Modeling of aerosols in the marine mixed-layer

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ABSTRACT

A model is developed to calculate the vertical variation of aerosol extinction coefficients throughout the marine atmospheric boundary layer. It is a mixture of empirical and physical models, formulated to describe the often observed non-uniform, but also non-logarithmic, profiles. The physical model is based on the dynamical processes affecting the production, mixing, deposition and size of the aerosol within the marine atmosphere. A status report is presented including a critical evaluation.

1. INTRODUCTION

The height variation of electromagnetic scattering and absorption at wavelengths in the visible to the infrared is of considerable interest in many cases associated with vertical and slant path observations with electro-optical (EO) systems. Problems arise, however, in the evaluations of EO propagation characteristics because existing empirically derived expressions for aerosol scattering and absorption contributions to extinction were formulated for single levels. Variations in the vertical may be very large. The Naval Aerosol Model (NAM)[1] as found in Lowtran VI is an example of this limitation. NAM should be restricted to horizontal path calculations because all data used in its development was derived from deck level measurements, and no real provision was made for vertical structure in the aerosol concentration.

When vertical structure is required for slant path calculations, assumptions are required to extend surface values to higher levels. A usual approach in existing empirical models is to assume a logarithmic decrease with height using effective scaling heights.[2] This does not allow for the use of the extra information available from observed meteorological profiles, which are often non-logarithmic. This additional information is expected to improve the accuracy of the predicted extinction profiles.

This paper describes an approach being formulated to put vertical structure into the extinction prediction using a mixture of empirical and dynamical models. Prediction in this context does not imply prediction in time but rather an estimate of optical extinction given a set of atmospheric parameters which can be used with the empirical-dynamical model. The model is referred to as the Naval Oceanic Vertical Aerosol Model (NOVAM).[3]

The model for the structure in extinction was designed to describe non-uniform but also non-logarithmic aerosol distributions which are known to exist throughout the marine atmospheric boundary layer. The model is restricted to the marine boundary layer, hence the designation Oceanic in its title. The model is distinct from land-based models, because of the marine type of scaling used for the turbulent controlled processes near the sea surface, and because of the model used to determine the surface concentrations (NAM). The structure is a function of the growth of the particles due to height varying relative humidity and of turbulent controlled processes. The turbulent processes produce, deposit and mix the aerosol and also determine the depth of the mixed layer itself.

We address the following aspects of the multi-component model. The physical background of the turbulent controlled processes and of the growth features caused by relative humidity effects are presented in section 2. In addition, extinctions found under solid cloud decks will be discussed. Considerations of the physical constraints as they are treated in NOVAM are presented in section 3. A critical evaluation of the several crucial parts of NOVAM with reference to its intended use appears in section 4. Finally, conclusions on the present status and future of the approach and model will be given.

2. BACKGROUND ON PHYSICAL MODELS

2.1 Atmospheric Boundary Layer Turbulence

The concentration of aerosols at various levels in the marine boundary layer is determined by a number of inter-dependent complex processes. Multi-variable models of this behavior are still in a rather crude state of development.

The dynamic equations neglect advection, the effects of which are included through the air mass parameter. Thus, the sources and sinks for the aerosol particles present in the boundary layer are by transfer through the sea surface or by entrainment and gravitational fallout from the non-turbulent troposphere immediately above the marine boundary layer. The vertical distribution of aerosols throughout the mixed-layer is determined by the turbulent transport processes, which in turn are influenced by the relative humidity. The simplest case is the mid-latitude (as opposed to tropical) boundary layer with a strong inversion, which is well-mixed. When weak cumulus convection is present, a two layer model must be used to describe the aerosol structure.

In our empirical-dynamical approach the starting point for modeling aerosol properties is the continuity equation including source, sink, vertical transport and 'horizontal advection' terms for the domain. Since the marine boundary layer is of limited vertical extent, both the surface and the top of it are potential source or sink regions. In the simplest well-mixed case four scaling regimes exist within the marine boundary layer. These regimes are differentiated by the relative dominance of the different processes found within them. These are designated (see Figure 1) as the free troposphere above the mixed-layer (p), the mixed layer (f), the turbulent surface layer (c) and the diffusion sublayer (d).

The nature of the various atmospheric transfer processes permits us to identify certain height regimes where the analysis can be simplified by scaling arguments. For example, near the surface (within 10 meters of the ocean) the particle flux is generally considered independent of height. A thorough consideration of air-sea particulate transfer processes by Fairall and Larsen[4] addressed the relative importance of turbulent and diffusive transport mechanisms in this so-called constant flux layer and the diffusion dominated sublayer. Using a standard micrometeorological formalism, the surface source and sink properties can be described in a surface layer scaling context.

The mixed-layer constitutes about 90% of the boundary layer. Models based on its special properties are usually referred to as mixed-layer models. The classic mixed-layer

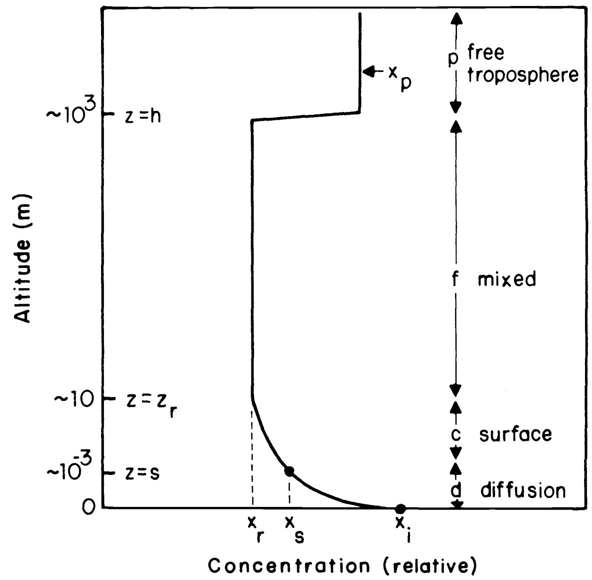

Figure 1. Schematic diagram of atmospheric scaling regimes (nonlinear scales)

model[5] is considered to be applicable to the mid-latitude marine regime where mixing in the boundary layer is dominated by reasonably homogeneous turbulence produced by surface shear and/or convection generated by warm water or cloud top radiative cooling. The mixed-layer model is one of the simplest because it ignores the details of the vertical transport processes by assuming that the turbulence is strong enough to maintain a well-mixed boundary layer. This implies that the fluxes in the boundary layer have a linear dependence on height and that we need only to specify the value of the flux at the bottom and top of the boundary layer.

The definition of the mixed-layer implies that particles of less than 30 μm radius are expected to obey mixed layer scaling[6] which is usually taken to mean the absence of a vertical gradient. Since the mixed layer formulation only requires that the gradient be constant with respect to time, clearly a constant vertical gradient is permissible. Davidson and Fairall,[7] using physical arguments of Wyngaard and Brost,[8] show that a mixed layer gradient for a surface generated aerosol component (e.g. sea salt) would be given by

$$\partial X_{sr}/\partial z = -1.5\ (S_r - V_d X_{sr} + 2.5\ W_e X_{sr})/hw_* \quad (1)$$

where X_{sr} is the concentration of the sea-salt aerosol in the mixed-layer at height z, S_r is the surface flux, V_d is the effective fall velocity, W_e is the entrainment rate, h is the height of the boundary layer, and w_* is the convective scaling velocity.

With representative values for the scaling parameters, gradients predicted by Eq. 1

would be dependent on particle size. For the very small particles under typical conditions the height variations are usually neglected. For larger particles this is generally not true. Also it is important to note that the role of relative humidity, which affects the gradient through both V_d and X_{sr}, has not been considered. This will be discussed in more detail in section 2.2.

Another climate regime is also globally important. This regime, which is visually characterized by "fair weather" or scattered cumulus clouds, is common over the ocean in the trade-wind latitudes. Physically, the presence of the cumulus towers significantly modifies the transport properties of the boundary layer. The cumulus towers dominate the upward transport of moisture, heat, and aerosols. This upward transport, which is confined to narrow columns that represent only a few per cent of the horizontal area, is balanced, in part, by a much more broadly spread downward transport (between cloud subsidence). Albrecht[9] has developed a model that is the trade-wind equivalent to the mid-latitude mixed-layer model. The application to aerosols is described by Davidson and Fairall.[7]

2.2 Aerosol humidity effects

In the mixed-layer the relative humidity varies with height and the dispersing particles adjust to the humidity by evaporation and condensation. In NOVAM, the modal[3] aerosol concentration profile is determined for the size distribution at 80% relative humidity. For simplicity the humidity growth effects are only taken into account to adjust sizes and refractive index to derive the extinction coefficients, but not to alter the modal profile concentrations.

2.3 Extinction in marine stratus clouds.

The stratus case in NOVAM is distinctly different from the physical profile models described above. It was developed from detailed measurements in marine stratus cloud layers when the surface wind was low.[10] The stratus case bypasses estimates of aerosol entrainment, generation or deposition rates and is based only on the physics of aerosol growth with changes in relative humidity. The extinction properties are determined using Fitzgerald's[11] approximation formulas.

The stratus model applies to the case when low level mixing is present and an inversion exists below 3 km, the cloud cover is greater than 0.8 and the wind speed does not exceed 5 m/s. The major limitation of this marine stratus model is the need to restrict the wavelength to the infrared range of 1-11 μm, compared to the wavelength of 0.2-40 μm for the other categories.

3. THE NAVAL OCEANIC VERTICAL AEROSOL MODEL (NOVAM)

3.1 Intended use of NOVAM

NOVAM was formulated to be used with an equilibrium surface layer aerosol model such as NAM,[1] to estimate the effect of the vertical variation of the aerosol concentration on slant path extinction. As such NOVAM is an extension of NAM. The NAM version found in LOWTRAN6 has been updated since new scientific data has become available after its introduction in 1983. These include the following developments:

1) A much more accurate parameterization of the wind dependence of large size aerosol, based on a new set of measurements.[12]
2) The development of an improved multispecie aerosol growth formulation.[13]
3) The inclusion of different chemical composition of the individual populations of marine aerosols. This affects both the optical properties of the aerosol and their growth properties.[3]
4) An improved parameterization technique which will eliminate the necessity of knowing the air mass parameter.[14]

3.2 NOVAM's input and output.

NOVAM has a comprehensive default system coupled with a method of estimating the "goodness" of the prediction. The philosophy behind this idea is that the model ought to be usable by everyone, even if the required input data is incomplete. However, the statistical reliability of the output should decrease as the quality of the input decreases, since that requires best estimates from other models with their inherent accuracy. This is reflected in a quality factor.

Inputs from surface and radiosonde observations are requested by NOVAM. The set of surface observations are listed in Table 1. The radiosonde observations include the general profile for temperature and humidity.

Table 1. Surface observation data file

position	meteorological data
1	sea surface temperature (C)
2	air temperature (C)
3	mixing ratio (g/kg)
4	optical visibility (km)
5	local wind speed (m/s)
6	averaged wind speed (24 hr) (m/s)
7	air mass parameter [1..10]
8	cloud cover (tenths)
9	cloud type [1..10][3]
10	surface infrared extinction at 10.6 μm (1/km)
11	present weather in standard code [0..99]
12	height of lowest clouds (m)
13	zonal/seasonal category [1..6]

The product of NOVAM is primarily a file of the extinction and absorption coefficients at various levels of the marine atmosphere. In addition, an optional log file is produced for the user which allows an insight into what "decision" steps were taken by the model.

3.3 Model Architecture

The model is based on the physical processes affecting the production, mixing, deposition and size of the aerosol within the marine atmosphere. Individual groups of aerosol with similar origin are represented by separate lognormal size distributions. All the processes which we assume to be acting on a certain group are considered to have similar effects on all particles in that group. The net optical effect produced by the aerosol is the result of the superposition of all the groups.

3.3.1. Selecting the profile. To determine the aerosol size distribution at any particular level, one of a set of mixing profile models is used. The selection process for the profile model depends on the input data available, the meteorological conditions, and the wavelength at which calculations are to be done, see Figure 2.

Several of the modular processes in Figure 2 have yet to be formulated. The possibility for their future existence is planned for however in the selection process. These cases are now routed to the default mode of calculation. The modular processes which are now supported include a weak convection model, a simple mixed layer model, a sub-stratus model and a default model. The modular processes which are not now supported are the stable boundary layer model, a deep convection model, and a high wind stratus model.

3.3.2 Extinction calculations with selected profile model. In all but the sub-stratus model, the physical processes acting on the aerosol are accounted for at each level and the net aerosol size distribution at a nominal 80% relative humidity is determined. The relative humidity at each level is determined either directly from the radiosonde data or from a default relative humidity profile generator.[15] At this point, pre-calculated Mie integrals of extinction and absorption for the wavelengths and relative humidities are associated with the derived aerosol size distribution at each level. Thus the Mie theory of light scattering and absorption from a population of aerosol is used to determine the optical properties of the atmosphere once the aerosol size distribution at a particular level is determined. For the case of the sub-stratus model a simplified Mie calculation for each height in question is undertaken in a more specialized way.

3.4 Examples of the results

Figure 3 shows the dependence of the extinction profiles on the model selection. All profiles were calculated with the same surface input conditions. Figure 3a shows the well-mixed case, Figure 3b shows the weak convection case and Figure 3c shows the result for a default (no radiosonde) calculation.

4. CRITICAL EVALUATION

4.1 Approach

The NOVAM approach as presented above, is a mixture of models developed at the author's Institutes.[1,7,10,16] Considerable effort has been put into the model, but we realize that it has certain limitations. The individual codes were developed for special situations and extensions to other locations and other meteorological conditions are now major goals.

For evaluation of the model, data on mixed-layer profiles of aerosol concentrations and optical properties are available from various experiments. These include aircraft aerosol measurements over the North Atlantic and the East Pacific and lidar profiles of backscatter and extinction coefficients measured over the North Atlantic and the North Sea. The geographic spreading of the locations for these experiments and the variations in meteorological conditions guarantee a severe test on the usage of the model. The comparison of the calculated and observed profiles is expected to show both the strength and the weakness of the model. Improvements will be made accordingly. Some of the problems we are currently working on are discussed in the following sections.

4.2 Radiosonde soundings.

A major requirement for application of the complete model is good-quality radiosonde soundings to determine the height of the inversion capped mixed-layer, the temperature and humidity gradients from the surface to above the boundary layer (see Figure 1), and the cloud base. The interpretation of the soundings to obtain the input parameters is not always straightforward. Errors in the interpretation may result in NOVAM selecting a non-representative extinction profile model. To assist the user with the analysis of the radiosonde data, an automatic computer code is under development.

In cases when radiosonde data are not readily available, default humidity and temperature profiles are generated from the surface observation data.[15] Because these results cannot always be as good as an actual measurement the reliability of the calculated extinction profile decreases. This is expressed in the quality factor. In particular some profile parameters in Eq. 1, e.g. the entrainment rate (w_e), cannot be evaluated reliably from the default model.

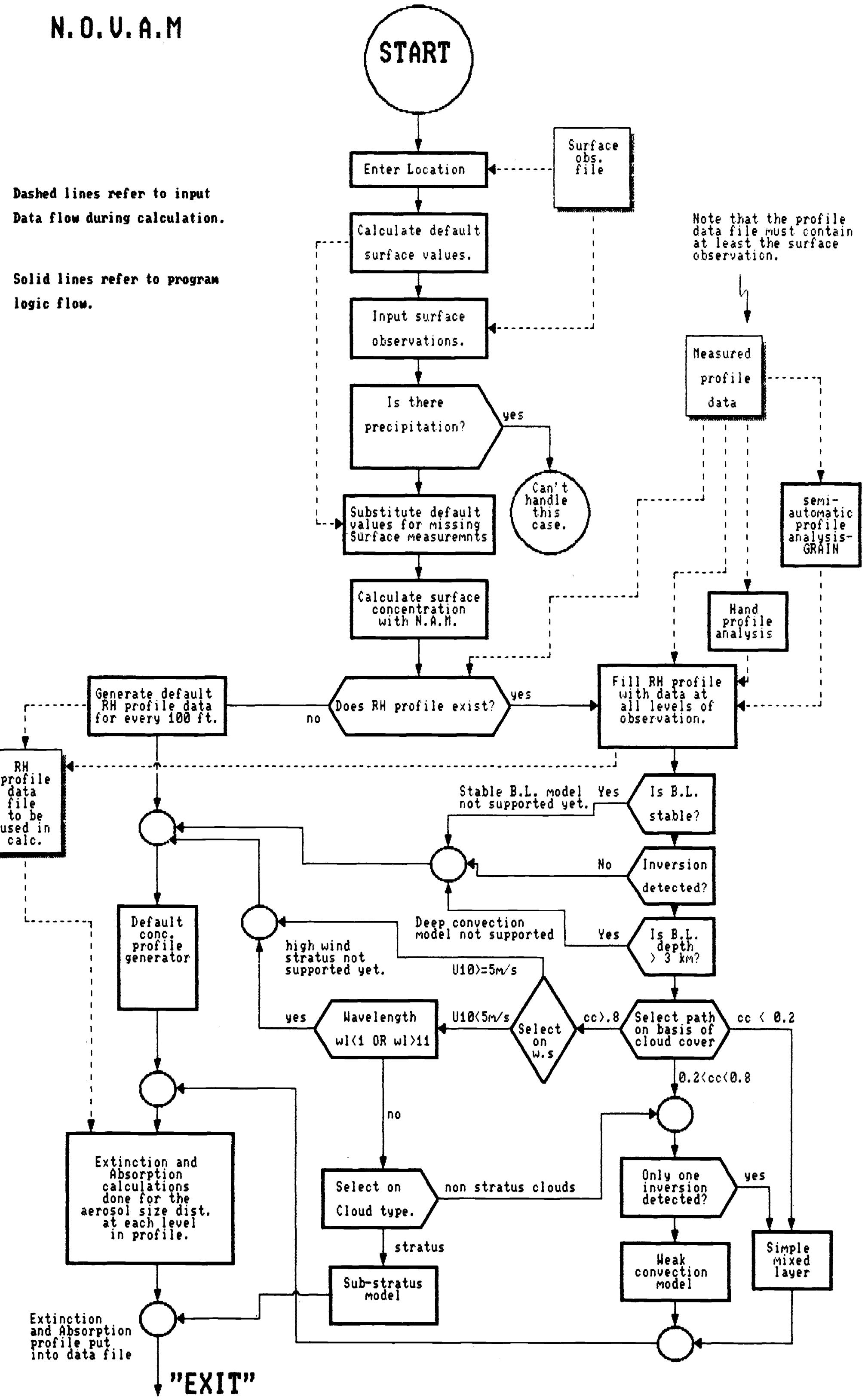

Figure 2. Flow chart for NOVAM 2.0

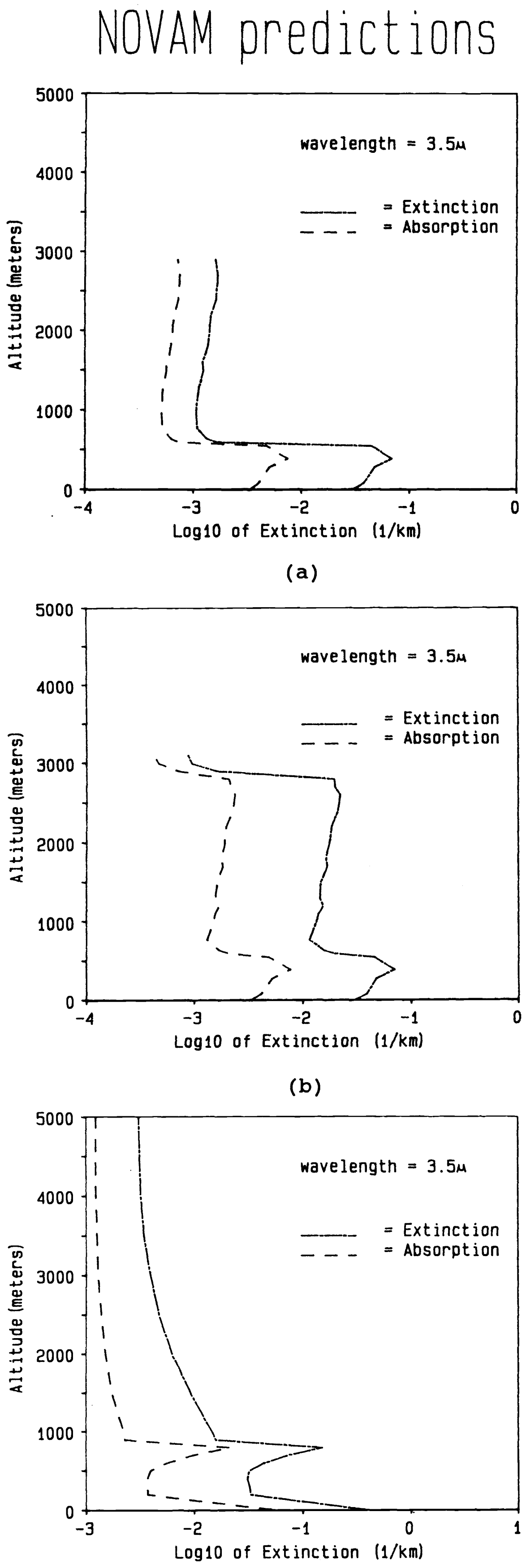

Figure 3. Examples of extinction and absorption profiles calculated with NOVAM for a wavelength of 3.5 μm, based on the same surface wind, temperature and humidity: (a) well-mixed case; (b) weak convection case; (c) default calculation.

4.3 Relative humidity effects.

In a well-mixed layer the profiles of scalar quantities can be described on the basis of surface fluxes and entrainment parameters. However, aerosol mass is not a conserved scalar quantity since the freshly produced surface droplets change size until they are in a dynamic equilibrium with ambient humidity. This process will predominantly take place in the surface layer.

The approach used by NOVAM to mix the particles throughout the boundary layer for a given size at 80% relative humidity (section 2.3) is too simplified. This is because the concentration gradients (Eq. 1) also change as the particle size varies with relative humidity. At least two effects should be considered.[16] The first effect is that the effective fall velocity V_d in Eq. 1 is affected through both the change in the Stokes fall velocity and the change in the deposition velocity. The Stokes fall velocity (V_S), e.g., increases by a factor 3-4 when humidity increases from 80% to 98%. For a particle with diameter D and density ρ, V_S is given by:

$$V_S = \frac{\rho \, D^2 \, g}{18 \, \eta}, \qquad (2)$$

where g is the gravitational acceleration and η is the dynamic viscosity. Eq. 2 shows that V_d varies with D^2, and with the particle density ρ. The particle density ρ changes with relative humidity, S, according to:

$$\rho = \rho_w + (\rho_d - \rho_w) \; g(S)^{-3} \qquad (3)$$

where ρ_w and ρ_d are the densities of pure water and of dry particles, respectively, and g(S) is the humidity correction factor that relates a particle with size D_{80}, at 80% relative humidity, to its size D at the actual ambient relative humidity:

$$D = D_{80} \; g(S) \qquad (4)$$

The second effect is the shift in the particle size distribution due to humidity effects. The shift in particle size is equal for all particles of the same NOVAM mode, while different growth factors apply to different modes. However, since the mixing varies with particle size, the shape of the size distribution should change in the vertical as well. These two effects are presently being formulated for NOVAM.

4.4 Aerosol size distribution model.

The aerosol size distribution used in NOVAM is similar to the one used in NAM.[1] In the last decade an appreciable number of other data on the marine aerosol has become available and has been incorporated into a new formulation of NAM, as described in section 3.1.

The largest particle mode has a mean radius of 2 μm. Consequently the present NOVAM aerosol extinction in both the 3-5 μm and the 8-12 μm transmission windows are predominantly determined by this mode. The transport properties of the 2 μm particles are quite different from those of the 10 μm particles which in fact determine primarily the IR extinction properties in the 8-12 μm transmission window. Therefore it might be desirable to add another mode that takes this into account. This will necessarily influence the vertical infrared extinction profile.

Surface layer aerosol size distribution profiles for particles larger than 5 μm were measured in a wide range of wind and stability conditions during the HEXOS experiments.[17] A parameterization of these particle size distributions will be attempted to take the influence of larger particles properly into account.

Additional improvements of the aerosol particle size distributions might be obtained from the inclusion of parameters other than mean and local wind speed, relative humidity and the air mass parameter. Monahan[18] has shown that whitecap coverage, which determines production, depends on atmospheric surface layer stability, water temperature and fetch, as well as wind speed. Further the wave properties should be considered. Wave breaking in a developing wave field is significantly different from wave breaking in an 'aged' wave field. In coastal regions the water depth and the fetch will influence the wave field.

The above considerations are important in the assessment of the present status of NOVAM. The inclusion of parameters such as fetch, stability, sea water temperature and 'wind duration' requires a new analysis of the available data. This is a major effort. On the other hand it might lead to a better parameterization of local influences and improve the applicability of NAM.

5. CONCLUDING COMMENTS

NOVAM is designed to provide realistic height variations of marine atmospheric boundary layer aerosol on the basis of dynamical and thermodynamical models for the region. There is no question, viewed from presented criticisms, that a formulation status still exists for NOVAM and that the model architecture has missing components. Data on vertical aerosol profile with complete meteorological information is needed for all future changes. In spite of this current formulation status, we believe NOVAM already has merit for providing vertical extinction profiles for many geographical and meteorological regimes.

NOVAM is a candidate for forecast purposes because rate equations describe the physical processes which determine the equilibrium boundary layer. A forecast is important because mean boundary layer processes and properties ,which are included in NOVAM, are continually evolving on time scales of hours.

6. ACKNOWLEDGEMENTS

We wish to acknowledge other people who provided significant contributions to NOVAM. These include Dr. Juergen Richter who manages the applied research program in which it is developed, which included the observations and the physical modeling, and Drs. Chris Fairall and Hermann Gerber who were involved in numerous discussions on the direction of the formulation. GdL participated in this program while he held a National Research Council Research Associateship at the Naval Postgraduate School.

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