



Tracing vessel-generated sea spray aerosols to support marine cloud brightening

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Abstract. The aim of Marine Cloud Brightening (MCB) is to purposely inject artificially generated sea spray aerosols (SSA_a) into low-level marine clouds, increasing their albedo with the objective of cooling the ocean surface beneath the clouds. Over the Great Barrier Reef (GBR), Australia, MCB is under research as a potential intervention to alleviate coral bleaching. Attributing cloud microphysical perturbations to MCB requires the ability to detect and trace SSA_a and distinguish that from other local sources, including the exhaust from research aircraft, research vessel, and generators used to power the MCB technology. Here, we present a novel “tracer” method using in situ airborne measurements of the non-volatile aerosol ratio to identify the aerosol plume emanating from the spraying vessel and apportion aerosols between SSA_a and combustion exhaust. A total of 14 flights were analysed using downwind, upwind, crosswind, or a combination of these sampling legs. Aircraft-sampled plumes of SSA_a and exhaust exhibited contrasting non-volatile properties. Aerosol plume detections were identified using aerosol number concentration, and a mixing ratio model was constructed to apportion the detected plumes between SSA_a and exhaust particulates. The “tracer” method reliably allowed tracking of the vessel-generated SSA_a across all sampling events as the plume spread downwind from the spraying vessel. The majority of sampled plumes were found to be mixed SSA_a and exhaust. This technique enables the tracking and identification of MCB aerosol in the natural marine atmosphere without the addition of a chemical tracer. Reliable plume identification is a prerequisite for studying the transport and dispersion of MCB aerosols and determining how they influence aerosols and clouds in the marine boundary layer.

Keywords: Marine Cloud Brightening (MCB), Non-volatile Aerosols, Airborne Measurements, Artificial Sea Spray Aerosols (SSA_a), Standard Deviation (SD) Method

1 Introduction

Climate change has caused an increase in sea surface temperatures, leading to more severe and frequent coral bleaching events (Cai et al., 2024; Johnson et al., 2022). Contemporary mass coral bleaching events have been recorded throughout the Great Barrier Reef in 2016, 2017, 2020, 2022, 2024 (GBR, Henley et al., 2024), and 2025 (Great Barrier Reef Marine Park Authority



et al., 2025). Anthropogenic climate change is predicted to continue, and unless mitigated, the threat of coral bleaching will increase (IPCC, 2022). To buffer this threat, solar radiation management methods have been designed to deliberately reduce the sunlight reaching the Earth's surface and have been proposed as a potential way to mitigate global warming (Biermann et al., 2022). One such method that has been proposed is marine cloud brightening (MCB, Latham, 1990; Latham et al., 2012). The idea of MCB is to purposely inject artificially generated aerosols, generally presumed to be sea salt (Claudel et al., 2024; Diamond et al., 2022), into the marine low-level clouds (Feingold et al., 2024; Haywood et al., 2023; Latham et al., 2012). The addition of artificially generated sea spray aerosols (SSA_a) is expected to increase cloud droplet number concentrations, increasing the cloud albedo (Latham et al., 2012), and leading to a negative radiative forcing that could counteract global warming (Ahlm et al., 2017).

Theoretical and modelling studies have emphasised the potential cooling effect from MCB based on the estimation of shortwave radiation reduction (e.g., Wood, 2021; M. Zhao et al., 2021; Chun et al., 2023). Over the GBR, pilot experiments on MCB have been conducted within a broader effort to investigate whether regional-scale MCB could help alleviate coral bleaching (Harrison, 2024). The MCB strategy can be achieved through vessel-mounted sprayers that produce and spray plumes of SSA_a into the marine boundary layer (Salter et al., 2008). As Latham et al. (2012) proposed, the initial phase of the outdoor MCB field experiment should focus on the detection and dispersion of vessel-generated SSA_a. Past experiments on hygroscopic seeding of warm clouds have investigated adding a tracer gas alongside sea salt particles to trace and identify the seeded clouds, such as black carbon (Russell et al., 2013) and sulphur hexafluoride (SF₆, Rosenfeld et al., 2010). However, black carbon was found to be ineffective, as its concentration showed no significant difference from the background level (PEACE, Russell et al., 2013), and SF₆, which exhibits a much higher global warming potential compared to carbon dioxide (Simmonds et al., 2020), requires vast emissions to trace ship generated MCB plumes over tens of km distance. Further, recent results over the GBR demonstrated that aerosol sources can shift between polluted continental air and clean remote marine air masses (Hernandez-Jaramillo et al., 2024) underscoring the need for a tracer that remains robust regardless of aerosol source. Therefore, an environmentally acceptable and effective “tracer” method is required to positively identify the MCB aerosols and MCB aerosol impacted air parcels to distinguish those air parcels from those that are impacted by background aerosols, ship or aircraft exhaust, or other local sources of pollution (Feingold et al., 2024; Latham et al., 2012).

Natural sea spray aerosols (SSA) are low-volatility and highly hygroscopic aerosols (Russell et al., 2023; Thomas et al., 2022) with complicated chemical compositions due to the source, including processes like debromination (Zhu et al., 2019) and chloride depletion (Xu et al., 2021). Rasmussen et al. (2017) used a thermodenuder to heat real and artificial seawater from 0 to 400 °C and measured the volume fraction remaining. Salt compositions, including NaCl, Na₂SO₄, and CaCl₂, still remain at 400 °C, because they have high melting and boiling points (Rasmussen et al., 2017). Inorganic compounds such as potassium and magnesium still persist at 300 °C (Poulain et al., 2014). Huffman et al. (2009) characterised the volatilities of different chemical species of aerosols, such that at a temperature of 230 °C, almost all ammonium, NH₂⁺, and NH₃⁺ were removed. Results from Clarke (1991) indicated sulfuric acid is volatilised by around 150 °C, while Johnson et al. (2004) refined this temperature to 139 °C, and determined that ammonium sulphate and ammonium bisulphate were completely volatilised at 202



°C and 205 °C, respectively. At a temperature of 280 °C, the volume fraction remaining of both artificial and natural SSA is approximately 90%, indicating that refractory inorganic salts are the dominant composition in SSA (Cravigan et al., 2015; Modini et al., 2010). While SSA contains an organic fraction, it remains a minor component compared to inorganic salts (Bates et al., 2012; Russell et al., 2023). Further, although some marine organics are semivolatile (evaporating by 250 °C), a significant portion is non-volatile, such as exopolymer gels, and can be thermally stable up to 550 °C (Mallet et al., 2016; Modini et al., 2010; Quinn et al., 2015). Therefore, a thermodenuder can be integrated with the aerosol measurement instrument to remove low-volatility aerosols (such as Cravigan et al., 2015; Wehner et al., 2002). Because marine aerosols consist primarily of organics, sulphates, and sea salts (Russell et al., 2023), thermal volatility can serve as a critical tool in characterising the mixing state of natural SSA. In the context of MCB, it is possible to separate MCB SSA_a from other aerosols due to its high thermal stability.

In the remote marine environment, including over the GBR, there are few significant local point sources of aerosols other than the exhaust from passing vessels, diesel-fuelled electricity generation on some small islands, and the research aircraft, sampling vessel, and MCB spraying vessel. Other present background sources of aerosol are generally well mixed within the marine boundary layer and can be subtracted, such that only plumes of enhanced aerosol concentration are used for analysis. In this work, we seek to test the hypothesis that MCB SSA_a has a sufficiently distinct volatility signature from other local sources and thus we will explore using the aerosol number concentration volatility ratio as a “tracer” of MCB-sprayed SSA_a.

2 Airborne data

2.1 Aircraft measurement

The field campaign was conducted in 2024 near Gould Reef, Queensland, Australia (19.35°S/148.75°E), and the research aircraft was operated from nearby Hamilton Island Airport (20.37°S, 148.95°E). The research aircraft was a Cessna 337, and a detailed description of the aircraft and instrumentation on board can be found in Hernandez-Jaramillo et al. (2024). The primary data for this study were aerosol concentration measurements obtained using airborne mixing condensation particle counters (MCPCs, Models 1720 and 9403, Brechtel, USA). These instruments condense butanol vapour on the particles to grow into larger sizes that can be detected optically and counted. The data was collected at 1 Hz, with a detection size range of 0.007-2 µm. The coincidence uncertainty of aerosol number concentration can be up to ±8% (Quinn et al., 2024). Meteorological data were collected by an ARIM-200 probe (Aventech, USA) at 1 Hz, including temperature, relative humidity, wind speed, and wind direction. A forward-facing camera was mounted on the instrument pod to record the flight (PodCam, Milesight, China), hereafter referred to as the PodCam. The cloud droplet probe (CDP, Droplet Measurement Technologies, USA) measured cloud droplet number concentration and was used to determine that the aircraft was outside of clouds, using the threshold of 10 cm⁻³, following Dong et al. (2022).



2.2 Measurements of non-volatile aerosol ratio

The non-volatile aerosol ratio was determined as the difference in aerosol number concentrations measured using two MCPCs. Both MCPCs drew air from a forward-facing aerosol inlet with total flow controlled by a microcomputer to maintain close to isokinetic flow during level sampling legs (see Hernandez-Jaramillo et al. (2024) for further information on the aircraft aerosol sampling inlet system). One MCPC sampled the ambient air, while the other MCPC was fitted after a thermodenuder (Flexotherm Industrial Heated Sample Line, USA), which heated the inlet to 300 °C. Both MCPCs were calibrated against each other for each flight using a period of data collection prior to the activation and heating of the thermodenuder (see Supplement S1). The thermodenuded MCPC measurement was taken as the number concentration of non-volatile aerosols ($N_{a,nonv}$), while the unheated MCPC measured the total number concentration of aerosols ($N_{a,tot}$). The ratio between non-volatile aerosol number concentration and the total number concentration is identified as the non-volatile ratio of aerosols (R_{nonv}). Aircraft passage through a “plume” of SSA_a was identified by a significant increase in number concentration above the background aerosol concentration at the same altitude. Periods of background aerosol and plume-induced aerosol were separated for each sampling leg, and the background aerosol number concentration was subtracted prior to calculating the ratio between non-volatile and total aerosol number concentrations. These instances of plume encounters were identified systematically by the standard deviation method described below. To characterise the nature of the MCB plume, this study uses the non-volatile aerosol ratio, calculated after subtracting the background aerosols ($R_{nonv-bk}$, equation 1). Unless stated otherwise, the non-volatile aerosol ratio shown in the following sections is calculated after subtracting the background aerosol concentration from both the measured total aerosol concentration and the measured non-volatile concentration. The non-volatile aerosol ratio will act as the “tracer” of the vessel-generated SSA_a. In this study, SSA_a refers to artificial sea spray aerosols generated by the MCB sprayers, whereas SSA refers to natural sea spray aerosols.

$$R_{nonv-bk} = \frac{N_{a,nonv} - N_{a,nonvbk}}{N_{a,tot} - N_{a,totbk}} \quad (1)$$

2.3 Aircraft sampling of vessel-generated plume

For this study, we isolated flight segments in which the research aircraft flew near the vessel at low altitude, targeting measurement of the vessel-generated SSA_a plume. Sampling was conducted using downwind, upwind, crosswind, or a combination of these approaches relative to the dispersion of the plume. Based on the aircraft altitude, aircraft location relative to the vessel, and PodCam, we could determine whether the aircraft was within or outside the visible SSA_a plume. Multiple plume encounters could occur during a single flight, depending on the sampling approach, plume dispersion, and vessel operation. Representative segments were selected and are shown in Table 1. Downwind, upwind, and crosswind refer to flying along the track of the plume in the same direction as the wind, opposite, or perpendicular, respectively. A total of 14 flights were analysed for plume sampling, including 12 flights in which the vessel was actively producing SSA_a and 2 control flights in which sprayers were operating but not producing SSA_a. Fogging refers to an alternative approach to generating SSA_a fog



(Hernandez-Jaramillo et al., 2024), while ship exhaust control was conducted when the vessel was not operating with SSA_a spraying.

Table 1 Summary of plume and exhaust sampling in the 2024 field campaign

Flight ID	MCB Operation	Plume/Exhaust	Plume/Exhaust
Date_Flight No.		Sampling	Sampling Method – Altitude
		Plume	Crosswind – 150 m
20240206	MCB	Aircraft Exhaust	Turning – 650 m
		Ship Exhaust	Crosswind – 150 m
20240207_F1	Fogging	Plume & Exhaust	Crosswind – 150 m
20240208_F1	Fogging	Plume & Exhaust	Crosswind – 150 m
20240208_F2	Fogging	Plume	Upwind – 150 m
20240209_F1	MCB	Plume	Downwind – 150 m
20240209_F2	MCB	Plume	Downwind – 150 m
20240210	MCB	Plume	Downwind – 175 m
20240212_F1	Fogging	Plume	Downwind then Upwind – 75 m
20240213_F1	Fogging	Plume	Downwind – 75 m
			Crosswind – 375 m
20240215_F1	Fogging	Ship Exhaust	Upwind then Downwind – 150 m
		Plume	Crosswind – 150 m
20240221_F1	MCB	Plume & Exhaust	Downwind then Upwind – 75 m
			Crosswind – 600 m
20240223_F1	MCB	Plume & Exhaust	Downwind then Upwind – 75 m
			Downwind – 75 ~ 450 m
20240225	Ship Exhaust Control	Ship Exhaust	Downwind then Upwind – 75 m
20240226_F2	Ship Exhaust Control	Ship Exhaust	Downwind then Upwind – 75 m

130 3 Standard deviation method

The setup of two MCPCs provides insight into the non-volatile fraction of SSA_a, but the sampled air contains both the background aerosols and vessel-sprayed SSA_a. A standard deviation (SD) method was used to identify plume encounters with sprayed SSA_a aerosols, defined as periods during which aerosol number concentration increases substantially above the well-mixed background in the marine boundary layer. The SD method has been shown to be an efficient automatic method to detect spikes in a time-series aerosol dataset (El Yazidi et al., 2018). In this study, we apply it to a time series of aircraft sampled aerosol data. This method simply adopts two factors that can automatically detect the spikes, the standard deviation of the selected dataset σ and a threshold parameter ∂ . The σ was calculated based on a selected range of the entire dataset. Due to different background aerosol levels, for each flight, this range was chosen differently to ensure that the SD method can



effectively flag the “spikes” of aerosol number concentration. In this way, the standard deviation can represent the smooth
variation of the background aerosol level. The spikes are defined by data values higher than a threshold, defined as C_{unf} plus
an additive value: $\partial \times \sigma + \sqrt{n} \times \sigma$, where ∂ is a parameter to control the selection threshold, and n is the number of points
between C_{unf} and C_i (equation 2). In this study, a “spike” is defined as, for each flight, whenever the non-volatile or total
aerosol number concentration was flagged by the SD method. A sensitivity test of selected combinations of σ and ∂ was
performed, and the result showed that the choice of data range used to calculate σ strongly influences the number of spikes
flagged by the SD method (see Supplement S2). After applying the SD method to each flight, the time series is separated into
background aerosols at different altitudes and spikes that identify aircraft passage through a plume with a higher aerosol
number concentration. Based on the SD method's sensitivity test results for each flight, the threshold parameter ∂ was set to 1
for consistency, while the dataset percentile range used to calculate σ was adjusted for each flight (see Table S3).

$$C_i > C_{unf} + \partial \times \sigma + \sqrt{n} \times \sigma \quad (2)$$

4 Constructing the mixing model of non-volatile aerosol ratios

End-member mixing analysis is often used to quantify the contributions of different sources using chemical or isotopic tracers
(e.g., Barthold et al., 2011; Sanders et al., 2014). The known tracer parameter values serve as input to a mixing model to
determine the relative contribution of the tracers in the measured sample. A simple mixing model typically includes two sources
(Kirchner, 2023). The proportion of the identified sources collectively sums to 1, allowing the calculation of the fractional
contribution from each source. Samples to identify the signature of the MCB spraying vessel diesel generator exhaust were
collected during low altitude passes downwind of, or overhead of the vessel, during periods when the vessel was acting as a
‘control’. During these periods, the energy intensive components of the MCB plant were operated (air compressors and high-
pressure water pumps), but the plant was not producing SSA_a. Ancillary equipment on the vessel, such as air conditioning
units, was also operated to make up the shortfall so that the diesel generators on the vessel were loaded to an equal level of
power output as during MCB spraying operations (Hernandez-Jaramillo et al., 2025). Aircraft exhaust signature samples were
collected by flying circling manoeuvres in clear air away from other aerosol sources. It was not possible to unequivocally
sample a pure SSA_a plume since producing the SSA_a required the generators to be producing exhaust. In practice, under low
wind and highly unstable atmospheric conditions, the SSA_a and exhaust plumes were observed to follow separate advection
trajectories and form separate plumes in close proximity to the vessel. This separation arose due to the difference in location
between the source of diesel generator exhaust (underwater midship) and the MCB SSA_a (at the stern, propelled aft by high-
power fans), along with the difference in buoyancy between the hot exhaust gases and evaporating seawater droplets. Because
pure MCB SSA_a could not be independently verified without relying on the tracer method itself, the SSA_a end-member was
instead defined based on the following theoretical considerations to avoid circularity.



Seawater composition can influence SSA volatility (Quinn et al., 2015; Rasmussen et al., 2017). At a temperature of 300°C, the volume fraction remaining of non-volatile seawater varied between 0.85 and 0.9 (Rasmussen et al., 2017). Organic matter could contribute to an amount of the volatile fraction up to 10% at temperatures between 230-250 °C (Mallet et al., 2016; Quinn et al., 2015). This would contribute to most of the volatile fraction of SSA. For artificial NaCl-only seawater, NaCl scarcely volatilises between 25-400 °C (Rasmussen et al., 2017). Under the simplifying assumption that the non-volatile component of sprayed SSA_a is dominated by NaCl, the theoretical non-volatile fraction of seawater is close to 1. In the dataset, we observed non-volatility ratios of up to ~1.2, which are theoretically impossible but can occur given known inaccuracies in MCPC-based measurements at high aerosol concentrations. We therefore adopted a threshold non-volatility ratio of 0.98, which is just below the theoretical maximum value. Thus, we selected a relatively small subset of samples with the highest measured volatility ratios as representing the SSA_a plume end-member (Fig. 1).

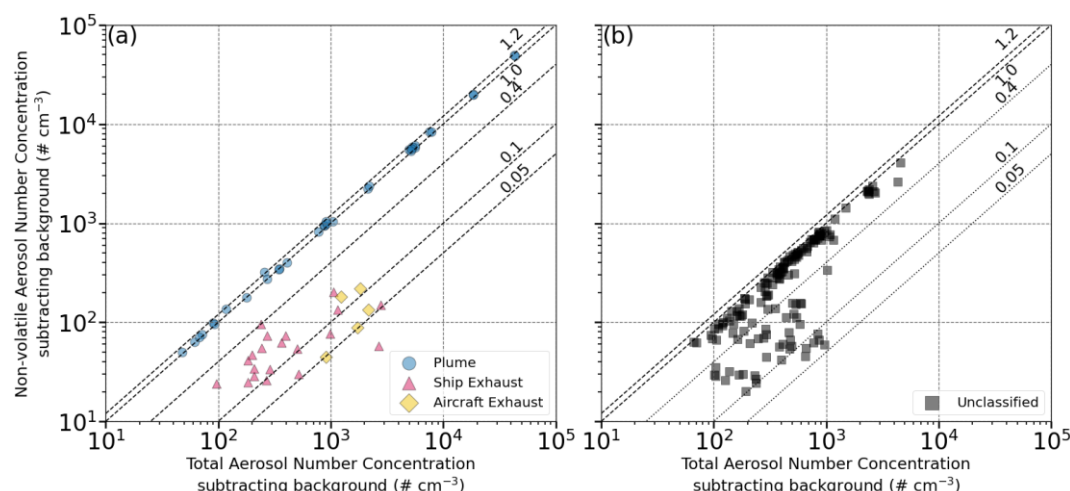


Figure 1 Scatter plot of total aerosol number concentration and non-volatile aerosol number concentration, both subtracting the background aerosols determined prior to the sampling, for (a) identified plume, ship exhaust, and aircraft exhaust, and (b) unclassified encounters during the sampling in the 2024 field MCB experiment

5 Results

5.1 Mixing ratio model of non-volatile aerosol ratio

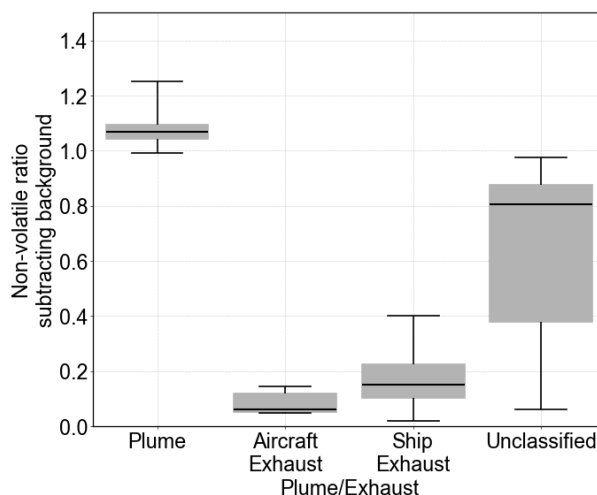
After removing the background aerosols, the non-volatile aerosol ratio increased for identified plume sampling but decreased during exhaust sampling. Across all isolated flight segments as detailed in section 2.3, only peak number concentrations of both total number concentration and non-volatile aerosol number concentration were selected during the plume and exhaust sampling periods. During the exhaust sampling, peak non-volatile number concentrations were often less distinct than total aerosol number concentrations, as a result of a much lower non-volatile aerosol ratio. When the aircraft flew to sample the vessel-sprayed SSA_a plume, it was expected that the total and non-volatile aerosol number concentration would be almost



identical (Fig. 1a). However, during plume sampling, it was found that the aircraft frequently captured exhaust emissions from both the spraying vessel and research aircraft. 2 out of 3 downwind-then-upwind samplings were found to capture exhausts, highlighting the importance of exhaust contamination and separating from SSA_a during future MCB campaigns. The non-volatile ratio for each identified aircraft encounter was calculated using Eq. 1, with background aerosols subtracted.

195 Considering the differences of MCPC number concentration up to $\pm 8\%$ (Quinn et al., 2024), the measured non-volatile aerosol ratios were all within the range. Although the non-volatile aerosol ratios varied across different vessel operations, the distribution of plume encounters and exhausts (ship and aircraft) exhibited distinct features (Fig. 2). Individual t-test results for plume vs. ship exhaust, plume vs. aircraft exhaust, and ship exhaust vs. aircraft exhaust were 1.1×10^{-26} , 5.6×10^{-9} , and 0.02, respectively, indicating a statistically significant difference between plume and exhaust. Because ship and aircraft exhaust

200 exhibited similar characteristics, they were jointly grouped as a single exhaust source for the mixing ratio model. Finally, vessel-generated SSA_a and exhaust emissions are the two sources identified and used to determine the non-volatile aerosol ratio, after subtracting the background aerosols, as inputs to the mixing model. The 50th percentile values of the non-volatile aerosol ratio are chosen as input to the mixing ratio model, with the plume encounter and exhaust ratios of 1.07 and 0.135, respectively.



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Figure 2 Boxplots of non-volatile aerosol ratios, subtracting the background aerosols, for identified plume encounters, identified aircraft exhaust, identified ship exhaust, and unclassified samplings

5.2 Tracing vessel-generated plume

The central question remains whether the non-volatile measurement can act as the “tracer” of the vessel-generated SSA_a plume.

210 Two examples are presented to demonstrate that the plume was successfully detected and traced downwind of the vessel as it rose to cloud base. On February 21st, aircraft sampled the SSA_a plume at 600 m, where the plume was well dispersed both downwind and vertically as shown in the PodCam video (Fig. 3a and 3e). The aircraft remained outside the clouds while capturing the SSA_a plume, and crosswind sampling was adopted. While the aircraft sampled the SSA_a plume, the spraying



vessel moved with the wind near the surface, which could explain why the vessel-generated SSA_a plume was located at 600 m elevation to the northwest of the ship. On February 23rd, the aircraft first flew overhead the vessel and sampled the MCB plume on a downwind leg at 75 m altitude (1st passage, Fig. 3b). After reaching the end of the dispersed plume and turning around, the aircraft then flew an upwind leg to resample the plume again at the same altitude (2nd passage, Fig. 3c). During the second sampling, the mixing ratio model indicated that the detected SSA_a plume was partially mixed with exhaust from the initial sampling. Although ship exhaust could mix into the SSA_a plume, as aircraft approached closer to the spraying vessel, mixing ratio model results showed the plume contained almost no exhaust. Further downwind, however, the plume contained an increasing contribution from exhaust, likely to be residual aircraft exhaust from the earlier sampling transect. As the SSA_a plume dispersed downwind away from the spraying vessel, the total aerosol number concentration decreased due to plume spreading and dilution. After another turn at 150 m, the aircraft ascended along the downwind plume from 75 to 450 m elevation (3rd passage, Fig. 3d and 3g). The mixing ratio model also indicates that the SSA_a plume is mixed with exhaust, which can be strongly influenced by the aircraft's own exhaust from the previous downwind and upwind sampling passages. The captured plume at heights of 75-450 m was also to the northwest of the vessel, above the previous two sampling paths at 75m. This also proved that vessel-generated SSA_a can travel upwards. The vertical transport and dispersion of the plume travelled along the wind direction (Fig. 3g). Moreover, the total aerosol number concentration decreased as the aircraft ascended and was much lower than the first sample near the vessel at 75 m. Standard temperature and pressure corrections for aerosol number concentration were not applied because the change in altitude (75-450 m) was relatively small compared with the much larger decrease in aerosol number concentration.

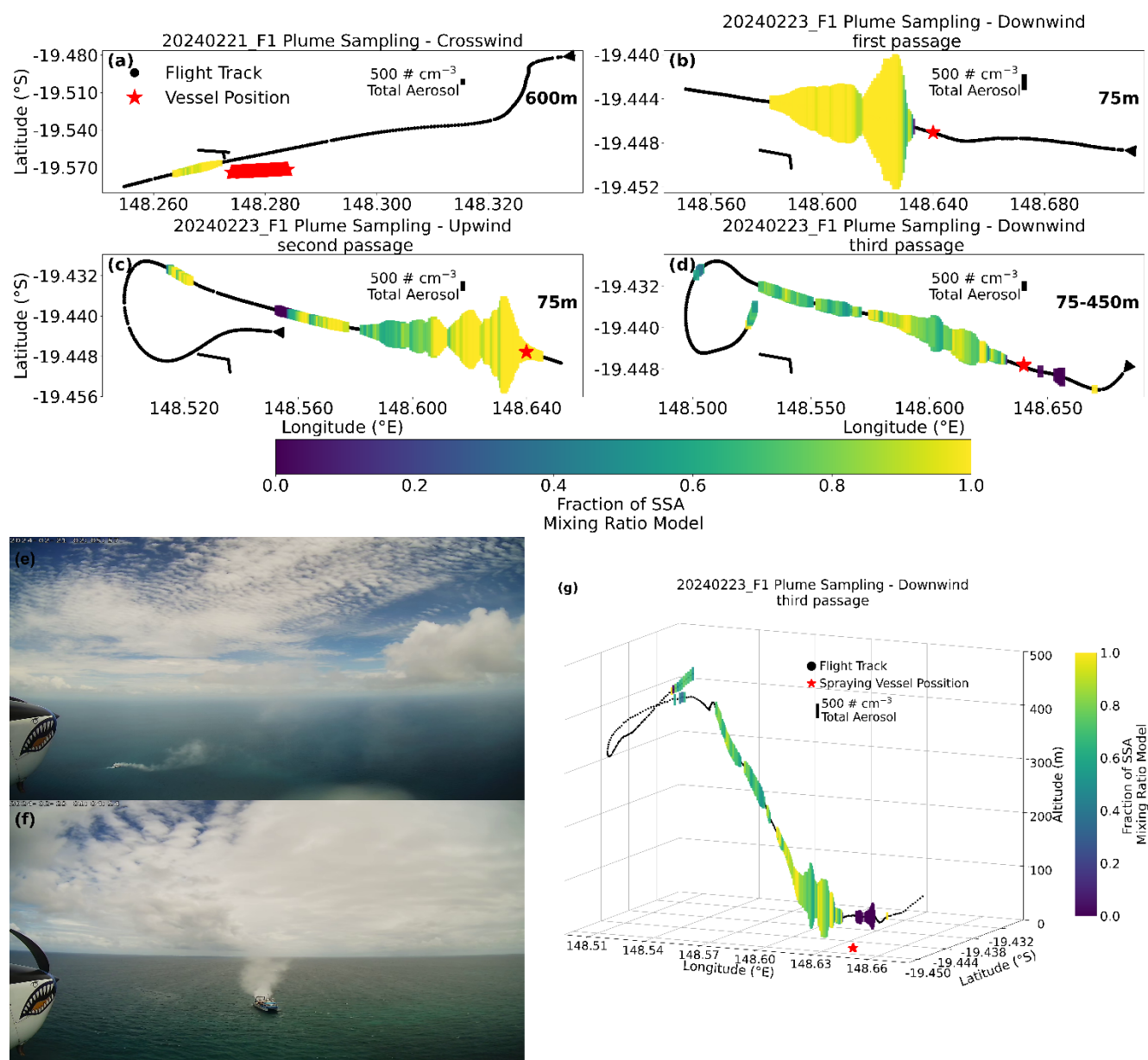


Figure 3 Mixing ratio model results of detection of vessel-generated SSA_a plume downwind of the vessel at different heights, where (a) is from February 21st, when the aircraft captured the SSA_a at 600 m below the cloud base; (b) and (c) are from February 23rd, where the aircraft sampled the SSA_a plume at 75 m from 1st downwind passage and subsequent upward 2nd passage, respectively; (d) is from February 23rd, where the aircraft ascended from 75 m to 450 m to capture the SSA_a plume dispersing upwards from the spraying source; (e) and (f) are from the PodCam video, the aircraft was approaching the dispersed SSA_a plume on the 21st and 23rd, February; and (g) is the from February 23rd, where the aircraft captured the SSA_a from 75 to 450 m, same sampling period as in (d). The red star indicates the location of the spraying vessel, and the black dots show the



240 actual research aircraft track. The colour scale shows the fraction of SSA_a , determined by the mixing ratio model, where the length of each coloured segment corresponds to the total aerosol number concentration. A vertical line at $500 \text{ \# cm}^{-3}$ is included as a reference in panels (a)–(d) and (g). The wind barb in (a)–(d) represents wind at 100 m, retrieved from the hourly ERA-5 reanalysis data.

5.3 Sampling vessel-generated plume

245 Despite the different strategies employed, the SD method consistently identified flagged “spikes” across all 14 flights, representing vessel-generated SSA_a plumes, that were sampled during the 2024 field campaign. Most of the “spikes” were due to the vessel-generated SSA_a , as determined by the mixing ratio model. On February 6th, most of the sampled aircraft exhausts were captured during aircraft turns (Fig. 4a). At these times, the aircraft was far away from the spraying vessel and the dispersed plume. The background aerosol number concentration on February 6th was high, which affected the spike flagging during the
250 sampling period. As a result, some spikes were not identified when the aircraft flew out of the plume. Further details of the spike-flagging limitation are provided in Supplement S2. The ship exhaust control on February 25th showed that ship exhaust can be detected at 150 m (Fig. 4b), and potentially mixed into the SSA_a plume sampling. The aircraft flew several legs that day, from overhead the vessel to the downwind end and back. However, most legs captured only a few exhaust-contributed data points, and the exhaust plume was not visible in the PodCam. Given that the aircraft repeatedly flew along the same leg
255 path, these samples were likely mixed with residual aircraft exhaust emissions from previous passes. On February 8th, the aircraft flew an upwind leg from the end of the downwind spread to sample the plume at 150 m (Fig. 4c). Prior to this upwind leg, the aircraft flew perpendicular to the plume spread, during which only a few plume data points were intercepted. Unlike on other days, the plume detected at 150 m was intermittent, and the total aerosol number concentration was lower than on other days, as shown in Fig. 4.

260 A total of five flights followed the sampling method used on February 12th: the 1st passage flew downwind from the vessel, and the 2nd passage flew upwind from the downwind end to capture the plume or ship exhaust (Table 1). On February 12th, the aircraft flew at an altitude of 75 m and first sampled the plume along the plume’s downwind spread (Fig. 4d). According to the mixing ratio model, the plane remained inside the plume; however, the SSA_a fraction was not consistently high near the vessel or further downwind. For many SD-flagged spikes, the mixing ratio model indicated that the sampled MCB plume
265 contained more than 20% exhaust. During the first sampling, several spikes were flagged before the aircraft turned toward the spraying vessel, and the fraction of SSA_a remained close to zero. The plume captured by subsequent sampling is most likely residual aircraft exhaust from the earlier downwind leg (Fig. 4e). As the aircraft flew closer to the spraying vessel, the newly sprayed plume remained at a high fraction of SSA_a , consistent with the 1st sampling.

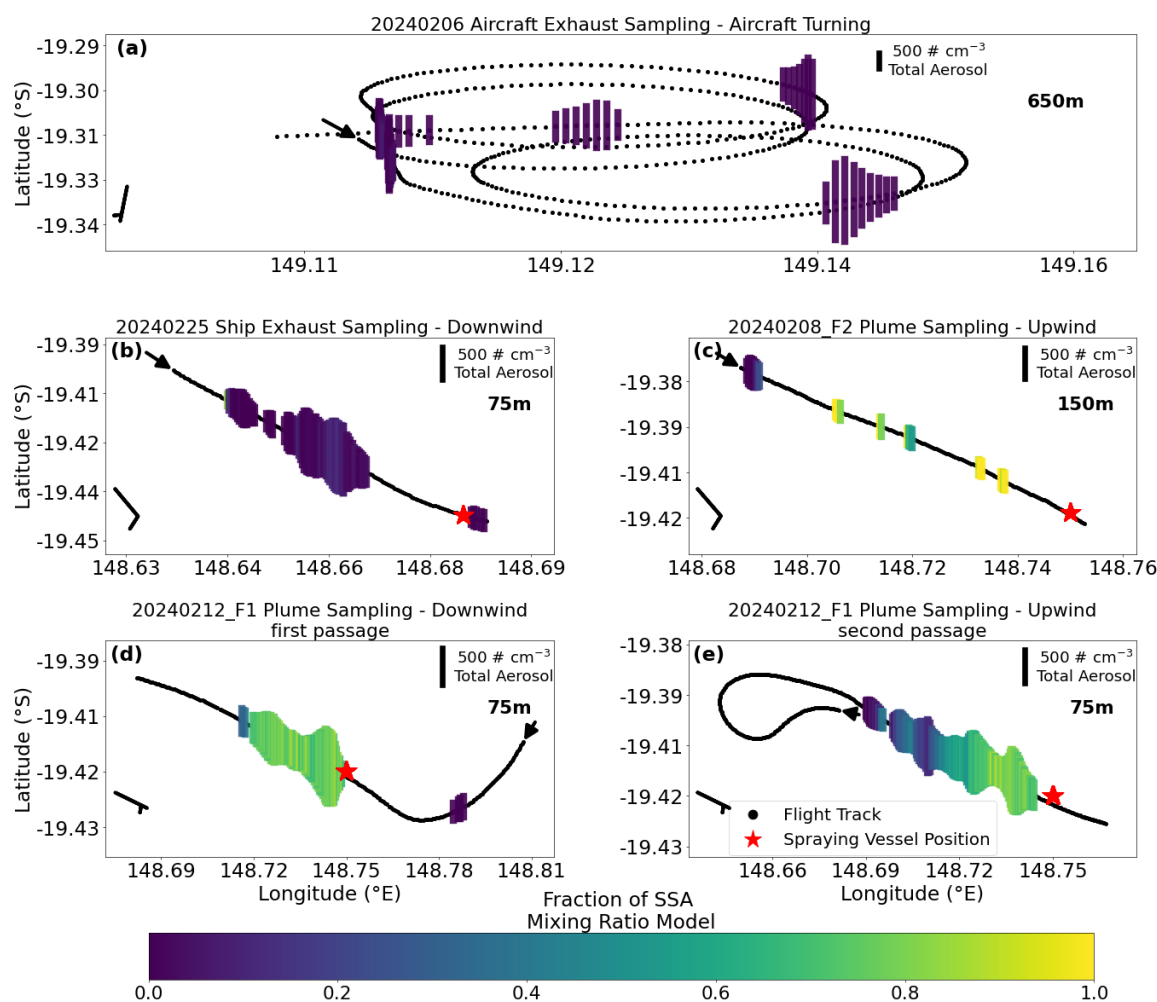


Figure 4 Mixing ratio model results of vessel-generated SSA_a fraction for selected examples for (a) aircraft exhaust sampling on February 6th; (b) downwind leg sampling of ship exhaust on February 25th; (c) upwind leg sampling of MCB plume on February 8th; (d) and (e) typical sampling method during the 2024 MCB field experiment, a first downwind passage followed by a second upwind passage on February 12th. Plotting conventions as in Fig. 3a–3d.

6 Discussions

This study demonstrated that the measurement of non-volatile aerosols can serve as the “tracer” for vessel-generated MCB plume. MCB-sprayed SSA_a was found to be mixed with emissions from the spraying vessel, thereby altering the volatile fraction of the sampled aerosols. The SD method effectively flagged the added SSA_a, separating them from background aerosols. However, the background aerosol level can affect the SD method's ability to flag spikes. A higher background aerosol number concentration may result in fewer spikes being flagged. The composition of marine aerosols can be different due to



long-distance transport (e.g., Ma et al., 2022; Roberts et al., 2006) or sulphur emissions (Quinn and Bates, 2011). Despite variability in background aerosol sources and concentrations, the proposed “tracer” method consistently identified vessel-sprayed SSA_a, demonstrating its reliability across various marine conditions. The mixing ratio model offered a straightforward fractional assessment of vessel-sprayed SSA_a, supporting tracing and detection of the MCB plume.

285 Results from the mixing ratio model showed that, for each sampling method, most aircraft-sampled MCB plumes were non-volatile SSA_a. Each sampling strategy demonstrated distinct strengths in capturing the plume. The crosswind sampling method was able to capture how the plume dissipated and eventually mixed with background aerosols, but only when the plume was already well dispersed and the aircraft flew precisely perpendicular to the axis of plume dispersion. Upwind and downwind sampling methods provided insights into how far the plume can spread downwind from the vessel, depending on the aircraft’s
290 sampling altitude. The upwind sampling method hardly captured the aircraft’s own exhaust if no prior sampling leg had been flown along the same path. Aircraft exhaust was highly associated with aircraft turning. When the aircraft circled the spraying vessel for crosswind sampling, it may also have captured its own exhaust from previous manoeuvres. A well-sampled plume for each vessel operation showed a relatively consistent non-volatile SSA_a fraction, unless the plume was mixed with ship or aircraft exhaust. The ship exhaust control sampling showed that such exhausts can be inevitable and might contain more
295 volatile aerosols, such as sulphate (Coggon et al., 2012). The size of MCB SSA_a is crucial for MCB to be effective, as SSA_a can easily act as CCN, whereas most studies have focused on this (such as Partanen et al., 2012). Below the cloud base, aerosols larger than 60nm can act as CCN under natural conditions over the GBR (Braga et al., 2025). Mixing with aerosols from ship exhausts could alter the size distribution of SSA_a through coagulation or even affect their hygroscopicity and their ability to act as CCN (Croft et al., 2021; O’Brien et al., 2015). The ship exhausts might not always be emitted into the MCB-sprayed
300 SSA_a, though separating those aerosols during MCB operations should be kept in mind. This will help assess the impact of MCB by determining how added SSA_a will finally alter radiative forcing, which could enhance the model’s accuracy in representing these parameters as discussed by McFiggans et al. (2006), Fan et al. (2016), and Rosenfeld et al. (2023).

The vertical dispersion of the MCB plume is closely linked to the turbulent mixing. Dhandapani et al. (2025) highlighted the importance of turbulent mixing in helping transport the MCB plume to reach the clouds. However, turbulence is often irregular
305 at small scales (Caldwell and Mourn, 1995), which could lead to intermittent dispersion of MCB SSA_a, as shown in Fig. 4c, and to fluctuating changes in the aerosol number concentration downwind of the spraying vessel (e.g., Fig. 3c and 4e). This naturally raises concerns about whether the MCB plume can be continuously uplifted to reach the cloud base. Although observations suggested that evaporative cooling from the vessel-sprayed SSA_a won’t significantly affect any uplift (Hernandez-Jaramillo et al., 2023), further investigation is still required to determine whether the underlying mechanism can
310 facilitate turbulent mixing and updraft motion, leading to improving the effectiveness of MCB.

Aerosols can be modified within clouds through microphysical processes such as activation, coalescence and scavenging, and evaporation (Fossum et al., 2020; IPCC, 2014; Zheng et al., 2024), as well as through aqueous-phase chemical reactions (IPCC, 2014; Tilgner et al., 2021). These processes alter the physical and chemical properties of aerosols, including but not limited to changes in size, morphology, composition, and hygroscopicity (Hudson and Noble, 2024; Liu et al., 2021; Zhang et al., 2024).



315 Cloud processing can modify volatile organic compounds and acids into low-volatility salts in the aqueous phase, and enhance
the hygroscopicity of processed aerosols with the addition of highly soluble sulphate and oxalate salts (Ervens et al., 2018;
Tilgner et al., 2021; Zhang et al., 2024). Above the cloud base, unactivated aerosols can also undergo secondary activation
related to the supersaturation condition (Efraim et al., 2024). Past studies have shown that the measurement of aerosols is the
overall result of all the above-mentioned processes, making it difficult to distinguish between primary emissions and secondary
320 mass addition (Fossum et al., 2020; Mayer et al., 2020; Zhang et al., 2024). In a natural marine environment, SSA is often
found as a complex internal mixture, aggregated with other aerosols, such as sulphate and organic matter (Fossum et al., 2020;
Mayer et al., 2020). Hence, challenges remain in measuring and separating the mixing processes of aerosols. When evaluating
the efficiency of MCB, jointly considering aerosol composition and the background cloud environment helps distinguish SSA_a
introduced by MCB from aerosols released through previously cloud processing. Our future work will focus on investigating
325 the cloud responses to MCB-sprayed SSA_a under varying aerosol conditions.

The proposed “tracer” method in this study was examined for detecting and tracing vessel-sprayed MCB SSA_a within the
boundary layer. In-cloud measurement of aerosols remains challenging because aircraft disturb the ambient environment and
sampling inlet systems inevitably misrepresent the true in-cloud aerosol population (Wendisch et al., 2004). The shattering of
cloud droplets caused by aerodynamic breakup or impaction with the aerosol inlet results in a large number of spurious “shatter
330 artifact” particles (Craig et al., 2013; Spanu et al., 2020). This process contaminates the aerosol samples and makes the analysis
of interstitial particle population nearly impossible (Craig et al., 2013). Consequently, the shattering leads to an overestimation
of measured aerosols, potentially overestimating the aerosol indirect effects on clouds, especially the Twomey and Albrecht
effect. Past studies have emphasised that both the instrumental factors and aerosol chemical composition can bias the aerosol-
cloud analyses (Hu et al., 2020; Schlosser et al., 2025). These uncertainties highlight the critical importance of closure
335 investigations to determine the precise fraction of aerosols activated into cloud droplets, a necessary step for accurately
assessing aerosol cloud interactions and constraining global climate model estimates.

7 Conclusions

A new “tracer” method was developed to identify the SSA_a fraction in aircraft-sampled, vessel-sprayed MCB plume over the
GBR in February 2024. This method was validated under various aerosol conditions and demonstrated consistent reliability.
340 Combining the aerosol number concentration spike detection (SD method) with a mixing ratio model successfully attributed
the vessel-sprayed MCB plume from sprayed SSA_a and from exhaust emissions contributions. Measurements of non-volatile
aerosol ratios at 300 °C showed that, for a subset of the data, the background-subtracted number concentrations of non-volatile
aerosols and total aerosols were nearly identical. This result suggests that the sprayed SSA_a could consist predominantly, or
even entirely, of non-volatile sea salts, in contrast to the volatile aerosols emitted from ship exhaust. However, the composition
345 of sprayed SSA_a can significantly alter its non-volatile properties. When incorporated into the mixing ratio model, accurate
knowledge of the actual sprayed SSA_a composition would help define a more precise boundary definition for sprayed SSA_a



based on the exact non-volatile aerosol ratio. Therefore, continuous monitoring of sprayed SSA_a non-volatile aerosol ratios is recommended to ensure accurate end-member representation and effective tracing in MCB operations. The proposed tracer method outlined here can provide a broadly applicable tool for studying solar radiation management methods and their impacts on aerosol cloud interactions in diverse marine environments, including but not limited to the GBR.

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