, atmospheric sulphur, cloud albedo and climate. *Nature* 326 (6114), 655–661. doi: 10.1038/326655a0
- Chen, Z., Schofield, R., Rayner, P., Zhang, T., Liu, C., Vincent, C., et al. (2019). Characterization of aerosols over the Great Barrier Reef: The influence of transported continental sources. *Sci. Total. Environ.* 690, 426–437. doi: 10.1016/j.scitotenv.2019.07.007
- Cropp, R. A., Gabric, A. J., van Tran, D., Jones, G., Swan, H., and Butler, H. (2018). Coral reef aerosol emissions in response to irradiance stress in the Great Barrier Reef, Australia. *Ambio* 47 (6), 671–681. doi: 10.1007/s13280-018-1018-y
- Curson, A. R., Liu, J., Martínez, A. B., Green, R. T., Chan, Y., Carrión, O., et al. (2017). Dimethylsulfoniopropionate biosynthesis in marine bacteria and identification of the key gene in this process. *Nat. Microbiol.* 2, 17001–17009. doi: 10.1038/nmicrobiol.2017.9
- Dave, P., Patil, N., Bhushan, M., and Venkataraman, C. (2019). "Aerosol influences on cloud modification and rainfall suppression in the south Asian monsoon region," in *Climate change signals and response*. Eds. C. Venkataraman, T. Mishra, S. Ghosh and S. Karmakar (Springer), 21–37. doi: 10.1007/978-981-13-0280-0_2
- Deschaseaux, E. S. M., Jones, G. B., Deseo, M. A., Shepherd, K. M., Kiene, R. P., Swan, H. B., et al. (2014). Effects of environmental factors on dimethylated sulfur compounds and their potential role in the antioxidant system of the coral holobiont. *Limnol. Oceanography*. 59 (3), 758–768. doi: 10.4319/lo.2014.59.3.0758
- Deschaseaux, E. S. M., Swan, H. B., Maher, D., Jones, G. B., Schulz, K., Koveke, E., et al. (2022). The interplay between dimethyl sulfide (DMS) and methane (CH₄) in a coral reef ecosystem. *Front. Mar. Science.: Coral. Reef. Res.* doi: 10.3389/fmars.2022.910441
- Fan, J., Rosenfeld, D., Zhang, Y., Giangrande, S. E., Li, Z., Machado, L. A. T., et al. (2018). Substantial convection and precipitation enhancements by ultrafine aerosol particles. *Science* 359 (6374), 411–418. doi: 10.1126/science.aan8461
- Fiddes, S. L., Woodhouse, M. T., Lane, T. P., and Schofield, R. (2021). Coral-reef-derived dimethyl sulfide and the climatic impact of the loss of coral reefs. *Atmospheric. Chem. Phys.* 21 (8), 5883–5903. doi: 10.5194/acp-21-5883-2021
- Fiddes, S. L., Woodhouse, M. T., Nicholls, Z., Lane, T. P., and Schofield, R. (2018). Cloud, precipitation and radiation responses to large perturbations in global dimethyl sulfide. *Atmospheric. Chem. Phys.* 18 (14), 10177–10198. doi: 10.5194/acp-18-10177-2018
- Fiddes, S. L., Woodhouse, M. T., Utembe, S., Schofield, R., Alexander, S. P., Alroe, J., et al. (2022). The contribution of coral-reef-derived dimethyl sulfide to aerosol burden over the Great Barrier Reef: A modelling study. *Atmospheric. Chem. Phys.* 22 (4), 2419–2445. doi: 10.5194/acp-22-2419-2022
- Fischer, E., and Jones, G. (2012). Atmospheric dimethylsulfide production from corals in the Great Barrier Reef and links to solar radiation, climate and coral bleaching. *Biogeochemistry* 110 (1), 31–46. doi: 10.1007/s10533-012-9719-y
- Gabric, A. J., Matrai, P. A., Kiene, R. P., Cropp, R. A., Dacey, J. W. H., DiTullio, G. R., et al. (2008). Factors determining the vertical profile of dimethylsulfide in the Sargasso Sea during summer. *Deep. Sea. Res. Part II: Topical Stud. Oceanography*. 55 (10–13), 1505–1518. doi: 10.1016/j.dsr2.2008.02.002
- Gabric, A. J., Qu, B., Rotstajn, L., and Shepard, J. M. (2013). Global simulations of the impact on contemporary climate of a perturbation to the sea-to-air flux of dimethylsulfide. *Aust. Meteorol. Oceanographic. J.* 63 (3), 365–376. doi: 10.22499/2.6303.002
- Gali, M., Simó, R., Vila-Costa, M., Ruiz-González, C., Gasol, J. M., and Matrai, P. (2013). Diel patterns of oceanic dimethylsulfide (DMS) cycling: Microbial and physical drivers. *Global Biogeochem. Cycles*. 27 (3), 620–636. doi: 10.1002/gbc.20047
- Gardner, S. G., Nielsen, D. A., Laczka, O., Shimmon, R., Beltran, V. H., Ralph, P. J., et al. (2016). Dimethylsulfoniopropionate, superoxide dismutase and glutathione as stress response indicators in three corals under short-term hyposalinity stress. *Proc. R. Soc. B.: Biol. Sci.* 283 (1824), 20152418. doi: 10.1098/rspb.2015.2418
- Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., et al. (2020). The ERA5 global reanalysis. *Q. J. R. Meteorol. Soc.* 146 (730), 1999–2049. doi: 10.1002/qj.3803
- Hodshire, A. L., Campuzano-Jost, P., Kodros, J. K., Croft, B., Nault, B. A., Schroder, J. C., et al. (2019). The potential role of dimethylsulfonic acid (MSA) in aerosol formation and growth and the associated radiative forcings. *Atmospheric. Chem. Phys.* 19 (5), 3137–3160. doi: 10.5194/acp-19-3137-2019
- Hoffmann, E. H., Tilgner, A., Schroedner, R., Bräuer, P., Wolke, R., and Herrmann, H. (2016). An advanced modeling study on the impacts and atmospheric implications of multiphase dimethyl sulfide chemistry. *Proc. Natl. Acad. Sci.* 113 (42), 11776–11781. doi: 10.1073/pnas.1606320113
- Hopkins, F. E., Bell, T. G., Yang, M., Suggett, D. J., and Steinke, M. (2016). Air exposure of coral is a significant source of dimethylsulfide (DMS) to the atmosphere. *Sci. Rep.* 6 (1), 1–11. doi: 10.1038/srep36031
- Hughes, T. P., Kerry, J. T., Baird, A. H., Connolly, S. R., Dietzel, A., Eakin, C. M., et al. (2018). Global warming transforms coral reef assemblages. *Nature* 556 (7702), 492–496. doi: 10.1038/s41586-018-0041-2
- IPCC (2013). *Climate change 2013: The physical science basis. contribution of working group I to the fifth assessment report of the intergovernmental panel on climate change*. Eds. T. Stocker, G. Qin, G. Plattner, M. Tignor, S. Allen, J. Boschung, et al (United Kingdom and New York, NY, USA: Cambridge University Press), pp. 1535.
- IPCC (2021). "Summary for policymakers. in: Climate change 2021: The physical science basis," in *Contribution of working group I to the sixth assessment report of the intergovernmental panel on climate change*. Eds. V. Masson-Delmotte, P. Zhai, A. Pirani, S. L. Connors, C. Pean, S. Berger, N. Caud, Y. Chen, L. Goldfarb, M. I. Gomis, M. Huang, K. Leitzell, E. Lonnoy, J. B. R. Matthews, T. K. Maycock, T. Waterfield, O. Yelekci, R. Yu and B. Zhou (United Kingdom and New York, NY, USA: Cambridge University Press), pp. 3–32.
- IPCC (2014). *Climate change 2014: Synthesis report. Contribution of working groups I, II and III to the fifth assessment report of the intergovernmental panel on climate change*. [Core Writing Team, R. K. Pachauri and L. A. Meyer (eds.)]. IPCC, Geneva, Switzerland, pp. 151.
- Jackson, R. L., Gabric, A. J., Matrai, P. A., Woodhouse, M. T., Cropp, R. A., Jones, G. B., et al. (2021). Parameterizing the impact of seawater temperature and irradiance on dimethylsulfide (DMS) in the Great Barrier Reef and the contribution of coral reefs to the global sulfur cycle. *J. Geophysical. Research.: Oceans*. 126 (3), e2020JC016783. doi: 10.1029/2020JC016783

- Jackson, R. L., Gabric, A. J., Woodhouse, M. T., Swan, H. B., Jones, G. B., Cropp, R. A., et al. (2020). Coral reef emissions of atmospheric dimethylsulfide and the influence on marine aerosols in the southern Great Barrier Reef, Australia. *J. Geophysical. Research.: Atmospheres*. 125 (7), e2019JD031837. doi: 10.1029/2019JD031837
- Jones, G. B. (2015). "The reef sulphur cycle: Influence on climate and ecosystem services," in *In ethmobiology of corals and coral reefs* (Switzerland: Springer), 27–57. doi: 10.1007/978-3-319-23763-3_3
- Jones, G. B., Curran, M., Broadbent, A., King, S., Fischer, E., and Jones, R. (2007). Factors affecting the cycling of dimethylsulfide and dimethylsulfoloniopropionate in coral reef waters of the Great Barrier Reef. *Environ. Chem.* 4 (5), 310–322. doi: 10.1071/EN06065
- Jones, G. B., Curran, M., Deschaseaux, E. S. M., Omori, Y., Tanimoto, H., Swan, H. B., et al. (2018). The flux and emission of dimethylsulfide from the Great Barrier Reef region and potential influence on the climate of NE Australia. *J. Geophysical. Research.: Atmospheres*. 123 (24), 13835–13856. doi: 10.1029/2018JD029210
- Jones, G. B., Curran, M., Swan, H. B., and Deschaseaux, E. S. M. (2017). Dimethylsulfide and coral bleaching: Links to solar radiation, low level cloud and the regulation of seawater temperatures and climate in the Great Barrier Reef. *Am. J. Climate Change* 6 (02), 328. doi: 10.4236/ajcc.2017.62017
- Kettle, A. J., Andreae, M. O., Amouroux, D., Andreae, T. W., Bates, T. S., Berresheim, H., et al. (1999). A global database of sea surface dimethylsulfide (DMS) measurements and a procedure to predict sea surface DMS as a function of latitude, longitude, and month. *Global Biogeochem. Cycles*. 13 (2), 399–444. doi: 10.1029/1999GB900004
- Korhonen, H., Carslaw, K. S., Spracklen, D. V., Mann, G. W., and Woodhouse, M. T. (2008). Influence of oceanic dimethyl sulfide emissions on cloud condensation nuclei concentrations and seasonality over the remote southern hemisphere oceans: A global model study. *J. Geophysical. Research.: Atmospheres*. 113 (D15). doi: 10.1029/2007JD009718
- Lana, A., Bell, T. G., Simó, R., Vallina, S. M., Ballabrera-Poy, J., Kettle, A. J., et al. (2011). An updated climatology of surface dimethylsulfide concentrations and emission fluxes in the global ocean. *Global Biogeochem. Cycles*. 25 (1), GB1004. doi: 10.1029/2010GB003850
- Lana, A., Simó, R., Vallina, S. M., and Dachs, J. (2012). Potential for a biogenic influence on cloud microphysics over the ocean: A correlation study with satellite-derived data. *Atmospheric. Chem. Phys.* 12 (17), 7977–7993. doi: 10.5194/acp-12-7977-2012
- Liss, P. S., and Merlivat, L. (1986). Air-sea gas exchange rates: Introduction and synthesis. in *Role. air-sea. exchange. geochem. cycling* (Dordrecht: Springer) 185, 113–127. doi: 10.1007/978-94-009-4738-2_5
- Mahajan, A. S., Fadnavis, S., Thomas, M. A., Pozzoli, L., Gupta, S., Royer, S., et al. (2015). Quantifying the impacts of an updated global dimethyl sulfide climatology on cloud microphysics and aerosol radiative forcing. *J. Geophysical. Research.: Atmospheres*. 120 (6), 2524–2536. doi: 10.1002/2014JD022687
- Mann, G. W., Carslaw, K. S., Ridley, D. A., Spracklen, D. V., Pringle, K. J., Merikanto, J., et al. (2012). Intercomparison of modal and sectional aerosol microphysics representations within the same 3-d global chemical transport model. *Atmospheric. Chem. Phys.* 12 (10), 4449–4476. doi: 10.5194/acp-12-4449-2012
- Mann, G. W., Carslaw, K. S., Spracklen, D. V., Ridley, D. A., Manktelow, P. T., Chipperfield, M. P., et al. (2010). Description and evaluation of GLOMAP-mode: A modal global aerosol microphysics model for the UKCA composition-climate model. *Geoscientific. Model. Dev.* 3 (2), 519–551. doi: 10.5194/gmd-3-519-2010
- McCormick, R. A., and Ludwig, J. H. (1967). Climate modification by atmospheric aerosols. *Science* 156 (3780), 1358–1359. doi: 10.1126/science.156.3780.1358
- McGowan, H., Sturman, A., Saunders, M., Theobald, A., and Wiebe, A. (2019). Insights from a decade of research on coral reef-atmosphere energetics. *J. Geophysical. Research.: Atmospheres*. 124 (8), 4269–4282. doi: 10.1029/2018JD029830
- McParland, E. L., and Levine, N. M. (2019). The role of differential DMSP production and community composition in predicting variability of global surface DMSP concentrations. *Limnol. Oceanography*. 64 (2), 757–773. doi: 10.1002/lno.11076
- Modini, R. L., Ristovski, Z. D., Johnson, G. R., He, C., Surawski, N., Morawska, L., et al. (2009). New particle formation and growth at a remote, sub-tropical coastal location. *Atmospheric. Chem. Phys.* 9 (19), 7607–7621. doi: 10.5194/acp-9-7607-2009
- Raina, J. B., Tapiolas, D. M., Forêt, S., Lutz, A., Abrego, D., Ceh, J., et al. (2013). DMSP biosynthesis by an animal and its role in coral thermal stress response. *Nature* 502 (7473), 677–680. doi: 10.1038/nature12677
- Ramanathan, V., Cess, R. D., Harrison, E. F., Minnis, P., Barkstrom, B. R., Ahmad, E., et al. (1989). Cloud-radiative forcing and climate: Results from the earth radiation budget experiment. *Science* 243 (4887), 57–63. doi: 10.1126/science.243.4887.57
- Rosenfeld, D., Dai, J., Yu, X., Yao, Z., Xu, X., Yang, X., et al. (2007). Inverse relations between amounts of air pollution and orographic precipitation. *Science* 315 (5817), 1396–1398. doi: 10.1126/10.1126/science.1137949
- Sanchez, K. J., Chen, C. L., Russell, L. M., Betha, R., Liu, J., Price, D. J., et al. (2018). Substantial seasonal contribution of observed biogenic sulfate particles to cloud condensation nuclei. *Sci. Rep.* 8 (1), 1–14. doi: 10.1038/s41598-018-21590-9
- Stefels, J. (2000). Physiological aspects of the production and conversion of DMSP in marine algae and higher plants. *J. Sea. Res.* 43 (3–4), 183–197. doi: 10.1016/S1385-1101(00)00030-7
- Sunda, W., Kieber, D. J., Kiene, R. P., and Huntsman, S. (2002). An antioxidant function for DMSP and DMS in marine algae. *Nature* 418 (6895), 317–320. doi: 10.1038/nature00851
- Swan, H. B., Crough, R. W., Vaattovaara, P., Jones, G. B., Deschaseaux, E. S. M., Eyre, B. D., et al. (2016). Dimethyl sulfide and other biogenic volatile organic compound emissions from branching coral and reef seawater: Potential sources of secondary aerosol over the Great Barrier Reef. *J. Atmospheric. Chem.* 73 (3), 303–328. doi: 10.1007/s10874-016-9327-7
- Swan, H. B., and Jones, G. B. (2017). *Atmospheric dimethylsulfide surface concentrations and supermicron particle number concentrations at heron island (23.44° s, 151.91° e) February 2016* (Southern Cross University). doi: 10.4226/47/59c460fb8322
- Swan, H. B., and Jones, G. B. (2020). *Database of surface level atmospheric dimethylsulfide (DMS) collected for the project "The Great Barrier Reef as a significant source of climatically relevant aerosol particles."* (Southern Cross University). doi: 10.25918/5c772187f39b6
- Swan, H. B., Jones, G. B., Deschaseaux, E. S. M., and Eyre, B. D. (2017). Coral reef origins of atmospheric dimethylsulfide at Heron Island, Southern Great Barrier Reef, Australia. *Biogeosciences* 14 (1), 229–239. doi: 10.5194/bg-14-229-2017
- Thomas, M. A., Suntharalingam, P., Pozzoli, L., Rast, S., Devasthale, A., Kloster, S., et al. (2010). Quantification of DMS aerosol-cloud-climate interactions using the ECHAM5-HAMMOZ model in a current climate scenario. *Atmospheric. Chem. Phys.* 10 (15), 7425–7438. doi: 10.5194/acp-10-7425-2010
- Trounce, H., Ristovski, Z., Miljevic, B., Cravigan, L., Alroe, J., Osuagwa, C., et al. (in preparation) Great Barrier Reef aerosol and cloud data from the reef to rainforest campaign. *Earth System. Sci. Data Discussions*.
- Twomey, S. (1974). Pollution and the planetary albedo. *Atmospheric. Environ.* (1967). 8 (12), 1251–1256. doi: 10.1016/0004-6981(74)90004-3
- Twomey, S., Piepgrass, M., and Wolfe, T. L. (1984). An assessment of the impact of pollution on global cloud albedo. *Tellus. B.* 36 (5), 356–366. doi: 10.1111/j.1600-0889.1984.tb00254.x
- Veres, P. R., Neuman, J. A., Bertram, T. H., Assaf, E., Wolfe, G. M., Williamson, C. J., et al. (2020). Global airborne sampling reveals a previously unobserved dimethyl sulfide oxidation mechanism in the marine atmosphere. *Proc. Natl. Acad. Sci.* 117 (9), 4505–4510. doi: 10.1073/pnas.1919344117
- Von Glasow, R., and Crutzen, P. J. (2004). Model study of multiphase DMS oxidation with a focus on halogens. *Atmospheric. Chem. Phys.* 4 (3), 589–608. doi: 10.5194/acp-4-589-2004
- Walters, D., Baran, A. J., Boutle, I., Brooks, M., Earnshaw, P., Edwards, J., et al. (2019). The met office unified model global atmosphere 7.0/7.1 and JULES global land 7.0 configurations. *Geoscientific. Model. Dev.* 12 (5), 1909–1963. doi: 10.5194/gmd-12-1909-2019
- Woodhouse, M. T., Carslaw, K. S., Mann, G. W., Vallina, S. M., Vogt, M., Halloran, P. R., et al. (2010). Low sensitivity of cloud condensation nuclei to changes in the sea-air flux of dimethyl-sulphide. *Atmospheric. Chem. Phys.* 10 (16), 7545–7559. doi: 10.5194/acp-10-7545-2010
- Woodhouse, M. T., Mann, G. W., Carslaw, K. S., and Boucher, O. (2013). Sensitivity of cloud condensation nuclei to regional changes in dimethyl-sulphide emissions. *Atmospheric. Chem. Phys.* 13 (5), 2723–2733. doi: 10.5194/acp-13-2723-2013
- Yang, M., Blomquist, B., Fairall, C., Archer, S., and Huebert, B. (2011). Air-sea exchange of dimethylsulfide in the Southern Ocean: Measurements from SO GasEx compared to temperate and tropical regions. *J. Geophysical. Research.: Oceans*. 116, C00F05. doi: 10.1029/2010JC006526
- Zavarsky, A., Booge, D., Fiehn, A., Krüger, K., Atlas, E., and Marandino, C. (2018). The influence of air-sea fluxes on atmospheric aerosols during the summer monsoon over the tropical Indian ocean. *Geophysical. Res. Lett.* 45 (1), 418–426. doi: 10.1002/2017GL076410