Observations of aerosols in the free troposphere and marine boundary layer of the subtropical Northeast Atlantic: Discussion of processes determining their size distribution

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Abstract. During July 1994, submicron aerosol size distributions were measured at two sites on Tenerife, Canary Islands. One station was located in the free troposphere (FT), the other in the marine boundary layer (MBL). Transport toward these two sites was strongly decoupled: the FT was first affected by dust and sulfate-laden air masses advecting from North Africa and later by clean air masses originating over the North Atlantic, whereas the MBL was always subject to the northeasterly trade wind circulation. In the FT the submicron aerosol distribution was predominantly monomodal with a geometric mean diameter of 120 nm and 55 nm during dusty and clean conditions, respectively. The relatively small diameter during the clean conditions indicates that the aerosol originated in the upper troposphere rather than over continental areas or in the lower stratosphere. During dusty conditions the physical and chemical properties of the submicron aerosol suggest that it has an anthropogenic origin over southern Europe and that it remains largely externally mixed with the supermicron mineral dust particles during its transport over North Africa to Tenerife. Apart from synoptic variations, a strong diurnal variation in the aerosol size distribution is observed at the FT site, characterized by a strong daytime mode of ultrafine particles. This is interpreted as being the result of photo-induced nucleation in the upslope winds, which are perturbed by anthropogenic and biogenic emissions on the island. No evidence was found for nucleation occurring in the undisturbed FT. The MBL site was not strongly affected by European pollution during the period of the measurements. The MBL aerosol size distribution was bimodal, but the relative concentration of Aitken and accumulation mode varied strongly. The accumulation mode can be related to cloud processing of the Aitken mode but also to pollution aerosol which was advected within the MBL or entrained from the FT. No bursts of nucleation were observed within the MBL.

1. Introduction

Subtropical oceanic atmospheres, adjacent to the western parts of the continents, are characterized by a distinct temperature inversion. This is a consequence of heating by compression of the subsiding air over a cold upwelling ocean. The inversion tops the marine boundary layer (MBL) at altitudes that range between 500 m, close to the center of the subtropical high, and 2000 m in the trade-wind region closer to the tropics. When the condensation level is lower than the inversion level, water vapor will condense on the available aerosol particles to form stratiform clouds. Because of the large scale of subtropical subsidence, the MBL stratiform clouds can extend over very large areas and cover 30% of the

world's oceans [International Satellite Cloud Climatology Project (ISCCP), 1992]. Charlson et al. [1987, 1992] drew attention to the possible role of these clouds in climate regulation and climate change. Assuming a constant liquid water content, these authors estimated that a 30% change in the cloud droplet number concentration of MBL stratus clouds could lead to a global-mean radiative forcing of -2 W/m^2.

Cloud droplets form on a fraction of the aerosol particles, called cloud condensation nuclei (CCN). The estimates by Charlson et al. have therefore focused a lot of aerosol research on the origin and behavior of MBL aerosols and CCN. Even for the simplest case of the clean MBL, where sea salt and sulfate derived from dimethylsulfide (DMS) are the major aerosol constituents, there is not yet an agreement regarding the following two issues: (1) What is the relative contribution of sea salt and DMS-derived particles to the total number concentration of MBL CCN? (2) Is the number of DMS-derived aerosol particles controlled by nucleation of condensable species within the MBL, or are they formed in the free troposphere (FT) and subsequently entrained into the MBL? In case of a polluted MBL, aerosol particles and CCN are provided by advection from the continents. Here the following question may be raised: (3) What is the relative importance of transport within the MBL, and long-range transport in the FT with subsequent entrainment, as a way of perturbing the MBL and its clouds? All these questions have

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to do with how to describe aerosol and, eventually, cloud droplet number concentrations in climate models; whether to focus more on transport issues, on atmospheric chemistry and aerosol dynamics, or on primary aerosol sources.

A quantitative answer to the questions will eventually require Lagrangian studies in order to untangle aerosol processes from transport processes, as well as long-term measurements yielding meaningful statistics about the type of conditions or transport patterns that are prevailing. However, simultaneous characterization of the aerosol in the MBL and in the overlying FT should also give insights in the processes that are possibly involved. With this in mind, we have performed measurements on Tenerife, an island located in the NE subtropical Atlantic, where we operated two stations, one in the MBL and one in the free troposphere (FT). During summer, both stations are subject to clean maritime conditions and to outbreaks of anthropogenic aerosols from Europe and mineral dust from northern Africa.

In this paper we focus on measurements of the size distribution of submicron aerosols and attempt to explain the observed variability (and regularities) in terms of aerosol dynamic processes and transport patterns.

2. Sites and Meteorology

Measurements were performed during July 1994, at two sites on Tenerife (Canary Islands, 16° 30' W, 28° 18' N): Punta del Hidalgo (PDH), (50 m above sea level (asl)) and Izaña (IZO), (2360 m asl).

Punta del Hidalgo (PDH)

Measurements were taken from the top of a 50 m high lighthouse. It is located on the shore of the NE extremity of the island and is directly exposed to the northeasterly trade winds. PDH is a new site and no climatology of trajectories exists. Calculations of 5 days back trajectories during July 1994 (see Figure 1a), 1995 and 1996 show that during this month the site is usually impacted by air masses subsiding around the high of the Azores. Often, these air masses will pass close to the Iberian Peninsula or even have their origin over western and central Europe, transporting enhanced levels of anthropogenic pollution through the MBL to Tenerife. On 1 out of 90 days, air masses in the boundary layer came straight from Africa.

Izaña (IZO)

Measurements were taken at the Instituto Nacional de Meteorologia (INM) - Global Atmosphere Watch (GAW) Observatory, which is located at 2360 asl, on the mountain ridge that crosses the NE part of the island. Climatologies of back trajectories [Sancho et al., 1992, Merrill, 1994, Cuevas et al., 1996] show that in summer the site is mainly impacted by air subsiding from the mid and upper troposphere over the North Atlantic and by continental air masses from northern Africa. Frequently, the latter have their origin over the Mediterranean or central Europe.

Radio soundings from Santa Cruz de Tenerife show that during July the subsidence temperature inversion is located between 1000 and 1500 m. This inversion separates the MBL air masses arriving at PDH from the FT air masses arriving at IZO. However, during daytime, when the (volcanic, dark) island is heated, upslope winds will bring air

from lower levels and possibly from below the inversion to IZO. Urban development and industry along the coast of the island as well as biogenic emission from the forests on the mountain up to 1900 m strongly alter the composition of the upslope winds, compared to what is sampled at PDH.

For the period discussed in this paper (July 7-18, 1994), back trajectories were calculated with a variety of models: two three-dimensional (3-D) models based on the European Centre for Medium Range Weather Forecast (ECMWF) and the National Meteorological Center (NMC) wind fields respectively, an isobaric model based on the United Kingdom Meteorological Office (UKMO) model and isentropic trajectories based on NMC wind and thermodynamic data. They all show the same general picture, however relevant differences also exist. Here we will show only the Royal Netherlands Meteorological Institute (KNMI) trajectories (Figure 1). They are based on ECMWF wind fields, which have a horizontal resolution of 0.5°, and are available every 6 hours. The relatively good performance of the ECMWF model over ocean regions has been demonstrated by Fuelberg et al. [1996]. The sensitivity of the KNMI trajectories to time and spatial resolution showed that a 6 hour, respectively, 1° x 1° resolution is largely sufficient to describe 3-D air mass trajectories in synoptic systems [Scheele et al., 1996].

In Punta del Hidalgo, air masses were mainly from the open Atlantic, except between July 11 (0600) and July 13 (0600) when they traveled closer, or according to the isobaric UKMO model, over the Iberian coast (F. McGovern, private communication, 1997; see also Figure 1a). Izaña, on the contrary was influenced by European-African air between July 8 and 14, followed by midtropospheric Atlantic air between July 15 and 18 (see Figure 1b and 1c). This analysis of the origin of air masses is confirmed by the chemical composition and loadings of the aerosol: mainly clean air at PDH and dust followed by clean air at IZO (F. McGovern, private communication, 1997). In this paper we will focus on the size distribution of the submicron aerosol, and collate them with the transport patterns and aerosol processes.

3. Instrumentation

At both sites, aerosol size distributions were obtained with identical differential mobility particle sizers (TSI DMPS model 3932) operating in the size range between 16 and 620 nm and giving a distribution every 20 min. The two systems were intercompared side by side using laboratory aerosol. The number concentrations resulting from integrating the number distribution were equal within 10%, and the geometric mean diameters of the distribution were within 5%. The DMPS at PDH failed after July 17.

Apart from the TSI DMPS, a second system was used at IZO, consisting of a short and medium length Vienna-type differential mobility analyzer (DMA), [Winklmayer, 1987] operated in parallel. The short, ultrafine DMA (UFDMA) was operated with a TSI 3025A particle counter, while the medium DMA (MEDMA) used a TSI 3010 counter. The size ranges scanned by he UFDMA and MEDMA were 3.5-20 nm and 20-620 nm, respectively. Both scans and hence the complete measurement from 3.5 to 620 nm is obtained every 15 min. During the campaign, the "Twin DMA" and TSI DMPS gave, in the size range of overlap, integrated number concentrations within 20% and geometric mean diameters within 5%. After July 19 the UFDMA malfunctioned. For the
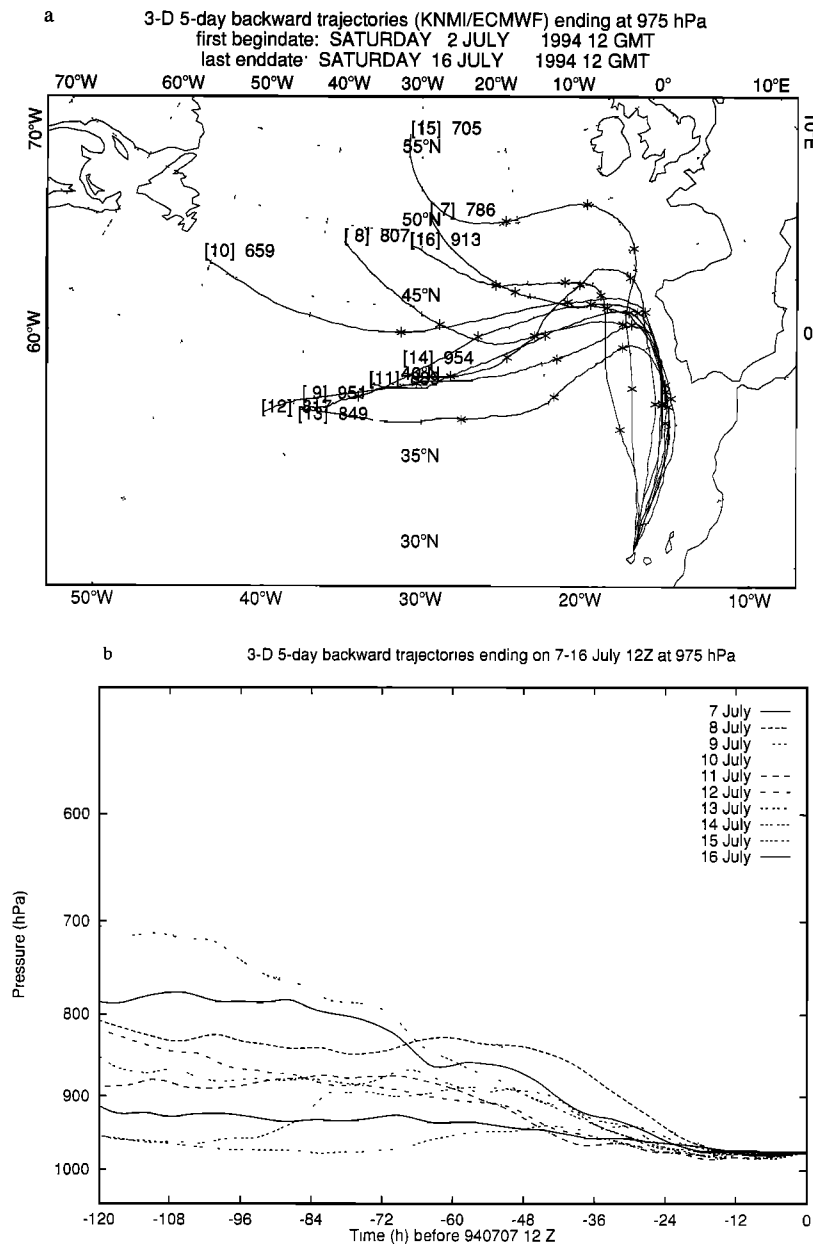
Figure 1a. Compilation of 5 days back trajectories ending at Tenerife, at 975 hPa. (Punta del Hidalgo (PDH)), at 1200 UTC between July 7 and 16 1994. (a) Horizontal projection: the numbers [xx] yyy given at the beginning of each trajectory indicate the end day (xx) and the pressure level (yyy) where they start. (b) vertical projection.

times and size classes where both IZO systems yielded valid data, they are presented in what follows as averages from the two systems. In other cases, data from one system are taken into account. At PDH, size distributions were measured at 40-50% RH, whereas at IZO the RH was always lower than 25%. These relative humidities were obtained from those recorded outside, taking into account the higher temperature in the instrumentation room. In what follows the shown size distributions can therefore be considered as those of the aerosol in the dry state.

Other measurements that will be used for interpreting the size distribution measurements are as follows: (1) at IZO: surface ozone, measured with a Dasibi UV absorption instrument, CO_2, with a nondispersive infrared Ultramat-2 analyzer, dry and wet temperature, and global radiation with a

Kipp and Zonen pyranometer; (2) vertical information of temperature, humidity, and wind over Tenerife, from sea level to 30 km height, from rawinsondes, released twice a day (0000 and 1200 UTC) at Santa Cruz de Tenerife; (3) National Oceanographic and Atmospheric Administration Advanced Very High Resolution Radiometer (NOAA/AVHRR) images over the NE Atlantic, obtained from the archive of the Dundee satellite receiving station (http://www.sat.dundee.ac.uk/).

4. Observations

Cloudiness From AVHRR Images

The images are taken at the receiving station of Dundee (United Kingdom) in the late afternoon. They cover the NE
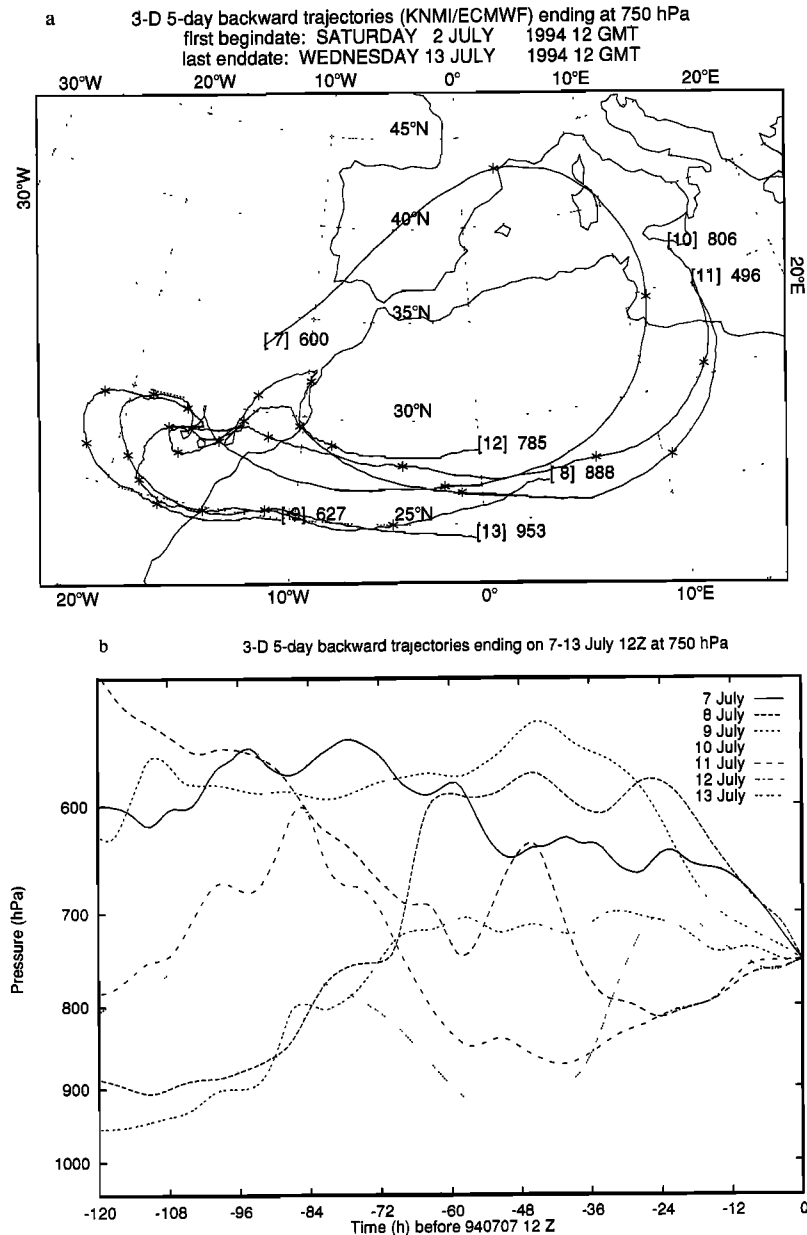
Figure 1b. Compilation of 5 days back trajectories ending at Tenerife, at 750 hPa (Izaña Station (IZO)), at 1200 UTC between July 7 and 13 1994, i.e., when dust was observed at IZO. (a) Horizontal projection, (b) vertical projection.

Atlantic and western Europe, and include the Canary Islands at their bottom end. The images pertaining to our measuring period were collected and can be seen at the WWW site http://rea.ei.jrc.it/~vandinge/sat94/sat94.html. By visually subtracting channel 2 (VIS) and channel 4 (IR) images, a rough impression of low-level cloudiness can be obtained. Detailed analysis of these satellite data is outside the scope of this study.

During the period discussed here, the cloudiness over the NE Atlantic was rather complex. We focus on the area between the Canary Islands and the coast of Portugal, i.e., the area crossed by the MBL air parcels during the last 36-48 hours before reaching Tenerife. This area is generally free of low-level clouds on July 8, 9, 10, and 11. On July 12, low-level clouds are present immediately upwind of Tenerife and

from July 13 onward the cloudiness becomes complex, with low-level clouds immediately north of the Canary Islands and often high-level, probably convective clouds near and over Portugal.

Structure of the Atmosphere above Tenerife

Plate 1a shows, as a 2-D contour plot, the time history of the vertical profile of the dew point, obtained from radio soundings at Santa Cruz de Tenerife. Santa Cruz is located at the lee side of the NE mountain ridge, hence protected from the trade winds. The first 600 m of the radio soundings are therefore not representative for the MBL upwind of Tenerife.

The soundings show the humid MBL extending up to 1300 m. The second clear structure is the extremely dry layer above 6000 m. The layer in between, where IZO is located, shows
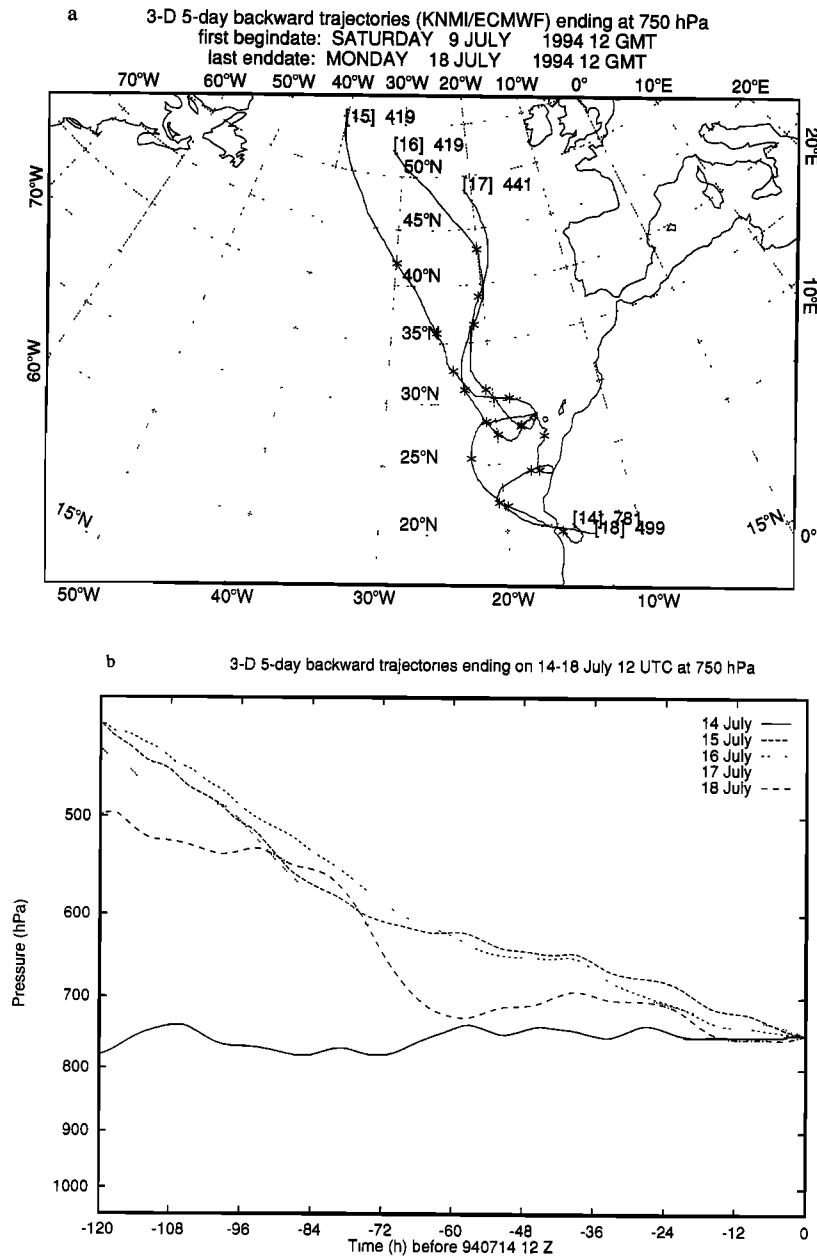
a   3-D 5-day backward trajectories (KNMI/ECMWF) ending at 750 hPa
first begindate: SATURDAY 9 JULY   1994 12 GMT
last enddate: MONDAY 18 JULY   1994 12 GMT

[15] 419
[16] 419
50°N
[17] 441
45°N
40°N
35°N
30°N
25°N
20°N
[14] 78
[18] 499

b   3-D 5-day backward trajectories ending on 14-18 July 12 UTC at 750 hPa

14 July ——
15 July ----
16 July ·· ·
17 July
18 July - - -

Pressure (hPa)

Time (h) before 940714 12 Z

Figure 1c. Compilation of 5 days back trajectories ending at Tenerife, at 750 hPa (IZO), at 1200 UTC between July 14 and 18 1994, i.e. when clean air was observed at IZO. (a) Horizontal projection, (b) vertical projection.

large variability in time and with altitude. The station is impacted by intrusions of dry upper tropospheric air on the night between July 10 and 11 and more clearly between July 14 and 17. During these intrusions a distinct layering is observed, and IZO is representative only for a relatively small portion of the free troposphere. During the other periods, which coincide with dust outbreaks from northern Africa, the free troposphere is more homogeneous, at least with respect to its thermodynamic properties.

FT Size Distributions at Izaña (Nighttime)

A good picture of the evolution and variability of the size distributions is given by the 2-D contour plot (Plate 1b), showing $dN/d\log(D_p)$ as function of D_p and time, for the

whole measuring period. The strong diurnal variations, with high concentrations of small particles during daytime stand out clearly. We will first consider the nighttime measurements, because they are least disturbed by upslope conditions and represent best the FT. A clearer view of the shape of the corresponding size distributions is given in Figures 2a and 2b, as a frequency plot of $dN/d\log(D_p)$ versus D_p The averages of the modal parameters of the distributions are given in Table 1.

A first observation is that FT size distributions are predominantly monomodal. This can be explained by the absence of cloud processing and the long residence times in the FT, during which coagulation will reduce any initial distribution into a monomodal one. The absence of cloud
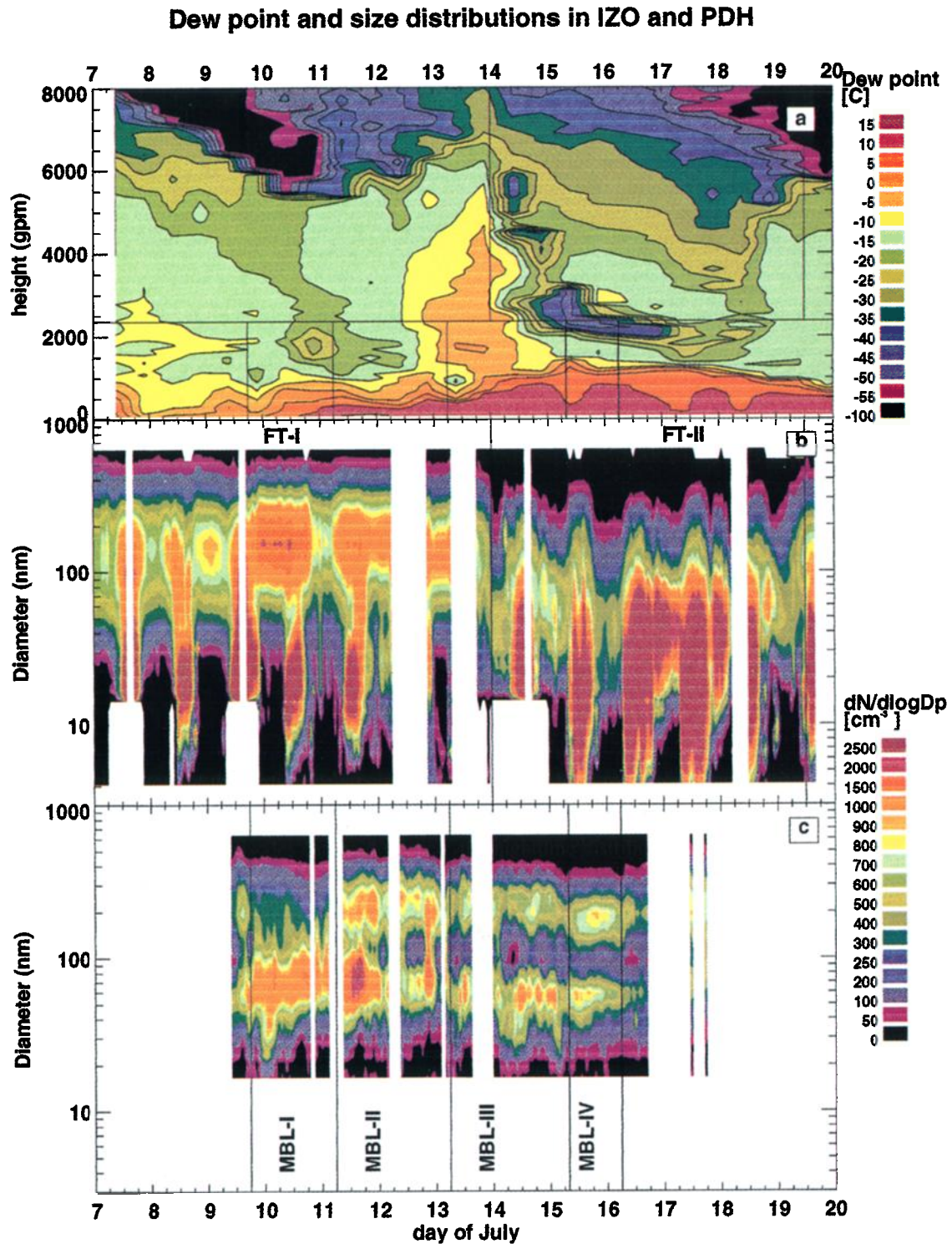
Dew point and size distributions in IZO and PDH

Plate 1. (a) Time history of the vertical dew point profile, obtained from radio soundings released twice a day at Santa Cruz de Tenerife. The horizontal line at 2360 m indicates the location of the Izaña Station (IZO). (b) Time history of the submicron size distribution measured at Izaña (IZO). The size distributions switch between unimodal during nighttime and bimodal during daytime. Two episodes can be discerned: the nighttime distributions change abruptly on July 14 (0000), which coincides with the arrival of dry upper tropospheric air at the station. (c) Time history of the submicron size distribution at Punta del Hidalgo (PDH) 50 m above sea level. The size distribution is always bimodal, with strong variations however in the relative concentrations of the accumulation mode and the Aitken mode. Four episodes can be discerned and are discussed in the text.

processing in the subtropical FT is supported by the ISCCP data, which show that the frequency of medium and high clouds in this area is typically $< 10\%$. It is worth noticing, however, that according to theory, coagulation eventually results in lognormal distributions with a geometric standard deviation of 1.4 [Lai et al., 1972], whereas coagulation plus condensation would result in even narrower size distributions. Here the observed value is around 1.9!

As mentioned before, all back trajectory models show that before July 14 the air masses arriving at IZO passed over the Sahara and, with some exceptions, over the Mediterranean before passing the Sahara. During the course of July 14, the transport pattern changed and by July 15 trajectories originated in the middle atmosphere above the North Atlantic. Contrary to this gradual change suggested by the trajectories and by the dewpoint (Figure 2a), the composition of the atmosphere changed more abruptly at July 14 0000. The implication of this change from dusty to clean conditions is a dramatic decrease of the geometric mean diameter of the submicron aerosol from 117 nm to 55 nm. We now discuss the dusty and clean episodes in more detail.

Episode FT-I, July 7, 0000 to July 14, 0000 (Figure 2a). The average diameter of the aerosol is fairly constant (117 ± 13 nm, see Table 1), but the number concentration is more variable. This variability can be related to the large variability in horizontal and vertical transport over northern Africa (Figure 1b). d'Almeida and Schutz [1983] report that during desert dust storms particles in the range 0.1-0.2 µm D_p can reach number concentrations up to 1500 cm^{-3}. It is therefore possible that the observed 400-600 cm^{-3} submicron particles at IZO are mineral dust particles. However, several other observations support the possibility that the submicron mode consist of aged anthropogenic sulfate exported from southern Europe: (1) some of the back trajectories show transport out of southern Europe (Figure 1b). (2) the chemical analysis of the aerosol at IZO shows that during dust events, sulfate loadings are usually high [Prospero et al., 1995], that the majority of this sulfate mass is in the submicron fraction, and that the Ca^{++}/SO_4^{-} ratio is smaller in the submicron than in the supermicron fraction, suggesting different origins for these fractions (D. Savoie and J.P. Putaud, personal communications. 1997), (3) during the episode discussed here, the regression of nighttime (12 hour averages) submicron aerosol volume on sulfate yields $r^2 = 0.84$, suggesting that most of the sulfate was indeed in the submicron fraction.

The decrease of the number concentration and appearance of smaller particles at July 11 0000, coincides with drier air around Tenerife (see Plate 1a), which is likely to be a remainder of air transported from higher altitudes, as is shown by the back trajectory ending on July 11 1200. Such downward transport was more clear after July 14.

Episode FT-II, July 14, 0000 to July 18, 1200 (Figure 2b). The sudden decrease in the particle size coincides with a decrease in water vapor and a strong increase with O_3 (see Figure 3c). Plate 1a shows that a layer of extremely dry air is subsiding over Tenerife, and the trajectories show that this layer has its origin in the middle troposphere. The monomodality and relatively small average diameter of the aerosol distribution show that the aerosol is relatively fresh and formed by nucleation and subsequent condensation and coagulation higher up in the troposphere. An aged aerosol
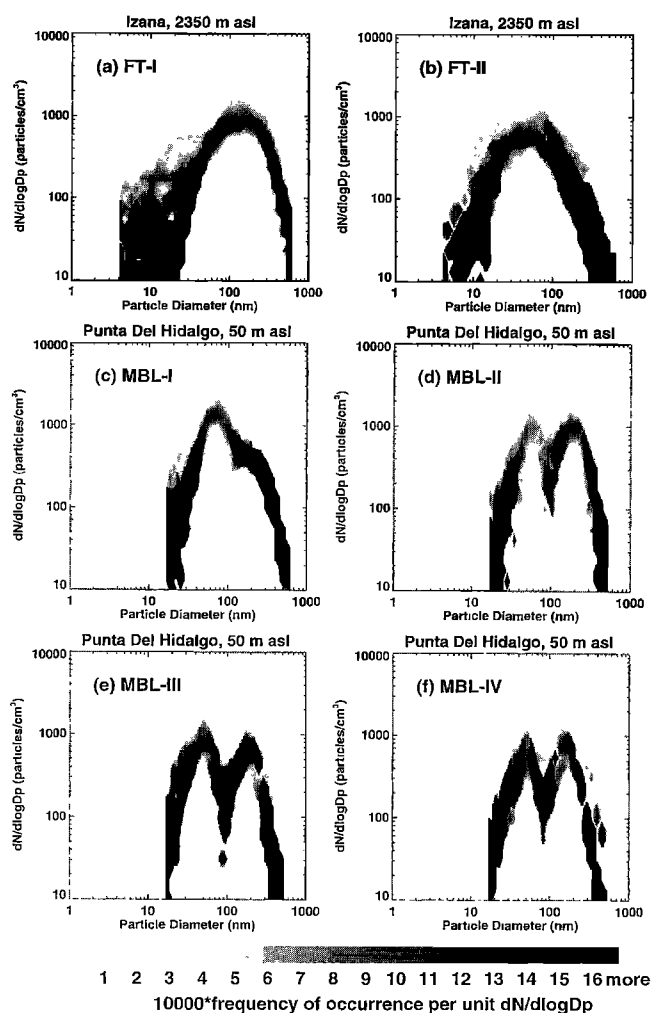

Figure 2. Frequency plots of aerosol size distributions in the free troposphere (IZO) (a, b), and in the marine boundary layer (PDH) (c, d, e, f) during the episodes discussed in the text.

originating over the continents or in the stratosphere would indeed show geometric diameters in the range 100-200 nm. The values of all parameters vary distinctly from one night to the other. For the first three nights this is explained by IZO being inside or outside the shallow subsiding dry layer (Plate 1a). During the nights of July 16-17 and July 17-18 the particle concentration remains exceptionally high, which is explained by daytime aerosol that is staying around the island at the level of IZO (see next section).

Table 1. Averaged FT Aerosol Properties, Based on Fits of One Lognormal Distribution to the Measured Distributions

Episode	Type	N, cm^{-3} STP	D_{pg}, nm	σ_g
FT-I, night	dust	570 (70)	117 (13)	1 95 (0 2)
FT-II, night*	clean	425 (115)	53 (8)	1 89 (0 1)

FT, free troposphere; STP, standard temperature and pressure
Averages, excluding particles $D_p < 15$ nm and $D_p > 620$ nm.
Values in parentheses: 1 standard deviation
*excluding nights of July 16-17 and 17-18 (see below).

Diurnal Variation of the Aerosol at IZO

The most prominent feature of the aerosol at IZO is the diurnal variation in the number concentration and size distribution of the aerosol. This diurnal variation in particle concentration has been noticed before. In fact, diurnal variations have been documented for most of the atmospheric constituents measured at IZO. They are explained by upslope winds that evolve during daytime and that bring air from lower levels and probably from within the MBL to the observatory. It has also been noticed before that during dusty conditions the upslope winds are weaker, and IZO is less influenced by air at lower levels [*Cuevas et al.*, 1991]. Measurements of humidity, ozone. and CO_2 during the period discussed here are shown in Figure 3 and confirm the previous analyses. For what concerns aerosol particles, our measurements show for the first time that the diurnal variation in number concentration is predominantly due to an increase in the ultrafine particles (Plate 1b, Figure 3d). During clean conditions (FT-II), nucleation seems to happen in the upslope air near IZO, since particles were detected down at the lower detection limit of 3 nm. During dusty conditions (FT-I), nucleation seems to have happened and subsequently quenched in the upslope air before it arrived at IZO, since the smallest particles detected were significantly larger than in clean conditions. This difference can be explained by the known slower upslope transport during dusty conditions, and/or by quenching of nucleation when the upslope air mixes with the dust layer.
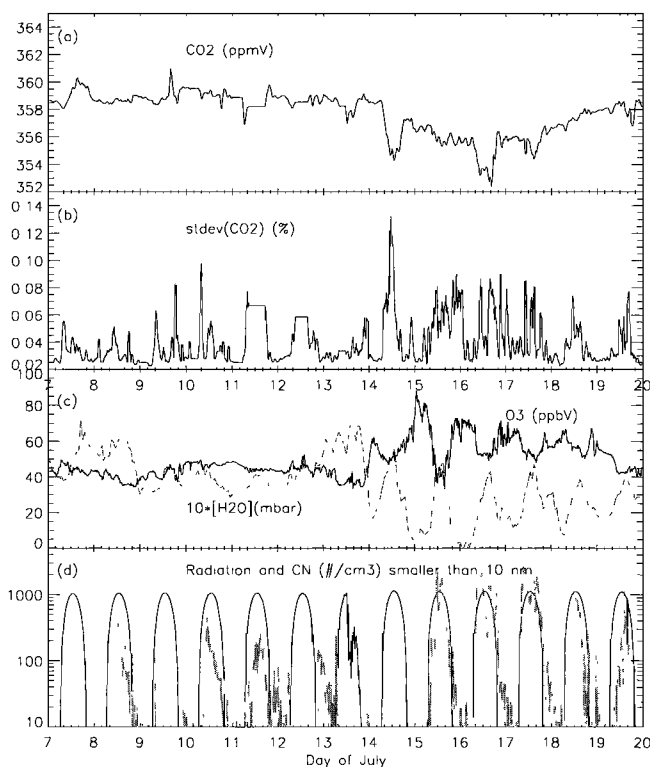

Figure 3. Diurnal variations of a variety of parameters measured at IZO observatory, documenting the upslope transport of air during daytime: (a) half hourly CO_2 values (ppmv) and (b) corresponding standard deviation (%), (c) ozone and water vapor, (d) global radiation and aerosol particles smaller than 10 nm.

Although the observed nucleation is most likely a local phenomenon, induced by the photochemistry in the perturbed air masses reaching IZO, it must be looked at with care since it might mask in situ nucleation over large areas in the undisturbed free troposphere at the altitude of IZO. There is no evidence in our data set that the latter is the case. The ultrafine particles never appeared before humidity, ozone or CO_2 signaled the arrival of upslope winds at IZO, and they usually disappeared abruptly after the sun sets and the upslope wind collapses. The latter is true except on the evenings of July 16 and July 17 (see Plate 1b). During these nights the winds at IZO drop below 2 m/s, and the back trajectories show local transport near Tenerife. It is therefore likely that the air that has been pumped up during the previous days is lingering on during nighttime.

MBL Size Distributions at Punta del Hidalgo

Plate 1c shows the time history of the aerosol size distributions at Punta del Hidalgo, and Figures 2c, 2d, 2e and 2f the corresponding distributions as frequency plots. Table 2 gives the average modal parameters of the aerosol.

An immediate observation is that the MBL size distribution is bimodal most of the time, showing an Aitken mode around 60 nm D_p and an accumulation mode around 200 nm D_p. High concentrations of ultrafine particles are not apparent, showing that bursts of nucleation did not occur in the MBL upwind of and close to PDH. This lack of nucleation bursts raises the problem of the origin of the Aitken mode particles, which we will discuss later. The accumulation mode is commonly explained by the cycling and chemical processing of the Aitken mode aerosols in the stratiform cloud deck [*Hoppel et al.*, 1986, 1994]. However this explanation does not seem to apply to the accumulation modes observed before July 12, when AVHRR images do not show obvious low-level clouds upwind of Tenerife. This and the generally large variation in the relative importance of Aitken and accumulation mode asks for a closer look into the origin and behavior of these modes. We have used the KNMI model to calculate 5 days back trajectories ending in the MBL near Tenerife at 900, 925, 950, and 975 hPa. In the discussion below we will only refer to the 925 and 975 trajectories. The trajectory analysis and the patterns seen in Plate 1c allow identification of four episodes.

Episode MBL-I, July 9, 1800 to July 11, 0600 (Figure 2c). The distributions are characterized by a sharp Aitken mode around 60-70 nm. The 925 and 975 trajectories ending on July 10 1200 originated five days earlier in the middle troposphere at 560 and 659 hPa, respectively. They reached the inversion (taken as the 900 hPa level) at about 40°N. 24 to 36 hours prior to arrival in PDH. On the basis of these trajectories an average subsidence velocity of 0.8-1.0 cm/s can be inferred, which is within the range of entrainment velocities determined during the Atlantic Stratocumulus Transition Experiment (ASTEX): i.e., 0.3-1.0 cm/s [*Bretherton et al.*, 1995]. It is therefore likely that FT air entered and mixed with the MBL without much resistance. The most straightforward explanation for the MBL Aitken mode is therefore that it originated from the preexisting FT aerosol that entered the MBL and evolved by condensational growth. Condensational growth would increase the size of the aerosol but also make the size distribution narrower, in particular when particles are smaller than the mean free path

Table 2. Average MBL Aerosol Properties, Based on Fits of Two Lognormal Distributions to the Measured Distributions

| Episode | Type | Aitken Mode | | | Accumulation Mode | | |
		N, cm^{-3} STP	D_{pg}, nm	σ_g	N, cm^{-3} STP	D_{pg} nm	σ_g
MBL-I	mixed	319 (152)	66 (9)	1.38 (0.16)	349 (115)	120 (40)	2.00 (0.28)
MBL-II	mixed	447 (327)	65 (7)	1 41 (0.12)	325 (73)	217 (14)	1 38 (0.08)
MBL-III	clean	321 (169)	52 (6)	1 47 (0.09)	207 (51)	209 (16)	1.35 (0.07)
MBL-IV	clean	228 (44)	52 (2)	1.40 (0 05)	249 (23)	178 (5)	1.34 (0.02)

MBL, marine boundary layer
Values in parentheses: 1 standard deviation.

length of the condensing molecules, as is the case here [*Friedlander*, 1977, p. 253]. This would explain the significantly lower value of the geometric standard deviation of the MBL Aitken mode (1.38 ± 0.16) compared to that of a clean FT aerosol (1.89 ± 0.1). We cannot exclude nucleation occurring upwind of Tenerife, during the initial mixing of dry FT air with humid MBL air [e.g. *Hegg et al.*, 1992] and that the freshly nucleated aerosol evolved and contributed to the Aitken mode. Apart from the sharp Aitken mode, a broad accumulation mode is also present (see Figure 2c). Cloud processing is not a viable explanation, since no low-level clouds were present between the point where the trajectories entered the MBL and PDH. The FT aloft, however, was heavily laden with aerosol, which we interpreted as anthropogenic sulfate. The AVHRR pictures and back trajectory analysis show that this FT aerosol extends NE of Tenerife toward Portugal. It is thus possible that entrainment has partially flushed the clean MBL with the aerosol from the FT, and that the latter is at the origin of the observed accumulation mode. This scenario is supported by the correspondence between the geometric mean diameter and standard deviation of the MBL accumulation mode particles and the FT aerosol aloft. A narrowing of the size distribution is indeed not expected since the particles are now larger than the mean free path length. The scenario is also supported by the chemical analysis of the MBL aerosol, which shows traces of mineral dust (D. Savoie, personal communication, 1996).

Episode MBL-II, July 11, 0600 to July 13, 0600 (Figure 2d). Although low-level clouds are still not clearly present upwind of PDH, the aerosol distributions are characterized by prominent Aitken and accumulation modes. The 925 trajectory ending on July 11 1200 still originated at 580 hPa, and could still explain the large Aitken mode. The 975 trajectory, on the other hand, remained below the 800 hPa level during the previous 5 days (see Figure 4). Although the latter trajectory remains over the ocean, the isobaric 1000 hPa trajectory of the UKMO model was located more eastward and passed over the Portuguese coast. This situation must have resulted in mixing of clean FT air with polluted BL air, since black carbon and sulfate concentrations in PDH were found to be higher during this period (McGovern, personal communication, 1997). Anthropogenic aerosols rapidly develop an accumulation mode [e.g. *Raes et al.*, 1995], so the bimodal distribution during episode MBL-II can be explained as the result of mixing of two air masses: one with a prominent Aitken and one with a prominent accumulation

mode. Both modes hide the contribution of FT aerosol entrained from aloft, which according to the aerosol chemical analysis is still present. The trajectories ending on July 12 1200 show a similar pattern. During the afternoon of July 12 low clouds are present just upwind of Tenerife. so that the aerosol observed at PDH could have been minimally cloud processed.

Episode MBL-III, July 13, 0600 to July 15, 0800 (Figure 2e). The distributions are characterized by prominent Aitken and accumulation modes. The 975 trajectories ending on July 13 and July 14 1200 stayed below the 850 hPa level all the time, whereas the 925 trajectories stayed below the 740 hPa level. The trajectories are passing farther away from the Iberian coast. and the chemical measurements show minimal anthropogenic impact. The air masses ending at PDH thus stayed predominantly within an unperturbed MBL for more than 5 days, and cloud processing has likely been the main process shaping the bimodal size distribution. The short time variability in the Aitken mode must be explained by variability in the source of this mode. The variability in the accumulation mode is probably due to intermittent precipitation in the air mass. Hence episode MBL-III corresponds best to the classical picture of an undisturbed MBL, in which there is, on the average, a balance between the source of the Aitken mode particles, cloud processing to create the accumulation mode, and wet removal of the accumulation mode particles.

Episode MBL-IV, July 15, 0800 to July 16, 0600 (Figure 2f). There is again a prominent Aitken and accumulation mode, but the modal parameters changed slightly. This coincides with another change in mainly vertical transport. The 925 and 975 back trajectories ending on July 15 1200 originated again in the middle troposphere, at 520 and 700 hPa respectively, and enter the MBL 24- 36 hours upwind of Tenerife. The situation is very similar to Episode MBL-I, except for the clear presence of low level clouds along the trajectories toward PDH. It is most likely that cloud processing has eroded the aerosol entering the MBL to create the typical MBL Aitken and accumulation modes.

5. Discussion

FT

Mixing of dust with sulfate aerosols at IZO. The likelihood that the dust plume observed at IZO is mixed with sulfate from Europe is of interest, since the degree of

a

3-D 5-day backward trajectories (KNMI/ECMWF)
begindate: WEDNESDAY 6 JULY   1994 12 GMT
enddate: MONDAY   11 JULY   1994 12 GMT

b   3-D 5-day backward trajectories ending at 940711 12 Z

900 hPa ——
925 hPa – – –
950 hPa ·····
975 hPa

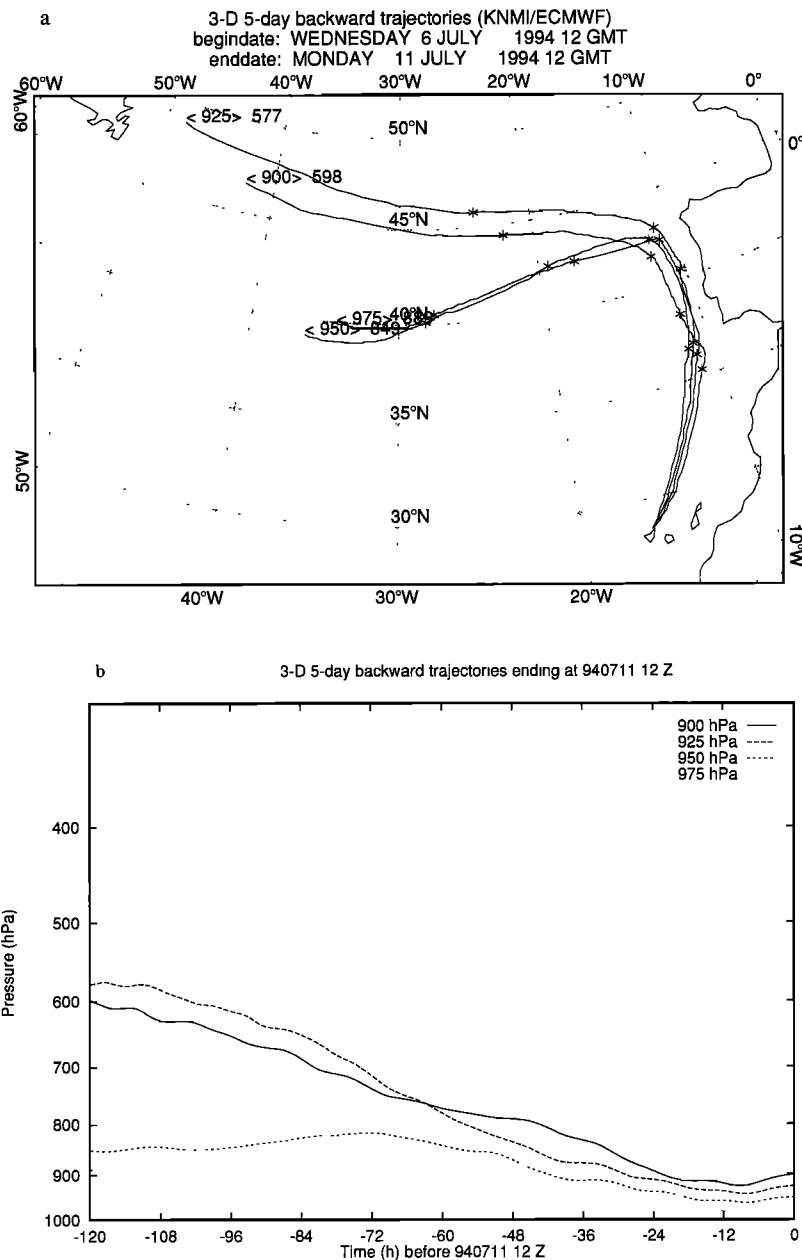Pressure (hPa)

Time (h) before 940711 12 Z

Figure 4. Five-day back trajectories ending on July 11, 1200 at four pressure levels within the marine boundary layer (MBL): 975, 950, 925 and 900 hPa. (a) horizontal projections, (b) vertical projections. They show that the composition of the MBL at PDH is made up of two air masses, one of them recently originating in the free troposphere.

internal/external mixing of these aerosols has implications for the radiative effect of the sulfate plume. We gave evidence for the fact that the sulfate particles observed during the dust event are predominantly in the submicron range, hence externally mixed from the supermicron dust particles. It is still possible that the sulfate is internally mixed with the submicron mineral dust aerosol that might be present. External mixtures of submicron pollution aerosol and supermicron dust aerosols have been observed by *Clarke et al.* [1996a] during one aircraft profile over the North Atlantic. They observed a transition from a predominantly pollution aerosol between 900 and 1200 m altitude, via an external pollution/dust aerosol mixture between 1200 and 2500 m, to a

predominantly dust aerosol between 2500 and 4200 m. It is not clear from their data whether the mixed aerosol layer resulted from a local vertical mixing between the pollution layer below and the dust layer aloft, or whether the various layers advected independently to the location of the aircraft descent. Both our observations and those of *Clarke et al.* [1996a] show that the anthropogenic sulfate remains largely in the size range that is most efficient for scattering light. Over the North Atlantic, this is probably due to the fact that the European sulfate aerosol is already well aged before it mixes with dust. The particles are therefore relatively large, have a low diffusivity, and will not interact with the dust particles. This is in contrast with a fresh sulfate plume, in which sulfur

dioxide is still being oxidized. Such a plume is expected to interact more efficiently with dust, e.g., by heterogeneous reactions or condensation. In this case the sulfate would be moved into the supermicron size range, which is much less effective for scattering light [e.g. *Dentener et al.*, 1996]. It is therefore important to study this degree of mixing in more detail, for example, using single-particle chemical analysis.

High ozone concentrations at IZO. There is an ongoing discussion whether the high O_3 levels at IZO, as seen for example on July 15, have an anthropogenic or natural origin [*Schmitt*, 1994; *Prospero et al.*, 1995; *Millan et al.*, 1996; *Cuevas et al.*, 1996]. The size distribution observed during the clean episode FT-II might shed some light on this. The back trajectories and vertical profile of dew point during this episode support the upper tropospheric origin of the air mass over IZO. Furthermore, the relatively small size of the aerosol particles during nighttime argues for an aerosol that results from nucleation (the day before) in the upper troposphere rather than transport of aged anthropogenic or stratospheric aerosol. However, this does not exclude yet that the ozone is anthropogenic. Ozone and insoluble gaseous aerosol precursors may have been pumped up over industrial areas by convective clouds into the FT, while precipitation in such clouds removed most of the pollution aerosol [*Prospero et al.*, 1995]. However, one further observation supports the natural origin of the air mass sampled at IZO, namely that the clean FT aerosol size distributions do not show an accumulation mode (see Figure 2b). This observation could only be made consistent with convective transport of polluted air, if convective transport would remove with 100% efficiency the anthropogenic aerosol precursors and the pollution aerosol. This is unlikely, since convective transport is not necessarily connected with precipitation. We therefore conclude that during episode FT-II the air masses sampled at IZO during nighttime were not influenced by anthropogenic pollution, and that the ozone and aerosol had a natural origin.

MBL

There are a number of similarities between our observations in the NE Atlantic, and observations made over the remote Pacific Ocean. *Covert et al.* [1996] reported a dominant Aitken mode in areas of large-scale subsidence and during subsidence events behind cold fronts. Their Aitken mode is shifted to smaller sizes compared to what we observed during, for example, MBL-I. This is possibly due a faster downward transport (in particular behind cold fronts), hence shorter growth times for the newly formed aerosol. In the Pacific trade-wind MBL, *Covert et al.*, [1996] and *Clarke et al.* [1996b] observed clearly distinct Aitken and accumulation modes, similar to what we observed during MBL-III. In all these cases it was the long residence and processing time within the MBL that resulted in the bimodal distribution. This distribution can be considered the result of a stationary state between a source of Aitken mode particles, cloud processing, and wet removal.

The fact that bursts of nucleation were not observed within the MBL is consistent with observations made elsewhere in subtropical or trade-wind regimes [*Clarke et al.*, 1996b, *Clarke et al.* 1996c, *Covert et al.* 1996, R. Van Dingenen, personal communication, 1995). These observations have risen the question about the origin of the Aitken mode particles observed in the MBL. Several hypotheses have been presented in the literature, which can be divided into "entrainment from the FT" [*Raes et al.*, 1993; *Clarke*, 1993; *Raes*, 1995; *Clarke et al.*, 1996b,c] and "nucleation within the MBL" [e.g. *Raes and Van Dingenen*, 1992, *Lin and Chameides*, 1993; *Kerminen and Wexler*, 1994; *Pandis et al.*, 1994; *Coffman and Hegg*, 1995].

Entrainment. Our observations during episodes MBL-I, -II, and -IV, in particular the back trajectories originating in the FT and the similarities between the MBL Aitken mode and the FT aerosol, argue for a FT origin of the MBL Aitken mode. During these episodes the transport from FT to MBL seemed to occur as an injection of FT aerosol into the MBL at a certain place. This is much different from the way *Raes* [1995] modeled entrainment, i.e., as a constant flux everywhere across the inversion. The latter type of entrainment can still be invoked for explaining the Aitken mode during episode MBL-III. The observed variability in the Aitken mode number concentration indicates however that the associated flux of particles is also very variable.

Other support for entrainment stems from the observation that despite the obvious decoupling of transport in the MBL and FT, and the differences in cloudiness in both layers, the total number concentrations of the MBL and FT aerosol are not very different. In Figure 5 we have plotted the frequency distributions of aerosol number concentrations at IZO and PDH for the whole duration of the period. It is shown that the background aerosol number concentrations at PDH and IZO were about equal at 600 cm^{-3} STP. A simple explanation could be that wherever and however the aerosol originates, coagulation will reduce the total number concentrations below 1000 cm^{-3} after several days of transport away from the source regions. The other explanation is entrainment, which provides enough aerosol surface for condensation and quenching in situ MBL nucleation. In absence of this in situ source of new particles in the MBL, the MBL number concentration must eventually equal the FT number concentration, or be lower in case of wet removal within the MBL. According to aerosol dynamics calculations, 15 $\mu m^2/cm^3$ of dry aerosol surface is sufficient to quench (classical) nucleation of H_2SO_4-H_2O aerosols at typical remote MBL conditions [*Raes*, 1995]. From measurements, *O'Dowd et al.* [1996] derived that a dry surface area of about 3 $\mu m^2/cm^3$ would be sufficient. Episodes FT-I and FT-II yield average dry FT aerosol surface areas of 22 and 4.4 $\mu m^2/cm^3$, respectively, with minimum values of 10 and 1.2 $\mu m^2/cm^3$. Considering that within the MBL, condensation, and cloud processing would further increase the size of the entrained FT aerosol, we can conclude that the injected or continuously entrained FT aerosol was probably sufficient to quench in situ nucleation in the MBL. We note also that in case of a continuous entrainment (e.g., episode MBL-III) the time scale of diluting the MBL is about 2 days (assuming an MBL height of 1000 m and an entrainment velocity of 0.6 cm/s), which is less than the (at least) 5 days the back trajectories stayed within the MBL before reaching PDH.

In-situ nucleation. Classical nucleation theory predicts nucleation to be critically dependent on the environmental parameters [e.g., *Jaecker-Voirol and Mirabel*, 1988]. Aerosol dynamic models based on this theory therefore predict the formation of aerosols in bursts, filling up the size range around and below 20 nm [e.g. *Raes*, 1995]. This was not observed at PDH, and it is unlikely that it happened randomly
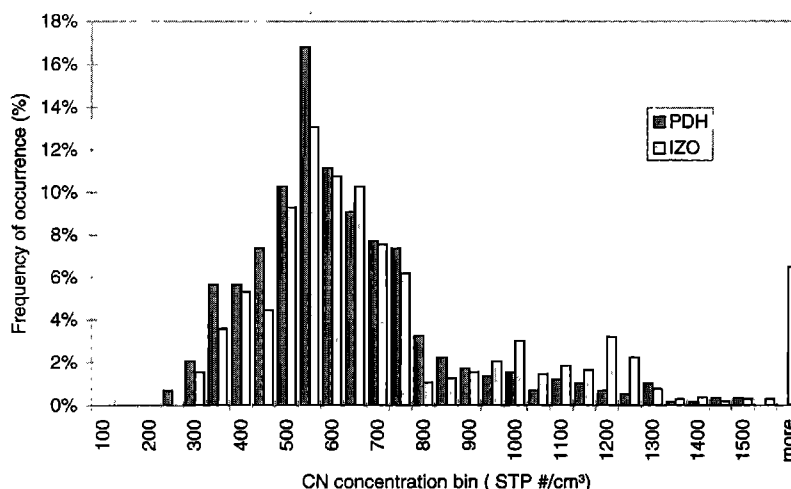
Figure 5. Frequency of measured values of total number concentration in Punta del Hidalgo (day and night) and Izaña (night only), showing that between July 9 and 17, the background number concentration in the free troposphere was essentially equal and around 600 cm^{-3} STP. Number concentrations were obtained by integrating the TSI DMPS size distributions.

upwind of PDH, as this would have resulted in a more variable mean diameter of the Aitken mode. There is, however, experimental evidence for a less critical (nonclassical) nucleation process [*Weber et al.*, 1996], which opens the possibility that nucleation occurs more continuously at low rates. Such a nucleation mechanism cannot be dismissed by our data. Applying a continuous low rate nucleation of the order of 2.10^{-3} cm^{-3}s^{-1} in our AERO2 model [*Raes*, 1995] can indeed lead to a mode that peaks around 65 nm D_p, but with a tail down to the size of the nucleating particles. This is because at a lower nucleation rate but constant formation rate of condensable material, more condensable material is available per particle. Hence each particle will initially grow very fast but eventually pile up around ≈ 65 nm where its growth becomes diffusion limited. The existence of an Aitken mode tail into the nanometer range could not be ascertained by the instrumentation operated at PDH. Low concentrations of nanometer particles would, in fact, be difficult to detect with any DMA system, because of the low sensitivity of these systems in the sub-10 nm range, and approaches like those of *Weber et al.* [1996] are essential.

Sea spray. During our measuring period, production of submicron particles by sea spray can be neglected as a source of aerosol number. In the area upwind of Tenerife the average wind speed is around 5 m/s, and according to *O'Dowd et al.* [1993], such wind speeds would contribute about 10 sea spray particles/cm^3 larger than 100 nm D_p. This is about 3 to 10% of the observed number of accumulation mode particles, and it is unlikely that it would be more in the Aitken mode.

Van Dingenen et al. [1995] have interpreted the accumulation mode of marine aerosols as cloud condensation nuclei. Our measurements show that during episode MBL-II the accumulation mode very probably consisted of pollution derived aerosol, which contained enhanced levels of black carbon. Since carbon containing particles might not all be capable of being activated in clouds, the contribution of sea salt to actual CCN (rather than accumulation mode particles) might have been larger than 3-10%. Single-particle chemical

analysis of CCN is required to quantitatively address the question of the contribution of sea salt to CCN.

6. Summary

The observations of the aerosol size distributions at IZO and PDH allowed to identify various episodes, which were not always detectable with chemistry measurements only. For instance, the chemical composition of the aerosol at PDH distinguished only between the polluted episode MBL-II on the one hand, and the clean periods before and after (McGovern, personal communication, 1997). Back trajectories, radio soundings, and AVHRR images, were essential to interpret the observations in terms of the origin of the air mass, and to show that FT-MBL transport, cloud processing, and mixing of air masses were the main processes shaping the aerosol distribution of the MBL aerosol. Our measurements do not give quantitative answers to the three questions mentioned in the introduction but yield the following information:

1. There is a close link between the variability of MBL aerosol and the changes in vertical transport patterns. This supports that entrainment or injection of aerosols from the FT is a major source of MBL aerosol. The entrainment process and its effect on MBL aerosol dynamics is more complex than originally proposed by *Raes* [1995]. For instance, the variability in the number concentration of the MBL Aitken mode suggests that the flux of aerosols by entrainment is not constant. This can be due to variations in the entrainment velocity, which is related to the structure of the MBL and the turbulent forces, and/or it can be due to the layered structure of the FT, which we observed.

2. When injection of FT aerosol occurred, it occurred at a rather fixed distance upwind of the MBL site. It cannot be excluded that at the moment of injection, when dry FT air is mixed with humid MBL air, new particles were formed by rapid nucleation, which subsequently grew and contributed to the observed Aitken mode at the site.

3. No bursts of nucleation did occur at the MBL measuring site, and it is unlikely that they happened at random upwind of the site. A continuous slow production of nanometer particles, according to a nonclassical nucleation process cannot be excluded. Measuring such a slow production is at the limit of present measuring capabilities.

4. During desert dust outbreaks, major submicron aerosol loadings were found in the free troposphere, which were very likely associated with anthropogenic aerosol originating over southern Europe. We have some evidence that entrainment of these aerosols affected the MBL aerosol composition during some of the episodes. Advection in the free troposphere and subsequent entrainment must be considered as a pathway to affect MBL aerosols, and possibly CCN, far away from pollution sources.

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