

Vertical profiles of aerosol scattering and absorption measured in situ during the North Atlantic Aerosol Characterization Experiment (ACE-2)

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ABSTRACT

In situ measurements of dry aerosol optical properties were made during the North Atlantic Aerosol Characterization Experiment in the vicinity of the Canary Islands. Profiles of aerosol scattering in 3 wavelengths and absorption coefficients are presented for 2 cases where North African mineral dust layers were found above a marine boundary layer (MBL) influenced by varying pollution level. The scattering and absorption coefficients showed large differences in magnitude and variability; not only between the 2 cases of mineral dust (e.g., the mean scattering in the green wavelength was $9.8 \times 10^{-6} \pm 1.6 \times 10^{-6} \text{ m}^{-1}$ and $2.1 \times 10^{-5} \pm 7.3 \times 10^{-6} \text{ m}^{-1}$), but also within a given layer (e.g., from less than 1×10^{-5} to $3.1 \times 10^{-5} \text{ m}^{-1}$ during the second flight). This was also true for the polluted MBL where it is likely due to origin, age and history of the air mass as well as on the mixing state of the MBL. The scattering, backscattering and absorption coefficients have been used to calculate aerosol optical properties such as hemispheric backscatter fraction, single scattering albedo, aerosol optical depth and the Ångström exponent, which are also presented. The hemispheric backscatter fraction was found to be smaller in the mineral dust layer than in the MBL, as was the wavelength dependence on scattering. The single scattering albedo of dry aerosols was found to vary between 0.73 ± 0.12 and 0.91 ± 0.12 in the dust layer and showed large variability within and between the dust cases. This might be due to the mixing state of the dust layer, but could also be explained by the dust having different optical properties depending on local origin within the same major source region.

1. Introduction

Aerosol particles influence the Earth's radiative balance directly by scattering and absorption of the incident solar radiation (Ångström, 1929; cf. Table 1 in Charlson, 1997). Their effect on the heat budget of the atmosphere has gained increasing attention, since it has been estimated (Charlson et al., 1990; Charlson et al., 1992) that the scat-

tering of short-wave radiation by anthropogenic aerosols creates a global radiative perturbation of comparable magnitude but opposite in sign to the forcing by anthropogenic greenhouse gases. This magnitude of radiative forcing and climatic impact of aerosols is still highly uncertain. For improved estimates and increased understanding of the effect aerosols have on climate, their optical properties need to be established over a wide range of aerosol types and atmospheric conditions. The ability to model climate and to assess impact of anthropogenically produced pollutants and desert dust events would be substantially increased with atmospheric measurements.

The net radiative effect of aerosol is probably

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most important over the oceans, due to the low albedo of the sea relative to land. Measurements of aerosol light scattering coefficients and absorption in marine locations (especially over the open ocean) are scarce. In those cases where in situ measurements have been made, they are mainly at ground level and they are unlikely to be representative of the whole air-column above the sampling site (Clarke et al., 1996) as both aerosol composition and humidity change with height in the atmosphere and both influence light scattering.

There is practical need for reliable interpretation of satellite-derived radiances (e.g., the Advanced Very High Resolution Radiometer (AVHRR); (Husar et al., 1997)), particularly for marine environments. To calibrate these retrieval products, comparisons with in situ measurements in the vertical are needed. Due to the suitability of marine regions for the satellite retrieval of aerosol properties, an improved characterisation of aerosols found in the marine environment is of particular interest.

Mineral dust and sea salt aerosols have among the highest atmospheric mass loadings on a global basis and are comparable in magnitude to each other (Jaenicke, 1993). Mineral dust aerosols can be a dominant feature in satellite observations of aerosol optical thickness where plumes from arid North African areas advect over the tropical and sub-tropical North Atlantic Ocean. The aerosol optical depth from African dust outbreaks over the North Atlantic has been measured to be up to 0.65–0.7 (Kidder and Vonder Haar, 1995) which is substantial compared to the global annual average of 0.12 and the unperturbed Southern Hemisphere value of 0.08 (Husar et al., 1997).

Since desert dust is found in the free troposphere its forcing has the potential to be more important than that of the aerosols found in the marine boundary layer (MBL). This effect is possible since the radiative forcing is reduced when aerosols are found at lower layers, due to the greater amount of scattering and absorption by the overlying atmosphere. Hence, less solar radiation is incident upon the aerosol and any radiation that is scattered in the upward direction by the aerosol has a greater chance of being scattered or absorbed before reaching the top of the atmosphere.

The measurements described here were all taken during “The North Atlantic Aerosol Characterization Experiment” (ACE-2) which was carried out

between Portugal and the Canary Islands in June and July 1997 (Raes et al., 2000). The focus of this paper is 2 outbreaks of dust aerosols from North Africa, found in the free troposphere, at the same time as the MBL was influenced by anthropogenic pollution from Europe. We have chosen to investigate 2 flights from the CLEARCOLUMN project (Russell and Heintzenberg, 2000) made during these episodes to try to answer the following questions.

- What is the vertical and horizontal variability in aerosol scattering and absorption for clear sky conditions in an atmosphere with a polluted MBL and a dust layer at higher altitudes over the Atlantic ocean in the ACE-2 area?
- What are the differences in optical properties within and between MBLs having different pollution levels?
- What are the differences in optical properties within and between the two North African mineral dust layers in the 2 cases we encountered during the ACE-2 campaign?
- What are the differences between the MBL aerosol optical properties and the same properties in the dust layer?
- What is the magnitude and variability in derived parameters such as aerosol extinction, aerosol single scattering albedo, hemispheric backscatter fraction, and aerosol optical depth over the Atlantic in the ACE-2 area in the same column of air?

2. Platform and instrumentation

The instrumentation used for this study was installed on the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) platform called “*Pelican*”. Details of the platform and its instrumentation during ACE-2 can be found in Bluth et al. (1996) and Russell and Heintzenberg (2000), and only the primary instrumentation used in the analysis in this paper will be mentioned here.

The instrument used for aerosol particle scattering coefficient measurements was a TSI® Inc. 3563 3-wavelength integrating nephelometer (Bodhaine et al., 1991; Heintzenberg and Charlson, 1996). It measured aerosol light scattering coefficient at 1 Hz sampling frequency in 3 wavelengths (450, 550 and 700 nm) at 2 ranges of angular integration (7° – 170° , total scattering σ_{sp} ; 90° – 170° ,

hemispheric backscattering σ_{bsp}). The nephelometer was calibrated before and during the campaign using CO_2 and particle free air. Before each flight and during flights the instrument was purged with particle free air to obtain zero baseline measurements. The scattering data presented here have been corrected for angular nonidealities (Anderson and Ogren, 1998), e.g., to allow for extinction budget studies and comparisons with satellite data, and have been smoothed by using a 30-s running average. The relative humidity (RH) of the sample air was measured within the nephelometer with the TSI supplied RH sensor using the manufacturers original calibration.

Aerosol light absorption coefficient (σ_{ap}) was measured with a sampling frequency of 1 Hz at 565 nm with the filter-based Particle Soot/Absorption Photometer (PSAP) continuous light absorption photometer from Radiance Research, Seattle, WA (Bond et al., 1999). The instrument uses the basic principle of Beer's law in which the change in optical transmission of a filter caused by particle deposition is related to the optical absorption coefficient and a calibration transfer coefficient. Due to the signal to noise ratio the absorption coefficient was later re-calculated using 30-s boxcar averages, and corrected with calculated volumetric flow and actual measured filter sample area. The absorption coefficient has also been corrected for artificially high absorption on the filter using the corrections given in Bond et al. (1999). This correction is necessary since filter-based methods will report an artificially high absorption coefficient if transmission through the

filter is reduced by the presence of scattering particles on the filter (Bond et al., 1999).

The air was sampled through an isokinetic inlet at the front of the aircraft and passed through cyclonic impactors. The cut size and the collection efficiency of the impactors are assumed to be the same as for those used at a number of NOAA background aerosol monitoring stations from which the design was copied, i.e., a nominal 50% cut size of $2.5 \mu\text{m}$ diameter for a unit density particle at STP. This is a reasonable assumption for aqueous particles in the MBL, but for the dust particles which have densities in the order of 2.6 g cm^{-3} (Tegen and Fung, 1994) and were encountered at pressures below 800hPa and at low RHs, the cut size will be reduced by about 40% (Collins et al., 2000). The air was then heated to maintain a RH below 40% (below 30% for 93% of the time during the profiles for the first flight and 77% for the second; see Table 1 for examples of RH during the investigated days). Light scattering measurements are dependent upon the RH at which they are made, since scattering increases as a function of increasing RH. However if the particles are maintained at RHs of 10–40% this variation is minimised (see, e.g., Anderson and Ogren, 1998), so even if the particles are not chemically dry at or below 30% they can be considered dry from a scattering point of view. This means that the measurements we made of absorption and scattering coefficients are of dry aerosol particles and that all the derived aerosol optical properties are for dry aerosol particles when not indicated otherwise.

Table 1. The average dry aerosol single scattering albedo (ω_0) during the horizontal runs in each of the two flights TF15 and TF20; SD is the standard deviation of ω_0 , RH is the relative humidity including its standard deviation and the altitude at which each of the runs were flown

Run no.	TF15 — 8 July 1997				TF20 — 17 July 1997			
	ω_0 (dry)	SD	RH (%)	altitude (m)	ω_0 (dry)	SD	RH (%)	altitude (m)
1	0.732	0.12	4.3 ± 0.14	3881	0.869	0.37	7.3 ± 0.31	3885
2	0.806	0.11	37.4 ± 0.71	40	0.961	0.19	41.1 ± 0.90	30
3	0.751	0.43	15.0 ± 1.9	980	0.805	0.17	32.5 ± 0.90	626
4					0.866	0.64	4.4 ± 0.15	1586
5					0.959	1.02	7.4 ± 0.10	1918
6					0.988	0.66	6.7 ± 0.26	1968
7					0.910	0.11	8.2 ± 0.23	3250

The combination of aerosol total scattering ($\sigma_{\text{sp}}(\lambda)$), hemispheric backscattering ($\sigma_{\text{bsp}}(\lambda)$), and absorption ($\sigma_{\text{ap}}(\lambda)$) coefficient measurements allows determination of key parameters needed to evaluate the direct forcing of climate by aerosols, including the aerosol light extinction coefficient ($\sigma_{\text{e}} = \sigma_{\text{sp}} + \sigma_{\text{ap}}$), the single scattering albedo ($\omega_0 = \sigma_{\text{sp}}/(\sigma_{\text{sp}} + \sigma_{\text{ap}})$) and the hemispheric backscatter fraction ($b = \sigma_{\text{bsp}}/\sigma_{\text{sp}}$).

The optical properties of aerosol particles can be divided into 2 groups (Ogren, 1995): those that depend on aerosol concentration (extensive properties) and those that are independent of concentration (intensive properties). The extensive parameters discussed in this paper are: $\sigma_{\text{sp}}(\lambda)$, $\sigma_{\text{ap}}(\lambda)$, $\sigma_{\text{e}}(\lambda)$, $\sigma_{\text{bsp}}(\lambda)$ and aerosol optical depth (δ), defined as the vertical column integration of the extinction coefficient $\sigma_{\text{e}}(\lambda)$. We have used the approximate equation $\delta = \Sigma \sigma_{\text{sp}} \times \Delta z$ with the summation carried out from the bottom to the top of the layer of interest. The intensive state properties to be discussed are: the hemispheric backscatter fraction ($b(\lambda)$), the aerosol single scattering albedo ($\omega_0(\lambda)$) and the Ångström exponent ($\tilde{a}(\lambda_1/\lambda_2)$) defined as

$$\tilde{a}(\lambda_1/\lambda_2) = -\log[\sigma_{\text{sp}}(\lambda_1)/\sigma_{\text{sp}}(\lambda_2)]/\log[\lambda_1/\lambda_2],$$

where λ_1/λ_2 denote the measurement wavelength of 450/550, 450/700, or 550/700 nm.

3. Case studies

2 of the 21 *Pelican* flights during ACE-2 have been chosen as case studies of Saharan mineral dust layers. The flights TF15 and TF20 on 8 and 17 July 1997 provided descent profiles in cloud free airmasses with similar vertical structure. Both had 3 distinctly different layers in the vertical: a mineral dust layer at high altitudes, a polluted MBL separated by a very clean layer. These 2 flights provide a survey of vertical and horizontal variability of the optical properties of the aerosols that dominate the direct aerosol radiative forcing of climate, especially during Saharan mineral dust events with a polluted MBL.

Fig. 1 gives details of the location of the 2 flights. The profiles we will discuss in this paper were centred around 15°58'W, 27°51'N on TF15 and 16°33'W, 27°48'N on TF20 (indicated with P15 and P20 in Fig. 1) and were between altitudes

of 3900 and 30 m above sea level (ASL). The profile flown on 8 July took place between 1018 and 1100 UTC, while the profile on 17 July was between 1525 and 1606 UTC.

3.1. TF15 — 8 July 1997

3.1.1. Meteorology. At 1200 UTC on 8 July there was a surface low (1015 hPa central pressure) over Spain and a surface high (1026 hPa in the centre) over the Azores. The satellite image at 1533 UTC from the NOAA-14 AVHRR shows that there was an extensive sheet of stratocumulus clouds to the north and west of Tenerife. The airflow was from the north-west in the MBL and there were no clouds in the area to the south of Tenerife where the *Pelican* was flying.

The 5-day back trajectories for this day from the Royal Dutch Meteorological Institute (KNMI; Scheele et al., 1996) ending at 1200 UTC on Tenerife estimate that the air in the lowest part of the MBL originated over Spain and the British Isles where it had been exposed to pollution sources. The clean air between the dust layer and the MBL had its origin over the Atlantic to the west of Tenerife and had subsided in the free troposphere for several days. The air at the level of the dust layer had its origin over Libya and has spent several days over North Africa enroute to Tenerife.

The balloon sounding from the measuring site Punta del Hidalgo (marked with an X in Fig. 1) at 0600 UTC indicated an inversion height of 1025 m. A slightly stable layer at 600 m was indicative of the surface layer being de-coupled from the top of the MBL. Between 720 and 520 hPa there was a well mixed dry layer capped by a temperature inversion. At these altitudes it is a clear indication of a dust layer, where the “dust top” cooling effect and the warming by absorption within the layer creates a well mixed layer. The wind speed showed maxima at 880 m of 8.6 m s⁻¹ from the NE and at 4580 m (16.1 m s⁻¹, SSW). There was a minimum at 1875 m (3.4 m s⁻¹, SSE).

3.1.2. Aerosol optical properties

3.1.2.1. Vertical profile of scattering. The optical characteristics of the aerosol particles encountered during this day varied considerably with height as shown in Fig. 2. There was a clear 3-layer

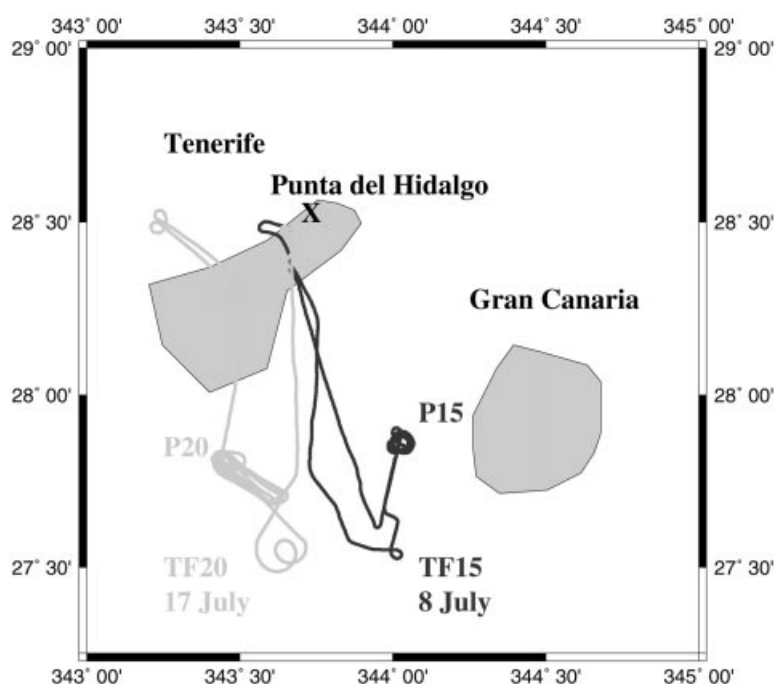


Fig. 1. A map showing Tenerife and the area where flights TF15 and TF20 were flown on 8 and 17 July. The locations of the profiles are indicated with P15 and P20. The X marks the measuring site at Punta del Hidalgo, where the balloon soundings were made.

structure. A MBL extended to 1000 m with elevated dry aerosol scattering levels (the mean scattering for the green wavelength (all scattering values are for 550 nm if not stated otherwise and are denoted $\sigma_{sp,g}$) was $2.37 \times 10^{-5} \text{ m}^{-1}$). Above the MBL there was a clean layer between 1000 and 2720 m with virtually no aerosol scattering (the mean $\sigma_{sp,g}$ was $7 \times 10^{-7} \text{ m}^{-1}$) and a dust layer above 2720 m with scattering levels between the MBL and clean layer (mean $\sigma_{sp,g}$ was $9.81 \times 10^{-6} \text{ m}^{-1}$).

The magnitude of the $\sigma_{sp,g}$ was about $3.4 \times 10^{-5} \text{ m}^{-1}$ in the lowest part of the MBL. The scattering decreased by more than a factor of 3 with altitude up to a layer with virtually no aerosol scattering (effectively the noise level of the nephelometer) at 980 m (900 hPa). This layer extended to about 2720 m (740 hPa) where the scattering increased again. The MBL aerosol scattering coefficients decreased with increasing wavelength. This is as expected for pollution aerosols where the major fraction of particle number is in the same size region or smaller than the wavelengths used in the measurements.

The dust layer started at 2720 m (735 hPa) and extended above the *Pelican*'s ceiling of 3.9 km. The 0600 UTC Punta del Hidalgo sounding indicates that it extended to 5.2 km. At the lower edge of the dust layer the scattering increased from $0.15 \times 10^{-5} \text{ m}^{-1}$ to about $1.25 \times 10^{-5} \text{ m}^{-1}$. The scattering coefficients within the interior of the dust layer were fairly constant with altitude, varying between $0.9 \times 10^{-5} \text{ m}^{-1}$ and $1.25 \times 10^{-5} \text{ m}^{-1}$. There is little difference between the scattering coefficients in the different wavelengths and this behaviour is what would be expected if a significant fraction of the particles were larger than the wavelength used and the particle size distribution was wider than the wavelength difference, keeping in mind that only the small particle range of the dust aerosols is measured due to the cut size of the cyclone.

3.1.2.2. Horizontal variability in scattering coefficients. The diamonds in Fig. 2 show the mean values of the scattering coefficients measured during horizontal runs flown adjacent to the profile. The run at 40 m (1000 hPa) ASL was flown

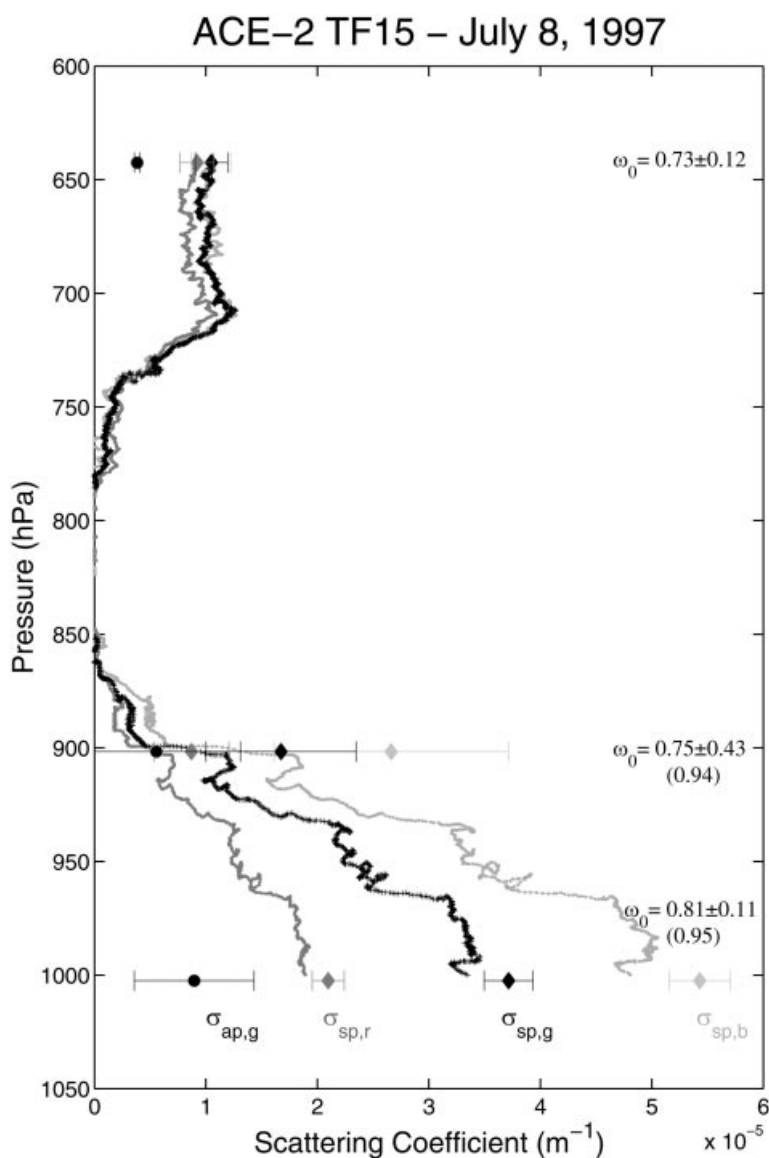


Fig. 2. The scattering coefficients at 450, 550 and 700 nm ($\sigma_{sp,b}$, $\sigma_{sp,g}$ and $\sigma_{sp,r}$, respectively) for the flight on 8 July are plotted as a function of pressure. The filled diamonds indicate the averages of scattering coefficients during the horizontal runs, while the solid circles indicate the absorption coefficient at 565 nm ($\sigma_{ap,g}$) during the same runs. The variability bars indicate one standard deviation from the measurement. The dry single scattering albedos ω_0 and their standard deviation for each of the runs are given together with the wet single scattering albedo (in brackets) calculated for the MBL assuming a growth factor of 2.2.

immediately after the vertical profile and the average $\sigma_{sp,g}$ was $3.7 \times 10^{-5} \text{ m}^{-1}$. This is higher than the $\sigma_{sp,g}$ measured at the same altitude in the profile. The variability in the horizontal runs can

be estimated by the standard deviation (SD) of the measurements ($0.22 \times 10^{-5} \text{ m}^{-1}$) and is shown as "error bars" in the figure. In the 980 m (900 hPa) run the $\sigma_{sp,g}$ was $1.7 \times 10^{-5} \text{ m}^{-1}$ with a variability

of $0.14 \times 10^{-5} \text{ m}^{-1}$. This run was flown after the profile in an area south of the profile and the other 2 horizontal runs. The MBL top often sloped during the experiment which may explain why the average σ_{sp} in this horizontal run was higher than the value from the profile at the same pressure level. The average scattering coefficients within the horizontal run in the dust layer at 3880 m (640 hPa) was $1.05 \times 10^{-5} \pm 1.4 \times 10^{-6} \text{ m}^{-1}$ for the green, $1.04 \times 10^{-5} \pm 1.7 \times 10^{-6} \text{ m}^{-1}$ and $0.92 \times 10^{-5} \pm 1.5 \times 10^{-6} \text{ m}^{-1}$ for blue and red wavelengths, respectively. The horizontal variability in the dust layer was less than that observed in the MBL and the difference between the mean values in the run and the profile were negligible.

3.1.2.3. Aerosol absorption. The solid circles in Fig. 2 are the absorption coefficient measurements during these runs. Again, the variability bars reflect 1 SD. The absorption in the MBL at 40 m was $8.9 \times 10^{-6} \text{ m}^{-1}$, at 980 m (900 hPa) it was $5.6 \times 10^{-6} \text{ m}^{-1}$ and at 3880 m (640 hPa) it was $3.8 \times 10^{-6} \text{ m}^{-1}$. The absorption by particles showed the same trend with altitude as the scattering by particles during this flight and the absorption coefficient in the dust layer was between 43% and 67% of that in the polluted MBL.

3.1.2.4. Aerosol single scattering albedo. Using the measured dry aerosol scattering and absorption coefficients in the green wavelengths (550 nm for scattering and 565 nm for absorption), the single scattering albedo for dry aerosol particles (smaller than the impactor nominal cut size of $2.5 \mu\text{m}$ diameter) can be calculated. The values are given in Fig. 1 and in Table 1 for each of the horizontal runs, together with the SDs. The values for the dried aerosol particles in the MBL were 0.81 ± 0.11 at 40 m and 0.75 ± 0.43 at the transition level. These values are within the previous measured values for polluted air masses (Japar et al., 1986). The ω_0 was 0.73 ± 0.12 in the dust layer.

Normally, MBL aerosol particles are hygroscopic and grow to larger sizes at high ambient RHs, which increases their scattering coefficients (Charlson et al., 1984; Hegg et al., 1993, and Covert et al., 1972). This increase will affect the single scattering albedo so that the ω_0 for dry aerosol particles we report here will actually be the lower limit for the ambient ω_0 . In order to obtain an upper limit, we need to estimate the

effect of RH on ω_0 . We have chosen to do this by using the relationship given by Hänel (1976) and Charlson et al. (1978):

$$\sigma_{\text{sp}}(\text{RH})/\sigma_{\text{sp}}(\text{dry}) = [D(\text{RH})/D(\text{dry})]^2,$$

where $D(\text{RH})/D(\text{dry})$ is the growth factor (GF) of the aerosol particles (e.g., how much their size changes between 30% and 80% RH). In order to use this relationship we need to know the growth factor of the aerosols, and we assume that the absorption component of extinction does not change with RH.

Swietlicki et al. (2000) measured GFs of 1.5 to 1.7 at a ground based site on Tenerife. Gassó et al. (2000) derived an aerosol hygroscopic component γ from measurements onboard the *Pelican*. Using their relation (from Kasten, 1969), we derive GFs of between 1.4 and 1.6 for the conditions we report here. Schmeling et al. (2000) used measured chemical composition and size distributions from the *Pelican* combined with a relationship from Howell and Huebert (1998) to calculate GFs for *Pelican* data. They derive values of between 1.4 and 1.9. Previous observations on the east coast of the North Atlantic give GFs (as defined here) of between 1.2 and 1.7 (Kotchenruther et al., 1999). Similar GF values were also observed during ACE-1 by Berg et al. (1998).

We use GFs of 1.6, 1.7 and 2.2 to calculate the effect of increased scattering on ω_0 . The highest GF value comes from measurements on sea salt and pure NaHSO_4 particles (Tang, 1996). It could be considered representative for clean MBLs not influenced by pollution. We use this value (which leads to an $\sigma_{\text{sp}}(80\%)/\sigma_{\text{sp}}(30\%)$ ratio of 4.8) to put an upper bound on the effect of relative humidity on the scattering coefficients.

With these GFs the ω_0 for the lowest horizontal run (at 40 m) increased from the dry value of 0.806 to 0.914 with a GF of 1.6, to 0.923 for a GF of 1.7, and to 0.953 for a GF of 2.2. The wet aerosol ω_0 in the MBL are given in Table 2.

The Punta del Hidalgo sounding indicated that the RH in the dust layer was lower than 20% below 4000 m. It also showed that the RH increased with height from about 8% at 2720 m to 18% at 4000 m. Because of the low RH we have chosen not to calculate the ω_0 of wet dust particles even though measurements onboard the *Pelican* suggest a GF (between 30% and 80%

Table 2. Dry single scattering albedo and the estimate of wet single scattering albedo in the MBL during TF15 and TF20

TF15 — 8 July 1997					TF20 — 17 July 1997				
ω_0 (dry)	ω_0 (GF = 1.6)	ω_0 (GF = 1.7)	ω_0 (GF = 2.2)	altitude (m)	ω_0 (dry)	ω_0 (GF = 1.6)	ω_0 (GF = 1.7)	ω_0 (GF = 2.2)	altitude (m)
0.806	0.914	0.923	0.953	40	0.961	0.985	0.986	0.992	30
0.751	0.885	0.897	0.936	980	0.805	0.913	0.922	0.952	625

We have assumed growth factors of 1.6 and 1.7 and 2.2 for 90% RH. The values given are for the two lowest runs for each flight. Also given is the altitude for the horizontal runs.

RH) of about 1.1–1.2 in the dust layer (Gassó et al., 2000).

3.1.2.5. Contribution of aerosol scattering to total atmospheric scattering. The contribution of the aerosol particles to atmospheric scattering can be visualised by plotting the scattering by particles normalised with the Rayleigh scattering of air (σ_{rg}), and is shown in Fig. 3. The left panel shows the ratio between the total scattering by particles and Rayleigh scattering by gases for the profile and the horizontal runs (diamonds). The ratio follows the vertical structure of the scattering coefficient. In the MBL the ratio varied between 1.0 and 3.0. It was effectively zero in the intermediate clean layer and varied between 1.2 and 1.56 in the dust layer.

3.1.2.6. Hemispheric backscattering fraction. The right panel in Fig. 3 shows the backscatter to total scattering ratio (also called the hemispheric backscatter fraction (b)) and is only shown for the part of the profile where the scattering coefficient in the green wavelength (550 nm) was larger than $5 \times 10^{-6} \text{ m}^{-1}$. The hemispheric backscatter fraction was slightly higher in the MBL than in the dust. The mean value in the MBL was 0.17 with an SD of 0.03 in the green wavelength. There was no clear trend with altitude within the MBL even though the scattering coefficient decreased by a factor of 3 from close to the sea surface to the top of the MBL. The hemispheric backscatter fraction was more variable in the dust layer where the mean value was 0.082 and the SD was 0.023. The averages for the green wavelengths for the 2 layers together with variability bars denoting the width of the 99.9% confidence interval are shown in Fig. 3 in between the 2 profiles. The layers were statistically significantly different from each other, although the magnitude of the difference was

small. The mean values of hemispheric backscatter fraction in each of the 2 layers, together with the SDs and sample size and the confidence intervals for 99.9% level are given in Table 3.

3.1.2.7. The Ångström exponent. The profile of the Ångström exponent for the blue, green and red wavelengths ($\hat{a}_{(450/550)}$, $\hat{a}_{(450/700)}$, and $\hat{a}_{(550/700)}$) are shown in Fig. 4. As in Fig. 3, only the values for $\sigma_{sp,g}$ larger than 5×10^{-6} are shown. The $\hat{a}_{(550/700)}$ values varied between 1.71 and 2.41 in the MBL, while $\hat{a}_{(550/700)}$ varied from 1.76 to 2.3. The values for $\hat{a}_{(550/700)}$ show a wider range with values between 1.51 and 2.63. The largest change in all 3 Ångström exponents can be found in the transition region into the clean layer above the MBL. Except for this region they are quite constant with height within the MBL. The $\hat{a}_{(450/550)}$ values in the dust layer varied from -0.86 to 0.79 , while $\hat{a}_{(450/700)}$ and $\hat{a}_{(550/700)}$ varied between -0.33 to 0.78 and 0.096 to 1.17 , respectively. The variation within the dust layer for all 3 wavelengths was higher than in the MBL. The blue and green Ångström exponent in the dust layer had values reaching into the negative and the red is close to zero at its lower range. The differences between the 2 layers is likely due to differences in the aerosol particle size distribution with the larger values of $\hat{a}$ corresponding to cases with smaller particles sizes. That would mean that the dust layer had larger particles (among the sampled aerosols) than the MBL, which is confirmed by the simultaneously measured size distributions (Collins et al., 2000).

3.1.2.8. Dry and wet aerosol optical depth. Table 4 shows the calculated dry aerosol optical depth for the MBL (between 40 m and 1000 m), the dust layer (between 2720 m and 3890 m) and the whole column (integrated over the whole

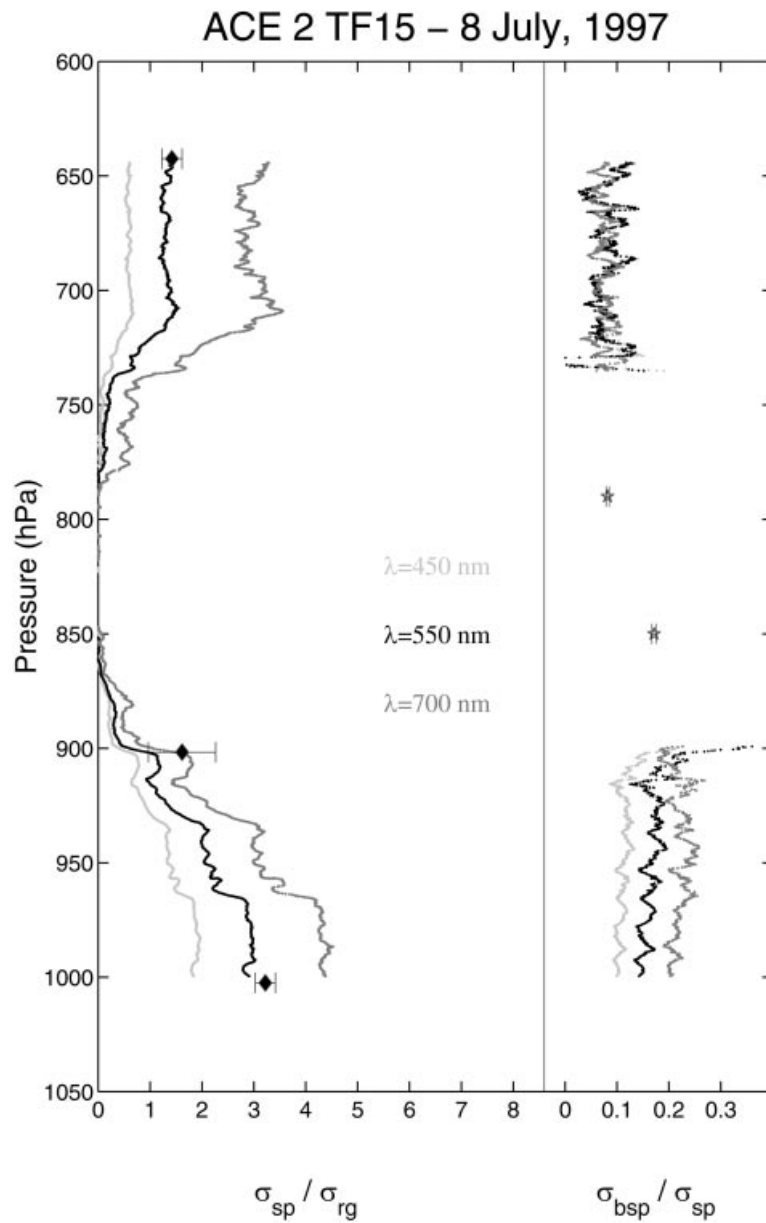


Fig. 3. The left panel shows the ratio of scattering by particles for 450, 550 and 700 nm to the Rayleigh scattering of gases at the same wavelengths as a function of pressure for the flight on 8 July. This ratio is a measure of the relative contribution of aerosols to the total atmospheric scattering. The right panel shows the hemispheric backscatter fraction for $\sigma_{\text{sp},g} > 0.5 \times 10^{-5} \text{ m}^{-1}$. The stars indicate the averages within each layer and the “error bars” indicate the significance level of 99.9% CI.

Table 3. Summary of the dry aerosol hemispheric backscatter fractions in the MBL and in the dust layer for flights TF15 (8 July) and TF20 (17 July)

Layer	TF15 — 8 July 1997						TF20 — 17 July 1997					
	$\sigma_{\text{bsp}}/\sigma_{\text{sp}}$ (450 nm)		$\sigma_{\text{bsp}}/\sigma_{\text{sp}}$ (550 nm)		$\sigma_{\text{bsp}}/\sigma_{\text{sp}}$ (700 nm)		$\sigma_{\text{bsp}}/\sigma_{\text{sp}}$ (450 nm)		$\sigma_{\text{bsp}}/\sigma_{\text{sp}}$ (550 nm)		$\sigma_{\text{bsp}}/\sigma_{\text{sp}}$ (700 nm)	
	mean	SD	mean	SD	mean	SD	mean	SD	mean	SD	mean	SD
MBL	0.116	0.019	0.171	0.03	0.222	0.018	0.107	0.021	0.150	0.030	0.193	0.024
dust	0.082	0.027	0.082	0.027	0.076	0.016	0.087	0.014	0.092	0.014	0.108	0.013
MBL 99.9% CI	0.0025		0.0040		0.0024		0.0035		0.0022		0.0024	
dust 99.9% CI	0.0032		0.0031		0.0018		0.0021		0.0010		0.0017	

The sample size was 593 in all three wavelengths for the MBL and 794 for blue and green wavelengths in the dust layer and 809 in the red wavelength during TF15. For TF20 the sample size was 647 for the MBL and 1118 for the dust layer.

Table 4. Summary of the dry and wet aerosol optical depths as estimated from a growth factor of 1.6, 1.7 and 2.2 at a relative humidity of 90% in the MBL and no growth factor in the dust layers

Layer	TF15 — 8 July 1997				TF20 — 17 July 1997			
	δ (450 nm)	δ (550 nm)	δ (700 nm)	altitudes (m)	δ (450 nm)	δ (550 nm)	δ (700 nm)	altitudes (m)
MBL (dry)	0.033	0.022	0.012	40–1000	0.021	0.015	0.009	45–1020
MBL GF = 1.6	0.083	0.056	0.032	40–1000	0.054	0.038	0.022	45–1020
MBL GF = 1.7	0.094	0.064	0.036	40–1000	0.061	0.043	0.025	45–1020
MBL GF = 2.2	0.157	0.106	0.060	40–1000	0.102	0.072	0.042	45–1020
dust	0.011	0.011	0.010	2720–3890	0.044	0.041	0.032	1925–3890
MBL + dust (dry)	0.046	0.035	0.024	40–3890	0.066	0.057	0.041	45–3890
MBL + dust; GF = 1.6	0.096	0.069	0.043	40–3890	0.099	0.080	0.055	45–3890
MBL + dust; GF = 1.7	0.107	0.076	0.047	40–3890	0.106	0.085	0.058	45–3890
MBL + dust; GF = 2.2	0.171	0.119	0.071	40–3890	0.147	0.114	0.075	45–3890

The optical depths for the whole profile (MBL + dust) are calculated from a MBL with a growth factor of 1.6, 1.7 and 2.2 and no growth of the particles in the rest of the profile.

profile from 40 m to 3890 m) by using the dry scattering coefficients in the 3 different wavelengths. Since the scattering coefficients are for dry particles the calculations using these values are lower limits of the optical depth. The optical depth for the MBL was 0.022 at 550 nm, which is 2 times higher than the optical depth of 0.011 for the dust layer and would be even larger if the MBL optical depth was calculated for particles at ambient RH rather than dry particles. The optical depth decreases rapidly with increasing wavelength if the layer contains primarily submicrometer particles. We clearly see this for the MBL

while the optical depth for the dust layer was between 0.010 and 0.011 for the 3 different wavelengths. This is another sign that the aerosols were larger in the dust layer.

What is more important for radiative transfer comparisons with, e.g., satellite retrievals is the aerosol optical depths at ambient RH. One way of estimating the upper limit of aerosol optical depths for the flights reported here is to calculate the aerosol optical depth for wet particles. These have been calculated and shown in Table 4 assuming the same GFs for the MBL (1.6, 1.7 and 2.2). Optical depth for the entire column when the RH

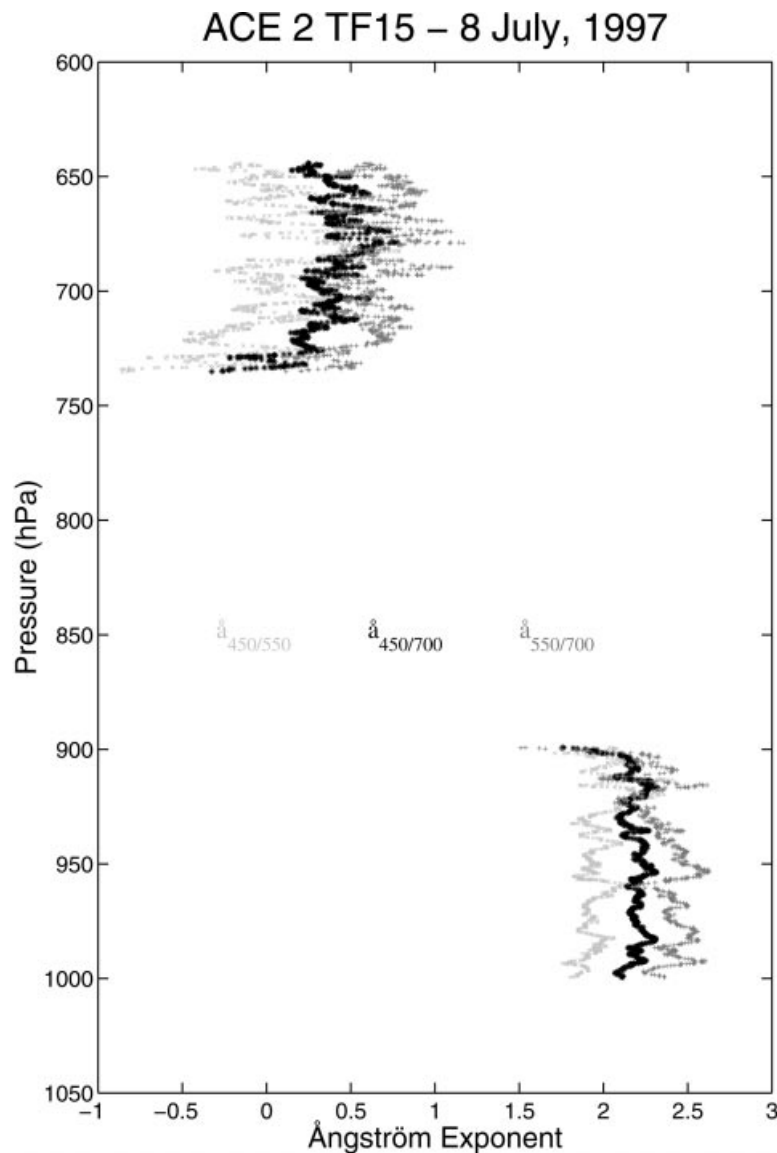


Fig. 4. The Ångström exponents for 8 July are plotted as a function of pressure for $\sigma_{sp,g} > 0.5 \times 10^{-5} \text{ m}^{-1}$.

effects in the MBL are considered increases by 97% for a MBL GF of 1.6 and by a factor of 2.2 for a GF of 1.7.

3.2. TF20 — 17 July 1997

3.2.1. Meteorology. At 1200 UTC on 17 July 1997, there was an extensive high pressure (1025 hPa) system to the south and east of the

Azores and a low over the African continent. That caused the air to flow out over the Iberian Peninsula from Europe into the ACE-2 area at 850 hPa. The satellite images from a NOAA-14 overpass at 1535 UTC showed an extensive sheet of stratocumulus clouds to the north of Tenerife. It was mostly clear to the south of the Canary Islands.

The KNMI 5-day back trajectories ending at Tenerife at 1200 UTC indicate a marine source

for the air at 975 hPa. The 850 hPa trajectory started over the Iberian Peninsula at low levels and ascended up to 700 hPa where it started to descend $2\frac{1}{2}$ days prior to arrival. The 770 hPa trajectory passed over the Portuguese coast at high altitudes before reaching Tenerife. The trajectory ending at 500 hPa had originated at 395 hPa over Senegal in Africa. The Punta del Hidalgo sounding at 1200 UTC identified 2 inversions, one at 890 hPa (1120 m) and a weaker one at 800 hPa. There was no clear adiabatic layer aloft during this profile which suggests that the sounding may have missed the dust plume. There were wind speed maxima at 300 m of 7.6 m s^{-1} from the ENE and at 3230 m (16.2 m s^{-1} , WSW) and a minimum at 1586 m (1.3 m s^{-1} , S).

3.2.2. Aerosol optical properties

3.2.2.1. Vertical profile of scattering. The optical characteristics of the aerosol particles again varied with height, but showed a different behaviour compared with flight TF15. As can be seen from the profile of aerosol scattering shown in Fig. 5 the MBL extended from the sea surface up to about 900 m (910 hPa). The MBL during this day contained mainly maritime air at low levels, more polluted air at higher levels interspersed with a band of local pollution (Collins et al., 2000). The scattering coefficient at 550 nm decreased with altitude from $2.1 \times 10^{-5} \text{ m}^{-1}$ at the sea surface to about $1.5 \times 10^{-5} \text{ m}^{-1}$ at 800 m, then increased to $1.7 \times 10^{-5} \text{ m}^{-1}$ at 900 m. The mean $\sigma_{\text{sp,g}}$ in the MBL was $1.67 \times 10^{-5} \text{ m}^{-1}$. The layer immediately above the MBL was clean with low aerosol scattering (mean $\sigma_{\text{sp,g}}$ of $0.1 \times 10^{-9} \text{ m}^{-1}$) and started at about 1050 m (890 hPa) extending up to 1900 m (815 hPa). A high scattering dust layer ($\sigma_{\text{sp,g}}$ was generally higher than $2 \times 10^{-5} \text{ m}^{-1}$ and the mean $\sigma_{\text{sp,g}}$ was $2.09 \times 10^{-5} \text{ m}^{-1}$) started at 1925 m (805 hPa) and was estimated (from lidar measurements at Izaña; Welton et al., 2000) to have its top at 4.5 km. Even though the ceiling of the *Pelican* is 3.9 km, the top of the profile was thought to be close to the top of the dust layer, since the Island tends to force layer structures upward relative to open ocean locations (Welton et al., 2000). The dust layer had higher scattering coefficients than the MBL. The scattering by blue and green light were close to each other in the dust layer (the mean $\sigma_{\text{sp,b}}$ was $2.49 \times 10^{-5} \text{ m}^{-1}$,

the mean $\sigma_{\text{sp,g}}$ was $2.09 \times 10^{-5} \text{ m}^{-1}$) while the red scattering coefficient was lower (mean $\sigma_{\text{sp,r}}$ was $1.83 \times 10^{-5} \text{ m}^{-1}$). The $\sigma_{\text{sp,g}}$ varied from $0.47 \times 10^{-5} \text{ m}^{-1}$ just underneath the dust layer to a maximum of $3.12 \times 10^{-5} \text{ m}^{-1}$ at 3500 m (670 hPa).

3.2.2.2. Variation in scattering measured during horizontal runs. Fig. 5 also shows the average scattering coefficients (diamonds) measured in 7 race track patterns in the vicinity of the profile and extending to the south east. The variability bars show 1 SD of the measurements and give an estimate of the horizontal variation. The scattering coefficient at 30 m (1000 hPa) was 2.41×10^{-5} with an SD of $0.29 \times 10^{-5} \text{ m}^{-1}$. All the average values during the horizontal runs were higher than the values at the same level in the profile, but they all follow the general scattering profile, except in the clean layer just below the dust layer. Here the average scattering coefficients in the horizontal runs showed a significantly higher value than in the profile. This was probably due to variation in the vertical position of the layer with higher scattering that can be seen just above 800 hPa. The lower boundary of this layer may have been sloping so that it was higher at the location of the profile and lower at the location of the runs.

3.2.2.3. Aerosol absorption. The absorption coefficients for the runs are shown in Fig. 5 as circles and they were higher at 630 m than 30 m. The higher absorption at 630 m could be due to influences from sources on Tenerife that were detected in the fine particle aerosol number concentration (Collins et al., 2000) measured at the same time by the *Pelican*. The absorption was factor of 2.8 to 2.9 higher in the dust layer than at 30 m in the MBL. The variability was lower in the dust layer than in the rest of the profile. Compared to the highly absorbing layer in the MBL caused by local sources on Tenerife the absorption in the dust is a factor of 0.65 to 0.7 smaller. This is still a high absorption for the dust and shows that it has the potential to be important for radiative transfer calculations in dust.

3.2.2.4. Aerosol single scattering albedo. Also shown in Fig. 5 (and given in Table 1) are the calculated dry aerosol single scattering albedos for the race tracks. The values (at all levels) were higher than during TF15. The highest values were found at 30 m in the MBL and at 1920–1970 m in the very clean layer just below the dust layer. These had dry ω_0 in the range 0.96–0.99. The lowest ω_0 was at

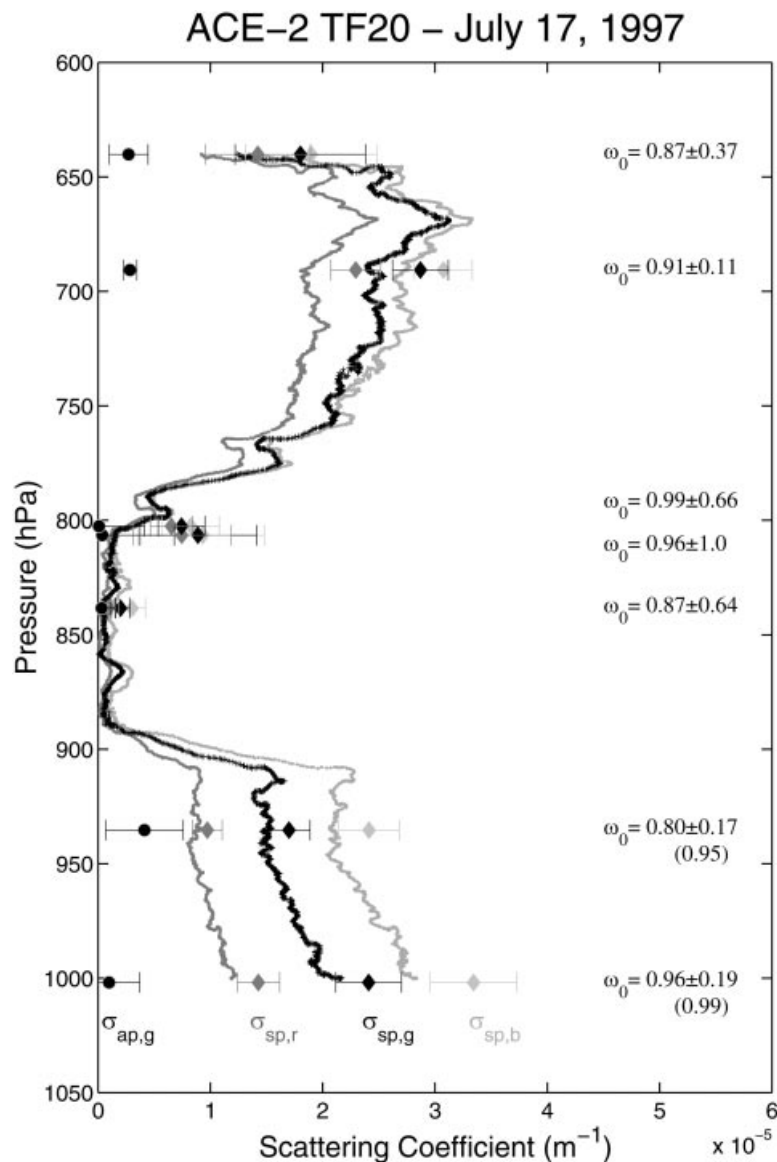


Fig. 5. Same as Fig. 2 but for 17 July.

630 m in the locally influenced part of the MBL ($\omega_0 = 0.80$). The dust layer itself had a dry single scattering albedo of 0.91 ± 0.11 at 3250 m (690 hPa). The horizontal variability in the dry aerosol ω_0 was lowest again in the dust layer.

The wet aerosol single scattering albedo (Table 2) calculated assuming the same growth factors, increases from the dry value of 0.961 to

0.985 for $\text{GF} = 1.6$, 0.986 ($\text{GF} = 1.7$) and reaches the value 0.992 for a GF of 2.2 at 30 m. The same assumption about aerosol growth in the dust layer can be made as for TF15, i.e., that the dust particles will not grow due to RH, since it during this day was also below 25%.

3.2.2.5. Contribution of aerosol scattering to total atmospheric scattering. The contribution by

aerosols to the total scattering is shown in the left panel of Fig. 6. The diamonds are the averages for the 7 different race tracks with the variability bars being 1 SD. In the lowest part of the MBL the aerosol scattering to Rayleigh scattering ratio was close to 1.8 and decreased with height to 1.3 at

750 m (920 hPa). There was a slight increase again up to the transition to the clean layer. The value of the ratio was always lower than 0.2 in the clean layer, and in the dust layer it varied between 0.47 and 4.1 with a maximum at 3500 m (670 hPa). We can see that the contribution of aerosol scattering

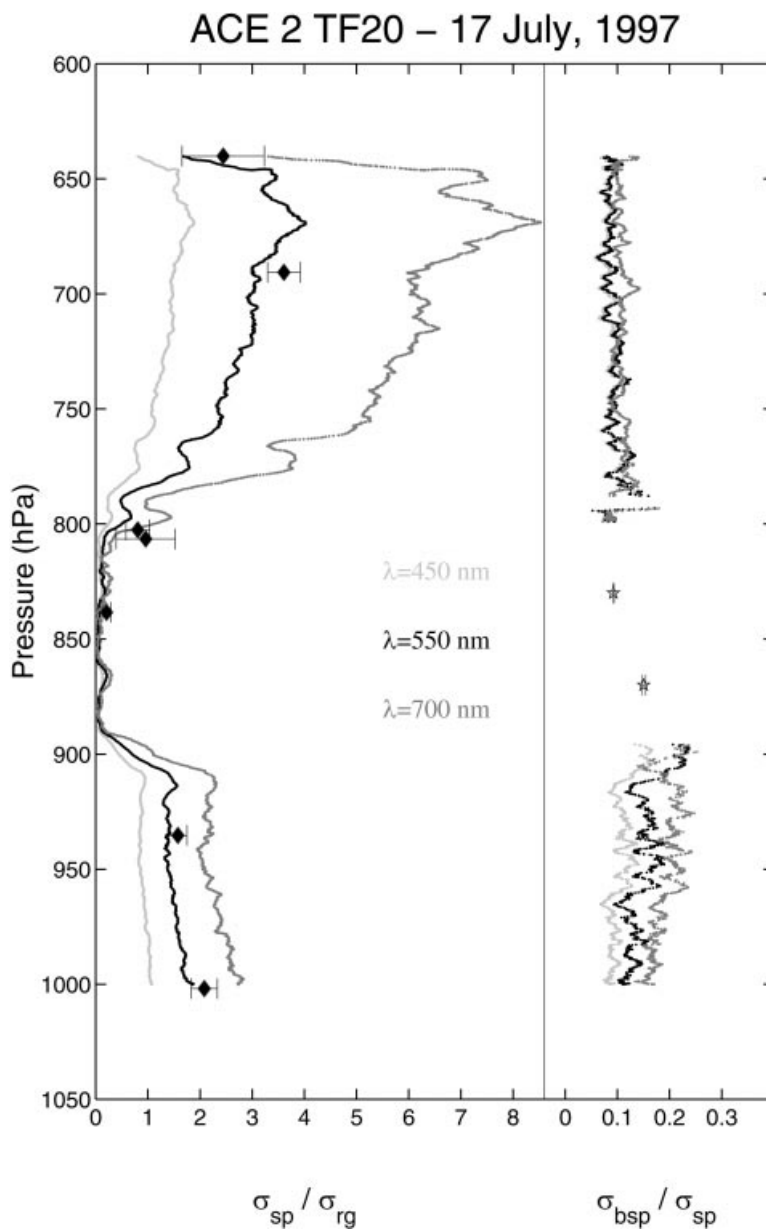


Fig. 6. Same as Fig. 3 but for 17 July.

to total atmospheric scattering was higher in most parts of the dust layer than in the MBL by a factor of 2 at times. The horizontal variation in the ratio is highest for the race track at 3885 m (640 hPa), and was similar in magnitude to the variation in the vertical within the dust layer.

3.2.2.6. Hemispheric backscattering fraction. The right panel in Fig. 6 shows the hemispheric backscatter fraction for the parts of the profile where $\sigma_{\text{sp,g}}$ was larger than 5×10^{-6} . The average values in the dust and MBL together with the confidence intervals for 99.9% significance level are shown in the figure between the 2 profiles and are presented in Table 3. As in TF15 there is a statistically significant difference between the hemispheric backscatter fraction in the dust layer and the MBL during this flight, although the magnitude of the difference was small. The hemispheric backscatter fraction in the MBL was larger than for the dust layer. The average in the MBL was 0.15 with an SD of 0.03 and there was a slight increase with height from 0.1 at 50 m (1000 hPa) to 0.24 at 800 m (900 hPa). In the dust layer the average was 0.092 with an SD of 0.014.

3.2.2.7. Ångström exponent. The Ångström exponent for the profile is plotted in Fig. 7 where $\sigma_{\text{sp,g}}$ was larger than $5 \times 10^{-6} \text{ m}^{-1}$. There was a clear difference between the MBL and the dust layer. In the MBL the $\hat{a}_{(450/550)}$ varied between 1.3 to 2.1, the $\hat{a}_{(550/700)}$ between 1.65 and 2.55, and the $\hat{a}_{(550/700)}$ between 1.52 and 2.6, while the range in the dust layer for $\hat{a}_{(450/550)}$ was between -0.02 and 0.72 . For $\hat{a}_{(550/700)}$ and $\hat{a}_{(550/700)}$ the variation was between 0.38 to 0.95 and 0.60 and 1.4, respectively. The spread in the data within each layer was similar. The differences in $\hat{a}$ between the dust layer and the MBL indicate that the size distributions of the particles in the 2 layers were not the same. It is striking that, even though the scattering in the blue wavelength was not too different in the 2 layers, the $\hat{a}$ was much lower in the dust layer than in the MBL. It was even negative in the lowest part of the dust layer, i.e., the aerosol particles scattered more at longer wavelengths.

3.2.2.8. Dry and wet aerosol optical depth. Table 4 shows the calculated dry aerosol optical depths. In the MBL between 30 m and 1020 m for 550 nm it was 0.015 while in the dust layer between 1925 m and 3885 m it was 0.041. For the other 2 wavelengths the dry aerosol optical depths were 0.021 in the MBL for 450 nm and 0.009 for 700 nm.

In the dust layer the dry aerosol optical depths were 0.044 (450 nm) and 0.032 (700 nm).

The estimated wet aerosol optical depths are also given in Table 4. The optical depth at 550 nm for the whole vertical profile increased by 40.4% for a GF of 1.6 and 49.1% for a GF of 1.7. The wet optical depth in the green wavelength for the MBL is the same as for the dust layer assuming a GF of 1.65 for the MBL at 90% RH and no growth of the dust aerosol particles.

4. Discussion of differences and similarities

We have investigated the optical properties of aerosols during 2 cases with North African mineral dust above an anthropogenically influenced MBL, in both cases they were separated by a clean layer with low aerosol scattering coefficients. This has made it possible to compare the optical properties of mineral dust and pollution aerosols in the marine environment, and also the optical properties within similar layers on different days.

The MBL optical properties were highly dependent on the source region and history of the air mass and how effectively the mixing took place in the MBL. Both of the MBLs investigated had a height of about 1000 m and contained anthropogenic pollution aerosols. On 8 July a major pollution outbreak influenced the Tenerife area with high pollution levels. On 17 July local pollution sources were affecting the sampling area. The aerosol scattering coefficients in the MBL were higher on 8 July than 17 July and as a result the MBL dry aerosol scattering optical depth was also higher on 8 July (0.021) than on 17 July (0.018). The absorption coefficients and the hemispheric backscatter fraction were also higher in the MBL on 8 July. The highest variability in absorption and scattering was found in the transition region between the MBL and the clean layer above the MBL on 8 July. This could be due to a sloping MBL top. Apart from that specific run the horizontal variability in the scattering was often smaller or comparable to the variability in the vertical.

The dry single scattering albedo was lower in the more polluted MBL (0.75 and 0.81 on 8 July; 0.96 and 0.80 on 17 July), while the calculated wet aerosol single scattering albedo for the highest assumed GF showed less difference with height in

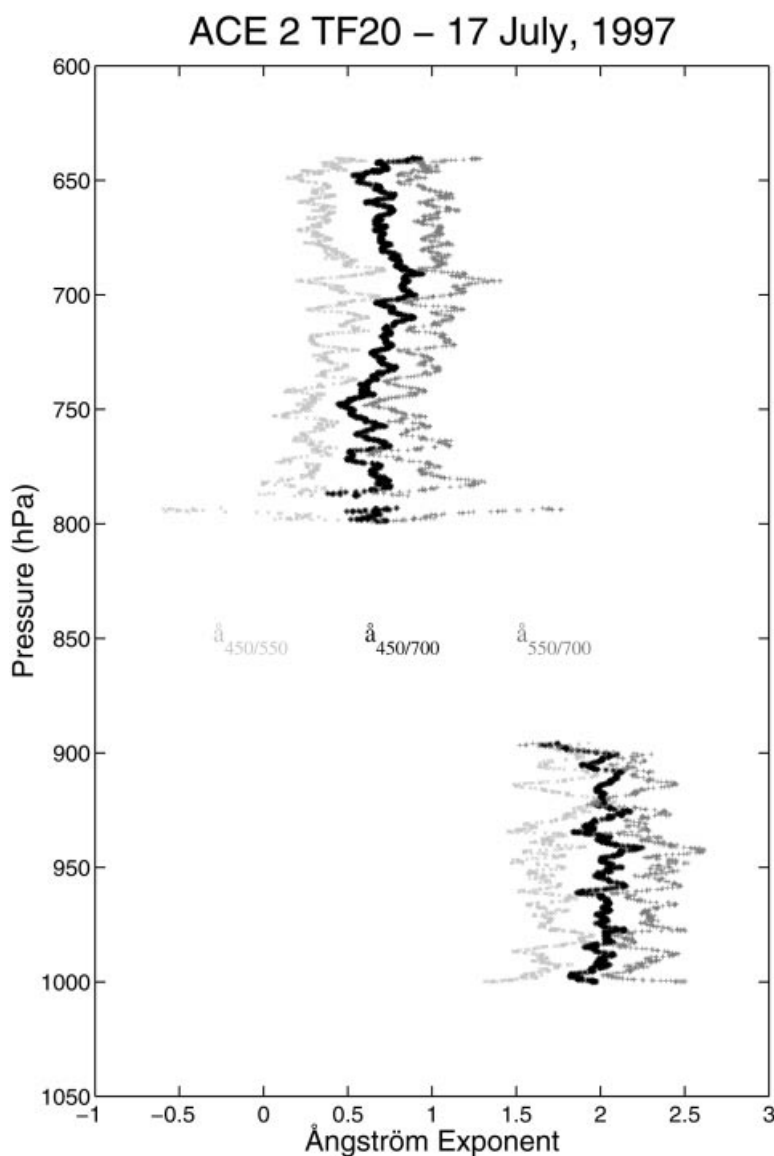


Fig. 7. Same as Fig. 4 but for 17 July.

the MBL within one and the same day, as well as less difference between the MBLs on the 2 days. To illustrate the difference in the dry aerosol single scattering albedo in the MBL during the 2 days it can be mentioned that to obtain the same ω_0 on 8 July to that found on 17 July the particle diameters would have to grow by more than a factor of 4.8.

The 2 dust cases encountered during ACE-2

have few similarities. On 8 July, the scattering and absorption coefficients showed little spatial variation, while on 17 July there was substantial variation both in the vertical and horizontal. The wavelength dependence of scattering for the 2 cases also has few similarities. On 8 July the Ångström exponent for all 3 wavelengths showed that there was little difference in the scattering between the wavelengths ($a_{(450/550)}$ has a mean of

0.023, $\hat{a}_{(450/700)}$ a mean of 0.36 and $\hat{a}_{(550/700)}$ of 0.65), while on 17 July the $\hat{a}_{(450/550)}$ has a mean of 0.36, $\hat{a}_{(450/700)}$ a mean of 0.70 and $\hat{a}_{(550/700)}$ of 0.98, a larger difference between the wavelengths. The backscatter fraction was also different in the 2 cases. It was lower on 8 July, when the scattering coefficient in the dust layer was also lower. The measured dry aerosol single scattering albedo in dust varied between 0.73 to 0.91. This is within the range used by models in the past. For 550 nm the single scattering albedo in various dust optical models (Foucart et al., 1987; Sokolik and Golitsyn, 1993; Tegen et al., 1996) varied between 0.63 and 0.97. There is a considerable difference between different models and was hypothesised by Sokolik and Toon (1996) as being caused by an inability to model the physical processes responsible for dust optical properties, measurement uncertainties, and also by actual differences in properties of mineral aerosols originating from various geographical regions. They also conclude that this can be a serious complication in assessment of regional dust effects.

Comparison of our measured single scattering albedos in the dust layer on 17 July to the values calculated for different effective radii of mineral dust particles by Tegen et al. (1996), assuming a refractive index of $1.56 + 0.006i$ at 550 nm gives good agreement for the model particles with an effective diameter of 2 μm . They calculate that 0.8 μm effective diameter particles would have a ω_0 of 0.956 whereas particles of 2 μm effective diameter size would have a ω_0 of 0.887, the latter value being close to our measured ω_0 s. For Tegen et al. (1996) to obtain a single scattering albedo of 0.73 however, they need an effective particle diameter slightly larger than 8 μm , which is much larger than the cut size of the cyclone. We have seen that the dry aerosol single scattering albedo in dust can vary considerably between events, and that it can even vary within the same dust layer (0.87–0.91; for 17 July). The difference between the 2 cases may be explained by the lower scattering coefficients on the 8th, even though the absorption coefficient was 1.4 times higher than during TF20. This could be the effect of different evolution of the dust layer aerosol size spectra by processes such as coagulation and cloud processing. It may also be due to the dust having different optical properties depending on its local origin within the same major source region. This

idea is supported by a study by Herrmann et al. (1999) which shows that the chemical composition of mineral dust particles can vary substantially within the Sahara and Sahelian regions as a function of local source area and time of year. The dust aerosols in this study originated from Libya and had spent a considerable time over Africa before arriving to Tenerife on 8 July, while on 17 July the dust originated in Senegal and had been transported a much shorter distance to Tenerife. Filter measurements of aerosol chemical composition made onboard the *Pelican* by Schmeling et al. (2000) also support these findings, as they found different chemical composition of the aerosol particles on these 2 days.

The contribution by the aerosols to the scattering by a particle-free atmosphere (as estimated by the ratio $\sigma_{\text{sp}}/\sigma_{\text{rg}}$) was largest for the aerosols in the MBL on 8 July. On 17 July the situation was the opposite and the dust layer made the largest contribution. The dry aerosol optical depth was much lower in the MBL on 17 than 8 July, 0.015 and 0.022 respectively. The MBL depth over which the aerosol optical depth was determined was almost the same (960 m and 975 m, respectively). The difference was a factor of 1.5 for the same integration depth for these 2 days. The dust layer aerosol optical depth on the other hand was much higher on 17 July. The depth of the dust layer was also larger, 1965 m instead of 1170 m for 8 July. Most of the scattering material in the dust layer for 17 July can be found above 2375 m. When calculating the optical depth for the layer between 2375 m and the top of the flight profile on 17 July it becomes 0.036, which is still a factor of 3.3 higher than 8 July. The dry aerosol optical depth for the whole air column in the profiles was 1.6 times higher on 17 than 8 July due to the relatively high dry aerosol optical depth in the dust layer on 17 July which shows the importance of dust layers on the radiative properties of the atmosphere. We estimated the upper limit on aerosol optical depth by growing the size of the MBL aerosols which increases with the square of the GF, due to RH effects.

Comparison of extinction optical depths retrieved by a sunphotometer onboard the *Pelican* (Schmid et al., 2000) gives closure (within the estimated uncertainties, see Schmid et al., 2000) between the sunphotometer retrieved MBL aerosol optical depth at 550 nm wavelength and our

MBL aerosol scattering optical depth if we assume a GF of 1.9. This indicates that we sampled most of the aerosols important for radiative transfer in the MBL. In the dust layer however there is a factor of 6 to 7 difference, which suggests that a large fraction of the dust aerosols important for light attenuation had a size larger than $2.5\text{ }\mu\text{m}$ at ambient RH and therefore their scattering coefficients were not measured by the nephelometer.

5. Summary and conclusions

In situ measurements, on board the *Pelican* aircraft, of aerosol scattering and absorption coefficients over the Atlantic ocean in the ACE-2 area have been used to address the questions about aerosol optical properties asked in the introduction. The major intention has been to gain more knowledge of the vertical and horizontal variability of the optical properties of the aerosols that are actually present in the marine atmosphere, especially for dust and pollution aerosols after transport to the marine environment, in order to increase the ability to model global climate and to assess impact of anthropogenically produced pollutants and dust events.

The measurements made during the 2 days in this study, where dust layers were observed above MBLs with different pollution levels, show high variability in the aerosol scattering and absorption for particles smaller than $2.5\text{ }\mu\text{m}$ diameter ambient size. The vertical variation in scattering and absorption was highly dependent on the origin and history of the air mass, and varied from layer to layer and case to case.

The magnitude of the scattering and absorption coefficient in the MBL was a function of the pollution level and how effectively the mixing within the MBL took place. On 8 July, where the vertical mixing was poor, due to de-coupling of the MBL, the scattering coefficient showed a step-like profile with decreasing scattering with altitude. On 17 July, the absorption coefficient varied substantially with a low value at the sea surface and much higher in the middle of the MBL caused by local pollution from Tenerife. The dry single scattering albedo was found to be lower in the MBL with higher pollution levels (TF15) and decreased with height on both days.

The optical properties for the dust layers were

complex in the sense that the 2 dust layers encountered showed very different behaviour for the particles below $2.5\text{ }\mu\text{m}$ ambient size. During TF15 the absorption and scattering within the dust layer showed little variation in the vertical and horizontal and there was little difference in scattering as a function of wavelength. During the second event on 17 July the variability in the optical parameters were larger both in the vertical and in the horizontal. The wavelength dependence of scattering was larger, but the scattering at 550 nm and 450 nm were similar. The absorption coefficient in the dust layer was between 43 and 66% of the highest absorption coefficient in the MBL. The single scattering albedos encountered in the dust varied considerably between events (0.73 in TF15 and 0.87–0.91 in TF20). It was fairly constant for TF15, but varied within the dust layer for TF20. This indicates that the optical properties of dust particles could depend on its local origin within one and the same source regions, in this case Northern Africa.

On 17 July, the dry scattering coefficient in the dust layer and MBL were comparable in magnitude and even larger in parts of the dust layer. The dust layers were vertically more extensive than the MBL so that the optical thickness of the dust layer was comparable to that of the MBL aerosols at ambient RH. The absorption coefficient in the dust layer was between 65% and 2.8 times of that found in the MBL. This shows the significance of absorption by dust aerosols and stresses the need for inclusion of absorption by mineral aerosols when modelling the effect of mineral aerosols on climate.

We have shown that ground-level measurements of aerosol optical properties are likely not to be representative of the MBL, especially in cases of boundary layer de-coupling, let alone of the whole air-column above the Atlantic Ocean. This is in agreement with the measurements made near the Azores by Clarke et al. (1996), but different from the findings by Ogren and Sheridan (1996) over the North American continent.

Due to the complexity of the vertical structure and variability, both in the vertical and the horizontal, more in situ measurements of aerosol optical properties in the vertical are needed in order to tell if, when and where ground-based measurements can be representative of the whole column of air above the sample site. More meas-

urements are also needed of dust aerosol optical properties in order to verify dust optical models and assess the effects of enhanced mineral dust concentrations on climate and its potential role in the last glacial maximum. More measurements of the MBL optical properties of aerosols are needed to find a simple relationship between aerosol scattering and boundary level pollution levels.

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