Aerosol extinction in coastal zone

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ABSTRACT

The performance of electro-optical systems can be substantially affected by aerosol particles that scatter and absorb electromagnetic radiation. A few years ago, an empirical model was developed describing the aerosol size distributions in the Mediterranean coastal atmosphere near Toulon (France). This model has been coupled with Mie theory to yield the code MEDEX (MEDiterranean EXtinction) for the aerosol extinction. This contribution deals with the evaluation of MEDEX for aerosol data recorded near the Black Sea coast. For this site, MEDEX correctly predicts the aerosol extinction as function of wavelength, albeit with minor discrepancies below one micron. These differences are attributed to the uncertainty in predicting the concentrations of submicron particles. The comparison shows that MEDEX may be more generally applicable than to the Toulon area.

Keywords: coastal aerosols, optical propagation, fetch

1. INTRODUCTION

The effective range of electro-optical devices is largely determined by the molecules and aerosol particles suspended in the atmosphere. These atmospheric constituents scatter and absorb radiation, resulting in transmission losses and hence a reduction of the effective range. While the impact of the molecules can be relatively well assessed, the effects of aerosols are more difficult to infer due to large spatial and temporal variations in concentration and composition. In particular, in coastal areas, concentrations and optical properties of aerosols are not well-characterized due to the occurrence of several specific processes.

The aerosol in coastal zone consists of a complex mixture of particles generated at the sea surface by the interaction between wind and waves, and a continental contribution emitted from natural and/or anthropogenic sources. The exact concentration and composition depends on a multitude of (meteorological) parameters, such as wind speed and wind direction, and the distance to the various sources. In this matter, the fetch parameter (the distance an air mass has traveled over sea) can be useful for assessing the relative contribution of marine and continental aerosols.

The model that is most frequently used for the prediction of aerosol extinction in the marine atmosphere is the US Navy Aerosol Model (NAM)[1]. However, although NAM provides reasonable predictions for the extinction over the open ocean, experimental evidence shows that the model is less reliable in coastal regions.[2,3] To remedy this deficiency, empirical models were developed for the prediction of aerosol concentrations in the coastal atmosphere, notably the West Coast of Ireland near Inisheer [4] and the Toulon region in the Mediterranean Sea.[5] The latter model was coupled with Mie theory to provide the extinction code MEDEX (MEDiterranean EXtinction),[5] which shows good performance for coastal extinction in the Mediterranean.[5] In view of the large variability of aerosols in coastal areas, the question is to what extent the predictions of MEDEX can be extended to other geographical coastal locations. As a first step, the present parameterization based on data obtained in the Mediterranean sea will be applied to another geographical location, namely the Northern Black Sea coast. A comparison between the Black Sea coast data and the Mediterranean model was proposed by Piazzola and Kaloshin[6] since the two coastal sites are often exposed to air mass transport with a large continental trajectory, and hence, with characteristics of mixed origin from the aerosol point of view. Indeed, the Black Sea is a quite closed sea with a strong influence of continental air masses coming from East or Southwest directions. This is quite similar to most of the wind conditions encountered in the Mediterranean site.[5] The present paper discusses a comparison between the extinction coefficient as inferred from aerosol measurements at the Black Sea coast and the predictions of MEDEX.

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2. FIELD SITE AND INSTRUMENTATION

Aerosol extinction in the Black Sea coast was obtained on the basis of an extensive series of measurements in summer season in Cape Tarhankut off the western part of Crimea Black Sea (Fig. 1). The site is characterized by marine winds coming from Black Sea, and hence, the fetch has to be considered. The extinction coefficient in the 0.44-11.9 µm band was obtained using a transmissometer with a spectral resolution of about 0.02 µm in the band 0.44-1.06 µm and 0.15-0.20 µm in the band 1.06–11.9 µm. Atmospheric extinction coefficients along the horizontal propagation paths were calculated from to the ratio between power of the received and the transmitted pulses. The received power was recorded every 30 nsec. The sensor data were recorded every 10 minutes. The propagation paths were located at heights of 2.5, 4 and 20 m above the sea surface. The length of measuring paths varied from 2 to 7.5 km. In addition, simultaneous measurements of visibility were achieved using nephelometric technique. In addition, the temperature, humidity, pressure and wind speed were provided using meteorological sensors installed on the top of the receiver.

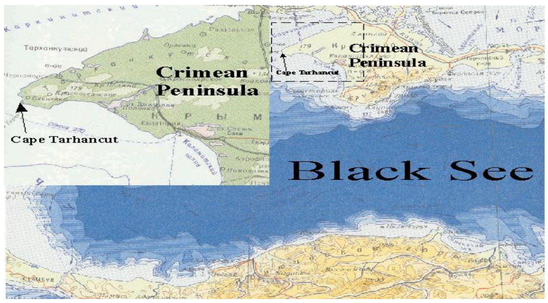

Fig. 1 : Map of the Black Sea with a zoom of the Crimea coast showing the location of the experimental site.

3. THE CODE MEDEX

The code MEDEX[5] consists of the Mediterranean coastal aerosol model (see below) coupled to a Mie program. The extinction coefficient, σ(km^{-1}), of electromagnetic radiation by aerosol particles is given by :

$$\sigma = \int Q(r,\lambda,m)\cdot N(r)\cdot \pi r^2 dr \qquad (1)$$

where $N(r)$ is the particle size distribution, i.e., the number of particles per cubic centimeters per microns, and $Q(r,\lambda,m)$ is the extinction efficiency of a particle (assumed to be spherical) with radius r and complex refractive index m at wavelength λ. The choice of the values for the refractive index m has an important impact on the calculation of the aerosol extinction. Because the prevailing winds at the Mediterranean coastal site causes the air masses to spend considerable distance over water, MEDEX assumes that marine aerosols dominate the distribution. Therefore, the refractive index for sea salt as proposed by Volz[7] is used in the model. As mentioned above, The MEDEX model calculates the particle size distribution $N(r)$ from the Mediterranean aerosol model, but it is also possible to enter a measured aerosol size distribution. The output of MEDEX consists of the aerosol size distribution and extinction at a height of 25 meters. In addition, MEDEX offers an option to calculate the vertical profile of aerosol extinction, from 0 to 25 m height. The format of the output is compatible with the input format of MODTRAN, which allows MEDEX to supply MODTRAN with aerosol extinction parameters.

3.1 Aerosol size distribution in coastal environment

The prediction of the aerosol extinction depends on the accuracy of the aerosol size distribution model. The aerosols in coastal zones consist of a complex mixture of aerosol particles, i.e., a marine component generated at the sea surface by the interaction between wind and waves, and a continental contribution emitted from natural and/or anthropogenic sources. The aerosol mixture in coastal zones depends critically on the wind direction. Changes in the wind direction are accompanied by variations in meteorological parameters such as relative humidity, atmospheric stability and boundary layer height. In turn, this results in changes in the physical processes affecting aerosols (generation, transport, deposition), thus causing a great variability of the aerosol size distribution and composition. In particular, the variation in wind direction is accompanied by a change of fetch, i.e., the distance traveled over water by an air mass before reaching the measurement location. The fetch influences the whitecap coverage and hence the sea surface-generated particle concentration, since wave breaking is different for a fully developed sea compared to growing wave field periods.

For example, we have reported in Fig. 2 polynomial representations of aerosol distributions measured for different fetches at a wind speed of about 10 m/s in the Mediterranean coast by Piazzola et al.[5] The concentrations of coarse particles, which are assumed to be dominated by sea-salt particles, increase with the fetch. This effect becomes more prominent as particle size increases. In contrast, the concentrations of sub-micrometer particles decrease at larger fetch. Fig. 2 then shows the transition from a continental to a marine aerosol as the air mass is advected over the sea.

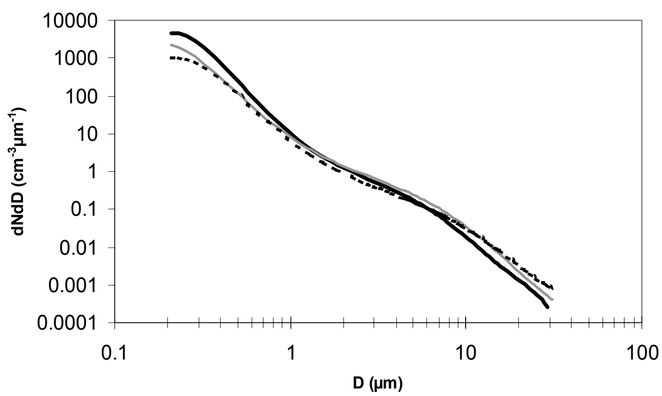

Fig. 2 : Examples of particle size distributions recorded for a wind speed of 10 m/s at various fetches in a Mediterranean coastal zone.[5] The thick black curve represents the size distribution recorded for a fetch of 13 km, the grey line is the data measured for a fetch of 25 km and the dashed line is for a 100 km fetch.

3.2 The model for the Mediterranean coastal aerosol size distribution

The coastal aerosol model is based on a Mediterranean dataset that was acquired on the island Porquerolles near Toulon (France) between 2000 and 2001. The experimental data from Porquerolles were statistically analyzed to develop an empirical coastal aerosol model as a modification of the Navy Aerosol Model (NAM) published by Gathman.[1] A comparison between the Black Sea coast data and the Mediterranean model was proposed by Piazzola and Kaloshin[6] since the two coastal sites are often exposed to air mass transport with a large continental trajectory, and hence, with characteristics of mixed origin from the aerosol point of view. Indeed, the Black Sea is a quite closed sea with a strong influence of continental air masses coming from East or Southwest directions. This is quite similar to most of the wind conditions encountered in the Mediterranean site.[5]

As in NAM[1], the particle size distribution *N(r)* of the coastal aerosol model, is calculated as the sum of modified lognormal functions (Eq. (3)), but the amplitudes of the various modes are parameterized as functions of fetch. Furthermore, as suggested by De Leeuw[8] and in accordance with the more recent Advanced Navy Aerosol Model (ANAM),[9] a fourth mode has been introduced to model the largest sea spray particles, i.e., *N(r)* is calculated as the sum of four modified lognormal functions:

$$N(r)=\sum_{i=1}^{4}\frac{A_i}{f}\exp(-C_i(\ln(r/fr_{oi}))^2) \quad (3)$$

where $r_{o1} = 0.03$, $r_{o2} = 0.24$, $r_{o3} = 2$ µm, $r_{04} = 10$ µm, f represents the humidity growth factor, A_i denotes the i^{th} mode amplitude (in $cm^{-3}\mu m^{-1}$) and C_i is the width of the i^{th} mode.

Empirical relations for the amplitudes and widths as function of fetch and wind speed have been given by Piazzola et al.[5] These relations were obtained from regression analysis of the aerosol concentration and wind speed or fetch. As discussed in detail elsewhere,[4,5] the variation of the particle concentrations with the wind speed results from the relative contribution of both marine and land-originated aerosol. For larger fetch, the marine-originated particles prevail, resulting in a larger (positive) slope of the concentration/wind speed plots (increasing production with increasing wind speed). At shorter fetch, a relatively large fraction of the aerosol originates from overland. This is evident in the intercept of the concentration/wind speed plots, which exhibit a larger intercept with decreasing wind speed. This is due to the less efficient dispersion of the land-originated particles at lower wind speeds. In contrast with the Navy Aerosol Model (NAM) where the continental influence was limited to the first mode describing the smallest particles, the Mediterranean coastal model introduces the fetch in all modes. In this manner, coastal effects on the (larger) marine aerosols can also be taken into account. Such effects include the fetch-dependence of the wave field resulting in enhanced or reduced wave-breaking, and hence, marine aerosol production.

The fetch dependence of mode amplitudes A_2, A_3 and A_4 for the Mediterranean aerosol model was determined on the basis of the Porquerolles dataset using using linear regression on the concentration *vs*. wind speed for particles of 0.24 µm, 2 µm and 10 µm radius. Having obtained the parameterization of mode amplitudes A_2, A_3 and A_4, the four widths, C_i, were obtained from a multi-variable fit to the experimental size distributions as function of wind speed and fetch. Optimization of the widths was obtained by assuming a limited range of variations of the mode radii, since they correspond to a physical characteristic of the aerosol size distributions. Subsequently, plots of the regression parameters (slopes and intercepts of the concentration/wind speed plots) versus fetch were fitted to an exponential function. This procedure resulted in mode amplitudes as functions of wind speed and fetch.
On the basis of more recent measurements on the island of Porquerolles,[10] the Mediterranean model was slightly modified to better take into account the aerosol size spectra measured for very short fetches, which represent complex conditions to model. This has been published in Piazzola and Kaloshin.[6] Fig. 3 shows the particle size distribution as measured in the Mediterranean coastal zone and the corresponding model (upgraded version) prediction for different fetch and wind speed conditions. The figure shows an excellent agreement between the (upgraded) Mediterranean coastal aerosol model and the measurements.

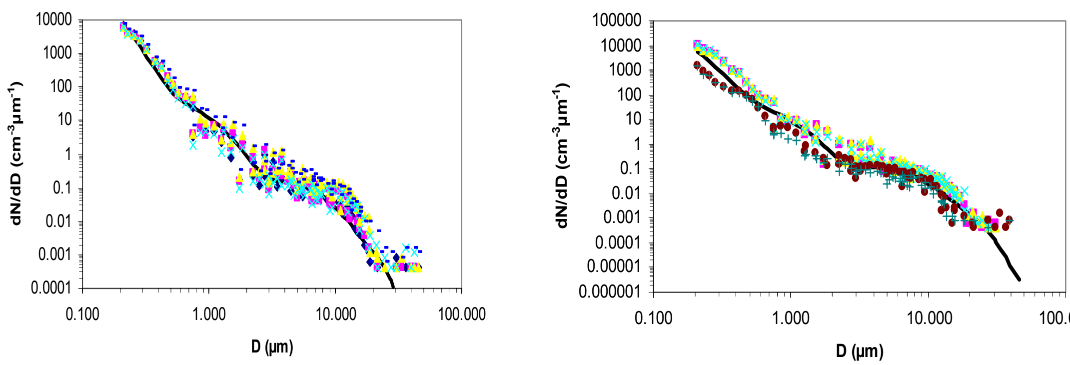

Fig. 3 : Measured particle size distribution and the predictions of the latter version of the coastal aerosol model (solid line) after Piazzola and Kaloshin[6] for a 3 km fetch and for wind speeds between 6.5 and 8 m/s (left) and for a 13 km fetch and for wind speeds between 9.5 and 11 m/s (right).

4. AEROSOL EXTINCTION AT THE BLACK SEA COAST

In this section, the aerosol extinction obtained at the experimental site in the Black Sea is compared to the predictions of the code MEDEX. This comparison represents a true test for the representativeness of MEDEX, since the model that has been developed on the basis of Mediterranean datasets is now applied to a different geographical location.

4.1 Extinction during light wind periods

Fig. 4 shows a comparison between the aerosol extinction measured at the Black Sea coast and the prediction by MEDEX for a low wind speed scenario. Fig. 4 (left) concerns a fetch of 1 km and a wind speed of 3.3 m/s while Fig. 4 (right) deals with a fetch of 30 km.

Slight differences between MEDEX and the measured extinction are noted for the lower wavelengths due to the difficulty of predicting the concentration of sub-micron aerosol particles, i.e. the heavily continentally influenced first mode of the modeled aerosol spectrum. Above 3 µm in wavelength, the present figures reveal a remarkably good performance of MEDEX. This indicates that the inclusion of fetch allows for a good characterization of coastal effects.

This gives MEDEX a definite advantage over NAM for moderate to low off-shore wind speeds when land-originated aerosols dominate the distribution. Moreover, the figures suggest that the MEDEX parameterization can be applied to different geographical sites. However, this last conclusion must be viewed with caution since MEDEX is a purely empirical model,. The MEDEX parameterization is likely to break down under extreme conditions, such as heavily polluted sites or very shallow waters.

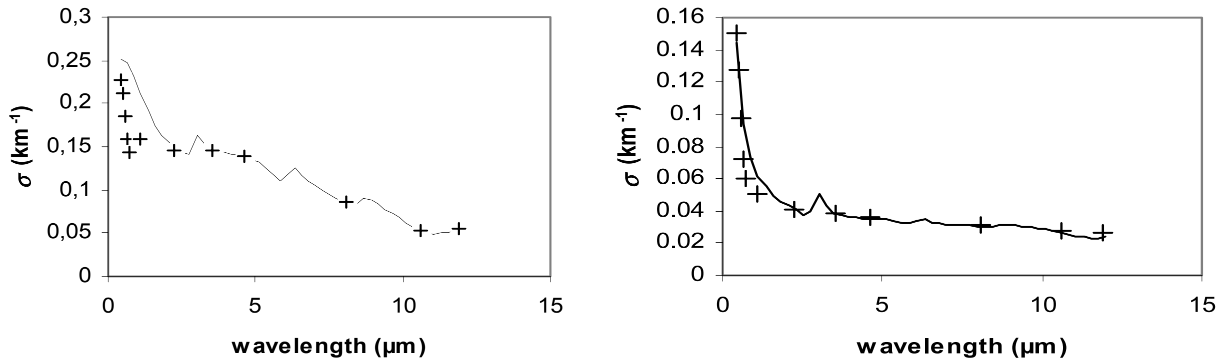

Fig. 4: Comparison between the aerosol extinction recorded in the Black Sea coastal zone and the predictions of MEDEX for a wind speed of 3.5 m/s and a relative humidity of 85 %. The crosses represent the Black Sea measurements while the line is the MEDEX output. Figure 4 (left) : Fetch of 1 km ; Figure 4 (right) : Fetch of 30 km.

4.2 Extinction during high wind speed periods

For high wind speeds, the predictions of MEDEX also show an excellent agreement with the measured extinction at the Black Sea coast (Fig. 5). This is probably not only due to the inclusion of fetch, but also to the addition of the fourth lognormal mode in the model. This mode yields a relatively high number of large particles, representing (locally generated) marine aerosols. This kind of production is expected at higher wind speeds. Of course, at such a short fetch the presence of surf-generated aerosols cannot be ruled out. Whatever the origin of the aerosols, the fourth mode causes MEDEX to yield a higher concentration of large aerosols as compared to NAM. This is especially important for the prediction of the extinction at thermal infrared wavelengths. For example, the extinction at 10.6 µm calculated by the ANAM model is about a factor of 2 higher than the one given by NAM due to the additional 4th mode.[9]

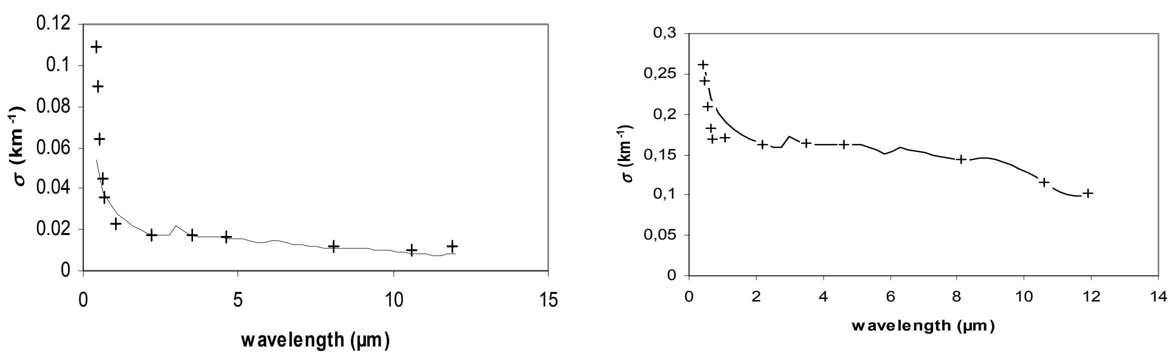

Fig. 5: Comparison between the aerosol extinction recorded in the Black Sea coastal zone and the predictions of MEDEX for a wind speed of 15 m/s and a relative humidity of 85 %. The crosses represent the Black Sea measurements while the line is the MEDEX output. Figure 5 (left) : Fetch of 1 km ; Figure 5 (right) : Fetch of 30 km.

5. FUTURE IMPROVEMENT OF MEDEX

The results shown in the previous section suggest that MEDEX is more generally applicable than to the Mediterranean coast near Toulon. This would significantly increase the value of the MEDEX code and merits further improvements in the code. The previous section shows that MEDEX predicts the aerosol extinction for the Black Sea site well. The discrepancies at smaller wavelengths are attributed to the uncertainty in predicting the concentrations of submicrometer particles. However, it is also possible that the refractive index plays a role here. One of the main improvements of MEDEX planned for the future deals with the role of the refractive index *m* in the extinction calculation. For an aerosol distribution of mixed origin as encountered in the coastal zone, the differences in refractive index could induce quite different absorption and scattering effects according to the exact composition of the particle. The imaginary part of the coefficient *m* represents the absorption effect while the real part of m is concerned with the scattering. The imaginary part, usually called *n* and the real part *k* of the refractive index *m* for an aqueous droplet are given by :

$$n = n_w + (n_0 - n_w)(\frac{r_w}{r_0})^{-3} \quad (1)$$

$$k = k_w + (k_0 - k_w)(\frac{r_w}{r_0})^{-3} \quad (2)$$

where the indices *w* and *0* refer to the wet phase (water) and the dry phase, respectively.

As mentioned in section 3, MEDEX uses the refractive index of sea salt.[7] For sea salt particles, the absorption coefficient is very small compared to the scattering coefficient. As a consequence, the single scattering albedo is nearly unity. In an urbanized coastal zone, such as the French Mediterranean coast, the particles transported in the atmosphere will not only be of marine, but also of rural and anthropogenic origin. The latter particles have refractive indices differing considerably from that of sea salt. In the urban zone, anthropogenic sources emit predominantly sulfate aerosols[11,12] and carbonaceous aerosols, i.e., black Carbon.[13,14] For the sulfur compounds, it is well-recognized that the absorption coefficient is quite low,[15] as is the case for sea salt. However, for black Carbon, things are quite different since the soot compounds are known to be one of the most absorbing aerosols.[15] This implies that these particles are not well represented by the refractive index of sea salt. Since the sizes of these rural and anthropogenic carbonaceous particles generally fall in the submicrometer part of the aerosol spectrum, a misfit in refractive index will mostly effect the smaller wavelengths, below 1 microns. Thus, the influence of smaller anthropogenic particles could explain the relative poor fit between MEDEX and observations at smaller wavelengths as compared to larger wavelengths. The above discussion shows the importance of assessing the impact of soot (and other rural and anthropogenic compounds) on the extinction in the coastal areas. For a correct calculation of the extinction, both the refractive index and the concentrations of these aerosols must be established. Experimental efforts are currently planned to provide these properties. Aerosol particle counters and impactors will assess the concentrations and also allow for a chemical analysis of the aerosols. The data will subsequently be used to upgrade MEDEX for multi-component mixtures with the ultimate goal to better model the extinction in the coastal zone.

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