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The 2nd Aerosol Characterization Experiment (ACE-2): meteorological and chemical context

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ABSTRACT

A review is given of the climatological and actual meteorological conditions in the sub-tropical northeast Atlantic, during June–July 1997, when the 2nd Aerosol Characterization Experiment (ACE-2) took place. Surface pressure maps, trajectory calculations and in-situ measurements show how the outflow of European pollution into the marine boundary layer of this area is determined by the location of the Azores high pressure cell. Observations during ACE-2 and 3 preceding summers show that pollution outbreaks both from the Iberian peninsula and from northern or central Europe can occur during such situations. During ACE-2, an unusually low number of strong North African dust outbreaks were recorded at the free tropospheric station of Izaña (Tenerife, 2360 m asl), although dust was recorded aloft the station.

1. Introduction

The 2nd Aerosol Characterization Experiment (ACE-2, Raes et al, 2000) took place from 16 June till 24 July, 1997, over the sub-tropical northeast Atlantic, roughly between Portugal and the Canary Islands (Fig. 1). Like any sub-tropical oceanic atmosphere adjacent to the western part of a continent, the area is characterized by a distinct temperature inversion. This is a consequence of heating by compression of the subsiding air over a cold upwelling ocean. This inversion separates the marine boundary layer (MBL) from the free troposphere (FT) aloft. Transport in these 2 major layers can be totally decoupled, resulting in different properties between these layers, including differences in chemical composition. Earlier observations have shown that in the MBL of the sub-tropical NE Atlantic clean air alternates with anthropogenic pollution from Europe, and possibly from N. America (Huebert et al., 1996; McGovern et al., 1999). In the FT aloft, clean air masses alternate with mineral dust, transported out of N. Africa (Prospero et al., 1995; Raes et al., 1997; Fisher et al., 1998). The area thus offers

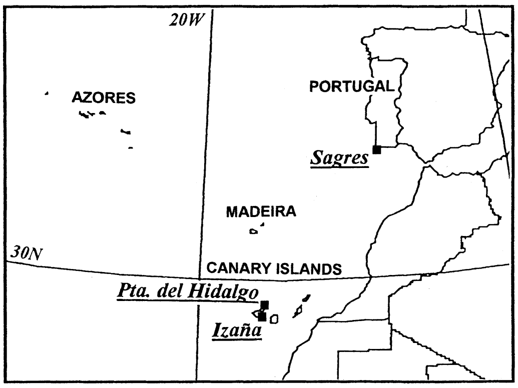

Fig. 1. The ACE-2 intensive area, with indication of the ground based measuring stations at Sagres (40 m asl), Punta del Hidalgo (50 m asl) and Izaña (2360 m asl).

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opportunities for studying the characteristics of various aerosol types. In particular, the area offers good opportunities to study the interaction of anthropogenic aerosols from Europe with the extensive low level stratiform clouds forming at the top of MBL.

The goal of this paper is to give a general meteorological context for the observations presented in this issue. In particular, we will compare the observed transport patterns and cloudiness during ACE-2, with the general climatology of *the area* (rather than in specific sites). We will look for relationships between the observed meteorology and some aerosol parameters measured during ACE-2, focusing in particular on the conditions leading to the outflow of anthropogenic aerosols from Europe, and dust aerosols from North Africa. Finally, we will try to identify specific source areas that affected the ACE-2 area.

Meteorological conditions for particular sites and/or observations are discussed in the individual papers. Back trajectories ending at various altitudes above the major ACE-2 sites (Tenerife (Canaries, ES), Sagres (PT), Mt Foia (PT), St Miguel (Azores, PT), Madeira (PT) and the ship (R/V *Vodyanitsky*) can be found in the ACE-2 archive. Information and request for access to this archive can be obtained from

http://ace2.ei.jrc.it or aceadmin.dm@jrc.it

2. Meteorological conditions during ACE-2

2.1. *Transport towards and within the marine boundary layer of the ACE-2 area*

2.1.1. General. In the lower troposphere, transport is governed by the location of the Azores High, the sub-tropical anticyclone over the north Atlantic. During summer this anticyclone is normally located to the west of the Azores, giving northerly and north easterly trade winds over most of the ACE-2 area, transporting relatively clean maritime air into the region. After the passage of a cyclone at mid-latitudes, the Azores High may temporarily extend over west Europe. This is expected to lead to an outflow of continental air into the ACE-2 area. Outflows of European pollution should thus be modulated by the passage of mid-latitude cyclones, and ridges of high pressure in between. Hence, in the sub-tropical northeast Atlantic, clean and polluted conditions are generally expected to alternate every 3 to 5 days on average. The latter reflects indeed the synoptic time scale where most of the variability in the atmosphere is found.

2.1.2. During ACE-2. We compared meteorological fields produced by the NCEP/NCAR re-analysis project (*http://www.cdc.noaa.gov/Composites/beta.html*). They show that during June–July 1997 the north Atlanic mid-latitude storm tracks as well as the sub-tropical anticyclone were located more to the south, compared to the 1968–1996 long-term average. This might be attributed to the strong onset of El Niño in 1997, as a similar displacement is not seen in non-El Niño years (e.g., 1990).

Between 16 June and 24 July, the Azores high extended 5 times into the Gulf of Biscay and over France. The evolution of one such an event, leading to the first ACE-2 pollution period, is shown by the series of ECMWF* surface pressure maps in Fig. 2. Aerosol measurements in the ACE-2 area (Fig. 3), show that with each of these events outflow of European pollution did occur, although not throughout the whole ACE-2 area. The first outflow occurred in the beginning of the experiment (17, 18 June). Pollution was detected in Sagres (SE Portugal) but not in Punta del Hidalgo (Canary Islands). The second outflow (23, 24 June) occurred after the passage of a cyclone from Iceland over the UK towards Scandinavia. Again, pollution was detected in Sagres but not in Punta del Hidalgo. The absence of pollution in Punta del Hidalgo during these 2 outflows will be discussed below. From 25 June till 3 July, a cyclone persisted over W. Europe bringing clean polar maritime air into the ACE-2 area. The cyclone weakened and disappeared on 3 and 4 July. After this, a ridge of high pressure developed, covering most of the north Atlantic with extensions over France between 4 and 10 July (Fig. 2). The resulting outflow of pollution was recorded first in Sagres and about 36–40 h later at Punta del Hidalgo. This was the first pollution event throughout the ACE-2 domain. Between 11 and 13 July, the high pressure retreated again and a

* Pressure maps and trajectories are obtained with the operational ECMWF (T212) model. For the calculations analysed fields of $1^\circ \times 1^\circ$ horizontal resolution, and 31 vertical layers were used.

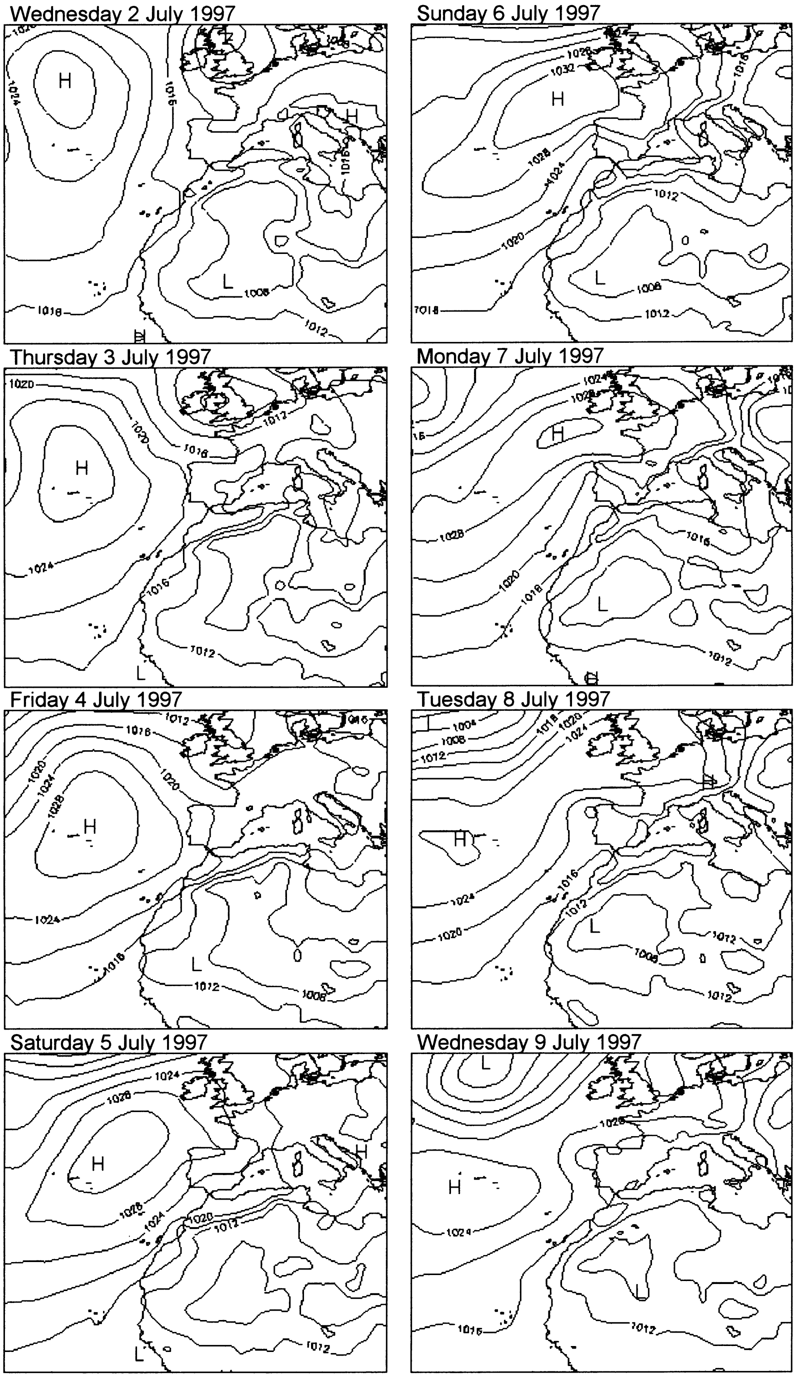


Fig. 2. Sequence of ECMWF surface pressure fields at 00 UTC, from 2 July till 9 July 1997. Shown is the transition from the conditions resulting in clean polar air masses arriving in the ACE-2 area, to the extension of the Azores high over west Europe, resulting in the first ACE-2 pollution event.

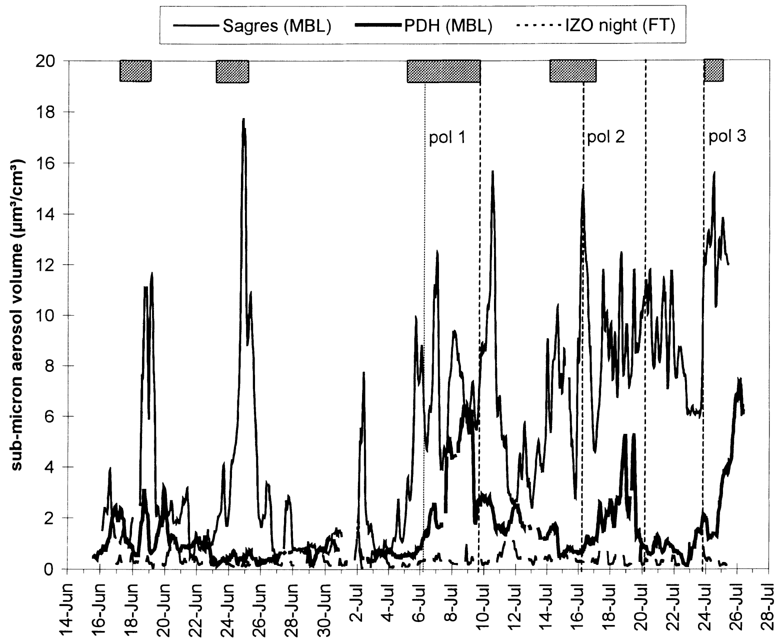


Fig. 3. Record of the sub-micron ($D_p < 1$ μm) aerosol volume in Punta del Hidalgo and Izaña (Putaud et al., 2000) and Sagres (Wiedensohler, ACE-2 data archive), for the whole ACE-2 period. The volume was measured with identical instrumentation at the 3 stations. Trajectory analyses (see Fig. 4, and also in Putaud et al., 2000) show that large increases in sub-micron aerosol volume at these stations are related to transport from the continents. The gray bars indicate when the Azores high extended into the Gulf of Biscay and France.

weak depression persisted west of the UK, bringing clean maritime air over the ACE-2 area. Pollution levels at Punta del Hidalgo dropped, but those in Sagres remained relatively high. Between 14 July and the end of the experiment on 24 July a huge high pressure area stretched from the Azores to the North Sea, initiating an outflow of European pollution to the south west. This led to 2 more pollution events that were recorded both in Sagres and Punta del Hidalgo. During this period, from 18 till 21 July, unusually heavy rains and floods accompanied the passage of a cyclone over central Europe.

The discussion above shows that the position of the Azores high determines the outflow of European pollution towards the sub-tropical north Atlantic. However, 2 points must be noted.

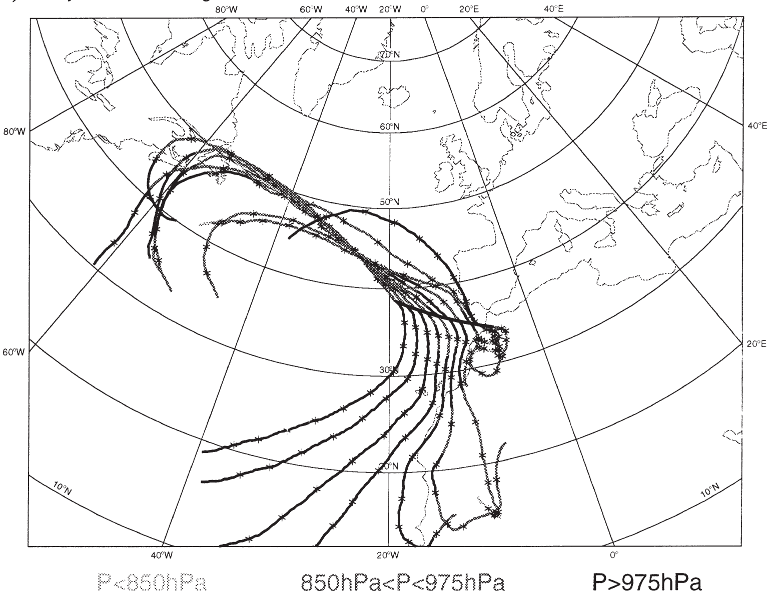
(i) As seen in Fig. 3, pollution events in Sagres are not always explained by the position of the north Atlantic anticyclone. That was the case on 18 June, on 1–2 July, and between 10–13 July. Trajectory analyses show that in these cases the air masses originated over the north Atlantic, and when moving along the Iberian west coast must have been picking up local pollution. (ii) Fig. 3 further shows that pollution arriving in Sagres did not always travel to Punta del Hidalgo. This is due to a heat low that is present in the lower troposphere over north-west Africa and that can draw the polluted air mass over the African continent before it reaches the Canaries.

The 3 pollution events detected both in Sagres and Punta del Hidalgo will be called the “ACE-2 pollution events”. They are indicated by the dotted lines in Fig. 3.

The trajectories in Figs. 4a–e show in more detail 5 situations of transport towards and within the MBL of the ACE-2 area. Each trajectory is a composite of a 5 days backward trajectory ending at the dashed line at 975 hPa., and a 3 days forward trajectory starting at the same line. They are 3-D trajectories based on the ECMWF model (Scheele et al., 1996).

(a) Fig. 4a is representative for the first week of ACE-2. Air masses were relatively clean. The trajectory analysis does not exclude that the

a) Trajectories crossing the line at 975 hPa on SATURDAY 21 JUNE 1997 12 GMT



b) Trajectories crossing the line at 975 hPa on THURSDAY 3 JULY 1997 12 GMT

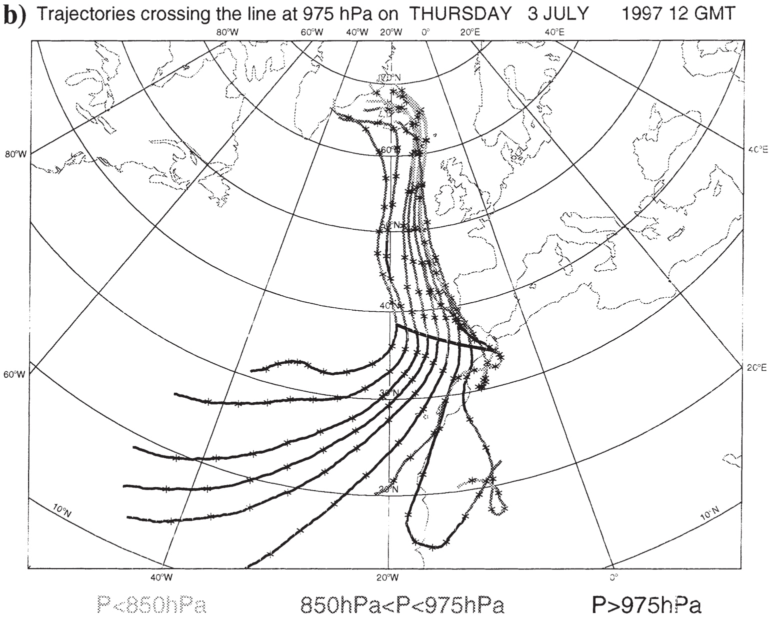


Fig. 4. (a) Trajectories based on ECMWF meteorology. Each trajectory is a composite of a 5 days backward trajectory ending at the line at 975 hPa, and a 5 days forward trajectory starting at that same line. Pieces of the trajectories that stay above 975 hPa (hence within the MBL) are plotted black. Others are grey and light grey. In this plot the crossing of the dashed line occurs on 21 June, 12 UTC. (b) Same as Fig. 4a, but for 3 July, 12 UTC. See Fig. 2 for the corresponding surface pressure fields. (c) Same as Fig. 4a, but for 7 July, 12 UTC. See Fig. 2 for the corresponding surface pressure fields. (d) Same as Fig. 4a, but for 18 July, 12 UTC. (e) Same as Fig. 4a, but for 24 July, 12 UTC.

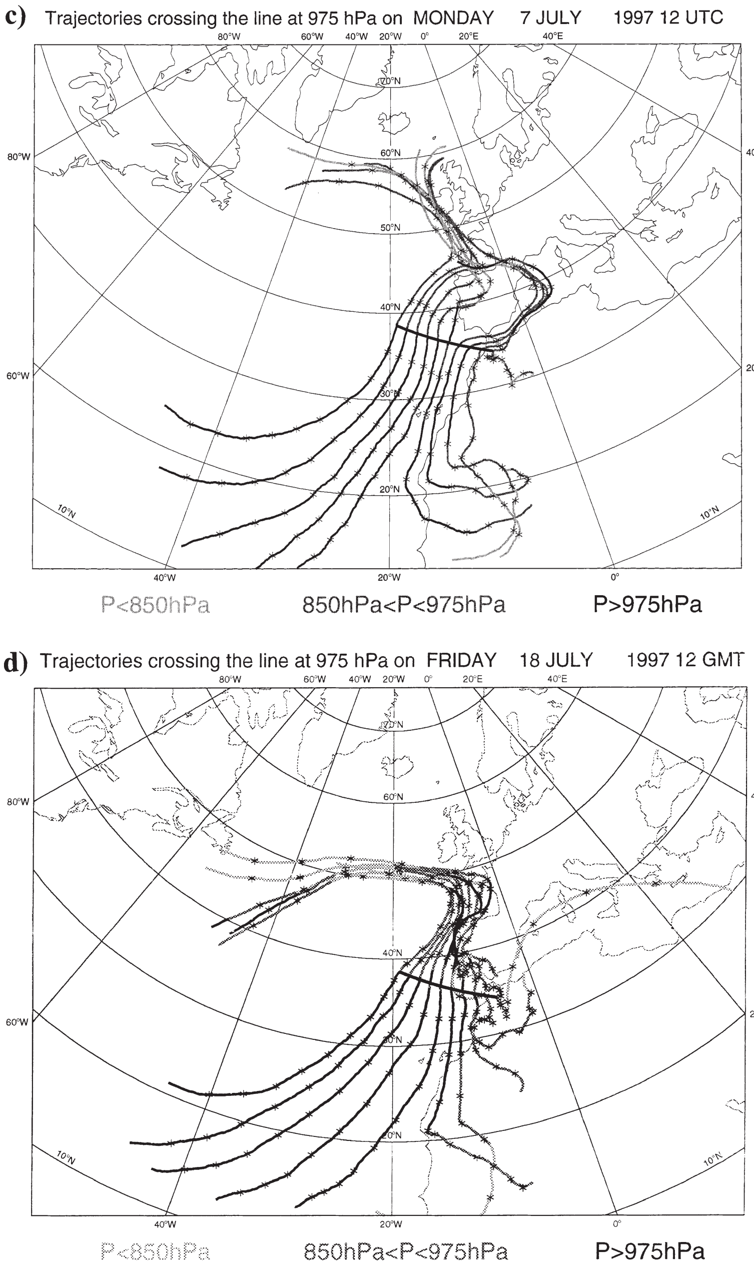


Fig. 4 (cont'd).

increased background concentration of aerosols in Punta del Hidalgo is the result of transport from north America. Transport across the Atlantic occurred near the surface.

(b) Fig. 4b shows the transport of polar air masses during the end of June/beginning July. This situation corresponds with the first pressure field shown in Fig. 2. It resulted in the cleanest air masses of the experiment in Sagres, Punta del Hidalgo, and the free tropospheric station of Izaña.

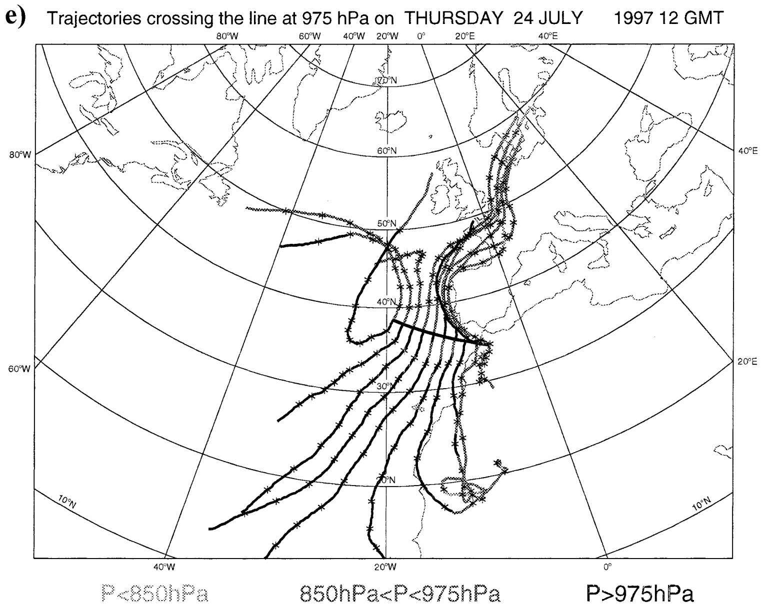


Fig. 4 (cont'd).

According to the trajectories, large scale subsidence occurred, which can also be seen from the divergence of the trajectories south of the dashed line. At the end of this period the first Lagrangian experiment was performed, following a clean air mass (Johnson et al., 2000).

(c) During the first pollution event (Fig. 4c), the air-mass crossed the UK and Ireland, and was split by the mountain ridges of the Pyrenees. One branch continued over the Atlantic, whereas the other crossed France and entered the Mediterranean. As suggested by Fig. 4c (and confirmed by back trajectories from the sites), Sagres detected the latter air mass when it left the Mediterranean through the Strait of Gibraltar, whereas Punta del Hidalgo detected pollution coming along the western Iberian coast. Measurements on the R/V *Vodyanitsky*, which sailed southwestwards of Sagres showed clear differences between these 2 air masses (Quinn et al., 2000, Bates et al., 2000). Many of the pollution/cloud interaction studies in Hillcloud (Bower et al., 2000) and CloudyColumn (Brenguier et al., 2000) took place near or on Tenerife, sampling the Western branch of that polluted air mass.

(d) During the second pollution event (Fig. 4d), the trajectories traveled along the Iberian coast and the pollution detected at both sites probably originated there. Air masses transported in the anti-cyclone (the 5 most western trajectories) were subject to subsidence. During this event the second Lagrangian experiment was performed, following a polluted air mass, and more Hillcloud and CloudyColumn experiments were carried out in the same air mass.

(e) During the 3rd pollution event (Fig. 4e), the trajectories for Sagres and Punta del Hidalgo both originated over north Europe, and spent some days over the ocean before reaching the sites. It is likely that both sites experienced the same aged pollution plume. According to the trajectory analyses, the air mass was only slightly subject to subsidence. During this event the 3rd Lagranian experiment was performed, following the aged polluted air mass.

The trajectories in Figs. 4c–e indicate that through the action of the extended Azores high, the airflow tended to hug the European coastline. This resulted in rather narrow plumes of pollution, which were subsequently spread and diluted in the divergent flow over the NE Atlanic. The latter dilution is thought to be responsible for the systematically lower aerosol concentrations at Punta del Hidalgo, compared with Sagres (see Fig. 3, Van Dingenen et al. (2000)).

As a final remark regarding the MBL, it should be mentioned that the representation of the MBL as one layer is too simplistic. Earlier work in ASTEX (Huebert et al., 1996) has elucidated the process of decoupling of the MBL when the air moves southward over a warmer sea surface. The resulting multi-layered MBL and its effect on the aerosol has been studied in depth during the ACE-2 Lagrangian experiments (Johnson et al., 2000).

2.2. *Transport within the free troposphere of the ACE-2 area*

2.2.1. General. Transport in the FT is governed by geostrophic and subsiding flow around the sub-tropical anticyclone in the mid-troposphere. At 500 hPa, this anticyclone usually extends from the sub-tropical north Atlantic to deep over the Sahara. In this case, Tenerife is located at the northern side of the anti-cyclone and at high altitudes it will experience air masses from the west and northwest. Trajectories in the FT are roughly parallel to those in the MBL during clean conditions (Figs. 4a,b). The air masses are clean but may contain layers of pollution originating from the North American continent. Dust that is lifted over Niger and Chad, travels along the southern side of the anti-cyclone westwards to the Atlantic. As a response to easterly waves over west Africa and the tropical Atlantic (Diedhiou et al., 1998), the anti-cyclonic flow often splits in an oceanic (Azores) cell and a continental (Libyan) cell, with a mid-latitude trough in between that extends somewhat to the south (see Fig. 6, for an example during ACE-2). In this case, Tenerife can be at the western side of the Libyan high pressure cell, and at high altitudes it will experience air masses from the south that originated over the dust source areas. The trajectories in the FT are totally decoupled from those in the MBL. These conditions lead to observations of dust at the FT station of Izaña (2360 m asl) on Tenerife during the summer months.

2.2.2. During ACE-2. Satellites detected dust over de Sahel and Saharan region and over the tropical Atlantic throughout the ACE-2 experiment. Sunphotometers detected 3 clear dust events aloft the Izaña station (Formenti et al., 2000), however dust was rarely and only weakly detected at the Izaña station itself (Fig. 3). During June the continental high pressure cell remained deep over the Sahara and dust was transported northwards over the western Sahara, rather than towards Tenerife (Fig. 5a). On 7 and 8 July, this high pressure cell shifted to the west, leading to the first occurrence of dust over Tenerife 2 days later (Fig. 5b). A major dust event was recorded at Izaña between 26 and 31 July, and resulted from a break up of the high pressure cell as described in the General section above, and as shown in the sequence of maps in Fig. 6. During most of the ACE-2 period, air masses arriving at Izaña originated in the middle or higher troposphere over the north Atlantic or Arctic, and the (night-time) aerosol concentrations at Izaña remained extremely low (Fig. 3).

3. Cloudiness in the ACE-2 area

Figs. 7a–e are Meteosat VIS images of the ACE-2 area, for the same dates and hours as when the trajectories arrive at the dashed line in Figs. 4a–e. A clear distinction in cloudiness is visible between the clean periods (Figs. 7a,b) and polluted periods (Figs 7c–e). In the former period a strong stationary depression over the UK kept the Azores high over the north Atlantic. Broken low-level clouds formed upwind the ACE-2 area and move in with the trade winds. During the each of the European outflows a cloud free air mass left the continent and cloud formed downwind within the ACE-2 area. This is particularly clear in Fig. 7c. The detailed observation of the transition of the continental boundary layer into a (new) marine bounday layer where cloud formation occurs, is one of the main achievements of the Lagrangian experiments (Johnson et al., 2000).

4. Origin of the pollution detected within the MBL, during ACE-2

Fig. 8 shows the sulfur emission inventory for 1995, assembled by the co-operative programme for monitoring and evaluation of the long range transmission of air pollutants in Europe (EMEP, 1997). Along the western and northern coast of Iberia important point sources of sulfur exist.

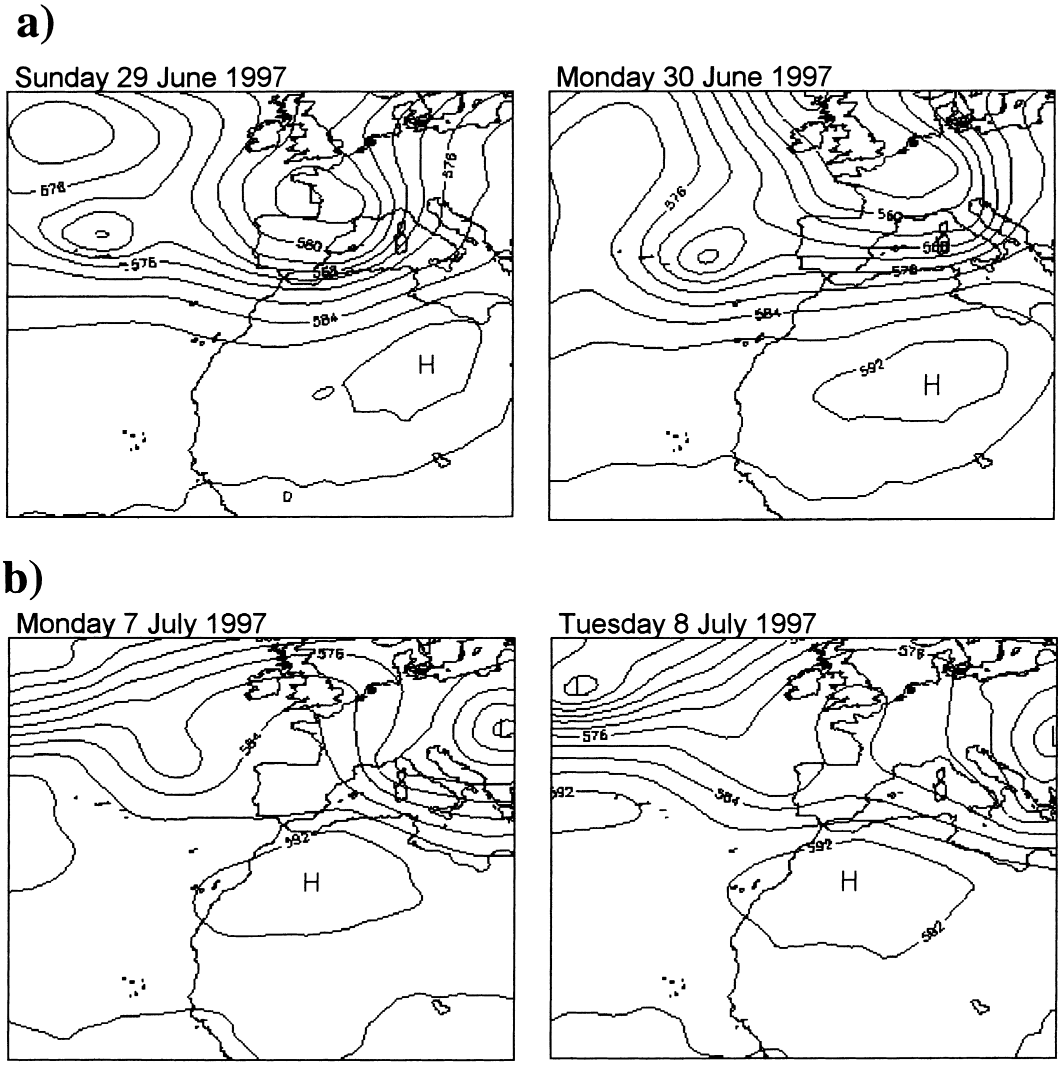


Fig. 5. ECMWF 500 hPa geopotential height maps (a) for 29 and 30 June, corresponding with dust free conditions at Izaña (Tenerife, 2360 m asl) and (b) for 7 and 8 July, which situation leads to the first dust event over Tenerife on 9 and 10 July.

According to the trajectories in Figs. 4c,d, it is likely that these sources were responsible for the pollution observed in the ACE-2 area during the first 2 pollution events. Another concentration of sulfur emissions stretches from the English Channel to the German Ruhr area. According to the trajectories in Fig. 4e, this is likely to be the source of the pollution measured in the third pollution event. This area has also particularly high NO_x and ammonia emissions, compared to elsewhere in Europe. Fig. 8 also shows the emissions of ships. The contribution of ships going from Gibraltar to the English Channel might not be insignificant, as many of the air masses ending in Sagres or Punta del Hidalgo follow the same route and could have integrated those emissions. A recent modelling study, however, estimates the contribution of ship emissions to the nss sulfate

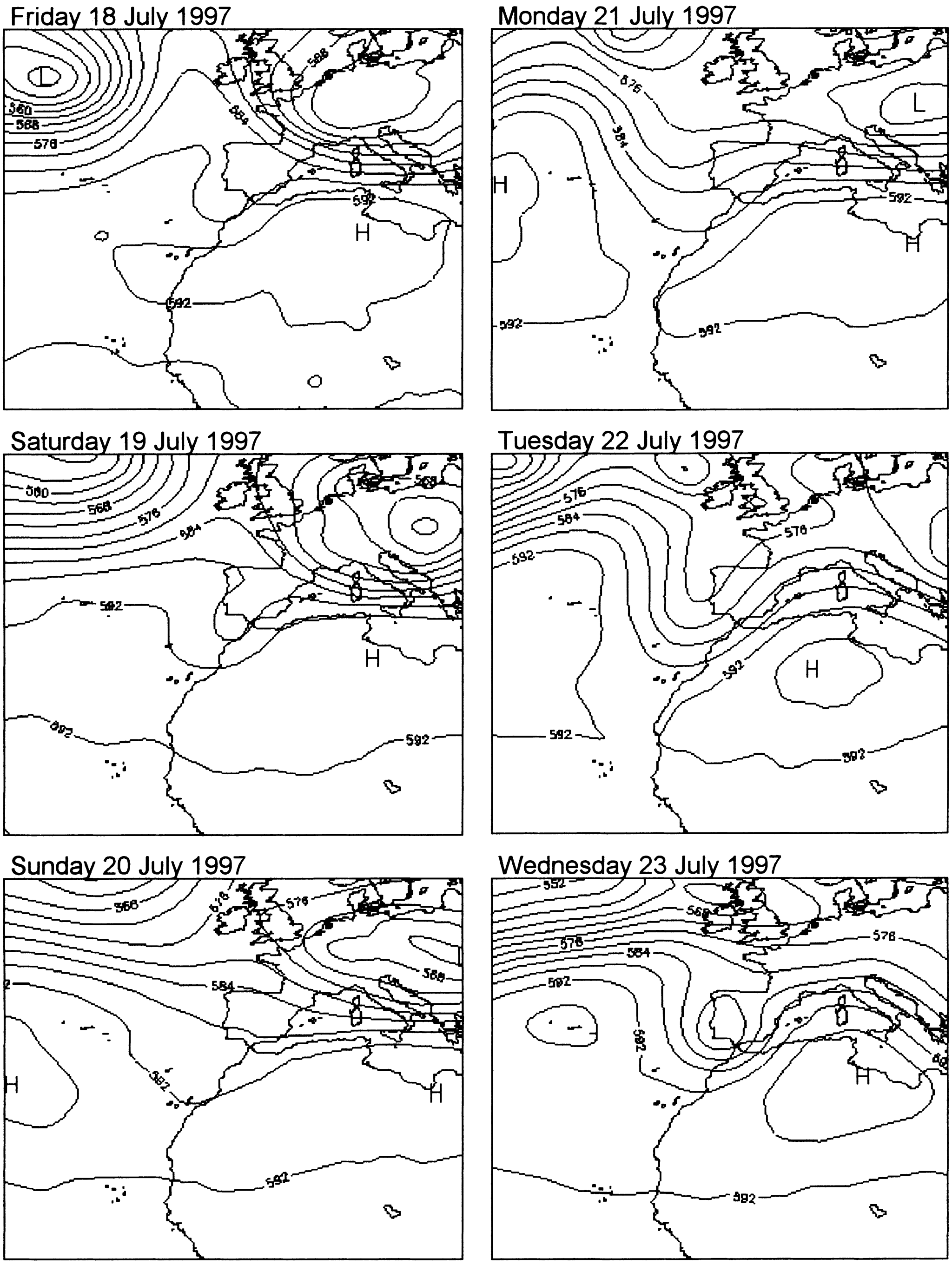


Fig. 6. Sequence of 500 hPa geopotential height maps from 19 till 23 July. The break up of the high pressure cell in an oceanic and continental one leads to the major dust event over Tenerife on 25 July.

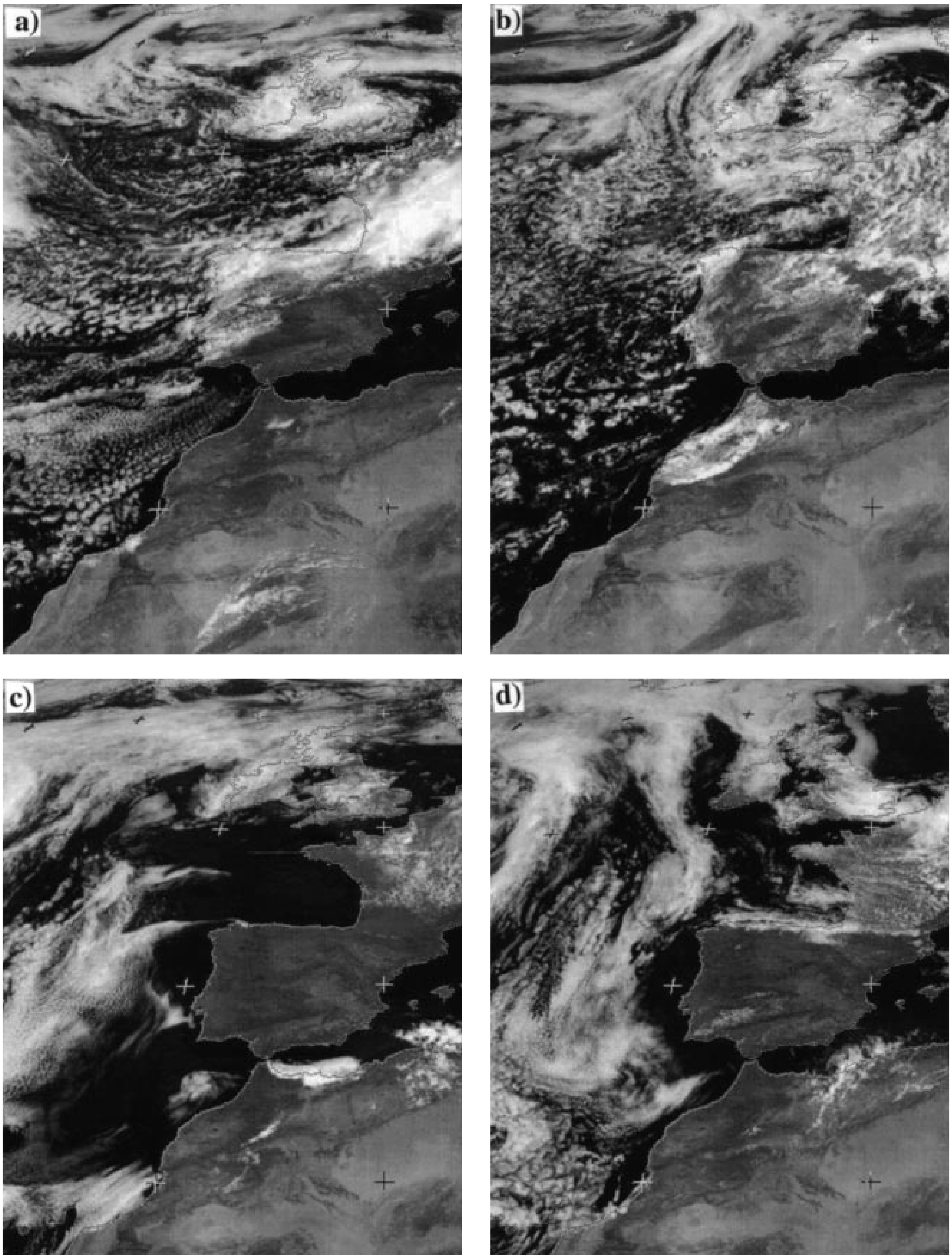


Fig. 7. (a) Meteosat VIS image for 21 June, 12 UTC (same time Fig. 4a). (b) Meteosat VIS image for 3 July, 12 UTC (same time Fig. 4b). (c) Meteosat VIS image for 7 July, 12 UTC (same time Fig. 4c). (d) Meteosat VIS image for 18 July, 12 UTC (same time Fig. 4d). (e) Meteosat VIS image for 24 July, 12 UTC (same time Fig. 4e).

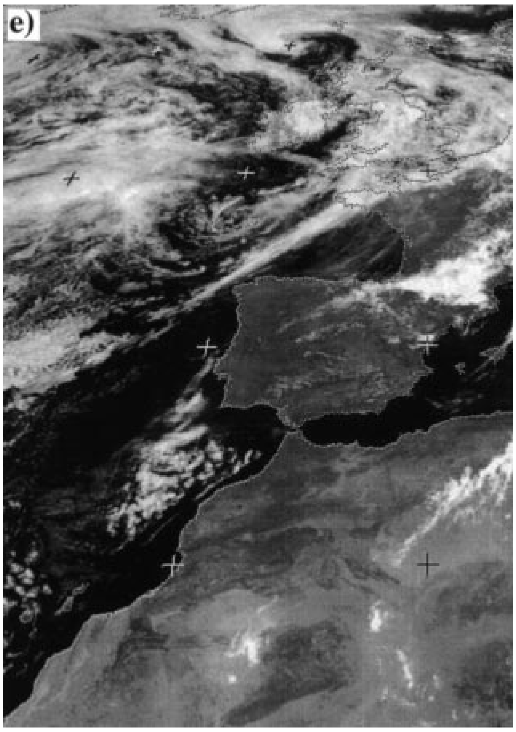


Fig. 7 (cont'd).

concentration not higher than 20% in this area (Capaldo et al., 1999).

The above discussion might explain the relatively high ($\sim$70%) sulfate + ammonium content in the sub-micron aerosol during the ACE-2 pollution events (Putaud et al., 2000, Neususs et al., 2000). It should be noted however, that the few mediterranean air masses sampled during ACE-2, contained equal amounts of nss sulfate, but significantly larger concentrations of carbonaceous aerosol (Quinn et al., 2000). The effects of the large sulfur emissions areas in central UK and in former East Germany were probably not detected, neither in Sagres nor in Punta del Hidalgo.

5. Perturbation of the ACE-2 area by anthropogenic pollution and dust

Following Putaud et al. (2000) we used the aerosol absorption coefficient (b_{ABSU}) to define background and polluted air masses. In the MBL, where absorption is mainly due to elemental carbon, a threshold of $b_{ABSU} > 1.2 \cdot 10^{-6}\,m^{-1}$ has been chosen to define continental aerosol. In the FT, where absorption is more related to the presence of dust, the same threshold has been taken. Using these threshold it is possible to calculate the percentage of the time the ACE-2 sites were affected by continental air masses during the campaign The resulting values are summarized in Table 1, and compared with measurements performed during pre-ACE-2 campaigns in 1994, 1995 (Raes et al., 1997; McGovern et al., 1999) and 1996 (unpublished results). For ACE-2, only the month of July is considered, to be consistent with the previous years.

The % of time of pollution within the MBL was not unusually high or low during ACE-2. During the preceding years pollution from the Iberian peninsula was more common than from northern or central Europe (McGovern et al., 1999). This was also the case during ACE-2. Hence, the effect of a more southern location of the mid-latitude jet and sub-tropical anti-cyclones during ACE-2 (see above), was not very influential on the frequency and type of pollution outflow from Europe. The % of time of pollution in Punta del Hidalgo and Sagres is large, in the light of the AVHRR aerosol optical depth (AOD) values in the ACE-2 area (Durkee et al., 2000). They show that the area has been particularly clean, i.e., its average AOD (630 nm) = 0.162 (SD = 0.109) compared to 0.130 (SD = 0.028) in ACE-1. The low average AOD indicates that European pollution or N. African dust outbreaks covered only a small fraction of the area. As mentioned before, the pollution drawn out of Europe tends to stay rather close to the continental boundaries. The higher standard deviations if the AOD, in the case of ACE-2, indicate how the continental outflows result in a large aerosol variability both in space and time.

Table 1 indicates that the % of time of dust detected at Izaña during ACE-2, was lower than during the preceding years. However, considering the sunphotometer measurements of Formenti et al. (2000), which probe the aerosol in the atmospheric column above Tenerife, a percentage of time of 40% can be estimated, which would be in agreement with the "in-situ" percentages of the

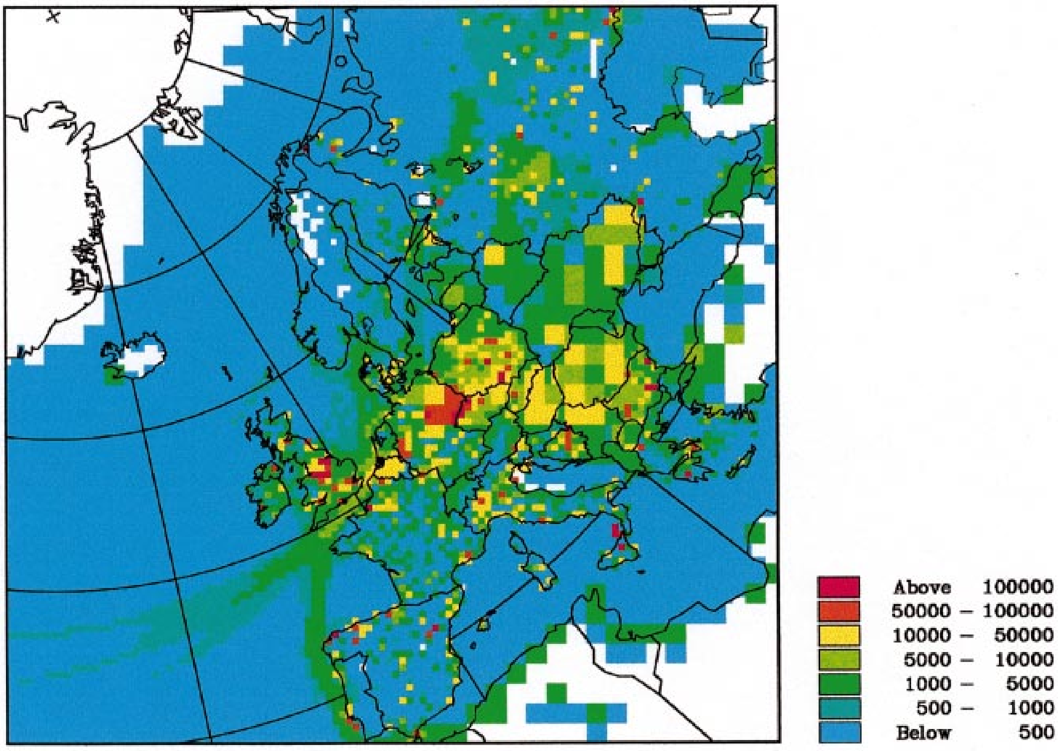


Fig. 8. Sulfur emissions in Europe during 1995 (EMEP, 1997). Units are tonnes of SO_2.

Table 1. *% of time in July, during which each site was affected by continental air masses, i.e., anthropogenic pollution at Punta del Hidalgo and Sagres, and mineral dust at Izaña*

	ACE-2			
	July 1997	July 1996	July 1995	July 1994
Punta del Hidalgo	45	60	45	35
Sagres	95	75	—	—
Izaña	15 40*	35	40	65

Values are based on "in-situ" measurements of the aerosol absorption coefficient. The value marked * is estimated from sunphotometer measurements above Tenerife (Formenti et al., 2000).

previous years. Hence, the difference between ACE-2 and previous years might have been only the height of the dust layer, and not its frequency of occurrence.

6. Conclusion

The choice of the ACE-2 area turned out to be excellent for the study of the evolution of pollution aerosols in the MBL. After 3 weeks of clean conditions in the MBL, the expected outflows of European pollution occurred at regular intervals.

This allowed for the study of aerosol characteristics and processes in contrasting clean/polluted environments. The polluted air masses were cloud free when leaving the continent, allowing for clear air radiative transfer studies. As the air mass moved over the ocean, cloud developed and allowed for the study of aerosol/cloud interactions. In the FT, dust transport seem to have occurred at a normal frequency. However, for reasons not explained, the base of the dust layers remained at higher altitudes and above the Izaña station. Hence there were few opportunities to characterize the mineral dust with the complete suite of techniques deployed at that station.

7. Acknowledgements

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