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# Marine aerosol in Aotearoa New Zealand: implications for air quality, climate change and public health

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#### ABSTRACT

Particulates emitted from the ocean's surface such as sea salt and byproducts of marine biogenic activity form atmospheric aerosols. Aerosols are important for climate change because they have offset some of the historical warming caused by greenhouse gases. Aerosols are also significant for human health: they are small enough to be inhaled and contribute to respiratory problems and other illnesses. Marine aerosol is the primary source of natural aerosol present in urban areas of Aotearoa New Zealand and, as part of the natural aerosol background, cannot be managed. Here, we review the production and presence of marine aerosols in New Zealand's air, and the implications for human health and climate change. Because marine aerosol is sensitive to physical changes in climate such as sea surface temperature and winds, production is likely to be affected by climate change. Overall, marine aerosol is unlikely to become a smaller contributor to urban atmospheric aerosol loading in New Zealand towns and cities under future climate change scenarios. Continued assessment of anthropogenic aerosols will be necessary to ensure that air quality targets are met.

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# Introduction

More than half of New Zealand's population lives within 5 km of the coast. This population is exposed to typical urban air quality issues from traffic-related air pollution, biomass burning for home heating and industrial emissions. Unlike many other countries, New Zealand's urban air quality is heavily influenced by the ocean. The haze that is sometimes visible when viewing the ocean is a result of tiny particulates entering the atmosphere when waves break and bubbles burst. These particulates–also

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known as aerosols-are amalgamations of solid, liquid or mixed-phase particles suspended in air (Colbeck 1998; Baron and Willeke 2001).

Aerosols mostly consist of sulfates, nitrates, mineral dust, sea spray and carbonaceous aerosols (organic aerosol and black carbon). Their lifetimes range from minutes to weeks, meaning that they can be transported over hundreds to thousands of kilometres through the atmosphere. However, atmospheric transport and aerosol residence times can be complicated by synoptic conditions such as blocking highs (see e.g. Jiang et al. 2004, 2005 and references therein). In general, fine aerosol particles (those smaller than 2.5  $\mu$ m in diameter) remain in the atmosphere for longer than coarse particles (larger than 2.5  $\mu$ m; Seinfeld and Pandis 2006).

Aerosols are typically classified into different modes depending on their size (Seinfeld and Pandis 2006; Figure 1). The nucleation mode (spanning particles  $\approx 0.001-0.01 \,\mu$ m in diameter) contains very fine primary particles emitted directly from chemical or physical processes, or secondary particles that have nucleated from the gas phase. The Aitken mode ( $\approx 0.01-0.1 \,\mu$ m) contains agglomerates of nucleation mode particles that have coagulated and particles that have grown from the nucleation mode by condensation. Aerosols in the accumulation mode ( $\approx 0.1-1 \,\mu$ m) are subject to rainout and washout, but sediment out very slowly, unlike coarse mode particles ( $\approx 0.5-10 \,\mu$ m). Accumulation mode aerosols can be activated to cloud condensation nuclei (CCN; Tatzelt et al. 2022; Regayre et al. 2020; Section 'Climate impacts of marine aerosol') and take up water vapour to form cloud droplets. The coarse mode contains mostly primary particles from dust, anthropogenic emissions, sea spray, volcanoes and plants (Jacobson 2005).



**Figure 1.** Typical aerosol size distributions, showing the number of particles relative to their size. The peaks, from left to right, indicate the maxima in the nucleation, Aitken, accumulation, and coarse modes. The blue line shows the global average, and the black line shows the average over New Zealand (36–47°S, 167–178°E). Data source: atmosphere-only configuration of the UK Earth System Model (UKESM1-AMIP); Sellar et al. (2019).

While accumulation mode particles are more numerous than coarse particles and constitute the majority of CCN, coarse mode particles can also act as CCN.

From a health perspective, aerosols are classified as  $PM_{10}$  and  $PM_{2.5}$ , which correspond to particulate matter (PM) with an aerodynamic diameter less than 10  $\mu$ m and 2.5  $\mu$ m, respectively. Particles of these sizes can be inhaled and contribute to respiratory and cardiac illnesses. The World Health Organisation's (WHO) guideline levels for annual average concentrations of airborne PM are  $15 \,\mu$ g m<sup>-3</sup> for PM<sub>10</sub> and  $5 \,\mu$ g m<sup>-3</sup> for PM<sub>2.5</sub> (WHO 2021). In New Zealand, air quality targets are set through the National Environmental Standards (NES) for Air Quality and monitored by regional councils. The PM<sub>10</sub> guideline is  $50 \,\mu$ g m<sup>-3</sup> as a 24-hour mean; currently, there is no PM<sub>2.5</sub> guideline (Ministry for the Environment 2021).

As an annual mean, total aerosol concentrations over New Zealand are relatively low compared with other countries (Gui et al. 2021). In consequence, New Zealand urban areas generally experience good air quality: in the 2022 list of the world's most polluted countries, based on annual-mean PM<sub>2.5</sub> concentrations, New Zealand ranked 121 out of 131 (IQAir 2023). Other countries have higher populations and population densities than New Zealand, and correspondingly higher levels of anthropogenic activities. These produce primary aerosols such as industrial dust and soot from fossil fuels (Jacobson 2005). Secondary anthropogenic aerosol can be formed from sources including biomass burning, sulfur dioxide from power stations and coal burning, and nitrates from fossil fuel burning and agricultural fertilisation. While these aerosol sources are present in New Zealand, they are smaller than in many other countries. New Zealand therefore has a unique aerosol environment because of relatively low anthropogenic emissions, a long coastline and proximity to the Southern Ocean, which, as one of the windiest regions on the planet, is a significant producer of marine aerosol. Here we review the current state of knowledge regarding marine aerosol in New Zealand's air and its implications for health and climate change.

### Marine aerosol production

### Sea spray aerosol

Sea spray is the largest source of natural aerosols in the atmosphere (Saliba et al. 2019), and consists of sea salt and primary marine organic aerosol. Primary marine organic aerosol is an umbrella term for more than 10,000 organic compounds in the sea surface microlayer, such as phytoplankton, zooplankton and bacteria. Exudates from these organisms, including fats, proteins and carbohydrates, are converted through wave action into a marine gel. Breaking waves form underwater air bubbles which move to the surface. Air bubbles scavenge organic material from marine gel, as well as fragments of the bodies of organisms in the sea surface microlayer, and emit them into the atmosphere as aerosols (Ovadnevaite et al. 2011). Primary marine organic aerosol is relevant for climate change over New Zealand and the Southern Ocean due to its interactions with clouds (Section 'Climate impacts of marine aerosol'; Burrows et al. 2022; Gantt and Meskhidze 2013).

Sea salt aerosol (SSA) is ejected into the atmosphere when waves break and bubbles burst in a dynamical process that occurs when the wind speed is higher than  $\sim 5 \text{ m s}^{-1}$ 

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(Grythe et al. 2014). Such high wind speeds are common over the ocean surrounding New Zealand, with the strong westerly winds between 40–50°S known as the Roaring Forties. In general, higher wind speeds lead to larger SSA emissions (Gong 2003; Hartery et al. 2020). In addition to wind speed, some studies indicate that the sea surface temperature may also influence SSA generation (Jaeglé et al. 2011; Liu et al. 2021). However, the nature of its effect on SSA is still poorly constrained.

### Secondary sulfate aerosol

Secondary sulfate aerosol has multiple natural and anthropogenic sources, as discussed in Section 'Air particulate matter composition analysis and source apportionment in New Zealand'. An important natural source is the ocean. Marine phytoplankton, algae, corals and other biota produce dimethylsulfoniopropionate (DMSP), which undergoes bacterial breakdown to form dimethylsulfide (DMS; Groene 1995). An approximate indication of the DMS distribution in seawater can be obtained by analysing oceanic chlorophyll *a*, which can be detected by satellite instruments. High levels of chlorophyll *a* generally indicate high levels of marine biogenic activity, and therefore of oceanic DMS (Galí et al. 2018; Bhatti et al. 2023). As shown in Figure 2, chlorophyll concentrations around New Zealand are elevated in nutrient-rich areas such as coastal regions and the Chatham Rise off the east coast of the South Island. Large DMSP and DMS abundances are also associated with springtime breakup of Antarctic sea ice (Trevena and Jones 2006; Webb et al. 2019).

DMS is emitted into the atmosphere with a flux dependent on the seawater DMS concentration and wind speed (Bell et al. 2015; Blomquist et al. 2017). Once in the atmosphere, DMS undergoes a series of chemical reactions to produce  $SO_2$  and methane sulfonic acid (MSA).  $SO_2$  undergoes gas-to-particle reactions to produce secondary sulfate aerosol (Figure 3). MSA condenses on existing particles, therefore increasing their size (von Glasow and Crutzen 2004; Hoffmann et al. 2016).

DMS chemistry is highly complicated and the reactions involved are not fully understood (Cala et al. 2023; Revell et al. 2019). The reactions involved can take hours to days depending on the reaction pathway followed, temperature, relative humidity and the strength of solar radiation (Seinfeld and Pandis 2006). Therefore, concentrations of sulfur-containing particulate matter from secondary sulfate sources are likely to be highest some distance downwind of a precursor gas emission source (Polissar et al. 2001).

The seasonal cycle of oceanic DMS is driven by marine biogenic activity, which at southern high latitudes is influenced by the loss of sea ice cover (Trevena and Jones 2006; Lana et al. 2011). This seasonality is also seen in measurements of secondary sulfate in New Zealand urban PM: secondary sulfate concentrations generally maximise during summer and are at a minimum during the winter months (Figure 4), reflecting the seasonality in the marine DMS source and the influence of solar forcing on atmospheric reaction pathways.

# Air particulate matter composition analysis and source apportionment in New Zealand

The Institute of Geological and Nuclear Sciences (GNS Science) has used an air particulate matter composition analysis and receptor modelling approach to identify sources of



**Figure 2.** Ocean chlorophyll *a* concentrations observed via satellite. High chlorophyll *a* concentrations are indicative of marine biogenic activity, which leads to marine aerosol production via primary marine organic aerosol and DMS emission. Data are averaged over December 2022–February 2023. Data source: Moderate Resolution Imaging Spectroradiometer (MODIS)-Aqua (Hu et al. 2019; O'Reilly and Werdell 2019).

airborne particles in New Zealand airsheds since 1996. Data are compiled into the National Air Particulate Matter Speciation Database (NAPMSD) held by GNS Science (GNS Science 2020; Davy and Trompetter 2020). Elemental concentrations in filterbased particulate matter samples are determined by two nuclear analytical techniques at the New Zealand National Isotope Centre: Ion Beam Analysis and/or X-ray fluorescence (Trompetter et al. 2005; Barry et al. 2012). Since these techniques are nondestructive, most of the collected sample filters are now archived (>50,000 samples) at the National Isotope Centre in Lower Hutt. Each source or type of particulate matter has a distinctive particle size range and chemical composition (Figure 5).

Filter-based PM samples have been collected and analysed for approximately 40 sites across New Zealand, with multiple sites included in some urban areas (Figure 6). Filterbased sample collection has been conducted primarily on behalf of regional councils



**Figure 3.** Marine biogenic activity leads to the production of dimethylsulfide (DMS). When emitted into the atmosphere, DMS undergoes chemical reactions which ultimately lead to the formation of sulfate aerosol and cloud condensation nuclei (CCN; see Section 'Climate impacts of marine aerosol').







**Figure 5.** Scanning electron microscope images of particulate matter showing differences in their structures (3500–10,000× magnification; Davy 2007).

seeking to understand the sources that lead to air pollution episodes so that air quality can be managed to protect human health as required by the National Environmental Standards for Air Quality (Resource Management Regulations, 2004). Accordingly, the majority of the sampling campaigns ran for 1–2 years collecting 24-hour time integrated particulate matter samples to better understand the local drivers of air pollution for air quality management purposes. The exception is the Auckland Council multi-site air particulate matter speciation database that has been running since mid-2004. The long time series of data collected in Auckland allows for inter-site comparisons, trend analysis and all-of-urban assessment of PM composition and source contributions to both  $PM_{2.5}$  and  $PM_{10}$  size fractions (Davy and Trompetter 2019).

Receptor modelling analysis of filter samples held in the NAPMSD shows that marine aerosol is the primary source of natural aerosol present in urban areas of New Zealand. Secondary sulfate aerosol-that is, sulfate particles that have nucleated from the sulfur-containing gas  $SO_2$  -is also present in New Zealand urban air. Secondary sulfate aerosol has a range of sources; some natural and some anthropogenic. An important natural source is the activity of marine phytoplankton (Section 'Secondary sulfate aerosol'). The elemental analysis of filter samples shows that the ocean contributes significantly to air quality at many New Zealand urban sites. For example, in Auckland, the contribution of marine aerosol and secondary sulfate aerosol to measured  $PM_{10}$  and  $PM_{2.5}$  is around 50% and 30%, respectively (Figure 7).

Sodium and chlorine are the primary constituents of sea salt, and are also significant elemental contributors to both  $PM_{2.5}$  and  $PM_{10}$  mass at New Zealand monitoring sites along with the more minor components of sea salt (potassium, calcium, magnesium and sulfur). Figure 8 presents box plots for the sea salt contribution to particulate matter concentrations, indicating that sea salt aerosol is ubiquitous in New Zealand urban air. This is also visible in the size distribution for New Zealand shown in Figure 1. The coarse mode aerosol number concentration is approximately one order of magnitude greater over New Zealand compared with the global mean, reflecting the abundance of sea salt in New



**Figure 6.** Urban particulate matter speciation sampling locations in New Zealand (Davy and Trompetter 2020). The inset shows the Auckland sampling sites included in Figure 7.

Zealand's air. Sodium and chlorine are highly correlated in PM samples (as shown for the Auckland data set in Figure 9) and present in the same ratio at peak concentrations as found in sea salt ([Na] = 0.56[Cl]; Lide 1992). Although chloride depletion and its removal from sea salt in the form of HCl can alter the Na/Cl ratio in sea salt aerosol,



Figure 7. Composite average for sources contributing to particulate matter at five Auckland monitoring sites (Takapuna, Penrose, Queen Street, Khyber Pass Road, and Henderson; Davy and Trompetter 2019).

the analytical results presented in Figure 8 and 9 demonstrate the relative influence of this natural aerosol source on urban particulate matter concentrations in New Zealand, even for inland locations, due to the isolated oceanic location of the New Zealand landmass.

As discussed in Section 'Sea spray aerosol', the concentration of primary marine aerosol shows a strong dependence on wind speed across the ocean surface. In general, primary marine aerosol fluxes increase at higher wind speeds, and the Southern Ocean is one of the windiest regions on Earth. Therefore, marine aerosol concentrations in New Zealand urban areas are largely influenced by meteorological and long-range



**Figure 8.** Box plots for sea salt contributions to  $PM_{10}$  and  $PM_{2.5}$  at NZ monitoring locations. Note that the monitoring periods vary from site to site.

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Figure 9. Scatterplot for sodium and chlorine in all Auckland PM samples (Davy and Trompetter 2020).

transport mechanisms (Davy et al. 2011). The marine aerosol component of urban air particulate matter is part of the natural background and cannot be managed. It has been shown previously that the primary source regions of marine aerosol in Takapuna's air are in the Southern Ocean below Australia and to the northeast of Auckland (Figure 10; Davy et al. 2017). Similar plots generated for other New Zealand locations show analogous source regions depending on predominant local air mass trajectories.



**Figure 10.** Potential source contribution function plot (using 96-hour back trajectories) for the Takapuna  $PM_{10}$  marine aerosol source contribution data (2006–2013) showing that the most likely source regions are in the Southern Ocean below Australia and the Pacific Ocean to the northeast of Auckland. From Davy et al. (2017).



**Figure 11.** Box plots for secondary sulfate contributions to  $PM_{10}$  and  $PM_{2.5}$  at NZ monitoring locations. Note that the monitoring periods vary from site to site.

Secondary sulfate aerosol is produced from natural and anthropogenic sources. Natural sources include marine biogenic (e.g. phytoplankton) activity and volcanic emissions. Anthropogenic sources include the combustion of sulfur-containing fuels and gas-to-particle conversion of precursor gases such as sulfur dioxide (SO<sub>2</sub>). Source apportionment data indicates that there are both natural and anthropogenic sources of secondary sulfate aerosol in New Zealand urban air (Figure 11). The relative contribution depends on local emissions, atmospheric chemical reactions and long-range transport of secondary sulfate (Davy et al. 2017; Davy and Trompetter 2017).

Overall, the significant contribution of primary marine aerosol and secondary sulfate aerosol to particulate matter measured in New Zealand's air presents challenges to meeting air quality guidelines, as discussed in the following case study.

### Case study: Marine aerosol in Hawke's Bay

Awatoto is a 3 km long coastal airshed in Hawke's Bay. The airshed is mostly industrial; local industrial activities include fertiliser production, composting, pet food manufacturing, wastewater treatment, engineering and asphalt production. Hawke's Bay Regional Council's air quality monitoring site is 200 m from the coast and most of the airshed lies within 800 m of the shoreline.

As discussed in the Introduction, New Zealand's  $PM_{10}$  guideline is 50 µg m<sup>-3</sup> as a 24hour average. One exceedance per monitoring site is allowed in a 12-month period. In general, the highest  $PM_{10}$  concentrations in Awatoto are measured when the wind is



Figure 12. Sources of PM<sub>2.5</sub> and PM<sub>10</sub> in Awatoto airshed.

blowing onshore (from the east). PM exceedances average 2–3 per year; they can occur in any season and predominantly when the wind is blowing onshore.

Particulate source apportionment indicates that despite the local industrial activities at Awatoto, marine aerosol (sea spray and secondary sulfate) makes up approximately 75% of measured PM<sub>10</sub>, and 60% of PM<sub>2.5</sub> (Figure 12). In the summertime, when biomass burning emissions from heating are negligible, the measured PM<sub>2.5</sub> is often greater than  $4 \mu \text{g m}^{-3}$ . While New Zealand currently does not have a PM<sub>2.5</sub> guideline, the World Health Organisation's annual guideline for PM<sub>2.5</sub> is  $5 \mu \text{g m}^{-3}$ . If New Zealand adopts this guideline then meeting it consistently at Awatoto will be challenging. In particular, very low wintertime anthropogenic PM<sub>2.5</sub> levels will be required.

# **Climate impacts of marine aerosol**

Unlike greenhouse gases, which warm the atmosphere by absorbing longwave (infrared) radiation emitted by the Earth, aerosols have complex influences on atmospheric temperature. Aerosol 'direct effects' involve scattering and absorption of shortwave (solar) radiation. 'Indirect effects' involve aerosol-cloud interactions: depending on particle size, composition and water solubility, along with meteorological conditions, aerosols can interact with clouds by acting as CCN (Section 'Introduction') or ice nucleating particles (INP; Forster et al. 2021). Aerosol particles that are large enough to act as CCN, such as sea salt and secondary sulfate aerosol, take up water vapour as it condenses around them and form cloud droplets. In general, aerosol-cloud interactions cause clouds to contain more CCN and therefore more but smaller cloud droplets. Such clouds scatter radiation efficiently and may be longer lived, leading to a cooling at Earth's surface (Twomey 1977; Albrecht 1989; Forster et al. 2021).

A small subset of aerosols can act as INPs (Kanji et al. 2017). INPs are sparse over the Southern Ocean; however, primary marine organic aerosol (Section 'Sea spray aerosol') is a key local source (Vergara-Temprado et al. 2017). INPs interact with supercooled liquid clouds, which form at temperatures between  $0^{\circ}$ C and  $-38^{\circ}$ C (Pruppacher and Klett 1997). In this temperature range, the liquid is cold enough to form ice but requires a surface upon which the first ice crystals can form. INPs can provide such a surface. The

transition from supercooled liquid cloud to ice cloud is significant because the two cloud types have different radiative properties. In a cloud containing the same amount of water, droplets in liquid clouds are smaller and more numerous than ice crystals in an ice cloud (Murray et al. 2021). As a result, liquid clouds reflect more sunlight back to space, leading to greater surface cooling than ice clouds do.

The effects of aerosols on climate are typically quantified by the effective radiative forcing (ERF) metric, which represents the energy gained or lost by the Earth system following an imposed perturbation (Forster et al. 2021). Since the pre-industrial period, the ERF of aerosols calculated relative to the present day is  $-1.3 \pm 0.7$  W m<sup>-2</sup>, implying a cooling effect at Earth's surface. In other words, aerosols have offset some of the warming caused by anthropogenic greenhouse gases, which have an effective radiative forcing three times larger in magnitude of  $3.84 \pm 0.38$  W m<sup>-2</sup> (Forster et al. 2021). Most of the aerosol radiative forcing comes from aerosol-cloud interactions (indirect effects) rather than aerosol-radiation interactions. It should be noted, however, that aerosol-cloud interactions are highly complex and constitute an active area of current research. Along with aerosol-radiation interactions, they contribute large uncertainties in determining the magnitude of human impacts on climate between pre-industrial times and the present day (Szopa et al. 2021). In a New Zealand context, understanding local sources of INP and CCN and representing them accurately in Earth system models is critical because they determine the cloud phase, which is the balance of ice and liquid water clouds. The cloud phase over the Southern Ocean has been shown to significantly influence the equilibrium climate sensitivity, which is the simulated change in global-mean surface air temperature following a doubling of the atmospheric carbon dioxide concentration (Bjordal et al. 2020; Zelinka et al. 2020). Current Earth system models simulate Southern Ocean cloud phase poorly (Cesana et al. 2022), leading to errors in sea surface temperature which affect the accuracy of climate change projections for New Zealand. Future marine aerosol projections are shown in Section 'The future of marine aerosol in New Zealand and the Southern Ocean'.

# Health impacts of marine aerosol

Aerosols are of concern for human health because they can be inhaled and cause respiratory and cardiac illnesses. In New Zealand, the key pollutants of concern are  $PM_{10}$  and  $PM_{2.5}$ , along with nitrogen dioxide gas (NO<sub>2</sub>). As shown in Figure 7, marine aerosols make a significant contribution to  $PM_{10}$  and  $PM_{2.5}$  in New Zealand urban air, along with anthropogenic sources such as fuel combustion in motor vehicles.

The latest assessment of health impacts resulting from air pollution in New Zealand (HAPINZ 3.0) was published in 2022, based on data sourced from 2016 (Kuschel et al. 2022a, 2022b). While the summary report focussed on the implications of air pollution due to anthropogenic sources because these can be controlled, the study assessed effects from natural PM sources as well.

The literature includes considerable discussion as to whether different kinds of PM are likely to have different toxicities. However, the HAPINZ 3.0 study applied the same exposure-response functions to all PM regardless of source. This reflected the approach taken by the World Health Organisation in the formulation of the latest (2021) global air quality guidelines where they decided not to formulate air quality guideline levels (AQG) for different types of PM (i.e. black carbon, elemental carbon, sand and dust storms):



Figure 13. Key steps involved in the Health and Air Pollution in New Zealand 2016 study (HAPINZ 3.0; Kuschel et al. 2022a, 2022b).

... because the quantitative evidence on independent adverse health effects from these pollutants was still insufficient at the time of deriving the AQGs' (WHO 2021).

The HAPINZ 3.0 study assessed human exposure to air pollution using population data and air quality data, and attributed the source contributions using elemental 'fingerprints' from PM samples (Figure 13).

Anthropogenic air pollution in New Zealand in 2016 was found to be responsible for approximately 3300 premature deaths and social costs of \$15.6 billion. This is more than ten times the corresponding annual road death toll (327) and more than 10% of all deaths (31,179) in 2016 (Ministry of Transport 2023; Statistics New Zealand 2023). By comparison, the health impacts of natural sources were found to be just over half of the anthropogenic impacts: approximately one-third of all  $PM_{2,5}$  effects in total (Figure 14).

Whether or not PM is the best marker for marine aerosol health effects is inconclusive. One hypothesis suggests that regular airborne exposure to biogenic compounds and microbiota in sea spray aerosols is health-promoting via the downregulation of a cellular signalling pathway involved in neurological diseases (Van Acker et al. 2020; Wong 2013). Other hypotheses focus on concerns regarding the ageing of sea spray aerosol resulting in chlorine depletion and enrichment in nitrates and sulfates (Su et al. 2022). Additionally,



**Figure 14.** Health impacts from PM<sub>2.5</sub> air pollution from all sources in New Zealand in 2016 (Kuschel et al. 2022a, 2022b).

the emission of sea spray aerosol from harmful algal blooms may enhance exposure to toxins (Lim et al. 2023): a massive toxic algal bloom on the east coast of New Zealand in 1998 resulted in hundreds of cases of respiratory distress (Chang et al. 2001).

### The future of marine aerosol in New Zealand and the Southern Ocean

As discussed in Section 'Marine aerosol production', marine aerosol emissions are sensitive to factors such as wind speed, sea surface temperature and marine biogenic activity, all of which are projected to change in a warming world. The Southern Ocean is projected to warm as greenhouse gas emissions increase (Bracegirdle et al. 2020), with implications for sea spray emissions and marine biogenic activity. Marine biogenic activity, which leads to primary marine organic aerosol and DMS emissions, is also sensitive to changes in sea ice and nutrients (Bock et al. 2021).

Wind speeds are projected to strengthen in some regions of the Southern Hemisphere and weaken in others, due to ongoing greenhouse gas emissions and recovery of the Antarctic ozone hole (Revell et al. 2021, 2022). In the Southern Hemisphere, the highest wind speeds are found just south of 50°S, and are known as the mid-latitude near-surface jet, or westerly jet. The westerly jet strengthened and moved poleward during the late 20th century as a consequence of Antarctic ozone depletion (Thompson et al. 2011), affecting marine aerosol fluxes (Bhatti et al. 2022). Assuming that the Antarctic ozone hole continues to recover from the effects of halogenated ozone-depleting substances, as projected following the Montreal Protocol (Robertson et al. 2023), the westerly jet will weaken and move north (i.e. toward New Zealand) under a very low greenhouse gas emissions scenario, or further strengthen and contract poleward under a high greenhouse gas emissions scenario (Goyal et al. 2021).

Figure 15 shows the change in Southern Hemisphere near-surface sea spray and DMS mass concentrations in the 21st century relative to 2015–2024 in Coupled Model Intercomparison Project phase 6 (CMIP6) Earth system models (Eyring et al. 2016). Under the high greenhouse gas emissions scenario SSP5–8.5 (Shared Socioeconomic Pathway 5–8.5; see Section 1.2 of Bodeker et al. (2022) for an explanation of the SSPs), Earth system models project significant increases in sea spray aerosol and DMS (implying increases in sulfate



**Figure 15.** (a) Change in Southern Hemisphere near-surface sea spray aerosol mass mixing ratios relative to the 2015–2024 average under three Shared Socioeconomic Pathways (SSP1–2.6, SSP2–4.5 and SSP5–8.5). The solid lines show the multi-model mean of CMIP6 Earth system models (see text for details) and shading indicates plus/minus one standard deviation on the multi-model mean. (b) As for (a) but showing near-surface DMS mass mixing ratios. Models used: CNRM-ESM2-1 (Voldoire 2023); GFDL-ESM4 (John et al. 2018); MIROC-ES2L (Tachiiri et al. 2019); MRI-ESM2 (Yukimoto et al. 2019); NorESM2 (Seland et al. 2019); UKESM1 (Good et al. 2019). CNRM-ESM2-1 did not provide DMS projections and is not included in (b).

aerosol) of up to 12% and 40%, respectively. A future enhancement in marine aerosol emissions would represent an example of anthropogenic enhancement of natural emissions, whereby human forcing of the climate system changes physical drivers of aerosol production, such as increases in sea surface temperature and changes in winds (Saiz-Lopez et al. 2023).

Under the moderate-to-low greenhouse gas emissions scenarios SSP1–2.6 and SSP2– 4.5, relatively small increases are seen in DMS and SSA, with large uncertainties (Figure 15). The Intergovernmental Panel on Climate Change assigned 'low confidence in the magnitude and changes in marine aerosol emissions in response to shifts in climate and marine ecosystem processes' (Szopa et al. 2021). This is because there are large uncertainties in process-based understanding of the mechanisms controlling DMS and sea spray emissions (Bock et al. 2021; Liu et al. 2021).

Because future marine aerosol emissions around New Zealand and across the wider Southern Hemisphere are uncertain, future projections of marine sources of CCN and INP are uncertain too. Any future increases in CCN over the Southern Ocean (e.g. from sea salt and DMS-derived sulfate aerosol) are hypothesised to make clouds more reflective (Twohy et al. 2021), while future increases in primary marine organic aerosol could lead to an increase in INPs, thus changing more supercooled liquid clouds to ice clouds. However, ice clouds are also sensitive to future atmospheric temperature changes (Murray et al. 2021).

# **Outlook and conclusions**

The marine environment is the largest source of natural aerosol in New Zealand urban environments. Previous studies have shown that marine aerosol (sea spray and secondary sulfate aerosol) is a significant contributor to  $PM_{10}$  and  $PM_{2.5}$  in New Zealand and contributes to around one-third of premature deaths related to  $PM_{2.5}$  air pollution. However, further research is needed to conclusively determine the toxicity of marine aerosol versus anthropogenic sources of particulate matter.

Sea spray and secondary sulfate aerosol are significant for climate change through their direct radiative effects and interactions with clouds by acting as CCN. Both processes lead to localised cooling at the Earth's surface. Primary marine organic aerosols, although less abundant than sea spray and secondary sulfate aerosols, are important for determining cloud phase as they are the leading source of ice nucleating particles over the Southern Ocean.

Fluxes of marine aerosol are expected to be sensitive to future climate change. A warming sea surface, changes in wind patterns and increases or decreases in marine biogenic activity will all influence future marine aerosol emissions. Changes in winds will be particularly important: all marine aerosols are emitted in greater concentrations at higher wind speeds. The dominant westerly wind pattern over the Southern Ocean has changed in recent decades due to the Antarctic ozone hole, and it will change in future as the ozone hole recovers and greenhouse gas concentrations increase. However, Earth system models disagree on the magnitude of change we might see in marine aerosol emissions under different greenhouse gas emission scenarios. Further process-based studies are needed to inform model development; in particular to understand the role of temperature in sea spray aerosol production, and to develop better proxies to represent the production of DMS from marine biota.

Earth system models project that marine aerosol concentrations across the Southern Hemisphere will either remain relatively constant through the 21st century (under low and moderate greenhouse gas emission scenarios) or increase (under a high emissions scenario). For New Zealand, with its long coastline and close proximity to the windy Southern Ocean, this implies that marine aerosol is not expected to become a smaller contributor to airborne PM concentrations in New Zealand towns and cities. Continued PM monitoring is important, as well as source attribution studies to understand trends in natural marine aerosol emissions (for example in response to climate change).

Lastly, it should be noted that while marine aerosol is important for New Zealand's air, there are other sources of natural and anthropogenic aerosol about which relatively little is known in the New Zealand context. These include biogenic volatile organic compounds (Paton-Walsh et al. 2022), nitrogen-containing aerosols (Jones et al. 2021) and micro- and nano-plastics (Chen et al. 2023; Knobloch et al. 2021). Given the importance of aerosols for human health and climate change, and the projected responses of natural aerosol emissions to climate change, ongoing research is needed.

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