Supplementary Information: Ship fuel sulfur content regulations may exacerbate mass bleaching events on the Great Barrier Reef

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1 Model evaluation

We evaluated the output from the FSC05 simulations (representing the contemporary state of shipping emissions for 2022) against a range of observational datasets to examine the suitability of WRF-Chem to interrogate atmospheric chemistry and aerosol, cloud and radiation impacts over the GBR and Coral Sea regions. We provide a summary of the results here.

For meteorology, we firstly compare WRF-Chem to in-situ meteorological observations at Davies Reef near Townsville in the GBR (Figure 1). Davies Reef is at 18.83 ^{o}S, 147.63 ^{o}E. Observations for Davies Reef are collected by, and available through, the Australian Institute of Marine Science (https://weather.aims.gov.au/). The temperature, relative humidity and wind observations are made close to the ocean surface by a weather station attached to a mooring. We also compare the model to to observations taken on the RV Magnetic during fieldwork as part of the Reef Restoration and Adaptation Program's Cooling and Shading subprogram during February 2022 (Figure 2). The voyage left from and returned to Townsville, collecting atmospheric measurements between Townsville and the Davies Reef area.

WRF-Chem captures most hour-to-hour variations in surface air temperature in both datasets, underestimating temperature observations by 2.1 % on average on the Magnetic and 0.1 % at Davies Reef. We compare temperature from WRF-Chem using the "T2" variable rather than the mean temperature in the lowest model layer. Day to day variations in wind speed and direction are very well captured by the model at both Davies Reef and on the ship. Relative humidity proves more challenging, with the model overestimating the Davies Reef observations by 14 % on average. The overestimate is only 3 % for the ship dataset.

Total particle numbers and cloud condensation nuclei (CCN) counts were also measured on the RV Magnetic voyage. Particle numbers were measured using a Scanning Electrical Mobility Spectrometer (SEMS, Brechtel Manufacturing Inc.) The SEMS instrument measures total particle concentration in 151 size bins ranging from 10 nm to 2 μm. For the comparison to WRF-Chem in Figure 3(a), total particle counts in all size bins have been summed. WRF-Chem captures the observed total particle numbers during periods in the time series, for example on 19 and 20 February, while underestimating the observations at other times especially on 21 and 22 February. The same periods of good agreement and underestimation are seen for CCN at 0.5 % supersaturation (Figure 3(b)), measured using a CCN counter. The normalised mean bias for WRF-Chem compared to the observations, for both CCN and total particle counts, was -28 %. This indicates that despite the addition of updated shipping emissions in this study, the model may underestimate aerosol numbers over the GBR. This is likely driven largely by an underestimate of black carbon, which was 50 % lower on average in the model than measured using a Tricolor Absorption Photometer on the RV Magnetic. We note that the underestimate of surface-level aerosol concentrations is consistent underestimates of sulfate and black carbon in, to our knowledge, the only previous WRF-Chem study focusing on the GBR [1]. With this indication of underestimated land-based anthropogenic and/or wildfire emissions considered, we do not take the model's 28 % underestimate of total particle numbers and CCN to preclude the use of WRF-Chem in this study of marine aerosol/cloud interactions.

This approach is backed by strong agreement between model and satellite observations for domain-wide solar radiation and cloud variables. This is shown in Figure 4, where we compare the model to satellite datasets of shortwave and longwave radiation, as well as cloud liquid water path and aerosol optical thickness. For short and longwave radiation we use data from the Clouds and the Earth's Radiant Energy System (CERES) project, which uses multiple satellite datasets to constrain radiation fluxes [2, 3]. Specifically, we download the CERES synoptic scale short and longwave radiation at top of atmosphere at $1 \times 1^{\circ}$ spatial resolution ("SYN1deg", details and data download at https://ceres.larc.nasa.gov/data). Figures 4(a) and (b) show that the spatial agreement between both short and longwave modelled and observed products is strong. The WRF-Chem and satellite datasets agree both domain-wide, and over the ocean only, within 5 % for shortwave radiation and within 8 % for longwave radiation.

Cloud liquid water path (LWP) validation is crucial for this study because it indicates not only whether the model does a good job of representing the location of clouds, but also their optical properties. We use WRF-Chem LWP as the basis of calculating cloud optical depth, and thus diagnosing changes in cloud optical propterties attributable to ship emissions. We use LWP data from the Moderate Resolution Imaging Spectroradiometer (MODIS) level 3 $1 \times 1°$ product (MMYD08_D3, from the Aqua platform, https://modis.gsfc.nasa.gov/data/dataprod/mod08.php). As the daily Aqua overpass is 13:30, we average WRF-Chem output from 13:00 and 14:00 to compare with MODIS LWP. Figure 4(c) shows good agreement between model and satellite observations, especially over the ocean. For example, lower LWP close to the coas around Townsville, Cairns and Gladstone, compared to higher values over the Coral Sea, is captured by the model. Overall, the model and observations agree within 2 % over the oceanic part of the domain.

The comparison of modelled aerosol optical thickness (AOT) with observed satellite data (Figure 4(d)) is more challenging, again highlighting discrepancies in the WRF-Chem's aerosol emissions over this region. AOT output for WRF-Chem is calculated from modelled aerosol mass concentrations and the mass-extinction method outlined in [4]. We compare WRF-Chem with output from the Modern-Era Retrospective analysis for Research and Applications Version 2 (MERRA-2, https://gmao.gsfc.nasa.gov/reanalysis/merra-2/). Average AOT for the period 18-27 February 2022 was downloaded downloaded from the NASA data hub 'Giovanni' (https://giovanni.gsfc.nasa.gov/giovanni/). We used reanalysis data instead of satellite observations because satellite AOT measurement relies on cloud-free conditions, and as shown in the LWP maps, cloud was very common in this region during the analysis period. The spatial pattern of modelled and measured AOT do not agree well, especially over the land. Despite this, the WRF-Chem AOT averaged over the simulation domain is only 5 % higher than MERRA-2.

Finally we compare modelled and measured tropospheric column amounts for the key trace gases nitrogen dioxide (NO_2) and formaldehyde (HCHO). The observations used for this comparison come from the Tropospheric Monitoring Instrument (TROPOMI) aboard the Sentinel-5P satellite (https://www.tropomi.eu/) [5, 6]. As with MODIS Aqua, the overpass time for TROPOMI is 13:30 local time so for the comparison, we average model output from 13:00 and 14:00 local time. As shown in Figure 5(a), the spatial patterns of modelled and observed NO_2 agree well and across the domain, the mean column amounts agree within 5 %. This 5 % discrepancy comes largely from WRF-Chem underestimating NO_2 in the Gladstone region, in the south-eastern part of the domain. Figure 5(b) shows the TROPOMI comparison to WRF-Chem HCHO. Again the spatial patterns agree quite well although WRF-Chem produces slightly more HCHO over the GBR and Western Coral Sea than seen in TROPOMI. The mean values agree within 9 %.

In summary, while it appears that WRF-Chem aerosol counts and optical thickness could be improved by optimising terrestrial emission inventories, we see generally good agreement between modelled and observed values for surface meteorology and

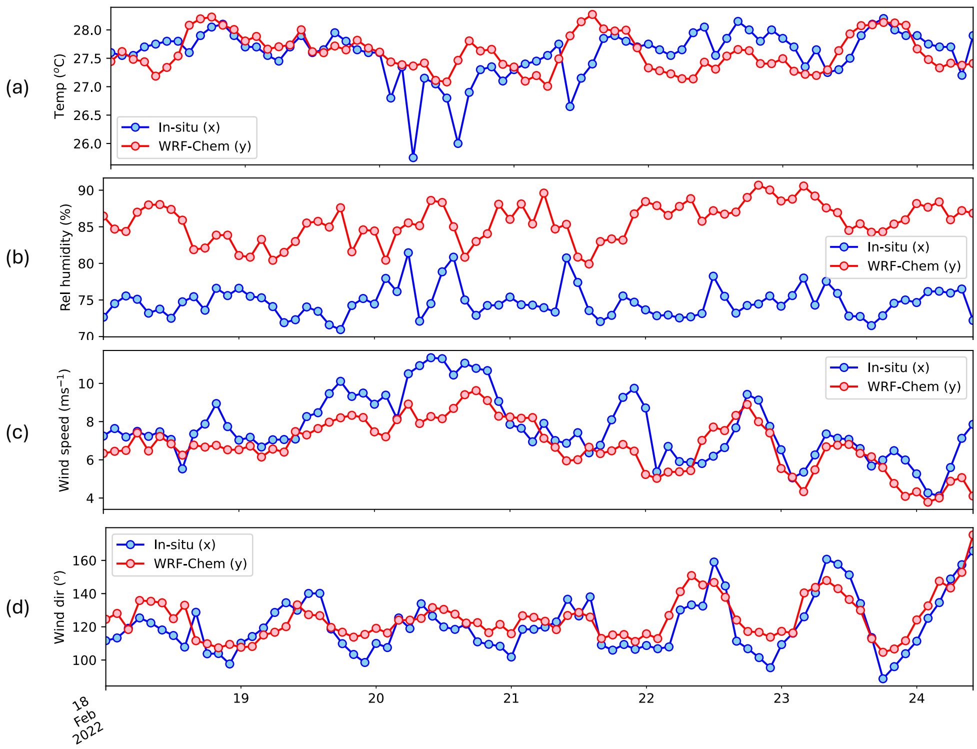

Fig. 1 Comparison of WRF-Chem output with surface in-situ meteorological observations at Davies Reef in the GBR. Timeseries panels show (a) temperature, (b) relative humidity, (c) wind speed and (d) wind direction. The model is sampled in the lowest altitude layer (0-34 m) and in the closest 9x9 km gridbox to the location of Davies Reef (18.83 ^{o}S, 147.63 ^{o}E).

tropospheric columns of trace gases integral to the oxidative chemistry budget. Most importantly, we see strong consistency in spatial pattern and magnitude for the model-measurement comparison of shortwave and longwave radiation as well as cloud liquid water path. This gives confidence in interpreting changes in radiation and cloud optical thickness attributable to ship emissions.

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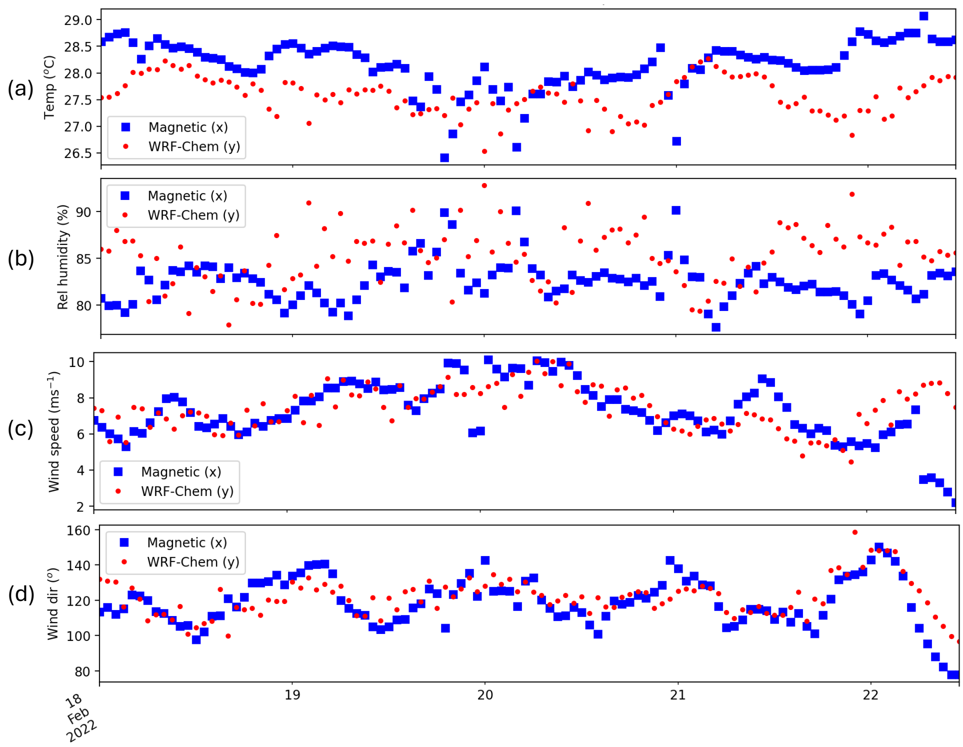

Fig. 2 Comparison of WRF-Chem output with in-situ meteorological observations taken on the RV Magnetic voyage, transiting between Townsville and Davies Reef in the GBR. Timeseries panels show (a) temperature, (b) relative humidity, (c) wind speed and (d) wind direction. The model is sampled in the lowest altitude layer (0-34 m) and in the closest 9x9 km gridbox to the location of the ship at each timestep.

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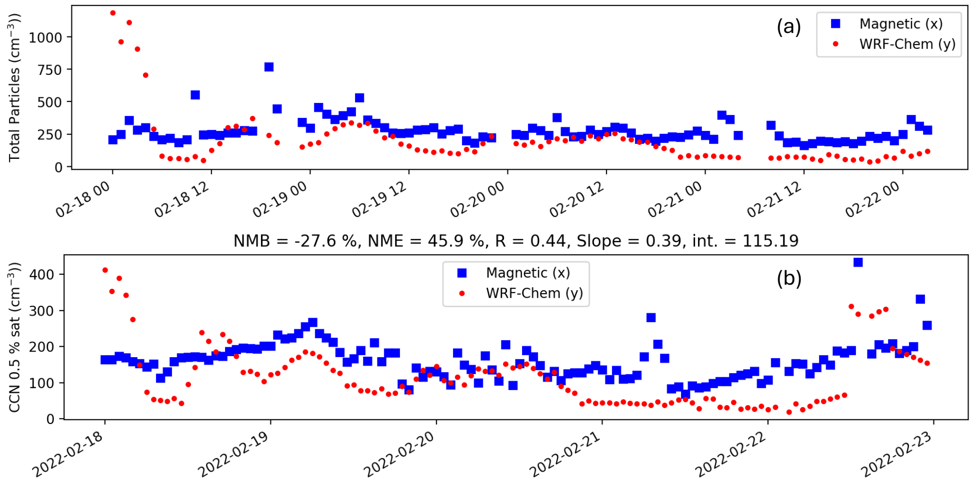

Fig. 3 Comparison of WRF-Chem output with in-situ aerosol observations taken on the RV Magnetic voyage, transiting between Townsville and Davies Reef in the GBR. Timeseries panels show (a) total particle counts and (b) CCN at 0.5 % supersaturation. Model sampling details are the same as in Figure 2.

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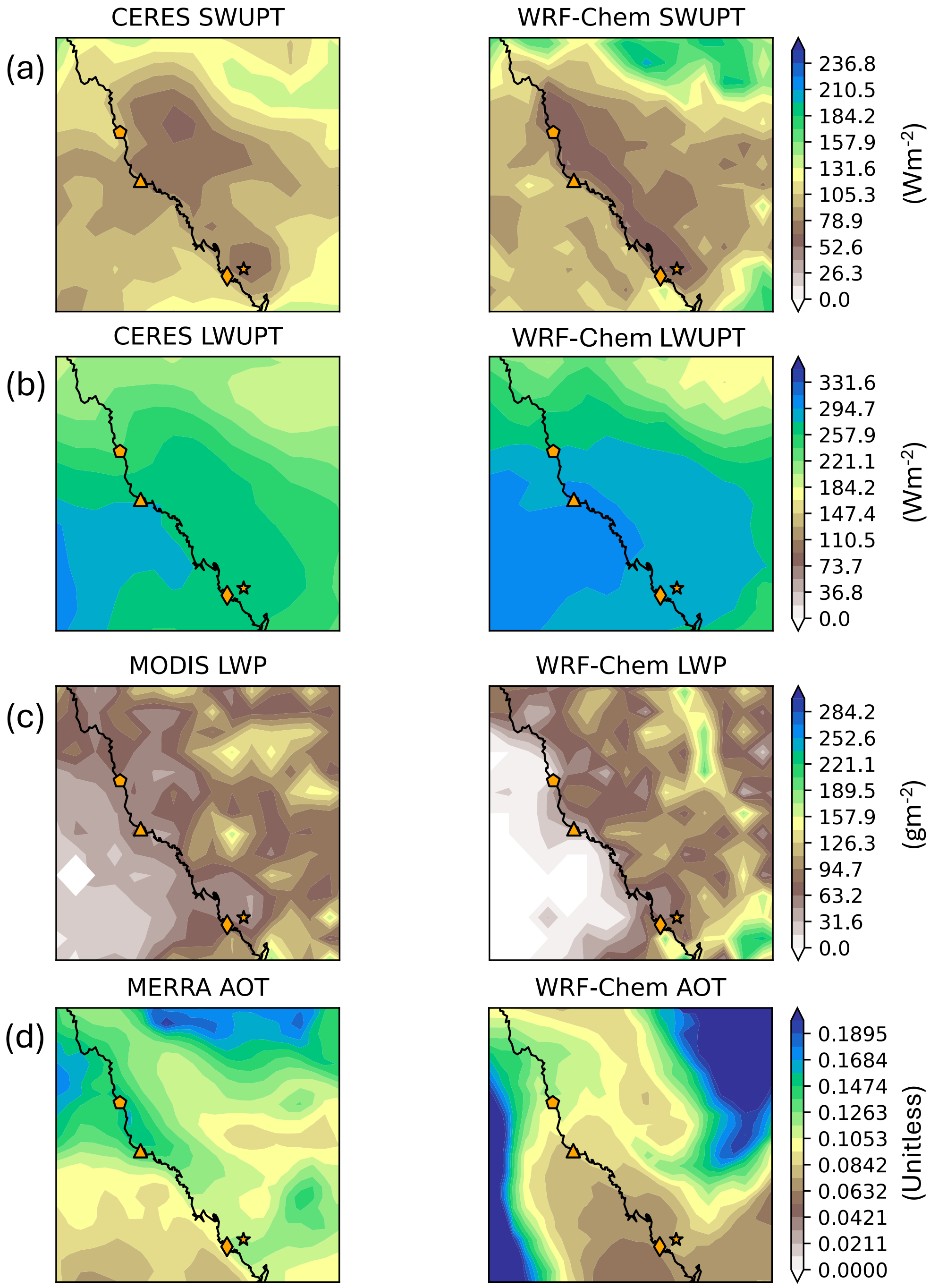

Fig. 4 Comparison of WRF-Chem radiation and cloud variables across the simulation domain. (a) Shows the comparison of shortwave upwelling radiation at the top of the atmosphere (SWUPT) from CERES, (b) shows the same as (a) but for long wave radiation (LWUPT). (c) Shows the comparison of cloud liquid water path (LWP) from MODIS while (d) shows the comparison to aerosol optical thickness (AOT) from MERRA reanalysis. Location markers are (star) Heron Island, (diamond) Gladstone, (triangle) Townsville and (pentagon) Cairns.

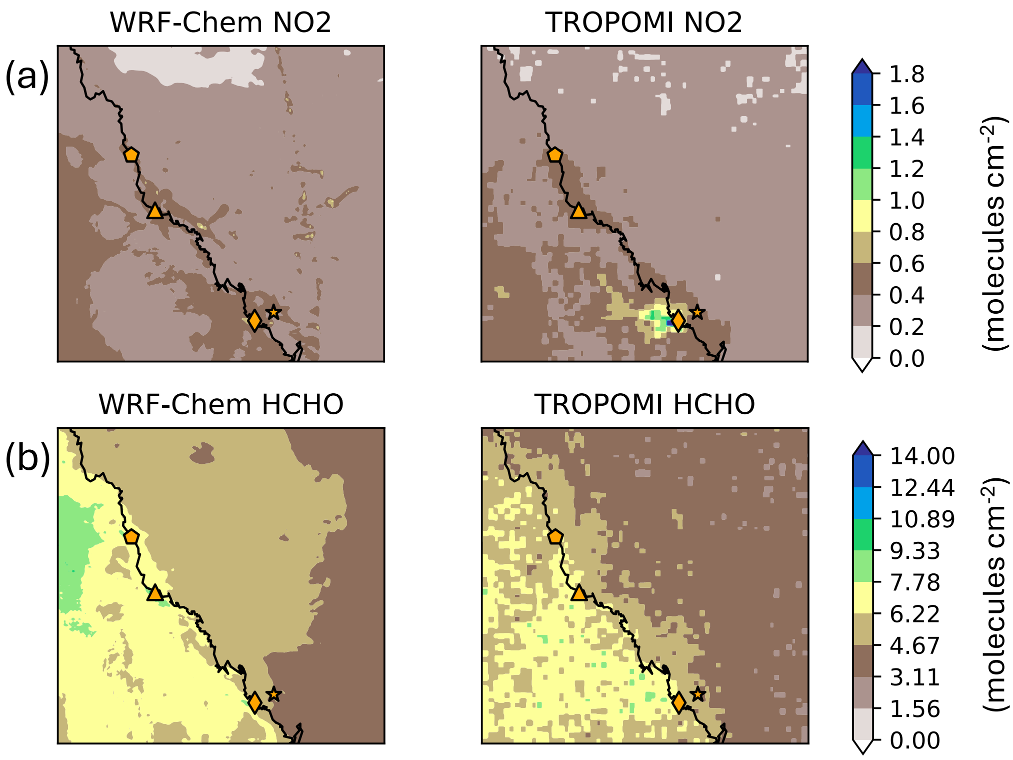

Fig. 5 Comparison of WRF-Chem trace gas tropospheric columns to TROPOMI, for (a) NO_2 and (b) HCHO. Location markers are the same as in Figure 4.

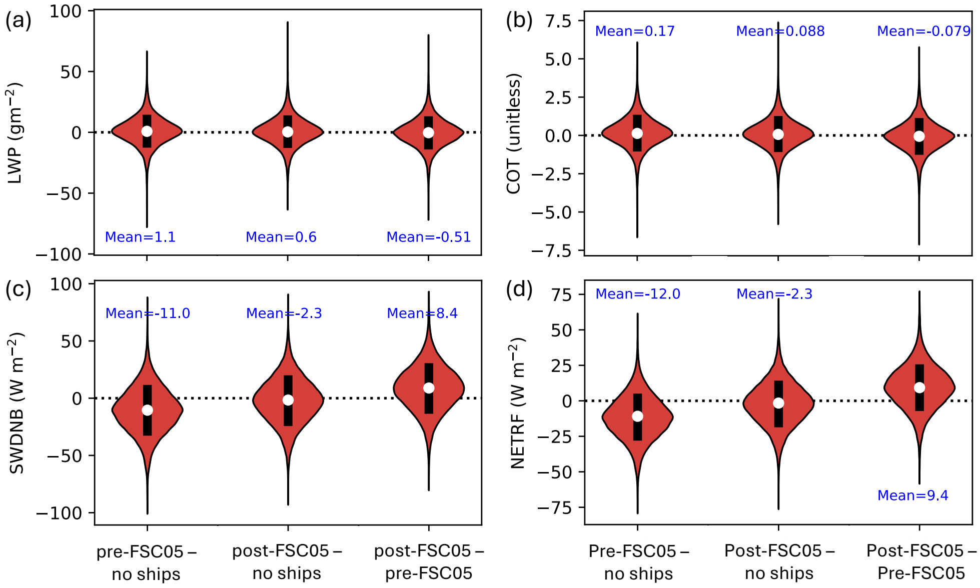

Fig. 6 Summary of daytime cloud and radiation changes over the whole oceanic part of the domain, under different ship emission scenarios: for (a) liquid water path, (b) cloud optical depth, (c) shortwave downwards radiation at the surface and (d) net radiative forcing at the top of the atmosphere. In each panel, simulation scenario results are (left to right) pre-FSC05 regulation minus no ships, post-FSCO5 regulation minus no ships and post- minus pre-FSC05 regulation. The shape of each violin plot indicates the frequency distribution of all results across the Coral Sea part of the domain. The black bars indicate the 90 % confidence interval, the white dots show the median value, and the means are annotated in blue text.

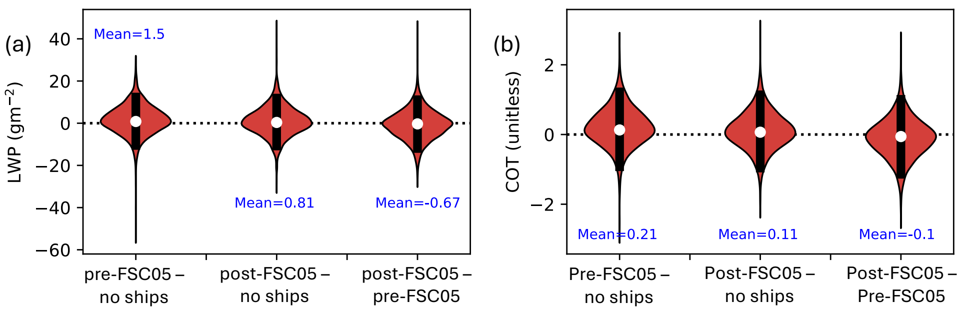

Fig. 7 Summary of daytime cloud changes over the GBR part of the simulation domain, under different ship emission scenarios: for (a) liquid water path and (b) cloud optical depth. In each panel, simulation scenario results are (left to right) pre-FSC05 regulation minus no ships, post-FSCO5 regulation minus no ships and post- minus pre-FSC05 regulation. The shape of each violin plot indicates the frequency distribution of all results across the Coral Sea part of the domain. The black bars indicate the 90 % confidence interval, the white dots show the median value, and the means are annotated in blue text.