A Sea-Salt Flux Parameterization Derived From Field Measurements at a Remote Coastal Site on Oahu

A THESIS SUBMITTED TO THE GRADUATE DIVISION OF THE UNIVERSITY OF HAWAI‘I AT MANOA IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF

MASTER OF SCIENCE

IN

METEOROLOGY

AUGUST 2004

By:

Steven Ray Owens

Thesis Committee:

Antony D. Clarke, Chairperson
Gary M. Barnes
Barry J. Huebert
John N. Porter
ACKNOWLEDGMENTS

To my Mom, Granny and Paw-Paw to whom without none of this would have been possible.

Even though my name appears on the cover of this thesis I owe thanks to many people who made this research possible.

First of all I would like to express my sincere gratitude to my academic advisor Dr. Antony D. Clarke for his advice, patience and support during this work and sticking with me during my first year of graduate school. Without him I never would have experienced the fascinating world of sea-salt aerosol. I would also like to thank the other members of my committee Dr. Gary M. Barnes, Dr. Barry J. Huebert and Dr. John N. Porter for their honest criticism and suggestions on how to improve my thesis as well as Dr. Steven Businger for acting as a proxy member for Dr. Barnes during my defense. I wish to give special thanks to the entire HiGEAR team Volodia Kapustin, Vera Brekhovskikh, Mitchell Pinkerton, Steve Howell and Jingchuan Zhou for their guidance on various issues from lab and instrumentation work to GRL manuscript submission problems that I experienced during the course of my study. My fellow Clarke group aerosol graduate researchers in the MSB 404 Yohei Shinozuka and Cameron McNaughton you guys were good officemates and a lot of fun to work with, but most importantly you guys are great friends. The never-ending support, encouragement and care packages from of my family and the Ryan family back in Memphis provided invaluable motivation to keep me pushing along through the trials and tribulations I experienced during my studies these past 2.89 years. Last but certainly not least my ohana in Hawaii; I never would have survived this journey through graduate school without you guys.

This work was supported by a grant from the Office of Naval Research (N00014-96-1-0320).
Particles produced exclusively from coastal breaking waves have been identified over a detectable size range of $0.01 \leq D_p \leq 8 \mu m$, about an order of magnitude smaller than previously reported, with a peak in number concentration near $0.035 \mu m$. The thermal stability of particles produced by typical oceanic breaking waves and similarities with synthetic ocean spray size distributions is consistent with them being sea-salt. These size distributions were used in conjunction with the aerosol field generated by coastal breaking waves to develop new flux estimates for oceanic whitecaps that extend down to $0.01 \mu m$. The derived source flux compares favorably with previously published estimates and it was found that nuclei $\leq 0.1 \mu m$ dominate the number flux.
# TABLE OF CONTENTS

Acknowledgments.................................................................................................................. iii
Abstract........................................................................................................................................ iv
List of Tables................................................................................................................................ vi
List of Figures............................................................................................................................ vii

I. Introduction.............................................................................................................................. 1
   1.1 Historical Perspective of the Evolution in Sea-Salt Measurements................................. 2
   1.2 Generation Mechanisms of Sea Spray Droplets: Formation of Sea-Salt Aerosol............ 5
   1.3 Physical Properties Sea-Salt Aerosol................................................................................ 7
   1.4 Sea-Salt Aerosol Generation Function.............................................................................. 9
   1.5 Methods of Determining Size-Dependent Aerosol Production Fluxes........................... 10
   1.6 Estimating a Sea-Salt Flux from Coastal Breaking Waves............................................... 11

II. Measurements......................................................................................................................... 17
   2.1 Coastal Experiments at BAFS....................................................................................... 17
   2.2 Instrumentation............................................................................................................... 18
   2.3 Results of Coastal Experiments: Linking Surf Zone Particle Production and Coastal Measurements................................................................................................................. 22

III. Formulation of Source Function.......................................................................................... 25
   3.1 Deriving the Sea-Salt Flux............................................................................................... 25
       3.1.1 Description of Terms in $N_{100}$........................................................................... 29
       3.2 Parameterization and Scaling to Oceanic Conditions................................................. 35

IV. Discussion and Conclusion of Research.............................................................................. 37
   4.1 Comparison With Current Source Functions................................................................... 37
   4.2 Conclusions of Research................................................................................................. 42

Appendix A: Previous Flux Estimates....................................................................................... 44
Appendix B: Derivation of Source Function.............................................................................. 52
Appendix C: Uncertainty Analysis/Propagation......................................................................... 54
References................................................................................................................................... 58
## LIST OF TABLES

<table>
<thead>
<tr>
<th>Table</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Table 1.6.1 Processes Affecting Sea-Salt Aerosol Particle Production &amp; Measurement</td>
<td>15</td>
</tr>
<tr>
<td>2. Table 3.2 Coefficients ($\beta_0 - \beta_3$) for $A$ in Equation 3.2b describing $N_{100}$ over three size intervals in $\mu$m</td>
<td>36</td>
</tr>
<tr>
<td>3. Table 4.11 Total Fluxes Over Entire Valid Size Range</td>
<td>38</td>
</tr>
<tr>
<td>4. Table 4.12 Total Fluxes Over Applicable Size Range as Defined by Other Investigators</td>
<td>38</td>
</tr>
<tr>
<td>5. Table A.4 Coefficients for the parameterization of $A_k (c_4 - c_0)$ and $B_k (d_4 - d_0)$ for the three size intervals ($k$)</td>
<td>50</td>
</tr>
</tbody>
</table>
### LIST OF FIGURES

<table>
<thead>
<tr>
<th>Figure</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. <strong>Figure (1.21)</strong> Bubbles on the ocean surface have a thin liquid cap. Upon bursting, this bubble cap burst into many small droplets called film drops.</td>
<td>62</td>
</tr>
<tr>
<td>2. <strong>Figure (1.22)</strong> Jet droplets are produced by the collapse of a spherical cavity after bursting.</td>
<td>62</td>
</tr>
<tr>
<td>3. <strong>Figure (1.23)</strong> High winds tear droplet directly from the wave crests and eject large spume drops into the marine boundary layer.</td>
<td>62</td>
</tr>
<tr>
<td>4. <strong>Figure (1.24)</strong> Sea spray sizes depend on the generation mechanism, and cover about 4 orders of magnitude. The sea spray droplet size spectrum covers several orders of magnitude.</td>
<td>63</td>
</tr>
<tr>
<td>5. <strong>Figure (1.25)</strong> Taken from Andreas [1998] (his Figure 1). Various estimates of the sea spray generation function in terms of the volume flux, for a 10 m wind speed ($U_{10}$) of 15 m/s.</td>
<td>63</td>
</tr>
<tr>
<td>6. <strong>Figure (1.61)</strong> Taken from Mårtensson et al. [2003] (their Figure 6). Number distributions of primary marine aerosol produced at water temperatures of -2°C (green solid), 5°C (blue dash), 15°C (black dot-dash) and 23°C (red solid). Here the salinity was 33‰.</td>
<td>64</td>
</tr>
<tr>
<td>7. <strong>Figure (1.62)</strong> Taken from Mårtensson et al. [2003] (their Figure 5). Number distributions of primary marine aerosol produced from bubbles in water with the salinity of 0.0‰ (green solid), 9.2‰ (blue dash) and 33.0‰ (red solid). The water temperature was 23°C.</td>
<td>64</td>
</tr>
<tr>
<td>8. <strong>Figure (2.11)</strong> Map of Oahu with the location of Bellow Air Force Station indicated.</td>
<td>65</td>
</tr>
<tr>
<td>9. <strong>Figure (2.12)</strong> a) The University of Hawaii Coastal Aerosol Facility located at BAFS on the northern portion of Waimanalo Bay (21°21' N, 157°42' W) has a 20 m tower situated on the site around 37 m inland and is oriented into the prevailing trade winds. b) Near surface aerosol plume inlet.</td>
<td>65</td>
</tr>
<tr>
<td>10. <strong>Figure (2.21)</strong> Images of sea-salt collected during ACE-1, reported in Buseck and Posfai [1999] (their Figure 5) taken with a transmission electron microscope. (a and b) Subhedral halite (NaCl) and euhedral sulfate crystals.</td>
<td>66</td>
</tr>
</tbody>
</table>
11. **Figure (2.22)** Relevant available meteorological variables at 10 m associated with aerosol measurements during SEAS: April 21-27, 2000. Mean values of each are also displayed.

12. **Figure (2.31)** Vertical gradient in CN Hot and scattering from 1 m above the beach up to 20 m on the tower. Measurements < 5 m are most influenced by coastal breaking wave plumes while those at 20 m reflect background oceanic values.

13. **Figure (2.32)** Vertical distribution of lognormal average scattering and standard deviation over the breaking waves at Bellows Air Force Station.

14. **Figure (2.33)** **Top panel:** A simple linear regression linking breaking wave scattering and CN Hot during SEAS after removal of background oceanic values measured at 20 m. Dataset covers a typical 9-minute period. **Bottom panel:** Time series of breaking wave scattering and CN Hot after subtracting oceanic values measured at 20 m on the tower.

15. **Figure (2.34)** Photograph of surf zone at BAFS superimposed with some parameters that make up $N_{100}$.

16. **Figure (2.35)** Mean breaking wave size distribution during SEAS (left y-axis) with a cumulative number plot (right y-axis).

17. **Figure (3.12)** Graphical representation of the calculated total relative uncertainty (i.e. ± 54%) in the derived $N_{100}$ number flux. Note log (left graph) and linear (right graph) y-axes are for same data.

18. **Figure (3.13)** Example analysis of the *Pick Peaks* function from OriginPro 6.1, which was used to determine the mean refractory breaking wave CN excursion measured at 5 m on the tower.

19. **Figure (3.21)** **Bottom Panel:** The original breaking wave number flux is shown in heavy black while the individual fit over each size range is displayed on top of it. **Top Panel:** Simple linear regression of the original breaking wave flux to the 5th order polynomial fit.

20. **Figure (3.22)** Monahan's empirical relation for whitecap coverage (%) for a given wind speed at 10 m, shown for winds ranging 5 - 30 m/s.

21. **Figure (4.11)** A comparison of the derived sea-salt source function (± 54%) determined from SEAS normalized breaking wave refractory size distributions to others available in literature. All flux estimates have been scaled to oceanic conditions based upon estimated whitecap coverage for an assumed wind $U_{10} = 9$ m/s (§ 4.1) and are represented as dry diameters.
I. Introduction:

Sea spray plays important roles in air-sea exchange, the scattering of light in the marine boundary layer (MBL), and affects the microphysical properties of marine clouds [Andreas, 1998]. With 70% of the Earth’s surface covered by ocean, sea-salt has the highest mass flux of all naturally emitted aerosols [Buseck and Posfai, 1999] and determines the optical properties characteristic of a clean background atmosphere unperturbed by the existence of anthropogenic aerosols. In clean marine conditions these sea-salt aerosol dominate electro-optical attenuation and thus affects the transmission of wavelengths from the ultraviolet to the infrared (i.e. 0.06 μm - 1000 μm). Studies have shown that sea-salt aerosol produced from sea spray can be an important source of cloud condensation nuclei (CCN) [Smith et al., 1998] thus, being essential to the understanding and modeling of climate forcing. Additionally, sea-salt particles can provide large surface areas for heterogeneous atmospheric reactions. Understanding the magnitude and variation of this natural source is necessary in order to identify the effects of aerosols on a global scale. This contribution is also important to both satellite retrieval and modeling effects used to resolve the relative influence of continental and anthropogenic sources.

Aerosol particles affect the Earth’s climate by two processes referred to as the aerosol direct and indirect effect. The main contribution to direct forcing from aerosols in the MBL is the scattering of sunlight back into space, reducing the amount that reaches the Earth’s surface, thus increasing the planetary albedo. This increase in planetary albedo has a net cooling effect on climate (negative forcing). In terms of radiative forcing the sub-micrometer aerosol size range, with its high mass scattering efficiency, is most
relevant to light scattering because it comprises particles with diameters comparable to
the wavelength of visible solar radiation.

Anthropogenic influences upon radiative forcing due to aerosols can be calculated as the
difference between the radiative effects of all aerosols and the effects of naturally
produced aerosols. The validity of this calculation depends on our ability to accurately
model the concentration of naturally produced aerosols (i.e. sea-salt) in the atmosphere.
This natural baseline is characteristic of an atmosphere free of anthropogenic aerosols.
Furthermore, marine aerosols affect the global energy balance indirectly by acting as
CCN. Cloud albedo, cover and lifetime are controlled by the size and concentration of
cloud droplets. The effectiveness of an aerosol particle as a CCN depends on its size and
response to water; sea-salt particles (being very hygroscopic) can act as efficient CCN.
The presence of aerosol acting as CCN can modify cloud microphysical properties,
allowing clouds to form at a lower supersaturation. Quantification of these effects poses
one of the largest uncertainties on the calculation of global radiative forcing [IPCC,
2001].

1.1 Historical Perspective on the Evolution of Sea-Salt Measurements:
In the late 19th century a Scottish physicist named John Aitken [1881] demonstrated that
particles of water vapor do not coalesce to form cloud particles, rather they must have
something solid on which to condense. He further hypothesized that the world’s oceans
were an important source of such nuclei. Interest in sea-salt aerosols has continued to the
present day, thanks in part to the findings of Wilson [1897] and his invention of the cloud
chamber in 1911. He showed that water vapor would not condense and form clouds at normal humidities in air without these hygroscopic nuclei. For his continued work with the cloud chamber he shared the Nobel Prize for Physics in 1927. Jacobs [1937] pioneered the concept that mechanical dispersion of ocean water by breaking waves and bursting bubbles was responsible for the generation of sea-salt aerosol. Boyce [1951] concluded that the majority of coastal airborne droplets are produced from bursting bubbles through the analysis of spots formed through chemical action on salt sensitive paper held above breaking waves and beyond the wave break point as well as between breaking waves and the beach. In his early study Woodcock [1950] reports findings on oceanic airborne sea-salt concentrations sampled at the top of the Hillsboro lighthouse (38 m asl) in Florida in 1948. The mass and number of nuclei were calculated from exposing 20 silver rods (1 mm²) normal to the ocean during high onshore winds (24 - 30 m/s). Later he applied the same exposure technique during aircraft measurements windward of the Hawaiian Islands, but with microscope observations of sea-salt aerosol on glass slides, to model the relationship between airborne sea-salt mass and wind speed [Woodcock, 1953]. Blanchard [1966] and Gathman and Trent [1968] measured the space charge of newly generated marine aerosols using Faraday cages on the east coast of Hawaii and over the open ocean respectively. They were able to calculate the amount of jet drops produced, however electrical techniques such as this are unable to resolve physical properties i.e. size distributions. In 1970 Blanchard and Syzdek [1972] measured the mass concentration of sea-salt at the top of a 15 m tower on the windward coast of Oahu using a 10 cm long platinum wire. Reanalysis of measurements and observations presented in Woodcock [1953], Blanchard and Woodcock [1980] established one of the
most comprehensive vertical sea-salt mass distributions to date. Using a Tala kite to
measure the vertical distribution of sea-salt in the lowest 150 m at three locations on
Oahu Daniels [1989] arrived at a similar conclusion to Blanchard and Woodcock [1980],
finding the largest mass concentrations are confined near the ground and a very weak
gradient exists in a well mixed region above 20 m.

Early interest in sea-salt aerosol was concerned with its ability to act as condensation
nuclei promoting fog and cloud development and reducing visibility [Aitken, 1881,
Wilson, 1897 and Jacobs, 1937] then attention shifted to its role in rain formation
[Woodcock, 1952]. Other less popular concerns dealt with enrichment of heavy metals,
viruses, bacteria, inert organic material and harmful effects on ship turbines [Blanchard,
1980 and references therein]. Marine aerosol concentrations and fluxes over the ocean
continue to be of interest for a variety of reasons. Most recently a new theory on the
origin of life proposed by a team of international researchers suggest that bubble bursting
mechanisms at the ocean surface and subsequent marine aerosols could have played a key
role in creating the chemical building blocks of living cells [Tuck, 2002]. Over the last
half-century speculation on and desire to model the relationship between sea spray and
air-sea heat and momentum fluxes [Andreas, 1998] (in addition to global aerosol climate
forcing, chemical interactions and electro-optical scattering) has necessitated the
development of the sea spray generation function. In order for models to accurately
reproduce sea-salt aerosol distributions, such a function should be based on
microphysical, meteorological and oceanographic variables. Furthermore, it is crucial to
have representative sea-salt size distributions and concentration measurements over a wide range of environmental conditions.

Since the 1960’s numerous experimental investigations have been undertaken to establish a quantitative relationship between sea-salt aerosol concentrations, size distributions, chemical composition and their various atmospheric effects. Considerable advancements in particle measurement technology in the last 25 years or so have made such estimates possible. Ongoing improvements in open volume techniques (e.g. LIDAR, FSSP) will allow more measurement of the largest particles without fear of particle loss by impaction.

Numerous generation functions are available in literature, however they are primarily limited to describing sea-salt particles with dry diameters \( D_p \geq 1 \mu m \). The main objective of this study is to develop a new parameterized fit to the sea-salt number flux that extends estimates into the sub-micrometer particle size region. Later in this text, the generation function will be discussed in greater detail (§ 1.5) along with a few previous attempts to quantify the sea spray production mechanisms (§ 1.6).

### 1.2 Generation Mechanisms of Sea Spray Droplets: Formation of Sea-Salt Aerosol

Sea-salt aerosols are small salt-loaded particles formed by the evaporation of suspended seawater called sea spray. These particles are ejected into the marine boundary layer (MBL) from physical processes occurring at the ocean surface. Primarily two distinct mechanisms are responsible for the generation of sea-salt aerosol. The most important are
the indirect production mechanisms (bubble mediated), resulting from the entrainment of air bubbles by a breaking wave. These bubbles are highly concentrated in whitecaps associated with breaking waves. When these bubbles burst two different types of droplets are produced. Bubbles floating on the surface have a thin liquid cap. Eventually this thin cap gives way and bursts. Upon bursting, the bubble cap shatters into many small film droplets (Figure 1.21) ranging $0.25 \leq D_p \leq 2.5 \mu m$. After bursting the bubble cavity subsides and forms a water jet. Once the jet becomes unstable a droplet is ejected off the top and forms a jet drop (Figure 1.22) typically in the range of $1.5 \leq D_p \leq 25 \mu m$ [Andreas, 1998]. The rate of production of these droplets is generally assumed to be directly proportional to the fraction of the sea surface covered by whitecaps [Monahan et al., 1986].

Direct mechanisms of drop formation occur when high winds ($\approx 10 \text{ m/s}$) tear much larger spume drops (Figure 1.23) $D_p \geq 10 \mu m$ directly from the wave crest [Monahan et al., 1986]. The relative sizes of all droplets cover several orders magnitude (Figure 1.24). Film, jet and spume droplets are collectively known as sea spray or sea-salt aerosol. After formation these droplets are ejected into the air where their composition is similar to that of seawater. If the relative humidity (RH) of the surrounding environment is less than 100% evaporation will take place and the droplets will eventually reach equilibrium and shrink forming sea-salt aerosol [Blanchard, 1983]. This thesis will focus on sea-salt aerosol with dry diameters over the size range $0.01 \leq D_p \leq 8.0 \mu m$ produced from breaking waves in the surf zone.
1.3 Physical Properties Sea-Salt Aerosol:

**Aerosol Particle Size Distribution:**

Perhaps the most important physical characteristic of an aerosol population is the particle size distribution. The size distribution can reveal characteristics of the formation and evolution of the aerosol as well as size dependent optical properties needed to interpret their radiative effects. The aerosol number size distribution \( \frac{dN}{dD_p} \) refers to number density of particles per unit volume with diameters between \( D_p \) and \( D_p + dD_p \) (i.e. as a function of the particle size). Here \( D_p \) is the dry diameter particle measured in micrometers. Since the number size distribution often covers several orders of magnitude it is often expressed as \( \frac{dN}{d \log D_p} \), where dN is the differential number of particles per cm\(^3\) of air in the size range \( \log D_p \) to \( \log D_p + d \log D_p \). A typical feature of the sea-salt aerosol size distribution is a decrease in particle number concentration with increasing diameter. When plotted in the linear format as \( \frac{dN}{d \log D_p} \) vs. \( \log D_p \), the area under the curve is proportional to the total number of particles and best reveals the contribution of small particles. The logarithmic format (x-axis) provides information over a larger range of particle sizes. The total number of particles per unit volume can be expressed as [after *Seinfeld and Pandis, 1998*]:

\[
N = \int_{-\infty}^{\infty} \left( \frac{dN}{d \log D_p} \right) d \log D_p \quad (1.3a)
\]

Assuming the sampled particles are spherical the number distribution (Equation 1.3a) can be converted to surface area, volume and mass distributions by multiplying the number at
each diameter by \( \pi D_p^2, (D_p^3 \pi / 6) \) and \((D_p^3 \rho_p / 6) \) respectively and integrating over size. We assume a density \( \rho_p \) of 2.2 g/cm\(^3\) for dry sea-salt particles [Hess et al., 1998].

From the area of a sphere the aerosol surface area distribution can be calculated as:

\[
\frac{dS}{d \log D_p} = \frac{dN}{d \log D_p} \left( \frac{\pi D_p^2}{6} \right)
\]  (1.3b)

\[
S = \int_{-\infty}^{\infty} \left( \frac{dS}{d \log D_p} \right) d \log D_p = \int_{-\infty}^{\infty} \left( \frac{dN}{d \log D_p} \right) \left( \frac{\pi D_p^2}{6} \right) d \log D_p
\]  (1.3c)

Similarly, the aerosol volume distribution can be calculated from:

\[
\frac{dV}{d \log D_p} = \frac{dN}{d \log D_p} \left( \frac{\pi D_p^3}{6} \right)
\]  (1.3d)

\[
V = \int_{-\infty}^{\infty} \left( \frac{dV}{d \log D_p} \right) d \log D_p = \int_{-\infty}^{\infty} \left( \frac{dN}{d \log D_p} \right) \left( \frac{\pi D_p^3}{6} \right) d \log D_p
\]  (1.3e)

The aerosol mass distribution can be derived from the volume and density \( \rho_p \) of the particle.

\[
\frac{dM}{d \log D_p} = \frac{dN}{d \log D_p} \left( \frac{\pi D_p^3}{6} \rho_p \right)
\]  (1.3f)

\[
M = \int_{-\infty}^{\infty} \left( \frac{dM}{d \log D_p} \right) d \log D_p = \int_{-\infty}^{\infty} \left( \frac{dN}{d \log D_p} \right) \left( \frac{\pi D_p^3 \rho_p}{6} \right) d \log D_p
\]  (1.3g)

Because particle area and volume (mass) are proportional to the square and the cube of the diameter respectively, a few large particles can contribute significantly to the total area and volume (mass). Larger particles that contain the bulk of the total volume have relatively quick settling velocities; consequently much of the sea-salt mass has a short residence time in the MBL and is quickly deposited back into the ocean.
1.4 Sea-Salt Aerosol Generation Function:

The sea-salt aerosol generation function describes the surface flux of sea-salt aerosol, which can be defined as the number of sea-salt particles produced per unit of ocean surface area per unit time. Andreas [1998] reviewed a collection of currently used source functions available in literature. It was shown that the various estimates of the sea spray generation function in terms of the volume flux at any given radius vary by six orders of magnitude for a 10 m wind speed of 15 m/s (Figure 1.25). While the individual particle generation mechanisms are relatively well understood, the sea-salt aerosol number source flux remains uncertain. This has been in part due to limitations in instrumentation for measuring dry sea-salt particles with $D_p < 0.1 \mu m$.

Our measured refractory (heated to 360°C) aerosol size distributions from coastal breaking waves will be used to establish the aerosol source generation function under 100% breaking wave bubble coverage. The derived generation function will be valid for sea-salt particles between $0.01 \leq D_p \leq 8.0 \mu m$. This function will be scaled to oceanic fractional whitecap coverage and compared to other currently used functions available in literature (§ Appendix A). Particles larger than a few micrometers are less well characterized here and can comprise most of the sea-salt mass, but have negligible influence on the number flux. In this work sea-salt aerosol are assume to be spherical.
1.5 Methods of Determining Size-Dependent Aerosol Production Fluxes:

Numerous approaches have been developed from previous efforts to derive a complete sea spray generation function; two of the most commonly used methods are briefly described below:

**Whitecap Method** - This method determines the size dependent sea-salt production flux from measurements of laboratory-simulated whitecaps (e.g. Monahan et al., 1986, Mårtensson et al., 2003) or from surf zone aerosol generation (e.g. de Leeuw et al., 2000). The oceanic sea-salt aerosol production flux is calculated by scaling these measurements with field observations of the oceanic whitecap ratio $W$ (§ 3.2), the fraction of the sea surface covered by whitecaps. This approach assumes that the size-dependent sea-salt aerosol production flux per unit white area is independent of the nature and extent of the white area and of its means of production. Hence, the size dependent sea-salt aerosol production is the same for oceanic, laboratory and active surf zone whitecaps. Three variations of the whitecap method (i.e. discrete laboratory, continuous laboratory and surf zone whitecap method) are discussed in greater detail in section 4.1.

**Steady State Dry Deposition Method** - The steady state dry deposition method determines the size-dependent sea-salt number concentration from field measurements at a given reference height (usually $\approx$10 m) and modeled values of the size-dependent dry deposition velocity (e.g. Smith et al., 1993). To use this method it is essential to assume that the local sea-salt production flux and removal flux by dry deposition are equal and opposite during the time and location of measurement.
Investigations employing these four methods (i.e. Monahan et al., [1986], Mårtensson et al., [2003], de Leeuw et al., [2000] and Smith et al., [1993]) to quantify the sea spray generation function are discussed in greater detail in Appendix A. The first two whitecap methods are restricted due to scaling difficulties, and all four are restricted by difficulties associated with the measurement of large drops. Due to these measurement difficulties, the derived function in this work will only be compared with previous investigations concerned with bubble-mediated production mechanisms (i.e. film and jet droplet production).

1.6 Estimating a Sea-Salt Flux from Coastal Breaking Waves:

There are a number of factors that need to be considered that influence the variability of sea-salt aerosol concentrations. Since wave breaking is an irregular process, it is expected that bubble concentrations and subsequent sea spray generation will naturally fluctuate with time. The production of the coastal breaking wave bubble spectrum is highly variable due to a variety of environmental and meteorological factors such as surfactant concentration, viscosity, Langmuir circulations, turbulence, wave breaking characteristics influenced by fetch and swell, atmospheric thermal stratification [de Leeuw et al., 2003], sea surface temperature and salinity [Mårtensson et al., 2003]. The strength of many of these factors has not been adequately determined due to measurement difficulties. The following is a description of various factors that may influence marine aerosol production and concentration. These factors are summarized in Table 1.6.1 at the end of this section.
Wind Direction

Wind direction is mainly a transportation issue, however particle concentrations can also be influenced by wind direction. As the angle of transport becomes more parallel to the shoreline more particles have the opportunity to be transported to the instrumentation, thus artificially inflating the measured concentration. Due to the shorter time over the surf zone, perpendicular onshore flow allows for a more representative measurement of coastal concentrations.

Wind Speed

Processes governing breaking waves in the open ocean (deep water waves) are different from those in the surf zone (shallow water waves). Since waves do not break in deep water, the visible whitecaps on oceanic waves are created by high winds breaking their crest tip because the wind is driving the wave forward faster than the initial wave speed. In short, wind speed must be greater than wave speed if energy is to be transferred. However, this is not the case for coastal breaking waves. When coastal wave breaking occurs it is due to the interaction between the wave field and topography and is not directly driven by the wind field.

Because coastal breaking waves are forced by topography the near surface aerosol concentrations will exhibit a different dependence on wind speed compared to the open ocean. During high onshore winds more air passes over the surf zone per unit time, thus diluting the airmass resulting in lower measured particle concentrations at the tower site [Clarke and Kapustin, 2003]. Oceanic background sea-salt concentrations are not
significantly affected by the coastal winds, however high wind speeds over the open ocean will increase the background concentration. In the surf zone wind primarily plays the role of transport and dilution rather than particle generation like in the open ocean.

**Sea-Surface Temperature and Salinity**

Mårtensson et al. [2003] found that their measured bimodal number distribution behaved differently with variations in salinity and water temperature. Bubbles were generated in water at different temperatures and salinities in a closed bubble chamber by pumping filtered air through a submerged sintered glass filter. In this controlled setting they were able to investigate the effects of water temperature and salinity on the bubble and aerosol spectra. Increasing water temperature resulted in a decrease in the number concentration for \( D_p < 0.07 \) and an increase for \( D_p > 0.35 \) μm (Figure 1.61). Furthermore, [Mårtensson et al., 2003] investigation of the effects of salinity on the dry number concentration suggests different formation processes for \( D_p \) smaller and larger than 0.2 μm Figure (1.62). For \( D_p < 0.2 \) μm salinity only affects the residual dry aerosol particle size not the original droplet formation, for \( D_p > 0.2 \) μm the droplet production is affected by salinity. Higher salinities will increase the concentration of particles with \( D_p > 0.2 \) μm while smaller \( D_p \) show little change when the salinity is varied from 9.2% to 33%. While this variation in droplet formation and subsequent particle production is most likely a result of modifying the surface tension, the individual mechanisms responsible are unclear.
Surfactant Concentration

A surface-active agent, or surfactant, is a substance that when added to liquid has the ability to reduce the surface tension of that liquid. The single largest natural source of surfactants in the world’s oceans is primarily thought to be production by phytoplankton, which gives off natural surfactants as metabolic byproducts [Zutic et al., 1981]. This buildup of organic molecules decreases the number of hydrogen bonds at the ocean surface, thereby lowering the surface tension of the water. This is important because surface tension determines the tendency for surfaces to establish contact with one another. In short the lower the surfactant concentration the higher the surface tension, resulting in a stronger attraction between the molecules within the liquid.

Ironically, a lower (higher) surface tension will prolong (shorten) the lifetime of a bubble floating on the ocean surface. Since surface tension will reduce the surface area of a liquid this film contracts and compresses the air inside, the magnitude of this compression is determined by the surface tension. This compressed air will break through the thin water film and rupture the bubble. However, if the surface tension is low the water film will be more elastic and the bubble will float on the ocean surface longer (Munzing Chemie Performance Chemicals, 2001).

Surfactants could potentially play a major role in the size and lifetime of bubble populations in the ocean by modifying the surface tension of the bubble film. The presence (absence) of surfactants lowers (increases) surface tension, extends (shortens) bubble lifetime and allows (prevents) bubbles to merge easily, increasing the number of
large \textit{(small)} bubbles, droplets and sea-salt aerosol. More laboratory and field experiments focusing on the effects of surfactants on the breaking wave bubble spectra are needed. However, dissolved surfactant concentrations are difficult to measure. Including a surfactant concentration parameter to the surface source function would have the effect of introducing a variable surface tension term to the assumed bubble population.

Table 1.6.1. Processes Affecting Sea-Salt Aerosol Particle Production and Measurement

<table>
<thead>
<tr>
<th>Process</th>
<th>Variation</th>
<th>Effect</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wind Direction</td>
<td>On Shore</td>
<td>Measured Concentrations More Representative of Surf Zone Production</td>
</tr>
<tr>
<td></td>
<td>Along Shore</td>
<td>Additional Particles Detected from Sources Adjacent to Sampling Station (\rightarrow) Artificial Increases Measurements</td>
</tr>
<tr>
<td>Wind Speed</td>
<td>High</td>
<td>Dilution of Breaking Wave Plume (\rightarrow) Lower Measured Particle Concentration</td>
</tr>
<tr>
<td></td>
<td>Low</td>
<td>Decrease in Near Surface Particle Transport</td>
</tr>
<tr>
<td>Sea Surface Temperature</td>
<td>High</td>
<td>Decrease Concentration for (D_p &lt; 0.07 \mu m)</td>
</tr>
<tr>
<td></td>
<td>Low</td>
<td>Increase Concentration for (D_p &gt; 0.35 \mu m)</td>
</tr>
<tr>
<td>Salinity</td>
<td>High</td>
<td>Increase Concentrations for (D_p &gt; 0.2 \mu m)</td>
</tr>
<tr>
<td></td>
<td>Low</td>
<td>Little Change in Concentrations (D_p &lt; 0.2 \mu m)</td>
</tr>
<tr>
<td>Surfactant Concentration</td>
<td>High</td>
<td>Lower Surface Tension (\rightarrow) Extends Bubble Lifetime (\rightarrow) Bubbles Merge Together Easily (\rightarrow) Increase Large Bubble Population/Droplets/Particles</td>
</tr>
<tr>
<td></td>
<td>Low</td>
<td>Higher Surface Tension (\rightarrow) Shorten Bubble Lifetime (\rightarrow) More Difficult to Make Bubbles (\rightarrow) Increase Small Bubble population/Droplets/Particles</td>
</tr>
</tbody>
</table>

From the few processes mentioned in Table 1.6.1 it is evident that a great number of processes influence the production and transport of sea-salt aerosol; the complete source flux equation will be a complex function of such parameters. However, future research
will determine which of these parameters should be considered in the development of the complete source function that will be used to accurately model the effects of sea spray aerosol.
II. Measurements:

2.1 Coastal Experiments at Bellows Air Force Station:

Coastal aerosol production was investigated from datasets collected during the Shoreline Environment Aerosol Study (SEAS, 21 - 30 April, 2000) [Clarke et al., 2003] and similar measurements were taken from 25 January to 8 February 2004. The University of Hawaii Coastal Aerosol Facility at Bellows Air Force Station (BAFS, 21°21' N, 157°42' W) located on the windward coast of Oahu, Hawaii (Figure 2.11) has a 20 m tower located about 37 m inland and is oriented into the prevailing trade winds (Figure 2.12a). This site offered a stable environment for marine aerosol study and ensured that instrument performance would not be influenced by excessive aerosol variability. The beach slopes gradually into the ocean for ≈35 m to where the water depth is roughly 1.5 - 2 m. All instruments (§ 2.2) sampled the same airmass that was collected through inlets located at 5, 10, 15 and 20 m on the tower. Airmass sampling cycled through each inlet at regular intervals to reveal the temporal and vertical variation in marine aerosol properties.

An A-frame ladder was also located on the beach (Figure 2.12b) facing the onshore flow about half way between the tower and shoreline (17 m) and equipped with an inlet that could be varied between 1 - 5 m over the beach to establish the near surface aerosol vertical gradient generated by coastal breaking waves. Measurements were made continuously at 1 m intervals over a period of 30 minutes taking 2.5 hours to complete one profile. Data were collected 5 minutes after changing the inlet position, allowing the sampled volume to stabilize.
2.2 Instrumentation:

An assortment of instruments was used during the two campaigns to establish the size distribution of aerosols generated from coastal breaking waves, as well as to formulate a coastal generation flux for sea-salt aerosol that can be applied to oceanic conditions. All instruments sampled the same airmass collected through inlets mounted at various heights on the tower. A complete description including sampling procedures and loss corrections can be found in Clarke et al. [2003]; a brief summary follows.

Condensation Nuclei (CN) Counters

Two condensation nuclei (CN) counters (TSI Inc. 3010, 50% detection at 0.012 \( \mu \)m) simultaneously recorded particle number concentrations. One operated at 50°C (CN Cold) while the other collected data after heating the sample volume to 360°C (CN Hot) to burn off volatile components such as sulfates (Figure 2.21), leaving only a residual, refractory aerosol (i.e. sea-salt or soot). Particles entering the CN counter are grown to an optically detectable size by passing through a region that is supersaturated with butanol vapor. Particles are counted as they travel through the optical chamber where a light source is disrupted by the particles passing between it and a photodetector. The CN Hot concentration indicates the total sea-salt particles present in the Aitken and accumulation modes. Values for CN Hot have also been corrected for thermophoretic losses determined to be about 10%. Thermophoresis is the tendency of particles in a temperature gradient to move from hotter to colder regions due to molecules colliding with the particle on its “hot” side [Seinfeld and Pandis, 1998].
Nephelometer

Scattering coefficients ($\sigma_{sp}$) from all particles was measured by a three-wavelength (450, 550, 700 nm) integrating nephelometer (TSI Inc. 3563) over an angular range from about 7° - 173°. Truncation errors from angular nonideality of the instrument were corrected according to Anderson and Ogren [1998] specifications. A single wavelength (530 nm) Radiance Research (M903) nephelometer was also used to capture the near surface scattering gradient above the beach and have been corrected for large particle impaction losses following Porter et al. [2000].

Radial Differential Mobility Analyzer (RDMA)

In order to measure refractory particles (0.007 ≤ $D_p$ ≤ 0.250 μm) thermal analysis was used in conjunction with the RDMA. The RDMA measures aerosol size information based on particle mobility in an electric field [Zhang et al., 1995]. Sampled aerosols (RH ≈ 25%) are charged with a Boltzmann charge distribution prior to heating and particles with mobilities in a certain narrow range are detected at the sampling slit under the influence of a given voltage. After scanning the voltage range the associated particle counts are inverted mathematically to obtain a size distribution. Due to the diffusive loss of smaller particles and the charging efficiency of larger particles only aerosols in the size range 0.01 ≤ $D_p$ ≤ 0.15 μm were considered in this analysis [Clarke et al., 2003].

Laser Optical Particle Counter (OPC)

A modified 256 channel laser optical particle counter (OPC) spectrometer (Particle Measurement Systems LAS-X, Boulder, CO) with optional thermal analysis (360°C) to
resolve volatile and refractory (i.e. sea-salt) [Clarke, 1991] components was used to measure particles covering \(0.1 \leq D_p \leq 7.0 \mu m\) [Clarke et al., 2003]. Polystyrene latex spheres (PLS) and glass beads of known diameters and refractive index were used during calibration of the OPC. Thus, the measured particle diameters are effective optical sizes (i.e. size of a PSL sphere that scatters the same amount of light as the aerosol particle assuming both refractive indices are the same).

In an OPC a collimated HeNe laser (632 nm) is directed into a viewing volume where aerosols pass and scatter the incident laser beam. The amount of light scattered from a single particle is measured by a photodetector. The particle size distribution is then interpreted from the measured intensity of the scattered light. The light power that a particle scatters is a function of its size, refractive index and shape [Gebhart, 1993]. During SEAS the heating and tubing system used prevented the efficient transfer of particles to the OPC for \(D_p\) roughly above 0.75 \(\mu m\). Thus only those measurements in the size range \(0.1 \leq D_p \leq 0.75 \mu m\) will be considered for this study [Clarke et al., 2003].

**Aerodynamic Particle Sizer (APS)**

As particles \((0.5 \leq D_p \leq 12 \mu m)\) flow through the APS (TSI Inc. 3320, 3321) they cross the path of a two-laser system, momentarily obstructing the beams and causing a light pulse. These light pulses are detected by a photomultiplier, which in turn measures the time between the detected pulses. The particle velocity is then compared to a calibration curve and translated into an aerodynamic diameter. These translated velocities were converted from aerodynamic to geometric diameters assuming a density for wetted sea-salt at the measured APS RH [Tang et al., 1997]. Internal heating from the APS (30 -
32°C) resulted in an RH around 50%, but above the efflorescence point of sea-salt [Clarke et al., 2003]. Due to poor inlet efficiency for large particles only APS measurements from \(0.75 \leq D_p \leq 8.0\) μm will be used.

All size distribution data were adjusted to dry diameters \((D_p)\) corresponding to an RH = 40% using the accumulation mode [Swietlicki et al., 2000] and sea-salt [Tang et al., 1997] growth factors. Sizing measurements of the RDMA and APS overlap with the lower and upper limit of the OPC size range respectively. A cutoff \(D_p\) for each instrument was chosen as the midpoint between overlapping regions yielding the best statistics over each size range. The combined size distribution provides information over the following ranges \(0.007 \leq D_p \leq 0.15\) μm (RDMA), \(0.15 \leq D_p \leq 0.75\) μm (OPC), and \(0.75 \leq D_p \leq 8.0\) μm (APS). The DMA portion of the number distribution is good to about 15% while the OPC and APS data were compared to light scattering and agreed within flow uncertainties of about 5-10% [Clarke 2004, personal communication].

**Meteorological Data**

Meteorological conditions experienced during the SEAS campaign (Figure 2.22) were typical of the mild spring season on Oahu. Temperature, relative humidity, atmospheric pressure, wind speed and direction were recorded on the tower at a height of 10 m. Variables most relevant to coastal aerosol concentrations and measurements are wind speed and direction (§ 1.5). Onshore trade wind flow, associated with a high pressure system North of the island chain, was well developed and had a mean around 7.3 m/s at an angle roughly 81° to the tower. The air temperature was around 22°C and it is assumed
the sea surface temperature was within the climatological range of 24 - 27°C [Flament et al., 1996].

2.3 Results of Coastal Experiments: Linking Surf Zone Particle Production and Coastal Measurements

Profile measurements from the ladder system on the beach revealed a strong vertical gradient in CN Hot and scattering (Figure 2.31) between the surface and 5 m. Comparison with the tower measurements showed a gradual decrease up to 10 m with little change to 20 m. This scattering profile was very similar to that derived from prior vertical LIDAR measurements by Porter et al. [2003] (Figure 2.32) along the coast at BAFS. Hence, the surf zone strongly influenced the near surface aerosol plume dominating coastal refractory CN and scattering. These findings revealed that measurements made at 20 m were more characteristic of oceanic values free of influences from coastal particle production, while those below 5 m were dominated by particles recently generated from coastal breaking waves [Clarke et al., 2003]. Therefore, the difference between the 5 m and 20 m number distributions measured at the tower enabled us to directly assess the influence of coastal breaking waves on aerosol generation as a function of particle size.

This is clearly demonstrated in Figure (2.33), a time series of well-defined breaking wave events observable in corresponding excursions in refractory CN and scattering after subtracting the background values measured at 20 m on the tower. Wide overlapping peaks like those between 350 - 425 s were consistent with variable production from multiple waves breaking along the shore. The period of the breaking wave aerosol pulses
resulted from the passage of each breaking wave to the beach (Figure 2.34) roughly over a period of 22 s. Initially the wave broke and plunged to produce the wave front that generated the leading concentration of each pulse. The concentration increased as the bubble coverage of the wave front and surfacing bubbles behind it increased, until a maximum bubble density was achieved. As the wave moved to shore the width of the breaking wave front and bubble density behind the front diminished with time, decreasing the resulting aerosol production and concentrations.

The highly correlated ($r = 0.95$) excursions in breaking wave CN Hot and scattering (Figure 2.33) was significant because small particles ($D_p \leq 0.1 \, \mu m$) dominated excursions in number, while larger particles ($0.3 \leq D_p \leq 1.0 \, \mu m$) dominated scattering. Hence, the breaking wave process produced all sizes proportionally across the entire distribution. To ensure these 9 minute data were not anomalous events, similar analysis conducted over longer periods resulted in a mean $r = 0.85$. Nearly equal excursions in CN Cold and CN Hot concentrations from breaking waves were consistent with refractory sea-salt particles being produced in the Aitken and accumulation modes [Clarke et al., 2003]. Once the background concentrations were subtracted, the mean heated breaking wave size distribution (Figure 2.35) revealed that particles produced from coastal breaking waves ranged $0.01 \leq D_p \leq 8.0 \, \mu m$ and were refractory at 360°C. The majority (60%) of particles produced by coastal breaking waves had $D_p \leq 0.1 \, \mu m$, while larger particles ($0.3 \leq D_p \leq 1.0 \, \mu m$) that dominated scattering comprised only about 12% of the total number concentration.
Through size distributions and vertical profiles obtained at BAFS we have demonstrated that waves breaking in the surf zone substantially influenced coastal sea-salt aerosol concentrations and scattering, especially in the lowest 5 m. Furthermore, these particles spanned the entire detectable size range (0.01 \( \leq D_p \leq 8.0 \) \( \mu \)m). Thermal analyses of these distributions revealed similar breaking wave distributions for aerosol heated to 360°C suggesting that these were refractory sea-salt particles down to 0.01 \( \mu \)m [Clarke et al., 2003].
III. Formulation of Source Function:

If we can relate the measured refractory size distributions and concentrations to the breaking wave bubble coverage we can derive a function describing the sea-salt aerosol generation flux produced from coastal breaking waves that extends well into the submicrometer region. We assume that the primary generation mechanism of sea-salt aerosol is the bursting of bubbles resulting from breaking waves. Generalization of this flux to oceanic conditions requires that we also assume it can be scaled appropriately through an empirical expression such as that derived by Monahan et al. [1986] relating whitecap coverage to wind speed.

3.1 Deriving the Sea-Salt Flux:

The vertical gradient measured at BAFS (Figure 2.31) was a combination of measurements between 1 - 5 m over the beach and 5 - 20 m on the tower. These measurements demonstrate that CN Hot concentrations and scattering at 4 m above the beach is comparable to that measured at 5 m on the tower. Presumably the LIDAR cabin located at the base of the tower obstructs onshore flow and mixes near-surface air up to 5 m on the tower. Hence, variations in tower measurements at 5 m reflect variations in coastal breaking wave production. Beach profiles measured in conjunction with 5 m tower data revealed that the 5 m values, after subtracting background values measured on the tower, could represent the mean scattering and CN Hot concentration of the near surface breaking wave plume when multiplied by a factor \( k \) of 2.25. Values at 5 m, after subtracting background values measured at 20 m, are associated with production solely from coastal breaking waves. The sea-salt generation function \( (N_{100}) \), which is defined as
the number of sea-salt aerosols generated per unit area of ocean surface covered by bubbles (100% coverage) per unit time can be obtained from the mean refractory CN excursions measured at 5 m on the tower \( \langle CN \rangle \) for the average bubble coverage that gives rise to it. Consequently, shoreline measurements of \( N_{100} \) at BAFS can be appropriately scaled by oceanic bubble coverage to estimate oceanic sea-salt fluxes.

**Bubble Coverage**

The total breaking wave sea-salt particle concentration measured at BAFS is produced from bubbles rising to the ocean surface both in front of and behind a breaking wave moving toward shore. Digital video of incoming wave sets taken with a Cannon PowerShot S110 from various positions above the beach was used to estimate the bubble coverage of the wave front and the area of surfacing bubbles behind the advancing wave (Figure 2.34). This estimate was based upon relative "whiteness" in the image relative to the breaking wave front. The initial width of the wave front measured from back to front of wave \( w_o \approx 2 \) m was clearly defined and is assumed to be 100% bubble coverage. On average, the initial breaking point for a wave was measured to be \( L \) 35 m from the shoreline (Figure 2.34) and took roughly \( \tau \) 22 s \((\pm 2\) s\) to reach the beach. From this we conclude the mean wave speed \( \langle V_{wave} \rangle \) was about 1.6 m/s. During advancement to shore the initial wave front diminished linearly with time \( F_{avg} \), but rarely completely dissipated. Behind the advancing wave is an irregular, but extended area of rising bubbles. As the wave makes its way to shore, the area between it and its initial break point increases, while the fraction of that area covered by bubbles gradually decreases. Visual examinations of similar images suggest that the mean percentage of the surf zone
occupied by bubbles over the total distance traveled by the wave \( (A_{avg}) \) is roughly 40% in addition to the bubble coverage of the wave front. Hence, the mean area covered by bubbles for a breaking wave can be expressed as \[ \left( A_{avg} L + \frac{w_o}{2} \right) dl \]. Where \( dl \) is an element of the length along the breaking wave.

If one visualizes an effective “box” of height, \( h \), above an individual breaking wave in the surf zone then the total sea-salt production can be calculated by integrating the CN produced over the total time required for a wave to break and reach shore \( (\tau) \). As the wave breaks sea-salt aerosols are ejected upward and introduced into the box through an effective surface area determined by the difference in the mean wind and wave speed \( (\vec{V}_{wind} - \vec{V}_{wave}) \) multiplied by the total lifetime of a breaking wave \( (\tau) \) for an incremental length along the wave front. This is given as the numerator in Equation (3.1a). We assume negligible particle loss since deposition velocities for the size range that dominates particle number are very low over the short transit time \(< 10 \text{ s}\) from initial production to detection.

Equation (3.1a) relates the number of sea-salt aerosols produced in the surf zone per unit area (100% bubble coverage), per unit time to \( CN \) measured at 5 m for coastal breaking wave plumes; and after simplification (§ Appendix B) \( N_{100} \) can be written as:

\[
N_{100} = \frac{[\text{Total # of CN into Box}]}{[\text{Total Bubble Area}]\tau}
\]
\[ N_{100} = \frac{[CN][k][V_{\text{wind}} - V_{\text{wave}}][h][\tau]}{\left[ (A_{\text{avg}})[L] + \frac{W_o}{2} \right]} = \frac{[CN][k][V_{\text{wind}} - V_{\text{wave}}][h]}{\left[ (A_{\text{avg}})[L] + \frac{(F_{\text{avg}})[W_o]}{2} \right]} \]  

(3.1a)

Where:

- \( \bar{CN} \) = Measured mean refractory CN excursions at 5 m (40 cm\(^{-3}\) ± 6 cm\(^{-3}\))
- \( k \) = Constant of proportionality; multiplier for gradient scaling (2.25 ± 0.45)
- \( V_{\text{wind}} \) = Mean wind speed in the surf zone (7.3 m/s ± 1.1 m/s)
- \( V_{\text{wave}} \) = Mean wave speed in the surf zone (1.59 m/s ± 0.19 m/s)
- \( h \) = Height of plume layer (4 m ± 0.6 m)
- \( A_{\text{avg}} \) = Mean bubble coverage for area between waves (40% ± 12%)
- \( L \) = Total distance wave travels to shore (35 m ± 3 m)
- \( F_{\text{avg}} \) = Width of wave front as fraction of \( w_o \) (0.5)
- \( w_o \) = Initial width of breaking wave bubble front (2 m ± 0.5 m)
- \( \tau \) = Total lifetime of a breaking wave (22 s ± 2 s)

The measured \( \bar{CN} \) depends on the difference in \( V_{\text{wind}} \) and \( V_{\text{wave}} \) and \( A_{\text{avg}} \) under given breaking wave conditions, but for the data and conditions compared here a value of 40 cm\(^{-3}\) was found to be representative. Uncertainties in \( V_{\text{wind}} \) and \( \tau \) were calculated as the standard deviation of the measured quantity while \( L, V_{\text{wave}} \) and \( w_o \) were estimated from physical measurements in the surf zone. Repeated measurements of profiles on the beach (Figure 2.31) suggest uncertainties in \( k \) and \( h \) of 20% and 15% respectively. The uncertainty for \( A_{\text{avg}} \) was estimated from the variations in wave sets captured by digital imagery. An overall uncertainty of 44% was calculated (§ Appendix C) from the accumulated uncertainties of the 10 terms that make that make up \( N_{100} \).

We recognize that this flux may be sensitive to other meteorological and environmental factors that may influence production of coastal CN. Some of these include, sea surface temperature, salinity [Mårtensson et al., 2003] and surfactant concentration. However,
the effects of these variables are currently not well understood and are difficult to quantify.

3.1.1 Description of Terms in $N_{100}$:

This section will outline the terms that contribute to $N_{100}$ and how the individual uncertainties were calculated in more specific detail. All uncertainty calculations can be found in Appendix C. As presented graphically in Figure (3.12) an overall uncertainty of 54% was calculated from the accumulated uncertainties of the all terms in $N_{100}$.

Measured Mean Refractory CN Excursions at 5 m ($\overline{CN}$)

Values for excursions in CN at 5 m are unique to the BAFS field site. It is expected that these values would vary depending on the individual coastal sampling location and upon different dilution caused by different wind speeds. For instance, BAFS has a gradually sloping beach that is covered with sand, while much of the Oahu coast is characterized by an irregular reef structure. Wave breaking characteristics can greatly differ at each site, therefore if similar measurements were taken at two different locations one would expect to measure different values for $\overline{CN}$ however. We assume that all bubble patches, independent of their size or means of production, have the same instantaneous production flux of sea-salt aerosol for all particle sizes. Hence $\overline{CN}$ must be related to specific measurement situations. Values for $\overline{CN}$ have also been corrected for thermophoretic losses (§ 2.2).
The mean refractory CN excursion (e.g. Figure 2.33) is needed in order to normalize the mean breaking wave number distribution (Figure 2.35) to the specific measurement conditions experienced during the SEAS campaign. The $\overline{CN}$ was calculated using the Pick Peaks function available in the OriginPro 6.1 software package. It is particularly useful when the data do not seem to have a definable baseline. This process requires the user to define a set of parameters, which will be used to determine if a peak is present in the data under the defined conditions. Parameters were chosen so that the program would find all maximum and minimum points for the desired period. The mean refractory CN excursion was determined to be 40 cm$^{-3}$. An example of the output from this process for the same 9 minute period investigated in Figure (2.33) is shown in Figure (3.13).

Now that we know the $\overline{CN}$ during SEAS we can normalize the mean breaking wave number distribution obtained during SEAS (Figure 2.35, Clarke et al., 2003) to reflect the specific measuring situation during our measurement periods. This was achieved by multiplying it by the ratio of its integral number (i.e. 27 cm$^{-3}$) and our mean breaking wave number deviations, $\overline{CN}$ (i.e. 40 cm$^{-3}$). In this way, the number flux obtained from this deviation in breaking wave CN can be represented as a size distribution flux.

**Constant of Proportionality; Multiplier for Gradient Scaling ($k$)**

The constant $k$ is necessary in our source function because measurements were taken on the tower where flow perturbations existed due to the LIDAR cabin located at the base of the tower. Thus, we need to scale the breaking wave number and particle size distributions measured at 5 m on the tower to be representative averages for the near
surface breaking wave aerosol plume layer characterized over the beach (i.e. \( h \), the depth of the near surface breaking wave plume 0 - 4 m above the beach). In order to do this we must establish a multiplication factor (\( k \)) that links the 5 m tower measurements to the mean near surface breaking wave plume concentration. This was achieved by taking simultaneous measurements of CN Hot concentrations at 5 m on the tower and 1 - 4 m over the beach (Figure 2.31). The mean concentration of the near surface breaking wave plume layer was calculated after subtracting background values measured at 20 m on the tower. The magnitude of the breaking wave deviations seen at 5 m is comparable with that of the plume layer seen over the beach if they are multiplied by a factor of 2.25. An uncertainty of \( \pm 20\% \) was assigned to \( k \) based upon the standard deviation in the measurements of the near surface breaking wave plume (i.e. Figure 2.31). These measurements were recently repeated (June 2004) with two heated (360°C) CN counters (TSI Inc. 3760). One sampled through a garden hose inlet located at various positions on the tower while the other, using a similar garden hose inlet system, took simultaneous profiles over the beach. The resulting \( k \) was 2.20. Again within the uncertainty range for \( k \) originally presented as 2.25 \( \pm 0.45 \).

**Mean Wind** \( (\bar{V}_{\text{wind}}) \) **and Wave Speed** \( (\bar{V}_{\text{wave}}) \) **in the Surf Zone**

The mean wind speed was calculated from averaging measurements taken by the meteorological instrumentation located at 10 m on the tower. The uncertainty was calculated as the standard deviation of the measured quantity. The mean wave speed and its uncertainty was calculated from physical measurements of the distance a breaking wave travels to shore and the total time it took to reach the beach and dissipate. These
calculations are necessary because their difference determines the size of the bottom of the “box” which sea-salt particles are ejected into.

**Height of Plume Layer (h)**

The height of the plume layer (i.e. effective “box”) determines the magnitude of vertical mixing near the surface due to turbulent eddies. This vertical mixing over the beach is further complicated by the presence of the LIDAR cabin at the base of the tower Figure (2.21a). From profile measurements on the beach and tower it was concluded that particles produced in the surf zone mix up to about 4 m before entering the complex flow regime created by the LIDAR cabin and being intercepted at the tower. The height of the near surface plume layer was estimated to fluctuate ± 0.6 m. Comparison of excursions of CN Hot at the tower to those measured in the profile over the beach indicate that the datum at 5 m on the tower is effectively the same as 3.5 m above the beach.

**Mean Bubble Coverage for Area Between Waves (A_{avg})**

Digital video and still imagery from various positions on the tower and heights above the beach were used to estimate the mean bubble coverage as a percentage (A_{avg}) for a single breaking wave. It is expected that our measurements of breaking wave CN Hot are proportional to the relative bubble coverage present in the surf zone. These images were time paired with measurements of breaking wave CN to help determine the relative contribution of the bubbles produced by a breaking wave. Repeated analysis of incoming wave sets led to an estimated maximum coverage of 50% and a minimum of 30%
averaged over the total lifetime of a breaking wave. An uncertainty in $A_{av}$ of ± 12% was taken to estimate the upper and lower limit of surf zone bubble coverage.

While visually assessing the bubble population it became clear that a number factors influence the uncertainty in $A_{av}$. Some that were considered in the ± 12% include bubble density, camera resolution, available sunlight and azimuth. Due to limited camera resolution, bubble populations further behind the breaking wave front were almost indistinguishable from the ocean surface, while populations in and just behind the wave front were clearly recognized. Imagery closer to the ocean surface (which did not capture the entire surf zone) revealed that some bursting bubbles were present all the way up to the shoreline. This discrepancy between imagery taken at different altitudes is attributed to the variation in bubble density over the 35 m a breaking wave travels in the surf zone. In addition images of the breaking wave bubble field were much easier to resolve when the sun was overhead providing more light for reflectance. Consequently, our best estimate of $A_{av}$ above should be reexamined with improved technologies when they become available.

**Total Distance Wave Travels to Shore ($L$)**

At BAFS wave breaking typically occurred at the end of a rocky embankment that extends into the ocean like a pier perpendicular to the shoreline. A 200 ft. tape measure was used to measure the distance from the tower to shoreline and to the breaking point. An uncertainty of ± 3 m is assigned to $L$ because the coastal waves did not always break in the same exact location.
**Initial Width of Breaking Wave Bubble Front** (*w_o*)

The initial width of a breaking wave in the surf zone at BAFS was estimated from physical measurements and observations in the ocean. Here width is defined as the distance from the back to the front of the wave once breaking occurs.

**Width of Wave Front as Fraction of *w_o* (**F_avg**)**

*F_avg* stems from a simple estimate assuming a linear decrease in the breaking wave front width with time. Because most bubble coverage occurs behind the wave front the uncertainty associated with this linear assumption (*F_avg* = 0.5) has negligible effect on the final result.

**Total Lifetime of a Breaking Wave** (*τ*)

The lifetime of a breaking wave was measured on the beach at BAFS with a stopwatch. Particular attention was given to the exact moment breaking occurs and was kept consistent for each observation. The uncertainty of ± 2 s was calculated from the standard deviation of the 110 time measurements that were made over several days.

**Size Distribution**

As described above, the shape of the size distribution was obtained from SEAS measurements. Because it is normalized we are only concerned with uncertainties in the integral number that describe the average SEAS concentrations. Smaller aerosol sizes depend upon DMA sizing uncertainties that are estimated at about 10% while larger sizes
were shown to yield calculated optical that are also within about 10% of those measured during SEAS (Clarke et al., 2003). Hence, we assume that our use of the normalized SEAS distributions contributes about 10% to our flux uncertainty.

3.2 Parameterization and Scaling to Oceanic Conditions:

For practical purposes and to support future research with intended applications to aerosol modeling an equation describing the breaking wave surf zone generation flux would be useful. $N_{100}$, the integral number flux (§ 3.1), was used to scale the normalized refractory size distribution determined for breaking waves at BAFS (Figure 2.35). Comparison with independent measurements of total number (small particles) and calculated $\sigma_{sp}$ (large particles) during SEAS [Clarke et al., 2003] constrains the overall uncertainty of this mean normalized distribution to about 10%. This flux distribution was fit with a 5th order polynomial regression of the form

$$A_i = \beta_0 + \beta_1 D_p + \beta_2 D_p^2 + \beta_3 D_p^3 + \beta_4 D_p^4 + \beta_5 D_p^5$$  (3.2a)

for the three size intervals (57, 49 and 42 channels respectively) given in Table 3.2. The product of each term in Equation 3.2a results in units of $\mu m$ when $D_p$ is in $\mu m$. The coefficients are given to four significant figures to preserve the shape of the distribution. The mean number flux per log $D_p$ interval can be calculated by summation of Equation 3.2a for each size range. It was found that the indicated coefficients were able to reproduce the observed distribution within about 1% uncertainty (Figure 3.21).

$$\left( \frac{dF_N}{d \log D_p} \right)_{BW} = \sum_{i=1}^{3} A_i$$  (3.2b)
Table 3.2. Coefficients ($\beta_0 - \beta_5$) for $A$ in Equation 3.2b describing $N_{100}$ over three size intervals in $\mu m$

<table>
<thead>
<tr>
<th>Size Interval ($\mu m$)</th>
<th>$\beta_0$</th>
<th>$\beta_1$</th>
<th>$\beta_2$</th>
<th>$\beta_3$</th>
<th>$\beta_4$</th>
<th>$\beta_5$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$0.01 \leq D_p \leq 0.132$</td>
<td>$-7.502E^3$</td>
<td>$1.212E^6$</td>
<td>$-2.971E^7$</td>
<td>$3.283E^8$</td>
<td>$-1.716E^9$</td>
<td>$3.435E^9$</td>
</tr>
<tr>
<td>$0.132 \leq D_p \leq 1.2$</td>
<td>$5.78E^3$</td>
<td>$1.753E^4$</td>
<td>$-9.858E^4$</td>
<td>$1.505E^5$</td>
<td>$-9.610E^4$</td>
<td>$2.240E^4$</td>
</tr>
<tr>
<td>$1.2 \leq D_p \leq 8.0$</td>
<td>$6.74E^2$</td>
<td>$1.259E^3$</td>
<td>$-8.092E^2$</td>
<td>$1.828E^2$</td>
<td>$-1.820E^1$</td>
<td>$6.771E^1$</td>
</tr>
</tbody>
</table>

The derived mean breaking wave number flux (3.2b) can be scaled to oceanic conditions by multiplying it by the open ocean fractional whitecap coverage, $W$. An empirical expression (3.2c) derived by Monahan et al., [1986] describes whitecap coverage as a function of wind speed where $U_{10}$ is the mean wind speed at a height of 10 m (Figure 3.22), this function is described in further detail in Appendix A.

$$W = (3.84E^{-6})(U_{10}^{3.41})$$

(3.2c)

Multiplying Equation 3.2c by the normalized flux parameterization (Equation 3.2b) results in an expression (Equation 3.2d) describing the oceanic mean size-resolved number flux from the area of the ocean surface covered by whitecaps.

$$\left( \frac{dF_N}{d \log D_p} \right)_{oceanic} = W \sum_{i=1}^{3} A_i$$

(3.2d)

Equation 3.2d is valid for particles ranging $0.01 \leq D_p \leq 8.0 \mu m$ and can be converted to surface area, volume and mass fluxes by multiplying the appropriate factors (§ 1.3) and integrating over size.
IV. Discussion and Conclusions of Research:

4.1 Comparison With Current Source Functions:

Here I will compare the derived source function of this work with those from the four methods first mentioned in section 1.5. A thorough description of these techniques for each referenced investigator can be found in Appendix A. Because these functions were reported in different formats they will be presented here graphically as dry diameter ($D_p$) measured in micrometers. Particle dimensions were converted assuming the radius at formation ($r_o$) at 98% RH is approximately twice that at RH = 80% ($r_{80}$). Hence, our $D_p$ is numerically equivalent to $r_{80}$. For instance, a particle with a $r_o = 0.2 \mu m$ would have a $D_p = 0.1 \mu m$ and a $r_{80} = 0.1 \mu m$. For ease of comparison all number fluxes have been scaled to oceanic conditions corresponding to $U_{10} = 9 \text{ m/s}$ according to (3.2c) and have been converted to $\frac{dF_N}{d \log D_p}$ in $[\text{cm}^{-2} \text{s}^{-1}]$. The derived mean number flux and these published source functions described in Appendix A are presented graphically in Figure (4.11) over their reported applicable size range. It is of interest to compare the number, area, volume and mass fluxes for these various approaches. The total number flux can be calculated by a summation of the number flux over the total applicable size range:

$$dF_N = \sum_{i=1}^{146} \left( \frac{dF_N}{d \log D_p} \right)_i \left( \Delta \log D_p \right)_i$$

(4.1a)

where $\left( \Delta \log D_p \right)_i$ is the width of each size bin. This same process can be used to calculate the total surface area, volume and mass flux. The total calculated fluxes over all applicable dry sizes for each cited estimate is displayed in Table (4.11), all have been scaled to oceanic conditions under a $U_{10} = 9 \text{ m/s}$. Table (4.12) is the same as Table
(4.11), but fluxes derived in this thesis are calculated over the reported applicable size range of each referenced work.

Table 4.11. Total Fluxes Over the Entire Measured Size Ranges

<table>
<thead>
<tr>
<th>Parameterization</th>
<th>Size Range (µm)</th>
<th>$\frac{dF_N}{d \log D_p}$ [# / cm² s]</th>
<th>$\frac{dF_A}{d \log D_p}$ [cm² / cm² s]</th>
<th>$\frac{dF_V}{d \log D_p}$ [cm³ / cm² s]</th>
<th>$\frac{dF_M}{d \log D_p}$ [µg / m² s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Owens and Clarke</td>
<td>0.01 ≤ $D_p$ ≤ 8.0</td>
<td>91</td>
<td>133</td>
<td>86</td>
<td>189</td>
</tr>
<tr>
<td>Monahan et al. [1986]</td>
<td>0.302 ≤ $D_p$ ≤ 8.0</td>
<td>5</td>
<td>24</td>
<td>11</td>
<td>25</td>
</tr>
<tr>
<td>Mårtensson et al. [2003]</td>
<td>0.02 ≤ $D_p$ ≤ 2.88</td>
<td>69</td>
<td>29</td>
<td>8</td>
<td>17</td>
</tr>
<tr>
<td>Modified</td>
<td>1.0 ≤ $D_p$ ≤ 8.0</td>
<td>0.8</td>
<td>26</td>
<td>17</td>
<td>37</td>
</tr>
<tr>
<td>Smith et al. [1993]</td>
<td>0.398 ≤ $D_p$ ≤ 5.0</td>
<td>14</td>
<td>125</td>
<td>63</td>
<td>139</td>
</tr>
</tbody>
</table>

Table 4.12. Comparison of Fluxes From This Work With Other Investigators Over Applicable Size Ranges

<table>
<thead>
<tr>
<th>Size Range (µm)</th>
<th>Parameterization</th>
<th>Total Number [# / cm² s]</th>
<th>Total Area [cm² / cm² s]</th>
<th>Total Volume [cm³ / cm² s]</th>
<th>Total Mass [µg / m² s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.288 ≤ $D_p$ ≤ 8.0</td>
<td>Monahan et al. [1986]</td>
<td>5</td>
<td>24</td>
<td>11</td>
<td>25</td>
</tr>
<tr>
<td></td>
<td>Owens and Clarke</td>
<td>17</td>
<td>131</td>
<td>86</td>
<td>189</td>
</tr>
<tr>
<td>0.02 ≤ $D_p$ ≤ 2.88</td>
<td>Mårtensson et al. [2003]</td>
<td>69</td>
<td>29</td>
<td>8</td>
<td>17</td>
</tr>
<tr>
<td></td>
<td>Owens and Clarke</td>
<td>81</td>
<td>52</td>
<td>14</td>
<td>32</td>
</tr>
<tr>
<td>1.0 ≤ $D_p$ ≤ 8.0</td>
<td>Modified</td>
<td>0.8</td>
<td>26</td>
<td>17</td>
<td>37</td>
</tr>
<tr>
<td></td>
<td>Smith et al. [1993]</td>
<td>5</td>
<td>121</td>
<td>65</td>
<td>187</td>
</tr>
<tr>
<td></td>
<td>Owens and Clarke</td>
<td>14</td>
<td>125</td>
<td>63</td>
<td>139</td>
</tr>
<tr>
<td>0.398 ≤ $D_p$ ≤ 5.0</td>
<td>de Leeuw et al. [2000]</td>
<td>12</td>
<td>88</td>
<td>41</td>
<td>90</td>
</tr>
</tbody>
</table>

Monahan et al. [1986] estimated the generation flux using the discrete laboratory whitecap method. In this variation of the whitecap method, drop production is measured from laboratory-induced whitecaps created in a wave tank by the collision of two moving parcels of water. This method assumes that all whitecaps, independent of their size or
means of production, have the same instantaneous production flux of sea-salt particles for all sizes and that the area of all whitecaps decrease exponentially with the same characteristic decay time. One concern with this method is the extent to which natural wave-breaking processes can be replicated in a laboratory situation, and thus the resulting sea-salt flux distribution. Our derived source flux (Figure 4.11) agrees within an order of magnitude of Monahan et al. [1986] for the largest particles (3.0 \( \leq D_p \leq 8.0 \) \( \mu \)m), while much better agreement is achieved for smaller sizes (0.3 \( \leq D_p \leq 3.0 \) \( \mu \)m). The laboratory generated Monahan et al. [1986] flux underestimates the number source flux over its entire applicable size range by 12 cm\(^{-2}\) s (Table 4.12) compared to the present study. Details of the Monahan et al. [1986] wave tank experimental set up are vague. For instance there is no mention as to the origin of the water that was used during wave simulations. From the findings of Mårtensson et al. [2003] (§ 1.6), specific water properties and temperature during measurement could contribute to the discrepancy in Monahan et al. [1986] estimates. In addition the areas of the whitecaps that were simulated in the wave tank were reportedly small compared to that of typical oceanic whitecaps and the Monahan et al. [1986] function was derived from measurements taken in 1982. Particle counting techniques have also advanced considerably in the last 20 years.

Smith et al. [1993] employed the steady state dry deposition method (§ 1.5) to estimate the number flux for particles ranging 1.0 \( \leq D_p \leq 8.0 \) \( \mu \)m. This method determines the size-dependent sea-salt number concentration from field measurements at a fixed height and modeled values of the size-dependent dry deposition velocity based on that of Slinn and
This method assumes local production and dry deposition are in balance. Size distributions were measured at 14 m asl in the Outer Hebrides on the west coast of Scotland. A major concern with this formulation is that concentrations at the lower end of the Smith et al. [1993] applicable size range would not have attained their steady state values with respect to dry deposition. Assuming a local balance between production and removal is only reasonable for the largest drops as smaller particles have much longer residence times. This is revealed by the rapid decrease in concentration at $D_p \leq 2.5 \mu m$. Andreas [1998] suggests that the Smith et al. [1993] function is low by at least a factor of 3.5. He continues that Smith through personal communication, concurs his function may be low by a factor of 2 or 3. When the Smith et al. [1993] function is increased by a factor of 3.5 (Modified Smith et al., [1993]) it is shifted up to the tail end of Mårtensson et al. [2003] meeting at $D_p \approx 3.0 \mu m$ and giving the appearance of a combined distribution extending from $0.02 \leq D_p \leq 8.0 \mu m$ (Figure 4.11). The Modified Smith et al. [1993] is within an order of magnitude of our source flux for particles between $3.0 \leq D_p \leq 8.0 \mu m$. However, because of the unrealistic decrease in concentration for particles with $D_p \leq 2.5 \mu m$, which is not evident in other flux estimates, the Modified Smith et al. [1993] function still greatly underestimates the number flux over the size range (Table 4.12).

The premise of the surf zone variant of the whitecap method ($\S$ 1.5) used by de Leeuw et al. [2000] is that production of sea-salt aerosol particles in a surf zone is sufficiently similar to that of oceanic whitecaps and that the production flux in the surf zone can be equated to the flux over oceanic whitecaps. This method relies on measurements of the
change in the size-dependent sea-salt number concentration produced by surf zone breaking waves over some length \( L \) during onshore winds. This is done under the assumption that the sea-salt particles produced by the surf zone breaking waves are uniformly distributed throughout an air parcel of some height \( H \). Analogous to the discrete laboratory whitecap method, a concern with the surf zone variant is the degree of similarity between the oceanic breaking wave sea-salt flux distribution and that of the surf zone. de Leeuw et al. [2000] took concentration measurements upwind and downwind of the surf zone on the 10 m high and 350 m long Scripps pier in California. Measurements were assumed to representative of background and surf zone production respectively. Coastal breaking waves production was examined after subtracting the background concentration measured on the upwind side of the pier. Our derived source flux (Figure 4.11) compares very well to that of de Leeuw et al. [2000] over the entire applicable size range \( 0.4 \leq D_p \leq 5.0 \mu m \), which was collected on the coast of California using optical particle counters under similar conditions to that experienced during the present study. Note the number flux presented as Equation 4 in de Leeuw et al. [2000] contains an error. A factor of \( 10^6 \) is required for the expression presented by these investigators, their Equation 4, to yield the values shown in their Figure 7a. By the calculations represented in Table 4.12 it is clear that the de Leeuw et al. [2000] flux is in good agreement with that of this study over its entire applicable size range.

Using the continuous laboratory whitecap method Mårtensson et al. [2003] measured the size-dependent whitecap droplet production in a closed bubble chamber. Fundamental assumptions and concerns for this method are essentially the same as the discrete
laboratory whitecap method used by Monahan et al. [1986]. Bubble populations were generated by bubbling filtered air through a sintered glass filter that was submerged in synthetic seawater. The size-dependent drop production was measured at different salinities 0.0, 9.2, and 33.0‰ (Figure 1.52) and different water temperatures -2, 5, 15, and 23°C (Figure 1.51). Our flux estimate is most consistent with that of Martensson et al. [2003]. Their distribution, covering a wide range of particle sizes, was generated from bubble chamber simulations with dissolved synthetic sea-salt in pure Milli-Q water, similar to environmental conditions (salinity, water temperature) experienced at BAFS. The seasonal sea surface temperature range near Oahu is only about 3°C, from a low in the winter of 24°C to a high in the summer 27°C; additionally the mean salinity of the surrounding water is ≈35‰ [Flament et al., 1996]. Agreement between the two flux estimates is within a factor of two over the entire size range (Figure 4.11). Although the shape of Martensson et al. [2003] deviates slightly from this study for particles between 0.04 ≤ Dp ≤ 0.20 μm, similarities with our natural ocean breaking wave flux suggests that these processes are representative of breaking waves and our observed particles are likely to be sea-salt over all sizes. In addition the total number flux over the Martensson et al. [2003] size range agree surprisingly well (Table 4.12) considering the differences between laboratory and coastal environments.

4.2 Conclusions of Research:

Measurements during SEAS and those of early 2004 established refractory aerosol size distributions between 0.01 ≤ Dp ≤ 8.0 μm produced solely from coastal breaking waves based on distributions averaged over 600 hours representing the mean coastal breaking
wave sea-salt production. This distribution was combined here with assessments of near surface particle concentrations associated with the observed bubble coverage from coastal breaking waves to derive a source function for the size resolved number flux per unit of bubble covered surface. Once scaled to oceanic conditions the derived source function agrees favorably with several previously published estimates (e.g. Monahan et al., 1986, Modified Smith et al., 1993, de Leeuw et al., 2000 and Mårtensson et al., 2003). This source function extends particle production down to $D_p = 0.01 \mu m$, an order of magnitude smaller that previously characterized for natural oceanic breaking waves. Similarities in the distributions from the wave tank studies of synthetic sea-salt and our natural ocean waves suggest that these processes are representative of breaking waves and that our observed particles are likely to be sea-salt over all sizes. The extension of particle flux estimates down to these sizes provides a new parameterization for sea-salt aerosol that we hope will lead to new understanding of the production of sea-salt number concentration over the remote oceans.
Appendix A: Previous Flux Estimates

A.1 Description of Monahan et al. [1986] Source Function:

Monahan et al. [1986] derived one of the most widely recognized source functions describing the indirect production mechanisms of marine aerosol. To calculate the oceanic particle production Monahan et al. [1986] utilizes a combination of laboratory-simulated waves and observations of fractional whitecap coverage ($W$) in the ocean as a function of the wind speed at a height of 10 m ($U_{10}$).

To describe $W(U_{10})$, photographs of whitecaps taken during field observations aboard ship were investigated while values for the time it took a whitecap to decay were derived from wave tank simulations. Since analysis of $W$ is subjective some uncertainty is associated with calculations incorporating this value, but were not reported. The resulting empirical relation for the whitecap coverage (in %) as a function of wind speed is expressed as:

$$W = \left(3.84E^{-6}\right)U_{10}^{3.41}$$  (3.3c)

The wind speed dependence (Figure 3.22) illustrates that at higher wind speeds the production rate of sea-salt particles increase, due to the increase in whitecaps. An explicit expression that describes the indirect (bubble mediated) generation mechanisms (i.e. film and jet drops) was found to be:

$$\frac{dF_\phi}{dr_{80}} = 1.373U_{10}^{3.41} \cdot r_{80}^{-3} \left(1 + 0.057r_{80}^{1.05}\right) \cdot 10^{1.19\exp\left[-\frac{0.380-\log(r_{80})}{0.650}\right]^2}$$  (A.1a)

where $U_{10}$ is the wind speed at 10 m in m/s, $r_{80}$ is the particle radius at 80% RH in μm.
Equation (A.1a) models sea-salt aerosol production over the size range $0.3 \leq r_{80} \leq 10 \ \mu m$ ($r_{80} \approx D_p$) as a function of wind speed and particle size, $\frac{dF}{dr_{80}}$ has units of [m$^{-2}$ s$^{-1}$ $\mu m^{-1}$].

A.2 Description of Smith et al. [1993] Source Function:

Smith et al. [1993] measured marine aerosols produced from bubbles bursting within whitecaps at ambient humidity for radii ($r_{80}$) between $1 \leq r_{80} \leq 25 \ \mu m$ at wind speeds up to 30 m/s in the Outer Hebrides, off the west coast of Scotland in 1986. The derived sea-salt flux assumes that the particle concentration is under equilibrium. In this steady state assumption, the total loss of particles is replenished by a proportional new particle production. In order to validate the equilibrium assumption Smith et al. [1993] argues that the given size spectra segregated into different wind speeds regimes came from a substantial dataset comprised of some 700 hours of sampling. Size distributions for each wind speed regime came from averaging datasets under conditions where the atmospheric stability was near neutral or slightly unstable, and at a steady RH with a mean of 77%.

The total derived source flux is described by the summation of two lognormal distributions:

$$\frac{dF}{dr_{80}} = \sum_{i=1,2} A_i \exp \left[ -f_i \left( \frac{r_{80}}{r_{0i}} \right)^2 \right]$$

\begin{align*}
  f_1 &= 3.1 \mu m & r_{01} &= 2.1 \mu m \\
  f_2 &= 3.3 \mu m & r_{02} &= 9.2 \mu m \quad (A.2a)
\end{align*}

$$\log(A_1) = 0.0676U + 2.43$$
$$\log(A_2) = 0.959U^3 - 1.476$$
where \( r_{80} \) is the particle radius at 80% RH in \( \mu \text{m} \), and \( U \) is the wind speed at 10 m in m/s.

\[
\frac{dF}{dr_{80}} \text{ is given in } [\text{m}^{-2} \text{s}^{-1} \mu\text{m}^{-1}]. \text{ The coefficients } A_1 \text{ and } A_2 \text{ are highly dependent on wind speed; } \log(A_1) \text{ is associated with the jet drop mode and } \log(A_2) \text{ with the spume drop mode. The mode is related to the mode radius through } f_1, f_2, r_{01}, \text{ and } r_{02}. \text{ Meteorological parameters and size distributions were collected from a 10 m scaffolding tower that set approximately 14 m asl on the beach. Equation (A.2a) is valid over the size range } 1 \leq r_{80} \leq 25 \mu\text{m} \left( r_{80} \approx D_p \right). \text{ Fluxes obtained by this approach are only valid for particles that reach a minimum height equal to the measurement height (14 m for this experiment). The actual surface flux would be higher, thus Smith et al. [1993] is more representative of the surface flux lower limit.}

**A.3 Description of de Leeuw et al. [2000] Source Function:**

de Leeuw et al. [2000] derived a quantitative source function for sea spray produced by surf zone breaking waves from measurements taken during the EOPACE Surf-1, Surf-2, and Surf-3 experiments on the coast of California. They found that surf produced concentrations and plume heights vary with particle size and wind speed.

Surf-1 (January 24 - February 5, 1996) and Surf-3 (April 1 - 11, 1997) concentration measurements were taken by optical particle counters at the base and end of a 10 m high pier in La Jolla, San Diego. The Scripps Institution of Oceanography (SIO) pier reaches 350 m beyond the surf zone. Wind speed, direction, atmospheric pressure, air and water temperature were recorded by the SIO meteorological station located near the end of the pier. Wave data and video recording were also made by SIO. Background aerosol
concentrations were measured at the end of the pier 11 m above mean water level. This background concentration is assumed to be well mixed and representative for the concentrations throughout the atmospheric boundary layer. Measurements during Surf-2 (March 4 - 14, 1997) were taken at the Moss Landing Marine Laboratories (MLML) pier in Moss Landing, Monterey Bay. Unlike the SIO pier the MLML pier at 60 m does not usually reach past the surf zone. However, during Surf-2 waves did not break beyond the pier at Moss Landing. Data collected at the end of the pier were considered to be representative of the open ocean during onshore flow. Since the MLML pier is lower than the one located at SIO, background concentrations were measured at the end of the pier at a height of 6 m above mean water level.

Data collected during onshore flow were segregated according to wind speed at time of collection (0 - 2, 2 - 4, 4 - 6, and > 6 m/s). For each wind speed regime the background aerosol concentration was subtracted from the size distributions (A.3a) resulting in the surf zone breaking wave size distribution.

\[
\left( \frac{dN(D,z)}{dD} \right)_{surf} = \frac{dN(D,z)}{dD} - \left( \frac{dD(D)}{dD} \right)_{bg} \quad (A.3a)
\]

Where \( \frac{dN(D,z)}{dD} \) is the number concentration of particles with diameter at formation \( D \) per size interval \( \Delta D \) measured at a height \( z \), and \( \left( \frac{dD(D)}{dD} \right)_{bg} \) is the background number concentration. Sea spray aerosol number concentrations produced in the surf zone in each wind speed regime were determined by integration of the particle size distributions over the plume height.
\[
\left( \frac{dN(D)}{dD} \right)_{surf} = \int_0^{z_{\text{max}}} \left( \frac{dN(D,z)}{dD} \right)_{surf} \, dz \quad (A.3b)
\]

The surf zone aerosol surface flux \( \left( \frac{dF(D)}{dD} \right)_{surf} \) (Equation A.3c) was calculated by multiplying the surf produced aerosol number concentration (A.3a) with the wind speed \( u \), resulting in the advective flux, and dividing the flux by the width of the surf zone \( L \).

\[
\frac{dF(D)}{dD} = \frac{u}{L} \left( \frac{dN(D)}{dD} \right)_{surf} \quad (A.3c)
\]

Equation (A.3c) gives the mean surface number flux of sea spray aerosols per size interval, per square meter, per unit time \( [\mu m^{-1} \, m^{-2} \, s^{-1}] \).

To develop an equation describing the source function a simple power law fit to the calculated aerosol number flux for each wind speed regime. Equations were fitted of the type \( \frac{dF_N}{dD} = aD^{-b} \), for diameters at formation up to 20 \( \mu m \) (\( D_p \approx 5 \mu m \)), values for \( a \) and \( b \) were obtained from fitting the four wind speed regimes. A single Equation (A.3d) describing the surf source functions for wind speeds up to 9 m/s was found to be:

\[
\frac{dF_N}{dD} = 1.1 \exp(0.23U_{10})D^{-1.65} \quad (A.3d)
\]

Equation (A.3d) applies for particles with a diameter at formation between 1.6 - 20 \( \mu m \) (0.4 \( \leq D_p \leq 5 \mu m \)) where \( D \) is the diameter at formation in \( \mu m \), and \( U_{10} \) is the wind speed at 10 m in m/s. Note the number flux presented as Equation 4 in de Leeuw et al. [2000] contains an error. A factor of \( 10^6 \) is required for the expression presented by these investigators, their Equation 4, to yield the values shown in their Figure 7a.
A.4 Description of Mårtensson et al. [2003] Source Function:

Mårtensson et al. [2003] measured the size distribution produced from bubbles generated in a closed chamber by pumping filtered air through a sintered glass filter in a 2.0 L flask filled with dissolved synthetic sea-salt in pure Milli-Q water ($S_w = 0.0\%$) at different salinities ($S_w$) and water temperatures ($T_w$). Salinities used represented the Baltic Sea ($S_w = 0.0\%$ and $9.2\%$) and typical ocean ($33\%$). Most of the measurements were taken at water temperatures of $-2^\circ$C, $5^\circ$C, $15^\circ$C and $25^\circ$C. A differential mobility particle sizer (DMPS), condensation particle counter (CPC), and a differential mobility analyzer (DMA) were used to measure the size distribution of dry particle diameters ranging $0.020 - 0.135 \ \mu m$. Larger particles with a dry diameter between $0.135 - 20.0 \ \mu m$ were measured with an optical particle counter (OPC), and a passive cavity aerosol spectrometer probe (PCASP-X). The procedure was repeated for the different combinations of $S_w$ and $T_w$ in addition to different flow rates ($q_b$) 10, 60, and $120 \ mL \ min^{-1}$. All flow rates demonstrated the same two mode structure in the aerosol spectra, one around $0.05 - 0.1 \ \mu m$ and a second around $2 - 3 \ \mu m$. A flow rate of $10 \ mL \ min^{-1}$ was selected for the study because it appeared to most closely correspond to bubble concentrations during real whitecap conditions.

Mårtensson et al. [2003] derived a parameterization (A.4a) describing the number of particles produced per second, logarithmic size increment, and water surface area with bubbles ($A_b$) for each temperature and size interval ($j = 1 - 41$):

$$\Phi_j = \frac{1}{A_b} \frac{dn_j}{d \log D_p} \quad (A.4a)$$
Where $A_b \times 3.0172E^{-4}$ m$^2$ for $q_b = 10$ mL min$^{-1}$ and $n_p'$ (s$^{-1}$) is the measured particle number concentration produced at the water surface in each size interval during a certain time period. A linear fit was applied to the data collected for each DMPS and OPC size channel for each $T_w$. From these linear fits, $\Phi_j$ can be written as:

$$\Phi_j = a_j T_w + b_j \quad (A.4b)$$

Where the coefficient $a_j$ describes the temperature dependence of $\Phi_j$. Polynomials were fitted to $a_j(D_p)$ and $b_j(D_p)$ in order to describe the size dependence of $\Phi_j$,

$$A_k = c_4 D_p^4 + c_3 D_p^3 + c_2 D_p^2 + c_1 D_p + c_0$$

$$B_k = d_4 D_p^4 + d_3 D_p^3 + d_2 D_p^2 + d_1 D_p + d_0 \quad (A.4c)$$

for the three size intervals $k = 1$ (0.020 - 0.145 $\mu$m), $k = 2$ (0.145 - 0.419 $\mu$m) and $k = 3$ (0.419 - 2.8 $\mu$m). The coefficients for $c_l$ and $d_l$ are given in Table A.4.

**Table A.4. Coefficients for the parameterization of $A_k (c_4 - c_0)$ and $B_k (d_4 - d_0)$ for the three size intervals (k)**

<table>
<thead>
<tr>
<th>Size Interval ($10^{-6}$ m)</th>
<th>$c_4$</th>
<th>$c_3$</th>
<th>$c_2$</th>
<th>$c_1$</th>
<th>$c_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.020 - 0.145</td>
<td>-2.576E$^{35}$</td>
<td>5.932E$^{28}$</td>
<td>-2.867E$^{21}$</td>
<td>-3.003E$^{13}$</td>
<td>-2.881E$^{6}$</td>
</tr>
<tr>
<td>0.145 - 0.419</td>
<td>-2.452E$^{33}$</td>
<td>2.407E$^{27}$</td>
<td>-8.148E$^{20}$</td>
<td>1.183E$^{14}$</td>
<td>-6.743E$^{6}$</td>
</tr>
<tr>
<td>0.419 - 2.8</td>
<td>1.085E$^{29}$</td>
<td>-9.841E$^{23}$</td>
<td>3.132E$^{18}$</td>
<td>-4.165E$^{12}$</td>
<td>2.181E$^{6}$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Size Interval ($10^{-6}$ m)</th>
<th>$d_4$</th>
<th>$d_3$</th>
<th>$d_2$</th>
<th>$d_1$</th>
<th>$d_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.020 - 0.145</td>
<td>7.188E$^{37}$</td>
<td>-1.616E$^{31}$</td>
<td>6.791E$^{23}$</td>
<td>1.829E$^{16}$</td>
<td>7.609E$^{8}$</td>
</tr>
<tr>
<td>0.145 - 0.419</td>
<td>7.368E$^{35}$</td>
<td>-7.310E$^{29}$</td>
<td>2.528E$^{23}$</td>
<td>-3.787E$^{16}$</td>
<td>2.279E$^{9}$</td>
</tr>
<tr>
<td>0.419 - 2.8</td>
<td>-2.859E$^{31}$</td>
<td>2.601E$^{26}$</td>
<td>-8.297E$^{20}$</td>
<td>1.105E$^{15}$</td>
<td>-5.800E$^{8}$</td>
</tr>
</tbody>
</table>

The resulting particle flux per whitecap area (Equation A.4d) in [m$^2$ s$^{-1}$] can be estimates by combining Equations (A.4b) and (A.4c) for each size bin.

$$\frac{dF_p}{d \log D_p} = \Phi = A_k T_w + B_k \quad (A.4d)$$
The total oceanic flux \( (F_o) \) of particles with dry diameters in the range of \( 0.02 \leq D_p \leq 2.8 \) \( \mu \text{m} \) can be estimated by combining Equation (A.4d) with the Monahan et al. [1986] whitecap coverage (Equation 3.3c):

\[
\frac{dF_o}{d \log D_p} = \Phi W \quad (A.4e)
\]
Appendix B:
Derivation of Source Function

\[
\frac{CN_{\text{height}}}{N_{100}} = \frac{k \left[ (N_{100}) \left( A_{\text{avg}} \right) \left( L \right) (\tau) \right] dl + \int_{t_1}^{t_2} (N_{100}) \left( t \right) w \left( d \mbox{l} \right) dt}{\int \left[ w + (V_{\text{wind}} - V_{\text{wave}})(\tau) \right] dl h} \tag{B.1}
\]

Where:

\[
w = w_o - \frac{w_o}{\tau} t 
\]
Assume linear decrease with w

\[w_o = \text{Initial whitecap coverage of a breaking wave in the surf zone} \]
\[\tau = \text{Total time it takes for a wave to hit shore} \]

Assume:

\[N_{100} = \text{Constant} \quad w_o = \text{Constant} \]

Integrating the right hand side of the numerator in (B.1):

\[
\int_{t_1}^{t_2} \left( N_{100}(k) \int_{0}^{\tau} \left( \frac{w_o - \frac{w_o}{\tau} t}{\tau} \right) dt \right) = \left( N_{100}(k) \right) \left( \frac{w_o \tau}{2} \right)
\]

Now combine result with (B.1) and simplify

\[
CN_{\text{height}} = \frac{\left[ (N_{100}) \left( A_{\text{avg}} \right) \left( L \right) (\tau) (k) \right] + \left( N_{100}(k) \left( \frac{w_o \tau}{2} \right) \right)}{\left( \bar{V}_{\text{wind}} - \bar{V}_{\text{wave}} \right) \left( h \right) (\tau)} \tag{B.2}
\]

Simplify and solving (B.2) for \(N_{100}\) yields the source function:

\[
N_{100} = \frac{\left[ \left( CN \right) \left( k \right) \left( \bar{V}_{\text{wind}} - \bar{V}_{\text{wave}} \right) \left( h \right) \right]}{\left[ (A_{\text{avg}}) (L) \right] + \left( \frac{w_o}{2} \right)} \tag{B.3}
\]
Final form of the \( N_{100} \) source function:

\[
N_{100} = \left( \frac{CN}{A_{\text{avg}}} \right) \left( k \left( \bar{V}_{\text{wind}} - \bar{V}_{\text{wave}} \right) (h) \right) \left( \frac{L}{L_{\text{avg}}} \right) + \left( \frac{F_{\text{avg}}}{W_{\text{w}}} \right) (h) \]

（B.4）

**Units:**

[\#/cm^2 s] \( \rightarrow \) Number of aerosols produced per unit area (100% bubble coverage), per unit time

**Where:**

- \( CN \) \( \rightarrow \) Mean refractory excursions in CN at 5 m (40 cm\(^{-3}\) ± 6 cm\(^{-3}\))
- \( A_{\text{avg}} \) \( \rightarrow \) Mean bubble coverage (40% ± 12%)
- \( L \) \( \rightarrow \) Distance wave travels (35 m ± 3 m)
- \( \bar{V}_{\text{wind}} \) \( \rightarrow \) Mean wind speed in the surf zone (7.3 m/s ± 1.1 m/s)
- \( \bar{V}_{\text{wave}} \) \( \rightarrow \) Mean wave speed in the surf zone (1.59 m/s ± 0.19 m/s)
- \( k \) \( \rightarrow \) Constant of proportionality (2.25 ± 0.45)
- \( h \) \( \rightarrow \) Height where sample size distribution was taken (4 m ± 0.6 m)
- \( F_{\text{avg}} \) \( \rightarrow \) Width of wave front as fraction of \( W_{\text{w}} \) (0.5)
- \( W_{\text{w}} \) \( \rightarrow \) Initial width of whitecap bubble front (2 m ± 0.5 m)
Appendix C:
Uncertainty Analysis/Propagation

Uncertainty analysis is the process of estimating the uncertainty in a result calculated from measurements with known uncertainties. Uncertainty analysis uses the equations by which the result was calculated to estimate the effects of measurement uncertainties on the value of the result.

**Uncorrelated Uncertainty** - The error in one variable is **NOT** related to the error in another variable

**Correlated Uncertainty** - The error in one variable **IS** related to the error in another variable

**Rules for Calculating Uncertainty:**

1. For **addition** or **subtraction** the numbers must be expressed in terms of **absolute uncertainties** \([X \pm \delta x(#)]\). The absolute uncertainty of independently determined quantities is the sum of the squares of the absolute uncertainties of the quantities. Expressed with units.

   \[
   \text{Absolute Uncertainty} = X \pm \delta x(#)
   \]

2. For **multiplication** or **division** the numbers must be expressed in terms of **relative uncertainties** \([X \pm \delta x(\%)]\). The relative uncertainty of independently determined quantities is the square root of the sum of the squares of the percent uncertainties of the quantities. Expressed without units.

   \[
   \text{Relative Uncertainty} = X \pm \delta x(\%)
   \]

   All formulas assume \(\delta x\) is an **absolute uncertainty** (i.e. \(X \pm \delta x(#)\)):

   **Addition**
   \[
   (A \pm \delta A) + (B \pm \delta B) = (A + B) \pm \sqrt{\delta A^2 + \delta B^2}
   \]

   **Subtraction**
   \[
   (A \pm \delta A) - (B \pm \delta B) = (A - B) \pm \sqrt{\delta A^2 + \delta B^2}
   \]

   **Multiplication**
   \[
   (A \pm \delta A)(B \pm \delta B) = (AB) \pm \sqrt{\left(\frac{\delta A}{A}\right)^2 + \left(\frac{\delta B}{B}\right)^2}
   \]

   **Division**
   \[
   \frac{(A \pm \delta A)}{(B \pm \delta B)} = \left(\frac{A}{B}\right) \pm \sqrt{\left(\frac{\delta A}{A}\right)^2 + \left(\frac{\delta B}{B}\right)^2}
   \]

   \[
   \# \text{ to } \% = \left[\frac{\delta x(100)}{x}\right] \quad \%	ext{ to } \# = \left[\frac{\delta x(x)}{100}\right]
   \]
First we need to calculate mean wave speed \( (V_{\text{wave}}) \). Since we are dividing, according to Rule 2 we must work with relative uncertainties (decimal %). \( \delta L \) and \( \delta \tau \) are expressed as absolute uncertainties (3 m, 2 s) so we convert both to a relative uncertainty. The relative uncertainties are \( \delta L = 0.0857 \) and \( \delta \tau = 0.0909 \).

\[
\frac{V_{\text{wave}}}{\tau} = \frac{L}{\tau} = \frac{3500 \text{ cm} \pm 0.08}{22 \text{ s} \pm 0.09} = \frac{(3500 \text{ cm})}{(22 \text{ s})} \pm \sqrt{\left(\frac{300 \text{ cm}}{3500 \text{ cm}}\right)^2 + \left(\frac{2 \text{ s}}{22 \text{ s}}\right)^2} = 159 \text{ cm/s} \pm 0.12
\]

\[V_{\text{wave}} = 159 \text{ cm/s} \pm 19 \text{ cm/s}\]

\( N_{100} \) relates the number of aerosols produced in the surf zone per unit area, per unit time to \( CN \) for breaking wave plumes:

\[
N_{100} = \frac{\left[CN \left(\frac{k}{L} - \frac{h}{F_{\text{avg}}}ight)\right]}{\left[\frac{A_{\text{avg}}}{h} + \left(\frac{F_{\text{avg}}}{w_o}\right)\right]}
\]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Units</th>
<th>Relative (%)</th>
<th>Absolute (#)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( CN )</td>
<td>40</td>
<td>#/cm(^3)</td>
<td>± 15%</td>
<td>± 6</td>
</tr>
<tr>
<td>( k )</td>
<td>2.25</td>
<td>#/cm(^3)</td>
<td>± 20%</td>
<td>± 0.45</td>
</tr>
<tr>
<td>( V_{\text{wind}} )</td>
<td>730</td>
<td>cm/s</td>
<td>± 15%</td>
<td>± 110</td>
</tr>
<tr>
<td>( V_{\text{wave}} )</td>
<td>159</td>
<td>cm/s</td>
<td>± 15%</td>
<td>± 19</td>
</tr>
<tr>
<td>( h )</td>
<td>400</td>
<td>cm</td>
<td>± 15%</td>
<td>± 60</td>
</tr>
<tr>
<td>( A_{\text{avg}} )</td>
<td>40</td>
<td>%</td>
<td>± 15%</td>
<td>± 12%</td>
</tr>
<tr>
<td>( L )</td>
<td>3500</td>
<td>cm</td>
<td>± 8%</td>
<td>± 300</td>
</tr>
<tr>
<td>( F_{\text{avg}} )</td>
<td>0.5</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( w_o )</td>
<td>200</td>
<td>cm</td>
<td>± 25%</td>
<td>± 50</td>
</tr>
<tr>
<td>( \tau )</td>
<td>22</td>
<td>s</td>
<td>± 9%</td>
<td>± 2</td>
</tr>
</tbody>
</table>

We need to find the absolute uncertainties so we can calculate

\[
N_{100} = \left[\frac{40 \text{#/cm}^3 \pm 6 \text{#/cm}^3 \times 2.25 \pm 0.45 \times (730 \text{ cm/s} \pm 110 \text{ cm/s}) - (159 \text{ cm/s} \pm 19 \text{ cm/s}) \times (400 \text{ cm} \pm 60 \text{ cm})}{(40\% \pm 12\%) \times (3500 \text{ cm} \pm 300 \text{ cm}) + [(0.5) \times (200 \text{ cm} \pm 50 \text{ cm})]}\right]
\]

\[
(A \pm \delta A) - (B \pm \delta B) = (A - B) \pm \sqrt{\delta A^2 + \delta B^2}
\]

55
Now we will multiply the values in [brackets] in the denominator and get two new values and relative uncertainties

\[ N_{100} = \left[ \frac{[40 \text{#/cm}^3 \pm 0.15][2.25 \pm 0.20][571 \text{ cm/s} \pm 0.19][400 \text{ cm} \pm 0.15]}{[(0.40 \pm 0.12)(3500 \text{ cm} \pm 0.08)] + [(0.5)(200 \text{ cm} \pm 0.25)]} \right] \]

\[ (A \pm \delta A)(B \pm \delta B) = (AB) \pm \sqrt{(\delta A)^2 + (\delta B)^2} \]

\[ \frac{[(0.40 \pm 0.12)(3500 \text{ cm} \pm 0.08)]}{(0.40 \times 3500 \text{ cm}) \pm \sqrt{0.12^2 + 0.08^2}} = \frac{[(0.5 \pm 0.0)(200 \text{ cm} \pm 0.25)]}{(0.5 \times 200 \text{ cm}) \pm \sqrt{0.25^2}} \]

Now we will multiply the four terms in the numerator according to Rule 2 to get a single value and relative uncertainty

\[ N_{100} = \left[ \frac{[40 \text{#/cm}^3 \pm 0.15][2.25 \pm 0.20][571 \text{ cm/s} \pm 0.19][400 \text{ cm} \pm 0.15]}{[1400 \text{ cm} \pm 0.31] + [100 \text{ cm} \pm 0.25]} \right] \]

\[ (A \pm \delta A) + (B \pm \delta B) = (A + B) \pm \sqrt{(\delta A)^2 + (\delta B)^2} \]

\[ (1400 \text{ cm} \pm 0.31) + (100 \text{ cm} \pm 0.25) = (1400 \text{ cm} \pm 364 \text{ cm}) + (100 \text{ cm} \pm 25 \text{ cm}) \]

\[ (1400 \text{ cm} + 100 \text{ cm}) \pm \sqrt{(364 \text{ cm})^2 + (25 \text{ cm})^2} \]

\[ 1500 \text{ cm} \pm 434 \text{ cm} \]

\[ 1500 \text{ cm} \pm 0.28 \]

Now we will multiply the four terms in the numerator according to Rule 2 to get a single value and relative uncertainty

\[ N_{100} = \left[ \frac{[40 \text{#/cm}^3 \pm 0.15][2.25 \pm 0.20][571 \text{ cm/s} \pm 0.19][400 \text{ cm} \pm 0.15]}{[1500 \text{ cm} \pm 0.28]} \right] \]
\[ \frac{A \pm \Delta A}{B \pm \Delta B} \cdot \frac{C \pm \Delta C}{D \pm \Delta D} = (ABCD) \pm \sqrt{\left(\frac{\Delta A}{A}\right)^2 + \left(\frac{\Delta B}{B}\right)^2 + \left(\frac{\Delta C}{C}\right)^2 + \left(\frac{\Delta D}{D}\right)^2} \]

\[ (40 \text{#/cm}^3 \times 2.25 \times 571 \text{ cm/s} \times 400 \text{ cm}) \pm \sqrt{(0.15)^2 + (0.20)^2 + (0.19)^2 + (0.15)^2} = 20,556,000 \text{#/cm s} \pm 0.34 \]

Finally, by dividing according to Rule 2 we get the final value and relative uncertainty associated with \( N_{100} \).

\[ N_{100} = \frac{(20,556,000 \text{#/cm s} \pm 0.34)}{1500 \text{ cm} \pm 0.28} \]

\[ \frac{A \pm \Delta A}{B \pm \Delta B} = \frac{A}{B} \pm \sqrt{\left(\frac{\Delta A}{A}\right)^2 + \left(\frac{\Delta B}{B}\right)^2} \]

\[ \frac{(20,556,000 \text{#/cm s} \pm 0.34)}{(1500 \text{ cm} \pm 0.28)} = \left(\frac{20,556,000 \text{#/cm s}}{1500 \text{ cm}}\right) \pm \sqrt{(0.34)^2 + (0.28)^2} \]

\[ N_{100} = 13,704 \text{#/cm}^2 \text{s} \pm 0.44 \]

Comparison with independent measurements of total number (small particles) and calculated scattering (large particles) during SEAS [Clarke et al., 2003] constrains the overall uncertainty of the mean normalized number distribution used to scale \( N_{100} \) to about 10%.

**Uncorrelated Relative Uncertainty**

\[ N_{100} = 13,704 \text{#/cm}^2 \text{s} \pm 0.54 \]

**Uncorrelated Absolute Uncertainty**

\[ N_{100} = 13,704 \text{#/cm}^2 \text{s} \pm 6029 \text{#/cm}^2 \text{s} \]
References:


Figure (1.21) Bubbles on the ocean surface have a thin liquid cap. Upon bursting, this bubble cap burst into many small droplets called film drops.

Figure (1.22) Jet droplets are produced by the collapse of a spherical cavity after bursting.

Figure (1.23) High winds tear spume drops directly from the wave crests and eject them into the marine boundary layer.
**Figure (1.24)** Sea spray sizes depend on the generation mechanism, and cover about 4 orders of magnitude. The sea spray droplet size spectrum covers several orders of magnitude.

**Figure (1.25)** Taken from *Andreas* [1998] (his Figure 1). Various estimates of the sea spray generation function in terms of the volume flux, for a 10 m wind speed ($U_{10}$) of 15 m/s.
Figure (1.61) Taken from Mårtensson et al., [2003] (their Figure 6). Number distributions of primary marine aerosol produced at water temperatures of -2°C (green solid), 5°C (blue dash), 15°C (black dot-dash) and 23°C (red solid). Here the salinity was 33‰.

Figure (1.62) Taken from Mårtensson et al., [2003] (their Figure 5). Number distributions of primary marine aerosol produced from bubbles in water with the salinity of 0.0‰ (green solid), 9.2‰ (blue dash) and 33.0‰ (red solid). The water temperature was 23°C.
Figure (2.11) Map of Oahu with the location of Bellow Air Force Station indicated.

Figure (2.12) a) The University of Hawaii Coastal Aerosol Facility located at BAFS on the northern portion of Waimanalo Bay (21°21' N, 157°42' W) has a 20 m tower situated on the site around 37 m inland and is oriented into the prevailing trade winds. b) Near surface aerosol plume inlet.
Figure (2.21) Images of sea-salt collected during ACE-1, reported in Buseck and Posfai [1999] (their Figure 5) taken with a transmission electron microscope. (a and b) Subhedral halite (NaCl) and euhedral sulfate crystals.

Figure (2.22) Relevant available meteorological parameters associated with aerosol measurements during SEAS: April 21-27, 2000. Mean values of each are also displayed.
Figure (2.31) Vertical gradient in CN Hot and scattering from 1 m above the beach up to 20 m on the tower. Measurements < 5 m are most influenced by coastal breaking wave plumes while those at 20 m reflect background oceanic values.

Figure (2.32) Vertical distribution of lognormal average scattering and standard deviation over the breaking waves at Bellows Air Force Station.
Figure (2.33) **Top panel:** A simple linear regression linking breaking wave scattering and CN Hot during SEAS after removal of background oceanic values measured at 20 m. Dataset covers a typical 9-minute period. **Bottom panel:** Time series of breaking wave scattering and CN Hot after subtracting oceanic values measured at 20 m on the tower.

![Figure 2.33](image)

Figure (2.34) Photograph of surf zone at BAFS superimposed with some parameters that make up $N_{100}$.  

![Figure 2.34](image)
Figure (2.35) Mean breaking wave size distribution during SEAS (left y-axis) with a cumulative number plot (right y-axis).

Figure (3.12) Graphical representation of the calculated total relative uncertainty (i.e. ±54%) in the derived $N_{100}$ number flux. Note log (left graph) and linear (right graph) y-axes are for same data.
Figure (3.13) Example analysis of the *Pick Peaks* function from OriginPro 6.1, which was used to determine the mean refractory breaking wave CN excursion measured at 5 m on the tower.

Figure (3.21) **Bottom Panel:** The original breaking wave number flux is shown in heavy black while the individual fit over each size range is displayed on top of it. **Top Panel:** Simple linear regression of the original breaking wave flux to the 5th order polynomial fit.
Figure (3.22) Monahan's empirical relation for whitecap coverage (%) for a given wind speed at 10 m, shown for winds ranging 5 - 30 m/s.

$$W = \left(3.84 \times 10^{-6}\right) U_{10}^{3.41}$$

Monahan et al., [1986]

Figure (4.11) A comparison of the derived sea-salt source function (± 54%) determined from SEAS normalized breaking wave refractory size distributions to others available in literature. All flux estimates have been scaled to oceanic conditions based upon estimated whitecap coverage for an assumed wind $U_{10} = 9$ m/s (§ 4.1) and are represented as dry diameters.