

**THE INFLUENCE OF WIND AND RAIN ON NEAR SURFACE SEA-
SALT AEROSOL AND VISIBILITY OVER THE OPEN-OCEAN**

**A THESIS SUBMITTED TO
THE GLOBAL ENVIRONMENTAL SCIENCE
UNDERGRADUATE DIVISION IN PARTIAL FULFILLMENT
OF THE REQUIREMENTS FOR THE DEGREE OF**

BACHELOR OF SCIENCE

IN

GLOBAL ENVIRONMENTAL SCIENCE

DECEMBER 2007

By

Ross E. Timmerman

Thesis Advisor

Antony D. Clarke

I certify that I have read this thesis and that, in my opinion, it is satisfactory
in scope and quality as a thesis for the degree of Bachelor of Science in
Global Environmental Science.

THESIS ADVISOR

Antony D. Clarke
Department of Oceanography

ACKNOWLEDGEMENTS

I would like to thank my senior thesis advisor, Antony Clarke, for his aid and guidance throughout the project. I would also like to show appreciation for Cameron McNaughton and Yohei Shinozuka for their time, encouragement, and expertise. I would like to thank Vladimir Kapustin for his help. I would like to express gratitude to the Global Environmental Science faculty with special thanks to Jane Schoonmaker and Margaret McManus for their encouragement and support. I would also like to thank Rene Tada for her assistance and delicious snacks. I would lastly like to thank my fiancée, Amanda Vinson, for her unconditional love and motivational support throughout the progression of my thesis. All of these people have made this project possible.

ABSTRACT

Aerosols have been recognized as a major source of uncertainty in climate change models. Wind and precipitation can influence the near surface sea-salt aerosol visibility and satellite derived radiances over the open ocean. Quantifying this role of sea-salt is important for a more complete understanding of aerosols and their effect on climate change (aerosol indirect and direct effects). Here, autonomous ship-based instruments measured meteorological parameters, in-situ and column aerosol light extinction, aerosol size distributions and number concentrations. There was a correlation between wind speed, aerosol production, and light extinction. These relationships must be accounted for in models of radiative transfer and climate for marine regions.

TABLE OF CONTENTS

ACKNOWLEDGMENTS.....	iii
ABSTRACT.....	iv
LIST OF FIGURES.....	vi
CHAPTER 1: INTRODUCTION TO AEROSOLS	1
1.1 AEROSOL PRODUCTION	1
1.2 IMPORTANCE OF AEROSOLS IN CLIMATE.....	2
1.3 AEROSOL EFFECTS ON THE RADIATION BUDGET	4
1.3.1 DIRECT RADIATIVE INFLUENCE	5
1.3.2 SEMI-DIRECT RADIATIVE INFLUENCE	5
1.3.3 INDIRECT RADIATIVE INFLUENCE.....	6
1.4 SEA SALT AEROSOL.....	7
1.5 AEROSOL REMOVAL MECHANISMS.....	9
1.5.1 DELIQUESCENT AND EFFLORESCENCE.....	9
1.5.2 DRY DEPOSITION.....	10
1.5.3 WET DEPOSITION	10
CHAPTER 2: METHODOLOGY	12
2.1 INSTRUMENTATION	13
2.1.1 AEROSOL SIZE DISTRIBUTION.....	13
2.1.2 CEILOMETER	14
2.1.3 VISIBILITY MONITOR	15
CHAPTER 3: RESULTS AND DISCUSSION.....	16
3.1 AEROSOL PRODUCTION	16
3.2 AEROSOL REMOVAL	21
CHAPTER 4: CONCLUSION	24
REFERENCES.....	26

LIST OF FIGURES

Figure	Page
1.1 IPCC Forcing.....	3
1.2 Bubble Bursting.....	8
2.1 PLUME Cruise Track.....	12
2.2 Ceilometer.....	14
2.3 Visibility Monitor.....	15
3.1 PLUME Time Series..... wind speed, aerosol area, visibility, RH, and precipitation	17
3.2 Aerosol Production and Light Extinction Dependencies.....	19
3.3 Visibility and Extinction Dependencies.....	20
3.4 11 May 2007 PLUME Time Series..... wind speed, aerosol area, visibility, RH, and precipitation	22

CHAPTER 1: INTRODUCTION TO AEROSOLS

Aerosols are solid or liquid particles suspended in the atmosphere, and are produced naturally and by anthropogenic activities. Examples include sea salt aerosols from sea spray, sulfate from volcanoes, from various chemical reactions, industrial processes, and fuel combustion (Raes et al., 2000). These different production pathways produce different aerosol sizes, spanning approximately four orders of magnitude (Seinfeld and Pandis, 1998). In general, aerosols are considered to be larger than 2 molecules but less than 100 μm in diameter. Most sizes that interact effectively with light and determine their optical properties discussed here lie between 0.1 and 10 μm . Aerosols also differ in composition and behavior in the atmosphere.

1.1 AEROSOL PRODUCTION

The sources of aerosols can be divided into two types: formed at the surface (primary) and formed in the atmosphere (secondary production). Primary formation of particles occurs due to a mechanical force or disturbance. For example, mineral dust aerosol is produced naturally by high winds over the Earth's deserts, and sea salt aerosol is produced by wind and wave action over the Earth's oceans. Primary aerosols are emitted directly to the atmosphere as particles. By convention, soot aerosol from combustion is considered a primary aerosol as they condense at high temperatures near the emission source.

Secondary aerosol production describes particle formation from existing precursors through chemical reactions. One example is the formation of sulfate aerosols from the biogenic precursor gas, Dimethyl sulfide ((CH₃)₂S). Marine phytoplankton emit Dimethyl sulfide (DMS) on a large scale in the surface waters, making this the dominant source of sulfur (in the form of sulfate) over the ocean (Charlson et al., 1987). The flux of DMS from the ocean to the atmosphere occurs due to the lower atmospheric concentration. DMS is oxidized upon entering the atmosphere, producing various sulfur compounds, which include sulfuric acid and sulfur dioxide. These compounds undergo further reactions that form particles (gas to particle conversion). Secondary production pathways produce fine aerosol sizes (in comparison to primary production pathways), influencing particle behavior and characteristics.

1.2 IMPORTANCE OF AEROSOLS IN CLIMATE

The mounting importance in understanding the effects of climate alteration by anthropogenic perturbations has been recognized by the IPCC. Since the Industrial Revolution, carbon dioxide and other greenhouse gas concentrations have risen drastically due to fossil fuel combustion and industrial processes. Aerosol emissions have also increased as a result of these activities. The difference between these two types of emissions is greenhouse gases tend to heat the atmosphere by allowing incoming solar radiation to enter and trapping outgoing radiation. Aerosols on the other hand tend to reflect incoming radiation, thus cooling the atmosphere. The impacts of these emissions on the planetary radiation budget are speculative due to the variable nature of aerosol

properties and relationships. The recent IPCC estimate for the year 2007 has stated that the effect of an increase in aerosols on the radiation budget is complex and still relatively uncertain (Figure 1.1) (IPCC, 2007).

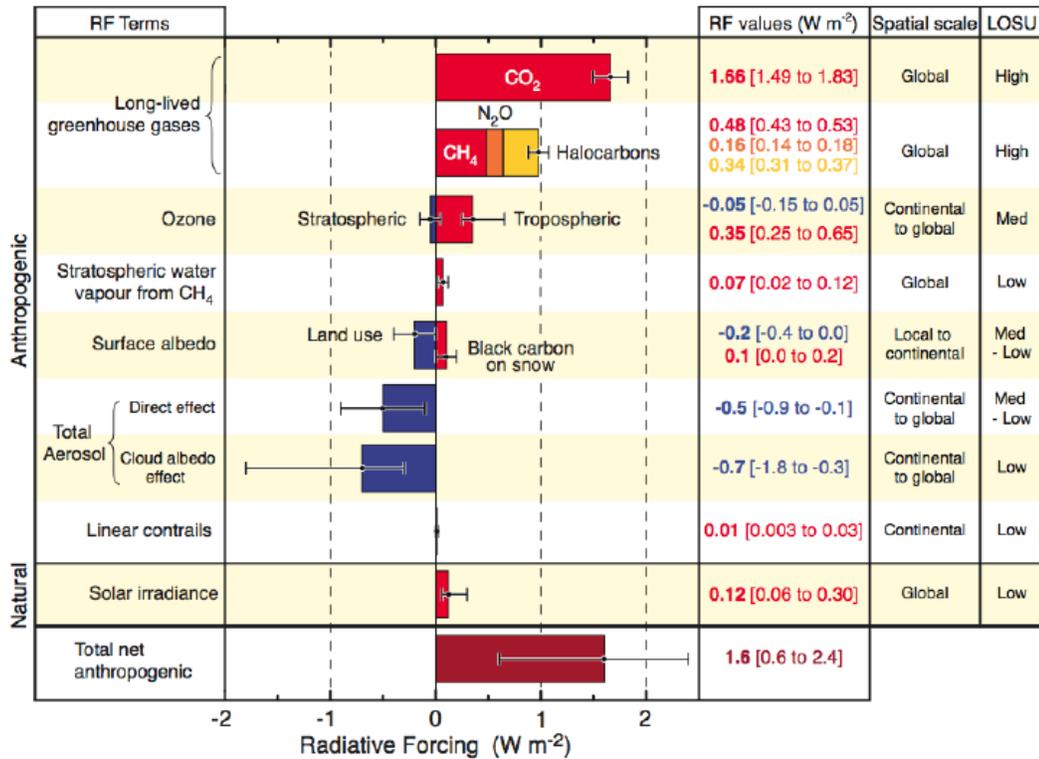


Figure 1.1. IPCC summary for the year 2005 of global-mean radiative forcing (RF) estimates. The red and blue bars indicate the amount of expected forcing along with the uncertainty (lines). The right-most column indicates the level of scientific understanding (LOSU). (provided by <http://www.ipcc.ch/SPM2feb07.pdf>)

Aerosols pose a significant uncertainty in today's climate models (Penner et al., 2006; Kaufman et al., 2002; Lewis Schwartz, 2004, and others). This is due to the variable nature of aerosol properties and characteristics. For example, light scattering and absorption are a function of particle size, with smaller diameters behaving differently from larger diameter particles. Size affects their atmospheric residence time (§1.5), as

well as chemical and physical properties that can influence their incorporation into clouds and precipitation. In particular, the source of uncertainty in radiative forcing lies in the lack of understanding the aerosol direct effect and the more undefined indirect effect. The geographical distribution of aerosol sources is widely dispersed and therefore concentrations and compositions are highly variable, making them difficult to study. A better understanding of the properties and influences of aerosols in the environment would allow for more complete and accurate modeling of the future climate.

1.3 AEROSOL EFFECTS ON THE RADIATION BUDGET

The global radiation balance involves many components but a key influence comes from clouds. Aerosols serve as sites for condensing water vapor, also known as cloud condensation nuclei (CCN), without which clouds and precipitation could not form under normal conditions. The presence of clouds leads to scattering and absorption of radiation. In particular, high clouds tend to warm the planet by trapping outgoing long wave radiation. Low clouds tend to cool the planet by insulating the atmosphere beneath and reflecting most incoming solar radiation back to space (Penner et al., 2006). Thus, the mixing and cycling of aerosols in the atmosphere influences the height of cloud formation, precipitation, and the radiation budget.

1.3.1 DIRECT RADIATIVE INFLUENCE

The *direct effect* refers to the absorption and scattering of incoming shortwave solar radiation by suspended particles, causing net cooling of the atmosphere (Penner et al., 2006). Long wave (infrared) radiation emitted from the earth is also absorbed by these particles (Coakley et al., 1983). On one hand, sea salt and other large particles contribute to this effect significantly at some locations; however the residence time of these aerosols is short due to gravitational settling. Smaller particles, such as sulfate aerosols, are more efficient at contributing to the direct effect. Diameters between 0.1 μm and 10 μm are found to contribute most to the direct effect due to the efficient scattering of light by this size range (Clarke and Kapustin, 2002). Pollution hazes often associated with urban regions tend to reflect light back to space, thereby cooling the planet (negative radiative forcing).

1.3.2 SEMI-DIRECT RADIATIVE INFLUENCE

An intermediate effect of aerosols on the radiation budget occurs due to the increased concentration of CCN, known as the *semi-direct effect*. Cloud formation is dependent on the temperature profile of the atmosphere and its moisture content, which is in part, governed by the presence of fine aerosols (Charlson et al., 1992). Cloud formation can be inhibited when the absorption of solar radiation by particles heats the surrounding atmosphere, however below this level, an increase in clouds may occur

Ackerman et al., 2004). Atmospheric warming prevents water vapor and other gases from condensing onto particles (CCN), making cloud formation less efficient.

1.3.3 INDIRECT RADIATIVE INFLUENCE

The *indirect effect* pertains to a modification of cloud properties as a result of increased concentrations of CCN (Charlson et al., 1992). An increase in the number of fine particles entrained in clouds increases the concentration of CCN. The result is a reduction in cloud droplet size due to the same liquid water content being redistributed over a larger number of particles per unit volume. As a result, higher supersaturation levels are required for growth and precipitation to occur (Penner et al., 2006; Twomey et al., 1984). Individually, these smaller droplets are slow to condense and coagulate into larger droplets necessary for the formation of precipitation. Smaller droplets tend to scatter more light than an equivalent mass of larger droplets. Aerosols in the size range of 0.05 μm and 1.0 μm influence droplet size, number and reflectivity (Clarke and Kapustin, 2002). Within the cloud, the increased scattering increases cloud optical depth, thus increasing the overall albedo (reflectivity) of the cloud (Penner et al., 2006; Twomey, 1977a).

A second indirect effect is associated with changes in the precipitation efficiency of clouds. For example, Takahashi [1976] performed simulations that showed precipitation decreased extensively when CCN concentrations were above 150 cm^{-3} . When there is a delay in energy released (latent heat), clouds persist longer, exerting a

greater influence over the planet by reflecting more short wave radiation over a longer time (Charlson et al., 1992; Wallace and Hobbs, 2006).

The aggregate effect of an increase in CCN concentration is enhanced reflectivity and therefore cooling of the atmosphere. Charlson et al. [1992] estimates that a 30% increase in the aerosol number concentration in marine clouds will result in a global average cooling of 1 W m^{-2} . The estimate for warming due to the increase in GHG concentrations since the Industrial Revolution is 1.6 W m^{-2} , suggesting aerosols offset a significant amount of heating (IPCC, 2007).

1.4 SEA SALT AEROSOL

This study will focus primarily on sea salt aerosol (SSA) production over the remote North Pacific Ocean. Globally SSA is estimated to contribute 1300 Tg yr^{-1} to the total aerosol mass in the atmosphere, second highest to soil dust contribution (Seinfeld and Pandis, 1998). Continentally derived aerosols are found in the remote Pacific; however, most particles originate from the ocean (Savoie and Prospero, 1989).

Production occurs when wind blowing across the ocean creates stress on the surface and on the underlying water column. Waves are generated and will start to break when the force of the wind blows the crest off the wave base, or when the steepness of the wave is too great for the base to support. Breaking waves entrain air bubbles, which rise to the surface and burst, ejecting droplets into the air in two ways (Figure 1.2).

The first process in which droplets are ejected into the atmosphere is known as *film* drops. When the bubble bursts, the film collapses and shatters (Figure 1.2b and 1.2c).

The second way aerosol droplets are produced is by the *jet* that forms after the bubble bursts. Once the film breaks the remaining cavity fills in, producing a column of water (Figure 1.2d) (Lewis and Schwartz, 2004, Wallace and Hobbs, 2006). Both pathways produce droplets and SSA with the same or approximate chemical composition as seawater (Glass and Matteson 1973). Boyce [1951] found that bubble bursting produced more drops than the initial breaking waves. Lewis and Schwartz [2004] report that jet and film drops comprise all small ($D_p < 1 \mu\text{m}$) and medium ($1 \mu\text{m} < D_p < 25 \mu\text{m}$) SSA particle diameters (D_p).

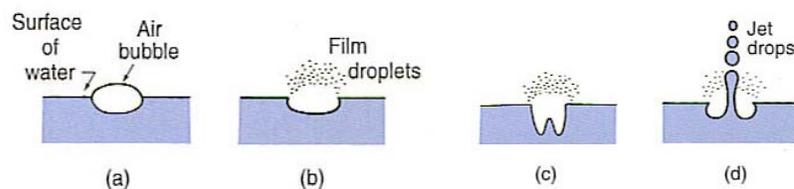


Figure 1.2. Illustration of the bubble bursting mechanisms for SSA formation. Air bubbles rise to the surface (a) and burst (b) and (c), creating film drops. Water fills in the cavity, producing jet drops (d). (Wallace and Hobbs, 2006).

The formation of whitecaps (the remnants of breaking waves) typically begins at wind speeds in excess of 7 m/s, and results in increased aerosol production. White caps are dense clusters of bubbles near the surface of the ocean; the scattering of light by these bubbles produces the coloration. The breaking of these groups of near-surface bubbles produces elevated concentrations of SSA in the methods described. Thus the higher the surface area of the region covered by whitecaps, the higher the level of aerosol production. Additionally, the amount of aerosol production depends on the fetch, or the

distance and duration over which the wind has blown. A longer fetch provides more time and distance in which to increase aerosol concentrations.

1.5 AEROSOL REMOVAL MECHANISMS

Two general pathways for removal of aerosols from the atmosphere are dry and wet deposition. The residence time of most aerosols varies from hours to weeks and can depend on particle size. The residence time of most green house gases is on the order of years to millennia.

1.5.1 DELIQUESCENT AND EFFLORESCENCE

Changes in aerosol properties can result from fluctuations in humidity and precipitation events. The responses to meteorology are not instantaneous, often requiring additional time to achieve equilibrium. Aerosol growth occurs when the partial pressure of a compound in the gas phase is higher than the vapor pressure of that compound in aerosol form (Dorsey, 1940). In other words, aerosol particles grow in size when humidity reaches a critical level and water accretes on the surface. Particles that undergo this transition, known as *deliquescence*, produce a solution (Lewis and Schwartz, 2004). As water vapor continues to condense on the particle, the solution concentration decreases, thus changing the size and optical properties of the particle. For sea-salt this transition occurs near 75 % relative humidity (RH).

In contrast, aerosol properties can change through evaporation. Particles undergo a reduction in size when humidity drops below a critical level, known as *efflorescence*. The concentration of the solution increases until supersaturated, possibly forming a dry particle under low humidity (Lewis and Schwartz, 2004).

1.5.2 DRY DEPOSITION

Dry deposition occurs due to gravitational settling of particles directly to the Earth's surface, which does not involve precipitation in any form (i.e. rain, snow, fog). Because the rate of settling depends on the mass of the aerosol particle, larger particles ($D_p > 1 \mu\text{m}$) settle more efficiently than smaller particles ($D_p < 1 \mu\text{m}$). Therefore, the residence time of smaller particles with respect to dry deposition is longer than larger particles. In other words, turbulent forces such as wind or convection loft finer particles more efficiently. Other factors that affect the rate of deposition are the particle shape, density, and RH.

1.5.3 WET DEPOSITION

There are two mechanisms for removal of aerosols by wet deposition, both transferring particles to the Earth's surface in aqueous form (i.e. rain, snow, fog). The first is by *rainout* (Seinfeld and Pandis, 1998); occurring when aerosols serve as CCN. Once activated, CCN form droplets that continue to grow by condensation, collision and

coalescence, forming cloud drops when the radius typically exceeds 20 μm (Wallace and Hobbs, 2006). If precipitation occurs, these particles are removed from the atmosphere.

The second removal process is *washout*, which occurs when existing cloud drops incorporate aerosol particles as they descend (Seinfeld and Pandis, 1998). These existing raindrops remove aerosol particles beneath the cloud. In comparison to dry deposition, wet deposition removes particles more efficiently; however the process is periodic rather than continuous.

CHAPTER 2: METHODOLOGY

In May 2007, Kilo Moana circled the Hawaiian Islands (Figure 2.1) during the Plume-Lithosphere Undersea Melt Experiment (PLUME). The cruise length was 27 days with continuous meteorology, aerosol, and satellite data available (Figure 2.1). The ship path is plotted in blue with the mid day locations in red. The numbers beside each red dot are the date (year, month, day). The vectors originating from the dots are the average daily wind speeds and directions.

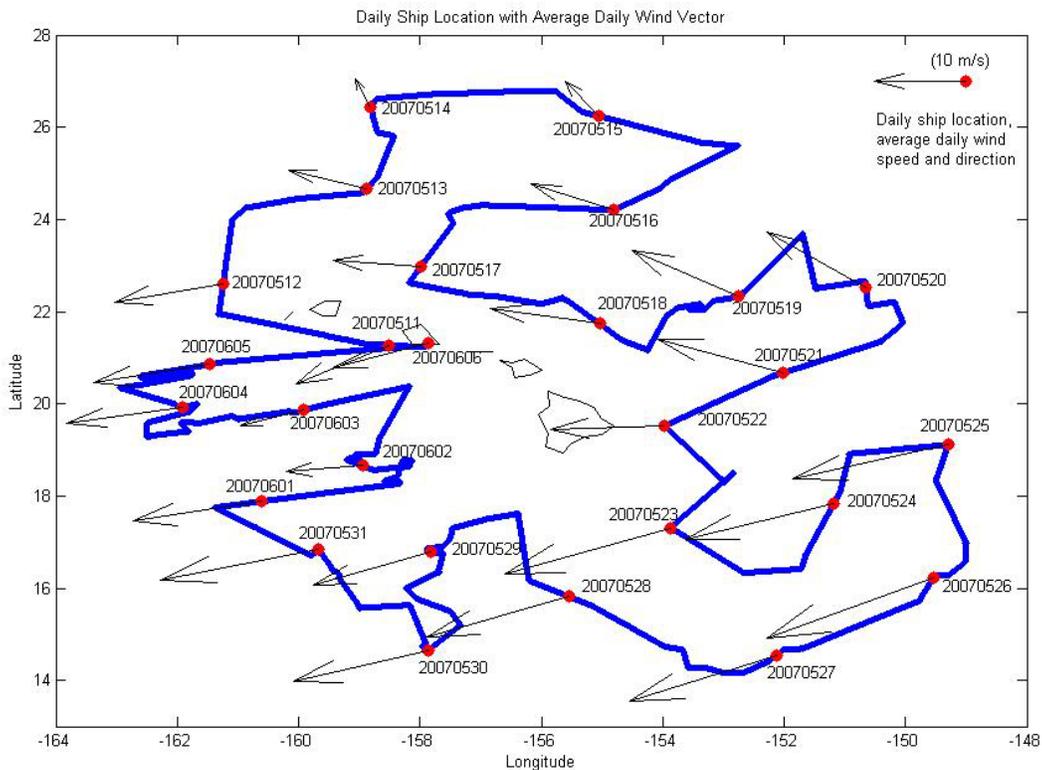


Figure 2.1. Ship path during 27 day long PLUME cruise. The red dots indicate the mid day locations of the ship. The vectors indicate the average daily wind speed and direction. The numbers beside each dot is the date.

2.1 INSTRUMENTATION

The instrument package was deployed on the University of Hawaii Research Vessel (RV) Kilo Moana. This allowed for the analysis of wind speed and precipitation dependencies of aerosol production and removal during the cruise. Data were examined using Matlab computer software (Math Works Inc.). Wind speeds were grouped from 0, 4, 6, 8, 10, and 12 meters per second (m/s).

2.1.1 AEROSOL SIZE DISTRIBUTION

Aerosol number and diameter were measured using a TSI model 3321 aerodynamic particle sizer (APS). This instrument measures aerosols in the 0.7 μm to 20.0 μm aerodynamic size range. For a sea salt density of 2.2 g cm^{-3} , this corresponds to a geometric spherical particle diameter of 0.4 μm to 13.5 μm under dry conditions (RH < 40%). When the ambient relative humidity increases to 80%, the SSA diameter increases by a factor of about 2. At 98% RH, the particle diameters are approximately a factor of 4 higher than under dry conditions (Lewis and Schwartz, 2004). The APS sampled at a height of 15 meters above sea level (MASL) at a resolution of 60 seconds per sample. Area and mass could be computed using particle diameter and number. Aerosol area is used here as it is roughly proportional to light extinction (Clarke et al., 2003).

Meteorological conditions have been found to influence aerosol measurements, making it necessary to couple both types of instruments (Clarke and Kapustin, 2003).

Measurements of meteorological conditions include temperature, humidity, wind speed and direction, visibility, precipitation, and location (latitude and longitude).

2.1.2 CEILOMETER

Two Vaisala ceilometers (models CT25K and CL31) were used to detect column aerosol extinction (airborne particles) using Light Detection and Ranging (LIDAR) (Figure 2.2). The instrument was also capable of detecting clouds and precipitation. The CT25K emits a laser at 905 nm (near infrared) which is reflected back to the sensor, indicating the presence of visual impediments (Vaisala Oyj, 2002). The spatial resolution of the CT25K is 15 meters with a maximum altitude of 7500 meters (Vaisala Oyj, 2002). The ceilometers were positioned at 15 MASL and pointed vertically.

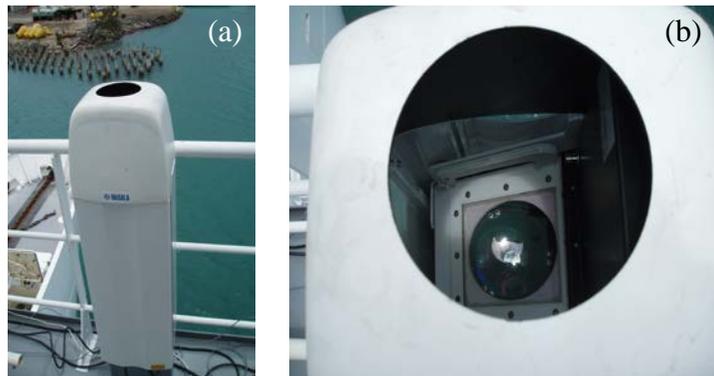


Figure 2.2. Ceilometers utilize lasers for Light Detection and Ranging (LIDAR) of column aerosol extinction. (a) One of the two ceilometers and (b) inside the casing, which prevents direct sunlight from contaminating data.

2.1.3 VISIBILITY MONITOR

A visibility monitor (Vaisala FD12P) was used to measure in-situ light extinction at a height of 15 MASL. Inferred visibility is obtained by measuring forward scattering at 33 degrees. This instrument (Figure 2.3) complements the two ceilometers. Extinction is a measure of visibility, and depends on the presence of suspended particles. The more light scattered, the greater the extinction (lower visibility) due to particles. Increases in humidity may increase extinction (reducing visibility) until eventually the formation of clouds or fog at 100% humidity. The monitor performs well in fog and mist but not as well during precipitation, although it is designed to measure precipitation by counting the pulses in light scattered by falling raindrops.



Figure 2.3. Vaisala visibility monitor (model FD12P). In-situ measurement of light extinction at 15 meters above sea level.

CHAPTER 3: RESULTS AND DISCUSSION

3.1 AEROSOL PRODUCTION

The 60 second high-resolution data do not allow for clear illustration of the aerosol production and removal dependencies on wind speed and precipitation. The data contain instrument noise (anemometer, visibility monitor, and APS) which distorts longer period trends such as fluctuations in wind speed and humidity occurring over tens of minutes. This noise affects the aerosol area and visibility on a similar time scale. Reducing the resolution by averaging over 30 minutes provides clearer trends in aerosol measurements (Figure 3.1). The top panel (a) shows the wind speed in meters per second (m/s) with missing data before 2007 May 9. The aerosol area is shown in (b) and visibility in (c) with arbitrary units (a.u.). Aerosol area and mass was computed from the APS instrument. Visibility was measured with the visibility monitor. Figure 3.1 (d) shows a time series of APS relative humidity (RH) in percent, and the bottom panel shows the time series of precipitation (e) in millimeters (mm).

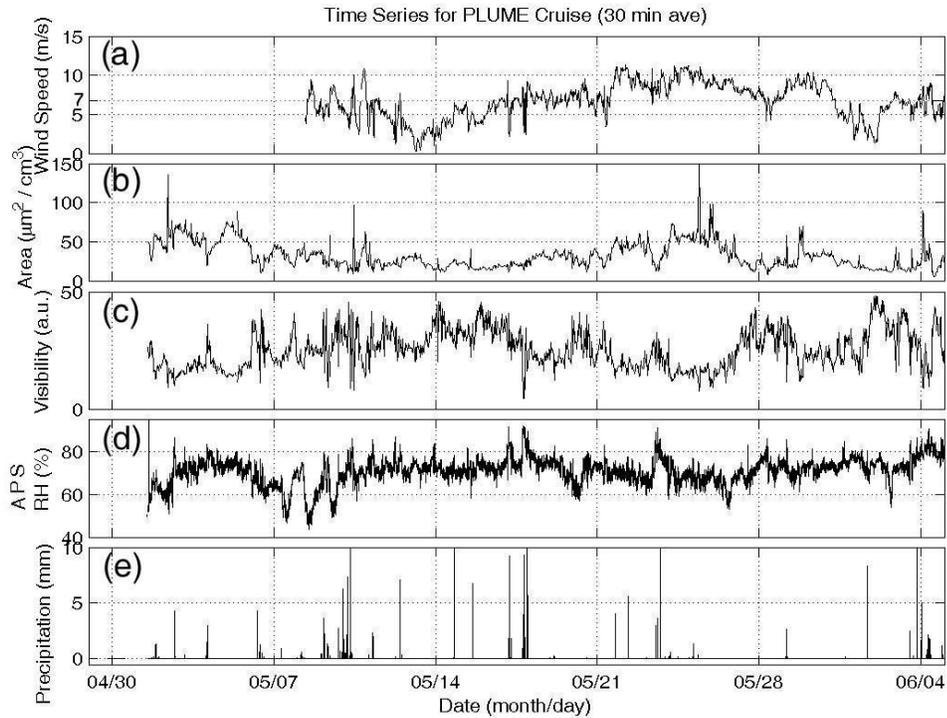


Figure 3.1. Time series showing wind speed, aerosol area, visibility, APS RH, and precipitation during the length of PLUME cruise. The 60 second high resolution data was averaged to 30 minutes to better illustrate longer temporal trends. This was done only for wind speed, aerosol area, and visibility.

The trend of increasing wind speed and increasing aerosol area can be seen in Figure 3.1. Aerosol production (measured as area) increases as the wind speed increases (in excess of 7 m/s, dashed line in plot) to start generating white caps. When humidity rises, the total aerosol area will increase by deliquescence. Note the production response to higher wind speeds can be delayed on a scale of hours due to the required time for wave development over the fetch and vertical mixing into the atmosphere to occur. As a result of higher winds and increased white capping, the bursting bubbles produce SSA and extinction increases (visibility decreases).

Figure 3.2 illustrates the aerosol production dependence on wind speed for PLUME. The averaged aerosol number (a), area (b), and mass (c) are plotted as functions of average wind speed. Also shown are the means (red lines) and standard deviations (red vertical bars) for the incremental wind speeds. The upward trend is a result of increased aerosol production with higher wind speeds. Note the bend in the means corresponding to wind speeds of approximately 7 m/s suggesting a more dominant role in SSA production. Figures 3.2 (d), (e), and (f) show average light extinction as functions of averaged aerosol number, area, and mass respectively. Figures 3.2 (b) and (c) demonstrate the visibility response to increased aerosol production (negative correlation). In other words, light extinction increases as the amount of suspended particles increases (positive correlation).

The secondary correlation (green oval) in Figure 3.2 (f) indicates lower light extinction with little change in aerosol mass. This occurred during periods in which RH was above 80%, which also corresponded to significant precipitation events. The days from May 8 to May 10, May 17, and May 24, 2007 recorded APS RH levels above 80%. RH causes particle growth to occur by deliquescence. Ambient RH during rain events is probably much higher than the APS RH. This makes extinction measurements during rainfall uncertain. Sizing by the APS depends on particle density that also decreases significantly as RH increases. These anomalous data points are related to uncertainty in extinction and APS sizing during precipitation events.

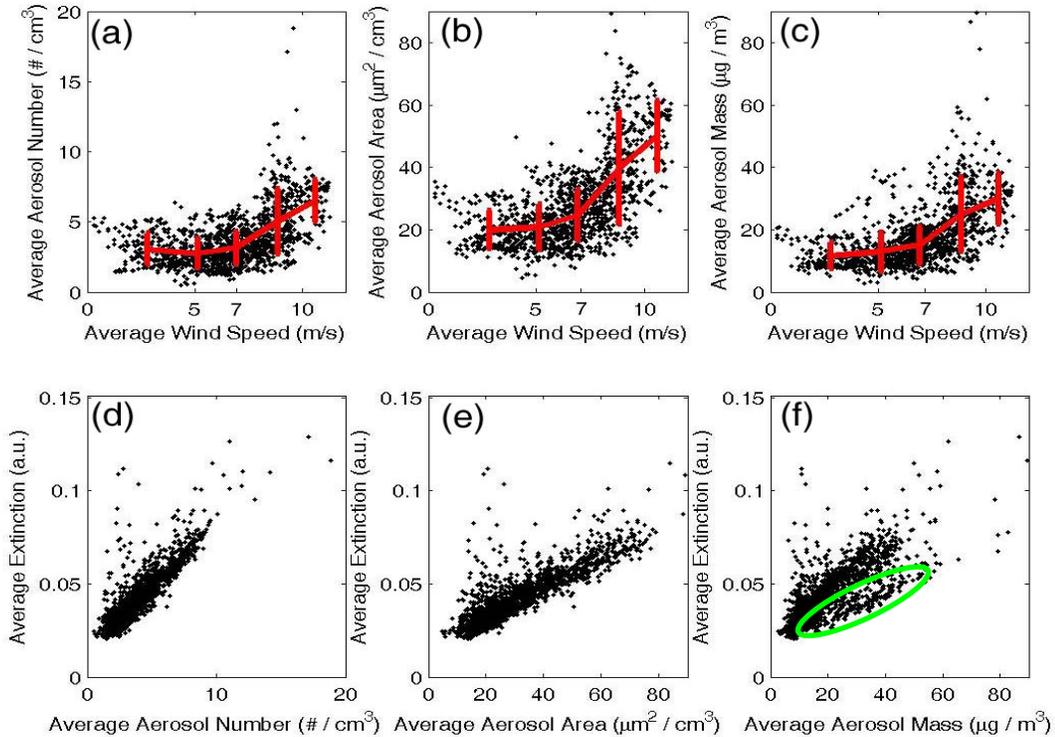


Figure 3.2. The averaged APS aerosol number (a), area (b), and mass (c) plotted to show the wind speed dependencies during PLUME. Red lines indicate the means and red vertical bars show the standard deviations for the wind speed increments. The relation between extinction and aerosol number is shown in (d), area in (e), and mass in (f). Most data reflects APS measurements near 75% RH. The green oval in (f) represents a trend during periods associated with rainfall when RH exceeded 80% (see text).

The average aerosol number measured as a function of wind speed in Figure 3.2 (a) shows the majority of the data lie below 7 cm^{-3} . This range of values for the wind speeds agree with the canonical average and range published in the review by Lewis and Schwartz (Lewis and Schwartz, 2004; and refs). In particular, these results agree with the number concentration measurements by Parungo et al. [1986b], which range from 2 to 7 cm^{-3} for wind speeds between 7 and 10 m/s. The measurements taken by Parungo et al. were shipboard at 15 meters above sea level in the Pacific Ocean.

The range of values for average aerosol mass in Figure 3.2 (c) fall between about 15 to 30 $\mu\text{g}/\text{m}^3$ for winds between 7 and 10 m/s. This agrees well with the canonical value of about 20 $\mu\text{g}/\text{m}^3$ from Lewis and Schwartz [2004]. This value represents data from 10 other investigators that vary by a factor of 3 from this value (Lewis and Schwartz, 2004; and refs).

Figure 3.3 relates visibility and light extinction to wind speed for PLUME. The visibility and wind speed dependency is shown in Figure 3.3 (a). The relation of wind speed and light extinction is shown in Figure 3.3 (b). The red lines indicate the mean and vertical bars indicate the standard deviations for each incremental wind speed. From these figures, it can be seen that an increase in wind speed produces lower visibility, which is equivalent to an increase in extinction. The reason for this was explained in Figure 3.2; aerosol area and wind speed are correlated, and thus visibility and wind speed. Also, the threshold for whitecap production of 7 m/s is apparent by the bends in the means fit to the data corresponding to these wind speeds. As seen in Figure 3.3 (a), the narrowing of the deviations in average visibility demonstrates the increasing wind dependency. In other words, higher wind speeds exert a dominant influence over visibility and its variability.

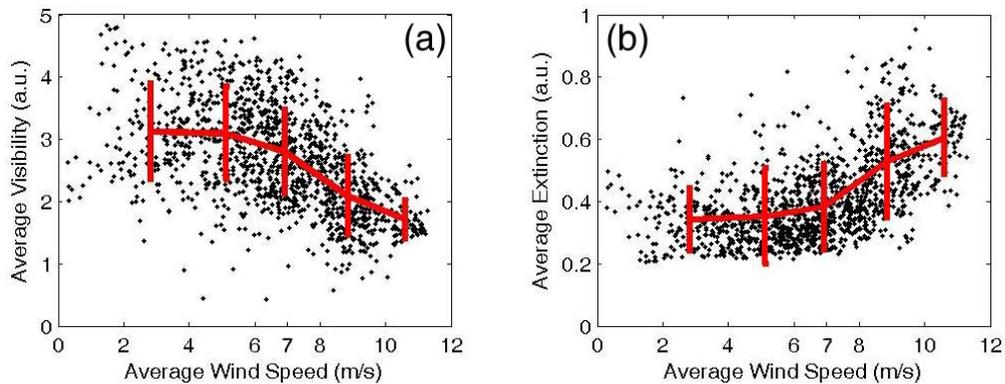


Figure 3.3. Average (30 minute) visibility as a function of wind speed (a); Average (30 minute) light extinction as a function of wind speed (b). Red lines show the means and red vertical bars are the standard deviations for each wind speed increment during PLUME. The trend shows that as wind speed increases, visibility decreases and light extinction increases.

3.2 AEROSOL REMOVAL

The relation of aerosol production and visibility to wind speed is clear in Figures 3.2 and 3.3. However, the near surface SSA and visibility are also dependent on precipitation and RH. During PLUME, rainfall and humidity removed aerosols as seen in Figure 3.4. The day is 11 May 2007 and spans 6 hours. The data (wind speed, aerosol area, and visibility) was averaged over a 15 minute interval to smooth the high frequency fluctuations but preserve temporal trends. The top panel shows a time series of averaged wind speed, aerosol area, visibility, ambient RH and precipitation. Some gaps exist for the precipitation and visibility data due to instrument failure.

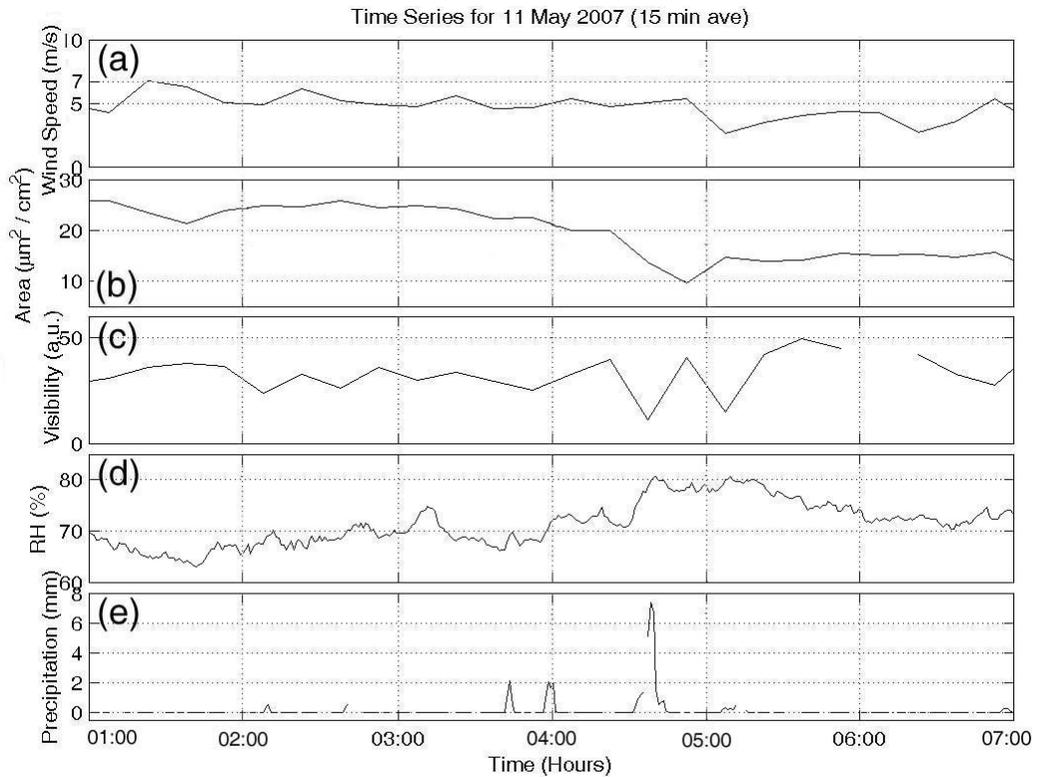


Figure 3.4. Time series of average (15 minute) data (excluding RH and precipitation) for 11 May 2007 showing the period preceding and following rainfall. The top panel shows the wind speed (a), aerosol area (b), visibility (c), RH (d), and precipitation in panel (e). Constant wind speed produced aerosol particles and caused a reduction in visibility. Particles were removed via wet deposition (precipitation and high RH), increasing visibility.

The wind speed was constant (between 5 and 6 m/s) during the period from 1:30 AM (HST) until approximately 5:00 AM, suggesting a nearly continuous rate of aerosol production. However precipitation occurred periodically from 3:45 AM until about 4:45 AM and RH increased at 4:45. The first two rain events (3:45 and 4:00 AM) had a combined total of about 10 mm of rainfall and the last and largest event (4:45 AM) had about 15 mm total (panel (e)). These rain events are reflected by the decreases in visibility (negative correlation) and spikes in RH. The reductions in visibility occurring at

about 2:10, 2:40, and 3:10 AM can be attributed to increases in RH occurring during this time (panel (d)). Visibility was reduced by about 25, 20, and 15% respectively.

The large rain event and high RH at 4:45 AM removed about 50% of the total aerosol area by wet deposition, and reduced visibility by 50%. The second large decrease in visibility (~50%) occurring shortly after 5:00 AM is due to high RH and precipitation. The ship recorded only a small amount of rain; however the decrease in visibility indicates a large rain event in the surrounding area (unmeasured). After about 5:15 AM, visibility increased, RH decreased, and no further precipitation was recorded. This led to the constant aerosol area measurements.

CHAPTER 4: CONCLUSION

Meteorology was found to influence near surface SSA and visibility during PLUME. Specifically, wind speeds exceeding approximately 7 m/s led to higher aerosol production by wave and bubble formation (white cap formation) and a reduction in visibility (increased light extinction). In addition, aerosol area was found to decline during precipitation and high RH via wet removal. Particles are incorporated into cloud droplets and rain drops and deposited at the ocean surface, thus reducing the aerosol concentration. High RH increases particle size and enhances gravitational settling. Surface visibility was reduced during precipitation and high RH due to increased light extinction, and increased following such events. The increase in visibility was due to the removal of aerosol particles.

These relationships have many implications, from maritime navigation and naval weapons systems, to climate studies. Navigational and guidance systems are sensitive to atmospheric visibility, making this a topic of further research. Climate models and observational instruments, such as satellites equipped with LIDAR, must account for natural aerosol production in order to estimate anthropogenic contributions. SSA is one component of total natural aerosols. Further research on wind speed dependencies over different terrains such as deserts, grasslands, and urban regions could be used to improve quantitative aerosol estimates. This would also result in a more complete understanding of the role natural aerosols have with the planetary radiation balance.

SSA influences cloud formation and precipitation by serving as CCN (indirect effect). Higher concentrations of CCN have been found to reduce the size of cloud

droplets and the development of precipitation. More numerous particles will scatter and absorb additional short and long wave radiation, with the overall effect of increasing regional cloud albedo and cooling. The dependency of aerosol production, and thus cloud formation, on wind speed provides insight on natural environmental mechanisms (air-sea interactions). The possibility of higher wind speeds over the ocean as a result of global warming may present a possible negative feedback process by reflecting more solar radiation. Further understanding of the dynamics of the atmosphere may lead to a more complete understanding of aerosols and the effect on climate change, including the aerosol indirect and direct effects.

REFERENCES

- Ackerman, A. S., M. P. Kirkpatrick, D. E. Stevens, and O. B. Toon (2004), The Impact of Humidity Above Stratiform Clouds on Indirect Aerosol Climate Forcing, *Nature*, 432, 1014-1017.
- Boyce, S.G. (1951), Sources of Atmospheric Salts, *Science*, 113, 620-621.
- Charlson, R.J., J. E. Lovelock, M. O. Andreae, and S. G. Warren (1987), Oceanic Phytoplankton, Atmospheric Sulphur, Cloud Albedo and Climate, *Nature*, 326, 655-661.
- Charlson, R.J., S. E. Schwartz, J. M. Hales, R.D. Cess, J. A. Coakley, Jr., J. E. Hansen, and D. J. Hofmann (1992), Climate Forcing by Anthropogenic Aerosols, *Science*, 255, 423-430.
- Clarke, A. D., and V. N. Kapustin (2002), A Pacific Aerosol Study. Part I: A Decade of Data on Particle Production, Transport, Evolution, and Mixing in the Troposphere, *J. Atmos. Sci.*, 59, 363-382.
- Clarke, A. D., and V. N. Kapustin (2003), The Shoreline Environment Aerosol Study (SEAS): A Context for Marine Aerosol Measurements Influenced by a Coastal Environment and Long-Range Transport, *J. Atmos. Oceanic Technol.*, 20, 1351-1361.
- Clarke, A., V. Kapustin, S. Howell, K. Moore, B. Lienert, S. Masonis, T. Anderson, and D. Covert (2003), Sea-Salt Size Distributions from Breaking Waves: Implications for Marine Aerosol Production and Optical Extinction Measurements during SEAS, *J. Atmos. Oceanic Technol.*, 20, 1362-1374.
- Coakley, J. A., Jr., R. D. Cess, and F. B. Yurevich (1983), The Effect of Tropospheric Aerosols on the Earth's Radiation Budget, *J. Atmos. Sci.*, 40 (1), 116-138.
- Dorsey, N. E. (1940), *Properties of Ordinary Water-Substance*, pp. 673, Reinhold Publishing, New York,
- Glass, S. J. J., and Matteson, M. J. (1973), Ion Enrichment in Aerosols Dispersed from Bursting Bubbles in Aqueous Salt Solution, *Tellus*, 25, 272-280.
- IPCC (Intergovernmental Panel on Climate Change) (2007), *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

- Kaufman, Y. J., D. Tanre, and O. Boucher (2002), A Satellite View of Aerosols in the Climate System, *Nature*, 419, 215-223.
- Lewis, E. R., and S. E. Schwartz (2004), *Sea Salt Aerosol Production, Geophys. Monog. Ser.*, vol. 152, pp. 413, AGU, Washington, D. C.
- Parungo, F. P., C. T. Nagamoto, J. Rosinski, and P. L. Haagenson (1986b), A Study of Marine Aerosols Over the Pacific Ocean, *J. Atmos. Chem.*, 4, 199-226.
- Penner, J. E., J. Quaas, T. Storelvmo, T. Takemura, O. Boucher, H. Guo, A. Kirkevåg, J. E. Kristjánsson, and Ø. Seland (2006), Model Intercomparison of Indirect Aerosol Effects, *Atmos. Chem. Phys.*, 6, 3391-3405.
- Raes, F., R. Van Dingenen, E. Vignati, J. Wilson, J.-P. Putaud, J. H. Seinfeld, and P. Adams (2000), Formation and Cycling of Aerosols in the Global Troposphere, *Atmos. Environ.*, 34, 4215-4240.
- Savoie, D. L., and J. M. Prospero (1989), Comparison of Oceanic and Continental Sources of Non-Sea-Salt Sulphate Over the Pacific Ocean, *Nature*, 339, 685-687.
- Seinfeld, J. H., and S. N. Pandis (1998), *Atmospheric Chemistry and Physics*, pp. 1326, John Wiley & Sons, New York.
- Takahashi, T. (1976), Warm Rain, Giant Nuclei and Chemical Balance – A Numerical Model, *J. Atmos. Sci.*, 33, 269-286.
- Twomey, S. A. (1977a) *Atmospheric Aerosols*, pp. 302, Elsevier Scientific Publishing Company, Amsterdam.
- Twomey, S. A., M. Piepgrass, and T. L. Wolfe (1984) An Assessment of the Impact of Pollution on Global Cloud Albedo, *Tellus*, 36B, 356-366.
- Vaisala Oyj: Ceilometer CT25K: User's guide, Vaisala Oyj, Vantaa, 2002.
- Wallace, J.M., and P. V. Hobbs (2006), *Atmospheric Science*, 2nd ed., 483, Elsevier, New York.