

Vertical ozone measurements in the troposphere over the Eastern Mediterranean and comparison with Central Europe

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Vertical ozone profiles measured in the period 1996–2002 in the framework of the MOZAIC project (Measurement of Ozone and Water Vapor by Airbus in Service Aircraft) for flights connecting Central Europe to the Eastern Mediterranean basin (Heraklion, Rhodes; Antalya) were analysed in order to evaluate the high rural ozone levels recorded in the Mediterranean area during summertime. The 77 flights during summer (JJAS) showed significantly (10–12 ppb, 20–40%) enhanced ozone mixing ratios in the lower troposphere over the Eastern Mediterranean frequently exceeding the 60 ppb, 8-h EU air quality standard, whereas ozone between 700 hPa and 400 hPa was only slightly (3–5 ppb, 5–10%) higher than over central Europe. Analysis of composite weather maps for the high and low ozone cases, as well as back-trajectories and vertical profiles of carbon monoxide, suggest that the main factor leading to high tropospheric ozone values in the area is anticyclonic influence, in combination with a persistent northerly flow in the lower troposphere during summertime over the Aegean. On the other hand the lowest ozone levels are associated with low-pressure systems, especially the extension of the Middle East low over the Eastern Mediterranean area.

1 Introduction

Tropospheric ozone gained attention 2–3 decades ago, when it was realized that increasing surface ozone levels observed in urban areas and at rural sites were attributable to enhanced photochemical production in the troposphere (Volz and Kley, 1988; Staehelin et al., 1994). Subsequently, research programs and extended monitoring networks provided a reasonably comprehensive picture of the spatial distribution of surface ozone in Northern and Central Europe, whereas a lack of long-term ozone measurements were noted to persist for the Mediterranean, especially the Eastern part (Beck and Grennfeld, 1993; Scheel et al., 1997). The analysis of ozone measurements made around Athens and at a rural site in Central Greece (Varotsos et al., 1993; Kalabokas and Bartzis, 1998; Kalabokas et al., 2000; Kalabokas and Repapis, 2004)

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revealed fairly high rural ozone levels; around 60 ppb during summer. This picture was confirmed by continuous measurements on the island of Crete (Kouvarakis et al, 2000) as well as measurements made during campaigns over the Aegean (Kourtidis et al., 2002; Kouvarakis et al., 2002; Lieleveld et al., 2002; Roelofs et al., 2003). The possibility for an influence of long range transport from the European continent, North America and Southeast Asia on ozone and its precursors over the Eastern Mediterranean has been discussed in several publications (e.g. Van Aalst et al., 1996; Lieleveld et al., 2002; Gros et al., 2003; Roelofs et al., 2003; Sheeren et al., 2003; Traub et al., 2003; Volz-Thomas et al., 2003). In addition, recent measurement and modelling studies suggest that polluted air masses exported from Switzerland towards the Mediterranean in summer exhibit high ozone production rates (Henne, 2005).

The weather conditions over the Eastern Mediterranean during summer are influenced by eastward extensions of the Azores anticyclone and the low pressure branch of the large South Asian thermal low. Modelling studies of the large-scale dynamics (Rodwell and Hoskins, 1996; Rodwell and Hoskins, 2001) also suggest a strong influence of the Indian Monsoon on the dry Mediterranean climate in summer, i.e. by Rosby wave interaction with the southern flank of the midlatitude westerlies producing adiabatic descent and hence anticyclonic conditions at the surface over the western Mediterranean. The resulting monsoon circulation over the Aegean Sea together with the strong pressure gradient, due to the surrounding mountains of the Greek peninsula in west and the Anatolian plateau in east, produces persistent northerly winds, the so called “Etesian winds” (annual winds). This flow of the low troposphere is most pronounced at the 850 hPa level (Repapis et al., 1977). As a result, the Eastern Mediterranean is influenced by advection from Europe in the lower troposphere associated with the Etesian winds and subsidence in the middle and upper troposphere associated with the westerly flow in the descending branches of the Asian thermal low and, to a lesser extent, of the East African monsoon. It was also shown that the day-to-day variations in these two main factors are linked to the Asian monsoon (Rodwell and Hoskins, 2001; Lieleveld et al., 2002; Ziv et al., 2004).

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The aim of this work is to investigate the high ozone background in the boundary layer over Greece and the Aegean Sea, using vertical ozone profiles collected over the Eastern Mediterranean and Central Europe within the MOZAIC project (Marenco et al., 1998; Thouret et al., 1998; Sauvage et al., 2005; Thouret et al., 2006) and by analysing the characteristic meteorological conditions of the days with the highest and the lowest ozone mixing ratios, respectively.

2 Results and discussion

The MOZAIC data base (<http://mozaic.aero.obs-mip.fr/web/>) was screened for summer flights between Central Europe and the northern part of the Eastern Mediterranean basin. In total, ozone profiles from 77 flights exist: 26 to Heraklion (35.3° N, 25.2° E), 14 to Rhodes (36.4° N, 28.1° E) and 37 to Antalya (36.8° N, 30.8° E). The departing Central European airport for most flights was Vienna (48.1° N, 16.6° E) in addition to 6 flights from Frankfurt (50.1° N, 8.5° E). The flights cover the period 1996–2002 and took place between June and September, almost exclusively during weekends. Both ascent and descent data have been used.

2.1 Average ozone profiles

For the following analysis the ozone profiles collected over the Mediterranean and the corresponding profiles over Vienna or Frankfurt were averaged over intervals of 100 hPa (50 hPa for the lowest and highest levels): 1000 (1000–950), 900 (950–850), 800 (850–750), 700 (750–650), 600 (650–550), 500 (550–450), 400 (450–350) and 300 (350–300) hPa.

Figure 1 displays median, 75 and 90 percentiles of the frequency distributions of the average ozone values for each pressure level over the Eastern Mediterranean (Heraklion, Rhodes and Antalya) and the corresponding values obtained over Central Europe (Vienna and Frankfurt) from the same flights. In both regions the 90 percentiles exceed the 60 ppb threshold at the 900 and 800 hPa levels. In the Eastern Mediterranean, even the 75 percentiles are still around 60 ppb, about 10 ppb higher than over Central Eu-

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rope. The median value over the Eastern Mediterranean is about 55 ppb between 900 and 700 hPa (Fig. 1a). Over Central Europe, the median at 700 hPa is similar (55 ppb) whereas significantly lower median values (42 ppb), are observed in the lower levels (Fig. 1b). Table 1 summarises the differences between the average ozone profiles collected over Central Europe and the Eastern Mediterranean. Clearly, the differences in ozone mixing ratios between the two regions are more pronounced in the boundary layer (9–12 ppb higher ozone values in the Eastern Mediterranean, whereas only 3–5 ppb higher ozone mixing ratios are observed in the middle troposphere). The picture remains unchanged when the flights to Antalya are excluded from the analysis. The results in Fig. 1 and Table 1 suggest that boundary layer processes should be responsible for the enhanced surface ozone levels over the Mediterranean.

In order to investigate the influence of the prevailing meteorological situation on the enhanced ozone values in the Aegean boundary layer, four groups of profiles were selected for further analysis: a) the quartile with the highest ozone values in the Eastern Mediterranean boundary layer (19 profiles), b) the quartile with the lowest ozone values in the Eastern Mediterranean boundary layer (19 profiles), c) the quartile with the highest ozone values in the Central European boundary layer (17 profiles) and d) the quartile with the lowest ozone values in the Central European boundary layer (17 profiles). The average ozone values for these four groups are presented in Tables 2 and 3. It is observed that the average ozone concentrations in the lower troposphere (700–900 hPa) for the highest ozone days in both regions are quite similar at 60–65 ppb while for the lowest ozone days the average ozone values in the Eastern Mediterranean are significantly higher: about 11 ppb at 900 hPa and 6 ppb at 800 hPa.

2.2 Composite weather maps

Figures 2 and 3 show composite 925 hPa weather maps for each of the four groups of profiles. The composite weather maps were constructed from the NCEP/NCAR reanalysis, based on grids of 2.5×2.5 degrees, for the days of the flights in each group following the procedure of Kalnay et al. (1996). The composite weather map (Fig. 2a) for the highest quartile of ozone values in the Eastern Mediterranean boundary layer

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(EMED) shows weak anticyclonic systems over the Central Mediterranean and over the Balkans inducing stagnant conditions. A well-established northern current occurs in the boundary layer over the Aegean due to the combined influence of the Central Mediterranean and the Balkan highs with the Middle East low-pressure system. High surface air temperatures are also recorded in the area during this group of days. The average 900 hPa ozone value in Eastern Mediterranean is 63 ppb with little variability (Table 2). The corresponding 900 hPa average value in Central Europe (CEUR) is about 50 ppb where a rather strong westerly airflow driven by the Scandinavian low occurs. The 13–15 ppb difference observed in the boundary layer between Eastern Mediterranean and Central Europe is reduced to 3–5 ppb in the 600–700 hPa pressure levels resulted from the uniform westerly flow over the whole Europe in the middle troposphere (not shown here). The lowest EMED boundary layer ozone levels (40 ppb) are recorded when the Middle East low-pressure system is extended to the West Mediterranean with a weak gradient (Fig. 2b). Low ozone values are observed throughout the troposphere. It is known that contrary to the high pressure systems linked with clockwise subsidence of tropospheric air, the low pressure systems are linked with upward counterclockwise movement of air masses, rising in fact the boundary layer air into the free troposphere. This upward air movement is enhanced by the fact that the weak pressure gradients in Mediterranean summers are linked with vertical instability due to thermal convection (Ziv et al., 2004).

The highest 25% of ozone values in CEUR are associated with a strong and extended anticyclone over the Central and Eastern European continent, associated with weak westerly winds (Fig. 3a) and high temperatures. The average 900 hPa value of the highest ozone values in CEUR is 60 ppb with little variability. On the other hand the lowest values in CEUR are observed when the area is under the influence of low-pressure system (Fig. 3b). The difference between EMED and CEUR is maximized (about 20 ppb) when low-pressure systems prevail over Central and Eastern Europe (Table 3).

From the above analysis it comes out that a key factor leading to high tropospheric

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ozone values in the Eastern Mediterranean but also in Central Europe is the anticyclonic influence. Summer anticyclones are rich in ozone as they transport downwards the upper troposphere ozone, which has a marked mid-summer peak over Europe (Thouret et al., 2006). At the same time anticyclones create stable conditions close to the surface, enabling the ozone precursors to travel over long distances horizontally. Under such low dispersion conditions fresh emissions added during the transiting of air masses over the abundant pollution sources of the European continent might offset the deposition of ozone precursors leading to pollutants accumulation. In the Aegean the frequent presence of the Central Mediterranean and the Balkan high pressure systems in combination with the Middle-East low is associated with a persistent northerly flow in the boundary layer, frequently quite strong and containing high ozone amounts.

2.3 Back-trajectories

From our data-set, four cases from the quartile of the highest ozone values in the Eastern Mediterranean and five cases from the quartile of the lowest ones have been recorded during summer 2002. For that year some CO profiles from the MOZAIC project are also available. The meteorological conditions for the high and low ozone days in that particular year are similar to those of the entire data-set. Likewise, a significant difference of 20–25 ppb is observed in the average ozone differences between EMED and CEUR in the lower levels, which is maximized at 900 hPa inside the boundary layer, indicating that the selected subgroup of profiles is quite representative.

For the determination of the air mass origin, the Lagrangian backward simulation model FLEXPART (version 6.2; Stohl et al., 1998, 2005) was used. The trajectories were started at vertical points along the MOZAIC profiles over Rhodes and were calculated backwards in time for ten and five days using ECMWF analysis data as input.

The backward trajectory simulations for the 4 cases of high ozone and the 5 cases of low ozone of the year 2002 in Rhodes are presented in Figs. 5 and 6 correspondingly. The percentage contribution of each geographical area separated in three atmospheric layers (0–2 km, 2–4 km and >4 km) was calculated for the air parcels arriving at the 0–2 km layer over the Rhodes airport during the high ozone cases (10-day backward

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simulation, Fig. 5). The corresponding percentage contribution of each atmospheric layer to the air arriving in Rhodes at 0–2 km on high ozone days, for the 10-days and 5 days backward FLEXPART simulations are shown in Table 4: For the 10-day trajectories about 55% of the air masses arriving at 0–2 km at Rhodes on the high ozone 5 days come from altitudes below 4 km, the contribution of this layer increases to 70% for 5-days trajectories. In the 10-days for the lower altitudes (0–2 km) most of the contribution comes from the Aegean area and especially the Turkish coast with a small contribution from Central Europe (Fig. 5). In the 2–4 km layer the contribution of the Aegean area is still significant but also a substantial contribution from the Balkans, Italy and Central Europe is observed. 10

The contribution of each atmospheric layer to the air arriving in Rhodes at 0–2 km on low ozone days for the 10-days and 5 days backward simulations is shown in Table 5. The corresponding geographical contribution for the different atmospheric layers during the low ozone days for the 10-days and FLEXPART backward trajectory simulations is shown in the Fig. 6. Most of the contribution comes from the lower layers of the Central and Eastern Mediterranean, advecting rather clean air to the measuring site, as it was also suggested by the composite weather maps. 15

2.4 Profiles of carbon monoxide

Because of its low solubility and its photochemical lifetime of the order of weeks, carbon monoxide is a good tracer of anthropogenic pollution. CO profiles have been used for tracking pollution episodes in the boundary layer or even in the free troposphere (c.f., Seiler, 1974, Seiler and Fishman, 1981; Nedelec et al., 2003). From the MOZAIC project and for the examined period, 6 CO profiles exist collected over Rhodes during summer 2002 on the following dates: 28 July, 25 August, 1 September (2 profiles), 15 20 September and 22 September. One CO profile falls into the group of the highest ozone quartile (August 25) and two CO profiles fall into the group of the lowest ozone quartile (28 July and 15 September). The significant contribution of the lower atmospheric layers, influenced by the pollution emissions of the adjacent northern areas, to the boundary layer ozone at Rhodes during the highest ozone day (25 August 2002), is 25

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reflected in the vertical CO profile of the day shown in Fig. 7a. In comparison to the CO profile of a low ozone day (28 July 2002) with western (clean) circulation (Fig. 7b), which could be considered as CO background conditions, there is an increase of 30–50% in the 700 and 800 hPa pressure levels, which sharply increases even further after entering the boundary layer reaching the 250 ppb close to the surface (Fig. 7a).

The shape of the vertical CO profile during high ozone days is very similar to the relative increases (in %) of ozone at the various pressure levels between the low ozone and the high ozone days (Fig. 8). Summarising from that Figure at first an ozone large difference (about 20–25%) in the upper tropospheric levels is observed, which is associated to the presence of the anticyclone and the consequent downward flux of ozone from the upper troposphere during the high ozone days. This downward flow is expected to cause similar ozone increases in the lower tropospheric layers down to the surface. A further ozone difference in the 700–800 hPa levels (about 30–35% in total) comparable to the corresponding CO increase (Fig. 7) is observed, which could be attributed to photochemical production at these layers due to the presence of primary pollutants and favourable conditions for photochemical production induced by the high solar irradiation under the anticyclonic conditions. Finally an additional ozone sharp increase of the difference at the 900 hPa level, inside the boundary layer (about 55–60% in total), evident in the CO too, could be related to the boundary layer photochemistry, which is expected to be high as the northern boundary layer flow, induced by the “Etesian” winds is transporting considerable amounts of primary pollutants from the north (the Aegean coasts and the Balkans in the first place) under optimum conditions for ozone photochemical production.

3 Conclusions

Based on 77 MOZAIC flights recording vertical ozone profiles, the differences between Eastern Mediterranean and Central Europe showed significantly (10–12 ppb, 20–40%) enhanced ozone levels over the Eastern Mediterranean for the 1000 hPa to 900 hPa levels, whereas ozone between 700 hPa and 400 hPa was only slightly (3–5 ppb, 5–

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10%) higher than over Central Europe. The composite weather maps for the days with the highest ozone mixing ratios observed in the Mediterranean boundary layer suggest a strong influence of anticyclonic weather conditions. The lowest ozone levels are associated with a westward extension of the Middle East low and weak pressure gradients over the Eastern Mediterranean. Summer midlatitude anticyclones influence ozone in two ways: (i) downward transport from the upper troposphere and (ii) by creating stable conditions close to the surface, thereby hindering vertical dispersion of ozone precursors and enabling them to travel over long distances horizontally thus leading to regional photochemical activity in the boundary layer. Especially in the Aegean area, northern airflow due to the combined influence of the Central Mediterranean and the Balkans anticyclones with the Middle East low predominates in the boundary layer, thus leading to southward transport of polluted air from the Balkans and Eastern Europe. The above arguments are supported by the results of backward trajectory simulations for days with high and low ozone mixing ratios and by the few CO profiles available for the Eastern Mediterranean.

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Table 1. Average difference in ozone (in ppb) between Central Europe (Vienna, Frankfurt) and the Eastern Mediterranean (Heraklion, Rhodes, Antalya) for summer (JJAS). Relative differences (in percent) are given in parenthesis.

Standard Pressure Levels hPa	Eastern Mediterranean – Central Europe (77 profiles) ppb (%)
1000	12.2 (40.1)
900	9.4 (22.0)
800	5.9 (12.1)
700	3.6 (6.6)
600	5.3 (9.1)
500	4.9 (7.8)
400	4.1 (6.0)

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Table 2. Comparison of ozone averages between Central Europe (Vienna, Frankfurt) and the Eastern Mediterranean (Heraklion, Rhodes, Antalya) for the highest and lowest 25% of ozone concentrations at 900 hPa in the Eastern Mediterranean (19 profiles in each group).

Standard Pressure Levels hPa	Average ($\pm$ sd) ozone of the highest 25% cases in Eastern Mediterranean (corresponding values in Central Europe for the same flights) ppb	Average ($\pm$ sd) ozone of the lowest 25% cases in Eastern Mediterranean (corresponding values in Central Europe for the same flights) ppb
1000	51.7 $\pm$ 8.8 (38.4 $\pm$ 13.7)	36.7 $\pm$ 7.3 (24.9 $\pm$ 11.2)
900	62.8 $\pm$ 3.6 (49.6 $\pm$ 12.8)	40.0 $\pm$ 4.3 (36.2 $\pm$ 7.6)
800	62.0 $\pm$ 9.0 (55.9 $\pm$ 9.4)	46.6 $\pm$ 6.6 (42.6 $\pm$ 7.6)
700	64.6 $\pm$ 12.4 (59.3 $\pm$ 7.4)	49.5 $\pm$ 8.4 (48.9 $\pm$ 7.8)
600	67.9 $\pm$ 9.2 (64.0 $\pm$ 9.5)	55.8 $\pm$ 10.2 (53.3 $\pm$ 10.2)
500	75.7 $\pm$ 14.5 (65.4 $\pm$ 8.4)	61.6 $\pm$ 15.2 (60.4 $\pm$ 12.9)
400	85.2 $\pm$ 18.7 (73.4 $\pm$ 18.4)	66.5 $\pm$ 22.2 (63.8 $\pm$ 14.0)

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Table 3. Comparison of ozone averages between Central Europe (Vienna, Frankfurt) and the Eastern Mediterranean (Heraklion, Rhodes, Antalya) for the highest and lowest 25% of ozone concentrations at 900 hPa in Central Europe (17 profiles in each group).

Standard Pressure Levels hPa	Average ($\pm$ sd) ozone of the highest 25% cases in Central Europe (corresponding values in Eastern Mediterranean for the same flights) ppb	Average ($\pm$ sd) ozone of the lowest 25% cases in Central Europe (corresponding values in Eastern Mediterranean for the same flights) ppb
1000	47.0 $\pm$ 9.4 (52.2 $\pm$ 6.3)	17.2 $\pm$ 8.5 (36.8 $\pm$ 7.3)
900	59.9 $\pm$ 4.9 (59.8 $\pm$ 6.3)	29.1 $\pm$ 4.1 (48.1 $\pm$ 8.7)
800	61.3 $\pm$ 6.9 (58.8 $\pm$ 7.7)	40.8 $\pm$ 7.7 (55.7 $\pm$ 12.1)
700	61.4 $\pm$ 6.5 (63.8 $\pm$ 6.5)	48.4 $\pm$ 8.6 (59.3 $\pm$ 12.7)
600	62.3 $\pm$ 8.2 (69.0 $\pm$ 7.4)	55.9 $\pm$ 10.2 (66.0 $\pm$ 14.5)
500	62.7 $\pm$ 11.3 (77.5 $\pm$ 13.1)	62.6 $\pm$ 11.5 (63.9 $\pm$ 17.2)
400	71.5 $\pm$ 16.6 (88.6 $\pm$ 14.2)	68.7 $\pm$ 18.8 (64.5 $\pm$ 17.2)

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Table 4. Contribution of each atmospheric layer to the air arriving in Rhodes at 0–2 km on the high ozone days (4 cases) for 10-days and 5 days backward simulations.

	10 days	5 days
0–2	29.3%	40.5%
2–4	25.6%	29.2%
>4	45.1%	30.3%
Total	100%	100%

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Table 5. Contribution of each atmospheric layer to the air arriving in Rhodes at 0–2 km on the low ozone days (5 cases) for 10-days and 5 days backward simulations.

	10 days	5 days
0–2	38.6%	52.4%
2–4	33.6%	38.8%
>4	27.8%	8.8%
Total	100%	100%

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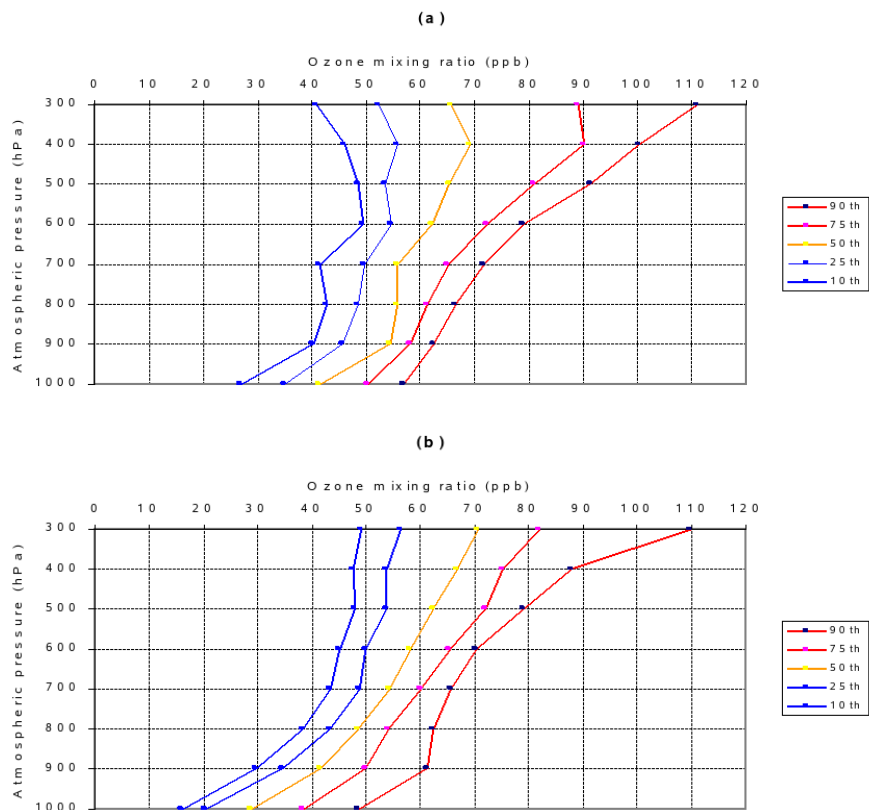


Fig. 1. Distribution statistics of summer (JJAS) vertical ozone averages at the standard pressure levels: **(a)** in the Eastern Mediterranean (77 profiles) and **(b)** in Central Europe (75 profiles).

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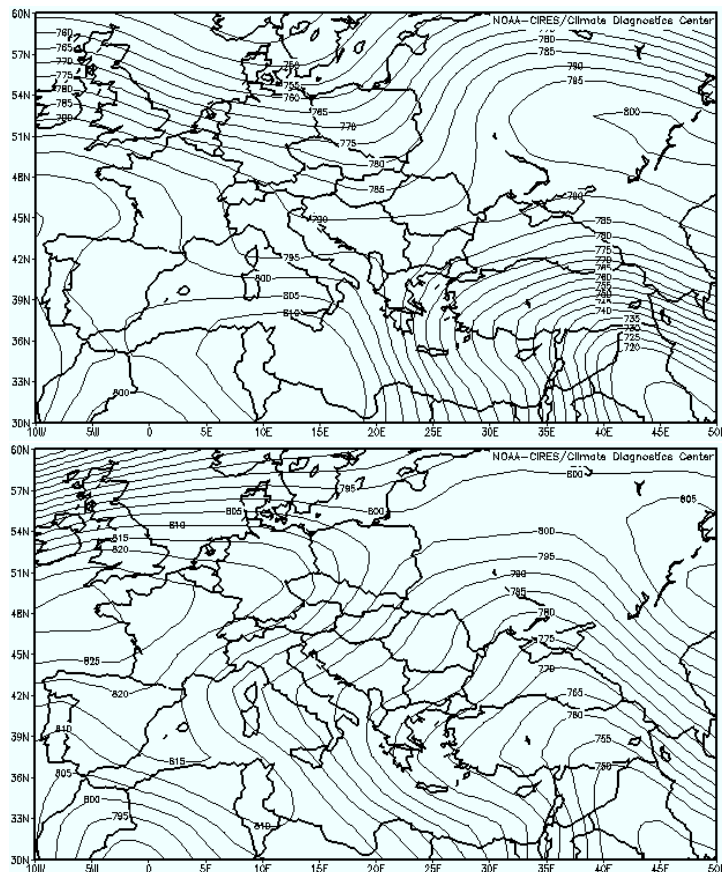


Fig. 2. Composite weather maps of geopotential heights at 925 hPa: **(a)** of the group of the 25% cases with highest ozone values at 900 hPa (upper panel), and (lower panel, **b**) of the group of the 25% cases with the lowest ozone values at 900 hPa in the Eastern Mediterranean (19 profiles in each group).

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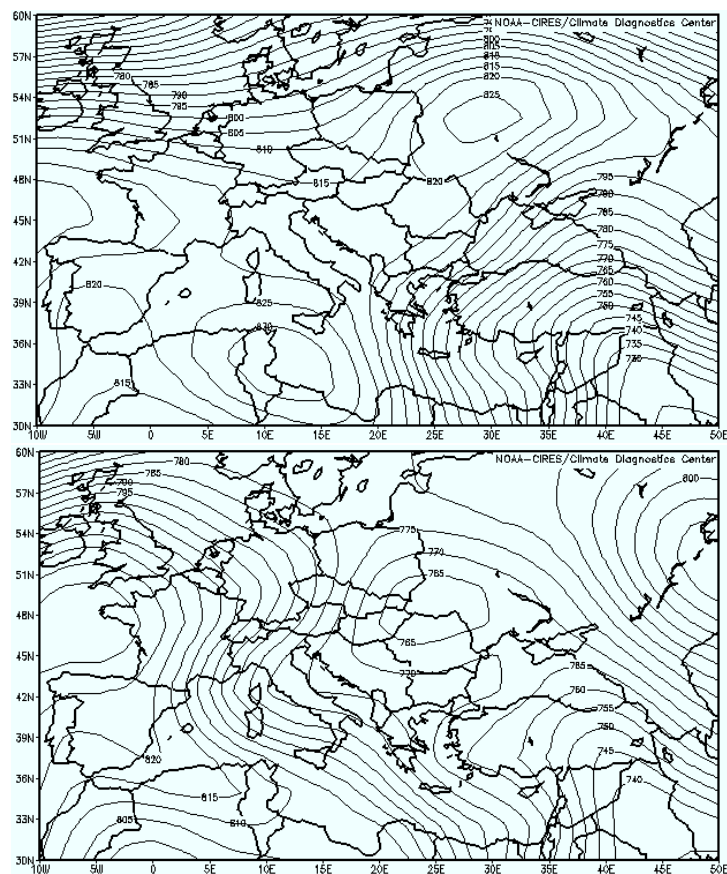


Fig. 3. Composite weather maps of geopotential heights at 925 hPa: (upper panel, **a**) of the group of the 25% highest ozone cases at 900 hPa, and (lower panel, **b**) of the group of the 25% lowest ozone cases at 900 hPa in the Central Europe (17 profiles in each group).

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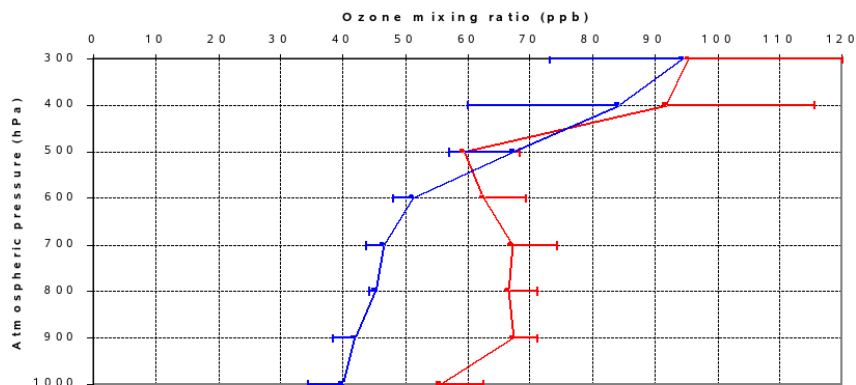


Fig. 4. Vertical ozone averages over Rhodes during the highest ozone days (4 cases, red line) and the lowest ozone days (5 cases, blue line) in summer 2002.

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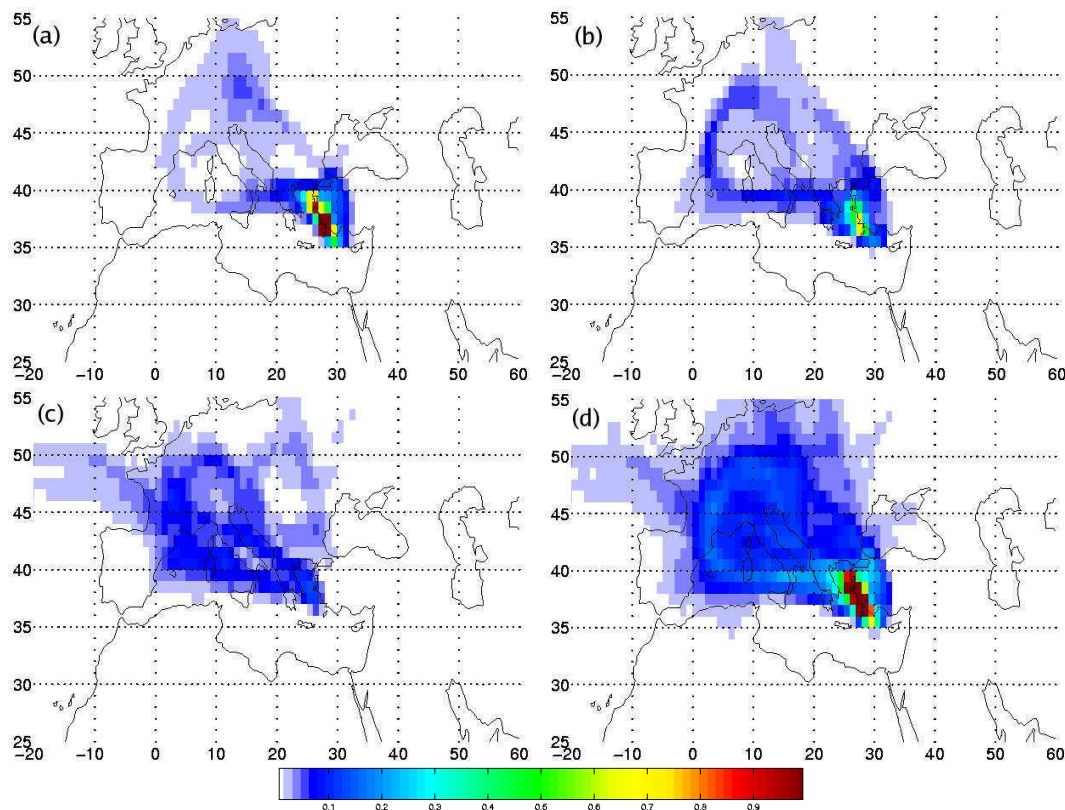


Fig. 5. Percentage contribution of geographical regions after 10-days FLEXPART backward simulation for the 4 high ozone cases and for the 0–2 km air parcels arriving at Rhodes. **(a)** for the 0–2 km layer, **(b)** for the 2–4 km layer, **(c)** for the >4 km layer and **(d)** for the total atmospheric column.

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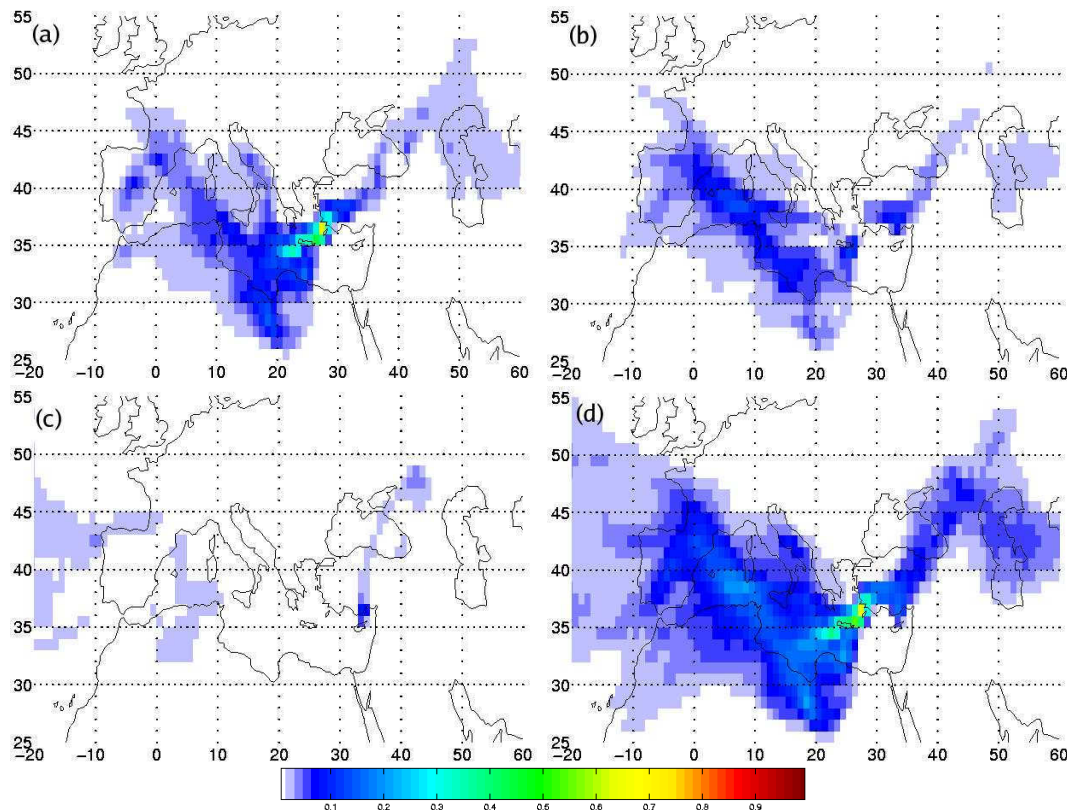


Fig. 6. Percentage contribution of geographical regions after 10-days FLEXPART backward simulation for the 5 low ozone cases and for the 0–2 km air parcels arriving at Rhodes. **(a)** for the 0–2 km layer, **(b)** for the 2–4 km layer, **(c)** for the >4 km layer and **(d)** for the total atmospheric column.

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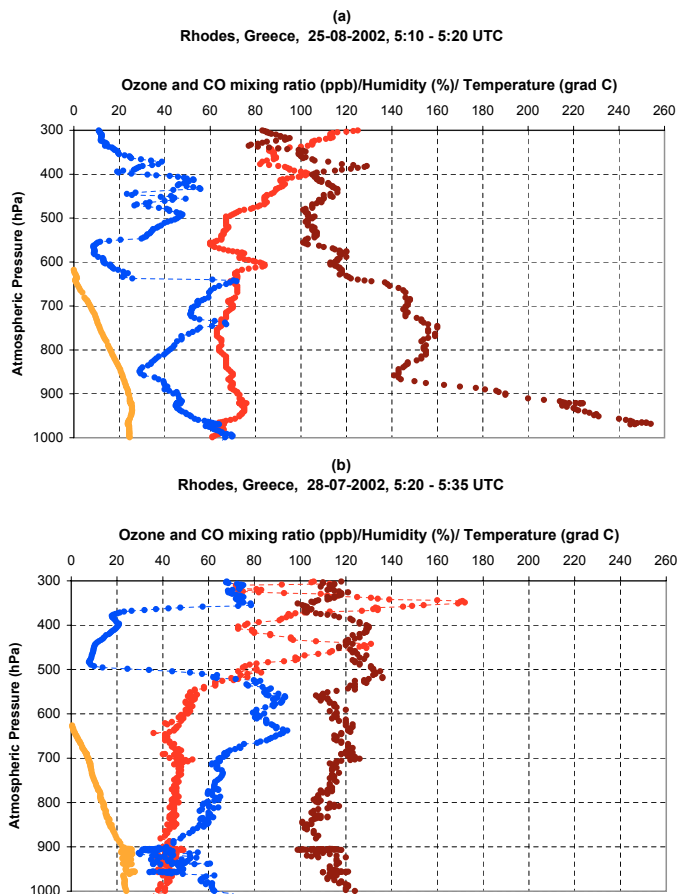


Fig. 7. Vertical profiles at Rhodes for a high ozone day (upper panel, **a**) and a low ozone day (lower panel, **b**), of CO (brown), ozone (red), relative humidity (blue) and temperature (orange).

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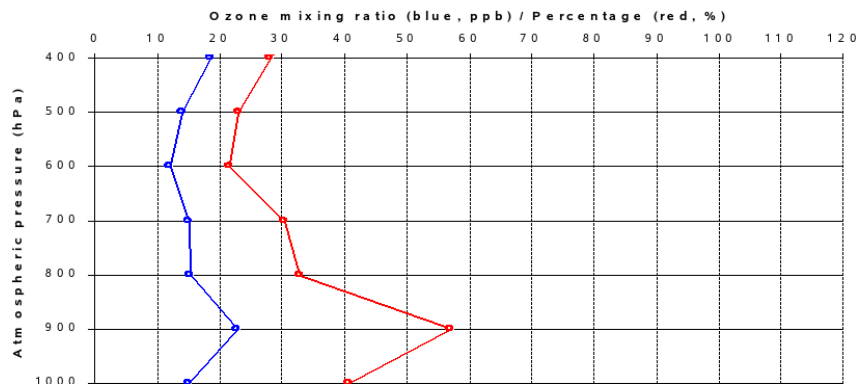


Fig. 8. Average vertical ozone summer (JJAS) absolute (blue, in ppb) and relative (red, in %) ozone differences in the Eastern Mediterranean (Heraklion-Rhodes-Antalya) between the 25% of the days with the highest ozone values at 925 hPa and the 25% of the days with the lowest ozone values (19 profiles in each group).

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