Characterizing large aerosols in the lowest level of the marine atmosphere

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ABSTRACT

This paper discusses techniques to describe the aerosol and the electro optical properties of the marine atmosphere from 15 meters down to the tops of the highest wave. Emphasis is placed on the experimental Rotorod technique to measure the concentrations of giant sea salt droplets. Data from these devices are parameterized using a lognormal function that is in turn related statistically to parameters such as wind speed, atmospheric stability and height above the surface. The new lognormal function is combined with the Navy Aerosol Model (NAM) to develop a first version of the Advanced Navy Aerosol Model (ANAM). Thus, ANAM allows the construction of an aerosol size distribution at any level from the wave tops to 15 meters and the assessment of electro optical parameters from this distribution using Mie theory. The first results of the ANAM model are compared to experimental data.

Keywords: marine atmosphere, aerosol model, rotorod technique, ANAM

1. INTRODUCTION

The Navy has a particular interest in determining the electro optical properties of the lower layers of the marine atmosphere for the assessment of the performance of electro optical surveillance systems. The propagation characteristics of the marine atmosphere are strongly affected by scattering and absorption of radiation by molecules and aerosols. Although the molecular composition of the air is relatively constant, the aerosol concentration in the marine boundary layer can vary by orders of magnitude. Consequently, the aerosol provides an extremely varied component to the deterioration of the propagation of electro optical energy.

The amount of deterioration of energy can be calculated from Mie theory. This requires a knowledge of the aerosol size distribution and the assumption that the scatterers are spherical in shape and that their composition is known. Within the marine boundary layer, the majority of aerosol which affect the transmission of infra red energy consists of large marine aerosol, i.e., hygroscopic droplets of water and dissolved salt with a spherical shape. The surrounding ambient relative humidity controls the actual composition ratio between salt and water in the drop and thus the index of refraction of the droplet.

The first generally available computer code used to calculate the atmospheric transmittance and radiance for a given atmospheric path at moderate spectral resolution was LOWTRAN-2 designed by Selby and McClatchey[1]. This code was produced by the US Air Force for Air

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Force applications. Over the years, this code improved in both accuracy and scope but still was not really applicable to many Navy needs. Finally with the advent of LOWTRAN-6,[2] the scope of the model was extended to include more of the needs of the tri-service community when an Army and a Navy aerosol model were added.

The Navy Aerosol Model (NAM)[3] determines the aerosol size distribution from a statistical analysis of many available aerosol size distributions obtained from the literature of the time and relates the characteristics of these size distributions to meteorological parameters which occurred during the sampling of these data. This model defines the marine aerosol size distribution as a set of three lognormal curves combined to represent the total distribution. The model is considered to represent the aerosol at shipboard level where all of the measurements that went into the construction of the model were obtained.

In a well mixed, shallow marine boundary layer, it is not too bad of an assumption to say that the aerosol size distribution is essentially independent of height. It was necessary however to determine in more detail the vertical structure of the aerosol extinction to ascertain the effects of aerosol on slant path ranges for shipboard systems. Such a model, the Navy Oceanic Vertical Aerosol Model (NOVAM), was constructed[4,5] and has recently been coded into the next LOWTRAN / MODTRAN series of codes and addresses the propagation characteristics at shipboard level and higher.

In recent years, the assessment of the propagation characteristics at altitudes below shipboard level has become more and more important. Close to the surface, a considerable amount of large aerosol (radius over 10 μm) is found, which severely hampers the propagation of infra red radiation. The large aerosol predominantly consists of freshly produced sea spray droplets. Their concentration is strongly related to the efficiency of aerosol production mechanisms such as bubble mediated production and direct wave tearing, which are a function of wind speed.

Measurements in this region are sparse and the resolution often limited because of the experimental difficulties involved, such as stability of the sensors (when mounted on buoys), motion (ships) and air flow distortion (ships, platforms). Pioneering efforts to measure the aerosol concentration close to the sea surface were made in the sixties[6] and vertical profiles were first measured in the seventies[7,8]; see De Leeuw[9] for an overview. Over the last decade, the Rotorod technique[10,11] used by TNO and more recently by SSC San Diego is responsible for the majority of the information about the (large) aerosol concentrations just above the sea surface up to a few meters. These measurements have shown that the aerosol concentration close to the surface is strongly influenced by the waves. The statistical relations between e.g., concentration and wind speed, which work well at a height of about 10 meters become increasingly inadequate at lower elevations.

The experimental data show a need to extend the modeled aerosol size distribution to larger diameters (as compared to the distribution used in NAM) and to introduce a dependence of the concentration on height, because close to the surface concentrations of very large particles are not negligible and vertical gradients of these particles may be important. The need to extend the modeling to larger diameters for a better assessment of long wavelength transmission properties of the marine environment had already been recognized,[12] but the sparse and scattered experimental data has so far hampered the actual extension of the NAM model.

The present contribution focuses on the extension of the empirical NAM model towards the sea surface. The extension consists of an element that specifically describes the concentration of the larger aerosol in terms of meteorological parameters. The governing relations are empirically determined from an analysis of experimental data.

2. OBTAINING AEROSOL DATA CLOSE TO THE WAVE TOPS

Close to the sea surface concentrations of smaller aerosols ("smaller" meaning sizes of less than 10 μm radius) can be measured by an optical device such as the Particle Measurement Systems devices. With these devices (and with certain assumptions) a size distribution can be obtained within a reasonable period of time because of the comparatively large quantities of these smaller aerosols. Larger aerosols are more difficult to characterize due to the limited sampling volume of the devices. The small concentration of large sea salt particles requires large samples of air in order to get statistically significant results. In addition the inertia of these large droplets makes the sampling of these droplets difficult in that because of their mass, they simply will not follow an air stream around sharp turns in sampling tubes. The simple rotorod device[10,11] turns out to be the best method developed up to the present of obtaining the required data. Such a device is shown symbolically in figure 1.

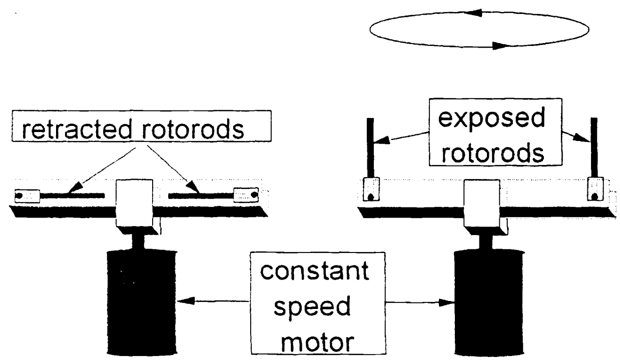

Figure 1: Diagram showing a rotorod device. The figure on the left shows the device configuration when the motor is stopped. The figure on the right shows the position of the rods as the motor is rotating at full speed.

2.1 Rotorod impaction samplers

The rotorod device consists of a commercially obtained retracting head, which supports two stainless steel rods, mounted on a stabilized constant speed motor. The rods are 1.6 mm in diameter with a polished mirror flat surface at one side over a width of 1.5 mm. Silicone is sprayed over the polished surface to collect the particles. Springs hold the rods into the retracted position when the device is not rotating. Thus it is protected from collecting aerosol until the motor is started. The square U shaped body of the device keeps the coated surface of the "D" shaped rotorod well hidden from the environment. However, once the motor is started, centrifugal force keeps the balanced rods vertical and the flat surface facing into the direction of rotation so as to impact the droplets in its path with the polished-coated face of the rotorod. Due to their own inertia, the particles suspended in the air collide with the moving rod leaving an image on the silicon coating that can be analyzed with a microscope afterwards. The volume that the device sweeps out is related to the rpm of the motor and the radius of the arm and the area analyzed under the microscope, and the length of time the device is rotating.

The Rotorod impactors can reliably sample the aerosol concentrations in the air if some conditions are met. The sample volume must be constantly refreshed. This condition is fulfilled because of the natural convection (wind) and because of the aspiration due to the rotation of the rods.[10] Secondly, the particles must be efficiently collected by the impaction rods. It was found both experimentally[13] and theoretically[14] that the rods efficiently (>90%)

collect particles larger than about 10 μm. Third, the sampling time must be such that sufficient particles are collected for sampling statistics, but that the individual impacts do not overlap.

The particles leave an imprint in the silicon coating which corresponds to their diameter at the time of impact. The imprints can subsequently be sized with a microscope and an imaging technique. A ruler with 10 micro meter grids is used for calibration of the scale. For correct sizing, it is important that the microscope is focused on the top of the silicon coating. During transport and further processing of the rods, the wet aerosols may partially evaporate since they find themselves in an environment with lower ambient humidity than close to the sea surface. Ultimately, the aerosols may evaporate to salt crystals deeply embedded in the silicone coating. These crystals are only seen when the microscope is strongly defocused and they do not interfere with the determination of the dimensions of the craters at the top of the silicon coating.

The microscope is equipped with a CCD camera and a frame grabber system. Typically, multiple microscopic images are saved on disk for each rod. Subsequent processing by image analysis software is used to reveal the particle size distribution. The software used for image analysis by TNO was developed by TNO itself,[15] whereas SSC San Diego uses a commercial package. One of the characteristics which can be determined by the SSC San Diego package is if a particular image is convex or concave. Obviously, images of droplets must be convex in shape. Other features of both software packages which are used in the analysis of the images, are the asymmetry factor and the mean diameter of the image. If a droplet is very asymmetric, it is dropped from the analysis.

The results of multiple microscopic images added together from the same set of rods is used to increase the sampling volume of the sample. A composite aerosol size distribution consisting of the total number of droplets counted in all of the microscopic areas are analyzed. The volume of space in which these droplets were encountered is determined by the radius of the rotation path, the total area of the microscopic analysis, the speed of rotation of the device, and the time of exposure to the atmosphere.

2.2 Reliability of the rotorod technique

Extensive tests have been conducted to determine the reliability of the rotorod technique. In 1983 when the technique was first used by TNO, the optimum experimental conditions (such as sample time and coating) have been determined, the sizing algorithm has been tested with mono-disperse particles of known diameter, and the concentrations of particles measured with rotorods and optical devices have been compared.[10,11] Nevertheless, in view of the importance of the rotorod measurements for the development of near-surface aerosol models, these tests have been repeated and extended.

In a first series of tests mono-disperse aluminum oxide particles of known diameter (10 and 20 μm) deposited on glass were sized with the microscope - imaging system. This resulted in a narrow (i.e., a few μm wide) distribution of diameters around the correct diameter. The system was also tested with calibration targets of circles and ellipses (black ink on paper) of known diameter (100 μm and up) and aspect ratio. In all cases the system sized correctly and determined the proper aspect ratio.

In the next series of tests the mono-disperse particles were collected on the coating of the spinning rotorods. This experiment was performed in a clean room to avoid contamination of the rods by dust and other particles, which would hamper the test. The system sized the 20 μm particles correctly, but slightly overestimated the size of the 10 μm particles. In addition, the standard deviation increased (i.e., the distribution around the center diameter had widened).This result shows that the presence of a coating has a small influence on the sizing. Possible causes are "smearing" of the coating upon impact and particles hitting the rod under an angle (leaving a non-circular imprint).

Two rotorod devices were then mounted under identical conditions on the beach of the North Sea (i.e., same height, exposure, and as close together as possible without interference). The use of pairs allowed for a study of reproducibility of rotorod measurements. In all, 25 pairs of measurements were compared. On the average, the two aerosol size distributions determined for each pair differed less than 10%. The mean difference between the pairs of each dN/dr measurement for a particular size and standard deviation of these values are plotted in figure 2. The figure shows high consistency in rotorod data for sizes up to 50 μm. The errors for the largest sizes is due to the very small concentrations of these large particles.

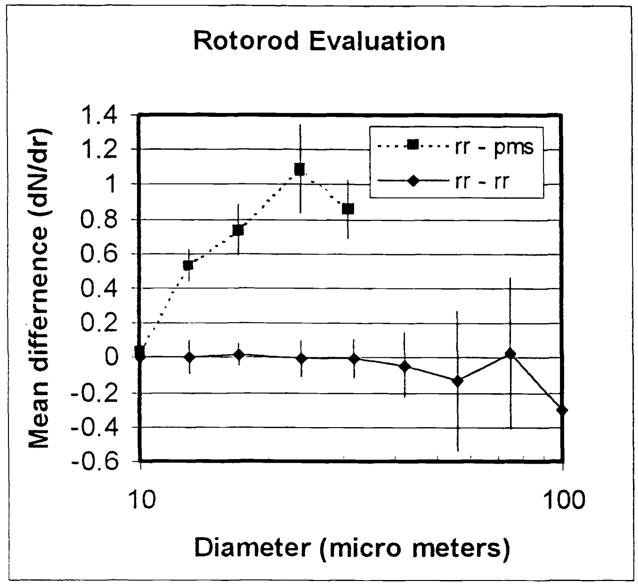

Figure 2: A plot showing the results of the series of experiments on the North Sea aerosol described in the text. The dashed line is the result of the comparison of the PMS probe and a near by rotorod system while the solid line represents the comparison between two rotorod observations.

In a final test, the rotorod devices were mounted next to a Particle Measurement Systems (PMS) optical aerosol counter (CSASP-100HV probe, 0.75-45.5 μm diameter). The PMS probe and the rotorod system have an appreciable overlap in diameter range (from 10-45 μm). At the start of the overlap region (10 μm), the aerosol concentrations as measured by rotorods and PMS were identical (to within a few percents). For larger diameters, the rotorod system yielded significantly higher concentrations than the PMS. The difference amounted to a factor of 10 for the largest particles. The results of these tests together with the standard deviation of the measurements (shown as error bars) are plotted in figure 2

We explain the observed discrepancy in measured concentration by the reduced efficiency of the PMS system for larger particles. At the end of its range, the statistical reliability of the PMS system reduces quickly due to the small concentrations of larger aerosol in combination with the small sampling volume of the probe. On the contrary, the rotorod devices which have a large sampling volume can reliably measure the small concentrations of larger particles.

2.3 An example of low altitude data: MAPTIP

The MAPTIP experiment (Marine Aerosol Properties and Thermal Imager Performance trial)[16] was dedicated to the evaluation of the 3-5 and 8-12 micrometer wavelength propagation characteristics near the ocean surface and their relationships with marine-generated aerosols, turbulence and meteorological factors. The key facility used in MAPTIP was the "Meetpost Noordwijk" (MPN), an oceanographic research tower situated at approximately 9 km from the Dutch coast (52° 11' 51.6" N, 04° 22' 57.6" E).

SSC San Diego and TNO operated PMS optical devices and rotorod samplers on MPN to characterize the aerosol concentration between 0.3 to 50μm radius below 15 meters. The SSC San Diego measurements were made with a box that could be raised and lowered to the water surface with its winch that was fastened to the overhang part of the MPN tower. This design tended to minimize any flow distortion about the legs of the tower. The box was equipped with optical devices and rotorod samplers. Due to the configuration of the system, rotorod measurements were restricted to four altitudes.

The TNO measurements were made using a horizontal mast extending 10 m North from the NW-corner of the platform. At this distance flow perturbation due to the platform is negligible[17] as well as the influence of waves breaking on, or reflected at the platform structure unless the platform shields the samplers from the wind. Rotorod samplers were attached to a 2 m long pole which was lowered from deck height (12 m) to about 1-2 m above mean sea level (fixed-level measurements), thus yielding measurements between these heights. In addition, the pole was attached to a small wave rider buoy. This allowed for measurements at heights between 50 cm and about 2 m with respect to the instantaneous sea level (wave-following measurements). A complete vertical profile from these devices (15 measurements) could be obtained in about 30-40 minutes.

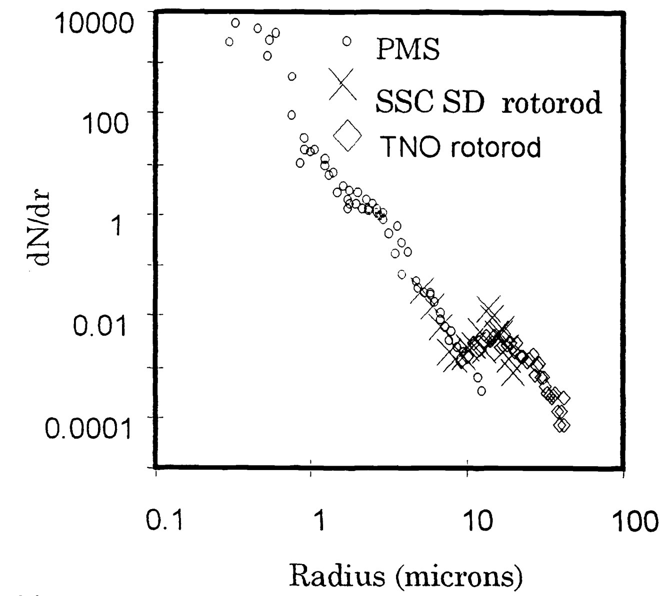

Figure 3: A data plot from a MAPTIP experiment in which the dN/dr values from three devices with overlapping range are measured at approximately the same place and time.

The simultaneous use of both optical devices and rotorods allows the construction of aerosol spectra over an appreciable range. This feature makes the MAPTIP data set particularly suited for the development of ANAM.[18] An example of a combined MAPTIP data set is shown in figure 3. On this plot are shown the data from an experiment on 25 October 1993 in which simultaneous samples were obtained by three independent systems at (07:10 GMT). The data shown in the figure were taken at an approximate altitude of 12 meters and include both types of rotorod systems as well as a Particle Measurement System (PMS) Active Scattering Spectrometer Probe (ASSP), type 100 with size range of from 0.2 to 15 micrometers radius. It is obvious from placement of points in the figure, that although the ranges of the devices differ from each other that there is sufficient overlap, which allows for a much broader composite spectrum of the aerosol size distribution than each of the individual distributions.

The application of rotorod devices is physically limited by the reduced collection efficiency of smaller particles and in practice also by the resolution of the microscope which is on the order of five microns diameter. The TNO system was designed to eliminate those particles smaller than 10 μm. The SSC San Diego system on the other hand (using a higher magnification) sized the smaller particles even though their images were not as sharp as for the larger particles. When compared to the PMS optical system, it seems that the alignment of these very small particles on the SSC San Diego rotorod slides with the PMS spectrum is not bad in the region of from five to ten microns radius. Of particular interest in the figure is the inability of the PMS system to see the "hump" of the aerosol size spectrum that was seen by the two rotorod devices. It is believed that this is due to the reduced statistical reliability of the PMS probe for larger particles as discussed above.

3. PARAMETERIZATION OF LOW LEVEL AEROSOL

The aerosol size distributions from MAPTIP were fitted by an analytical curve for parameterization purposes. These curves were analyzed for a maximum in dN/dr values and associated mode radii in the region between 5 and 20 microns in order to find the mode radius and mode amplitude represented by the large aerosol group (shown as the "hump" in Figure 3). Gathman[19] analyzed the characteristics of the mode radius and amplitude as a function of altitude and meteorological conditions. Whereas the mode radius did not seem to vary significantly over the range of altitudes and meteorological conditions, the mode amplitude was found to be a function of both altitude and wind speed.

The parameterization makes it possible to extend the NAM model with an element to describe the very large sea salt aerosol from the sea surface up to deck height (approximately 15 m). This element will be a lognormal function called the fourth mode. The total aerosol size distribution will then be the superposition of the three original lognormal functions of the NAM model, plus the fourth mode. The new model is named Advanced Navy Aerosol Model (ANAM).

The large sea salt aerosols in the model are represented by a single lognormal size distribution:

$$\frac{dN_4}{dr} = \frac{A_4}{f_4} \cdot \exp\left\{ -C_4 \cdot \log\left(\frac{r}{r_4 f_4}\right)^2 \right\}$$

where C_4 is the width of the lognormal distribution, f_4 is the growth factor, r_4 is the mode radius and A_4 is the mode amplitude. The parameterization yielded constant values $C_4 = -5$ and $r_4 = 15$ μm over the range of altitudes and meteorological conditions in the MAPTIP data set. In this first version of ANAM, the humidity effects on the droplet sizes are represented by a swelling factor, f_4. The swelling factor for sea salt aerosol is a strong function of relative humidity and can be represented by a simple approximation.[20]

The amplitude of the fourth mode, A_4, was found to be related to wind speed U, the height above the mean water level z and the atmospheric stability, characterized by Monin-Obhukov stability length L:

$$A_4(U,z,L) = A_4^{\,0}(U) \cdot p(z,L)$$

The functional dependence can be separated in a mode average amplitude $A_4^0(U)$ for a height of 10 meters, which is only a function of wind speed, and a scaling factor p(z,L) with respect to height above the water, which is a function of altitude and atmospheric stability. The function p(z, L) is based[21] in part on the classic non-dimensional profile function φ(z/L) of Businger and Dyer.[22] Only cases in which L is negative are allowed in this preliminary empirical model, as all of the data from MAPTIP were obtained under unstable conditions.

As mentioned in the introduction, the ultimate goal of this work is to assess the electro-optical properties. To this end, the aerosol extinction is calculated from the ANAM size distribution using Mie theory. This requires the Mie scattering coefficient, which is a function of the refractive index of the aerosol. ANAM assumes that all water produced aerosol consists of a solution of sea salt dissolved with water in equilibrium with the ambient relative humidity. With this assumption, the refractive index of the aerosol can be determined from look-up tables.

The first version of ANAM requires inputs consisting of regularly available meteorological parameters: air temperature, relative humidity and current wind speed at 10 meters height, the 24 hour average wind speed, sea surface temperature and an air mass parameter (derived from visibility, nephelometer data, radon concentration or condensation nuclei concentration).

4. THE CONSISTENCY OF THE MODEL

The objective of this work is to represent the aerosol distributions measured during MAPTIP by a model that should extend the existing applicability ranges of NAM/NOVAM. Although the empirical ANAM model was developed from MAPTIP data, it should be constructive to see how well the model represents the actual set of measurements.

The meteorological data at the time of the measurements plotted in figure 3 were used as input to ANAM. Figure 4 shows the experimental data, as well as the aerosol size distribution calculated by the four component ANAM model. The model result is represented by the solid line and adequately matches the measurements.

An example of a profile measurement is shown in figure 5 where the concentration of 15 μm particles as measured with TNO rotorod samplers on 22 October 1993 are plotted with respect to altitude. The oval symbols represent the data obtained at fixed height, whereas the solid square symbols represent the values from the wave rider buoy. Again, the solid line gives the ANAM prediction.

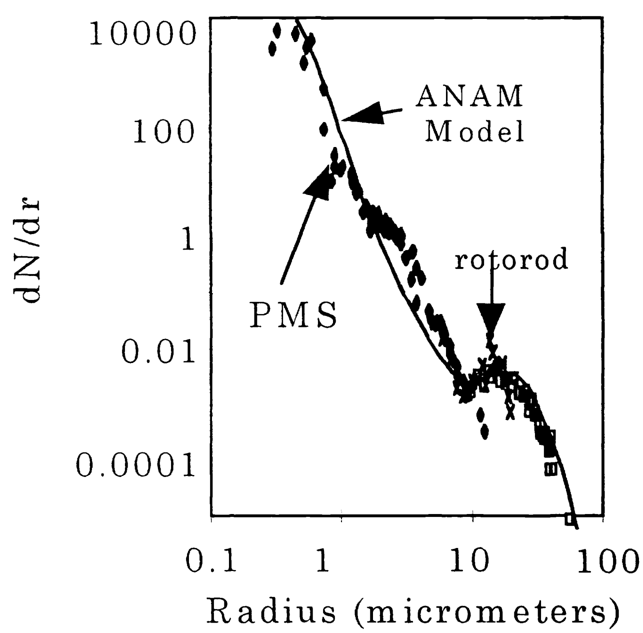

Figure 4: The dN/dr profile obtained during MAPTIP shown in figure 3 but with the modeled ANAM aerosol size distribution superimposed over that data as a line.

Figure 5 shows that the ANAM prediction matches the experimental data well down to about 2 meters above the surface. Below this height, the prediction and the experimental data do not agree. This may indicate that the profile function p(z,L) is not yet fully adapted for the presence of waves. However, the figure also seems to indicate a discontinuity between the fixed height measurements and those made with the buoy. The exact cause for this is not

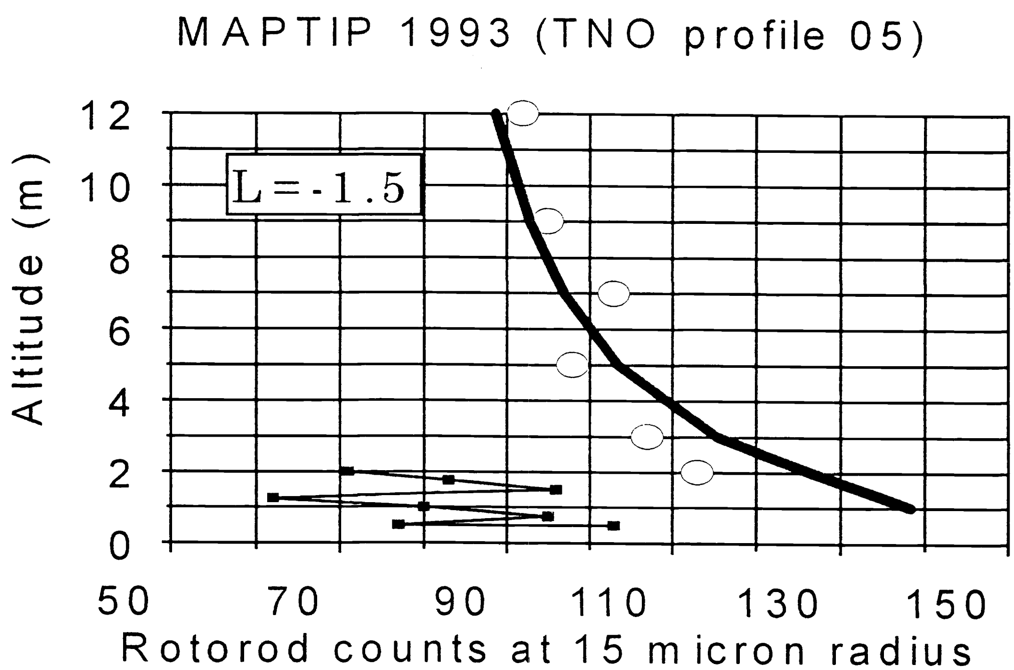

Figure 5: Profiles of TNO rotorod counts of 15 micron droplets and the ANAM prediction of these sizes for 24 October 1993 at 10:40 GMT. The stability for this case is represented by a mixing length, L=-1.5 meters. The large circles are the measured rotorod data from devices on the tower mounted at fixed heights above the water. The small square symbols connected by a line are those measurements done at the same time from the wave follower buoy. The thick line is the ANAM prediction for this case.

exactly known, but may arise from the difference in the definition of "altitude". The float measures at a fixed height above the actual water level, whereas the other measurements are made at a fixed height with respect to the mean water level. This means that the float measurements in part contain information of the aerosol concentration below the wave crests, whereas the fixed height measurements only contain information of the aerosol concentration above the wave crests. In addition, close to the water surface effects such as non-uniform production of aerosols from whitecaps in the moving waves may become important. This discussion shows that the interpretation of the data very close to the surface is not straightforward. Efforts are currently underway to improve the ANAM model at very low altitudes on a theoretical basis.

Figure 6 shows an example of the vertical variation of the electro optical properties of the atmosphere as predicted by the ANAM model for a typical unstable atmosphere (wind speed of 9 m/s and Monin-Obhukov length of –8.5 m). In this figure are plotted the volume extinction coefficients for the visual wavelength (0.55 micrometers), the mid IR, (3.5 micrometers), and the far IR (10.6 micrometers) as they vary from 10 meters down to the wave tops. On the top side of the plot are three arrows which represent the values that NAM gives for these wavelengths. We see first of all that the additional ANAM component for the larger aerosols primarily affects the mid and far infrared extinction values. Compared to the NAM values, ANAM predicts an increase of the mid and far infrared extinction values at 10 m, thus reflecting the influence of the larger aerosols even at the height of 10 m. The vertical variation of the extinction coefficient between 10 m and the wave crests is about 200% for the infrared wavelengths, whereas the variation for the visible wavelengths is about 25%.

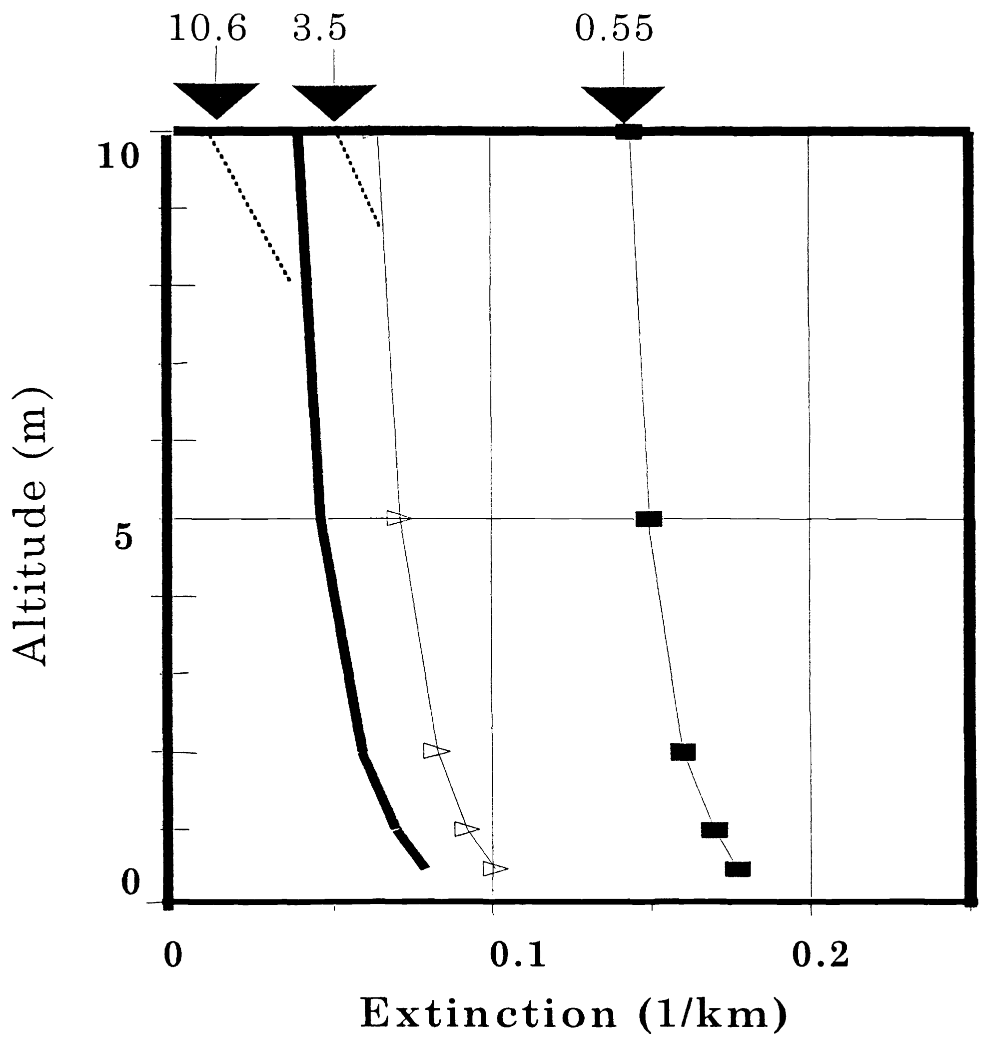

Figure 6: Extinction profile calculations from ANAM for the visible (solid square markers), 3.5 micrometer IR (open triangles), and 10.6 micrometer IR (heavy solid line) wavelengths. The wind speed is set at 9 m/s, and the mixing length is –8.5 meters. The arrows on top of the figure refer to the NAM predicted values for the same conditions.

As an indication of the effect of the vertical variation of the infrared extinction coefficient on the transmission over a 10 km path, we assume that molecular and aerosol extinction are of the same magnitude. In that case, the infrared transmission near the wave crests is only 36% of the transmission at 10 m height. This first example shows that the effect of large aerosols as represented by the ANAM component cannot be neglected when assessing the electro-optical propagation characteristics in the marine atmospheric surface layer.

5. CONCLUSIONS

The ANAM model matches the aerosol size distributions observed during the MAPTIP experiment within acceptable limits. The question is whether or not it will describe other sets of observations. This testing with other data sets will be the first round verification of the model.

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