The Advanced Navy Aerosol Model (ANAM):
Validation of small-particle modes

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ABSTRACT

The image quality of electro-optical sensors in the (lower-altitude marine) atmosphere is limited by aerosols, which cause contrast reduction due to transmission losses and impact on the thermal signature of objects by scattering solar radiation. The Advanced Navy Aerosol Model (ANAM) aims at providing a quantitative estimate of the aerosol effects on the basis of standard meteorological parameters such as wind speed and relative humidity. For application in coastal regions, the ANAM includes non-marine aerosols that are governed by an ill-defined tuning parameter: the air mass parameter (AMP). The present paper proposes a new parameterization for assessing the effect of these non-marine particles on the propagation. The new parameterization utilizes the Ångström coefficient, which can be experimentally obtained with a sun photometer, and introduces new types of aerosols in ANAM. The new parameterization was tested against experimental validation data acquired at Porquerolles Island at the French Riviera. The limited test data suggested that the new parameterization is only partially efficient in capturing the aerosol signature of the coastal environment. Nevertheless, the new Ångström coefficient algorithm avoids using the ill-defined AMP, and may thus be useful to the ANAM community.

Keywords: Navy Aerosol Model, ANAM, air mass parameter, Angstrom parameter, coastal aerosols

1. INTRODUCTION

Electro-optical sensors are presently used for a wide range of applications in the marine environment, e.g., for surveillance, ranging, and classification. The performance of state-of-the-art sensors is generally not determined by system design and technological limitations, but rather by the intervening atmosphere. Propagation of electro-optical radiation through the atmosphere can be severely limited by absorption and scattering of radiation (transmission losses). Absorption and scattering are caused by molecules and aerosols, which necessitates that the atmospheric composition and concentration of these constituents are known. The molecular part is relatively constant (apart from water vapor) and the scattering and absorption characteristics of the various molecular species are well-known. Hence, reliable codes, such as the USAF MODTRAN[1] code are available to assess the molecular component.

However, the composition and concentration of the atmospheric aerosols are much more variable, which has triggered substantial efforts to develop aerosol transmission codes. To predict aerosol transmission losses along a propagation path, the model must estimate the concentration of the various aerosol species, and must have knowledge about the refractive index of the individual species. Subsequently, Mie theory[2] provides the absorption and scattering coefficients from which extinction (and hence transmission losses along a path) can be calculated. Over the years considerable efforts have been made to express the aerosol extinction in the marine environment in terms of the meteorological conditions. In 1983 Gathman[3] published the first version of the Navy Aerosol Model (NAM) which allowed predicting the marine

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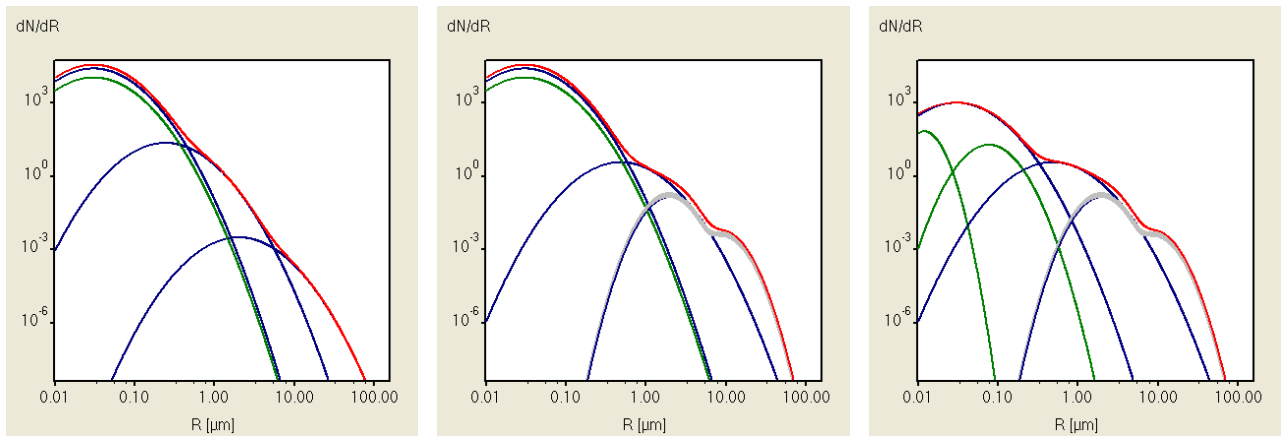[*] Lex.vaneijk@tno.nl; Phone: +31 88 866 4093; Web: www.tno.nl

Figure 1: Left panel: Original NAM model as provided through MODTRAN with (from left to right) superimposed 0th (green) and 1st modes, 2nd mode and 3rd mode. The overall distribution is given by the envelope (red). Middle panel: ANAM5 with fetch-dependent advection (2nd) mode, revised production mode (3rd/4th modes), and surf contribution (grey area superimposed on production mode). Right panel: ANAM6 with new black carbon (BC) and water-soluble (WS) modes (two left, green curves).

aerosol concentration at ship's deck height for open ocean conditions. It was considered to be a milestone. NAM is available to the community via the USAF MODTRAN code.[1]

The original NAM consists of 3 lognormal distributions (see left panel of Figure 1), which describe freshly produced marine aerosols, aged marine aerosols (produced elsewhere and advected to the measurement site) and a background concentration of marine aerosols. 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) 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 that constitute a marine background concentration. These particles have been aloft for such a long time that there presence is no longer governed by the instantaneous wind speed or even the wind speed history. Their concentration is given by the Air Mass Parameter (AMP), which can be given a value between 1 (low concentration) and 10 (high concentration). Since the AMP is not a measurable quantity, a relation between the AMP and the visibility have been established (and implemented in MODTRAN). In a trial and error process, the AMP is varied until the aerosol extinction ε (see chapter 2) at 0.55 μm corresponds to the specified visibility VIS, where VIS = 3.915 / $\varepsilon(\lambda=0.55)$.

To account for non-marine particles, a special lognormal mode representing "dust" particles was added to the NAM. The center radius of this 0th mode is at 0.03 μm (just like the 1st mode) and is not adjusted as function of humidity, because the dust particles are considered to be non-hygroscopic. This dust mode comes into play when the AMP exceeds a value of five. In that case, 30% of the aerosols of the 1st mode (marine background) are transferred to the dust mode. While this partitioning does not impact on the overall size distribution, it does impact on the aerosol extinction, because the dust particles have a different refractive index than those of marine origin.

Over the years, several improvements have been made to the original NAM. These improvements have primarily focused on the marine aerosol part of NAM, resulting in the addition of a height-dependent large particle mode (4th mode) centered around 8 μm.[4] This version was released as ANAM4. Furthermore, the production mode was reviewed,[5] as well as the advection mode, which resulted in the introduction of the fetch as a new input parameter.[6] Fetch accounts for the distance an air mass has travelled over water and allows taking into account the build-up of a marine aerosol concentration as the air mass advects out over the sea. Finally, a dedicated mode was added to describe the production of aerosols over the surf zone and their subsequent advection out to sea.[7] These improvements have been released in the ANAM5 series (see middle panel of Figure 1).

However, these improvements do not address the contribution of non-marine aerosols. Since the early 90s it is known that the performance of NAM is significantly reduced in the coastal zone.[8] The reduced performance arises from the rapid and drastic variation of the aerosol concentration and composition in the coastal zone, which is inadequately modeled by NAM. NAM only considers a 2-component aerosol mixture (marine and dust-like) and the AMP is not well enough related to meteorological observables to account for the variations in concentration. This marginal relation of the AMP to meteorological observables has bothered the NAM user community, because NAM users did not know how to set the AMP. Consequently, there have been requests to replace the AMP by a more accessible parameter. Piazzola et al. developed the Mediterranean extinction code (MEDEX),[9] in which the AMP is replaced by fetch. The rationale for this is that the contribution of aerosols generated over land decreases in the absence of sources and due to gravitational settling as the air mass advects out over the sea. However, MEDEX does not include a variety of aerosol species, and there is no scaling of the initial concentration that advects out over the land-sea interface.

More recently, Kusmierczyk-Michulec and Van Eijk demonstrated that the Ångström coefficient[10] (which describes the spectral variations of aerosol extinction) can be used as a tracer for the concentration of continental aerosols.[11] In this context, the word "continental" refers to aerosols that are generated over land, as opposed to marine aerosols. The initial ideas have been elaborated since,[12,13] resulting in a 3-component model (black carbon, water-soluble and sea-salt aerosols) for which individual concentrations are tuned as function of the Ångström coefficient. The Ångström coefficient can be directly measured and it has a physical relation to the aerosol size distribution. This makes it a good candidate to replace the Air Mass Parameter (AMP) in the ANAM for the assessment of continental aerosol contributions. An earlier paper[13] discussed an initial algorithm for the replacement of AMP by Ångström coefficient. The present contribution will elaborate on this initial approach and discuss its success on the basis of a validation trial in the Mediterranean. The model resulting from this effort is available as ANAM6.

2. METHODOLOGY

2.1 Aerosol extinction and Ångström coefficient

The total amount of radiation that is scattered and absorbed by aerosols is quantified by the wavelength-dependent extinction coefficient $\varepsilon(\lambda)$. The units of the extinction coefficient are generally [km^{-1}] and the transmission losses $T(\lambda)$ along a path of R [km] are then given by:

$$\frac{I(\lambda)}{I_0(\lambda)} = T(\lambda) = \exp\left[-\varepsilon(\lambda)R\right] \tag{1}$$

where $I_0(\lambda)$ is the radiant intensity of the light source and $I(\lambda)$ is the remaining intensity after propagating over R [km]. The extinction coefficient can be calculated from the number size distribution $dN(r)/dr$ in [μm^{-1}cm^{-3}], which specifies the number of aerosol particles N of size r present per unit volume of air:

$$\varepsilon(\lambda) = \pi \int_0^\infty r^2 Q_{ext}(r,n,\lambda) \frac{dN(r)}{dr} dr \tag{2}$$

where the integral should run over the full size spectrum. The extinction efficiency factor $Q_{ext}(r,n,\lambda)$ is a function of the complex refractive index of the particle $n(\lambda)$, particle size and the wavelength (or rather the ratio between size and wavelength) and is given by Mie theory.[2] The important implication of equation (2) is that the aerosol extinction is thus a function of particle size and composition (refractive index).

The variation of the extinction coefficient with the wavelength can be approximated by a power law function:[10]

$$\varepsilon(\lambda) = \gamma_c \lambda^{-\alpha} \tag{3}$$

where γ_c is a constant and α is the Ångström coefficient (also known as Ångström exponent or Ångström parameter). Usually, this parameter is determined in the spectral range from 440 nm to 870 nm using a sun photometer (provided that the sun is visible). The public-access Aerosol Robotic Network (AERONET)[14] performs passive measurements of

aerosol properties by ground-based sun photometers at more than 300 locations all over the world, and instantaneous (or delayed, quality inspected) measurements of the Ångstrom coefficient are available on-line.

It should be noted that a sun photometer provides the integrated spectral extinction of an atmospheric column of air (from the sensor towards the sun), which is generally referred to as the aerosol optical thickness $\tau_a(\lambda)$:

$$\tau_a(\lambda) = \int_0^\infty \varepsilon(\lambda, z)\, dz \approx \int_0^\infty \varepsilon(\lambda)\, f(z)\, dz \tag{4}$$

where $f(z)$ represents the vertical distribution of aerosols as function of altitude z. For practical purposes, it is often assumed that the majority of the aerosols are contained in the boundary layer, and that the concentration within the boundary layer is altitude-independent or follows an exponential decay. In any case, it should be kept in mind that the Ångström coefficient provided by a sun photometer thus reflects altitude-integrated aerosol properties and is not necessarily equal to the Ångström coefficient that would have been measured over a horizontal path.

2.2 Modeling aerosol mixtures by the Ångstrom coefficient

Figure 1 may illustrate how changes in the concentrations of various aerosol species impact on the Ångström coefficient. The Figure shows that continental aerosols (0th mode) are generally smaller than marine aerosols. Furthermore, Mie theory shows that the major contributions to the extinction efficiency factor at wavelength λ are due to particles that have a size that is more or less comparable to λ. Hence, the continental particles affect primarily the aerosol extinction at smaller wavelengths, whereas the marine aerosols have a stronger impact on longer wavelengths. Consequently, an increase in the concentration of continental particles will increase the aerosol extinction at shorter wavelengths more than at longer wavelengths, and thus result in a larger value of the Ångström coefficient.[11] This feature can be exploited to provide a relation between Ångström coefficient and the ratio of continental to marine aerosols.

The ANAM4 model was used to generate a relation between AMP and the Ångström coefficient, which is shown in Figure 2. The Figure shows the expected positive correlation between AMP and the Ångström coefficient, but it also shows that the exact relation depends on the other input parameters of the ANAM as well. Higher winds cause higher concentrations of marine particles, thereby flattening the aerosol distribution (cf. Figure 1) and thus lowering the Ångström coefficient. One curve (labeled X) shows the effect of replacing the dust-like particles in the 0th mode by soot, which has a higher absorption. This increases the Ångström coefficient, albeit by a relatively modest amount. Figure 2 shows that the maximum value of AMP = 10 is reached for Ångström coefficients of 0.5 – 1.3, depending on the other input parameters of the ANAM4. Since the Ångström coefficient can attain values as high as 2 to 2.5 in polluted conditions,[13] the model shown in Figure 2 is incomplete and does not provide a solution in a heavily polluted atmosphere.
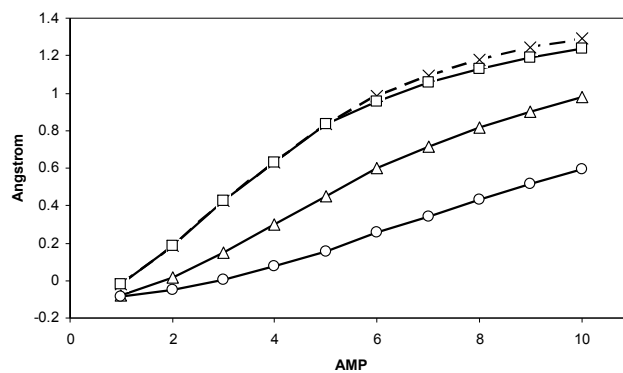

Figure 2: Relation between AMP and Ångström coefficient. Circles: $U_{10} = U_{24} = 20$ m/s; Triangles: $U_{10} = U_{24} = 10$ m/s; Squares: $U_{10} = U_{24} = 5$ m/s. X-signs: $U_{10} = U_{24} = 5$ m/s, with soot instead of dust-like particles in the 0th mode.

2.3 Extended modeling of aerosol mixtures by the Ångstrom coefficient

The extended model considers an aerosol mixture consisting of sea-salt aerosols (SSA), water-soluble aerosols (WS) and black carbon (BC). We are aware that these three components do not capture the full variability of the aerosol composition in the coastal zone, but we have selected WS and BC since these two have the greater impact on extinction. In contrary to the first two aerosol components (SSA and WS) which are hygroscopic, the latter aerosol component (BC) is assumed to be insoluble. The effects of changes in the relative humidity on the size distributions of hygroscopic species are taken into account by an approach described by Tang.[15]

It further assumed that the aerosol optical thickness can be modeled using the widely-used external mixing approach.[16] This means that each aerosol component of a given mixture is represented by its own single mode size distribution and a single complex index of refraction. The aerosol optical thickness is then the weighted average of the individual extinction coefficients using volume concentration percentages. The center radii r_n [μm] and the widths σ of the single mode number size distributions are the following: $r_n = 0.4$, $\sigma = 0.51$ for SSA, $r_n = 0.05$, $\sigma = 0.46$ for WS and $r_n = 0.0118$, $\sigma = 0.3$ for BC. The refractive indices of sea-salts, anthropogenic salts and black carbon are taken from McClatchey et al.[15]

Within this approach, the aerosol optical thickness is given by:[12]

$$\tau_a(\lambda, RH) = C_V^{tot}(RH) \left\{ \frac{C_V^{WS}(RH)}{C_V^{tot}(RH)} \psi^{WS}(\lambda, RH) + \frac{C_V^{SSA}(RH)}{C_V^{tot}(RH)} \psi^{SSA}(\lambda, RH) + \frac{C_V^{BC}}{C_V^{tot}(RH)} \psi^{BC}(\lambda) \right\} \qquad (5)$$

Where $C_V^{tot}(RH)$ in [μm³cm⁻²] is the total volume of all aerosols in the whole column:

$$C_V^{tot}(RH) = C_V^{SSA}(RH) + C_V^{WS}(RH) + C_V^{BC} \qquad (6)$$

and $\psi(\lambda, RH)$ in [cm²μm⁻³] is the ratio of aerosol extinction in [cm⁻¹] and volume concentration $\varsigma_V(RH)$ in [μm³cm⁻³]:

$$\psi(\lambda, RH) = \frac{\varepsilon(\lambda, RH)}{\varsigma_V(RH)} \qquad (7)$$

For various mixtures of the three aerosol species, and for various values of the relative humidity, the aerosol extinction at several wavelengths between 400 and 850 nm was calculated, and subsequently, the Ångström coefficient.[12] These simulations allowed parameterizing the volume contributions of individual species as polynomials of the Ångström coefficient α, e.g.:[13]

$$\frac{C_V^{BC}}{C_V^{tot}(RH)} = \sum_{i=0}^{5} A_i^{BC}(RH) \, \alpha^i \qquad (8)$$

where A_i^{BC} are the coefficients of a 5th degree polynomial.

2.4 Implementation in ANAM

The above model was implemented in ANAM, which permitted the removal of the Air Mass Parameter as an input parameter at the cost of introducing the Ångström coefficient as a new input parameter. In the presence of the Ångström coefficient model, the original dust-like 0th mode at nominal centre radius 0.03 μm is removed from the ANAM and replaced by two lognormal modes representing black carbon (BC) at centre radius 0.0118 μm and water-soluble aerosols (WS) at nominal centre radius 0.05 μm (cf. right panel of Figure 1). The values of the Ångström coefficient and relative humidity are subsequently used to determine the relative volume contributions of BC, WS and sea-salt aerosols (SSA) using the set of polynomials of which an example is given in equation (8). To convert these contributions into absolute mode concentrations, it is assumed that the advection mode (2nd mode) with a nominal center radius of 0.24 μm can be taken as the sea-salt aerosol (SSA) mode in Kusmierczyk-Michulec's analysis[12] (nominal mode radius 0.4 μm, see above). The absolute amplitude of the 2nd mode is calculated as function of wind speed and fetch[11] and thus provides the gauge for the relative amplitudes of SSA, BC and WS.

The above procedure then suggests removing the marine background (1st) mode from the ANAM, since sea-salt aerosols should now be covered by the 2nd mode. However, in pure marine air masses the Ångstrom coefficient becomes rather small and the contributions of BC and WS become virtually zero, which implies that ANAM provides only very few particles with diameters less then 0.1 μm as the contribution of the 2nd mode also rapidly decreases below 0.1 μm. This is not in agreement with experimental evidence suggesting that even a pure marine air mass contains a significant number of ultrafine particles. Hence, it was decided to retain the marine background (1st) mode in ANAM, as an independent mode that is not coupled to the Ångström coefficient and the relative contributions of SSA, BC and WS. The amplitude of the 1st mode was (rather arbitrarily) fixed at the amplitude previously corresponding to an AMP-value of 1.

Retaining the 1st mode is questionable, since one might argue that the Ångström coefficient model describes sea-salt aerosols as a single lognormal nominally centered at 0.4 μm. However, it is well-known that the size spectrum of marine aerosols extends to both shorter and longer diameters,[17] which suggests that the single lognormal is no more than an approximation. We feel that the presence of the 1st mode is justified as an omnipresent background concentration of ultrafine sea-salt aerosols that is not related to the meteorological scenario. This is equal to stating that the residence time of these particles is such that a homogeneous concentration can build up over the World's oceans.

3. EXPERIMENTAL DATA

To test the concept of the modeling of the smaller particle modes by the Ångström coefficient, a validation campaign was set up. The experiments took place over several timeframes between November 2010 and March 2011 at the island of Porquerolles, located near Toulon at the French Riviera (Mediterranean Sea). The measurement site at the extreme west of the island (indicated by a solid square in the left panel of Figure 3) allows for an obstructed view into all directions but due North. The air masses arriving at the station can be almost purely marine (win directions 160 – 200°N, corresponding to infinite fetch), but it is equally possible that the air mass originates from the heavily polluted Rhone delta (limited fetch conditions).

The primary instrumentation of the site consisted of 3 optical particles counters (SMPS TSI model 3081, PMS CSASP-200, PMS CSAP-100HV) providing a combined measurement range for particles ranging between 10 nm and (nominally) 40 μm over 195 (partially overlapping) diameter size bins (cf. right panel of Figure 3). Data of individual probes was sampled for 1 second and stored on disk, later reduced to 5 minutes and combined into a single size distribution by a software program. Prior to the experiment, the probes had been intercalibrated as to ensure smooth overlap between data of individual probes (note that this is not an absolute concentration calibration). The CSAP-100HV failed halfway through the experiment and was replaced by a PMS CSAP-100HV-ER to ensure continued collection of larger particles.

Meteorological data measured at the site were wind speed, wind direction, air and sea temperatures and relative humidity. The aerosol counters and meteorological sensors were located at heights of 22 and 30 metres, respectively,
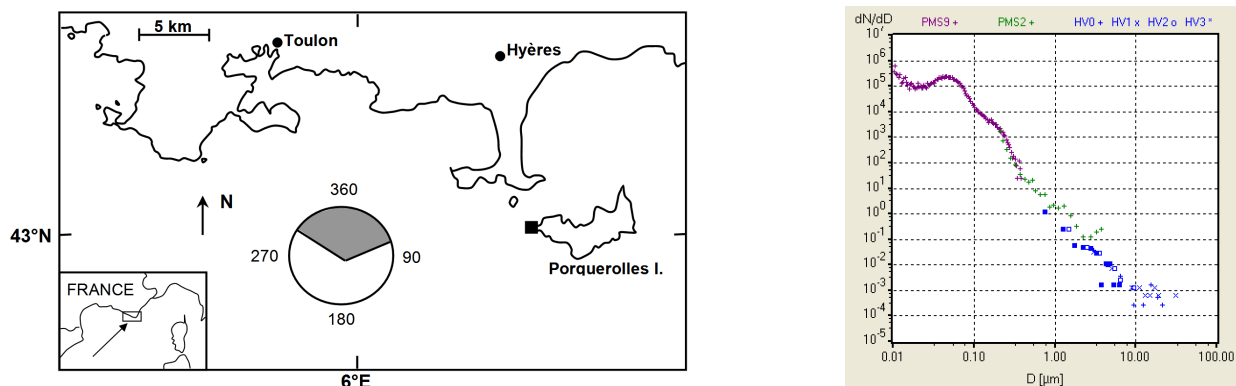

Figure 3: Left panel: Field site at Porquerolles Island (black squares) and wind rose showing wind directions principally corresponding to fetch-limited (gray area) and infinite fetch. Right panel: Typical composite aerosol distribution as measured at Porquerolles.

above the sea surface. The AERONET sun photometer of the University of Toulon was relocated to the measurement site at Porquerolles and provided aerosol optical thickness. These data were uploaded to the AERONET site, from which Ångström coefficients could be retrieved after initial data quality control.

Unfortunately, a substantial part of the trial timeframe was characterized by adverse weather severely limiting the deployment of the sun photometer as only very few days had cloud-free conditions. Furthermore, the aerosol probes ran off a power generator that required daily fuelling trips to the Porquerolles Island. Overall, 36 days of aerosol data was collected, but only 252 records (of 5-minute average, which amounts to 21 hours of data) coincided with measurements of the Ångström coefficient.

4.   RESULTS AND DISCUSSION

In view of the larger amount of aerosol data as compared to Ångström coefficients, the initial analysis focussed on the aerosol distributions. The 36 days of data provided a good range of meteorological conditions ($2 <$ Tair $< 15°$C, $35 <$ RH < 100%) with wind speeds ranging from 0 to 15 m/s (albeit with most data smaller than 10 m/s) and wind directions covering the full wind rose (with relatively more data in the 30-100°N and 220-280°N intervals). The lower values of the humidity and wind speed were found for the 30-100°N wind interval, whereas higher wind speeds and humidity were observed for the infinite fetch interval 220-280°N corresponding to a more marine origin of the air mass. The limited amount of Ångström coefficients was mostly recorded during lower wind speeds, and higher Ångström coefficients were observed for the fetch-limited wind directions in accordance with the more polluted origin of the air masses.

For marine wind directions, the concentration of larger aerosol particles (2 and 5 μm diameter) correlated positively with wind speed in agreement with the wind-driven production mechanisms for sea-salt aerosols. The slope of the regression lines increased when excluding very low wind speeds (U < 3 m/s), at which the production mechanism is inefficient. In contrast, the concentration of smaller particles (0.1, 0.2 and 0.5 μm diameter) showed marginal or negative correlation with wind speed. This suggest a wind-driven dispersion and deposition mechanism in the absence of sources, which corroborates with the predominantly non-marine origin of these smaller aerosols.[18] The ratio between smaller and larger particle concentrations increased for non-marine wind directions (suggesting more advection of land-originated particles and less production of marine particles), while the concentration of the 0.5 μm remained virtually constant. For these non-marine wind directions, the correlation between larger particles and wind speed almost vanished, whereas the correlation between smaller particles and wind speed became more pronounced. All these observations suggest that the aerosol data exhibits the well-established behavior of the coastal zone, which provides confidence in the measurements.

In the next step, aerosol extinction at the wavelengths of 0.5 and 1.0 μm were calculated from the experimental size distributions using Mie theory. For the calculation, the size distribution was integrated between 0.1 and 20 μm particle diameter, assuming a humidity-dependent refractive index for sea-salt aerosols. In addition, the meteorological conditions corresponding to each data record were fed into the ANAM to predict aerosol extinction at these same two wavelengths. Two set of predictions were made: one for the reference ANAM4 (assuming AMP-values ranging from 2 to 6 depending on wind direction), and one for the new ANAM6 (using the measured values of the Ångström coefficient). For ANAM6, the surf-aerosol mode was turned off, the fetch ranged from 50 to 300 km depending on wind direction, and the average fetch was set to 100 km. In all, ANAM4 provided results for 1300 data records, whereas ANAM6 only provided results for 210 data records due to the limited amount of sun photometer data (Ångström coefficient). The ANAM models were also used to predict the aerosol concentrations at 0.1 and 0.2 μm particle diameter.

Table 1: Performance factors for ANAM4 and ANAM6. N = number of records; C = concentration, ε = extinction

	N	C(0.1 μm)	C(0.2 μm)	ε (0.5 μm)	ε (1.0 μm)
ANAM4 (ref)	1300	3.2	7.4	4.2	5.1
ANAM6 (new)	210	39.5	12.8	8.6	8.6

A performance factor will be introduced to evaluate the success of ANAM in predicting concentrations and extinctions. The performance factor F is defined as $10^{\wedge}\sigma$, where σ is the standard deviation of a scatter plot of predicted and experimental values with respect to the identity line $x = y$. Differently phrased: F is the factor to within ANAM predicts the experimental concentration or extinction with 67% confidence. The performance factors for the various ANAM calculations results are summarized in Table 1. This Table shows that the reference version of ANAM4 scores a performance factor of 3.2 in predicting the concentration of 0.1 μm particles. The new ANAM6 with the Ångström coefficient scores much worse than ANAM4 in prediction aerosol concentrations, especially for the smallest particles of 0.1 μm diameter. ANAM6 does a better job in predicting extinctions, and the difference with ANAM4 is less pronounced, which demonstrates the beneficial effect of adding a strong absorber such as black carbon to the aerosol mixture. Nevertheless, the older ANAM4 outperforms ANAM6 on all accounts.

Since ANAM is an empirical model in which the relations between input parameters and aerosol concentration and/or extinction can be tuned to experimental observations, it is possible to redefine the relation between Ångström coefficient and the relative mode ratios of sea-salt, black carbon and water-soluble aerosols to better match the observations. There is reason to do so, since the theoretical model described in section 2 is based on 3 lognormal distributions, whereas the sea-salt contribution in ANAM spans a much larger size domain. However, the present dataset of 210 records seems rather minimal for empirical tuning.

Upon closer inspection, it was noted that ANAM4 tends to overestimate the aerosol concentration (especially for 0.2 μm particles) and extinction (especially for $\lambda = 0.5$ μm), whereas ANAM6 underestimates concentration (equally for 0.1 and 0.2 μm particles) and extinction (especially for $\lambda = 0.5$ μm). Thus, ANAM4 provides too many dust-like aerosols and ANAM6 provides too few black carbon and water-soluble aerosols to match the observations. This may suggest that neither model provides the exact composition and concentration of the actual aerosol loading at Porquerolles Island. This then may point a more fundamental limitation in the modeling of aerosol properties in the coastal zone on the basis of local meteorological parameters. As mentioned in the introduction, the aerosol loading in the coastal zone comprises a complex mixture of various types of aerosols originating from a large number of (localized) sources on the nearby land. Without a detailed map of these sources, and a detailed mesoscale dispersion model, it is almost impossible to predict the actual aerosol concentration and composition at a given coastal location. This leads to the conclusion that further tuning of the ANAM6 and/or the introduction of additional aerosol species may certainly improve the performance of the model for the Porquerolles dataset at hand, but may not result in an enhanced overall performance for any coastal location around the globe.

In conclusion, a new algorithm was introduced in ANAM to account for the contribution of (smaller) land-originated aerosols in the coastal zone. This extends the traditional ANAM-mixture of marine and dust-like particles to a 3-component mixture of black carbon, water-soluble particles and sea salt. The new algorithm is driven by the Ångström coefficient, which replaces the ill-defined air mass parameter (AMP) that has since long complicated the use of ANAM in the electro-optical propagation community. A limited validation trial at the French Riviera shows that the new algorithm has limited capabilities for predicting the aerosol properties. On the one hand, this suggests a need for including even more aerosol species, on the other hand it is questionable whether a model driven by local meteorological parameters will ever be able to capture the variability of the aerosol loading in the coastal zone that is governed by mesoscale processes. This latter hesitation is underlined by simulations using the older ANAM4, which reproduces the observed data better than ANAM6, using only a 2-component model and the "guestimate" parameter AMP. Nevertheless, we recommend using ANAM6, because this model is driven by the Ångström coefficient, which has physical relevance and can be measured experimentally. Additional validation efforts are needed to conclude whether the currently better performance of the older ANAM4 is incidental or structural.

REFERENCES

[1] Kneizys, F.X., L.W., Abreu, G. P. Anderson, J.H. Chetwynd, E.P. Shettle, A. Berk, L.S. Bernstein, D.C. Robertson, P. Acharya, L.S. Rothman, J.E.A. Selby, W. O. Gallery and S. A. Clough, [The Modtran 2/3 report and Lowtran 7 model], Philips Laboratory PL/GPOS, Hanscom AFB, MA (1996).

[2] Mie, G., "A contribution to the optics of turbid media, especially colloidal metallic suspensions," Ann. Phys. 25, 377-445 (1908).

[3] Gathman, S.G., "Optical properties of the marine aerosol as predicted by the Navy Aerosol Model," Opt. Eng. 22, 57-62 (1983).

[4] Van Eijk, A.M.J., L.H. Cohen, L.J. Navarro and G. De Leeuw. "Near-surface aerosol transmission in the marine environment," SPIE Proceedings 4884, 160-169 (2002).

[5] Van Eijk, A.M.J. and D.L. Merritt, "Improvements in the Advanced Navy Aerosol Model (ANAM)," SPIE Proceedings 6303, (2006).

[6] Van Eijk, A.M.J. and J. Kusmierczyk-Michulec, [Review of the marine modes in ANAM], TNO report TNO-DV 2007 C382, (2007).

[7] Van Eijk, A.M.J., J. Kusmierczyk-Michulec, M.J. Francius, G. Tedeschi, J. Piazzola, D.L. Merritt and J.D. Fontana, "Sea-spray aerosol particles generated in the surf zone," Accepted for publication in Journal of Geophysical Research, doi:10.1029/2011JD016197, in press, (2011).

[8] Van Eijk, A.M.J., and G. De Leeuw, "Modeling aerosol extinction in a coastal environment," SPIE Proceedings 1688, 28-36 (1992).

[9] Piazzola, J., F. Bouchara, de Leeuw, G. and A.M.J. van Eijk, "Development of the Mediterranean extinction code (MEDEX)," Optical Engineering 42, 912-924 (2003).

[10] Ångström, A., "On the atmospheric transmission of sun radiation and on dust in the air," Geogr. Ann. 11, 156-166 (1929).

[11] Kusmierczyk-Michulec J. and A.M.J. van Eijk, "Ångström coefficient as a tracer of the continental aerosols," SPIE Proceedings 6708, (2007).

[12] Kusmierczyk-Michulec, J., "Ångström coefficient as an indicator of the atmospheric aerosol type for a well- mixed atmospheric boundary layer: Part 1: Model development,", Oceanologia 51(1), 5-39 (2009).

[13] J.T Kusmierczyk-Michulec and A.M.J. van Eijk. "The Angstrom coefficient as an indicator of non-marine particles in ANAM (Advanced Navy Aerosol Model)," SPIE Proceedings 7463, (2009).

[14] Holben, B. N., T.F. Eck, I. Slutsker, D. Tanre, J.P. Buis, A. Setzer, E. Vermote, J.A. Reagan, Y.J. Kaufman, T. Nakajima, et al., "AERONET-A Federated Instrument Network and Data Archive for Aerosol Characterization," Remote Sensing of Environment 66(1), 1–16 (1998).

[15] Tang I.N., "Chemical and size effects of hygroscopic aerosols on light scattering coefficients," J. Geophys. Res., 101 (D14), 19245–19250 (1996).

[16] McClatchey, R. A., H. -J. Bolle, K. Y. Kondratyev, J. H. Joseph, M. P. McCormick, E. Raschke, J. B. Pollack, D. Spänkuch, and C. Mateer, [A preliminary cloudless standard atmosphere for radiation computation], Intl. Radiat. Comm., Boulder CO, (1984).

[17] O'Dowd, C.D. and G. DeLeeuw, "Marine aerosol production: a review of the current knowledge," Phil. Trans. R. Soc. A 365, 1753-1774 (2010).

[18] Van Eijk, A.M.J. and G. de Leeuw, "Modeling aerosol particle size distributions over the North Sea", J. Geophys. Res. C97, 14417-14429 (1992).