

Airborne *in situ* characterization of dry aerosol optical properties in a multisource influenced marine region

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ABSTRACT

In situ data from the 2nd Aerosol Characterization Experiment (ACE-2) were used to describe the aerosol optical properties in a marine environment perturbed by continental sources, such as the outflow of European aerosol and dust outbreaks from North Africa. The data consist of airborne measurements made with an integrating nephelometer and absorption photometer.

The cases investigated in the ACE-2 experiment included vertical profiles flown in dusty, polluted and clean airmasses. While it was possible to describe the extensive optical properties (scattering and absorption coefficients) for the clean cases in a more general way, the polluted and dusty cases showed much variability from case to case. The intensive properties (single-scattering albedo and Ångström exponent) showed little variability with height, in agreement with other studies. A key result in this paper is the strong relationship between single-scattering albedo and transport time for the polluted cases.

Calculations of aerosol optical depth (AOD) made using *in situ* measurements showed that in the marine-dominated airmasses the contribution from the free troposphere (FT) to the total columnar AOD is not negligible. In the polluted cases, the marine boundary layer (MBL) dominated the contribution to the total AOD, when this was topped by a clean FT. In the cases when pollution outbreak in the MBL was accompanied by a dusty FT, most of the columnar AOD was due to the dust layer.

Assessment of the horizontal variability of the aerosol field, achieved by employing the dry scattering coefficient as proxy data, showed that there was variability in the aerosol concentration at scales below the airmass scale in which the aerosol was embedded. This shows that the concept that the aerosol concentration varies horizontally at the same scale as the airmass can lead to significant errors in the aerosol modelling schemes, in particular on the estimates of the impact of aerosols on the Earth's radiative balance.

1. Introduction

The impact of scattering and absorbing tropospheric aerosols on the Earth's radiative balance is recognized as a major climate forcing effect. In spite of this recognition, aerosol forcing estimates are still uncertain and preclude an accurate quantification of the contribution of aerosols to the total industrial-era climate forcing (IPCC, 2001).

The sources, sinks, spatial distribution, lifetime and fate of aerosols are highly variable over the Earth, making it (along with uncertainties in the radiative properties of aerosols) difficult

to assess their net effect on the climate of the planet. In order to improve the estimates and increase our understanding of the effects that aerosols have on the climate, it is important to more accurately establish the spatial, temporal and vertical variability of the properties of aerosols over a wide range of atmospheric conditions (Kaufman et al., 1997).

Atmospheric aerosols, regardless of their origin and size, can interact with solar radiation directly, through scattering and absorption processes, and indirectly, through the modulation of cloud properties, as a result of the particles acting as cloud condensation nuclei (CCN). Different aerosol types interact with radiation in a number of ways. Mineral dust, for example, affects not only the solar region of the spectrum by scattering and absorbing ultraviolet (UV) and visible radiation, but also interacts with infrared radiation and can have a similar effect to that of greenhouse gases due to absorption (Haywood et al., 2003). In addition, aerosols containing absorbing material, such as black

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carbon (soot), may reduce cloud cover and liquid water through an increase in atmospheric heating rates by absorption of solar radiation. The absorption may lead to evaporation of cloud and a reduction of clear air relative humidity. This mechanism has been hypothesized to enhance the daytime clearing of cloud, especially in polluted regions (Ackerman et al., 2000). Thus, aerosols exert a control over the amount of solar radiation that reaches the Earth's surface through different mechanisms.

Aerosol optical properties in remote areas are strongly dependent on the evolution of the synoptic airmasses and associated source regions, and to a certain extent can be characterized by the type and source of the airmass (Smirnov et al., 1995). Understanding the spatial and temporal links between aerosol optical features and airmass parameters is important in describing the mechanisms that control the optical state of the atmosphere.

The overall objective of this paper is to quantify the optical characteristics of the aerosols in the North Atlantic marine area, affected by different types of aged aerosols advected in over the region. A brief explanation of the data and instrumentation is given in Section 2. Section 3 reports on the different types of airmasses that reached the measurement area and the optical conditions relating to them. Results of the aerosol optical characterization are shown in Section 4, including closure tests in which collocated measurements of layer aerosol optical depth (AOD) made with a sun photometer installed on-board an aircraft are compared with AOD derived from *in situ* measurements. The results are discussed in Section 5.

The specific questions that we will address here are:

- (1) What is the vertical and horizontal variability in extensive and intensive aerosol properties?
- (2) What is the relationship between the variability in intensive aerosol properties and changes in signatures of different airmasses and atmospheric conditions?
- (3) Under what conditions are measurements of aerosol optical properties made in the marine boundary layer representative of the aerosol properties within the column?
- (4) What are the contributions of the marine boundary layer and free troposphere aerosols to the total AOD?

2. Experiment, data and relevant instrumentation

2.1. Experiment

To investigate the features of the aerosol optical properties in a marine environment we use data acquired during the "North Atlantic Aerosol Characterization Experiment" (ACE-2), which took place from 16 June to 24 July 1997 over the subtropical northeast Atlantic. The main goals of the experiment were to describe the radiative effects and controlling processes of aerosols in an area perturbed by continental sources, including the out-

flow of European aerosol and dust outbreaks from North Africa (Raes et al., 2000).

The ACE-2 study area exhibits typical characteristics of a subtropical oceanic atmosphere in the vicinity of a continent, as discussed by Verver et al. (2000). Among these characteristics the most notable is a temperature inversion as consequence of heating by compression of the subsiding air over a relatively cold ocean. As a result the marine boundary layer (MBL) and the free troposphere (FT) are decoupled, showing different chemical and thermodynamic characteristics. Earlier observations have shown that in the pristine MBL of the subtropical northeast Atlantic clean air alternates with anthropogenic contamination caused by pollution outflows from Europe and, on occasion, North America. Similarly, in the FT aloft, clean airmasses alternate with mineral dust, transported out of North Africa, as described by Prospero et al. (1995). Thus, the area offers a variety of meteorological conditions for studying the characteristics of different aerosol types.

2.2. Platform and instrumentation

All data presented in this study were obtained on-board the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) platform denoted *Pelican*. This aircraft is a modified Cessna 337 Skymaster with the forward engine eliminated, designed to perform low-altitude flights for atmospheric and oceanographic research (Bluth et al., 1996).

An isokinetic inlet was mounted at the front of the aircraft, through which the sample air was drawn and subsequently passed through a bank of cyclone impactors, constraining the sample so that only particles with diameter of less than $2.5 \mu\text{m}$ (fine particle size fraction) were measured. After the impactors the aerosol sample was gently heated to maintain the relative humidity (RH) at less than 40%. RH-induced variations in the scattering coefficient are minimized by maintaining a low sample stream RH of 10–40% (achieved by modest heating of 0–10 °C; Anderson and Ogren, 1998). Once the large particles had been removed and the air stream dried, the air stream was passed through the aerosol instruments. The aerosol optical properties measured consist of the wavelength-dependent dry scattering coefficient, σ_{sp} , and the wavelength-dependent dry absorption coefficient, σ_{ap} , each with units of inverse distance (10^6 m^{-1} , denoted by Mm^{-1} herein).

Measurements of aerosol scattering (7° – 170°) and hemispheric backscattering (90° – 170°) coefficients were made with an integrating nephelometer (model 3563, TSI Inc.) at wavelengths of 450, 550 and 700 nm. The hemispheric backscattering measurements are accomplished by use of a shutter that blocks illumination from regions corresponding to scattering angles of less than 90° . Sources of uncertainties associated with the use of the integrating nephelometer include photon-counting inaccuracies during measurements, zeroing and calibration. The nephelometer measurements were corrected for variations from standard temperature and pressure (STP), truncation of the

forward-scattered radiation and deficiencies in the illumination source, using the method of Anderson and Ogren (1998).

The aerosol light absorption measurements used a filter-based differential transmission device. σ_{ap} was measured at 565 nm with a particle soot absorption photometer (PSAP; Radiance Research, Seattle, WA), by monitoring the change in transmission through a filter. This device responds to differential light transmission through a Teflon filter as particles are loaded onto it. The measured values were corrected for the deposit spot size, the PSAP flow rate, the manufacturer's calibration and multiple scattering artefacts according to a calibration study of Bond et al. (1999). The latter correction is necessary since filter-based methods yield an artificially high absorption coefficient if transmission through the filter is reduced by the presence of scattering particles on the filter. The aerosol data were collected at 1 Hz, but data at this resolution are too noisy to be useful. We have thus run 30 s averages to eliminate most of the high-frequency noise while preserving all of the meaningful information in the measurements.

2.3. Flight patterns

During ACE-2 a total of 21 data-collecting missions were flown between 18 June and 20 July 1997. These dedicated flights consisted of straight and level runs, ascending and descending profiles, within, below and above (maximum altitude ~ 4 km) the aerosol layers, to enable characterization of the aerosol over a broad range of atmospheric conditions. The missions were typically 2 to 10 h in duration. The flight altitude ranged from 15 to 4000 m and the typical cruising speed was about 50 m s^{-1} . The aircraft altitude and location were determined from an on-board Global Positioning System (GPS).

3. Characterization of airmasses during ACE-2

Rather than depicting the aerosol optical parameters for each *Pelican* flight, we find that the results will be of greatest interest when stratified by airmass type. A potentially useful facet of the assessment of aerosol optical properties is to identify patterns in the altitude dependence of these properties, and their dependence on the airmass type in which the aerosols are embedded. Measurements performed on the eastern seaboard of the United States (Hegg et al., 1995) suggest that there can be a significant vertical gradient in the aerosol light scattering coefficient. Such gradients must be taken into account when estimating AODs for radiative forcing assessments.

During the ACE-2 measurement period it was possible to observe three different optical conditions associated with three main aerosol sources, which we define in terms of generalized source trajectories:

(1) Clean marine airmasses: defined as airmass trajectories confined to the Atlantic and Arctic Oceans containing aerosols

that had travelled under unperturbed conditions for the entire length of the 5 day transport time. Pure maritime Atlantic conditions predominated on 30 June to 4 July. During these days the *Pelican* sampled clean marine air that had spent several days over the North Atlantic and/or polar region, according to the trajectory analysis. Such clean airmasses were advected over the Canary Islands as a result of the presence of a low-pressure system over Western Europe. The location of the Icelandic Low, as well as the Azores High, affected whether the flow was Atlantic or polar (Quinn et al., 2000).

(2) Dust outbreaks: airmasses originating from North Africa were responsible for dust-laden air sometimes present in the FT. Dusty conditions in the FT prevailed on 9, 16 and 17–19 July.

(3) Pollution outbreaks: anthropogenic airmasses that originated in Europe injected high particle concentrations into the MBL. Three major pollution outbreaks were observed in the MBL during the ACE-2 campaign, and two of them (namely 9–10 July and 17–20 July) are described in more detail in this paper. The first episode resulted from flow originating in western Europe, where a ridge of high pressure stretched across the North Atlantic and extended over France. Northeasterly flow brought polluted air just west of the Iberian Peninsula to the Canary Islands. During the second event the Azores High and a low-pressure system situated over Europe were positioned such that east-northeasterly flow brought polluted air directly from the Iberian Peninsula (Quinn et al., 2000).

All back trajectories were obtained from the Royal Dutch Meteorological Institute (KNMI) (Scheele et al., 1996).

4. Aerosol characterization

The aerosol optical measurements obtained for each flight include the extensive properties, that is, properties that vary in proportion to the aerosol concentration, as well as the intensive properties, which are independent of the amount of aerosol present (Ogren, 1995). The extensive properties dealt with here are expressed in terms of the aerosol light scattering (σ_{sp}) and absorption coefficients (σ_{ap}). From the measurements of these quantities, two intensive aerosol optical properties have been derived: the single-scattering albedo (ω_0) and the Ångström exponent ($\text{\AA}$).

4.1. Extensive properties

4.1.1. Vertical profiles and straight level runs (SLRs)

(1) *Scattering coefficient.* The vertical distribution of the extensive properties of an aerosol in the atmosphere is closely related to the aerosol mass distribution, which in turn is related to the vertical structure of the atmosphere. As shown in other investigations, measurements of the light scattering coefficient present varying vertical patterns in different environments (e.g. Magi et al. (2003) from the SAFARI2000 experiment, Hartley

et al. (2000) from TARFOX and Öström and Noone (2000) from two case studies using ACE-2 data).

Quinn et al. (1996) have shown the dependence of the aerosol scattering coefficient on particle size for both marine and continental environments. For marine cases, scattering by the total aerosol is affected by the submicrometre and supermicrometre modes, but is largely controlled by the supermicrometre mode due to its large mass concentration. For continental cases, the submicrometre mode has a larger effect on the scattering compared with the marine case. However, due to the lower mass concentration, the supermicrometre mode has a smaller effect.

The clean marine cases investigated here showed vertical profiles of light scattering typical of a boundary layer dominated mainly by sea salt. An upwards flux of sea-salt aerosol, formed by the bursting of small bubbles of the sea surface, is considered to enhance the concentration of submicrometre aerosol in the clean MBL (Hoell et al., 2000). As a result, aerosol light scattering is highest at the lower part of the column, generally below 950 hPa, where the concentrations of sea-salt particles are relatively large, and decreases with distance from the surface (virtually no aerosol scattering at the top of the layer, or lower than the noise level of the nephelometer) (e.g. Glantz et al., 2004). There is little difference between the scattering coefficients in the three wavelengths (Fig. 1), when compared with similar measurements made under polluted conditions (Fig. 3).

Figure 2 shows the statistical summary of the green scattering coefficient σ_{spG} for flights TF08, TF09 and TF11, representative of clean background aerosols. The box-and-whisker plot shows the medians and ranges (as indicated by percentiles) of σ_{spG} . The line in the centre of each box represents the median of the distribution, while the edges of the box give the 25th and 75th percentiles. The whiskers extend to the 5th and 95th per-

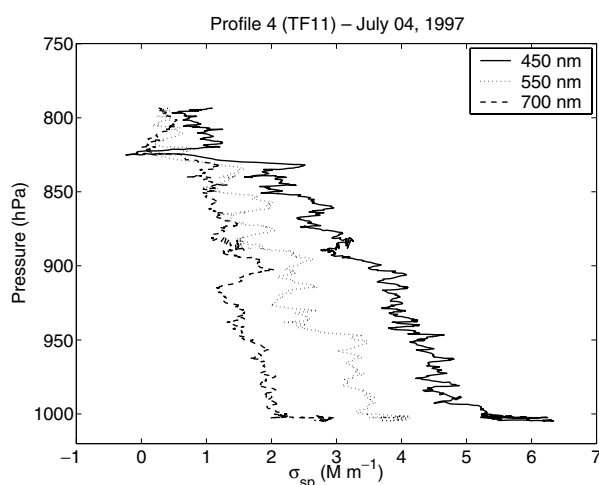


Fig. 1. The vertical profiles of dry scattering coefficient σ_{sp} at $\lambda = 450, 550$ and 700 nm for the *Pelican* flight TF11 on 4 July 1997 plotted as a function of pressure. During this flight a clean marine air mass extended from the MBL to the FT.

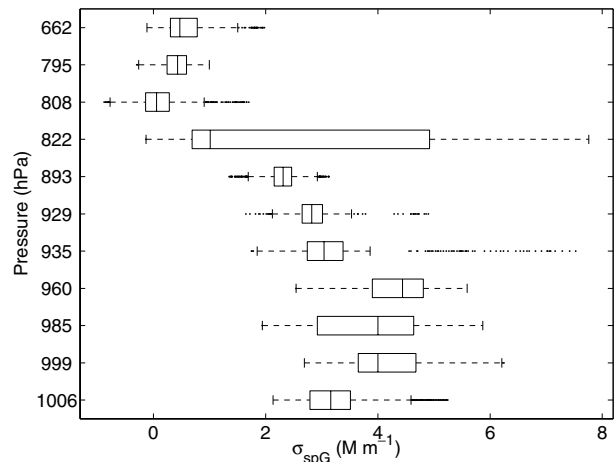


Fig. 2. Statistical summary of dry scattering coefficient σ_{sp} at green wavelength ($\lambda = 550$ nm) as a function of pressure, during the *Pelican* missions flown under clean marine conditions (i.e. TF08 on 30 June, TF09 on 1 July and TF11 on 4 July 1997). Each pressure level corresponds to an aggregate of data collected during the three flights. These box plots show the median (solid line), the interquartile range (box), the 5th and 95th percentile ranges of the data (whiskers) and outliers (dots outside the whiskers).

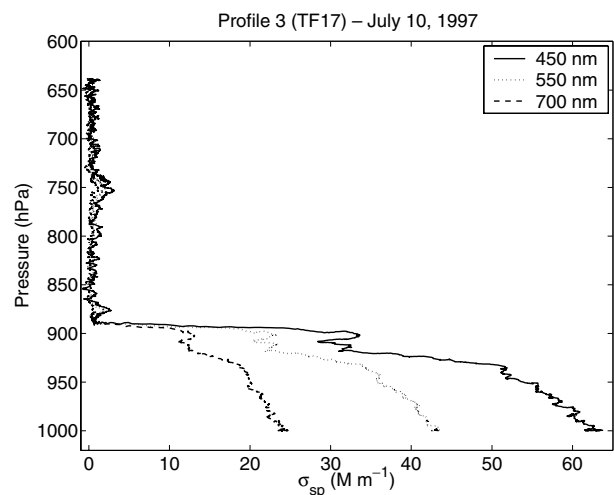


Fig. 3. The vertical profiles of dry scattering coefficient σ_{sp} at $\lambda = 450, 550$ and 700 nm for the *Pelican* flight TF17 on 10 July 1997 plotted as a function of pressure. During this flight, the polluted MBL was topped by the clean FT.

centiles and the dots outside them are outliers. The variability of σ_{spG} at any given level is greater than the vertical variability of the median values. For example, at 999 hPa the values of σ_{spG} range from 2.7 to 6.25 Mm^{-1} , while the median goes from 4.0 Mm^{-1} at 999 hPa to 4.4 Mm^{-1} at 960 hPa. Moreover, σ_{spG} becomes less variable with height due to the decrease of aerosol concentration (except at 822 hPa, where an elevated aerosol

layer, with relatively higher scattering coefficient, on flight TF08 introduced more variability at this level). Aircraft-based temperature measurements (not displayed here) showed that the MBL remained mostly unstable through the measurement period for the aforementioned cases. The general structure of the MBL can be described as deep and uniform in the measurement area, and dominated by the large-scale circulation. A weak inversion was sometimes observed along some of the profiles.

Pollution-influenced cases were generally (except during flight TF23) confined to the bottom of the MBL (typically under 1200 m), and topped by a strong inversion. Above the pollution layer, a layer with a different type of aerosol (either marine or dust) was observed, which is reflected in the measurements of the scattering coefficient (Fig. 3). In contrast to the clean marine cases, there was no functional dependence in the pollution cases (i.e. decrease of aerosol light scattering with height). The profiles varied considerably from case to case, showing complex stratification within the column and evidence of horizontal variability when comparing profiles from the same flight measured at different positions. Wood et al. (2000) discussed the existence of patches or differences in aerosol concentration in the MBL and the complexity of the evolution of polluted outflow from continental Europe when it is advected in over the oceans. The lower troposphere shows a complicated structure, with features inherent to the well-mixed European plume outbreak and to local processes taking place in a shallower marine internal boundary layer, which is formed when the continental air is transported over a relatively colder ocean. In light of this, the enrichment of continental air by maritime aerosol components can represent a source of variability, which depends both on the trajectory time over the ocean and on local factors. Additionally, the pollutants may undergo sedimentation, wash-out or chemical processes along the way, which lead to changes in their optical properties. The box-and-whisker plot in Fig. 4 shows that the scattering coefficient in the polluted air mass has a large range of values for a given flight level, and the median can differ by 1.5 orders of magnitude between different pressure levels (e.g. at 1000 and 937 hPa).

Dust-laden layers (when they appeared) were always capping the MBL (Fig. 5). Three major dust events affected the area during the measurement period, as evidenced by backtrajectory charts and corroborated by the AOD ($\lambda = 550$ nm) records on Mount Teide (Formenti et al., 2000). The distinction between the FT aerosol optical properties that contained mineral dust and those that did not was clear. As previously shown by Collins et al. (2000), the scattering coefficients in dust-laden layers were comparable to those resulting from boundary layer aerosols injected by continental outflows. The profiles within the dust layer also present a more stratified structure. The statistical summary for flight TF19, on 16 July 1997 (Fig. 6), when a dust layer was observed aloft, shows that the scattering coefficient is also variable within the dust layer, for a fixed pressure level. Between

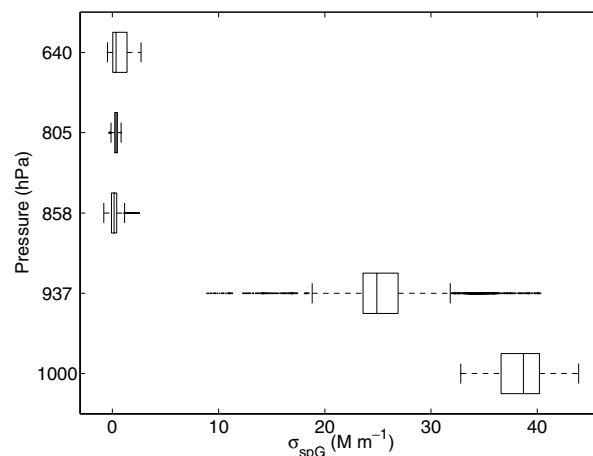


Fig. 4. Statistical box and whisker plots of dry scattering coefficient σ_{sp} at green wavelength ($\lambda = 550$ nm) plotted as function of pressure, during the *Pelican* mission TF17 on 10 July when the polluted MBL was topped by a clean layer that extended into the FT. These box plots show the median (solid line), the interquartile range (box), the 5th and 95th percentile ranges of the data (whiskers) and outliers (dots outside the whiskers).

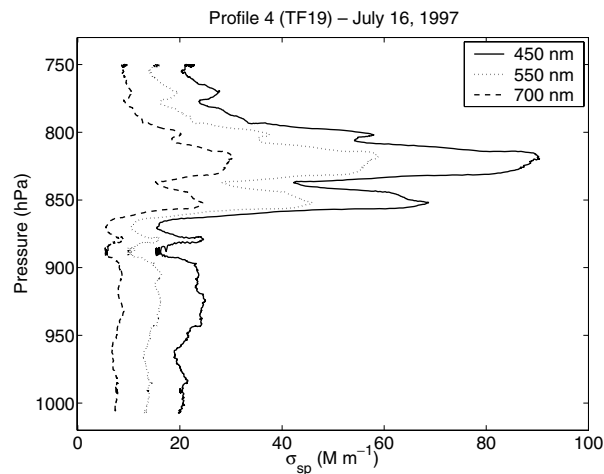


Fig. 5. The vertical profiles of dry scattering coefficient σ_{sp} at $\lambda = 450, 550$ and 700 nm for the *Pelican* flight TF19 on 16 July 1997 plotted as a function of pressure. During this flight, a clean layer MBL was topped by a dust layer extending into the FT.

individual levels of the same flight the values of σ_{spG} differ by two orders of magnitude.

(2) *Absorption coefficient.* The filter of the PSAP was sensitive to sudden changes in pressure and may have been deformed during vertical profiling flights. Therefore, the values of σ_{ap} are not fully reliable during aircraft ascent and descent. The data set was screened to extirpate these periods, and σ_{ap} data were used only from SLRs when the observed change in transmittance due to particle loading dominated over that caused by pressure variations.

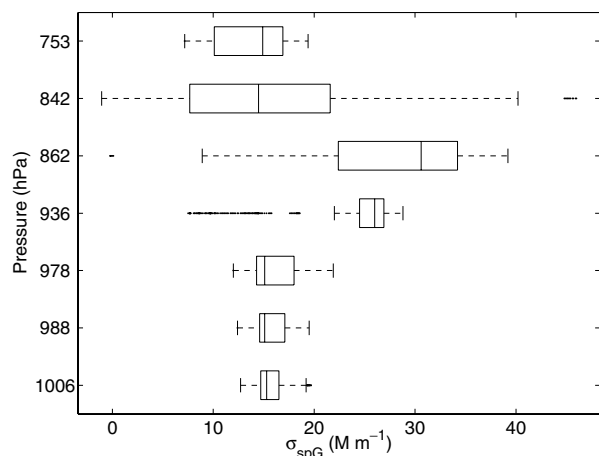


Fig. 6. Statistical box and whisker plots of dry scattering coefficient at green wavelength σ_{sp} ($\lambda = 550$ nm) plotted as a function of pressure, during the *Pelican* mission TF19 on 16 July 1997 when the clean layer MBL was topped by a dust layer. These box plots show the median (solid line), the interquartile range (box), the 5th and 95th percentile ranges of the data (whiskers) and outliers (dots outside the whiskers).

The variability of median σ_{ap} is surprisingly low throughout the same air column as well as when compared between flights (e.g. 1.0 and 1.1 Mm^{-1} in the clean marine column measured on 30 June 1997, and between 1.0 and 1.5 Mm^{-1} in the polluted case on 16 July 1997). While σ_{sp} follows the vertical distribution of the airmasses, the variation of the σ_{ap} coefficient is not as well correlated. Similar results had previously appeared in the study of Öström and Noone (2000), when they analysed two *Pelican* flights (TF15 on 8 July and TF20 on 17 July 1997), both with three discernibly different layers in the vertical: a mineral dust layer in the FT and a polluted MBL separated by a clean layer.

Putaud et al. (2000) carried out analysis of submicrometre aerosol chemical composition and aerosol absorption coefficient for two locations: Izaña (IZO), located at 2360 m along a ridge in

the central region of the island of Tenerife, and Punta del Hidalgo (PDH), located at the northern end of the island, at an elevation of 30 m. In their analysis, the change in aerosol chemical composition and mass is clearly seen in response to the advection of different airmasses. The measurements made at PDH showed an enhanced fractional contribution of sulfates (especially non-sea-salt sulfate, nssSO_4^{2-}), organic compounds (OC), NH_4^+ , and NO_3^- in the polluted cases, but little contribution in the black carbon (BC) mass, the most important light-absorbing aerosol species in the atmosphere at visible wavelengths (e.g. Bodhaine, 1995). A slight fraction of BC is seen in the total mass on 9, 11–13 and 16–18 July, and (associated with this signature) there is an increase in the absorption coefficient at PDH, ranging from 1.2 to 1.6 Mm^{-1} (Putaud et al., 2000). The absorption coefficients for *Pelican* flights TF16 on 9 July and TF20 on 17 July show a more pronounced variation in the vertical in these cases than on the other missions flown during continentally influenced airmasses, when evidence of intrusion of absorbing aerosol was not so evident. Measurements made with an aethelometer at PDH (not shown here) displayed a similar pattern, with the largest increase in σ_{ap} occurring when the BC mass increases and relatively low values ($\sigma_{ap} < 1.5 \text{ Mm}^{-1}$) when nss-SO_4 , NH_4 and NO_3 were the predominant species in the polluted airmasses. Table 2 presents the mean and range of the extensive aerosol properties for the different airmasses encountered during the ACE-2 *Pelican* missions.

4.1.2. Spatial variability. Knowledge of the spatial distribution of aerosols is relevant to climate issues, the mapping of pollutants, the study of the structure of air pollution patterns and tailoring of strategies for chemical sampling in aerosol field campaigns. At a planning level, the proper description of the spatial distribution of aerosols is also a necessary step for the design of an optimal aerosol monitoring network, and to streamline existing networks by identifying redundant stations.

The assumption that the aerosol concentration, and its associated extensive properties, vary horizontally at the same scale as the airmass scale and remain invariant within the airmass is ubiquitous in many aerosol modelling and observational activities. This practice is reflected in the studies of, for example, Schwartz (1989) and Benkovitz et al. (1994), the results of which showed that the aerosol and sulfate transport distances, in the lower troposphere, were related to synoptic-scale meteorological phenomena. Anderson et al. (2003), in analysing *in situ* light-scattering data from two ground stations, as well as airborne and space-borne AOD measurements, showed that aerosol plumes do occur in the atmosphere at scales of several hundred to a few thousand kilometres but are rarely internally homogeneous. In their study, they showed evidence that aerosol plumes occur at subsynoptic scales. The average characteristic scales were found to be less than 10 h (temporal) and 200 km (spatial). They attribute this variability to the mesoscale nature of both the major aerosol sources (e.g. dust storms, biomass fires and urban plumes) and the major sinks (e.g. precipitation), as well

Table 1. Summary of ACE-2 *Pelican* flights used in this study with the type of column and airmass found in each reservoir (marine boundary layer, MBL; free troposphere, FT)

Flight number	Data	Type of column	Type of aerosol
TF08	30 June	Clear	Clean marine (MBL and FT)
TF09	01 July	Clear	Clean marine (MBL and FT)
TF11	04 July	Cloudy	Clean marine (MBL and FT)
TF16	09 July	Cloudy	Polluted (MBL) and dust (FT)
TF17	10 July	Clear	Polluted (MBL) and marine (FT)
TF19	16 July	Cloudy	Marine (MBL) and dust (FT)
TF20	17 July	Clear	Polluted (MBL) and dust (FT)
TF22	19 July	Cloudy	Polluted (MBL) and dust (FT)
TF23	20 July	Cloudy	Polluted (MBL) and marine (FT)

Table 2. Summary of the extensive properties during the ACE-2 *Pelican* missions. $\sigma_{sp(550)}$ was evaluated during vertical profiling and $\sigma_{ap(565)}$ during the SLRs. Note that the summary for the FT polluted airmass was done using data from flight TF23 only

	Boundary layer		Free troposphere	
	$\sigma_{sp(550)}$ (Mm ⁻¹)	$\sigma_{ap(565)}$ (Mm ⁻¹)	$\sigma_{sp(550)}$ (Mm ⁻¹)	$\sigma_{ap(565)}$ (Mm ⁻¹)
Marine	9.20	1.1	1.4	1.0
	1.0–43.7	0.09–2.2	1–20.8	0.1–2.00
Pollution	21.0	1.2	16.6	1.1
	2.0–49.10	0.1–2.6	1–41.5	0.3–1.7
Dust	–	–	17.6	1.1
			1.0–46.1	0.1–3.1

as sporadic vertical transport of clean air from the FT into the boundary layer.

Following Anderson et al. (2003), we assess the horizontal variability of the dry aerosol concentration during ACE-2 *Pelican* flights by using the dry scattering coefficient at 550 nm with 10 s resolution. This parameter is highly correlated to low RH aerosol mass concentration ($r \sim 0.9$), with a mass scattering efficiency close to 3.0 m² g⁻¹ for submicrometre-dominated industrial haze (Waggoner et al., 1981; White et al., 1994) and considerably lower and more variable efficiencies for supermicrometre-dominated dust (White, 1990) and sea-salt (Quinn et al., 1995) particles. Thus, even though σ_{sp} is not a direct measurement of aerosol mass, it can be used as a good indicator of this quantity.

Assessment of the aerosol spatial variability is made by examining the autocorrelation coefficient $r(k)$, which is a measurement of the correlation between observations at different times. For evenly spaced data, the general autocorrelation coefficient for measurements x separated by k time intervals is

$$r(k) = \frac{\sum_{t=1}^{N-k} (x_t - \bar{x})(x_{t+k} - \bar{x})}{\sum_{t=1}^N (x_t - \bar{x})^2}, \quad (1)$$

where N is the number of observations and $\bar{x} = \sum_{t=1}^N x_t / N$ is the overall mean.

The spatial correlation is expected to change not only in time and space but also depending upon the aerosol concentration and aerosol type. Because of this, the scattering coefficient at 550 nm has been stratified by airmass type, according to previous classification, and whether it was measured within the MBL or in the FT. The data set was also restricted to SLRs, to assure that the variations in the spatial correlation were due to horizontal inhomogeneities and not because of the difference in aerosol concentration in the column. This eliminates the problem only partly, because even though the aircraft flew constant-altitude transects, in practice the pressure surfaces are sloped. A better approach to solving this problem would be by using measurements of scattering coefficient at isentropic surfaces. In addition, the analyses were restricted to flight transects long enough to cover a wide sampling domain (117–244 km in length). Repeated transects

have been discarded to avoid redundant information due to re-sampling. Owing to this restricted spatial coverage, the data will give more information about short spatial scales of variability. Using these criteria, the assessment of the horizontal variability can be done for the FT using data from two flights flown in dust layers, two flights in clean layers and one flight in a pollution outflow case.

With a typical cruise speed of 50 m s⁻¹, the *Pelican* was covering the distances of interest here fast enough to disregard the temporal evolution of the aerosol, and therefore the data are suitable for providing information about the spatial correlation structure of the aerosol. In other words, the difference between two samples depends only on the distance and not on the time. Thus, $r(k)$ is expected to be invariant in time.

A simple way of summarizing the autocorrelation in a time-series is by plotting the $r(k)$ statistic as a function of the lag. Ideally, the correlation decreases between samples located further apart. At distances shorter than a certain threshold, a correlation exists between two or more measurements in space. Anderson et al. (2003) adopted $r > 0.9$ to define “good correlation” and $r > 0.8$ to define “acceptable correlation” thresholds.

The spatial correlation coefficients as a function of time lag are plotted in Fig. 7 for the various cases studied here. The graph shows a constant positive value of r (high autocorrelation) that persists up to 1 km. Without making any rigorous statements, an inspection of this figure shows that there is practically no difference among the autocorrelation values for the distinct FT airmasses studied here at very short spatial lags (up to 1 km). The decay in r is quite abrupt at larger scales. As one moves towards lower autocorrelation regions, the spatial scales of variability lie between 3 and 8 km (for $r > 0.8$). Our analysis of correlation reveals much shorter spatial variability than the results of Anderson et al. (2003), who found spatial offsets between 60 km ($r > 0.9$) and 120 km ($r > 0.8$). Table 3 shows the length of the SLRs for each case and the 99% confidence intervals for the spatial offsets.

Some possible factors explaining the differences between our results and the results of Anderson et al. (2003) could be (1) the length of the legs used in our study (considerably shorter than

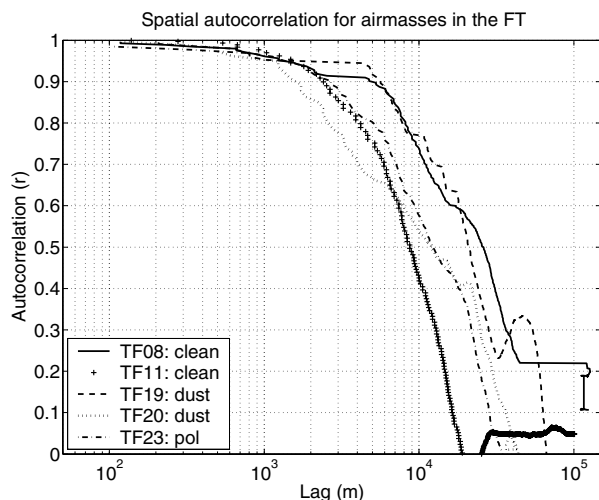


Fig. 7. Spatial autocorrelation of the dry aerosol concentration as a function of time lag for different airmasses in the free troposphere obtained during ACE-2 *Pelican* missions. This autocorrelation statistic is calculated using the dry scattering coefficient σ_{sp} at green wavelength ($\lambda = 550$ nm) as proxy data.

those used by Anderson et al. (2003), whose transect lengths were up to 648 km); (2) differences in the nature of the aircraft data (we used scattering coefficients measured in the FT, whereas Anderson et al. (2003) used boundary layer measurements). In our case, using shorter legs may preclude the assessment of the extent of longer scales of variability, although a large data set does not necessarily represent aerosol variability over the entire measurement domain. This basically depends on whether the data were collected randomly or in a region where certain aerosol events were anticipated. Finally, (3) the different geographical locations and airmass types can partly account for the dissimilarities between our results obtained with ACE-2 data and the ACE-Asia data employed by Anderson et al. (2003).

Charlson (personal communication, 2003) suggested that atmospheric instability could facilitate local vertical mixing and the creation of aerosol plumes in the boundary layer. Depending

upon their strength, these local plumes can penetrate into the FT and dilute the aerosols even when they (the plumes) occur sporadically and sparsely in the MBL. Instead of having a homogeneous horizontal aerosol distribution within the airmass, there can be patches distributed randomly into the aerosol field. The distance between these patches, which is directly related to the stability of the boundary layer beneath, defines the degree of horizontal variability within a given airmass.

It has also been hypothesized that the degree of this horizontal perturbation may also evolve in time. Once these aerosol bursts from the boundary layer reach the layer aloft, the patches may deform in response to the winds in the FT. While it is beyond the scope of this paper to evaluate the extent of the time evolution of these patches, we suggest that a Lagrangian study comprising detailed measurements of both boundary layer meteorology and aerosol would be a suitable way to assess more thoroughly the horizontal variability in the aerosol field.

4.2. Intensive properties

4.2.1. Ångström exponent. The Ångström exponent A describes the relationship between the aerosol scattering coefficient and the wavelength, and thus provides information on aerosol size distribution (Ångström, 1929, 1964). When evaluated in the vertical, A is an indicator of the vertical column size distribution. Typically, continental sites have scattering coefficients that are dominated by submicrometre aerosols and show higher values of A , while in remote sites, e.g. those affected by sea-salt aerosols, the supermicrometre fraction typically dominates and A has smaller values. Mean values of A for the 450–700 nm wavelength pair derived from

$$A = -\frac{\log(\sigma_{sp}(\lambda_1)/\sigma_{sp}(\lambda_2))}{\log(\lambda_1/\lambda_2)}, \quad (2)$$

are shown for the different airmasses and flights in Tables 4, 5 and 6. Since the nephelometer measurement included all particles with D_{aero} less than $2.5 \mu\text{m}$, only the fine mode has been measured and care must be taken when interpreting values of A .

Table 3. Information about the flight transects used to calculate the autocorrelation coefficient and the spatial offsets associated with the 0.9 and 0.8 autocorrelation indicator. The limits of the spatial offsets for the 99% confidence intervals are also shown

Flight	Airmass type	Length of the SLR (km)	99% confidence interval					
			Spatial offset (km)		$r = 0.8$		$r = 0.9$	
			$r = 0.8$	$r = 0.9$	Lower limit (km)	Upper limit (km)	Lower limit (km)	Upper limit (km)
TF08	Marine	127	8	5	7.57	8.36	4.87	5.11
TF11	Marine	166	4	2.8	3.81	4.16	2.74	2.85
TF19	Marine and dust	244	8	6	7.76	8.21	5.92	6.07
TF20	Polluted and dust	117	3	1.6	2.91	3.09	1.58	1.62
TF23	Polluted and marine	134	5	3	4.84	5.14	2.95	3.04

The values of A in clean marine airmasses displayed less variability and the highest mean of all cases within the MBL, indicating more small-diameter particles relative to other airmasses (Table 4). This suggests that A can be described in a more general sense and that the marine airmass tends to preserve its optical properties in the column. In the FT, due to the very small scattering coefficients, the values of A could not be retrieved.

Polluted airmasses had high values of A corresponding to generally larger extinction contributions from smaller particles, but these values were, however, smaller than A for the clean marine cases (Table 5). The vertical variation of A within the same flight

Table 4. Ångström exponent A derived for the 450–700 nm pair, single-scattering albedo and the corresponding flight levels during the ACE-2 *Pelican* missions flown under clean marine conditions

Flight	Level (hPa)	ω_0	A
TF08	823	0.83	2.8
	850	0.89	2.5
	960	0.82	2.6
	984	0.83	2.5
	999	0.80	2.5
TF09	963	0.81	2.2
	983	0.80	2.2
	999	0.81	2.1
TF11	893	0.69	2.0
	929	0.73	1.9
	935	0.75	2.0
	987	0.73	1.8
	1006	0.76	1.6
TF19	937	0.94	1.6
	978	0.94	2.4
	988	0.94	2.3
	1006	0.94	2.3

Table 5. Ångström exponent A derived for the 450–700 nm pair, single-scattering albedo and the corresponding flight levels during the ACE-2 *Pelican* missions flown under polluted conditions

Flight	Level (hPa)	ω_0	A
TF16	940	0.96	1.3
	960	0.95	2.0
	991	0.96	2.0
TF17	937	0.95	1.6
	1000	0.97	2.3
TF20	936	0.93	1.6
	1002	0.96	1.9
TF22	950	0.91	2.0
	1005	0.92	2.0
TF23	864	0.96	2.2
	1002	0.95	2.0

Table 6. Ångström exponent A derived for the 450–700 nm pair, single-scattering albedo and the corresponding flight levels during the ACE-2 *Pelican* missions flown under dusty conditions

Flight	Level (hPa)	ω_0	A
TF19	753	0.93	–
	842	0.93	–
	862	0.96	–
TF20	638	0.91	0.8
	690	0.96	0.7
	807	0.89	0.7
TF22	835	0.93	0.9

in the polluted cases was also considerable for some flights (e.g. from 2.3 at 1000 hPa to 1.6 at 937 hPa, on flight TF17), showing that even the intensive properties of the aerosol are not uniformly distributed throughout the column.

In contrast to the values observed in the two previous cases, one can note a significant decrease in A for the Saharan dust events due to the predominance of large particles (Table 6). Low levels of aerosol scattering and the sensitivity of the nephelometer prevented the derivation of A for some episodes. The paucity of values of A for dust-laden air hinders our ability to assess the degree of variability of this quantity within the same air column and between flights carried out under these conditions. On the flights where the lower layer was topped by a dust layer (e.g. TF20), there was a decrease in the value of A right at the height of the interface, probably due to the intrusion and dilution of large particles with the smaller particles of the lower layer.

4.2.2. *Single-scattering albedo.* The single-scattering albedo, ω_0 , calculated as

$$\omega_0 = \frac{\sigma_{sp}}{\sigma_{sp} + \sigma_{ap}}, \quad (3)$$

is a measure of the relative magnitude of scattering and absorption by the aerosol, and is a key parameter to determine whether a layer is cooled or heated. It is one of the parameters taken into account in the calculation of global mean, annually averaged aerosol forcing at the top of the atmosphere (Haywood and Shine, 1995). In eq. (3), σ_{sp} is the nephelometer-measured dry scattering coefficient corrected for angular non-idealities and σ_{ap} is the measured absorption coefficient corrected according to Bond et al. (1999).

A feature often quoted to be typical in the optical properties of *continental* airmasses is their variation as a consequence of the variety of aerosol sources, degree of contamination and difference in the airmass history. However, ω_0 does not show such variability in the polluted outflows that reached the ACE-2 area. Within the MBL the values of ω_0 varied very little between individual levels of the same flight (Table 5; e.g. from 0.95 to 0.96 on flight TF16 flown under polluted conditions). There is not a significant amount of variability in the values of ω_0 among profiles

flown during pollution outflow events. For example, the lowest value of ω_0 was 0.91 on flight TF22 and the largest was 0.96 on flight TF16. These values were derived from measurements carried out during two independent pollution outbreaks. Putaud et al. (2000) showed that nssSO_4^{2-} was the main submicrometre aerosol component and, together with NH_4^+ , accounted for up to 84% of the submicrometre aerosol mass. The predominance of nssSO_4^{2-} in the submicrometre pollution aerosol explains the almost invariant nature of the ω_0 , both within the MBL for the same flight and among flights during different pollution episodes.

For marine-influenced airmasses, ω_0 varied little between individual levels of the same flight (Table 4; e.g. ω_0 ranged from 0.80 to 0.83 on TF08). Conversely, when compared among different marine-influenced airmasses, ω_0 showed a significant amount of variability. During background conditions the chemical composition of the submicrometre aerosol was dominated by nssSO_4^{2-} (29%), sea salt (21%), OC (20%) and NH_4^+ (8%). The partitioning of the submicrometre aerosol by components with different mass-scattering efficiencies may explain to some extent the variability in the values of ω_0 within the MBL among the *Pelican* flights flown under background conditions. The values of ω_0 depend on the contribution of the individual dominating components to the total aerosol mass, which in turn are linked to local factors. For example, the injection of sea salt into the atmosphere is dependent on the wind speed; nssSO_4^{2-} concentration is dependent on the dimethylsulphide (DMS) oxidation and even on the entrainment from the FT aerosol. Moreover, Raes (1995) has suggested entrainment of submicrometre aerosol from the FT to be the main source of particle number in the background MBL. The almost invariable nature of ω_0 within the MBL of a given flight and the variability among flights is evidence of the influence of local factors on this property during marine-dominated conditions.

In the FT the mean ω_0 is much more variable, both among flights and between individual levels of the same flight. This degree of variability with altitude can be attributed to the low signal-to-noise ratios due to very low aerosol concentration. Large values of ω_0 (0.96 on TF19 and TF20) occurred in the FT only in the presence of dust (Table 6), because of the high-scattering nature of this type of aerosol. ω_0 varied from 0.89 to 0.96 in the dust layer of flight TF20. This variability of ω_0 in Saharan dust aerosols has been reported previously by AERONET measurements (Takemura et al., 2002) and by Carlson and Benjamin (1980) who obtained values of 0.95 and 0.86, respectively.

Figure 8 shows statistics for the single-scattering albedo derived for pollution-influenced airmasses as a function of transport time. We define transport time as the number of hours the air-mass travelled since it last passed over or only a short distance (arbitrarily taken as distances <50 km) from a densely populated or industrialized area in the European continent until it reached the measurement point. It was estimated by the inspection of 5 day backtrajectories arriving in Tenerife at 975 hPa.

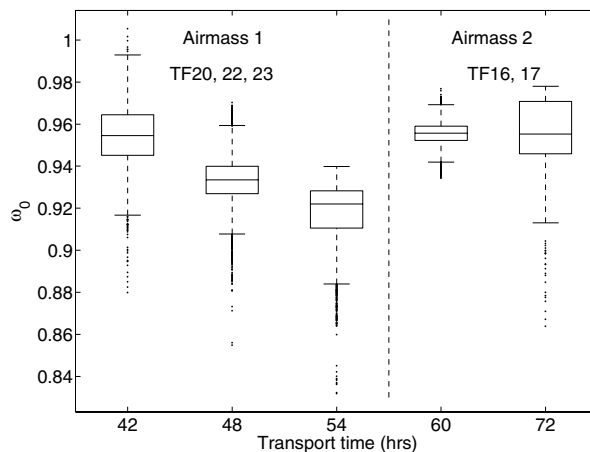


Fig. 8. Single-scattering albedo ω_0 for polluted cases as a function of transport time from Europe. Each box represents an independent *Pelican* mission flown in the MBL. These box plots show the median (solid line), the interquartile range (box), the 5th and 95th percentile ranges of the data (whiskers) and outliers (dots outside the whiskers).

In the figure, each distribution represents data of a flight segment carried out in the boundary layer. Inspection of Fig. 8 reveals that the five flights chosen are naturally grouped into two clusters. The first cluster consists of flights TF20, TF22 and TF23, the measurements of which were affected by a relatively weak pollution outbreak (on 17–20 July), that only touched the north and northeast coast of the Iberian Peninsula. There is a tendency for ω_0 to decrease with increasing transport time, as is evident by the median ω_0 changing from ~0.96 for a transport time of 42 h to ~0.92 for a transport time of 54 h. The ACE-2 second Lagrangian experiment (L2) (Johnson et al., 2000) was performed over a 29 h period between 16 and 18 July 1997, thus coinciding with part of the *Pelican* flight period we investigate here. Hoell et al. (2000) found that during L2 the fine mode concentration, centred at the dry particle diameter $d_{\text{dry}} = 60$ nm, decreased from 2700 to 1150 cm^{-3} , while the accumulation mode concentration (centred at $d_{\text{dry}} = 200$ nm) decreased from 670 to 430 cm^{-3} . By the end of the experiment, no growth in either the fine or the accumulation mode was observed. The decrease of concentration observed in both modes is an indicative that significant removal processes occurred over the period, thus explaining the decrease in the values of the scattering coefficients observed in our study. The reduction in concentration was explicable by dilution with free tropospheric air as the mixing layer deepened from 500 to 1100 m during the experiment, and to a lesser extent by coagulation.

The last two statistical boxes represent flights TF16 and TF17, which were influenced by a stronger pollution outflow. This episode lasted from 9–10 July, and was different from the first one because it remained for longer over the UK and western Europe. There is not much difference between the median of ω_0 for samples taken 60 and 72 h after last contact with the

European coast. These values are even slightly larger than the measurements made in the first polluted airmass at shorter transport times, despite more than 2.5 d since last contact with the land. The only two ways to increase the values of ω_0 are either by adding scattering material to or removing absorbing material from the airmass. Even though both scattering and absorbing materials may have been scavenged or deposited out along the transport track, the relatively large value of ω_0 in the second event can be attributed to the maintenance of the high-scattering nature of the continental plume through other mechanisms, e.g. gas-to-particle conversion. The characteristic timescale for the conversion of sulfur dioxide, the most predominant anthropogenic sulfur-containing air pollutant, to sulfates is approximately 100 h, which is comparable to the time that the plume in the second event needed to travel from its origin over the UK to the measurement point. While in the first event the transition towards a less scattering airmass occurred as the plume lost contact with the continent, in the second event removal and source processes competed to determine the nature of the plume after it lost contact with the source region. Aerosol formation mechanisms might have contributed to the maintenance of the almost steady values of ω_0 in the second pollution outbreak.

It is of utmost importance to accurately define the spatio-temporal variability of ω_0 , even though the variation in the values of this property is not large in some events studied here. The sign of the aerosol direct forcing is generally determined by the value of ω_0 and the surface albedo under clear-sky conditions. In the radiative transfer models problems can arise in the estimation of the direct aerosol radiative forcing by the improper assumptions of prescribed aerosol optical properties even if the aerosol distributions are properly simulated. Our preliminary results suggest that ω_0 has a dependence on the plume transport time and strength, but a larger number of observations are necessary to accurately assess the magnitude of this dependence. If a decrease in the value of ω_0 with increasing transport time can be shown to be characteristic of other pollution outbreaks, then this may provide a simple way to describe the time evolution of this quantity and assess regional- to global-scale aerosol effects.

4.3. Contribution of MBL and FT aerosols to the total AOD

In this section we present calculations of AOD and the contributions of the MBL and FT aerosols to the total AOD. To accomplish this we use *in situ* data (σ_{ap} and σ_{sp}) from four vertical profiles flown in cloud-free conditions. These vertical profiles are particularly suitable for this study because they provide continuous measurements from near sea-level to an altitude of approximately 4 km. Adding σ_{ap} and σ_{sp} allows us to estimate aerosol light extinction σ_e :

$$\sigma_e(\lambda, z, \text{RH}) = \sigma_{\text{sp}}(\lambda, z, \text{RH}) + \sigma_{\text{ap}}(\lambda, z, \text{RH}), \quad (4)$$

where z is the height, λ is the wavelength and RH is the relative humidity.

The AOD between heights z_1 and z_2 is given by:

$$\tau(\lambda) = \int_{z_1}^{z_2} \sigma_e(\lambda, z, \text{RH}) dz. \quad (5)$$

To calculate AOD we only use σ_{ap} data averaged over horizontal runs flown immediately before or after the vertical profiles discussed here. This assumption is strictly true for a homogeneous, time-invariant atmosphere.

Before AOD can be calculated, σ_{ap} and σ_{sp} must be adjusted to the ambient RH. The adjustment of σ_{sp} is relevant, especially when hygroscopic aerosol components are present. This adjustment is accomplished by using the methodology suggested by Gassó et al. (2000). The method uses measurements of σ_{sp} at two relative humidities: one slightly higher than the ambient humidity (RH_{wet}) and the other at lower humidity (RH_{dry}). The derivation of the correct ambient scattering coefficient ($\sigma_{\text{s[amb]}}$) with the two-point method assumes that scattering is a smooth function of RH between RH_{dry} and RH_{wet} , following the empirical equation of Kasten (1969):

$$\sigma_{\text{sp}}(\text{RH}) = (1 - \text{RH}/100)^{-\gamma}, \quad (6)$$

where γ is the aerosol hygroscopic exponent. γ depends upon the hygroscopic nature of the aerosol, and it has been shown to vary in ambient aerosols according to their chemical composition (Hänel, 1976).

Thus, the type of aerosol will affect the value of the derived γ as well as the validity of eq. (6). Gassó et al. (2000) used a simple method to identify the aerosol composition classes, which were mainly based on measurements that are correlated with the presence of certain airmass types (e.g. particle size distributions, airmass back trajectories, altitude of the aircraft and visual recognition). They classified their data in terms of three types of aerosols and obtained the following values of γ : γ (dust) = 0.23 ± 0.05 , γ (clean) = 0.69 ± 0.06 and γ (polluted marine) = 0.57 ± 0.06 . These values are employed in the present study.

Data from transects flown under clear air conditions were corrected for $\sigma_{\text{s[amb]}}$ whenever RH increased above the 40% threshold value adopted to characterize dry aerosol.

Little is known about the corrections that should be applied in order to adjust σ_{ap} to ambient conditions. However, since absorption contributed much less than scattering to the total aerosol optical depth, we assume here that the ambient absorption coefficient $\sigma_{\text{ap[amb]}} = \sigma_{\text{ap[dry]}}$.

Table 7 shows the layer AOD estimated from *in situ* measurements, and the contributions from the MBL and FT aerosols to the total columnar AOD. The results show that the clean and polluted MBL contribute more to the radiative forcing than the FT. However, in the cases when the clean marine condition extends from the bottom of the MBL up to the FT (i.e. TF08 and TF09), there is an important contribution from the upper layer aerosol to the total AOD. We attribute this to the fact that in the

Table 7. Summary of AOD estimated from airborne *in situ* ($\sigma_{\text{sp}(550)}$ and $\sigma_{\text{ap}(565)}$) measurements obtained during the ACE-2 Pelican flights

Flight	Airmass type	Layer AOD			
		MBL	FT	MBL (%)	FT (%)
TF08	Marine	0.0119	0.0068	63.6	36.4
TF09	Marine	0.0138	0.01148	54.6	45.4
TF17	Polluted and marine	0.0877	0.0016	98.2	1.8
TF23	Polluted and dust	0.0327	0.0404	44.7	55.3

clean cases the aerosol decreases monotonically with height and an important fraction of the aerosol burden is still found in the FT. Conversely, in the polluted cases most of the particles are confined to the bottom of the MBL, dominating the columnar AOD.

In the case of polluted MBL topped by a dusty layer (e.g. TF23), the results suggest that the MBL and FT contribute almost equally to the total AOD (44.7 and 55.3%, respectively). However, inspection of the vertical aerosol extinction plot makes it clear that the dusty layer has a larger aerosol burden than the polluted layer below, and should, therefore, account for a larger fraction of the columnar AOD.

Estimates using data from the collocated AATS-14 sun photometer (e.g. Schmid et al., 2000) show that the dusty layer contributes more than the polluted MBL to the total AOD (82 and 18%, respectively). Differences in AOD values for *in situ* and sun photometer measurements have been previously reported for case studies of ACE-2. Schmid et al. (2000) and Collins et al. (2000) have reported lower AOD values for *in situ* data than those acquired with sun photometer measurements. In their analysis, the degree of agreement between the derived and directly measured optical properties varied for different types of airmasses. Possible reasons for these differences are speculative, but it is thought that only a subset of the full population of particles reached the nephelometer and PSAP because particles larger than $2.5 \mu\text{m}$ were removed by the cyclone. Accuracy in the measurement of RH is another relevant factor accounting for a precise calculation of ambient scattering coefficient. At high RH, a small increase in the ambient RH leads to relatively large water uptake for the particles. This is especially important in the boundary layer where RH is typically higher than in the free troposphere and where most of the particle mass is normally found. For an accurate adjustment to the ambient RH, knowledge of the nature of the sampled particles is required. The coefficients used in eq. (6) were derived based on the assumption that the particles were internally mixed and no distinction is made among the different hygroscopicity of the different multicomponent aerosols.

These results concur with those of other studies (e.g. Hegg et al., 1996; Sheridan and Ogren, 1999) which showed that most aerosol forcing occurs in the lowest 900 m of the atmosphere (unless there are thick layers aloft), and individual vertical pro-

files are not always representative of the overall properties of the column.

5. Summary and conclusions

A survey of aerosol optical properties has been undertaken using data from the Aerosol Characterization Experiment (ACE-2) carried out in July 1997 in the northeast Atlantic Ocean. The main objective was to characterize the optical properties in a marine environment influenced by aged, polluted aerosol transported from Europe and dusty airmasses from North Africa.

The clean airmasses investigated had characteristics typical of clean marine aerosols, with the highest particle concentrations at the bottom of the layer and decreasing with height, as indicated by the measurements of aerosol scattering coefficient σ_{sp} . It was possible therefore to describe σ_{sp} in a more general sense, as depicted by the aggregate plot of the vertical structure of this property. The polluted airmasses showed a large degree of variability between cases, and because of this a general description of this category was not possible. The values of σ_{sp} in these cases were strongly dependent upon the strength of the pollution outbreak, transport time and the degree of mixing and transformation of the aerosols before arriving at the measurement point. Similarly, the dusty cases showed a large degree of variability and stratification.

The variability of median absorption coefficient σ_{ap} was surprisingly small throughout the same air column, and also when compared between flights. Whereas σ_{sp} followed the vertical distribution of the airmasses, the variation of σ_{ap} in connection with airmass type was not as evident. We attribute this to the almost invariant BC concentration during the experiment. During background conditions nssSO_4^{2-} , OC, NH_4^+ and sea salt were the major components, while BC accounted for less than 1% of the total mass. During pollution outbreaks, there was an enhanced contribution of nssSO_4^{2-} and NH_4^+ but little increase in the BC mass (up to 2%), yielding thus an increase of σ_{sp} but lack of variability and relatively low σ_{ap} .

Spatial variability of the FT aerosol at scales much lower than the airmass scale has been identified with values between 3 and 8 km (for an autocorrelation function $r > 0.8$). Sporadic vertical intrusion of aerosol from the boundary layer into the FT was thought to create perturbations in the aerosol field and might thus be one of the possible mechanisms able to create such variability.

The contribution from the MBL and FT to the total AOD was calculated using profile data flown within clean, polluted and dust-laden airmasses. The results showed that the clean and polluted MBL contribute more to the column radiative balance than the FT. However, in the cases when the clean marine aerosols expanded from the bottom of the MBL into the FT, the contribution of the latter to the total AOD was not negligible. In the polluted cases, most of the particles were confined to the bottom of the MBL, dominating the columnar AOD. In the cases of polluted MBL topped by a dusty layer, the results revealed that

the MBL and FT contributed almost equally to the total AOD (44.7 and 55.3%, respectively). These results do not agree with AOD estimated with a collocated sun photometer, which revealed that the dusty layer contributed more than the polluted MBL to the total AOD (82 and 18%, respectively). Losses of particles within the cyclone and accuracy of the RH measurements are some of the factors thought to account for such discrepancies.

The intensive properties (Ångström coefficient and single scattering albedo) had little variability within a given airmass and could be described in a more general sense. A key result in this study was the relationship between ω_0 and transport time of polluted airmasses. In the first episode studied there was a decrease in the values of ω_0 (from ~ 0.96 to ~ 0.92) with increasing transport time (from 42 to 54 h). The decrease in the value of ω_0 is related to the removal of scattering material and may have occurred by the reduction of aerosol concentration in the MBL through dilution with cleaner free tropospheric air as the mixing layer deepened, and, to a lesser extent, coagulation. In the second pollution outbreak there was not much difference between the median ω_0 for samples taken 60 and 72 h after last contact with the European coast. The longer contact this airmass had with anthropogenic sources may have contributed to the preservation of the scattering characteristics of the plume. A larger data set is, however, necessary to establish to what extent the values of ω_0 vary with transport time and possibly to formulate a simple way to describe the time evolution of this quantity.

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