

Spatial and Temporal Variability of Marine Primary Organic
Aerosols over the Global Oceans: A Review

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Abstract

Marine aerosols have a substantial impact on the radiative balance of over 70% of the Earth's surface through scattering of incident solar radiation. They are a source of cloud condensation nuclei (CCN) and create a net cooling effect at the Earth's surface. Each ocean basin has its own spatial and temporal variability which influence the radiative forcing seasonally. In the Atlantic Ocean, the concentration of sea salt has a seasonal pattern with a minimum in summer and maximum in winter because of a dependency of sea salt load on wind speeds. Increased average cloud condensation nuclei (CCN) activity are observed in the Pacific Ocean during periods of higher chlorophyll *a* levels i.e. during the summer months. A clear seasonal pattern was observed in the Indian Ocean for organic aerosols, with a maximum during the austral summer period and minimum concentrations during winter. In the Arctic Ocean, a strong decrease in sea spray aerosol production corresponded with increasing water temperature which was observed for water temperatures between -1°C and 9°C. Atmospheric aerosol particles resulting from the oxidation of dimethylsulfide (DMS) which are driven by ocean temperatures may have an impact on global climate through an enhancement in cloud concentration nuclei (CCN) number concentration and shortwave cloud albedo. The objective of this study is to combine previous marine aerosol experiments in order to attempt to create satellite retrieval emissions to ensure better accuracy looking in the future. These emissions will be put into both global and regional climate models to increase our collective understanding of the contribution of marine aerosols on the global climate forcing and its impact of surface cooling.

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List of Acronyms

ACE 1- Aerosol Characterization Experiment (1995)

ACE 2- Aerosol Characterization Experiment (1997)

CCN- cloud condensation nuclei

DMS- Dimethylsulfide

H₂SO₄- Sulfuric acid

IAOE- International Arctic Ocean Expedition (1991)

IGAC- International Global Atmospheric Chemistry

MSA- Methanesulfonate

MAGE- Marine Aerosol and Gas Exchange (1992)

MAP- Marine Aerosol Production (2006)

Na⁺- Sodium Cation

NH₄- Ammonium

nss-SO₄²⁻ - non-sea salt sulfate

POA- primary organic aerosol

PSI- Pacific Sulfur/Stratus Investigation (1991)

RITS- Radiatively Important Trace Species (1993, 1994)

SO₄- Sulfate

TOGA- Tropical Ocean-Global Atmosphere

Chapter 1. Introduction

1.1 Role of Marine Aerosols

Marine aerosols constitute one of the most important natural aerosol systems globally and affect the Earth's radiative budget as well as to regional air quality. Marine aerosols are classified as either primary or secondary. Primary marine aerosols are emitted directly into the atmosphere from the interaction of wind stress at the ocean surface, such as the mechanical production of sea-spray aerosol (O'Dowd et al., 2007). In terms of sea salt production, the global mass flux is estimated to be $1-3 \times 10^{16} \text{ g yr}^{-1}$ (Erickson et al., 1988). 32% of the global flux comes from the Northern Hemisphere and 92% of the mass flux can be attributed to the supermicrometer size range. An increase in sea salt mass is primarily associated with increasing wind speeds with mass concentrations being measured up to $1000 \mu\text{g m}^{-3}$ at wind speeds of $15-20 \text{ m s}^{-1}$ (Lewis and Schwartz, 2004). Sea-spray aerosol has traditionally been focused on large particles ($> 1 \mu\text{m}$) due to their influence on sea-air transfer of water vapor and latent heat at high wind speeds (e.g. Andreas 1998). Sea salt contributes 44% to the global aerosol optical depth. Estimates for top of atmosphere (TOA), global annual radiative forcing due to sea salt are -1.5 and -5.03 W m^{-2} for low and high emission values, respectively (IPCC 2001). The 4th IPCC Assessment Report elaborated on those findings and concluded that the largest uncertainty was associated with the indirect effect of atmospheric aerosols through the cloud albedo, which is displayed in Fig. 1.

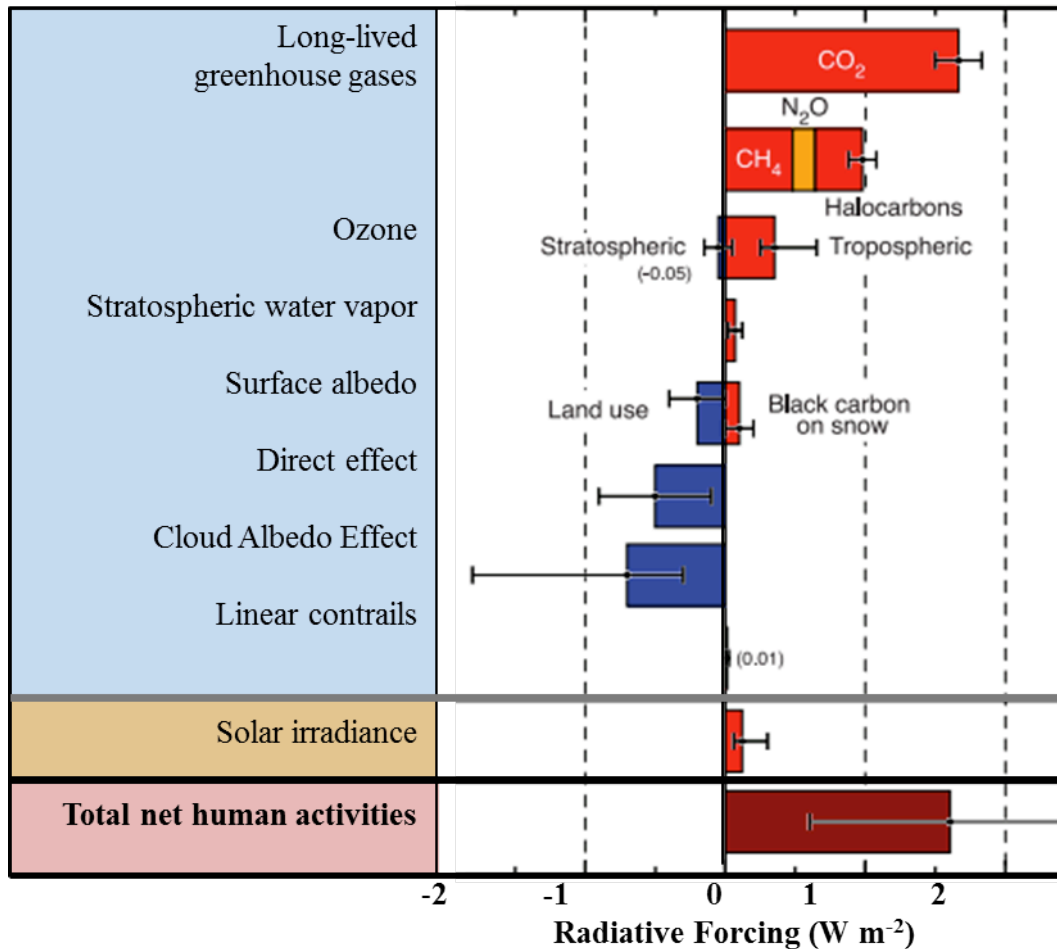


Figure 1. This bar graph displays the principal components of the radiative forcing of climate change. The values represent the forcings in 2005 relative to the start of the industrial era (about 1750). The forcings in blue are due to human activities while the forcing in orange is due to natural processes. The forcing in red is the total net of human activities. (Figure adapted from IPCC 2007 report.)

Sea spray scatters solar radiation, and acts as cloud condensation nuclei (CCN), contributing to the indirect aerosol effect and creating a net cooling effect as seen in Fig.

1. Sea salt has also been linked to the MBL cycle through the activation of halogens, leading to ozone depletion (Vogt et al., 1996).

Secondary marine aerosol production is defined as particle production resulting from gas to particle conversion processes. It has been thought that sulphur species have

been the primary chemical component involved. Secondary aerosol production can occur either by new particle formation via the nucleation of stable clusters of the order of 0.5–1 nm in size or they can also grow via heterogeneous reactions and aqueous phase oxidation of dissolved gases in existing aerosol particles. Dimethylsulfide (DMS) is a waste product produced by phytoplankton and is released from the ocean into the atmosphere where it undergoes oxidation by the OH radical to form SO_4 which is further oxidized to form H_2SO_4 and sulfate aerosols which is displayed in Fig. 2 below.

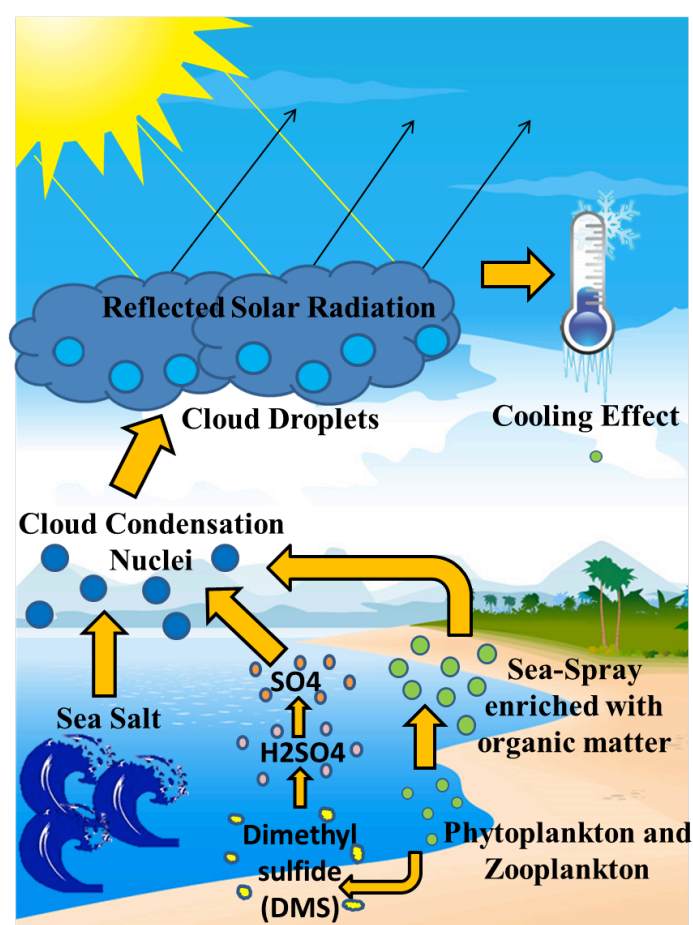


Figure 2. Diagram of how marine aerosols become CCN and add to the indirect aerosol effect in acting to cool the Earth's surface. Phytoplankton and Zooplankton also create sea-spray which is enriched with organic matter that leads to the creation of CCN. The breaking of waves releases sea salt into the

atmosphere which also create more CCN. This increases cloudiness, thus reducing the amount of solar radiation hitting the Earth's surface and creating a cooling effect.

Atmospheric aerosol particles resulting from the oxidation of DMS may have an impact on global climate through an enhancement in CCN number concentration and shortwave cloud albedo. Quinn et al. (1993) completed a study in the northeastern Pacific Ocean in April and May, 1991. The experiment involved taking simultaneous measurements of particulate non-sea salt sulfate, methanesulfonate (MSA), and ammonium mass size distributions. Simultaneous measurements of number size distributions of particles with diameters between 0.02 and 9.6 μm as well as CCN concentrations at 0.3% supersaturation, relative humidity, and temperature were taken. A doubling of non-sea salt sulfate, MSA, and ammonium (NH_4) mass corresponded to a 40–50% increase in number concentration in the accumulation mode. In a separate experiment, measurements of particulate non-sea salt sulfate, MSA, and ammonium mass size distributions over the central Pacific Ocean were made in February 1991 (Quinn et al., 1993). The percent of MSA in the supermicrometer particle size fraction was greater in those samples than in those collected over coastal waters of the northeastern Pacific.

Recent experiments have shown that aerosol production processes for inorganic species are insufficient to account for the entire aerosol mass that occurs in submicrometer sizes, and have pointed to the presence of significant concentrations of organic matter in marine aerosols. O'Dowd et al. (2004) sought to understand the contribution of organic matter to marine aerosol as a function of aerosol size. Measurements of the physical and chemical characteristics of submicrometer marine aerosol over the North Atlantic Ocean were taken during plankton blooms progressing

from spring through autumn. The organic fraction dominated and contributed 63% to the submicrometer aerosol mass during bloom periods. In winter, the organic fraction decreases to 15% when biological activity is at its lowest. Model simulations performed by O'Dowd et al. (2004) indicated that organic matter can enhance the cloud droplet concentration by 15% to more than 100%, and is therefore, an important component of the aerosol cloud climate feedback system involving marine biota.

Attempts to extend primary organic aerosol (POA) coverage both globally and seasonally have proved a difficult task, especially coverage over the oceans. This problem exists because most in-situ measurements are taken in populated areas, in which the oceans remain uninhabited. The chemical nature and origin of the fine particulate matter over oceanic regions are largely unknown because of the insufficient spatial and temporal coverage of in-situ measurements as well as the large variability in the emission from the ocean resulting from changing biological activity in seawater (Decesari et al., 2011). Our ability to measure organic aerosols has been hampered in the past by an incompatibility of sensors with different measuring principles as well as calibration sensors. For example, inlet losses, particle size, concentration calibrations, as well as compatibility of measuring principles have been the prime problems of size distribution measurements in both the marine and continental aerosol (Heintzenberg et al., 2000).

Marine primary organic aerosols have a substantial impact on the radiative balance of the Earth through scattering of incident solar radiation and as a source of cloud condensation nuclei (CCN). Sea spray aerosols represent the largest natural aerosol source on Earth by mass flux (Zábori et al., 2012). Different model estimates of the global annual mean clear sky direct radiative forcing due to sea salt range between -0.6 W m^{-2}

and -5.03 W m^{-2} (Ma et al., 2008). Sea salt emissions have been estimated to be on average $16,600 \text{ Tg yr}^{-1}$ (Textor et al., 2006). Sea salt aerosols may account for between 5% and 90% of the CCN in the marine atmosphere (Clarke et al., 2006). Submicron marine aerosols are an important controlling factor in the atmospheric radiative budget by dictating the number of CCN in the marine atmosphere (O'Dowd and de Leeuw, 2007).

The objective of this study is to combine previous marine aerosol experiments in order to determine an understanding of the current knowledge in this field. This will help create a wider foundation of previously obtained observations, which will be used to aid in the creation of future satellite retrieval emissions to ensure better accuracy. These emissions will be put into both global and regional aerosol models to understand the contribution of marine aerosols on the global climate forcing and its impact of surface cooling.

Chapter 2. Marine Aerosol Campaigns

2.1 Brief Overview of Past Marine Aerosol Experiments

There have been many marine aerosol experiments performed over the last ~25 years that have helped advance our understanding of marine aerosol observations in each of the ocean basins around the globe. The Atlantic and Pacific Oceans cover the most surface area of the world, and have been studied extensively. The purpose of this chapter is to summarize the field campaigns from which will be utilized to solidify and complete our current understanding of marine aerosols across the globe, in the hopes of better understanding their indirect radiative effect through the use of future satellite retrievals of marine aerosol emissions.

2.2 International Arctic Ocean Expedition (IAOE 91)

The goal of the International Arctic Ocean Expedition was to test the hypothesis that marine biogenically produced DMS gas could exert a significant global climatic control. DMS is transferred to the atmosphere and is oxidized to form airborne particles, in which some of these grow large enough to act as CCN. CCN have a strong influence on cloud albedo and the radiation balance of the area affected. The icebreaker, Oden, left Tromsø, Northern Norway on August 1st, 1991 and crossed the Barents Sea toward Fram Strait and the Greenland Sea before operating west and north of Spitsbergen into the marginal ice zones from August 4th–7th, 1991. On August 17th, 1991, the ship operated over the Nansen and Amundsen basins. The geographic North Pole was reached on September 7th, 1991. On October 5th, 1991 Oden reached the open sea in the same area as

on August 4–7, 1991 which allowed for a comparison of measurements in different seasons. The experiment ended on October 8th, 1991.

Research done by Leck et al. (1996) found that surface seawater DMS showed a clear seasonal progression, decreasing by about three orders of magnitude in concentration between August and October 1991. An agreement between apparent seasonal changes in seawater DMS, atmospheric DMS, MSA and nss-SO_4^{2-} measurements demonstrated the importance to DMS concentrations in seawater of the phytoplankton bloom and zooplankton abundance in the open water near the ice edge, in the wake of the receding ice and in the pack ice during summer ice-melts. Particulate concentrations were generally so low after mid-September that no direct radiational effects on the environment were likely. Indirect effects after mid-September are minor because of the rapid decline in cloud cover and the low sun.

2.2 Marine Aerosol and Gas Exchange (MAGE) 1992

MAGE 1992 was the first field project of the International Global Atmospheric Chemistry Program's Marine Aerosol and Gas Exchange project (IGAC-MAGE). The experiment had three main goals related to aerosol measurements using two vessels, the R/V Vickers and the R/V Wecoma in the Pacific Ocean. The first goal was to calculate the air-sea exchange of short-lived biogenic trace gases using shipboard measurements of the important seawater and atmospheric species, air-sea exchange models and micro-budget flux estimates. Comparisons between the atmospheric chemistry and aerosol properties over productive equatorial and oligotrophic 12°S waters were studied. Lastly,

the experiment calculated the cycling of trace gases in the upper water column using measurements of the key species as a function of depth and microbiological rate studies.

The experiment took place between February-March 1992 from 33°N–12°S along 140°W in the Pacific Ocean. The R/V Vickers departed Los Angeles, California on February 21st, 1992 and conducted a six day time series station at 12°S, 135°W, stopped in the Marquesas on March 11th, and returned to Los Angeles, California on March 25th, 1992.

2.3 RITS (Radiatively Important Trace Species) 93/94

The RITS 93 and RITS 94 cruises occurred in the Central Pacific Ocean and sought to assess regional and seasonal variations in both trace gases and aerosols along the latitudinal transect. The chemical, physical, and radiative properties of atmospheric aerosol particles were measured to develop an understanding of air mass sources. RITS 93 began on March 20th, 1993 and departed from Punta Arenas, Chile, crossed the Drake Passage toward Palmer Station on the Antarctic Peninsula. The ship continued southwest to approximately 67°S, 140°W, and then headed north to 57°N, 140°W and arrived in Seattle, Washington on May 7th, 1993. The RITS 94 cruise completed the reverse track of RITS 93. The ship departed Seattle, Washington on November 20th, 1993 and arrived in Punta Arenas, Chile on January 7th, 1994.

2.4 Aerosol Characterization Experiment 1

The First Aerosol Characterization Experiment (ACE 1) sought to determine and understand the properties and controlling factors of the aerosol in the remote marine atmosphere that were relevant to radiative forcing and climate. The purpose of the

experiment was to determine what factors control the formation, growth, and evolution of particles in the marine boundary layer (MBL). Oceanic emissions of dimethylsulfide (DMS) appeared to contribute to the growth of Aitken and accumulation modes while the coarse mode particles were comprised primarily of sea salt based on the measurements recorded on the NOAA ship *Discover* in the Southern Ocean south of Australia from November 16th, to December 12th, 1995 (Bates et al., 1998). The particles resulted from turbulence at the air-sea interface, but the instantaneous wind speed only accounted for one third of the variance in the coarse mode number concentration in the Tasman Sea in the Indian Ocean. Quinn et al. (1998) focused work on understanding the optical properties of a minimally perturbed natural aerosol system in terms of its chemical and physical properties on the ship during this campaign. Mass scattering efficiencies for submicron sea salt were higher due to the tailing of coarse mode sea salt into the particle size range most efficient for light scattering. Based on this study, it was evident that sea salt controls the aerosol optical properties in that Southern Ocean region.

A study done by Brechtel et al. (1998) was also completed during ACE 1 and recorded measurements on Macquarie Island, Tasmania. Observations at Macquarie Island were compared to those from a southern hemisphere mid-latitude site at Cape Grim as well as sites on the Antarctic continent. It was found that the average total number concentration observed during clean marine conditions was about 21% higher than values observed at Cape Grim. Evidence for possible cloud processing of aerosol was found during two Antarctic influenced periods (Brechtel et al., 1998).

2.5 Aerosol Characterization Experiment 2 (1997)

The Second Aerosol Characterization Experiment (ACE 2) sought to expand the understanding of the controlling factors of the aerosol in the remote marine atmosphere that were relevant to radiative forcing and climate. Putaud et al. (2000) measured organic and mineral content as well as mass concentration of the submicron aerosol from June 17th, to July 27th, 1997 on Tenerife Island (28°18'N, 16°30'W) in the Atlantic Ocean off of the coast of Africa, including measurements from the MBL and the free troposphere (FT). A simple budget calculation performed by Putaud et al. (2000) showed that during background conditions, the MBL and FT aerosol compositions were consistent with the hypothesis that the MBL aerosol was formed by the dilution of continental aerosol by FT air, modified by the deposition and condensation of species of oceanic origin. Table 1 displays values in the FT and MBL and compares the Arctic Ocean to the Atlantic Ocean as well as the observations taken during ACE 1.

Average nssSO₄²⁻ Concentrations in both the Atlantic and Arctic Oceans

	nssSO ₄ ²⁻ (FT) (μg m ⁻³)	nssSO ₄ ²⁻ (MBL) (μg m ⁻³)
Average Background	0.14 ± 0.001	0.30 ± 0.05
Background/ Arctic	0.10 ± 0.02	0.32 ± 0.14
Background/ Atlantic	0.15 ± 0.02	0.26 ± 0.7
Baseline Cape Grim (ACE-1) ^a	0.19	0.26

^aHuebert et al. (1998)

Table 1. Average concentrations (STP) observed in the FT and MBL aerosol samples compared with literature data; uncertainties in the average concentration are indicated. (Figure adapted from Putaud et al. 2000.)

The aerosol outbreaks in the FT were due to advection of desert dust which was probably mixed with pollution aerosol from Europe (Putaud et al., 2000). The sea salt submicron concentration at Tenerife Island was 0.23 μg m⁻³ and remained constant during pollution

events which suggest that the balance between sinks and sources for species emitted from the sea surface remained relatively constant. The ratio of MSA/ nssSO₄²⁻ (0.05 ± 0.01) observed during ACE 2 was significantly lower than that of 0.1 observed in the Southern Ocean. A latitude/temperature dependence of the MSA and nssSO₄²⁻ yields from DMS oxidation could explain the difference of measurements between ACE 2 and the Southern Ocean.

2.6 Marine Aerosol Production (MAP; 2006)

The Marine Aerosol Production (MAP) experiment sought to characterize the organic chemical composition of atmospheric submicron particles in the MBL over the northeast Atlantic Ocean in 2006 on and in the vicinity of Mace Head Island. Aerosol measurements were carried out onboard the research ship *Celtic Explorer* from June 12th–20th, 2006 as well as from June 25th–July 5th, 2006 (Decesari et al., 2011). The *Celtic Explorer* crossed oceanic regions full of chlorophyll northwest of Ireland between June 14th and 18th, as well as north of Ireland between June 29th and July 3rd, 2006. The organic aerosol measurements performed during MAP depicted a large pool of oxidized organic aerosol which couldn't be reconciled with the transport of aged continental polluted air. Future efforts need to be dedicated to extend the timeline of parallel online and off-line chemical determinations to find robust overlaps between chemical classes defined in statistical terms or isolated through chemical methods.

2.7 Summary of Marine Aerosol Measurements

Based on the results from the previous experiments, and including the Pacific Sulfur/ Stratus Investigation (PSI 91), sea salt dominates the aerosol mass concentration in the marine boundary layer in the central Pacific Ocean, especially in the submicron size range. This helps affirm that sea salt is a major contributor to scattering by the aerosol in marine regions. Submicron sea salt has a high scattering efficiency and lifetime which is why global transport models of sea salt distribution improved by the inclusion of this size (Quinn and Coffman, 1999). During PSI 91, it was found that open ocean regions, sea salt concentrations in particles with $D \leq 1.4 \mu\text{m}$ were relatively constant at $0.79 \pm 0.14 \mu\text{g m}^{-3}$ and higher than those measured in coastal regions at $0.33 \pm 0.29 \mu\text{g m}^{-3}$. This is because they depend on highly variable local sources, as well as the transport of air from continental regions, or how long the aerosol spends in the air prior to removal by rain. Values of two geometric mass diameters for each of the experiments located in the Pacific Ocean are displayed in Table 3 below.

Marine Aerosol Campaign Information

Experiment	Dates	Location	Sea salt ($\mu\text{g m}^{-3}$) $D \leq 1.4 \mu\text{m}$	Sea salt ($\mu\text{g m}^{-3}$) $D > 1.4 \mu\text{m}$
IAOE 91 ^a	08/01/91– 09/11/91 10/5/91–10/08/ 91	75°N–90°N, 20°W–180°E	N/A	N/A
PSI 91 ^b	04/1991– 05/1991	48°N, 110°W	0.33 ± 0.29	3.3 ± 2.3
MAGE 92 ^b	02/21/92– 03/25/92	12°S–20°N, 140°W	0.75 ± 0.55	12 ± 6.0
RITS 93 ^c	03/20/93– 05/07/93	70°S–40°S, 80°W–140°W 20°S–20°N, 140°W 30°N–54°N, 140°W	0.84 ± 0.64 0.56 ± 0.50 0.84 ± 0.96	4.8 ± 2.3 7.4 ± 3.8 7.3 ± 4.6
RITS 94 ^c	11/20/93– 01/07/1994	70°S–40°S, 140°W 20°S–20°N, 140°W 30°N–54°N, 140°W	0.70 ± 0.31 0.82 ± 0.90 0.71 ± 0.15	5.0 ± 2.0 8.8 ± 2.6 5.3 ± 2.5
ACE 1 ^d	11/16/95– 12/12/95	46°S, 145°E	1.0 ± 0.56	9.3 ± 5.4
ACE 2 ^e	06/17/1997–07 /27/97	28°18'N, 16°30'W	0.23	
MAP ^f	06/12/06– 06/20/06 06/25/06– 07/05/06	55.5°N–57.5°N, 9.5°W–13°W	N/A	N/A

^a Leck et al. (1996), ^b Quinn et al. (1995), ^c Quinn et al. (1996), ^d Quinn et al. (1998), ^e Putaud et al. (2000), ^f Decesari et al. (2011)

Table 3. Sea Salt at a variety of marine locations at two different size ranges are displayed. Particles with geometric mass diameters less than or equal to $1.4\mu\text{m}$ at 70% relative humidity, and particles with geometric mass diameter greater than $1.4\mu\text{m}$ but less than the upper size cut of the aerosol sampling inlet ($\sim 10\mu\text{m}$). (This table is courtesy of Quinn and Coffman, 1999.)

As a result of these experiments, sea salt is proven to comprise a significant mass fraction of both submicron and supermicron aerosol. Sea salt is also proven to dominate aerosol scattering compared to that of other major chemical components.

Chapter 3. Marine Aerosol Measurement Concentration

3.1 Overview

Seasonal and temporal variability varies based on latitude due to DMS production. Chlorophyll *a* is more plentiful during the summer months, which might have a greater impact in the future as the average global surface temperature rises, meaning a longer season favorable to chlorophyll growth. A longer summer season could be correlated with an increase in cloudiness due to the hypothesized overall increase in marine aerosols. Creating a larger understanding of how marine aerosols impact the radiative forcing in current conditions will allow us to run global climate models with more accuracy looking forward. Here we look at both the Atlantic and Pacific Oceans, which represent the largest oceans in the world, along with the Indian and Arctic Oceans which are located at higher latitudes and provide slightly different causes and effects on the global aerosol radiative forcing.

3.2 Atlantic Ocean

Research completed by O'Dowd et al. (2004) determined the primary marine aerosol flux to be about $2 \times 10^6 \text{ m}^{-2}\text{s}^{-1}$ based on typical North Atlantic wind speeds of 10 m s^{-1} . After transit over the Atlantic, primary aerosol generation contributes an input of about $700 \text{ particles cm}^{-3}$ into the marine boundary layer aerosol in which a primary source can explain the observed number concentrations and organic fraction in clean marine air.

A study performed by Yoon et al. (2007) analyzed the seasonal physiochemical characteristics of North Atlantic marine aerosols from January 2002 to June 2004 at Mace Head Island. Seasonal variations were found in the aerosol size distribution modal

diameters. The modal diameter was $0.031\mu\text{m}$ in winter and $0.049\mu\text{m}$ in summer for the Aitken mod and $0.103\mu\text{m}$ in winter and $0.177\mu\text{m}$ in summer for the accumulation mode. The accumulation mode mass showed a seasonal variation with a minimum in winter and maximum in summer. The concentration of sea salt had a seasonal pattern with a minimum in summer and maximum in winter because of a dependency of sea salt load on wind speeds.

3.3 Pacific Ocean

Based on the experiments in the central Pacific Ocean included in Chapter 2, sea salt dominates the aerosol mass concentration in the marine boundary layer which makes it a major contributor to scattering by the aerosol in marine regions based on an analysis by Quinn and Coffman (1999).

Sorooshian et al. (2009) took surface, airborne, and satellite measurements over the eastern Pacific Ocean off the coast of California between 2005 and 2007. The purpose of the experiment was to explore the relationship between ocean chlorophyll *a*, aerosol, and marine clouds. Increased average CCN activity was observed during periods of higher chlorophyll *a* levels which were most likely a result of aerosol size distribution and composition changes over the North Pacific Ocean. Increased CCN activity coincided with periods of increased chlorophyll *a* levels which likely resulted from both size distribution and aerosol composition changes.

3.4 Indian Ocean

Seasonal trends for marine aerosol collected at Amsterdam Island in the Southern Indian Ocean were observed by Sciare et al. (2009). A clear seasonal pattern was observed for organic aerosols, with maximum during the austral summer period and minimum concentrations during winter. It is believed that the seasonal variation of organic concentration was found to be almost entirely related to the water-insoluble carbon fraction which suggested a primary origin which was thought to be through bubble bursting processes. The summer maximum observed for organic aerosols was correlated with satellite-derived chlorophyll *a* concentrations averaged over an oceanic region upwind of the sampling site, and was characterized by relatively high biogenic activity. The biogenic marine organic source of primary origin in the Austral Ocean is fully consistent with similar observations previously reported for the Northern Atlantic Ocean.

3.5 Arctic Ocean

The Arctic Ocean is one of the most rapidly changing sea spray aerosol source areas and is impacted by changing water temperatures. Zábori et al. (2012) found that a strong decrease in sea spray aerosol production corresponded with increasing water temperature which was observed for water temperatures between -1°C and 9°C . Changes in aerosol emissions were most likely linked to changes of the physical properties of sea water at low temperatures. Experiments completed by Zábori et al. (2012) took place using Arctic Ocean sea water carried out at Ny-Ålesund ($78^{\circ}55'\text{N}$, $11^{\circ}56'\text{E}$), Western Svalbard in a marine laboratory. The experiment occurred during late Arctic summer

conditions from August 24th, 2010 to September 7th, 2009, as well as from February 15th, 2010 to March 7th, 2010.

Chapter 4. Past Observation, Satellite, and Modelling Efforts

4.1 Brief History of Surface Observations

As previously completed experiments explained above show, there is a lack of remote marine locations around the globe that aren't impacted by pollution emission. Looking forward, an array of instruments similar to the set of buoys deployed by the Tropical Ocean- Global Atmosphere (TOGA) experiment in the early 1990s in the equatorial Pacific Ocean would aid our ability to capture long term variability at more observational sites to use as validation for both satellite observations and model simulations. Increasing technology to be able to take air measurements on a buoy while keeping costs down would help tremendously in improving our understanding of marine primary organic aerosols and their impacts.

More detailed size-resolved information on marine organic aerosols in the remote MBL is needed to better define the contribution of organics to marine CCN (Rinaldi et al., 2010). The atmospheric fate of primary and secondary marine organic aerosols also needs to be further evaluated because it might significantly influence the ability of marine aerosols to act as CCN in unperturbed ocean regions. An increase in surface observations as well as an ability to take observations aloft would aid tremendously and prove invaluable.

4.2 Brief History of Satellite Observations

Satellite observations of global aerosol distributions started in the late 1970s and evolved technologically to include marine aerosols in the 1990s. From 1996–1997, the POLDER-1 instrument was able to measure aerosol and cloud properties for eight

months. Observational data for aerosol concentration, cloud optical thickness, and cloud droplet effective radius were used to establish statistical relationships among these parameters in order to analyze the first and second aerosol indirect effects (Quaas et al., 2004). A positive correlation between aerosol index and cloud liquid water path was found, which was particularly pronounced over the Northern Hemisphere mid-latitudes which could be interpreted as observational evidence for the second aerosol indirect effect on a large scale.

4.3 Brief History of Modelling Efforts

A global 3D sea salt simulation with a parametrization of a sea salt source function for both sub- and super-micron particles was developed based on the semi-empirical formulation of Monahan et al. (1986). This simulation had a much better dependence of sea salt on surface wind speed prediction than other schemes compared to observations. This scheme provided the most realistic number flux of sea salt particles between ocean and atmosphere using an indirect impact assessment of sea salt aerosols on climate where submicron particles may have a dominant contribution to aerosol-cloud interactions.

A sea salt aerosol model described by Gong et al. (1997) included sea salt generation due to surface winds and vertical transport by turbulence and convection. It was coupled with a one-dimensional climate model covering a grid square of 300 x 300 km to provide time-variant meteorological conditions including surface winds, precipitation, temperature and relative humidity. Gong et al. (1997) studied atmospheric sea salt aerosol concentrations using long term observations and model simulations of

Na^+ at seven stations around the globe. As a result of this study, a stronger seasonal variation occurred in the high latitude North Atlantic than in regions close to the equator and in high latitude southern hemisphere. In general, concentrations were found to be higher in both boreal and austral winters.

Observations studied by Gong et al. (1997) were taken at seven stations spaced around the globe. The 24 hour mean Na^+ concentrations were measured continuously for five years at Mace Head, Ireland from August 1988 to August 1994 as well as for three years from July 1991 to August 1994 at Heimaey, Iceland. No information was given about the Bermuda and Oahu, Hawaii data observation time frame (Gong. et al., 1997). At Cape Grim, Australia, measurements were taken for five years from December 1988 to May 1993. In Table 3, monthly mean sea salt (Na^+) concentrations of observations and simulations are displayed.

Monthly Sea Salt [Na^+] Concentrations between Observations and Simulations

Observation / Model	Location	Dec. ($\mu\text{g m}^{-3}$)	Jan. ($\mu\text{g m}^{-3}$)	Feb. ($\mu\text{g m}^{-3}$)	June ($\mu\text{g m}^{-3}$)	July ($\mu\text{g m}^{-3}$)	Aug. ($\mu\text{g m}^{-3}$)	Sept. ($\mu\text{g m}^{-3}$)
Mace Head, Ireland: Obs.	53.19°N, 9.54°W	4.21	6.06	5.91	2.95	2.98	2.95	3.64
Model		6.87	8.09	7.77	4.22	3.82	2.83	3.12
Heimaey, Iceland: Obs.	63.40°, 20.30°W	5.30	6.90	11.08	3.09	2.56	2.91	3.29
Model		5.91	8.89	6.61	1.98	2.68	2.15	3.87
Bermuda: Obs.	32.27°N, 64.87°W	5.38	6.37	6.87	4.21	3.99	3.57	5.24
Model		7.08	7.39	5.73	6.37	6.50	5.85	5.33
Oahu, Hawaii: Obs.	21.33°N, 157.70°W	5.26	5.54	4.92	3.96	3.78	4.18	3.47
Model		4.12	5.97	6.56	6.65	7.85	7.89	6.31
Cape Grim, Australia Obs.	40°41'N, 144°41'E	4.94	5.56	5.14	6.66	7.04	7.24	5.99
Model		4.08	3.13	4.66	7.61	7.04	10.27	6.47

Table 3. Comparison of monthly sea salt [Na^+] concentrations between observation and simulation for both winter and summer months. (This data is courtesy of Gong et al. 1997.)

According to the observations, the seasonal variation was stronger at Heimaey, Iceland than at Mace Head, Ireland, in which the minimum was lower for the model than for the observations. The seasonal variation and concentration for Bermuda and Oahu, Hawaii were lower than for the high latitude points. At the Bermuda and Oahu, Hawaii locations, a small decrease in the sea salt concentration in the summer time was observed. A peak of sea salt concentration was predicted by the model which opposed the observations. A general agreement was found between the observations and model predictions in terms of the annually averaged Na^+ mass concentration. The experiment was also performed in Alert, Canada, and Palmer Station, Antarctica in which the model was unable to replicate observations. These two locations are remote from ocean water in which the concentrations observed are not directly comparable to the one-dimensional model predictions which utilize a production flux of sea salt aerosols at Heimaey, Iceland. The model doesn't simulate removal during transport to the sties over ice-covered terrain.

New aerosol modules of global circulation and chemical transport distinguish among aerosol components including sea salt. Assumptions for the permitted water uptake contributed to optical depth differences of sea salt at higher latitudes. There were particularly large discrepancies over tropical oceans and oceans of the Southern Ocean which raised issues on the treatment of sea salt in models (Kinne et al., 2003).

An important source of organic matter from the ocean is omitted from current climate-modelling predictions and should be taken into account (O'Dowd et al., 2004). Given that the evolution of micro-algae is driven by environmental change, particularly increasing oceanic temperatures, the production of primary organic marine aerosol

represents a newly identified and potentially important component of the marine biota and climate feedback system involving aerosols and clouds.

Multi-decadal aerosol variations from 1980 to 2009 were evaluated using observations from multiple satellite sensors, available ground-based networks, and the Goddard Chemistry Aerosol Radiation and Transport (GO-CART) model (Chin et al., 2014). A decreasing dust trend in the North African dust outflow region of the tropical North Atlantic and the receptor sites of Barbados and Miami were closely associated with an increase of the sea surface temperature in the North Atlantic. The temperature increase might have driven the decrease in wind velocity over North Africa, which reduced the dust emission, and increased precipitation over the tropical North Atlantic which enhanced the dust removal during transport.

Comparisons to the Aerosol Robotic Network (AERONET) suggest an underestimate of source strength in many models while comparisons to the combined best of Moderate-Resolution Imaging Spectroradiometer (MODIS) and Total Ozone Mapping Spectrometer (TOMS) which indicate that away from sources, model simulations are usually smaller which can also be validated in the work performed by Gong et al. (1997) using Alert, Canada and Palmer Station, Antarctica surface observations compared to model simulations based on Heimaey, Iceland.

Chapter 5. Future Research

5.1 Effect of Marine Aerosols on Cloudiness

Marine aerosols are created due to processes attributed to sea spray and the conversion of DMS into H₂SO₄. These two processes ultimately create CCN which can elongate a cloud's lifetime as well as increase the volume and area of clouds. These processes are impacted by the ocean temperature and surface wind speed. Giant sea salt nuclei override the precipitation suppression effect of the large number of small pollution nuclei which aids in precipitation but decreases the solar radiation hitting the Earth's surface (Feingold et al., 1999; Rosenfeld et al., 2002). Large droplets initiated by large sea salt aerosols may grow to precipitation size by collecting small cloud droplets, thereby cleansing the air (Lohmann et al., 2005) which can have a beneficial aspect when connected to Climate Change. When the giant CCN are covered by film-forming compounds, then their impact would be less than previously estimated (Medina and Nenes, et al. 2004). It is important to understand the role of marine aerosols and their ability to impact the number and size of clouds as the climate evolves moving forward.

5.2 Effect of Marine Aerosols on Radiative Forcing over Oceans

Aerosol particles influence radiative forcing directly through reflection and absorption of solar and infrared radiation in the atmosphere. Aerosols can cause a negative forcing both directly and indirectly through the changes they cause in cloud properties. These changes include modifications to the microphysical properties, changes to the amount of clouds, and the lifetime of individual clouds as discussed in section 5.1. Many of these processes are shown in Fig. 3 below.

Various Radiative Mechanisms Related to Aerosols

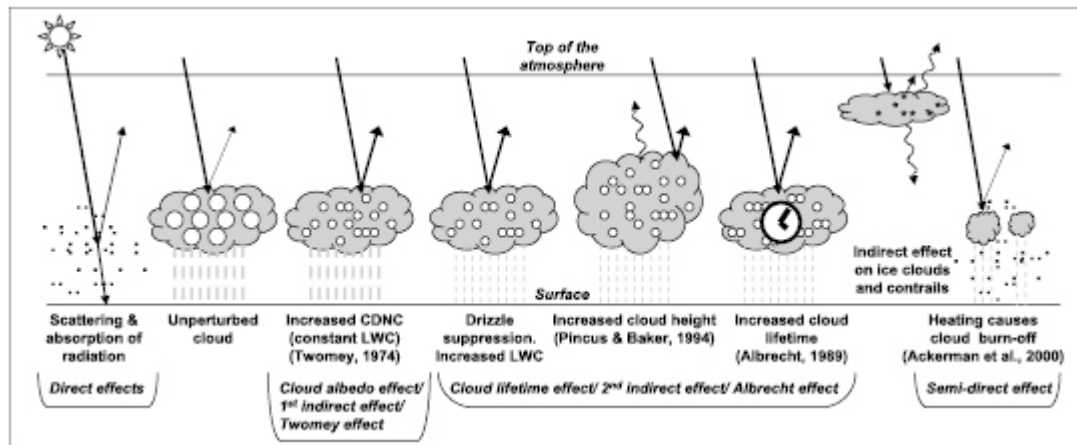


Figure 3. Diagram displays many radiative mechanisms associated with cloud effects that have been identified as significant in relation to aerosols. The black dots represent aerosol particles while the larger open circles represent cloud droplets. (Diagram courtesy of the IPCC 2007.)

Further analysis of marine aerosols will help increase our collective understanding on which processes are most related to marine aerosols and how both the direct and indirect aerosol effects will evolve in the future. Future research will utilize past surface observations from campaigns to create a satellite algorithm in the hopes of measuring global marine aerosol emissions from space at frequent time steps. This will allow us to

understand how marine aerosols currently impact the ocean basins in the hopes of making global climate models more accurate in utilizing the aerosol radiative forcing.

Chapter 6. Summary and Conclusion

Marine primary organic aerosols are currently underrepresented in global climate models and have a large impact on the Earth's radiative forcing. With the annual global averaged surface temperature projected to rise in the future years, marine aerosols will have an increasing impact as sea ice melts and the radiative forcing adjusts.

In the Atlantic Ocean, the accumulation mode mass showed a seasonal variation with a minimum in winter and maximum in summer. The concentration of sea salt had a seasonal pattern with a minimum in summer and maximum in winter because of a dependency of sea salt load on wind speeds (Yoon et al., 2007). Increased average CCN activity was observed in the Pacific Ocean during periods of higher chlorophyll *a* levels which were most likely a result of aerosol size distribution and composition changes over the North Pacific Ocean. Increased CCN activity coincided with periods of increased chlorophyll *a* levels which likely resulted from both size distribution and aerosol composition changes (Sorooshian et al., 2009). A clear seasonal pattern was observed in the Indian Ocean for organic aerosols, with maximum during the austral summer period and minimum concentrations during winter (Sciare et al., 2009).

Zábori et al. (2012) found that a strong decrease in sea spray aerosol production corresponded with increasing water temperature in the Arctic Ocean which was observed for water temperatures between -1°C and 9°C , and will have an increasing impact as the global climate continues to warm. Micro-algae is driven by environmental change, particularly increasing oceanic temperatures, so the production of primary organic marine aerosol represents a newly identified and potentially important component of the marine biota and climate feedback system involving aerosols and clouds.

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