Invited papers

The effects of the marine aerosol on infrared propagation over the World Ocean

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Abstract

In the modern world, where infrared systems are operated by the world's navies, it is important to understand the effect that large marine aerosols have on the propagation of these signals. This article reviews some of the work that has taken place to describe these aerosols and their scattering and absorption of infrared wavelength radiation. The paper describes those aerosols found in the marine environment which are produced by the whitewater phenomenon over the ocean, such as jet drops from breaking air bubbles at the sea surface and the shearing away of large droplets from cresting waves. These processes are the result of cresting of ocean waves and the breaking of swell and waves on the shore and are referred to as 'open ocean phenomena' and 'coastal processes'. The paper also presents some of the early results of the United States coastal aerosol research project, EOPACE (Electro Optics Propagation Assessment in the Coastal Environment).

1. Introduction

This article reviews much of the work done in the area of marine aerosols and their effect on the propagation of infrared energy in the first kilometre above the world ocean. It is not an exhaustive review as the focus of this

paper is on ways of determining swiftly and practically the effects of aerosols on electro-optical (EO) and infrared (IR) radiation, and as there are other applications of marine aerosol research being investigated world wide.

The world's navies have a particular interest in determining aerosol distributions throughout the lower layers of the atmosphere over the ocean, where most of their operations take place. The reason is that aerosols constitute an extremely varied component of the marine atmosphere and contribute to the deterioration in the propagation of EO energy in this region. Although the molecular composition of the air is relatively constant, the aerosol concentration in the marine boundary layer can vary by several orders of magnitude.

The marine aerosols which interact with IR energy propagation in the marine environment are especially emphasised in this paper. The aerosols of greatest significance in this case have radii between 0.5 and 40 microns. The largest aerosols in this range are mainly of marine origin. Furthermore, their chemical composition is known – they are considered to be chiefly a mixture of sea salt and water with an admixture of organic material. The structure and composition of the smaller 'background' aerosols, while extremely varied, do not for the most part affect IR propagation as much as the large particles because they are less efficient than these larger particles at scattering and absorbing IR energy. As a result the shapes of the individual small, dry 'background' aerosol particles, no matter whether they be cubes, spheres, elongated pencil shaped or disks, are relatively unimportant to the scope of this review. These larger particles can be assumed spherical, especially if they are hygroscopic and dissolved in water.

Another feature of this limited scope of aerosol research is that the size distributions of these background aerosols can usually be represented by a single log-normal function. In the case where we know there are both hygroscopic and non-hygroscopic small aerosols present, two functions can be used, one being a function of relative humidity and the other not.

Although this provides only a generalisation of the properties of the small background aerosols, this technique (representation by a log-normal function) does reduce the amount of knowledge (number of parameters) needed to describe the effect of this aerosol on IR propagation. In many cases the interaction of these background aerosols with the IR radiation is significantly less than that of the larger sea salt marine aerosol. These assumptions are most acceptable when talking about marine aerosols at distances far from the land sources of small background aerosols. When approaching land areas, then, these assumptions are less accurate and the nature of coastal marine aerosol needs to be addressed.

2. Background

The advent of infrared systems for naval use spawned the need to study and predict the propagation parameters in the marine environment (both for open ocean and coastal environments). The determination of atmospheric propagation at various IR wavelengths was at first addressed in a straightforward way: transmissions along an optical path were measured and the results recorded for certain geographical areas along with the meteorological conditions under which they were measured and recorded. This method worked for a limited set of fixed points such as over a concave shoreline or between the shore and a fixed platform at sea. However, this direct approach quickly ran into practical problems farther away from the shore because IR transmission at sea, *e.g.* IR transmission measurements at sea on a moving platform between two ships, is an extremely difficult measurement to make. Such transmission measurements only apply to a given place, time and meteorological conditions, and they require the construction of some sort of climatology if they are to be useful operationally in other geographic locations. Such a worldwide climatology compilation requires a statistically significant database for wavelengths of interest that is just too expensive to undertake. Thus a more physical and pragmatic approach was considered in which the molecular scattering and absorption of EO energy from the relatively constant atmospheric molecules is added to the EO properties of the more variable components of marine aerosols. In this approach (for the case of a sea salt aerosol), only the size distribution of the aerosol and the ambient relative humidity is needed to calculate the aerosol component of extinction at any wavelength of interest. An additional bonus is that the molecular component of extinction based on relatively constant ratios of elements in the lower atmosphere has its propagation properties well characterised by models such as LOWTRAN VI (Kneizys *et al.* 1983) and its successors. The problem then becomes one of determining the chemical composition and size distribution of the aerosols at representative locations throughout the world ocean.

This approach appears to be possible, as the marine aerosol concentration in the marine boundary layer should be predictable if one knows the physics involved in the aerosol sources, sinks and transport. In the case of a hygroscopic aerosol, the size of individual particles is a function of the ambient relative humidity, which allows the size distribution and diluted chemical composition of the aerosol to be known.

The science of small-particle measurements became more sophisticated with the advent of computerised particle-sizing instrumentation and thus much effort has been expended to determine aerosol characteristics of the

atmosphere experimentally. Aerosols have been shown to be an important and variable component of the atmosphere. Even though aerosol size distributions can be more easily measured from ships and aircraft than IR transmission, the construction of a statistical climatology of aerosol size distribution throughout the world ocean is still a prohibitively expensive effort. What is needed is a model that will describe the important EO parameters of the marine aerosol at a particular time and place and require only routine types of meteorological data.

It is well known that many of the large marine aerosols are produced by the injection of jet droplets into the atmosphere. Blanchard (1954) has shown that they leave the air/water surface with sufficient velocity that they are injected into the lowest parts of the turbulent boundary layers and hence mixed in the marine atmosphere where ships operate. The flux of these upward-projected aerosols is proportional to the percentage of the sea surface covered by bubbles or white water or whitecaps. It has been shown that the percentage of the sea surface covered by whitecaps is a function of the wind speed blowing over the waves (Blanchard 1963). Thus the source of the marine aerosol should be a function of the wind speed. In addition, Monahan *et al.* (1986) has shown the existence of 'spume' droplets, which are water droplets sheared off wave tops by strong winds, and these 'spume' droplets are added to the mixture.

3. Background Aerosols

Most small background aerosols are not produced by wind/wave interaction but are either produced *in situ* in the marine atmosphere with some sort of chemical conversion, or they may have been transported many miles from their generation sites by advection. A globally useful aerosol model would then take into account the various sources of aerosols, their residence time in the atmosphere, and their transport from the place of origin to the location where the calculation is being made. It should also have as input parameters those that are easily obtained either from remote sensors such as satellites, meteorological forecasting models or, for the case of constructing aerosol climatologies, from historical meteorological data bases.

The problem of small aerosol sources can then be broken down into two parts: the first refers to the many physical and chemical processes describing the 'life cycles' of small, non-sea salt aerosols in the marine atmosphere (Fitzgerald *et al.* 1998a,b). These sources are relatively uniform over the waters and are the main type of aerosol over the open ocean. A second class is that of aerosols advected downwind from land areas. In these cases, it seems that especially large clouds of various kinds of aerosol can be found in littoral zones near industrial areas. The variation between aerosol clouds may be

striking near land but they eventually lose their identity as these air masses are advected out to sea. An example of this was given by Gathman & Jensen (1995) (Figs. 1a and 1b). This illustration is from the MAPTIP experiment off the Dutch coast during 1993. A small aircraft flying at a constant altitude (below 150 metres) in a star-shaped flight pattern did the mapping shown in these figures. During these flights, the concentration of aerosol as measured by a Particle Measuring Systems model FSSP aerosol spectrometer was contoured in an 'onshore wind' (Fig. 1a) and an 'offshore wind' (Fig. 1b). These flights showed that the aerosol in the littoral zone can be dominated by land- or surf-produced aerosol.

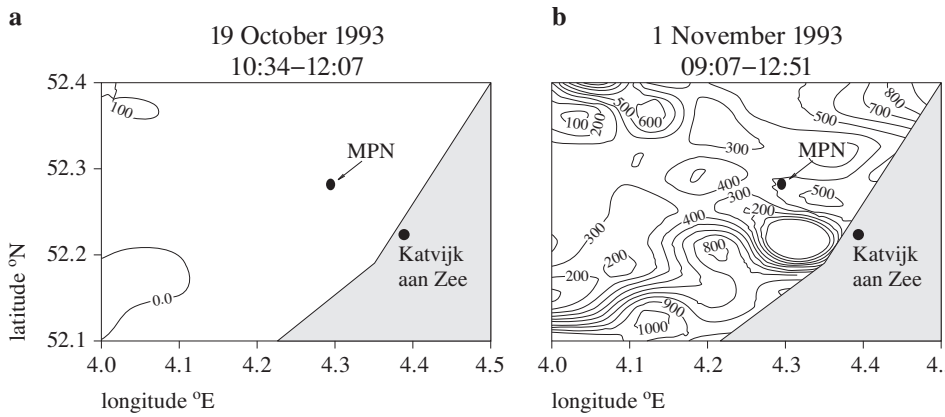

Fig. 1. An aerosol concentration map made on 19 October 1993 in which measurements at the MPN tower indicated an essentially 'open' ocean aerosol environment. The pockets of aerosol seen here may or may not be from ships traversing the region but it is felt that they could indeed be from noise sources either in the data itself or from the interpolation needed in the contouring of irregular data (a). The map of the MAPTIP area for 1 November 1993 shows an example of an offshore wind when the measurements at MPN indicated that there were indeed coastal aerosols in the region (b)

It is seen at once that if all the detailed physics and chemistry were taken into account, the model would require complicated and probably lengthy computations. Thus the idea of focusing only on the IR propagation problem helped produce small, quick, easy-to-use models which would approximately simulate nature. The idea is to use an approximation for a small aerosol that would be in the form of a simple log-normal function. The emphasis here would be on obtaining an equivalent log-normal function, which would have approximately the same scattering and absorption characteristics as a natural aerosol. This should minimise the small effect that differences in the size distributions of these aerosols have on IR propagation.

The log-normal is described by three parameters, and is thus very useful as a simple representation of the small aerosol component of the atmosphere.

4. Marine aerosols

The first approach in determining the characteristics of marine aerosols is to make *in situ* measurements of them. This has been done over the years although there have not been nearly enough measurements to enable the construction of any kind of adequate climatology. Measurements have been made from ships, islands, tower platforms located in the sea and aircraft of all kinds. Most of the measurements, however, were taken in several key areas of the world and an extension of these measurements to other parts of the world is not really advisable.

Accurate measurements from the layers just over the open ocean can be difficult to obtain. Issues such as the sampling time required to make an accurate measurement will obviously depend on the sizes of aerosol in question. We know that for the larger sizes, the aerosol concentration is very small. The problem here is essentially statistical in nature: a sufficient number of particles in a certain size range must be counted before statistically accurate measurements of the aerosol size distribution can be made. For instance, as in the case of a very large aerosol, concentrations of only 1 or 2 per cubic centimetre are meaningless unless a large sample is taken. If the time to take this large sample is too long, then the experimentalist is faced with the problem of losing the time or space resolution in his data. This problem and the problem of airflow about the sampling platform, be it a large object like a ship or platform at sea or a small, fast-moving aircraft or even a lighter-than-air craft, requires special care in order to ensure that an accurate sample is indeed taken.

Nevertheless, it seems that direct aerosol measurements are the basis of much of our knowledge of marine aerosols. Unfortunately, direct measurements have limitations in that they only describe what the situation is at a particular place and time. However, if enough world-wide observations could be put together, then statistical aerosol climatologies could be assembled which could be used to make statistical inferences on what usually happens at a particular place and time of year. But assembling enough data to provide an adequate aerosol climatology has been prohibitively expensive. Another approach would be to use remote sensing techniques involving satellites. With this method, the whole globe can be covered in a reasonable amount of time, but cloud cover and the accuracy of the inversion technique limit its utility. What is required is a means by which the aerosol can be determined at any place and time on demand. Models can, of course, be invoked here, but in order to initiate or develop such models, the experimental evidence must be available on which to base them.

5. Modelling the marine aerosol

5.1. First-principle models

It might seem that a first-principle model would best describe and simulate the various physical processes taking place in the marine atmosphere. However, even the simplest part of such a model, involving the production of a sea salt aerosol from waves, becomes very complicated. Even these first-principle aerosol models require preliminary data as well as a source function (based on measurements) and there are great differences in estimates of the source function for the marine aerosol. Full-component models, which take into account chemical species in addition to the sea salt, become very complicated indeed. Three-dimensional models are needed to adequately describe the effect of turbulence on the mixing of these aerosols, and such models are not available as yet, nor are they practical in the light of present computer power.

5.2. Examples of currently available limited-dimension models

One of these models, **SeaCluse** (Mestayer *et al.* 1996), describes and quantifies the non-linear interactions between spray droplet concentration and the scalar fields of water vapour concentration and temperature in the marine atmospheric surface layer over the waves in open or coastal regions. The main output of this code consists of vertical profiles of aerosol concentration, water vapour and temperature. While unique in its interactions of droplets with the scalar fields and the effects of the wave structure on these processes, this model is unfortunately only a step towards a final solution in that it does not include small aerosols, nor does it include enough dimensions to describe the advection of aerosols in this region.

Another approach (Fitzgerald *et al.* 1998) simulates in the marine boundary layer some of the multi-component aerosol dynamics (including various chemical interactions as well as gas-to-particle production). It shows in great detail these effects on the small components of the marine atmosphere, although it uses a simpler approach to the production of large sea salt aerosols than does SeaCluse. It is also a one-dimensional model, which cannot really describe the complex aspects of advection in the littoral zone.

In her Ph. D. thesis, Vignati (1999) proposed a two-dimensional aerosol model in which a column of air in the marine boundary layer is allowed to move with a constant wind speed. This two-dimensional model allows a more versatile application to the marine environment than is available in only one dimension. In addition, it allows for interactions between the marine aerosol and a few of the major chemical constituents of the marine

boundary layer. It does not, however, describe the details of the production of large sea salt aerosols and their interactions with water waves in the way SeaCluse does.

Modellers have thus addressed the problem of the marine aerosol from a physical process standpoint, yet none of these modelling approaches has reached the desired state of development.

5.3. Simplified models (semi-empirical approach)

Gathman (1983) first introduced the Navy Aerosol Model (NAM) to the community and various improvements have been added to this model over the intervening years. This model was developed from a set of oceanic aerosol size distribution measurements and the associated meteorological observations at the time of measurement. Once the aerosol size distribution has been found, then Mie theory is used to determine the optical and infrared properties of the aerosol in that atmosphere. This model was developed for and works well over open ocean conditions.

This model can be given standard meteorological input data such as relative humidity, wind speed, and visibility. The aerosol size distribution can thereby be estimated quickly, as can the extinction and absorption produced by this aerosol at any wavelength from 0.2 to 40 micrometres. Therefore, climatologies could be constructed for the optical/IR properties of the open ocean atmosphere based on historical meteorological records. The model could also be used to estimate the present optical/IR properties based only on a knowledge of present meteorological conditions. And finally, because future meteorological parameters can be forecast, the optical/infrared properties can also be forecast based on this model.

In general, the size distribution $n(r)$ of NAM is represented as a sum of three log-normal functions:

$$n(r) = \frac{dN(r)}{dr} = \sum_{i=0}^{3} k_i \, \frac{A_i}{f_i} \exp\left[-w_i \left(\log\left(\frac{r}{r_{0i}\, f_i}\right)\right)^2\right], \tag{1}$$

where the hygroscopic growth functions f_i are functions of the relative humidity and the chemistry of the droplet (Gerber 1985). The mode radii r_{0i} are determined by experiment and fixed at $r_{01} = 0.03$, $r_{02} = 0.24$ and $r_{03} = 2.0$ microns. The k_i's are correction functions that correct the surface values to direct measurements of visibility and/or IR extinction when available. The mode amplitudes A_i are proportional to the concentrations of the various populations in the size distributions as shown in Fig. 2. These mode amplitudes contain most of the variability of the model as they are

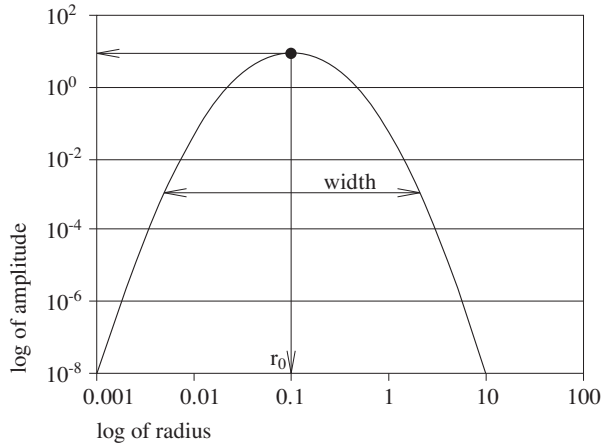

Fig. 2. The relationship of the three log-normal parameters, amplitude A, mode radius r_0, and width as plotted on a log-log scale

related to the production, dissipation and the advection of aerosols. It is also a function of altitude and is therefore related to the mixing turbulence and thermal stability of the atmosphere.

NAM uses the index of refraction of water from Hale & Query (1973) and makes an assumption on the chemical composition of each of the aerosol modes in the model. It is known that the chemical composition of the soluble droplets changes as a function of relative humidity. A perfectly dry particle has the complex index of refraction of the nucleus material. On the other hand, in a very high relative humidity environment, a hygroscopic particle will have changed size, taking on a considerable amount of water with the nucleus being dissolved in the water. The net complex index of refraction used in the Mie calculations for NAM is then assumed to be the volume average of the index of refraction of water and the index of refraction of the nucleus material using the method of Hänel (1976). Each of the classes of aerosol in the model is allowed to have its own complex index of refraction. The amount of growth, represented by the f factor, for each class also depends on the chemical composition of the nucleus of that class.

In NAM, an air mass parameter A_1 is defined, which relates the concentration of a small aerosol to a single number. A number of schemes were developed to estimate this parameter from measurable quantities. These techniques are described in more detail in Battalino (1998), Littfin *et al.* (1998), and Littfin & Goroch (1997).

The next part of the question is how the sea salt particle components of NAM are related to their generation physics and their concentration with respect to size at any place and time. Obviously, these larger aerosols will

have a more profound effect on the propagation of IR energy as these sizes are of the order of the wavelengths of radiation. In the original construction of NAM, the extensive analysis of the data available showed that the variations could be characterised by two additional log-normal modes. The mode amplitude of each of these modes is affected by wind speed. The smaller of these two modes seemed to be dependent on the long-term wind speed over the ocean. The idea here is that these smaller aerosols, having longer dwell times in the atmosphere than those of larger sizes, would be dependent on the wind speed history rather than on the current wind speed. On the other hand, the larger aerosol would have a much shorter dwell time and should therefore be a function of the current wind speed and predictions. During high wind speeds, these aerosols are kicked up into the atmosphere, only to fall again when the wind speed drops.

The initial data set had severe limitations on the larger sizes, causing errors in the parameterisation of the largest aerosol mode. The publication of rotorod data by de Leeuw (1986) allowed a much firmer parameterisation to be made. The effect of fetch is not taken into account in the formulation. For cases far from shore this is unimportant, but when exercising the model closer to shore, the fetch effect has been shown to be important (Piazzola *et al.* 1999). In fact, these authors have redefined the NAM parameters to include the effect of fetch for the specific coastal region of Galway Bay, Ireland, with good results. While this formulation works for this specific location, it has yet to be shown to be universally true for other locations.

6. Electro-optical calculations

Mie theory was used to calculate the volume extinction, scatter and absorption coefficients at a band of wavelengths from ultraviolet to 40 microns from a model population of spherical droplets of known chemical composition. These are functions of the wavelength, the radius of the sphere, and the index of refraction of the sphere. The volume extinction coefficient is expressed in eq. (2) as an integration over the whole population of aerosol sizes. The volume absorption coefficient can be calculated in a similar way, except that the Mie coefficient Q_{abs} must be used instead of Q_{ext}. The volume extinction coefficient is expressed as

$$\beta_{\text{ext}} = \frac{\pi}{1000} \int Q_{\text{ext}} \frac{dN}{dr} r^2 \, dr. \tag{2}$$

If we now substitute the log-normal formulation for dN/dr into eq. (2) above, our formulation of the size distribution in terms of the sum of these log-normal type functions becomes

$$\beta_{\text{ext}} = \frac{\pi}{1000} \int Q_{\text{ext}} \sum_{i=0}^{3} \frac{A_i}{f_i} \exp\left[-1 \left\{ \log\left(\frac{r}{r_{0i} f_i} \right) \right\}^2 \right] r^2 \, dr. \tag{3}$$

The Dave (1968) code was adopted to calculate Q_{ext} for these calculations. The integration and the summation can be switched as in eq. (4) so that we really have a sum of several integrals. This has the advantage that they can be precalculated to the required precision and their values stored in tables if necessary.

$$\beta_{\text{ext}} = \frac{\pi}{1000} \sum_{i=0}^{3} \frac{A_i}{f_i} \int Q_{\text{ext}} \exp\left[-1 \left\{\log\left(\frac{r}{r_{0i}\, f_i}\right)\right\}^2\right] r^2\, dr. \tag{4}$$

If the dry aerosol nucleus of a particular chemistry (say sea salt) is soluble then the growth factor f is a function of relative humidity. The index of refraction is determined after the method of Hänel (1976), where indices are calculated by using the ratio of the volumes for each population group.

7. Naval Oceanic Vertical Aerosol Model (NOVAM)

Most of the original data used in the development of NAM was taken in the open ocean environment, hence the model is principally useful at deck level for the open ocean scenario. A need exists, however, to extend the model to higher levels for determining the propagation properties along slant paths.

The need to know the optical/IR properties of slant paths resulted in the development of NOVAM (Gathman 1989, Gathman & Davidson 1993). It was based on NAM as a kernel and was the same as NAM at an altitude of 10 metres. It provides information on the optical/IR properties of the aerosol at any altitude from ship deck level to above the marine boundary layer. As its input, NOVAM uses information from profile data of temperature, humidity and surface meteorological data obtained from various types of measurement techniques. From this data, it automatically determines the significant profile parameters that it needs. However, a default case is currently being incorporated into NOVAM where world-wide climatological data can be used to determine the best estimate of this information.

The characteristics of the temperature profile can be classified in terms of the existence or non-existence of temperature inversions. Three cases recognised by NOVAM are:

1. No inversion – the free convection mode.
2. Two inversions – the weak convection mode.
3. One inversion – the developed boundary layer.

Shown in Fig. 3 is an example of a NOVAM extinction profile obtained during the KEY–90 experiments off the Florida Keys (Gathman *et al.* 1993). The *in situ* extinction profile data plotted in the figure are from Mie calculations, while the aerosol size distributions were measured from an aircraft making a spiral descent and from a boat on the water's surface.

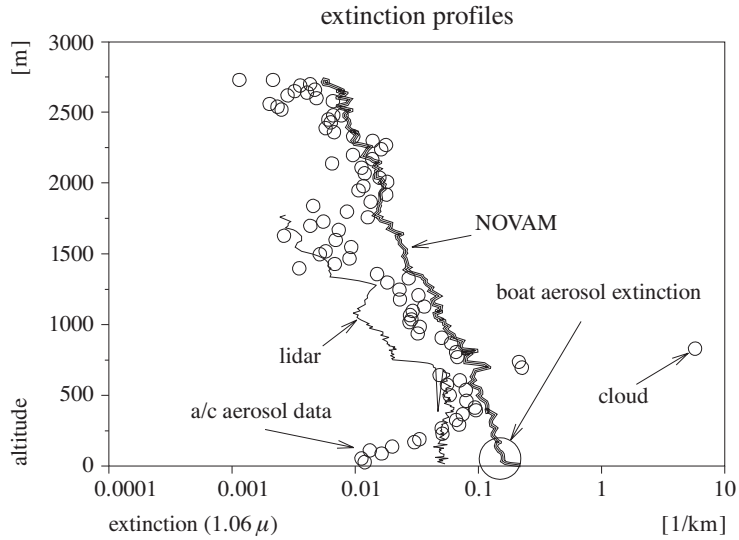

Fig. 3. A composite plot of various types of extinction data taken during KEY–90 on 14 July 1990 for a wavelength of 1.06 μm. The small circles are the data calculated from the aircraft aerosol size distribution measurements. The large circle at the surface is the extinction calculation from an hour average of the boat aerosol size distribution measurement. The thin line is the NRL LIDAR extinction estimate at approximately the same time and place. Finally, the thick line is the NOVAM estimate of extinction at 1.06 μm for the particular set of surface and radiosonde data available at the site. Note that the aircraft inadvertently entered a cloud in the process of making its profile and a few large extinction values are noted in the data

Remote sensing data is also shown in Fig. 3 – the thin line representing the profile of the estimated extinction from a downward-looking LIDAR located in a plane flying overhead at 6 kilometres. The NOVAM prediction is shown as the heavy dark line. Surface meteorological data from the boat and the profile data obtained from the aircraft were used as input for NOVAM.

The model does not attempt to describe the situations within clouds (as seen in this example when the spiralling aircraft penetrated a small cloud while making its profile). Likewise, if precipitation is present, the model is not capable of determining the optical/IR properties of the precipitation.

While existing Navy specific aerosol models such as NAM and NOVAM do a fairly good job of determining the IR/optical climate in the open

ocean environment at and above shipboard levels, experience has shown that altitudes below deck height levels and in coastal regions are not so well determined. In the coastal regions the aerosol has at least two additional sources that can, under certain conditions, overwhelm the aerosol generated in an 'open-ocean' manner. This is an area recently investigated under programs such as MAPTIP (Jensen *et al.* 1993) and EOPACE (Jensen *et al.* 1997).

7.1. Large marine aerosols between the wave tops and 10 metres

The large particles that occur just above the sea waves in high wind conditions affect IR propagation for paths that pass very close to the surface. These aerosols are produced by various air-sea interactions. Unlike smaller particles, the largest of these droplets will not mix throughout the marine boundary layer because of the overriding effects of gravity. This should result in significantly higher numbers of large aerosols close to the water surface than are found at higher altitudes. Mie theory says that the maximum scattering interaction of a spherical droplet with incoming radiation is at wavelengths of the same order of size as the diameter of the droplet. Thus the larger particles are quite effective at scattering IR radiation from a beam passing close to the tops of the wave crests. Therefore, a knowledge of the aerosol properties in this area is necessary to determine the optical/IR properties of aerosols near the ocean surface paths.

An effort has been made to extend the NAM/NOVAM series of models down to the sea surface. The measurements essential for this study need special instrumentation in order to be able to detect and count the largest droplets that might be encountered in nature. A device was needed which sampled a large amount of air in a relatively short period of time that also did not require changes in air flow into and through sampling tubes. Sampling problems occur with large-inertia droplets as they tend to resist changes in their direction of motion. In addition, the device needed to be expendable in case a rogue wave came through while sampling close to the wave crests. Such a device was found which fulfils the above requirements but which unfortunately is rather time consuming in the data analysis. The rotorod is an active impaction sampler as opposed to settling collectors such as glass plates. It impacts droplets as they are naturally advected through the sampling volume and then leaves an image on the flat surface of a rod which is D-shaped in cross-section. The machined flat surface has a mirror finish and is coated with silicone. Particles larger than 10 micrometres are collected by the rods with an efficiency factor greater than 90% (Vrins & Hofschreuder 1983). Advances in computer technology make the analysis of this data more reasonable, with, for example, the advent of frame grabbers

and statistical image processing techniques. A sample of rotorod aerosol size spectrum data is shown in Fig. 4, where two sets of the rotorod spectra from MAPTIP are plotted together with a simultaneous spectrum of a commercial scattering aerosol spectrometer (Particle Measurement Systems, model ASSP 100).

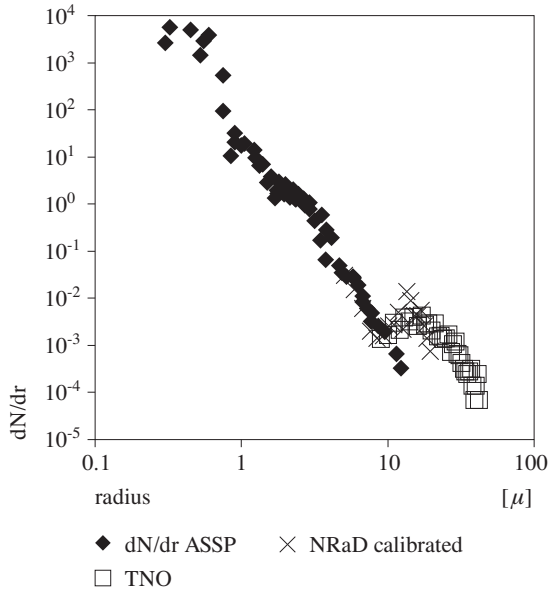

Fig. 4. A composite of three instruments making a simultaneous measurement close to each other of aerosol size distribution during MAPTIP

These data show that there does indeed seem (under certain conditions) to be a population of large aerosol droplets in the regions close to the wave tops that are not sampled with traditional methods. Not only are they not usually measured but they are not taken into account in the IR propagation calculations. The Advanced Navy Aerosol Model ANAM (Gathman & van Eijk 1997) allows for this by extending the number from three to four log-normal components to describe this population of aerosols.

At the present time, the ANAM has been constructed from a limited data set obtained during the MAPTIP experiment. The major feature of ANAM is the additional fourth mode log-normal function which describes the distribution of the very large particles close to the wave tops. Profiles of the fourth mode aerosol are (in this model) related to atmospheric stability, wind speed and generation rates. ANAM is presently being verified by comparison with other data from rotorod experiments and with numerical experiments using the SeaCluse model.

Reviewing the published information on large-size aerosols between the wave tops and 13 metres in altitude, one should note that Chaen (1973) concluded from his measurements at heights of 1 and 13 metres that the profile of the number concentration of large aerosols approached a power law form. Preobrazhenskii (1973) concluded from his measurements at 1 and 7 metres that the net particle volume decreased with height. De Leeuw (1986b) made rotorod measurements of the giant sea salt aerosol and concluded that the profiles were more complex than a strict power law or an exponential monotonically decreasing function of height, but that they were related to the micrometeorology of the marine atmospheric surface boundary layer.

A curious phenomenon occasionally exhibited itself in the rotorod profile records from MAPTIP and other experiments. Profiles made in the littoral zone occasionally show an apparent increase in aerosol concentration 10 metres above the sea surface compared with the measurements at 2 metres. Steady state models using a source function at the surface cannot be made to duplicate this phenomenon without the introduction of advection from a more distant source.

Of course, the question to ask is whether this is a real phenomenon or merely the result of errors in the rotorod sampling technique. In order to illustrate the behaviour of aerosols in a simulated atmosphere, an elementary two-dimensional model was constructed using a commercial spreadsheet program. In this model the source could be varied from a uniform source over the whole model (in order to simulate general whitecap aerosol production) or from an elevated source like an industrial chimney. The model was constructed by programming eq. (5) into a uniform grid:

$$\frac{dN}{dt} = w(y)\,\frac{\partial N}{\partial x} + V_g\,\frac{\partial N}{\partial y} + \frac{\partial}{\partial y}\,K(y)\,\frac{\partial N}{\partial y}. \tag{5}$$

The wind profile is represented by $w(y)$, the gravitational fall velocity of the droplets is V_g, and the turbulence mixing is represented by K theory as a function of altitude. N is the concentration of a single size of aerosol, and for the steady state condition, dN/dt is equated to zero. Each grid point in the model space contains the same equation, with the boundary conditions set to zero except for the insertion points which contain non-zero values. The results are plotted in three-dimensional style where the profiles of the steady state condition are presented as a surface. In these plots, the concentration is presented as values along the z-axis (vertical in this three-dimensional plot) and the altitude is plotted along the y-axis (essentially from right to left). The x-axis represents the position from the beach to the sea, and here the land is the surface farthest back into the page. Note that actual dimensions are not given in these results but only the trends of concentrations over

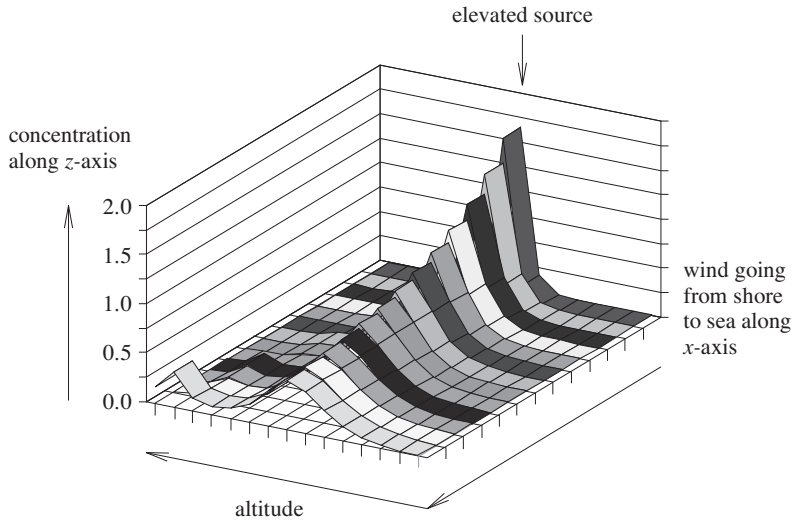

Fig. 5. Spreadsheet model showing the dispersal of aerosol from an elevated source. This is a simulation of coastal pollutants being advected to sea

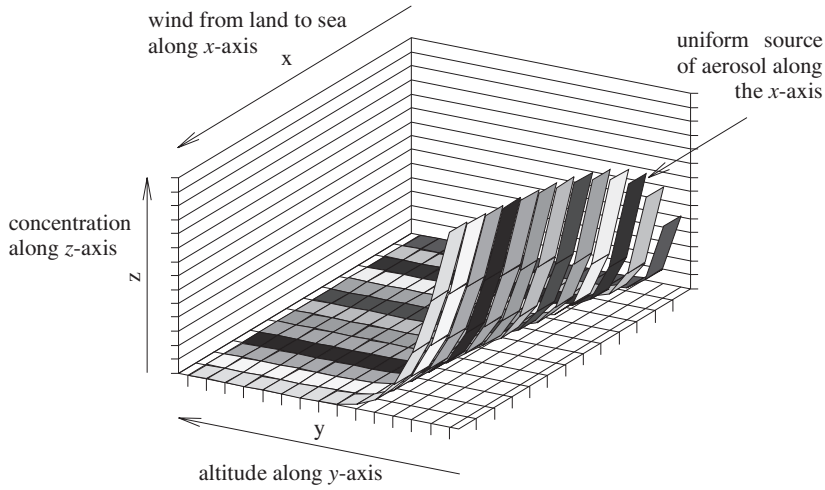

Fig. 6. A uniform aerosol source over the sea simulates the whitecap generation over the ocean as air leaves the land and heads to sea. This simulation shows a 'fetch' effect that is similar to what is experienced in real situations

a two-dimensional space. The particular view used here is shown so that the concentration details close to the water surface can be easily seen. In these cases the wind is blowing from land to sea. Fig. 5 represents the case of an elevated source which, like a chimney, produces a point source of aerosol that is injected into the atmosphere and becomes spread out as it is advected downwind. Note how the plume becomes less symmetrical as it blows out to sea because of the vertical variation in the K value and because there is a general downward drift owing to gravitational forces. The little wrinkles at the high altitude and seaward region are apparently an effect of the boundary conditions which in this elementary model are not real.

The result of another model orientation is shown in Fig. 6. The three-dimensional plot has the wind blowing from land to sea along the x-axis. The concentration of aerosol is plotted along the z-axis and the altitude is the y-axis with the surface being towards the right. In this figure, the results of a uniform source along the sea surface are seen. Here the situation simulates the effect of whitecaps on the marine atmosphere. As the air leaves the land, there is no aerosol of the size represented by this model. However, as it moves over the water surface, sources of aerosol are present which are introduced into the atmosphere. At distances further away from the shore, the aerosol upward flux balances the downward flux and a steady state situation is set up. The transition between land and the 'open sea' in this simulation represents the effect of fetch in the real atmosphere as the clear air picks up aerosol as it moves out to sea. All of the profiles have a maximum value at the water surface and decrease with altitude.

These two simple runs seem to indicate that if there are cases of increasing concentrations of aerosol as a function of increasing altitude above the water's surface, then this is an indication of remote production. Such remote production could be from a number of different sources such as distant surf-produced aerosol or the wake brought about by passing ships. It is apparent then that according to these models our unusual profile measurements in the littoral zone could have been the result of remote aerosol sources. Thus there is always the possibility of some form of coastal influence on the aerosol data when working in the littoral zone.

7.2. Coastal aerosol models

While existing Navy-specific aerosol models such as NAM and NOVAM do a good job of determining the IR/optical climate in the open ocean environment, coastal regions are not so well determined. In the coastal regions, the aerosol has at least two additional sources that can under certain conditions overwhelm the aerosol generated in an 'open ocean' manner. Additional types of aerosols in the littoral zone are those that have been

generated over the land and/or those which are surf-generated. Both of these types of aerosol must have been transported by advection from their points of generation to the coastal region.

Any model which is to predict accurately the paths of advected land generated aerosols in the littoral zone must come to grips with the net effect of anthropogenic aerosols being generated at various points on shore. This is essentially the air pollution problem being extended over neighbouring waters. Shore topology and variations caused by a non-uniform source function can cause large spatial variations in the concentrations in the littoral zone. As they are blown out to sea, however, the various mixing processes in the relatively homogeneous marine boundary layer cause these aerosols to be well mixed and more easily represented by simple models. The variations then merge as the air is advected over the water and as various natural cleansing processes eliminate these aerosols. In the author's opinion, the only way to really simulate this problem is with a large numerical modelling effort. This will require a fine grid, multi-layer meteorological simulation model which could estimate the source strengths of aerosol generators over land. The model would then be able to mathematically advect the aerosol from their sources to remote points of the coastal zone.

A stopgap measure will continue to be needed until such a model is constructed. These complex aerosols from land based sources are represented by NAM as the first single log-normal requiring only 3 parameters to completely describe it. A problem exists, however, in that downwind of cities and other land sources, much of the aerosol is not hygroscopic and could be made up of a number of unusual chemical compounds not expected in a marine model. Even if we determine the actual size distribution of these aerosols, it does not necessarily follow that an accurate Mie calculation can be made, owing to our ignorance of the chemical nature (and index of refraction) or the possible non-spherical shape of the aerosol.

7.3. Advected surf-generated aerosol

The breaking of the surf produces a large amount of white water. It is in this region where the deep ocean waves begin to touch the sea bottom and start to rise. When they break and spill over, air is entrapped by the falling water and mechanically pushed down until the buoyancy of the bubbles causes them to ascend to the water surface. When the air bubbles reach the surface, they burst producing jet droplets in the process. The rising jet drops are injected mechanically a few centimetres above the water surface and then disposed of by meteorological processes. Also in high winds, spume droplets are sheared off the tall surf waves. Thus the surf zone should be a productive source of sea salt aerosols.

8. Early surf measurements

Gathman & Hoppel (1970) used the electrical charge induced on the jet drops from breaking air bubbles in seawater to demonstrate the production of aerosols in the surf zone. Their experiments on the shores of Barbados measured space charge using a $2\,m^3$ Faraday cage mounted on a small fishing boat which crossed the surf with an onshore breeze from ocean to shore through a channel cut (for the local fisherman) in an offshore coral reef. These measurements showed conclusively that the surf zone was the source of the space charge and thus of the aerosol on which the charge resided.

Hooper & Martin (1999) used a scanning LIDAR at Wallops Island, Virginia, in a 1990 experiment. Their LIDAR was located in a region where aerosols were generated by breaking waves. They produced false colour LIDAR backscatter images showing views of both the horizontal and vertical extent of aerosol plumes being produced by the surf and blown to sea by the offshore wind. These images showed plume-like structures originating over the surf and being advected out to sea. These plumes grew from less than 10 metres to 100 metres over a kilometre out to sea but with a corresponding reduction in the backscattered signal. The structure here was seen to be non-uniform and plume-like.

The US Navy sponsored a programme in the 1990s called EOPACE (Electro Optics Propagation Assessment in the Coastal Environments). The objective of the surf production part of the EOPACE effort was twofold:

1) to determine the impact of surf-generated aerosols on visual and IR extinction in a coastal environment, and

2) to evaluate the measurable meteorological/physical oceanographic parameters of a surf zone by which surf aerosol production may be estimated.

Surf generated aerosols were investigated in EOPACE using a variety of platforms in San Diego, CA, Moss Landing, Monterey, CA, and Duck, NC. At each of these locations instruments were located on piers which extend seawards beyond the surf line allowing for a set of measurements to be taken both in and out of the surf during onshore situations. In addition simultaneous shore and boat measurements were made during the San Diego and Duck experiments.

The Applied Research Laboratory, Remote Sensing/Electrical Engineering Department of the Pennsylvania State University, obtained a visual confirmation of the Wallops Island non-uniform plume-like structure of the surf-generated aerosol. Penn State had developed and operated a laser scattering device during EOPACE at Scripps Pier, San Diego

(January/February 1996) and at the Marine Laboratory Pier at Moss Landing (Monterey Bay) in March 1996. This device illumined a wedge-shaped slice above the surf zone at night and allowed visualisation of the plumes through the scattering of the light. This gave a picture of the generation, density and movement of the surf-zone-generated aerosol plumes. Kiser (1997) used geometrical processing of images of two high-resolution CCD cameras to determine the geometry of these plumes. He found that the concentration of aerosols and the size and structure of the plumes are impacted by the air/sea temperature differences, the breaker type, and the local wind.

9. The San Diego surf production model

The situation for an onshore breeze is shown in Fig. 7 in which the boat is positioned just outside of the breaking surf and turned so that it is headed into the wind for its measurement of the aerosol content of the marine atmosphere prior to the addition of the surf aerosol. On shore, the other aerosol spectrometer was positioned to measure the same air sample after the addition of the surf aerosol. Thus, if the instruments were calibrated, the difference between them would be the aerosol produced by the surf and brought to the downwind instrument. A similar situation holds for the offshore breeze. With an offshore breeze the boat instrument would be able to measure the total aerosol (background plus surf) whereas the beach instrument would measure only the background component. The effect of the onshore and offshore breeze on the surf component of the aerosol is

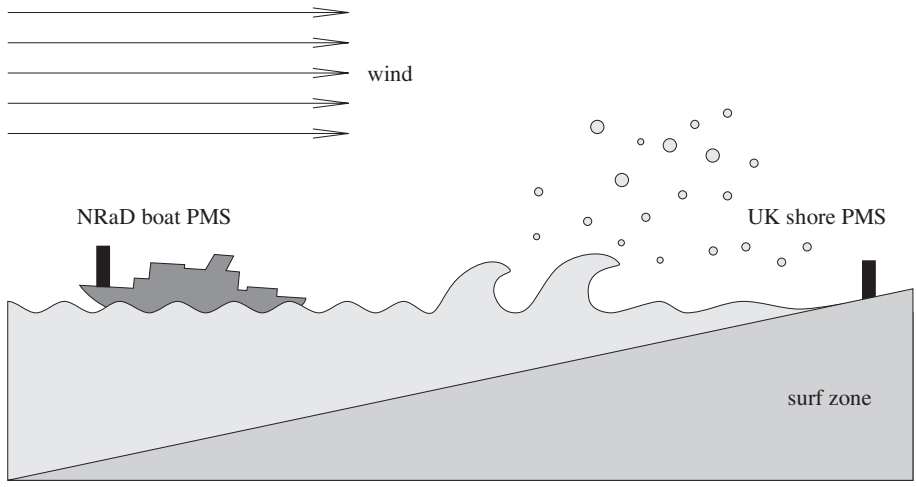

Fig. 7. Schematic diagram of the boat and shore instruments making aerosol measurements during an onshore breeze

illustrated in Fig. 8 where size distribution measurements in both situations are shown in the lower two panels. The usual time for the offshore breeze is during the evening and night hours as the land cools and the coastal aerosol is blown back out to sea. On the other hand, the situation during the day is usually that of the onshore breeze and here the surf aerosol is blown over the shore.

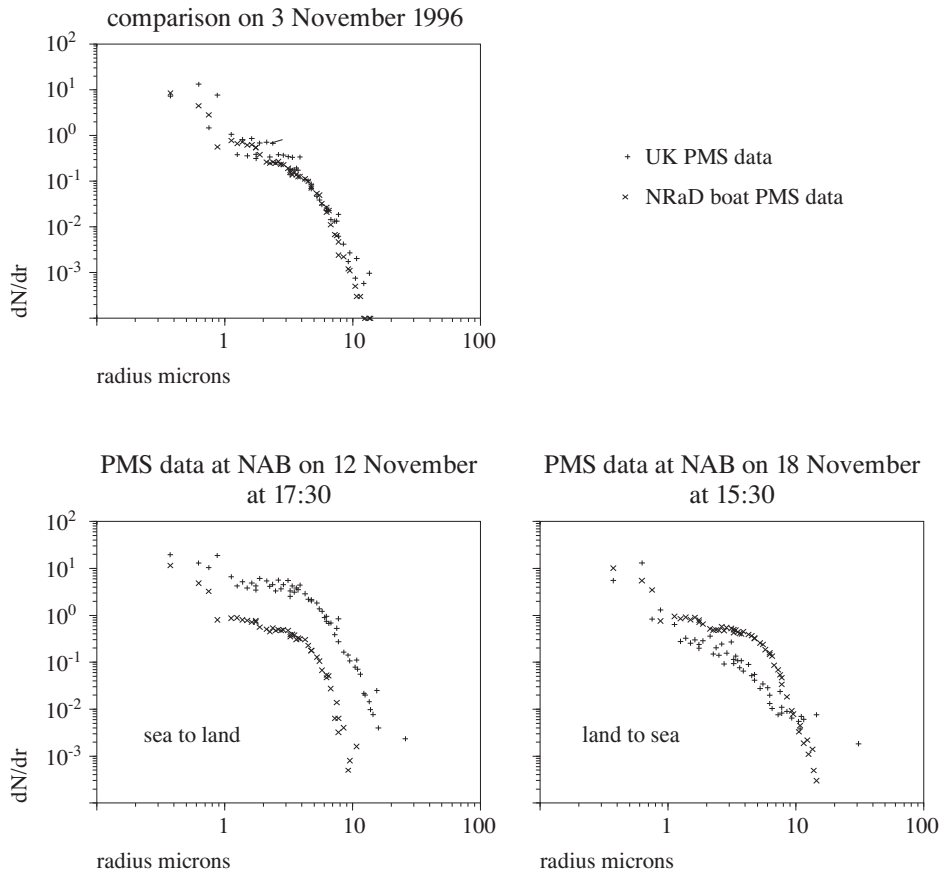

Fig. 8. Simultaneous aerosol size distribution measurements from the boat and the beach under various conditions. The top figure shows the comparison between the two instruments prior to the experiment

Figure 8 shows some average aerosol spectra obtained by Gathman & Smith (1997) using two spectrometers. The top panel shows the calibration comparison results when both of the instruments were located together on the cliff overlooking the Pacific Ocean at the SSC–SD laboratory. The instruments are distinguished by the style of the marks used. The plus sign '+' indicates the shore instrument and the X indicates boat

instrument data. It is seen that the two instruments agree reasonably well when in the comparison mode. When, however, the instruments are located on the boat and the beach, during an onshore breeze, it is seen that the land measurements are an order of magnitude higher than the boat measurements. On the other hand, during the few episodes in which after-dark measurements were made and in which an offshore breeze was blowing, the situation looked like that shown in the lower right hand side of the figure. It appears that on the whole, dN/dr values from the boat measurements were greater than on land. It appears that perhaps the airflow about the boat and box tended to discriminate against the largest aerosol and this instrumental factor would account for the crossing of these lines at the far end of the spectrum.

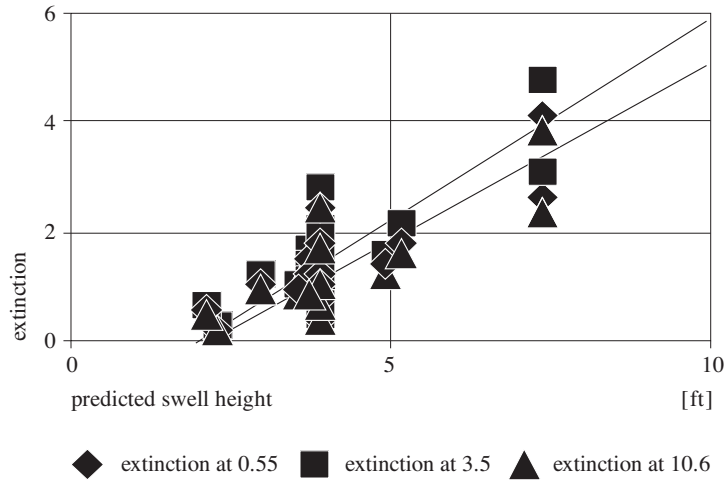

Fig. 9. Extinction calculations plotted against swell height predictions

Because of the predictable behaviour of the aerosol production on the beach based on the swell predictions for the San Diego area as shown in Fig. 9, it seems possible that a forecast model could be constructed for this region.

10. Summary and conclusion

Marine aerosol research over the last three decades has led to some important tools for approximating the propagation characteristics along IR paths over the world's oceans. Two lines of research have been in operation. Purely physical investigations have produced much insight into the mechanisms of marine aerosol production. Up to this time the problem has been found so complex that a truly integrated model has yet to be

developed. However, semi-empirical models exist which work over the world ocean and predict scattering and absorption from the wave tops to above the marine boundary layer to a reasonable degree. Open ocean regions are most accurately modelled at the present. In coastal areas there is the additional complication of further sources of aerosol, which must be correctly taken into account. In the littoral zone, fetch, surf- and land-generated aerosols must also be borne in mind.

The problem of fetch has been worked on with some success. The surf aerosol has been shown to be a significant source of aerosol in the littoral zone. Surf source functions appear to be dependent on wave height and bottom characteristics. Once generated, if surf aerosol is advected from the surf zone out to sea, it becomes a factor in propagation prediction. Land generated aerosols may also be transported from their regions of generation to the coastal waters where they affect propagation. It appears that to be completely successful in the construction of simulation coastal models, some form of fine grid numerical model must be developed whereby complex wind structure, diverse aerosol sources and atmospheric stability and humidity can be taken into account.

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