Aerosol transport in the coastal environment and effects on extinction

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ABSTRACT

The aerosol in the coastal environment consists of a complicated mixture of anthropogenic and rural aerosol generated over land, and sea spray aerosol. Also, particles are generated over sea by physical and chemical processes and the chemical composition may change due to condensation/evaporation of gaseous materials. The actual composition is a function of air mass history and fetch. At the land-sea transition the continental sources cease to exist, and thus the concentrations of land-based particles and gases will gradually decrease. At the same time, sea spray is generated due to the interaction between wind and waves in a developing wave field. A very intense source for sea spray aerosol is the surf zone. Consequently, the aerosol transported over sea in off-shore winds will abruptly change at the land-sea transition and then gradually loose its continental character, while also the contribution of the surf-generated aerosol will decrease. The latter will be compensated, at least in part, by the production of sea spray aerosol. A Coastal Aerosol Transport model (CAT) is being developed describing the evolution of the aerosol size distribution in an air column advected from the coast line over sea in off-shore winds. Both removal and production are taken into account. The results are applied to estimate the effect of the changing size distribution on the extinction coefficients. In this contribution, preliminary results are presented from a study of the effects of the surf-generated aerosol and the surface production.

Keywords: coastal aerosol, surf aerosol, sea spray, aerosol transport model, extinction, EOPACE

1. INTRODUCTION

The detection range of electro-optic sensors is determined by the properties of the propagation medium. One of the factors that need to be taken into account is scattering and absorption by molecules and aerosols determining the transmission of electromagnetic radiation at wavelengths in the visible and infrared. The effects of molecular extinction are much better modelled than those of aerosols. The main uncertainty is the description of the aerosol particle size distributions as function of meteorological parameters and geographical location. In particular coastal regions are a problem due to the large variety of sources ashore. At the land-sea transition, aerosol sources and sinks change abruptly. In off-shore flow continental sources cease to exist, and thus the concentrations of particles produced over land will gradually decrease. A very strong, but also very local, source for sea spray aerosol is the surf zone[1,2]. Thus, the composition of the aerosol changes instantaneously at the coast line, from a continental aerosol to a mixture of continental and surf-generated sea spray aerosol. The mixing ratio of both aerosol types depends on the concentrations advected from land, and on the source strength and dispersion properties of the surf-produced aerosol. This mixture is gradually removed due to deposition and other processes. Since the removal processes are size-dependent, not only the concentrations will change but also the size distributions. At the same time, at wind speeds exceeding 3-4 ms^{-1}, sea spray is produced in response to wave breaking (and thus the developing wave field) and dispersed into the Boundary Layer (BL). This mechanism compensates, at least in part, the loss of surf-produced sea spray particles.

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Meteorologically, the land-sea transition will result in a non-homogeneous condition in the atmospheric boundary layer due to changes in surface roughness, surface temperature, and surface fluxes of momentum, heat and moisture. In response, both the wind, temperature and humidity fields, and the thermal stratification will evolve until a new dynamic equilibrium has been reached. The influence of the land-sea transition may extend up to 100-200 kilometres from the coast line. A special meteorological feature is the development of surface waves in response to the action of the wind, resulting in a fetch-dependent wave field and surface drag or friction velocity, while also an internal boundary layer is expected to develop in response to the changing thermal structure and roughness.

Effects of meteorological parameters and fetch on the aerosol size distribution in coastal areas are evident from experimental data obtained, e.g., at the North Sea[3], the Danish coastal waters[4] and on the Irish Atlantic coast[5]. Processes were indicated that may contribute to the observed wind speed dependence of the size distribution[3], but the variation of production, dispersion and deposition with wind speed cannot explain the experimental results. A line source model was applied to estimate the effect of removal on the concentrations of surf-produced aerosol[1]. Since such an approach does not allow for the inclusion of extended surface sources, the development of the Coastal Aerosol Transport Model (CAT) was started. CAT describes the evolution of the aerosol size distribution and composition with fetch in a column that is advected out over the sea. The processes that are presently included in CAT are vertical mixing through diffusion and gravitation, surface production, removal by dry deposition and condensation/evaporation of water vapour. All these processes depend on the meteorological situation. CAT is still under development and other aerosol processes such as the interaction between aerosols and gases may be taken into account, while also a more extensive description of meteorological processes is planned.

Some preliminary results from CAT are presented which are focused on an analysis of the contribution of surf aerosol to the total sea spray size distributions. The results are applied to quantify the contributions of surf- and surface-produced aerosol to the extinction in the atmospheric transmission windows in the IR wavelength regions (3-5 and 8-12 μm). Results are presented for fetches of up to 25 km. These distances were chosen in compliance with the range of modern IR detection systems. The effect of the surf aerosol on the IR extinction in the atmospheric transmission windows is discussed.

CAT is under development and will be severely tested. In this contribution the preliminary results are presented. Full results will be presented in Vignati (1999)[6].

2. MODEL DESCRIPTION

In CAT, the aerosol is composed of continental and sea spray particles. Each component is characterised by a discrete size distribution with 26 classes ranging from about 0.04 to 12.59 μm dry radius. The geometrical mean radius of each class is given by $r_i=0.01\times10^{(i/10)}$, where i is an integer. The choice of the extremes of the interval is determined by the selection of the parameterisations used in the model, as will be described below.

The aerosol, an external mixture of continental and sea spray particles, is distributed over an air column divided in 10 vertical layers. In each layer the particle concentration is homogeneous. The depth of each layer increases with height to allow for a finer resolution close to the ground where the influence of the surface has a relatively large effect on the particle populations. The air column is advected along a horizontal path that is determined by the local wind field.

In the model, the fate of the particles is determined by a series of physical processes such as emissions, vertical turbulent diffusion, gravitational settling, and dry deposition. The time evolution of the particle concentrations $N_{type,i}$ in class i (type = 1 for continental particles and type = 2 for sea spray) is described in the generic layer L by the following equation:

$$\frac{dN_{type,i,L}}{dt} = \delta_{type,2}\delta_{L,1}E_i - (\kappa_{L-1}+\kappa_L)N_{type,i,L} + \kappa_L N_{type,i,L+1} + \kappa_{L-1}N_{type,i,L-1} + S_{type,i,L+1}N_{type,i,L+1} - S_{type,i,L}N_{type,i,L} - \delta_{L,1}\lambda_{type,i}N_{type,i,L} \quad (1)$$

where E is the sea spray production rate, κ_L is a parameter dependent on the exchange coefficient, S is the rate of particle gravitational settling, and λ is the deposition rate. δ is the Kronecker function.

This set of equations is solved numerically using the fully implicit Crank-Nicholson method[7].

In off-shore winds the continental aerosol is advected across the surf zone where their sources vanish. The aerosol size distribution and chemical composition change instantly due local production of sea spray aerosol in the surf. Recent experiments have shown that the surf production may be very significant[1]. A model for the surf flux is presented in Neele et al.[2] In this contribution, results are present using surf-generated aerosol concentrations derived from the experimental data[1], as described in section 3.

The concentrations in the continental/surf aerosol mixture decrease gradually with fetch due to removal by dry deposition (wet deposition and entrainment are presently not taken into account), while at the same time sea spray aerosols are produced at the sea surface. For the flux, F, of surface-generated particles the formulation of Smith et al.[8] is used:

$$\frac{dF}{dr} = \sum_{i=1,2} A_i \exp\left[-f_i\left(\ln\frac{r}{r_{0i}}\right)^2\right] \qquad (2)$$

where r is the particle radius, and the amplitudes A_i are wind speed dependent coefficients given by:

$$\mathrm{Log}(A_1) = 0.0676U + 2.43$$
$$\mathrm{Log}(A_2) = 0.959U^{1/2} - 1.476 \qquad (3)$$

The constants f_1, f_2, r_{01} and r_{02} have values of 3.1, 3.3, and 2.1 and 9.2 μm respectively. Equation (2) is valid for actual radii of up to 20 μm (at 80% relative humidity), corresponding to a dry radius of approximately 10 μm. According to the authors, care must be taken in extrapolating the formula to larger sizes. Although the source function is based on measurements of particles larger than 1 μm (in situ), in CAT Equation (2) is extrapolated down to a wet radius of 0.07 μm.

Close to the ground, particles are deposited with a rate depending on the wind speed and on particle size. The deposition velocities are parameterised using the two-layer model of Slinn and Slinn[9]. They are different for the two types of aerosols, because the actual particle size is determined by the chemical composition.

Vertical transport of aerosol particles due to turbulence is parameterised by an eddy diffusivity formulation. The computations presented in this contribution are made for a column with a depth of 260 m, which is representative for the marine boundary layer height off the Californian coast. The diffusivity coefficient K_L, is assumed to vary linearly with height in the surface layer (arbitrary chosen here as the layer adjacent to the surface up to 50m):

$$K_L = k u_* z_L \qquad (4)$$

k is the von Karman constant (equal to 0.4), u_* is the friction velocity and z_L the upper height of the layer L. Above this height the diffusivity coefficient is constant up to 163 m and then decreases to zero at the upper boundary of the column (Figure 1).

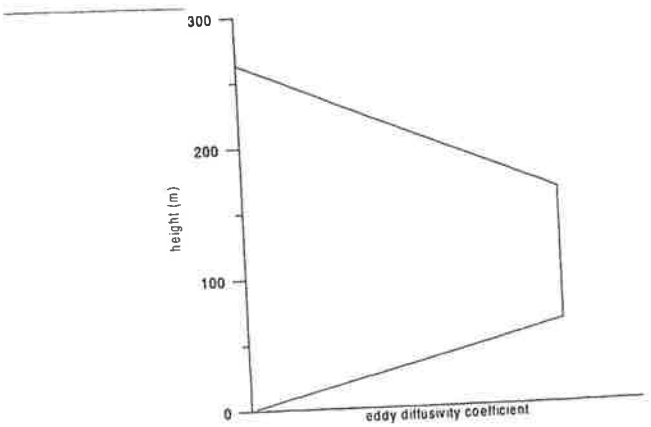


Figure 1. Profile of the eddy diffusivity coefficient used in the simulations.

The relative humidity varies with height above the surface. When particles are transported between different layers, these variations cause a change in the aerosol size distribution due to condensation or evaporation of water vapour. The response of the particles to the variations in relative humidity depends on their chemical composition.

In the model the particles are supposed to be always in equilibrium with the relative humidity in each layer. The characteristic time for a sea salt particle to reach equilibrium, as function of air temperature and relative humidity, was reported by Andreas[10]. The lower the temperature and the higher the relative humidity, the more time is needed for a sea salt particle to attain its equilibrium radius. For instance, at a temperature of 20°C, a particle emitted at the surface with a radius of 10 μm needs 10s to attain the equilibrium value at a relative humidity of 95%. The characteristic time determines the lower limit of the time step that can be used in the calculation. The higher limit is controlled by the turbulent diffusion, which is a fast process and therefore needs a short time step to be accurately described. The use of an implicit method allows to increase this low number to a value that can guarantee the equilibrium condition. In this work, a time step of 150s has been chosen. This is the time it takes for a particle emitted at the surface with a wet radius of 50 μm to come to equilibrium. This means that the particle population cannot extend to very large sizes, or the equilibrium condition is not fulfilled. The dry particle radius corresponding to a wet radius of 50 μm is circa 12.5 μm. Only particles with dry radius not larger than this value can be included in the model. According to Andreas it is uncertain whether droplets with initial radius larger than 40 μm stay in the air long enough to transfer moisture. This uncertainty only applies to particles belonging to the last class. With this upper limit of the particle size, also the proper application of (2) is insured.

The equilibrium between the droplets and the ambient relative humidity is calculated by[11]

$$S = a_w \exp\left(\frac{2M_w\sigma}{RT\rho_w r}\right) \tag{5}$$

where S is the saturation ratio, a_w the water activity, M_w and ρ_w the molecular weight and density of water, σ the surface tension, R the gas constant, T the temperature, and r the equilibrium radius. Water activity and surface tension depend on the particle chemical composition.

The choice of the continental aerosol chemical composition can be made based on measurements at the site or on emission inventories. One possible choice is to identify it with $(NH_4)_2SO_4$ (ammonium sulphate). Sea salt particles are more hygroscopic than ammonium sulphate. Therefore, for the same dry radius, wet sea salt particles are larger than wet ammonium sulphate. Sea spray particles are considered dry for relative humidity below 75%, while for ammonium sulphate particles this occurs for relative humidity below 80%.

The range of validity for the application of thermodynamic parameters used in the model determines the choice of the smaller particle class.

3. MODEL RESULTS

Preliminary results are presented from simulations with CAT. Since the model is still being developed as regards the meteorological description, these results must be interpreted with some care. For example, when an air mass moves from land out over sea, an Internal Boundary Layer (IBL) may develop. Vertical diffusion is therefore changing with the distance from the coastline. In the present simulation this variation is not included. Also effects of entrainment at the top of the marine boundary layer (MBL) and exchange between the MBL and the IBL are not yet included. Further, relative humidity, air temperature, wind speed, friction velocity, etc., are independent of fetch in the present simulations.

The results presented here focus on the contribution of surf produced aerosols to the evolution of the sea spray size distributions, therefore continental aerosols are not included in the calculations. Results are shown for the layer centred at 12 m above the sea level. All results are plotted for a wet radius.

Aerosols produced over the surf were measured during the EOPACE (Electro-Optical Propagation Assessment in Coastal Environment) experiment in La Jolla, California, in January and February 1996 and in March 1997[1,12]. Data are available for wind speeds up to 8 ms^{-1}. Plume heights extend up to about 30 m. The data were analysed to obtain size distributions as function of wind speed and height above the ground. In the following simulations, the results are used as inputs for the initial concentrations of surf-produced sea spray aerosol for wind speeds of 3 ms^{-1} and for 8 ms^{-1}. Also, to show the contrast,

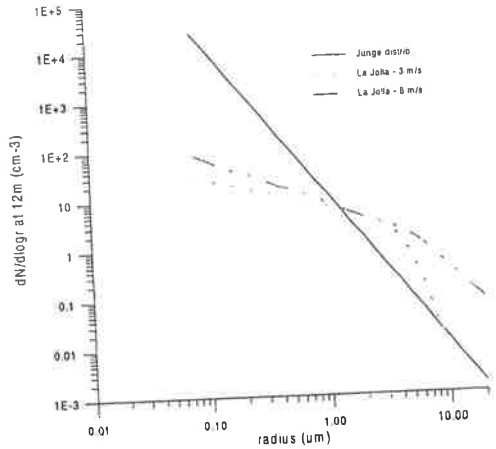
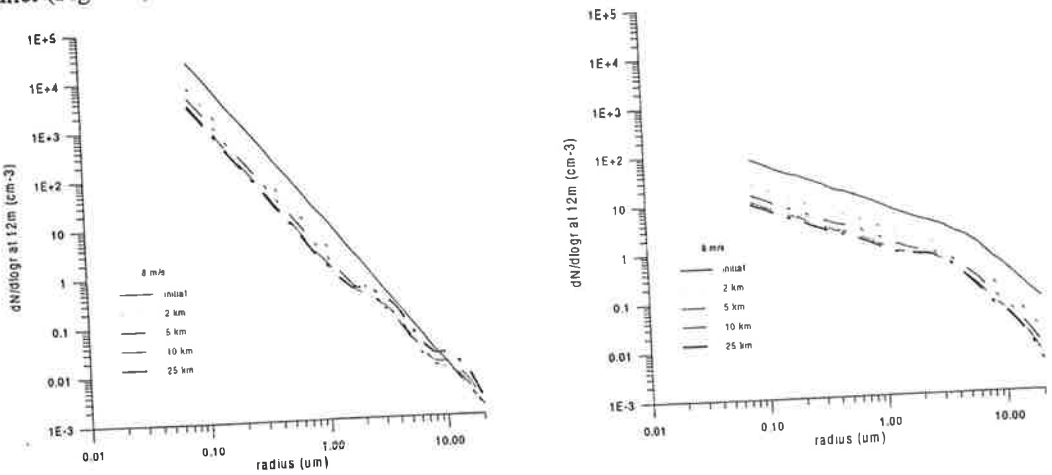


Figure 2. Initial sea spray size distributions over the surf.

simulations were made in which the surf-produced aerosol was described with a Junge distribution that is homogeneously distributed over the surf plume height of 30 m. The Junge distribution used here has a much steeper spectrum in which the number of sub-micron particles is much larger than for the EOPACE data while the concentrations of super-micron particles are much smaller (Figure 2).

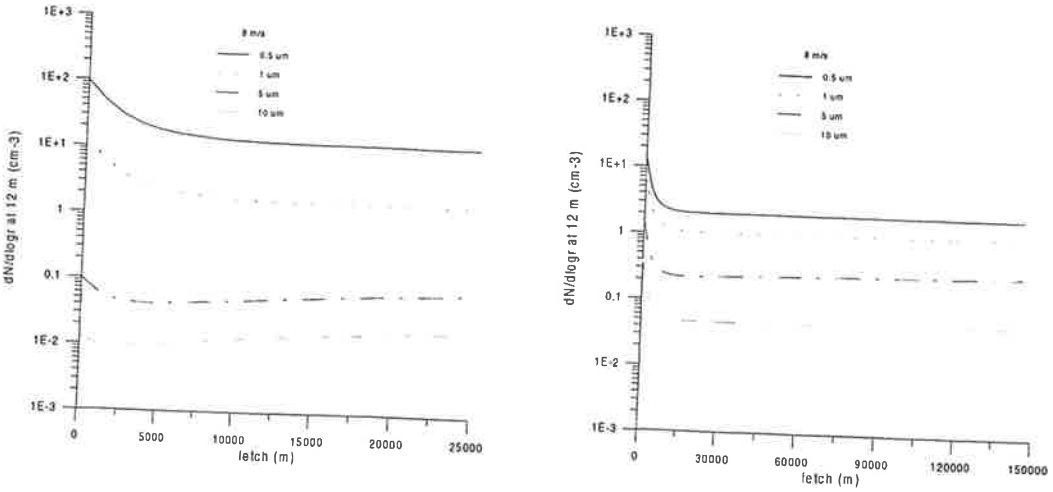


a) b)

Figure 3. Sea spray size distributions at 0, 2, 5, 10, 25 km from the coastline. Wind speed is 8 ms^{-1}. Initial conditions represented by a) a Junge distribution and b) EOPACE data.

In figure 3 the evolution of the sea spray size distribution is illustrated by the results obtained at fetches of 0, 2, 5, 10 and 25 km, for the two initial size distributions and a wind speed of 8ms^{-1}. For the initial Junge distribution (Figure 3a), the concentration of particles smaller than about 2 μm decreases due to diffusion to the upper layers where the initial particle concentrations were zero. For larger particles, the concentrations initially decrease due to diffusion and gravitational settling, but then increase due to the contribution of surface-produced aerosol in this size range. Already at 2 km fetch a mode appears at 10 μm, and at 5 km also another one at 2-2.5 μm. At 25 km the two modes are very pronounced. At the end of the simulation, a balance between production and removal seems to be reached, since the particle size distribution changes only little.

Figure 3b shows the evolution of the size distribution when EOPACE measurements are used as initial conditions. The concentrations decrease for the sizes. Nevertheless the two modes appear, although not as pronounced as the previous case.



a) b)

Figure 4. Particle concentrations as function of distance from the coastline for particles of 0.5, 1, 5, and 10 μm. Wind speed is 8 ms^{-1}. Initial conditions represented by a) a Junge distribution and b) EOPACE data. Note the difference in horizontal scales.

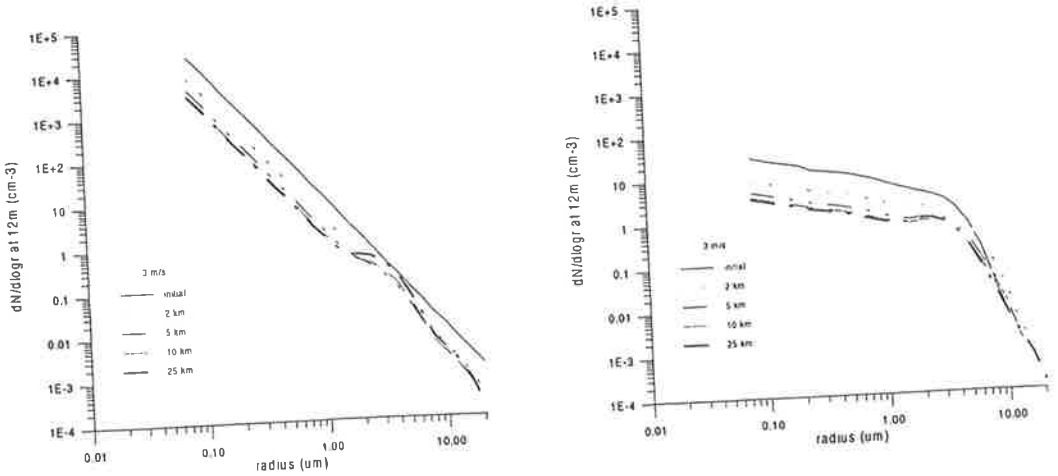
In Figure 4, particle concentrations at various wet radii are plotted as function of the distance from the coastline. The concentrations of particles of 1 μm and smaller initially decrease due to diffusion into the upper layers. When these small particles are uniformly distributed over the column, production and deposition, both small at these sizes, are more or less in balance. The concentrations do not visibly change during the rest of the simulation (total simulation time was 5 hours), which is in accordance with the long residence time of particles in these size ranges (days). These observations apply to initialisation of the surf-produced aerosol with both a Junge distribution and the EOPACE data.

For the larger particles there is a difference between these situations. Using the Junge distribution (Figure 4a), the concentrations of particles of 5 and 10 μm also decrease initially. After a few km they start to increase and exceed the initial concentrations at fetches of about 25 km and 7 km, respectively. This means that at larger fetches the effect of surface produced aerosol dominates the effect of the surf produced particles in this situation with a low surf production.

Figure 4b shows a situation with a large surf production as observed during the EOPACE experiments in La Jolla. In this case the surf-produced aerosol dominates the concentrations over the whole distance simulated. In fact, after 5 h of travel time the concentrations for all sizes change only little, indicating that the particle size distributions are not far from the steady state situation.

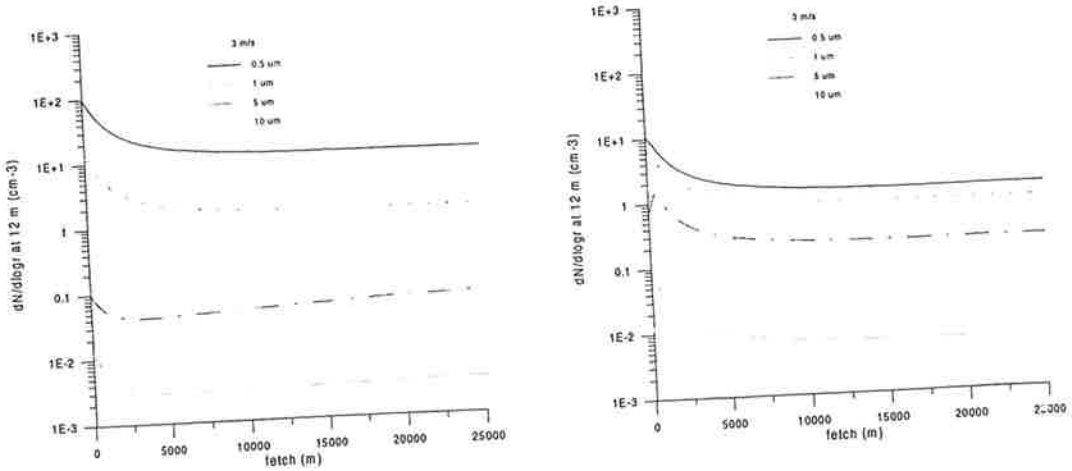
For a wind speed of 8 ms^{-1}, it takes 52 minutes for the column to be transported over a distance of 25 km. When the wind speed is 3 ms^{-1} it takes 2 hours to cover this distance. Therefore there is more time to produce particles of about 2-3 μm at the surface in a sensible concentration. No matter which initial distribution is chosen, the particle number concentrations at these sizes are slightly larger at 3 ms^{-1} than at 8 ms^{-1} (Figures 5a and 6a). It seems awkward that apparently at a wind speed of 3 ms^{-1}, where whitecapping is very small, larger sea spray concentrations are found than at a higher wind speed (8 ms^{-1}), for which significant whitecapping and thus also significant surface production is expected. Below is shown that at relatively long fetches, the sea spray size distributions calculated with CAT compare favourably with experimental size distributions obtained in similar conditions.

Comparison between Figures 3 and 5 shows a significant difference between the development of the sea spray modes at 3 ms^{-1} and 8 ms^{-1}. Apart from production, also transport plays a role. The upward transport is a balance between turbulent diffusion and gravitational settling. At low wind speeds, the upward diffusion is slow and gravitation tends to keep the particles in the lower levels. Because gravitational settling depends on particle size, and is effective only for particles larger than a few μm, it takes a higher turbulence intensity to keep larger particles airborne. Hence, at 3 ms^{-1} the turbulence is too weak, as compared to 8 ms^{-1}, to lift the particles in the 10 μm mode that was so well developed at 8 ms^{-1}.



a) b)

Figure 5. Sea spray size distributions at 0, 2, 5, 10, 25 km from the coastline. Wind speed 3 ms^{-1}. Initial conditions represented by a) a Junge distribution and b) EOPACE data.



a) b)

Figure 6. Particle concentrations as function of distance from the coastline for particles of 0.5, 1, 5, and 10 μm. Wind speed is 3 ms^{-1}. Initial conditions represented by a) a Junge distribution and b) EOPACE data.

When the surf produced aerosol is initiated with the EOPACE data, the concentrations of the larger particles initially increase when the wind speed is 3 ms^{-1}, see Figures 5b and 6b. This can be explained by the initial distribution of the surf-produced particles over the column. The particle size distributions in the lower five layers, as derived from the EOPACE experimental data for this wind speed, are shown in Figure 7. In each layer, identified in Figure 7 by the mean height, the aerosol is homogeneously distributed. The concentration gradients are very strong, in particular for particles larger than 1 μm. The concentration at 12m height is initially small. Due to diffusion, the particles from the surface layer are transferred into the upper layers, and the concentration there increases. As a result, the concentrations initially increase until after a few km the number concentrations decline to the initial concentrations and lower. At about 10 km the concentrations are almost constant.

As a preliminary test for CAT, size distributions calculated with the model at a fetch of 25 km, for wind speeds of 3 and 8 ms^{-1}, are compared with measurements reported by Smith et al.[8] (see Figure 8). To emphasise the surface produced contribution, the

model was initialised with the Junge distribution for which the concentrations of the large particles are relatively low. Comparing the model results for the two wind speeds, with the experimental data for 0, 5 and 10 ms^{-1}, collected at 14 m above the sea surface, shows that concentrations predicted by the model are only slightly larger, but the shape of the distributions are consistent with the measurements. The assumption that the mixing height does not vary with the wind speed may lead to an overestimation of the concentrations, specially at high wind speeds. Nevertheless, taking into account the simple meteorological approximations used in the model, the results are promising.

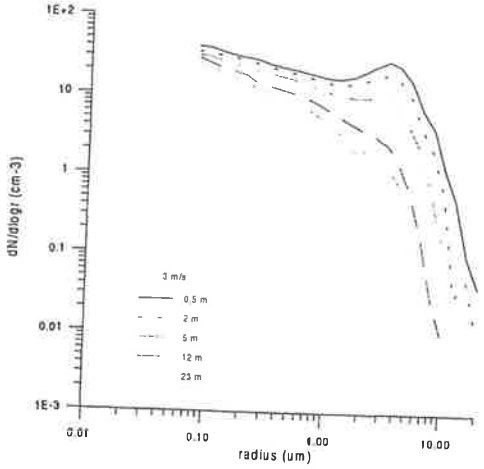


Figure 7. Mean particle size distributions as initialised in the lower five levels, derived from measurements during the EOPACE experiments for wind speeds of 3 ms^{-1}. The levels are identified by their mean height.

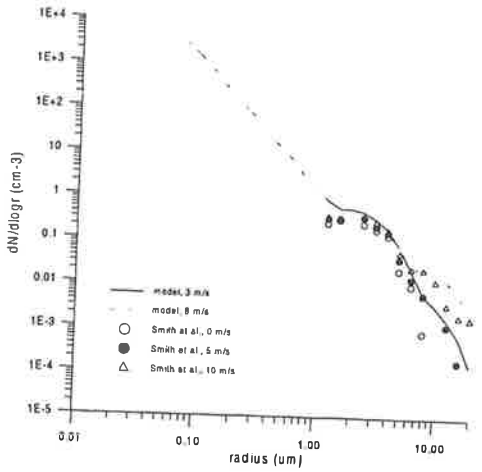


Figure 8. Comparison of model results with measurements reported by Smith et al.[8]

4. EFFECTS ON EXTINCTION

Extinction coefficients at various wavelengths in the visible and infra-red (IR) atmospheric transmission windows were calculated from the aerosol size distributions using an exact Mie code[13]. Results for wavelengths of 10.6, 4.0, 1.064, 0.6943 and 0.55 μm are shown in Figure 9, where they are plotted as a function of fetch at discrete distances from the coast. These five wavelengths were not only chosen because of their application in various kinds of sensor systems, but also to illustrate the influence of the different size classes.

The results with the experimental EOPACE surf size distribution show very little wavelength dependence, due to the small slope in the initial size distribution (Figure 2). Fitting of a power law distribution to this size distribution results in an approximate Junge coefficient of about -2.1 to -2.4. Thus the Ångström coefficient, giving the relation between the extinction coefficient and wavelength is close to zero.

When on the other hand the surf aerosol is initialised with a Junge distribution, with a Junge coefficient of -4, a strong wavelength dependence is observed. However, due to diffusion and deposition, the concentrations become smaller, while production is dominant for the larger particles. Hence the size distributions gradually change shape and the Junge coefficient increases (becomes less negative). As a result, the extinctions at the wavelengths in the infrared change faster than in the visible. In fact, the latter increase significantly in the 8 ms^{-1} case where a well-developed surface-produced mode is observed at the largest distance (Figure 3). After 25 km, the extinction has almost returned to it's initial value over the surf. It is further noted, that also in the visible the extinction increases between fetches of 10 and 25 km, due to the 3-5 μm mode.

In all cases the extinction initially decreases significantly with fetch, by a factor that varies with the initial condition and with the wind speed, of up to more than one order of magnitude. However, in contrast to the particle concentrations, the decrease continues over large distances from the coast, of 5-10 km in all cases. The reason for the different scales is that the extinction is an integrated effect over the whole size distribution. Thus the different behaviour of small and large particles is reflected in the variation of the extinction coefficients.

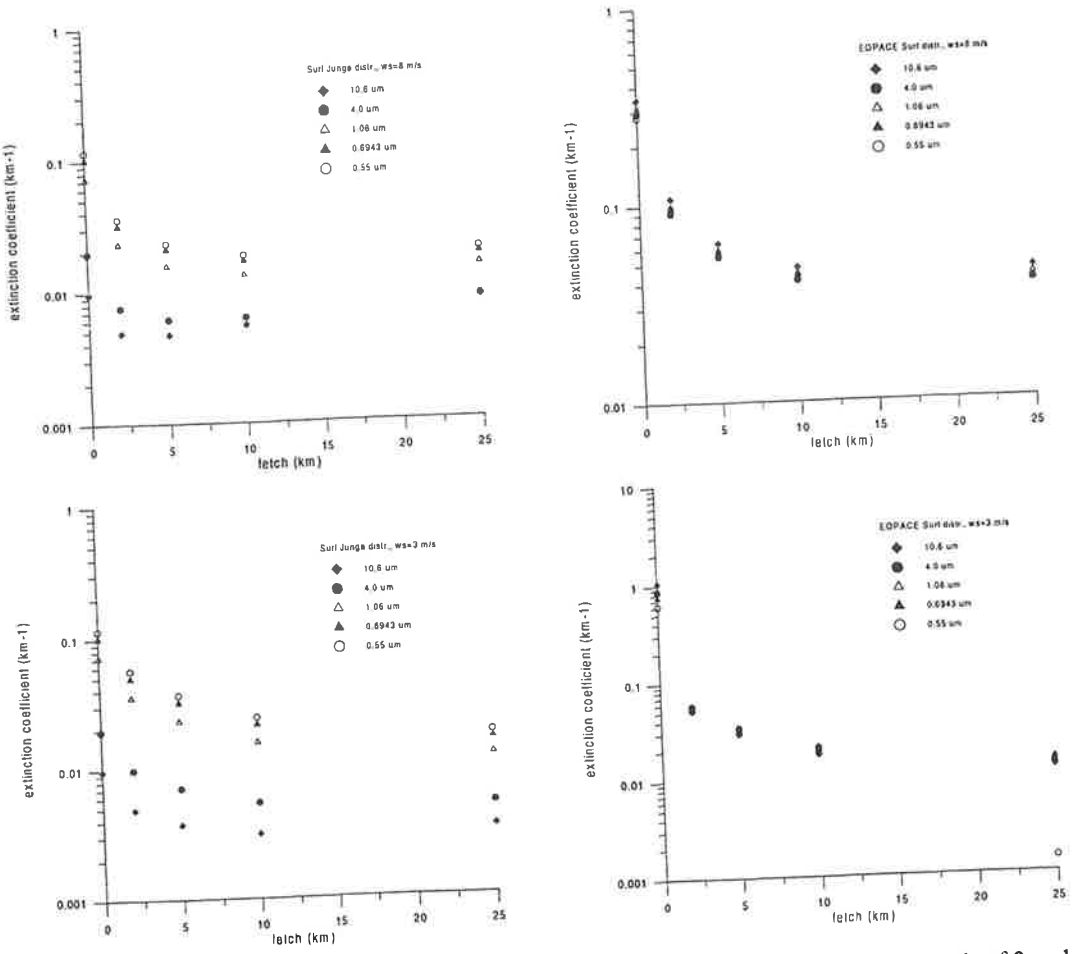


Figure 9. Extinction coefficient as function of fetch, for some visible and IR wavelengths, wind speeds of 3 and 8 ms^{-1} and different initial conditions (see legends).

5. DISCUSSION AND CONCLUSIONS

A Coastal Aerosol Transport model (CAT) has been developed and some preliminary results were presented. Only effects of sea salt production in the surf zone and at the surface by breaking waves were included in the present calculations. CAT was used to estimate the effect of the surf on the aerosol size distributions. Surf-produced concentrations for wind speeds of 3 and 8 ms^{-1} were derived from size distributions measured during the EOPACE experiments in La Jolla. Model calculations using these data were compared with results using an hypothetical Junge size distribution over the surf with relatively low concentrations in the super-micron range. For both cases, the influence of the surf on the size distributions extends to at least several km. When the surf production is low, the effect of surface production of the large particles (results were presented for 5 and 10 μm) becomes dominant. Also when the model is used with the measured surf-produced concentrations, the effects of surface production are visible in the size distributions. However, in that case the concentrations at fetches of up to 25 km are still much higher than in the case when surf-produced aerosol was described with the Junge distribution. This indicates that the surf has a significant effect on the sea spray concentrations at distances from the coast of at least 25 km, the largest fetch chosen in this work, but likely their influence will extend much further. Size distributions calculated with CAT at a fetch of 25 km compare favourably with experimental data measured in similar wind speeds.

Extinction coefficients were calculated from the size distributions obtained with CAT. At short fetches, the extinction coefficients are significant when the measured EOPACE surf source functions are used, but they decrease fast with fetch.

This fast decrease is initially mainly due to diffusion of the aerosol in the vertical, while later deposition and surface production determine the variation of both the aerosol concentrations and the extinction.

The present simulations were in the first place made to quantify the effect of surf-produced aerosols. Hence, continental aerosols were not included in the calculations. When passing the coast line, continental aerosols may be assumed to be uniformly and homogeneously distributed over the column. Hence, the observed initial strong decrease due to diffusion will not occur for the continental aerosol, and the changes in concentration will only be due to deposition and other removal processes. Thus, also the extinction due to continental aerosols is expected to vary much less with fetch.

Effects of continental aerosols on the total extinction will be included in future work. As indicated in the introduction, CAT is not finished yet. Nevertheless, the results are promising and CAT can be considered a very useful tool to study coastal aerosols and their effects on the propagation medium.

6. ACKNOWLEDGEMENTS

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