Near-surface aerosol transmission in the marine environment

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ABSTRACT

An extensive data set of aerosol vertical concentration profiles (height range 0.5-15 meters, diameter range 10-75 micron), acquired with the Rotorod[3] device at various geographical locations has been statistically analyzed. The analysis supplied a parameterization for the 4^{th} (large particle) lognormal mode in the Advanced Navy Aerosol Model (ANAM). The analysis revealed a positive correlation between concentration and wind speed, and a negative correlation between concentration and height. No clear dependence of radius and width on meteorological parameters has been found. Numerical simulations with the Dutch-French SeaCluse model supported the results of the statistical analysis. The present version of ANAM predicts the experimental aerosol concentration to within a factor of 3. This result was confirmed using an independent data set. Initial extinction calculations with the ANAM indicate that the addition of the 4^{th} mode changes aerosol extinction values by about 20% as compared to NAM calculations. Closer to the surface, this number is even higher due to the height dependence of the 4^{th} mode.

Keywords: NAM, ANAM, SeaCluse, marine aerosol, model

1. INTRODUCTION

Electrooptic and infrared (EO/IR) systems represent a critical Navy technology for the detection and tracking of high-precision and low-signature anti-ship sea-skimming missiles. It is imperative to know the effects of the atmosphere on EO/IR systems to ensure that these systems will perform as expected. The prediction of EO/IR system performance is particularly critical within the first several meters of the atmospheric surface layer. Within this thin surface layer there can be strong humidity, temperature and aerosol concentration gradients. All of these factors have a substantial impact on the detection of sea-skimming threats.

In recent years, it has become clear that the presence of waves and the production of aerosols at the sea surface have a distinct influence on near-surface propagation characteristics. The relative abundance of large aerosols may reduce the infrared transmission near the surface by a factor of 2 or 3 as compared to ship deck heights.[1] Consequently, the maximum detection and identification ranges of low-altitude targets may be significantly smaller than anticipated when near-surface effects are neglected.

In the marine environment, aerosol production from breaking waves and/or direct wind tearing from the crests constitutes an important source of aerosol. The actual concentration of marine aerosol in the atmosphere results from a delicate balance between production at the surface, vertical turbulent transport and deposition. Over the years, considerable effort has been spent to assess the marine aerosol concentration as function of the meteorological conditions. An important milestone was reached with the release of the Navy Aerosol Model,[2] which predicted the marine aerosol concentration at ship's deck height for open ocean conditions. The original NAM has been updated on the basis of new experimental[3,4] and theoretical[5,6] evidence, and is presently included in the widely used USAF MODTRAN transmission code.[7]

The NAM describes the aerosol size distribution by a superposition of three lognormal curves ("modes"). Each mode is characterised by a width (assumed constant), a centre radius and an amplitude. The centre radii of the modes are nominally 0.03, 0.24 and 2.0 μm, but are adjusted as function of the relative humidity. The largest or third mode (2 μm)

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consists of freshly produced marine aerosols. Its amplitude is determined by the instantaneous wind speed. The second mode (0.24 μm) consists of marine aerosols that have spent some time in the atmosphere ("aged" marine mode) and have adjusted their size to the ambient conditions. Since these particles have been produced elsewhere and transported to their present location, the amplitude of the second mode is determined by the wind speed history. Finally, the first mode (0.03 μm) consists of fine particles presumed continental in origin. Its amplitude is determined by the so-called air mass parameter, which in turn is related to the visibility at 0.55 μm. Depending on the value of the air mass parameter, the first mode is separated in a hygroscopic (centre radius adjusted according to humidity) and a non-hygroscopic (fixed centre radius of 0.03 μm) part. This last component is referred to as the 0th mode.

Over the years, it has become clear that the NAM code is less successful in coastal regions[8] and/or at other heights than deck height (10 m).[9] The latter shortcoming is partially addressed by the NOVAM model,[10] presently included in MODTRAN, which extends the NAM aerosol concentration to heights above 10 m. However, in view of the ever-increasing threat of sea-skimming missiles, an urgent need exists for a near-surface version of the NAM. Therefore, the Advanced Navy Aerosol Model (ANAM) is being developed to extend the NAM aerosol concentration from 10 meters down to the wave surface.

The initial development of the ANAM focuses on the behaviour of relatively large (radius in excess of 5 μm) aerosols. The rationale for this is twofold: first, it has been suggested[9] that the NAM underestimates the concentration of these larger aerosols resulting in an estimate of the (IR) extinction that is too low.[11] Secondly, turbulence is less efficient in transporting these heavy aerosols upward from the sea surface, which will (theoretically) result in steeper vertical concentration gradients.[12] The aim of the ANAM development is to extend the NAM by a height-dependent 4th mode, centred at a radius larger than 2 μm (nominal radius of 3rd mode in NAM). In a later phase of the development, the necessity of introducing height dependence in the other (NAM) modes will be assessed.

Like its predecessor NAM, the ANAM is being developed by establishing empirical relations between the aerosol concentration and meteorological parameters.[1] Unfortunately, measurements of vertical aerosol concentrations between the wave surface and deck height are sparse and almost exclusively limited to the Rotorod technique deployed by TNO-FEL.[3] Even when data is available, the uncertainty in the data is often considerable due to the difficulties encountered in making near-surface measurements. To further complicate the problem, the relatively simple relationships that exist at deck level between, e.g., wind speed and aerosol concentration, become more complex closer to the surface due to the influence of the waves.

In view of these complications, the further development of the ANAM requires a full analysis of all available near-surface aerosol data. However, even with a large data set questions remain as to whether a purely empirical ANAM may be accurate enough to reproduce all variations in the near-surface aerosol transmission. Therefore, numerical or physical models that capture the complex interplay of physical processes at the air-sea interface must support the ANAM development. Such models may be used to reveal trends that are currently hidden in the data (due to the limited amount of data or relatively large uncertainties), or to extend the ANAM to meteorological conditions not covered by the database.

Numerical modelling of the near-surface aerosol concentration requires knowledge on a multitude of processes ranging from a description of the wave surface, the effect of this surface on the airflow, aerosol production, turbulent mixing, evaporation and deposition. While state-of-the-art models are available for particular aspects of the problem (e.g., the distorted airflow over waves,[13] few models provide a complete description of the (near-surface) aerosol concentration. Recent advances include Gwaihir,[14] the NRL model,[15] the Kepert-Fairall model[16] and the Dutch-French SeaCluse model.[17,18]

The SeaCluse model is developed to study the non-linear interactions between the marine aerosol and the scalar fields of temperature and water vapour in the marine surface layer. Based on the "Couche Limite Unidimensionelle Stationaire des Embruns" (CLUSE) model,[19] the SeaCluse code simulates many aspects of the dynamics of sea spray droplets in the turbulent airflow over a wavy surface,[17] the thermodynamic transformations of the spray droplets,[20] and the influence of the droplets on heat and water vapour fluxes in the lower marine atmosphere.[18] The principal originalities of the model consist of explicitly taking into account the presence of the waves and of the simultaneous resolution of both dynamical and thermodynamic processes. The output of the model consists of vertical profiles of aerosol concentration, humidity and temperature, as well as water vapor and heat fluxes.

This contribution presents an overview of both the statistical analysis of Rotorod data and the numerical simulations with the SeaCluse model. The statistical analysis includes data from the recent RED (Rough Evaporation Duct, 2001) experiment near Oahu, Hawaii. These data represents one of the few sets acquired in open ocean conditions, whereas most Rotorod data have been acquired in coastal environment. Therefore, the RED data set is important to eliminate bias in the analysis caused by specific coastal effects.

A numerical model such as SeaCluse can be used in support of the statistical ANAM development. To this end, the model is run for a variety of meteorological conditions to yield relations between meteorological parameters and
It is anticipated that the ANAM development will be supported numerically by the Dutch-French SeaCluse model. This model calculates the distribution of aerosol in the atmospheric surface layer with explicit algorithms for the influence of the waves on the airflow and turbulence. In addition, the SeaCluse model yields the vertical profiles of humidity and temperature, which can be used to infer optical turbulence and refraction effects close to the sea surface.

2. STATISTICAL ANALYSIS

The development of the ANAM 4th mode requires concentration measurements of larger aerosols in the first few meters above the wave surface. The concentrations of these large aerosols are very small necessitating a sample device that takes in large samples of air in order to get statistically significant results. Additionally, the device should be small (for not disturbing the airflow) and preferably expendable (since it may get submerged or sprayed by water). The simple Rotorod device[3,21] satisfies these needs. The Rotorod technique is an active impactor sampling method, featuring silicon coated stainless steel rods that are mounted on a stabilized constant speed motor. The rods are spun to collect particles that leave an imprint in the silicon spray on the rods, which can be subsequently analyzed under a microscope. An important feature of the technique is that the size of the craters on the rods corresponds (closely) to the diameter of the particle at the time of impaction.[3] The thus counted particles are distributed over bins (typically 10 between 10 and 100 μm) to yield an aerosol size distribution. A more detailed description can be found elsewhere,[1] which also shows that the Rotorod technique reliable measures concentrations of particles with a radius larger than 5 μm. The upper radius limit is determined by statistics and ranges from 40 to 150 μm depending on the concentration (basically determined by wind speed for sea-salt particles).

The Rotorod technique has been deployed at various campaigns in different regions of the world,[22] but mostly at the MeetPost Noordwijk (MPN), a research tower located in the North Sea at 10 km off the Dutch coast. From these data, a relatively large composite MPN data set of 2078 observations (aerosol size distributions) can be formed, that should provide a good basis for a statistical analysis. The selection of MPN data implies that a bias could be introduced towards coastal conditions, even if only data acquired in long-fetch conditions are included in the analysis. Therefore, it is important to complete the analysis with open-ocean data, available from two experiments: the Cumulus trial in 1983 (117 observations),[3] and the more recent RED experiment (297 observations). The open-ocean data was not mixed with the composite MPN-set, but treated separately.

The aim of the present analysis being the extension of the three lognormal distributions of NAM with a 4th mode, a lognormal curve was fitted to each of the aerosol size distributions in the (composite) MPN and open ocean data sets:

$$\frac{dN}{dR} = A_4 \exp\left\{ - C_4 \left[\ln\left(\frac{R}{R_4}\right)\right]^2 \right\} \tag{1}$$

where the adjustable parameters A_4, R_4 and C_4 correspond to the amplitude, center radius and width of the lognormal curve. The success of each fit can be inferred from standard statistical parameters, such as the standard deviation and the correlation coefficient. With a few exceptions, the correlation coefficients of the fits were above 0.8, with 80% above 0.95. The lognormal parameters A_4, R_4 and C_4 were subsequently gathered in a spreadsheet, together with meteorological information (e.g., wind speed, wave height) at the time of observation. A micrometeorological model[23] was used to generate additional information (e.g., friction velocity) to be included in the spreadsheet. Linear and logarithmic correlation was then applied to the lognormal parameters and the (mirco)meteorological parameters.

The relations between lognormal and (micro)meteorological parameters are weak with correlation coefficients rarely exceeding 0.5. Several techniques were applied to maximize the correlation between two given parameters, basically consisting of minimizing the impact of other parameters. Thus, when correlating mode amplitude with wind speed, height dependence was minimized by only including data measured between 8 and 10 meters above mean sea level (AMSL). The dependence on humidity was minimized by conversion of all concentrations to a standard humidity of 80%.[24] Despite these efforts, the correlation between mode parameters and (micro)meteorological parameters remained modest. Reviewing the experimental data, the modest correlation was attributed to the relatively large standard deviation in the Rotorod measurements, combined with the limited variation of the aerosol concentration over the range of parameters such as wind speed and height.

The statistical analysis outlined above lead to the following conclusions:

(1) The lognormal amplitude or concentration increases exponentially with wind speed, wave height and friction velocity, factors that affect the production of aerosol.
(2) The lognormal amplitude or concentration decreases exponentially with height, albeit that the correlation is weak.
(3) No clear correlation exists for the lognormal center radius and/or width with any of the (micro)meteorological parameters.

The first two conclusions were also reached previously,[1,3,25] by analyzing individual data sets. The correlation is strongest for the RED data set, less for the composite MPN data set and weakest for the Cumulus data set. This last result may be explained by the relatively small size of the Cumulus set. For the MPN composite set, the overall correlation may be obscured due to differences between individual data sets. Such differences have been noted earlier[26] and raise questions as to whether a composite data set can and should be built from individual data sets acquired in various years and seasons. On the other hand, the individual MPN data sets are relatively small and the correlation efforts are often even less successful than for the composite data set.

As a next step, the NAM code[27] was extended with a 4th lognormal mode to create the basis of the ANAM model. Using the results of the statistical analysis as a starting point, the functional dependence of the 4th mode parameters A_4, R_4 and C_4 was varied by trial and error to obtain the best possible match between the aerosol concentrations as predicted by ANAM and as measured with the Rotorod impactors. The performance of a particular parameterization P_k of A_4, R_4 and C_4 was calculated from a scatter plot between the experimental and modeled aerosol concentrations (for the part of the size distribution larger than 10 μm diameter). In such a scatter plot, the standard deviation σ of the data with respect to the line y=x indicates the performance: the smaller σ, the closer experimental and modeled concentrations. Furthermore, a performance factor[28] F can be introduced ($F = 10^{\sigma}$) that is a measure how well the model predicts the experimental concentration: the prediction differs from the experimental concentration by a factor of F or less in 67% of the cases.

The MPN composite, Cumulus and RED data sets were optimized separately using a total of 87 different parameterizations. Table 1 summarizes the results, listing the best performance found with a parameterization P_k for each individual data set. Thus, for the MPN composite set parameterization P_{15} yielded the best result, i.e., a factor of 2.9 difference between experimental and modeled concentration.

The table shows that different parameterizations were required to obtain best performance for each individual data set. The difference is in the parameterization of the mode amplitude A_4 rather than in R_4 or C_4. These latter two could be taken equal for each data set, whereas the mode amplitude had to be increased from Cumulus to MPN to RED. That a larger mode amplitude was found for RED than MPN was to be expected, since more aerosol particles should be present over a fully developed oceanic wave field than in a limited fetch condition at the North Sea. However, the small mode amplitude in the Cumulus data set does not fit this picture and could be indicative for flow distortion at the measurement site, only a few meters away from the ship's hull.

A consensus parameterization was selected that yields acceptable performance factors for all data sets. In the search for this consensus, the Cumulus set was given a low priority for the reasons outlined in the previous paragraph. The current consensus parameterization P_{87} has mode amplitude A_4 that increases with wind speed and decreases with height, in accordance with the conclusions from the statistical analysis described above. Table 1 shows that P_{87} describes the

Table 1: Performance factors for various parameterizations and data sets:

	MPN composite	Cumulus	RED	FPN
Best parameterization	2.9 (P_{15})	2.6 (P_{42})	2.7 (P_{77})	---
Consensus (P_{87})	3.3	7.5	2.6	2.8
NAM	8.8	2.8	21.4	16.4

experimental aerosol concentration to within roughly a factor of three. At the time this contribution was prepared, the search for the best consensus parameterization was being finalized and therefore, the presentation of the 4th mode equations is being postponed until this work is finished.

For comparison, table 1 also shows the performance factors obtained with the NAM model. For MPN and RED, NAM performs (considerably) less than the present consensus ANAM parameterization. This is not surprising, since NAM does not take the contribution of larger aerosols into account and therefore underestimates the actual concentration of these larger particles. This is supported by the observation that NAM performs quite well for the Cumulus data set, which is characterized by a relatively low aerosol concentration, possibly due to the shielding effect of the ship's hull as mentioned above.

The performance of NAM and the consensus ANAM parameterization was also calculated for a Rotorod data set that had not been used in the statistical analysis or the trial-and-error procedure. For this test, an independent data set of 265 observations acquired at the Forschungsplatform Nordsee (FPN) in the German Bight of the North Sea was used. Table 1 shows that the performance of the consensus ANAM is comparable to that found for MPN and RED, whereas NAM again performs less.

3. NUMERICAL SIMULATIONS

The SeaCluse model has been developed to describe and quantify the dynamics of sea spray droplets and their non-linear interactions with the scalar fields of water vapour and temperature in the marine atmospheric surface layer.[17] The SeaCluse model thus simulates the dynamics of evaporating sea spray in the turbulent airflow over a wave surface.[18] The code has a 2-D pre-processor that builds an atmospheric surface layer over a wave surface, while explicitly taking into account the effect of the waves on the fields of wind, temperature and humidity. After averaging over a wave period, the 1-D Eulerian main processor numerically solves the coupled budget equations for the fields of droplet concentration, temperature and water vapor. Inertial effects are explicitly taken into account, and the thermodynamic interactions of the particles are modeled as source/sink terms in the budget equations. Essential to the code is the simultaneous resolution of both dynamic and thermodynamic processes. The output of the model consists of wave-averaged vertical profiles of temperature, humidity and aerosol concentration

Since the SeaCluse model focuses on the lower parts of the marine atmosphere where aerosols are most abundant, the model was originally built on surface layer theory. The initial vertical numerical domain was chosen to be 100 meters. It was realised that this height would often (if not always) supersede the actual height of the surface layer, and that only the results in the first tenths of meters should only be given physical significance. However, the domain was deemed necessary to introduce sufficient "space" for the aerosols to disperse freely while imposing a Dirichlet upper boundary condition of zero particles at the 100 meters level. In view of the relatively large aerosols handled by SeaCluse (lower diameter range of 10 μm) it was assumed that very few particles would disperse to the upper boundary, making the boundary condition a "natural" limit.

Unfortunately, the first simulations[17,18] showed that the upper boundary condition severally restricts the dispersion of the aerosol through the domain. It could thus not be ruled out that the boundary condition also affected the calculated aerosol concentrations and profiles close to the surface. If the upper boundary condition indeed has a marked effect on the dispersion of the aerosols in the lower part of the domain (the "true" surface layer), the extension of the vertical domain should reduce or remove this impact. Since it would be unrealistic to extend surface layer relations to even greater heights, it was decided to upgrade SeaCluse with a marine boundary layer module (which includes the surface layer). A second benefit of the introduction of a combined surface-boundary layer module is that the SeaCluse model may become more realistic for strong stable conditions. In those cases, the surface layer is quite shallow (sometimes only a few meters) and Monin-Obukhov similarity theory[29] is inadequate to describe the vertical profiles of wind, water vapour and temperature.

The extension of the vertical domain consisted of the introduction of a new meteorological pre-processor,[30] and a new turbulence module. For the latter, the $1^{1/2}$ order turbulence closure scheme of Galperin *et al.*,[31] variously described as the level $2^{1/4}$ or quasi equilibrium scheme in the Mellor-Yamada hierarchy,[32] was chosen in analogy with the Kepert-Fairall model.[16] Parameterisations were introduced for the variance of wind speed. With the extended domain, it could be demonstrated that the upper boundary condition does not affect the aerosol concentrations and vertical gradients in the first tens of meters in the domain. The initial steps in the upgrade of the SeaCluse model have been reported elsewhere[33,34] and a detailed report will be available shortly.[35]

The SeaCluse model was subsequently used to simulate the aerosol concentrations in the lower marine boundary layer. It was assumed that the wave field was fully developed (open ocean conditions) and the aerosol source function (the amount of droplets produced per unit area and time) as given by Andreas[36] was prescribed. Figure 1 shows some examples of vertical concentration profiles for particles in the 24 μm diameter size bin as calculated by SeaCluse. In this particular example, the wind speed at 10 meters $U_{10} = 20$ m/s and the relative humidity $RH_{10} = 80\%$. The atmospheric stability varied from unstable (Air-sea temperature difference ASTD = -5 °C, crosses) via neutral (plusses) to stable (ASTD = +3 °C, solid squares). The figure shows that vertical dispersion is favored under turbulent conditions (unstable atmosphere) and limited in the less-turbulent stable atmosphere. This example demonstrates how the SeaCluse model can simulate aerosol concentration profiles as function of meteorological conditions.

Figure 1 is plotted on a linear-logarithmic scale, which implies that an exponential decay of the aerosol concentration with height would result in a straight line. Clearly, the computed profiles do not decay exponentially with height above the wave crests. Exponential profiles have been suggested or postulated *ad hoc* by various authors, based on experimental evidence[37] [*Preobrazhenskii*, 1973] and early theoretical efforts [*Toba*, 1965].[38] Indeed, if only a small portion of the vertical concentration profile is considered (say, between 5 and 15 m, cf. Figure 2), an exponential fit to the profile would be indistinguishable from the true behavior.