### PHYSICAL MODELS FOR AEROSOL IN THE MARINE MIXED-LAYER

Gerrit de Leeuw, \* and Kenneth L. Davidson
Naval Postgraduate School, Department of Meteorology
Monterey, California 93943, U.S.A.

\*permanent address: Physics and Electronics Laboratory TNO P.O. Box 96864, 2509 JG The Hague, The Netherlands

Stuart G. Gathman
Naval Research Laboratory, Code 4117
Washington D.C., 20375, U.S.A.

R.V. Noonkester+

Naval Ocean Systems Center San Diego, California 92152-5000, U.S.A.

+present address: Innovative Meteorology, 4247 Mnt. Henry Ave, San Diego, CA 92117, USA

### SUMMARY

A model is presented to calculate the vertical variation of aerosol extinction coefficients throughout the marine atmospheric boundary layer. It is referred to as the Naval Oceanic Vertical Aerosol Model (NOVAM). NOVAM is a combination 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

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

When vertical structure is required for slant path calculations, the surface aerosol concentrations need to be extended to higher levels. This may be based on empirical models or on physical arguments. A usual approach in existing empirical models is to assume a logarithmic decrease with height using effective scaling heights.

Observed profiles are often nonlogarithmic, however. Therefore extra information available from observed meteorological profiles should be used to take into account the physical processes which influence the vertical aerosol structure and which are thought to be responsible for the observed variety of profiles. Existing empirical models do not allow for the use of this extra information.

In this paper we describe an approach being formulated to put vertical structure into the extinction prediction using a mixture of empirical and physical models which describe the aerosol dynamical behavior. 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).

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

The following aspects of the multicomponent model are addressed. The physical background of the turbulent controlled processes and of the growth features caused by relative humidity effects are presented in section 2, as well as the model used to predict extinctions found under solid cloud decks. The model architecture and considerations of the physical constraints as they are treated in NOVAM are presented in section 3. Examples of calculated profiles are presented and compared with observed profiles. 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. PHYSICAL BACKGROUND

# 2.1 TURBULENT TRANSPORT IN THE MARINE ATMOSPHERIC BOUNDARY LAYER

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.

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.

As they are presently used in NOVAM, 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 in the boundary layer are by transfer through the sea surface or by entrainment and gravitational fallout from the nonturbulent troposphere immediately above the marine boundary layer.

The vertical mixing of aerosols throughout the boundary 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 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 règimes 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

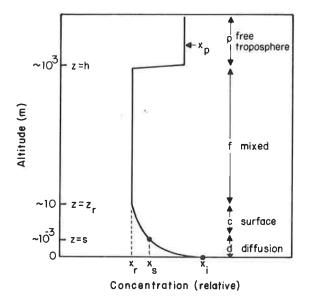


Figure 1. Schematic diagram of atmospheric scaling regimes (nonlinear scales).

thorough consideration of air-sea particulate transfer processes by Fairall and Larsen 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 model 5 is considered to be applicable to the midlatitude 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 mixedlayer 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  $\mu m$  radius are expected to obey mixed layer scaling 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, using physical arguments of Wyngaard and Brost, show that a mixed layer gradient for a surface generated aerosol component (e.g. sea salt) would be given by

$$\frac{\partial X_{sr}}{\partial z} = \frac{-1.5}{hw_{\star}} (s_r - v_d x_{sr} + 2.5 w_e x_{sr})$$

where  $X_{ST}$  is the concentration of the sea-salt aerosol in the mixed-layer at height z,  $S_T$  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  $w_{\star}$  is the convective scaling velocity.

Gradients predicted by Eq. (1) would be dependent on particle size. With representative values for the scaling parameters, the height variations for the very small particles can usually be neglected under typical conditions. This is generally not true for larger particles. It is also important to note that the role of relative humidity, which affects the gradient through both  $V_{\rm d}$  and  $X_{\rm gr}$ , 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 tradewind 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). A trade-wind equivalent to the midlatitude mixed-layer model was developed by Albrecht. Davidson and Fairall describe the application to aerosols.

### 2.2 AEROSOL HUMIDITY EFFECTS

The marine aerosol consists in large part of hygroscopic particles, the size of which varies by evaporation and condensation, in response to changes in the relative humidity. In the mixed-layer the relative humidity varies with height and the sizes of the dispersing particles change accordingly. In NOVAM, the modal<sup>3</sup> 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 model used in NOVAM to calculate extinction in marine stratus clouds is distinctly different from the physical profile models described above. 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.

It was developed from detailed

measurements in marine stratus cloud layers when the surface wind was low.  $^{10}$  This limits application of the stratus model to cases 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 extinction properties are determined using Fitzgerald's  $^{11}$  approximation formulas which apply at wavelengths in the infrared between 1 and 11  $\mu\mathrm{m}$ , as compared to the wavelength of 0.2-40  $\mu\mathrm{m}$  for the other categories. This is a major limitation of the marine stratus model.

# 3. THE NAVAL OCEANIC VERTICAL AEROSOL MODEL (NOVAM)

### 3.1 INTENDED USE OF NOVAM

NOVAM was formulated to estimate the effect of the vertical variation of the aerosol concentration on slant path extinction. It is intended to be used with an equilibrium surface layer aerosol model such as NAM. 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:

- A much more accurate parameterization of the wind dependence of large size aerosol, based on a new set of measurements.
- The development of an improved multispecie aerosol growth formulation.<sup>13</sup>
- 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.<sup>3</sup>
- An improved parameterization technique which will eliminate the necessity of knowing the air mass parameter.

### 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.

The inputs requested by NOVAM include the set of surface observations listed in Table 1 and the general profile for temperature and humidity as observed with radiosondes.

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

Table 1. Surface observation data file

position	meteorological data
1	sea surface temperature (C)
2	air temperature (C)
3	relative humidity (%)
4	optical visibility (km)
5	local wind speed (m/s)
6	averaged wind speed (24 hr) (m/s)
7	air mass parameter [110]
8	cloud cover (tenths)
9	cloud type [110]3
10	surface infrared extinction . at 10.6 $\mu$ m (1/km)
11	present weather in standard code [099]
12	height of lowest clouds (m)
13	zonal/seasonal category [16]

"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 is evident from the flow chart in Figure 2. The selected model depends on the input data available, the meteorological conditions, and the wavelength at which calculations are to be made.

Several of the modular processes in Figure 2 have yet to be formulated. These include the stable boundary layer model, a deep convection model, and a high wind stratus model. The possibility for their future existence has been 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.

# 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 to determine the net aerosol size distribution at a nominal 80% relative humidity. The actual 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 the Mie theory of light scattering and absorption from a population of aerosol is used to calculate the optical properties of the atmosphere with precalculated Mie integrals of extinction and absorption for the requested wavelengths and the appropriate relative humidity. This is achieved by associating these with the derived aerosol size distribution at each level. 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 AND COMPARISON WITH EXPERIMENTAL DATA

Figure 3 shows examples of how NOVAM estimates a profile of extinction at wavelengths in both the visible and IR bands. The meteorological profile data used as input for NOVAM was obtained form a tethered balloon platform on which also a nephelometer as well as a PMS (Knollenberg) particle spectrometer were located. Extinction at visible wavelengths is obtained directly from the nephelometer, whereas extinction at different wavelengths may be calculated, using Mie theory, from the aerosol size distributions obtained from the PMS system. The tethered balloon measurements presented in Figure 3 were taken on the upwind side of San Nicolas Island, California, on July 18, 1987.

Figure 3a is the extinction profile for wavelengths in the visible and contains extinction data measured directly with the nephelometer, extinction data calculated from the measured aerosol size distribution, and the extinction data estimated by NOVAM using the measured profile of air temperature and relative humidity. The data shows that there is a considerable amount of scatter in the measured extinction at the various altitudes. The NOVAM prediction at the visible wavelength is within the envelope of the scatter better than 75% of the time for this particular case.

Figure 3b shows the comparison between the calculations of the average extinction in the band between 3 and 5 μm using the Mie code on measured aerosol size distributions and the NOVAM prediction for 3-5  $\mu$ m. Because there was no direct measure of extinction in these IR bands, only the Mie calculations of the measured size distribution are shown. A similar comparison for the 8-12  $\mu m$  band and the NOVAM prediction at 10.6  $\mu m$  is shown in figure 3c. The present version of NOVAM is underestimating the apparent extinction in the IR bands in the region above the inversion. This is a result of larger particles from the sea surface being mixed into the atmosphere above the apparent inversion by the process of entrainment.

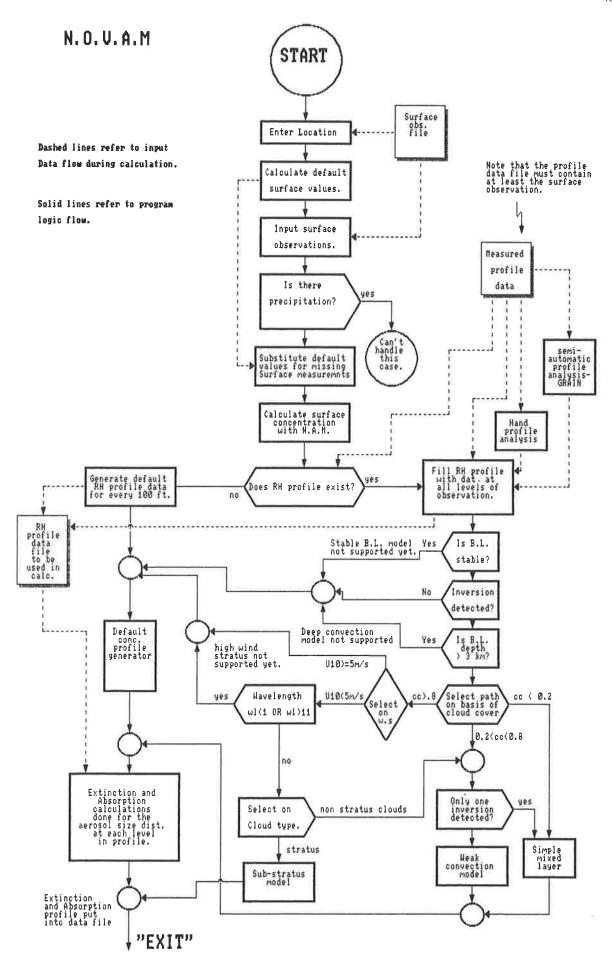
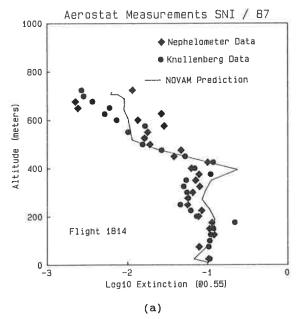
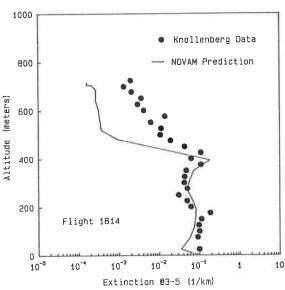


Figure 2. Flow chart for NOVAM 2.0.





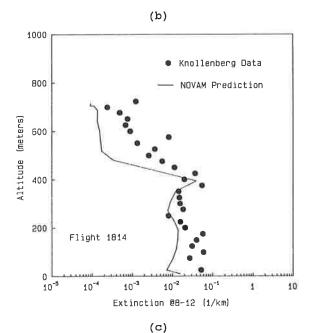


Figure 3. Comparison between extinction profiles predicted by NOVAM (solid lines) and experimental extinction profiles derived from nephelometer data (filled diamonds) or from particle size distributions using Mie theory (filled circles). The measurements were made from a tethered balloon.

(a) Comparison of the NOVAM prediction for a wavelength of 0.55  $\mu m$  with a Miecalculated profile at 0.55  $\mu m$  and the extinction profile derived from the nephelometer.

(b) Comparison of the NOVAM prediction for 3.5  $\mu$ m with the average Mie extinction in the 3-5  $\mu$ m band.

extinction in the 3-5  $\mu m$  band. (c) Comparison of the extinction profile as predicted by NOVAM for 10.6  $\mu m$  with the Mie calculated average extinction profile in the 8-12  $\mu m$  band.

### 4. EVALUATION AND FUTURE DEVELOPMENTS

### 4.1 APPROACH

The NOVAM approach as presented above, is a mixture of models developed at the author's Institutes. 1,7,10,16

The individual codes were developed for specific situations, which sets limitations to the applicability of the model. Extensions to other locations and other meteorological conditions are now major goals.

For evaluation, experimental data on the vertical structure of aerosol concentrations and optical properties are available from various experiments. Among these are 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 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 that are currently being worked on are discussed in the following sections.

### 4.2 RADIOSONDE SOUNDINGS

The availability of meteorological profiles is a major requirement for application of the complete model. Temperature and humidity profiles are needed 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. This information can be obtained from good-quality radiosonde soundings. 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. 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  $(W_{\rm e})$ , cannot be evaluated reliably from the default model.

### 4.3 RELATIVE HUMIDITY EFFECTS ON AEROSOL MIXING

In a well-mixed layer the profiles of scalar quantities can be described on the basis of surface fluxes and entrainment parameters. This does not apply to aerosol mass, because it is not a conserved scalar quantity since the size of the particles changes in response to changes in relative humidity. The freshly produced surface droplets evaporate until they are in a dynamic equilibrium with ambient humidity. This process will predominantly take place in the surface layer. In the mixed-layer the size of the aerosol particles changes because the relative humidity varies with height.

In NOVAM the particles are mixed throughout the boundary layer for a given size at 80% relative humidity (section 2.3). This is too simplified because the concentration gradients, cf. Eq. (1), also change as the particle size varies with relative humidity. At least two effects should be considered. The first effect is that the effective fall velocity  $V_{\rm d}$  in Eq. (1) is affected through both the change in the Stokes fall velocity and the change in the turbulent deposition velocity. The Stokes fall velocity  $(\mbox{V}_{\mbox{S}})$ , e.g., increases by a factor 3-4 when humidity increases from 80% to 98%. For a particle with diameter D and density  $\rho$ ,  $V_S$  is given by:  $V_S = \frac{\rho}{18} \frac{D^2}{\eta}$  (2)

$$V_{S} = \frac{\rho D^{2} g}{18 \eta}$$
 (2)

where g is the gravitational acceleration and  $\eta$  is the dynamic viscosity. Eq. (2) shows that  $V_d$  varies with  $D^2$ , and with the particle density  $\rho$ . The particle density  $\rho$  changes with relative humidity, S, according to:

$$\rho = \rho_{W} + (\rho_{d} - \rho_{W}) g(S)^{-3}$$
 (3)

where  $\rho_{\rm W}$  and  $\rho_{\rm d}$  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  $^{17}$ :

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

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 a combination of lognormal distributions describing the individual components, similar to the one used in NAM.  $^{\rm I}$  In the last decade an appreciable number of other data on the marine aerosol has become available. These were used for the new formulation of NAM that is now used in NOVAM, as described in section 3.1.

The largest particle mode in NOVAM has a mean radius of 2  $\mu\text{m}$  . The aerosol extinction in both the 3-5  $\mu\text{m}$  and the 8-12  $\mu$ m transmission windows are predominantly determined by this 2  $\mu m$  mode. The transport properties of the 2  $\mu$ m particles are quite different from those of the 10  $\mu m$  particles which in fact determine primarily the IR extinction properties in the 8-12  $\mu m$ transmission window. This is presently not taken into account in NOVAM and the profiles for wavelengths in both IR windows have similar shapes. To describe the extinction profiles in the 8-12  $\mu m$  window, it might be desirable to add another mode with a mean radius of about 10  $\mu$ m.

Data on these large particles are available from surface layer measurements of aerosol size distribution profiles for particles larger than 5 µm during the HEXOS experiments, 18 covering in a wide range of wind and stability conditions. A parameterization of these particle size distributions will be attempted to take the influence of larger particles properly into account in NOVAM.

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 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. For all future changes data on vertical aerosol profile with complete meteorological information is needed. 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.

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