Coral Reef Emissions of Atmospheric Dimethylsulfide and the Influence on Marine Aerosols in the Southern Great Barrier Reef, Australia

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Abstract Variability in atmospheric dimethylsulfide (DMSa) and the potential influence on atmospheric aerosols was investigated at Heron Island in the southern Great Barrier Reef (GBR), Australia. This work compiles previously published DMSa data (reported in Swan, Jones, Deschaseaux, & Eyre, 2017, https://doi.org/10.5194/bg-14-229-2017), with additional surveys of DMSa, atmospheric particle number concentration, and other oceanic and atmospheric data sets. DMSa was higher in summer (mean 3.2 nmol m−3/78 ppt) than winter (mean 1.3 nmol m−3/32 ppt), reflective of seasonal shifts in phytoplankton biomass and emissions from corals in the southern GBR. Seasonally extreme spikes in DMSa were detected during low tide and low wind speed, supporting findings that the coral reef can be an important source of DMSa above background oceanic emissions. A significant link was present between DMSa and aerosol concentration (ranging from 0.5 to 2.5 μm) during calm, daylight hours, when conditions were optimal for the local oxidation of DMSa to sulfate aerosol precursors. This link may reflect condensational growth of existing fine particles (< 0.5 μm), which is the dominant pathway by which biogenic trace gases influence aerosols in the marine boundary layer. Aerosol concentration significantly correlated with reduced surface solar irradiance and sea surface temperature, which is potential evidence of a local negative feedback mitigating coral physiological stress. These findings provide a step toward a better understanding of the processes influencing DMSa and aerosol concentrations and of the consequences for the local radiative balance over coral reefs; an increasingly important topic with ongoing ocean warming and coral bleaching.

1. Introduction

In the clean marine boundary layer (MBL) biogenic sulfates contribute to marine biogenic aerosol (MBA) formation and growth (Gabric et al., 2013; Korhonen et al., 2008; Modini et al., 2009; Sanchez et al., 2018). This MBA has important implications for the formation of cloud condensation nuclei (CCN) and the microphysical properties of low-level clouds (LLC) (Andreae & Rosenfeld, 2008). Marine emissions of the volatile gas dimethylsulfide (DMS) constitute 50% of natural global sulfur emissions (Barnes et al., 2006; Bates et al., 1992; Simó, 2001) and may therefore play an important role in the climate of the remote MBL (Broadbent & Jones, 2004; Burdett et al., 2015; Fiddes et al., 2018; Jones et al., 1994; Raina et al., 2013; Swan, Jones, Deschaseaux, & Eyre, 2017).

Sources of DMS in the ocean include marine macroalgae and free-living microalgae (Stefels, 2000; Van Alstyne & Puglisi, 2007), zooxanthellate Scleractinian (Raina et al., 2013) and Alcyonacea corals (Haydon et al., 2018), giant clams (Hill et al., 2000), and carbonate sediments (Deschaseaux et al., 2019). Reported concentrations of dimethylated sulfur compounds are typically higher in zooxanthellate corals than in other marine producers (Broadbent et al., 2002; Swan, Deschaseaux, et al., 2017). Corals and their endosymbiotic zooxanthellae upregulate the biosynthesis of dimethylsulfonpropionate (DMSp) during physiological stress caused by elevated sea surface temperature (SST) and irradiance, hyposalinity with rainfall or riverine discharge, and aerial exposure at low tide (Deschaseaux, Jones, et al., 2014; Hopkins et al., 2016; Jones et al., 2014; Raina et al., 2013). Zooxanthellate corals harbor several genetically distinct types of dinoflagellates, distinguished as zooxanthellae Clades (A–H), and DMSp biosynthesis varies between coral species and
endosymbiont composition (Deschaseaux, Beltran, et al., 2014; Deschaseaux, Jones, et al., 2014; Jones & King, 2015; Steinke et al., 2011). Acropora spp. are the dominant reef-building coral genus in the coral reef-dense region of the Indo-Pacific and contain among the highest reported concentrations of DMSP (Raina et al., 2013; Swan, Deschaseaux, et al., 2017). These species typically harbor temperature-sensitive endosymbionts (e.g., Clade C), which have been found to contain higher and more variable quantities of DMSP when exposed to thermal stress, compared to thermo-tolerant clades (e.g., Clade D) (Deschaseaux, Beltran, et al., 2014; Deschaseaux, Jones, et al., 2014; Jones & King, 2015).

DMSP is the major marine precursor of DMS and has a range of biological functions. In marine algae, DMSP and its derivatives DMS, acrylate, dimethyl sulfoxide (DMSO), and methane sulfonic acid (MSA) are involved in raising the oxidative stress threshold (Sunda et al., 2002). A similar antioxidant mechanism exists in corals (Deschaseaux, Beltran, et al., 2014; Deschaseaux, Jones, et al., 2014; Gardner et al., 2016). When photosynthetically active radiation (PAR) exceeds the absorption capacity of zooxanthellae photosystems, damage can occur to the photosynthetic apparatus (Jones et al., 2002; Lesser, 2010). Zooxanthellae may recover from photo damage at night, but if the rate of damage exceeds that of repair by photoprotective mechanisms, oxidative stress accumulates and reactive oxygen species (ROS) are leaked into host tissues (Jones et al., 2002; Lesser, 2006, 2010; Lesser et al., 1990). Higher than normal SST can exacerbate light stress by lowering the PAR absorption capacity, leading to more oxidative stress in the coral holobiont and coral bleaching if conditions persist (Lesser, 2010; Skirving et al., 2018; Yakovleva et al., 2009).

DMSP is cleaved to DMS and acrylate by enzymes collectively termed DMSP-lyases, present within coral zooxanthellae, marine microalgae, and coral-associated and free-living bacteria (Alcolombri et al., 2015; Bullock et al., 2017; Raina et al., 2009; Sun et al., 2016). DMS/P and acrylate rapidly scavenge these ROS forming DMSO, which may be further oxidized by ROS to MSA (Sunda et al., 2002). This results in a nonlinearity in coral DMS emissions with rising stress. For example, DMSP biosynthesis and cleavage to DMS has been shown to increase in corals with rising SST and irradiance (Jones et al., 2007; Jones et al., 2014; Raina et al., 2013), until SST and/or PAR exceed coral physiological tolerance thresholds. Beyond these thresholds, the rate of DMS/P oxidation to DMSO increases, resulting in a decline in ambient DMS concentrations (Deschaseaux, Beltran, et al., 2014; Deschaseaux, Jones, et al., 2014; Fischer & Jones, 2012; Jones & King, 2015).

When present in excess, DMS will diffuse into the water column and be ventilated to the MBL (Fischer & Jones, 2012; Hopkins et al., 2016). Atmospheric DMS (DMSa) is then oxidized primarily by hydroxyl radicals (OH) in the clean MBL to sulfur dioxide (SO2), which may then undergo gas phase oxidation to sulfuric acid (H2SO4) and homogenous nucleation to form new non–sea salt sulfate (nss-SO4) particles (Andreae & Crutzen, 1997; Andreae & Rosenfeld, 2008; Charlson et al., 1987). The rate of DMS conversion to sulfate aerosol precursors is critical in the formation of nss-SO4. However, this pathway requires high solar irradiance and OH concentrations (≥3×105 cm−3) (Andreae & Crutzen, 1997), high DMSa mixing ratios (≥100 pptv) (Chang et al., 2011), and relatively low humidity (~60%) (Modini et al., 2009), wind speeds, and preexisting particle number concentrations (<100 cm−3) (Chang et al., 2011). These are conditions typically found in the free troposphere (FT), and thus, entrainment to the MBL is an important source of new sulfate particles over remote oceans (Sanchez et al., 2018). The dominant pathways in the MBL are the aqueous-phase oxidation of DMS-derived sulfates within cloud droplets and the condensational growth of preexisting particles (Hoffmann et al., 2016; Woodhouse et al., 2013). Although these pathways suppress new particle formation, DMS-derived sulfates alter the chemical composition, size, and the climate forcing potential of atmospheric particles (Andreae & Crutzen, 1997; Hoffmann et al., 2016).

At Heron Island in the southern Great Barrier Reef (GBR), new particle formation events have been observed to coincide with DMSa spikes above the coral reef (Swan et al., 2016). These events occur during daylight, low relative humidity, and low wind speeds, when conditions for the local gas phase nucleation of DMS oxidation products are favorable. Newly formed particles in the submicron size range consisted of ~50% sulfate and 50% organics (Swan et al., 2016), similar to earlier findings which characterized nucleation mode particles (~0.02 μm) in the southern GBR as ~60% sulfate and 40% organics (Modini et al., 2009). Reef-scale micrometeorology is an important driver of coral physiological stress and bleaching in the GBR (McGowan et al., 2019; McGowan & Theobald, 2017). Local LLC cover reduces downward solar radiation and SST—both of which are major contributors to coral physiological stress and coral bleaching.
Evaporation and particle emissions over shallow, warm coral reef waters contributes to the formation of a convective coral reef internal boundary layer (~65–130 m), with typically higher temperatures and humidity than the atmosphere above, favorable for LLC formation (McGowan et al., 2019). Emissions of DMS and other volatile organic compounds (VOCs) may therefore establish a negative local climatic feedback to abate coral stress by influencing the chemical and physical properties of marine aerosols, CCN and LLC (Jones et al., 2017; Kleypas et al., 2008; Swan et al., 2016; Takahashi et al., 2010).

Several studies have demonstrated a significant relationship between DMS, aerosol optical depth (AOD), CCN, and cloud droplet radius in the remote MBL (Chang et al., 2011; Fiddes et al., 2018; Gabric et al., 2013; Gabric et al., 2018; Korhonen et al., 2008; Lana et al., 2012; McCoy et al., 2015; Sanchez et al., 2018; Vallina et al., 2007). A recent modeling study found that H₂SO₄ aerosol, LLC, and rainfall were sensitive to large perturbations in DMS (Fiddes et al., 2018). This response was greatest in remote Southern Hemisphere eastern ocean basin stratiform cloud decks. Here, enhanced DMS sea-air flux and DMS-derived aerosols reduced surface short-wave radiation (−5 W m⁻²) and precipitation (−10%), due to enhanced LLC cover and lifetime (Fiddes et al., 2018). Similarly, a correlation analysis of global monthly sea surface and atmospheric DMS concentrations positively correlated with satellite-derived fine-mode AOD and CCN number in pristine ocean regions (Vallina et al., 2007). This suggests that a regionally variable link exists between DMS emissions and atmospheric properties. Coral reefs are strong sources of DMS and may also bioregulate their local environment. However, the importance of biogenic compounds such as DMS in particle nucleation and growth and their effect on the local radiative balance in coral reefs remains uncertain.

Here, we investigate seasonal and diurnal variability in DMSₐ and potential sources of DMSₐ and atmospheric particles at Heron Island in the southern GBR. DMSₐ was measured by H.B. Swan above the Heron Island coral reef flat for four approximately 2-week periods during the summers of 2012, 2016, and 2018 and one winter (2013). Additional measurements of atmospheric particle number concentration in two size ranges (0.5–2.5 and > 2.5 μm) were taken during the 2016 and 2018 summer surveys. Seasonal variability in the 2012 and 2013 DMSₐ data sets is reported in Swan, Jones, Deschaseaux, and Eyre (2017). This study builds on those findings through further analysis of additional DMSₐ data, in conjunction with previously unanalyzed measurements of atmospheric particle number concentration and satellite derived observations of AOD and chlorophyll-a (CHL).

2. Materials and Methods

Heron Island (23.44°S, 151.91°E) is an outer-shelf lagoonal coral reef, located in the Capricorn Bunker region of the southern GBR (Figure 1). Surface level DMSₐ (nmol m⁻³) was measured above Heron Island at approximately 15-min intervals during four separate surveys: 6–20 March 2012, 18 July to 5 August 2013 (Swan, Jones, & Deschaseaux, 2017a), 5–18 February 2016 (Swan, Jones, & Deschaseaux, 2017b), and 27 January to 7 February 2018 (Swan et al., 2018). These data were averaged into an hourly time series (for 2012 n = 330; for 2013 n = 430; for 2016 n = 307; for 2018 n = 265). Measurements were taken using an
automated gas chromatograph fitted with a pulsed flame photometric detector (GC-PFPD), with a sample intake located on top of the Heron Island Research Station (~15 m above sea level [ASL]), less than 100 m from the coral reef flat (Figure 1b). Samples had a 0.1 nmol m$^{-3}$ (2 ppt) detection limit. Further details of the configuration, operation, calibration, and measurement uncertainty of the sampling method is described in Swan et al. (2015). Minute-averaged atmospheric particle number concentration in intermediate (0.5–2.5 μm) and coarse (> 2.5 μm) diameter size ranges were also measured during the 2016 and 2018 surveys (Swan, Jones, & Deschaseaux, 2017b; Swan et al., 2018) using a dual channel laser optical particle counter (Dylos DC1700 Air Quality Monitor, Dylos Corporation, Riverside CA, USA). Given the typical moist marine air flows at Heron Island, the particle diameters provided above are for hydrated particles. Seasonal variability and environmental factors affecting DMS$_a$ concentration in the 2012 and 2013 data sets are examined in Swan, Jones, Deschaseaux, and Eyre (2017). The current analysis builds on those findings by examining new 2016 and 2018 data and comparing DMS$_a$ with intermediate (D0.5) and coarse-mode (D2.5) atmospheric particle concentration, and satellite observations of CHL and AOD.

### 2.1. In Situ Physical Parameters at Heron Island

A wireless sensor network consisting of sensor floats (SF) and relay poles (RP), is deployed at Heron Island (Figure 1b) as part of the Australian Integrated Marine Observing System (IMOS) (2019). High-frequency (30-min) measurements of SST are available at varying depths ranging from 0.3 to 15 m, representative of depths across the coral reef flat, lagoon and reef slope. PAR is available from an above water LICOR™ LI-192 quantum sensor located in the NE coral reef slope (RP 8, Figure 1b). These data were obtained from the Australian Institute of Marine Science (AIMS) (http://data.aims.gov.au). Details of the sensors used in this analysis are provided in Table 1.

A time series of hourly SST for Heron Island was compiled from a collection of sensors chosen for their good temporal coverage (2009–2016) and depth (0.3–2 m to account for the reef flat and lagoon) (AIMS, 2019a). A time series of hourly surface PAR (mol m$^{-2}$ hr$^{-1}$) was obtained from RP 8 (AIMS, 2019b). Data from this sensor were not available for the 2012 or 2016 surveys. Data for these two periods were obtained from an equivalent above-water sensor located at One Tree Island ~10 km SE of Heron Island (AIMS, 2019c). Given the close proximity, PAR obtained from the One Tree Island sensor is representative of PAR measured at Heron Island. Hourly mean wind speed (WS: m s$^{-1}$) and wind direction (WD: degrees) were obtained from measurements made by an automatic weather station (AWS, model XC0348) mounted < 1 m from the DMS$_a$ sampling intake on top of the Heron Island Research Station ~15 m ASL (Swan, Jones, & Deschaseaux, 2017a; Swan, Jones, & Deschaseaux, 2017b; Swan et al., 2018). These data were unavailable for 2 days during the 2018 surveys (15:00 on 3 February to 12:00 on 5 February 2018). WS and WD for these days was obtained from an automated weather station (Vaisala™ WXT520) mounted on RP 5 ~2 m ASL (Bainbridge et al., 2010) located ~7 km east of the Heron Island Research Station (Figure 1b) (AIMS, 2019d). WS recorded by this weather station was on average 4 m s$^{-1}$ higher than WS recorded from the Heron Island Research Station, likely due to the Research Station being partially sheltered from prevailing southeasterlies by surrounding trees. For consistency with the remaining survey periods, the weather station WS on 3–5 February 2018 was first scaled to WS at 15 m using the logarithmic wind speed profile described in section 2.3 (Hoffman, 2011), and then reduced by 4 m s$^{-1}$. The range of adjusted wind speeds

| Table 1 | Details of In Situ Sensors at Heron Island (Shown in Figure 1b), Used to Compile the Time Series of SST, PAR, WS, and WD |
|-----------------|-----------------|-----------------|-----------------|
| **Sensor** | **Variable** | **Depth (m)** | **Data availability** |
| SF 1 | SST | 0.3 | 23-12-2008 06-08-2019 |
| SF 2 | SST | 0.3 | 23-12-2008 19-07-2016 |
| RP 3 | SST | 2 | 01-12-2008 06-08-2019 |
| RP 6 | SST | 1.5 | 02-12-2008 06-08-2019 |
| RP 8 | PAR | above-water | 09-09-2012 06-08-2019 |
| Weather Station (RP 5) | WS, WD (2018 only) | above-water | 02-12-2008 06-08-2019 |

*Note. Dates are formatted as dd-mm-yyyy.*
in 2018 were then similar in magnitude to wind speeds measured during the other surveys (< 15 m s⁻¹). Hourly tide height (Tide: m) was calculated using daily high and low tide predictions provided by the Australian Bureau of Meteorology. The predicted tide height was shifted +1.25 hr to reflect on-site observations of low tide over the Heron Island reef flat, which consistently occurred 1.25 hr later than the predicted time (Swan, Jones, Deschaseaux, & Eyre, 2017).

2.2. Remotely Sensed Data

To disentangle local from remote sources of observed DMSₐ, back trajectories of air masses arriving at Heron Island within 24 hr (the approximate atmospheric residence time for DMSₐ) were computed for each study day (supporting information Figure S1) by the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory HYSPLIT transport and dispersion model (Rolph et al., 2017; Stein et al., 2015). These show the origin of air masses in space and altitude (m) ASL. A daily area-averaged time series of CHL (mg m⁻³), a proxy for phytoplankton biomass, was obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS) aboard the Terra and Aqua satellites. CHL was downloaded from the NASA Ocean Color Distributed Active Archive Center (DAAC) (http://oceancolor.gsfc.nasa.gov) and averaged over a broad region encompassing Heron Island and the adjacent Coral and Tasman Seas (21.65–30°S; 150–160°E). HYSPLIT back trajectories show that low-lying air masses originated within this subset boundary on all study days, and thus, CHL provides an indication of the potential for phytoplankton as a DMSₐ source at Heron Island. We also obtained a global (1°) monthly climatology of ocean mixed layer depth (MLD: m), calculated using the density algorithm from Argo Float temperature and salinity data for January 2000–April 2018 (Holte et al., 2017). An area-averaged climatology of MLD was calculated for the CHL subset region to assess drivers of seasonal variability in CHL.

A daily time series of AOD (869 nm) was also obtained for each survey period for comparison with in situ measurements of DMSₐ. AOD was downloaded from the NASA Ocean Color DAAC at 4 km resolution and area averaged over a ~400 x 400 km grid centered on Heron Island (21.65–25.27°S; 150–153.93°E). It was expected that during low WS (≤ 2 m s⁻¹), locally emitted aerosols would remain within this subset boundary over a 24-hr period and be captured by the analysis. DMSₐ has an atmospheric residence time of ~1 day (Khan et al., 2016) before removal mechanisms such as oxidation to SO₂ occur (Andreae & Crutzen, 1997; Barnes et al., 2006). However, Berndt et al. (2019) suggest that DMS oxidation can occur far more rapidly via an isomerization pathway, as does Swan et al. (2016) who suggests that DMS oxidation can occur in a matter of hours over warm, well-lit tropical coral reef waters (Swan et al., 2016), where hydroxyl concentrations are high (Andreae & Crutzen, 1997). If DMS-derived sulfates significantly influence AOD over the southern GBR, a positive correlation is expected between daily mean DMSₐ and AOD during calm conditions (≤ 2 m s⁻¹).

2.3. DMS Sea-Air Transfer Velocity

The gas transfer velocity of DMS (k_DMS; cm hr⁻¹) controls the rate of exchange across the sea-air interface and is related to WS and SST (Ho & Wanninkhof, 2016; Yang et al., 2011). Rising wind speeds increase sea surface and atmospheric turbulence and enhance interfacial gas transfer between the ocean and atmosphere. Increasing SST also influences gas transfer by reducing diffusive resistance. Therefore, k_DMS can be used as an indicator of the potential importance of DMS emissions from coral reef lagoon waters and the surrounding ocean, relative to direct emissions from aerially exposed corals at low tide, which occurs independently of the DMS gas transfer velocity. We expect spikes in DMSₐ detected when k_DMS is low to indicate direct emissions from the coral reef.

Gas transfer velocity parameterizations use wind speeds at 10 m above the sea surface (U₁₀). Thus, to calculate k_DMS, WS measured at the Heron Island Research Station (15 m) and RP 5 (2 m) were first scaled to U₁₀ using a logarithmic wind speed profile, as per Hoffman (2011) (equation 1). For equation 1, U is the observed wind speed at height z (m), and z₀ is the sea surface roughness, calculated using equation 2 (Hoffman, 2011). For equation 2, the constants C₀ = 3.7 and C₁ = −1.165, were derived from a linear fit between ln (k_DMS) and ln (fl), where a is the Charnock constant (0.0185), k is the von Kármán constant (assumed to be 0.4 for seawater) and g is the acceleration due to gravity (9.8 m s⁻²) (see Hoffman (2011) for details).
\[ U_{10} = U_{(z)} \left( \frac{\ln(10/z_0)}{\ln(z/z_0)} \right) \]  
\[ z_0 = \frac{C_0}{\frac{U_{(z)}}{10}} \]  

Total gas transfer velocity is a function of both the air \((k_a)\) and water-side \((k_w)\) transfer velocities, which are largely controlled by \(U_{10}\) and SST (Liss & Merlivat, 1986). DMS is a soluble gas produced ubiquitously throughout the ocean and thus, the concentration in seawater is far higher than in the atmosphere. Therefore, \(k_w\) is the primary rate limiting process in total DMS gas transfer. Consequently, many studies exclude air-side resistance to DMS transfer and assume that \(k_{DMS}\) is equivalent to \(k_w\). McGillis et al. (2000) demonstrated that air-side DMS transfer velocity becomes important during high wind speeds and/or low temperatures, such as those which frequently occur in the Southern Ocean (e.g., Yang et al., 2011). More recently, various eddy covariance methods have shown that \(k_{DMS}\) is linearly related to wind speed and thus, \(k_a\) is important across all wind speeds and temperatures (Goddijn-Murphy et al., 2012). We calculated \(k_{DMS}\) as a function of both \(k_a\), using the parameterization of Johnson (2010) and \(k_w\), using the parameterization of Goddijn-Murphy et al. (2012). Given that WS at Heron Island rarely exceeds 10 m s\(^{-1}\) and SST is relatively high (typically > 20°C), we found that including \(k_a\) in our calculation of \(k_{DMS}\) did not substantially influence overall transfer velocity, with differences < 0.002 cm hr\(^{-1}\) between calculations including or excluding \(k_a\). Therefore, for simplicity, we only included \(k_w\) at Heron Island (equations 3 and 4). This was done by first calculating \(k_w\) for a Schmidt number (Sc) of 600, which was then normalized to the SST dependent SC for DMS (\(Sc_{DMS}\)), provided in Saltzman et al. (1993). Given that we do not have data on seawater DMS concentrations and thus, cannot calculate DMS sea-air flux, this simple calculation of \(k_{DMS}\) as a function of only water-side resistance is sufficient for this analysis.

\[ k_w(600) = 2.1U_{10} - 2.8 \]  
\[ k_w(DMS) = k_w(600) \left( \frac{Sc_{DMS}}{600} \right)^{-0.4} \]

### 2.4. Correlation Analysis

Seasonal and diurnal variability for each time series was examined for the four survey periods. To investigate local drivers of DMS\(_a\) and particle number concentration variability at Heron Island, the data was filtered to exclude moderate to high WS (>2 m s\(^{-1}\)) and the resulting time series were cross-correlated. Cross-correlation measures the similarity between two time series, where one is lagged before (negative lag) or after (positive lag) the other. For example, when physical conditions are lagged after DMS\(_a\) (positive lag), DMS\(_a\) is considered to be the dependent variable, thereby implying that physical conditions affect DMS\(_a\) at a later time. Similarly, when DMS\(_a\) is lagged after D0.5 or D2.5, particle number is considered to be the dependent variable. The data was first detrended by removing the temporal mean and any missing values were filled using a linear interpolation. As the 2012, 2016 and 2018 surveys were all taken during summer (January–March), these data sets were combined for the correlation analysis and will henceforth be referred to as the summer data set \((n = 902)\). The July 2013 data will be referred to as the winter data set \((n = 430)\). DMS\(_a\) was also compared with daily mean MODIS AOD. Given that MODIS provides only two measures of AOD around midday in the southern GBR, the average morning DMS\(_a\) was calculated for the hours 01:00 to 12:00, excluding data when WS > 2 m s\(^{-1}\). This maximized the chance that any coral reef-derived particles would remain within the subset boundary around Heron Island and resulting changes in AOD would be captured by the satellite observations. Given the small number of survey days (60), the summer and winter data sets were combined for comparison with the daily AOD observations. DMS\(_a\), SST, CHL, D0.5, and D2.5 were log-transformed to a normal distribution. A conservative significance level \((\alpha = 0.01)\) was used to account for nonnormality in PAR, which was not improved with any data transformation. All other data were approximately normally distributed.
3. Results

3.1. Sources of DMS$_a$ at Heron Island

DMS$_a$ concentrations at Heron Island (Figure 2) were typically higher in summer (mean 3.2 nmol m$^{-3}$/78 ppt) than winter (mean 1.3 nmol m$^{-3}$/32 ppt). Variability in DMS$_a$ was also higher during the summer months, with a diel range of $\sim$2 to 4 nmol m$^{-3}$ (49–98 ppt), compared to $\sim$1 nmol m$^{-3}$ (24 ppt) in winter. This was true with the exception of a single large spike of DMS$_a$ during the Austral winter, in the evening of 25 July 2013 (Figure 2b) which reached an hourly average of 16.2 nmol m$^{-3}$ (maximum 45.9 nmol m$^{-3}$) or 396 ppt (maximum 1,123 ppt). This spike occurred when rain (not plotted) fell onto the aerially exposed coral reef during the evening low tide on July 25 (Swan, Jones, Deschaseaux; & Eyre, 2017). Several smaller spikes in DMS$_a$ were detected during the four survey periods and also occurred during afternoon low and rising tides (Figure 2).

As noted, $k_{DMS}$ was largely driven by WS, yet was not a good overall predictor of DMS$_a$ ($r^2 = 0.15, p < 0.001$). This is likely due to the several large spikes in DMS$_a$ mentioned above, which were often detected during relatively calm conditions (WS $< 5$ m s$^{-1}$) (Figure 2). In terms of wind direction (see Figure S1), DMS$_a$ and particle number concentration were highest in the prevailing south to southeasterly winds, across the range of observed WS (Figure 3). Northerly winds were not as common and were slower in velocity. DMS$_a$ and particle number concentration were relatively low in these air masses (Figure 3). Interannual variability in summer DMS$_a$ was also present, with consistently higher concentrations detected during both 2012 (mean 3.9 nmol m$^{-3}$/95 ppt) and 2016 (mean 3.7 nmol m$^{-3}$/91 ppt), yet relatively low DMS$_a$ detected in 2018 (mean 1.8 nmol m$^{-3}$/44 ppt), similar to concentrations expected in winter (Figure S2). CHL was used as a proxy for phytoplankton biomass, to investigate the potential influence of phytoplankton emissions on observed DMS$_a$. Although CHL did not correlate ($p > 0.05$) with daily mean DMS$_a$, interannual variability in summer CHL (Figure S2) agrees with that of DMS$_a$, with consistently higher mean concentrations in 2012 (0.28 mg m$^{-3}$) and 2016 (0.23 mg m$^{-3}$) and lower concentrations in 2018 (0.16 mg m$^{-3}$).

The climatology of CHL and MLD for the study region shows a seasonal increase in CHL during winter, coinciding with a deepening mixed layer (Figure 4). Daily mean CHL observed during our study periods was well above average in 2012 and 2016, however was lower and remained largely within the expected range ($\pm$2 SE) in winter 2013 and summer 2018 (Figure 4). Phytoplankton are significant sources of DMS and thus, a shift in biomass may have driven the observed interannual variability in DMS$_a$.

On a diurnal timescale, summertime DMS$_a$ was lowest at 13:00 when PAR was high (Figure 5a) and increased through the evening, reaching a maximum at 05:00 when PAR (Figure 5a), SST (Figure 5c), and Tide (Figure 5e) were low. Conversely, in winter, DMS$_a$ was highest at 13:00, shortly after the peak in PAR at 12:00 (Figure 5b). DMS$_a$ then increased with SST (Figure 5d) and during afternoon low tides (Figure 5f) in winter.

Table 2 shows the Pearson’s linear correlation coefficients for the hourly time series. In summer, DMS$_a$ negatively correlated with SST and PAR, and positively with WS. As expected, SST positively correlated with PAR and negatively with WS, reflecting solar heating during the day and evaporative cooling of the sea surface, respectively. In winter, DMS$_a$ only significantly correlated with Tide and WS (Table 2). SST also negatively correlated with WS in winter, yet did not correlate with PAR. To investigate the drivers of local DMS$_a$ variability at Heron Island, cross-correlation coefficients were investigated during calm conditions (WS $\leq 2$ m s$^{-1}$) and are shown in supporting information Figure S3. In summer, the negative correlation between DMS$_a$ and both PAR ($r = -0.31, p < 0.001$) and Tide ($r = -0.27, p < 0.001$) increased in strength and were highest at zero time lag. In winter, DMS$_a$ significantly positively correlated with SST at zero-time lag ($r = 0.22, p < 0.001$). WS no longer correlated with DMS$_a$ in summer or winter ($p > 0.05$).

The positive correlation between SST and PAR also increased as PAR was lagged 4 hr after SST in summer ($r = 0.59, p < 0.001$), and by 3 hr in winter ($r = 0.70, p < 0.001$). This reflects the delayed response of SST to increasing solar irradiance shown in Figures 2a–2d. For the summer 2016 and 2018 data, particle number concentration negatively correlated with SST and PAR (Table 2). During low WS ($\leq 2$ m s$^{-1}$), the correlation coefficients strengthened as particle number concentration was lagged 4 hr after SST (for D0.5 $r = -0.30$,
Figure 2. Time series of hourly DMS$_a$ (-----), WS (••••), $k_{DMS}$ (-----), PAR (-----), Tide (-----) and SST (-----) at Heron Island during (a) 2012, (b) 2013, (c) 2016, and (d) 2018. Date axis indicates noon local time (UTC + 10 hr).
$p < 0.05$; for $D_{2.5}$ $r = -0.66$, $p < 0.001$) and with zero-time lag for PAR (for $D_{0.5}$ $r = -0.45$, $p < 0.001$; for $D_{2.5}$ $r = -0.70$, $p < 0.001$; see supporting information Figure S3). All other cross-correlation coefficients did not improve or were nonsignificant ($p > 0.05$).

### 3.2. Sources of Atmospheric Particles at Heron Island

Low-altitude marine air masses arrived at Heron Island on 77% (20 of 26 days) of the 2016 and 2018 study days, with the exception of 13–16 February 2016 and 30–31 January 2018 (Figure S1). Figure 6 shows the time series of $DMS_a$, WS and particle number concentration, all of which appear to follow similar trajectories and are therefore positively correlated (Table 2).

WS is an important driver of DMS and marine aerosol emissions and thus, increases in $DMS_a$ and particle number concentration were detected across the range of observed WS at Heron Island (Figure 3). Solar
radiation also influences the photochemical cycling of trace gases, for example by facilitating the oxidation of DMS$_a$ to various sulfate aerosol precursor compounds. Interestingly, the correlation between DMS$_a$ and D0.5 ($r = 0.58$, $p < 0.001$, $n = 572$) increased for wind speeds $\leq 2 \text{ms}^{-1}$ ($r = 0.60$, $p < 0.001$, $n = 59$) and further for daylight (PAR $\geq 1 \text{mol m}^{-2} \text{hr}^{-1}$) hours ($r = 0.82$, $p < 0.001$, $n = 20$). Conversely, the correlation between DMS$_a$ and the larger particles (D2.5) ($r = 0.55$, $p < 0.001$, $n = 572$) slightly improved for WS $\leq 2 \text{ms}^{-1}$ ($r = 0.57$, $p < 0.001$, $n = 59$), but not for daylight hours ($p > 0.05$). When time delays were considered, the cross-correlation coefficients were highest with zero-time lag, regardless of WS, and declined almost linearly as DMS$_a$ was lagged after both D0.5 and D2.5 (Figure S3). Similarly, a positive correlation was present between DMS$_a$ and MODIS AOD ($r = 0.33$, $p < 0.01$, $n = 60$), which strengthened on calm days when WS $\leq 2 \text{m s}^{-1}$ ($r = 0.71$, $p < 0.05$, $n = 11$). Figure S4 shows the coefficients for the correlations between DMS$_a$ and both D0.5 and AOD, as a function of WS, showing a clear increase in correlation strength during low wind speeds.

4. Discussion

Our findings support existing analyses of the 2012 and 2013 survey data, which identified a seasonal cycle in DMS$_a$ at Heron Island and a strong local source of emissions from the coral reef at low tide (Swan, Jones, Deschaseaux, & Eyre, 2017). We have built on these findings through analysis of additional measurements of DMS$_a$, SST, PAR, Tide and in situ and satellite observations of aerosol concentration, through which we have
identified seasonally dependent diel DMS$_a$ cycles and a significant link between DMS$_a$ and local oceanic and atmospheric parameters at Heron Island.

### 4.1. Sources of DMS$_a$ at Heron Island

A seasonal DMS$_a$ cycle was observed at Heron Island, with increases in summer driven by temperature and irradiance-dependent shifts in phytoplankton biomass and coral physiological stress (Broadbent & Jones, 2006; Jones et al., 2007). DMS$_a$ was highest during the prevailing south to southeasterly winds and positively correlated with WS, indicative of accumulating emissions from local and upwind coral reefs, lagoon waters and adjacent Coral and Tasman Seas. However, WS and $k_{DMS}$ were not good predictors of DMS$_a$ during relatively calm conditions (WS < 5 m s$^{-1}$), indicative of local emissions from the coral reef flat. This is supported by several large spikes in DMS$_a$ detected during afternoon low and rising tides. The coral reef flat often becomes aerially exposed during low tide and DMS may be directly transferred from the coral reef to the MBL (Hopkins et al., 2016; Swan, Jones, Deschaseaux, & Eyre, 2017). DMS emissions also increase on rising tides when DMS in coral mucous dissolves and accumulates in tidal slack waters (Hopkins et al., 2016; Jones et al., 2007; Jones et al., 2018; Swan, Jones, Deschaseaux, & Eyre, 2017). The highest spike occurred during the evening low tide of 25 July and Swan, Jones, Deschaseaux, and Eyre (2017) concluded that rainfall (hyposalinity) and rapid temperature change stressed the exposed corals. This combination of stressors most likely triggered an upregulation of DMSP catabolism to DMS, resulting in the detected DMS$_a$ spike above the coral reef (Deschaseaux et al., 2014; Gardner et al., 2016; Hopkins et al., 2016; Swan, Jones, Deschaseaux, & Eyre, 2017). Thus, although DMS$_a$ detected at Heron Island was largely within the range reported for other ocean regions (~100–200 ppt) (Lana et al., 2011), occasional spikes well above this range (>1,000 ppt) can occur during low and rising tides and persist for up to ~8 hr (Swan, Jones, Deschaseaux, & Eyre, 2017). Similarly, reported concentrations of DMS$_a$ detected above the nearby One Tree Reef lagoon are higher than that typically detected over remote oceans, reaching 23 nmol m$^{-3}$ (563 ppt) (Jones et al., 2007). Therefore, the coral reef is a much greater, albeit intermittent, source of DMS$_a$ compared to open ocean sources.

Sources of DMS$_a$ in coral reefs include zooxanthellate Scleractinian (Raina et al., 2013) and Alcyonacea (soft) corals (Haydon et al., 2018), crustose coralline algae (Burdett et al., 2015), marine macroalgae and free-living microalgae (Stefels, 2000; Van Alstyne & Puglisi, 2007), giant clams (Hill et al., 2000), as well as carbonate sediments dominated by benthic microalgae (Deschaseaux et al., 2019). Corals, particularly Acropora spp. (Broadbent et al., 2002; Broadbent & Jones, 2004; Swan et al., 2016), contain the highest reported...
concentrations of DMS/P in coral reefs. These species are abundant at Heron Island and are likely the dominant local source of DMS$_a$ in the coral reef ecosystem (Swan, Deschaseaux, et al., 2017). *Acropora* spp. release large quantities of DMS during aerial exposure (Hopkins et al., 2016), which can occur during low tide, and during exposure to thermal and irradiance stress (Deschaseaux, Beltran, et al., 2014; Deschaseaux, Jones, et al., 2014; Jones et al., 2014; Jones & King, 2015; Raina et al., 2013). However, DMS emissions from *Acropora* spp. have been shown to decline by 92% and 87% during exposure to SST $>$ 26°C and PAR $>$ 6 mol m$^{-2}$ hr$^{-1}$, respectively (Fischer & Jones, 2012). As oxidative stress increases in the coral holobiont, an upregulation of DMS/P oxidation to DMSO and a decline in ambient DMS concentrations occurs (Deschaseaux, Beltran, et al., 2014; Deschaseaux, Jones, et al., 2014). A similar trend was observed in late February 2016 (Figure 2c), when daily maximum DMS$_a$ declined prior to the coral bleaching event in March. During this period SST exceeded the estimated coral thermal stress threshold of 27.3°C, calculated as the climatological maximum monthly mean SST (Liu et al., 2006), for several consecutive days. WS and $k_{DMS}$ were low during this time, suggesting that the observed variability in DMS$_a$ was driven by a decline in biosynthesis and emissions from corals exposed to high levels of thermal and oxidative stress.

### 4.2. Interannual Variability in DMS$_a$ and CHL

Interannual variability in summer DMS$_a$ showed a decline in concentrations in January–February 2018. In the absence of data on seawater DMS concentration, we may only speculate as to the cause of this apparent decline. One possible cause is lower CHL (an indicator of phytoplankton biomass) in the southern GBR and surrounding ocean, which displayed similar interannual variability to that of DMS$_a$ (Figure S2). The seasonally low DMS$_a$ observed in summer 2018 may therefore have been driven by lower phytoplankton biomass. Another possible explanation is that reduced coral health and/or coral cover in the southern GBR after two consecutive coral bleaching events in the summers of 2016 and 2017, resulted in lower DMS emissions from the coral reef in 2018. Note that this explanation is less likely as coral mortality rates were relatively low in the far southern GBR (~5% at Heron Island) (AIMS, 2018).

Interestingly, the climatology of CHL displayed a seasonality that is not typical of subtropical–temperate latitudes. In temperate latitudes, increases in phytoplankton biomass typically occur in spring and autumn, coinciding with optimal PAR and adequate nutrients in the euphotic zone as the MLD shoals and deepens, respectively (Longhurst, 1995; Winder & Cloern, 2010). However, our analysis shows a clear winter increase in CHL (Figure 4). This seasonal cycle was previously documented in the southern Coral Sea adjacent to Heron Island, with seasonal increases in phytoplankton biomass occurring from May to September (Welch et al., 2016). Normally light is a limiting factor for phytoplankton growth in winter in temperate latitudes however, MLD remains relatively shallow ($< 85$ m) in our study region. Reduced photoinhibition, in combination with enhanced upwelling of nutrients with a deepening MLD may explain this seasonal cycle.

### 4.3. Diel Cycling of DMS$_a$

Seasonally dependent diel DMS$_a$ cycles were also observed. In summer, DMS$_a$ was lowest at 13:00 when SST and PAR (and the abundance of oxidative free radicals such as OH) were high and increased through the evening. Consequently, DMS$_a$ negatively correlated with both SST and PAR during summer. This trend likely reflects enhanced photo-oxidation of DMS$_a$ by free radicals, which are highest at solar noon. The correlation between DMS$_a$ and PAR increased during calm conditions, suggesting that PAR alone explains up to 10% of the diel variability in summer DMS$_a$. Photo-oxidation is a significant sink of both DMS$_w$ and DMS$_a$ (Gabric et al., 2008; Gali et al., 2013; Toole et al., 2003) and a source of aerosol precursors including MSA, SO$_2$ and H$_2$SO$_4$ (Andreae & Crutzen, 1997; Ayers & Gillett, 2000; Barnes et al., 2006). This diel cycle agrees with previous observations of DMS$_a$ in remote oceans (Ayers & Gillett, 2000; Bandy et al., 1996; Gali et al., 2013; Marandino et al., 2007; Warneke & De Gouw, 2001) and highlights the importance of photochemistry as a driver of diurnal DMS$_a$ variability.

Conversely, in winter, DMS$_a$ was highest at 13:00 and decreased through the evening. PAR was lower and midday levels persisted for a shorter period of time in winter ($>$ 4 mol m$^{-2}$ hr$^{-1}$ for ~2 hr in winter compared to ~5 hr in summer). Slower DMS$_a$ oxidation with lower solar irradiance, may have revealed enhanced biogenic DMS emissions with rising SST and during afternoon low tides. This is supported by the positive correlation between DMS$_a$ and SST, and negative correlation between DMS$_a$ and Tide in winter (Table 2; Figure S3). As discussed above, corals are a significant source of DMS$_a$ during low and rising tides. SST
was well below the estimated coral bleaching threshold in winter and thus, the positive correlation between DMS$_a$ and SST likely reflects increased coral DMSP biosynthesis and cleavage to DMS in response to rising SST (Deschaseaux, Beltran, et al., 2014; Deschaseaux, Jones, et al., 2014; Raina et al., 2013).

We recognize that the rate of DMS/P biosynthesis and loss to various sinks including microbial metabolism, vertical mixing and photochemical processes are important factors in determining DMS$_a$ concentration (Gabric et al., 2008; Gali et al., 2013; Toole et al., 2003). It is not possible to quantify the contribution of individual DMS/P sources and sinks without complementary DMS$_a$ data. However, the results presented here indicate that variability in DMS$_a$ at Heron Island is largely driven by seasonal shifts in phytoplankton biomass and coral physiological stress, and by photochemistry at a diel scale in summer.

### 4.4. Potential Feedback Between Marine Aerosols and Coral Physiological Stress

In both summer and winter, SST positively correlated with PAR, reflecting enhanced solar heating during the day; a contributor to coral physiological stress and bleaching (Lesser, 2010). Interestingly, increased particle number concentration in fine to coarse size ranges ($\geq 0.5 \, \mu m$) negatively correlated with both PAR and SST (Table 2, Figure S3). This suggests that increased particle number concentration enhances incoming PAR backscattering which consequently lowers SST. Particle number concentration accounted for up to 44% of variability in PAR and up to 49% of variability in SST. Marine aerosols may therefore contribute to the formation of a negative local climatic feedback over the coral reef, reducing solar irradiance and SST, thereby mitigating coral physiological stress. Other recent work has identified potential feedbacks between PAR, SST, AOD, and LLC in the GBR over several days using remotely sensed data (Cropp et al., 2018; Jackson et al., 2018; Leahy et al., 2013). Similarly, this study demonstrates that a short-term (< 1 day), local feedback may exist between atmospheric particles, SST and PAR over the southern GBR.

### 4.5. Sources of Atmospheric Particles at Heron Island

The concentration of atmospheric particles ranging in diameter from 0.5 to 2.5 and > 2.5 $\mu m$ was also measured at Heron Island during January and February in 2016 and 2018. Additional satellite-derived observations of total AOD were also obtained for all four survey periods and potential sources of these atmospheric particles was investigated.

Westerly winds were dominant on a small number of days, primarily in winter 2013 (Figure S1). On these days, particles were likely influenced by continental sources including industrial emissions (Junkermann & Hacker, 2015) from nearby Gladstone (Figure 1a), continental dust (Cropp et al., 2013), wildfires and biomass burning (Chen et al., 2019). On the remaining study days, prevailing southeasterlies transported marine aerosols from the GBR and adjacent Coral and Tasman Seas to Heron Island. This is supported by positive correlations between particle number concentration and WS, which suggests that particles in the observed size range ($\geq 0.5 \, \mu m$) were dominated by wind-driven emissions of sea spray aerosols (SSA) (Cochran et al., 2017).

Previous studies have characterized marine aerosols in the GBR as clusters of sea salt, organics (Mallet et al., 2016) and additional non–sea salt sulfate fractions largely derived from biogenic emissions of volatile sulfur compounds such as DMS (Modini et al., 2009; Swan et al., 2016). Biogenic sulfates influence atmospheric particles via two main pathways. When preexisting aerosol surface area is low, DMS-derived H$_2$SO$_4$ may undergo homogenous nucleation to form new nss-SO$_4$ particles (Andreae & Crutzen, 1997; Andreae & Rosenfeld, 2008; Charlson et al., 1987). This pathway is thermodynamically expensive and thus, predominantly occurs in the free troposphere where conditions are more favorable (Korhonen et al., 2008). Turbulent vertical transfer of DMS-derived sulfates to the free troposphere and subsequent entrainment to the MBL is therefore a significant source of new nss-SO$_4$ aerosols (Korhonen et al., 2008; Sanchez et al., 2018). Alternatively, heterogeneous oxidation of DMS-derived aerosol precursor compounds commonly occurs in the MBL, contributing to the growth of existing particles (Hoffmann et al., 2016; Woodhouse et al., 2013). Back trajectories show that marine air masses remained within the MBL (< 500 m ASL) within 24 hr of arriving at Heron Island (Figure S1), suggesting that the latter is the dominant process by which observed DMS$_a$ could potentially influence atmospheric particles during the surveys at Heron Island.

Previous studies in the GBR have demonstrated significant, positive correlations between estimates of coral physiological stress and AOD, which increase during calm conditions, suggestive of a local aerosol source.
above the coral reef (Cropp et al., 2018; Jackson et al., 2018). Other global studies have shown regionally variable sensitivity of fine-mode AOD and CCN number to DMS emissions, which is strongest in relatively remote, pristine ocean regions (Fiddes et al., 2018; Vallina et al., 2007; Woodhouse et al., 2013). Similarly, our analysis shows a positive correlation between DMS_0 and observed particle number concentration. Maximum cross-correlation coefficients occurred at zero-time lag and since the 0.5 μm minimum measured particle size is significantly larger than the < 0.1 μm size range expected for newly nucleated secondary aerosols, we do not expect that this positive correlation reflects DMS-derived new particle formation events. Furthermore, it is unlikely that DMS_0 underwent oxidation and homogenous nucleation to new nss-SO_4 within the MBL within 1 hr. Rather, the positive correlations reflect a common driver of variability, such as wind-driven emissions of SSA and DMS sea-air flux. Interestingly however, the correlation between DMS_0 and particle number concentration for particles in the smaller size range (0.5–2.5 μm), increased during low WS (≤2 m s⁻¹) and sunlight (PAR ≥ 1 mol m⁻² hr⁻¹), when conditions for the local oxidation of DMS to sulfate aerosol precursors were optimal. Wind-driven emissions alone do not explain this increase in correlation strength during calm, daylight hours. Photochemical oxidation is a strong sink of DMS_0 and source of sulfate aerosol precursors in summer at mid-day at Heron Island (Figure 3a). Condensation of DMS-derived sulfates on existing aerosol surfaces can alter the chemical composition of aerosols and contribute to growth to CCN activation sizes (Cochran et al., 2017; Grythe et al., 2014; O’Dowd & de Leeuw, 2007). Thus, existing fine particles consisting of fine SSA (< 0.5 μm), organics and biogenic sulfates from upwind oceanic sources, can continue to grow via aqueous-phase oxidation in cloud droplets and condensation of DMS-derived sulfates in the southern GBR. The relationship between DMS_0 and D0.5 may therefore reflect rapid oxidation of DMS_0 and subsequent condensational growth of existing fine particles (< 0.5 μm) to the detectable size range of this study (≥0.5 μm). DMS-derived aerosol precursors may therefore play an important role in the local radiative balance by facilitating particle growth and altering the climate potential of marine aerosols (Cochran et al., 2017; Grythe et al., 2014; O’Dowd & de Leeuw, 2007).

Furthermore, DMS_0 strongly positively correlated with MODIS AOD during calm, daylight hours. Processes driving the observed variation in DMS emissions could therefore account for up to 67% of the variability in situ particle number concentration and up to 50% of MODIS AOD variability. This is in agreement with previous work which characterized aerosols in the southern GBR as ~60% sulfates, likely derived from DMS (Modini et al., 2009) and provides further support for the hypothesis that a significant link exists between DMS emissions and marine aerosols in the southern GBR. Many studies focus on the role of DMS in new particle formation events however, this analysis suggests that DMS may be just as important in facilitating the growth of existing aerosols in the GBR.

### 4.6. Future Directions

The results presented in this analysis support existing findings that corals in the GBR are a significant source of DMS_0 (Fischer & Jones, 2012; Haydon et al., 2018; Jones et al., 2018; Raina et al., 2013; Swan et al., 2016; Swan, Jones, Deschaseaux, & Eyre, 2017). Ongoing ocean warming and coral bleaching may therefore lead to a change in DMS emissions from the GBR. However, it is not yet clear how coral DMSP biosynthesis and catabolism to DMS will be affected under future climate change scenarios. Furthermore, it is not known whether the DMS antioxidant system and/or the proposed DMS-aerosol-cloud feedback will assist corals in coping with these impacts. Corals may change their endosymbiont composition via symbiont switching and/or shuffling to favor thermo-tolerant species and lower oxidative stress (Bay et al., 2016; Smith et al., 2017). For example, Acropora spp. favor Clade D symbiont dominance when exposed to thermal stress (Jones & King, 2015), which can increase temperature tolerance by ~1.5°C (Berkelmans & Van Oppen, 2006). These temperature-tolerant clades are typically weaker producers of DMSP (Bay et al., 2016; Deschaseaux, Beltran, et al., 2014), and may result in a decline in DMS emissions from the GBR as these species become more abundant. Note that thermal tolerance thresholds are variable within zooxanthellae type and do not always predict the rate of DMSP biosynthesis (Steinke et al., 2011). The ability of the coral holobiont to acclimate to the rapidly changing climate will also affect changes to DMS emissions. DMSP-producing corals have a close association with a wide range of microbes and thus, harbor a diverse genome (Bourne et al., 2016). It is hypothesized that transgenerational epigenetic inheritance may facilitate rapid phenotypic change, allowing corals to acclimate to rises in SST (Torda et al., 2017). Further research is
needed to determine how the rate of coral acclimatization and changes to coral reef community structure will affect DMS emissions from the GBR in future and whether this will affect local aerosol emissions.

5. Conclusions

Both local and remote oceanic sources of DMS$_a$ were detected at Heron Island. Our results support previous findings that the coral reef is a strong source of DMS$_a$ above the background oceanic signal during calm conditions, particularly at low tide. Variability in DMS$_a$ is primarily driven by temperature and irradiance-dependent seasonal shifts in phytoplankton biomass and coral physiological stress and by photochemical processes at the ocean surface. The diel DMS$_a$ cycle varied between season at Heron Island, with daily maximums occurring at midday in winter, yet daily minimums at midday in summer. This is attributed to rapid photo-oxidation of DMS$_a$ with high solar irradiance in summer and a slower oxidation rate revealing enhanced biogenic emissions at midday in winter.

We also identified a significant link between DMS$_a$ and atmospheric particle number concentration at Heron Island. Particles in the observed size range (> 0.5 $\mu$m) were likely dominated by SSA. Wind speed is an important driver of SSA emissions and DMS sea-air flux and thus, this link reflects wind-driven variability. However, the correlation between DMS$_a$ and intermediately sized particles (0.5–2.5 $\mu$m) increased substantially during calm conditions and sunlight, when the rate of local DMS$_a$ oxidation to sulfate aerosol precursor compounds is highest. Although particles in this size range are larger than that of newly nucleated nss-SO$_4$ particles, this relationship could reflect DMS-derived growth of existing particles which is the dominant pathway by which biogenic trace gases influence marine aerosols in the MBL. Increased particle number concentration negatively affected PAR and SST, potentially forming a negative local climatic feedback over the coral reef. Our results provide a quantitative assessment of the strength and direction of the relationship between coral physiological stress, phytoplankton biomass, and DMS and marine aerosol emissions in the southern GBR. Further research is required to decipher the role of biogenic aerosol precursors and their potential to affect the growth, composition, and climate properties of marine aerosols. This is particularly important in remote coral reefs such as the GBR, where biogenic sulfates and other VOCs are an important source of secondary aerosol and a major determinant of local climatic variation.

Data availability

Atmospheric dimethylsulfide, wind speed, wind direction, and particle number concentration data are available from the Southern Cross University electronic publications website (https://epubs.scu.edu.au/data_collections/). The 2012 and 2013 data are available at doi: 10.4226/47/58781bfbca619 (Swan, Jones, & Deschaseaux, 2017a), the 2016 data are available at doi: 10.4226/47/59c460fde8322 (Swan, Jones, & Deschaseaux, 2017b), and the 2018 data are available at doi: 10.25918/5c772187f39b6 (Swan et al., 2018).

Sea surface temperature obtained from IMOS floats at Heron Island can be downloaded from https://apps.aims.gov.au/metadata/view/0abcf26e-f3de-4e2a-80b4-909090d9c91d (AIMS 2019a). Photosynthetically active radiation obtained from IMOS floats can be downloaded for Heron Island at https://apps.aims.gov.au/metadata/view/05cbcf31-8d43-4309-931c-b16603785be5 (AIMS 2019b) and for One Tree Island at https://apps.aims.gov.au/metadata/view/6a00144d-2aa4-4786-bd0d-dab24ea5aaaf (AIMS 2019c). IMOS weather station wind speed and wind direction at Heron Island can be downloaded from https://apps.aims.gov.au/metadata/view/45cf6bef-0999-4c82-ba31-7b9ff54e22db (AIMS 2019d).

Chlorophyll-a concentration and aerosol optical depth derived from the Moderate Resolution Imaging Spectroradiometer (MODIS) was downloaded from the NASA Ocean Color DAAC (http://oceancolor.gsfc.nasa.gov). Mixed layer depth data derived from Argo float measurements can be found at doi: 10.1002/2017GL073426 (Holte et al., 2017).

Conflict of interest

The authors declare no conflict of interest.
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