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OBSERVATIONAL EVIDENCE OF AN OZONE EPISODE OVER THE GREATER ATHENS AREA

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Abstract—Research flights have been performed over the Greater Athens Area (GAA) and southwards over the Island of Aegina and east of Peloponnisos in order to investigate the evolution of an ozone episode over GAA and the transportation of the urban pollution plume southwards from the Athens region. During the 3 day period of 6 July to 8 July 1994, the GAA was under the influence of an ozone episode with ground-level noontime concentrations of more than 120 ppbv. Upper-air ozone concentrations measured during the flights were as high as 135 ppbv. The interaction of the weak synoptic conditions over the area along with the development of a mesoscale thermal circulation created poor dispersion conditions during the period of interest and resulted in elevated ozone levels. The primary pollutants emitted in the GAA during the night and early morning hours, were funneled out to the Saronic Gulf and southwards along the southwestern Aegean Sea, near the coast of east Peloponnisos. Under the influence of strong sunlight these primary pollutants continued to undergo photochemical reaction giving rise to elevated ozone levels tens of kilometers downwind of the pollution emission sources. Further evidence of the photochemically-aged air masses was the high correlation ($R^2 = 0.8$) observed between NO_y and ozone. The ozone production efficiency in these transported air masses reached a value of close to six. © 1997 Elsevier Science Ltd.

Key word index: Ozone, nitrogen oxides, transportation, photochemical activity, Athens.

1. INTRODUCTION

The air-quality problem in the Greater Athens Area (GAA) which has received increasing attention during the last three decades, is primarily due to a population shift, increased industrialization and continuously increasing motorized fleet. Analysis of air-quality data available for several years in the Athens region has shown that ozone and nitrogen dioxide levels exceeded both national and EU standards on a significant number of days each year (Kallos *et al.*, 1993; Kassomenos *et al.*, 1995). In an attempt to improve air quality in central Athens, the Ministry of Environment imposes restrictions over the GAA during air pollution episodes. More precisely, on certain days private transportation is completely banned in central Athens and industrial activity in the GAA is reduced.

The GAA includes the cities of Athens, Pireus and their suburbs. Figure 1 shows that this area is located in a Basin which is surrounded by mountains on three sides and open to the sea with the Saronic Gulf on the fourth side. The main axis of the Basin is 25 km long, oriented south–southwest to north–northeast and its area is approximately 450 km^2. There are three main mountains around the Basin: Hymettus (1050 m) to the east, Pendeli (1100 m) to the north–northeast, and Parnitha (1400 m) to the north–northwest. Mountain Aegaleo, with an elevation of 450 m, is located to the west of the Basin.

The main sources of air pollution in the GAA are automobiles, industry and also central heating during the cold months. The GAA contains about 30% of the Greek industry (mainly small-to-medium-scale factories) and about 1.2 million automobiles, that comprise about 50% of all automobiles in Greece. The main industrial area is concentrated at the south–southwest edge of the Basin. Other important sources of air pollution are the Pireus harbor located to the west–southwest, and the Helinikon airport at the south–southeast edge of the Basin. Within the Athens Basin there are no large industrial sources with tall stacks. Significant industrial activity exists on the Thriassion Plain, west of Mountain Aegaleo. In the western part of the Thriassion Plain (near the Isthmus

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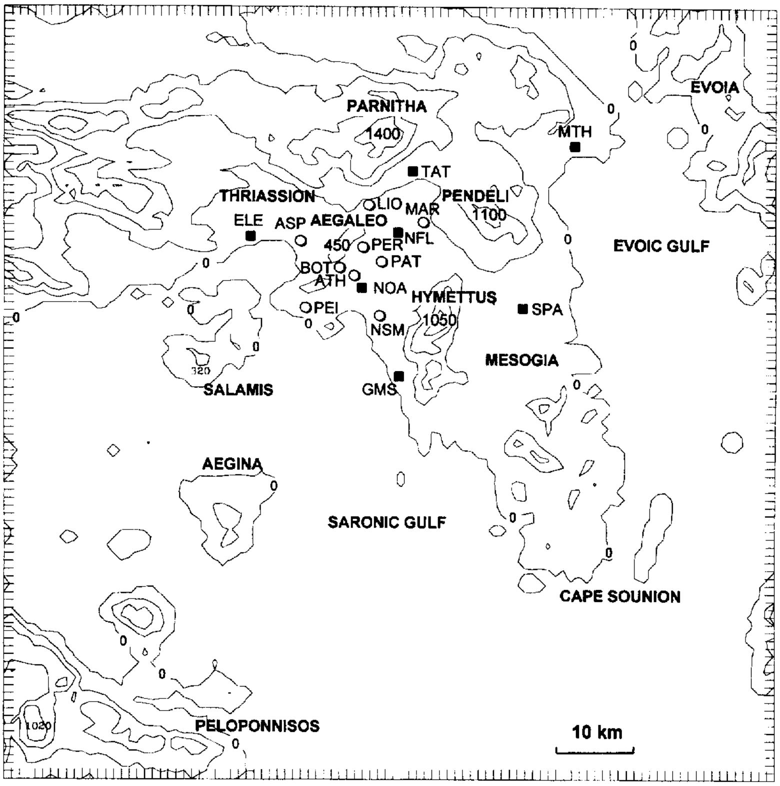

Fig. 1. Map of the Greater Athens Area. Elevations are every 300 m. Squares and circles indicate the location of the meteorological and air-quality surface stations, respectively.

of Korinthos) there are three refineries, two cement plants, a steel smelter, shipyards and a series of smaller industrial units. One of the refineries is located at the foothill of Aegaleo with stack heights varying from 40 to 120 m. The air pollutants released from these stacks often pass over Mountain Aegaleo and travel towards the Athens Basin (Asimakopoulos *et al.*, 1992; Kassomenos *et al.*, 1994). A major power plant is located southeast of Athens at the edge of the Attica Peninsula (at Cape Sounion). This power plant consists of several petrol burning units (thermoelectric units, turbines) with stacks varying between 40 and 140 m. As noted by Kallos *et al.* (1995), this power plant does not significantly contribute to the air pollution degradation of Athens. Another power plant is located at the central part of the island of Evoia at the Evoic Gulf (Aliveri). The air pollutants released from these stacks are directed over the Athens Basin under certain synoptic conditions, especially when relatively strong northerly wind flow prevail (Kallos *et al.*, 1995).

Atmospheric circulation within the Athens Basin, which determines good or poor ventilation in the area, is controlled by the interaction of different scales: synoptic, regional, meso and microscale. Generally, during the warm and dry period of the year (June through September), a high-pressure system covers the Mediterranean Sea and the surrounding area and a low-pressure system prevails over the Anatolian Plateau. The relative strength and extension of the anticyclonic system over the Mediterranean and Balkan area and the thermal low over the Anatolian Plateau, define the flow pattern over the Aegean Sea. A northerly flow regime is observed for most of the days during the summer period across the Aegean Sea. The strength of this northerly flow depends on the strength of the pressure gradient resulting from the interaction of the two synoptic-scale systems. When the pressure gradient is weak, the synoptic circulation is weak from the north favoring the development of local circulations and thus leading, in general, to poor dispersion conditions within the GAA. On the other hand, when the pressure gradient is strong, northerly winds (stronger during the day and weaker during the night) dominate, creating good ventilation in the Athens Basin (Kallos *et al.*, 1993; Pilinis *et al.*, 1993; Kassomenos *et al.*, 1995).

In the framework of the present paper, the collected meteorological and air-quality observations are analyzed. More specifically the ground and research aircraft measurements of ozone formation over the GAA are reported, and ozone transport southwards over the Saronic Gulf and the eastern border of Peloponnisos is studied. Section 2 is devoted to the description of the experimental settings. In Section 3 the meteorological conditions characterizing the case study are presented, while the surface and airborne air-quality observations are presented in Section 4. The last section is devoted to discussion of the results.

2. EXPERIMENTAL SETTINGS

A two-engine light aircraft (Cessna 310) was outfitted to perform continuous air-quality monitoring during flights. The instrumental package consisted of sulfur dioxide (TEII model 43S), nitrogen oxides (TEII model 42S) and ozone (Dasibi model 1008-AH) monitors. The sensitivity of the SO_2 and NO_y monitors was ±0.2 ppbv while for ozone it was ±1 ppbv. Calibration spans and zero checks were performed before the flights and the sulfur dioxide monitor was zeroed during the research flights using a PbO scrubber. Each monitor had its own Teflon sampling inlet which was located on the top front of the aircraft in order to prevent any contamination from the aircraft exhaust gases. The altitude of the aircraft was monitored using a MKS Baratron pressure gauge, and temperature and relative humidity were measured with a Campbell Scientific model 207 air-temperature and relative humidity probe. The aircraft position was continuously recorded using a Magellan NAV 5000A GPS unit. All instruments were operated

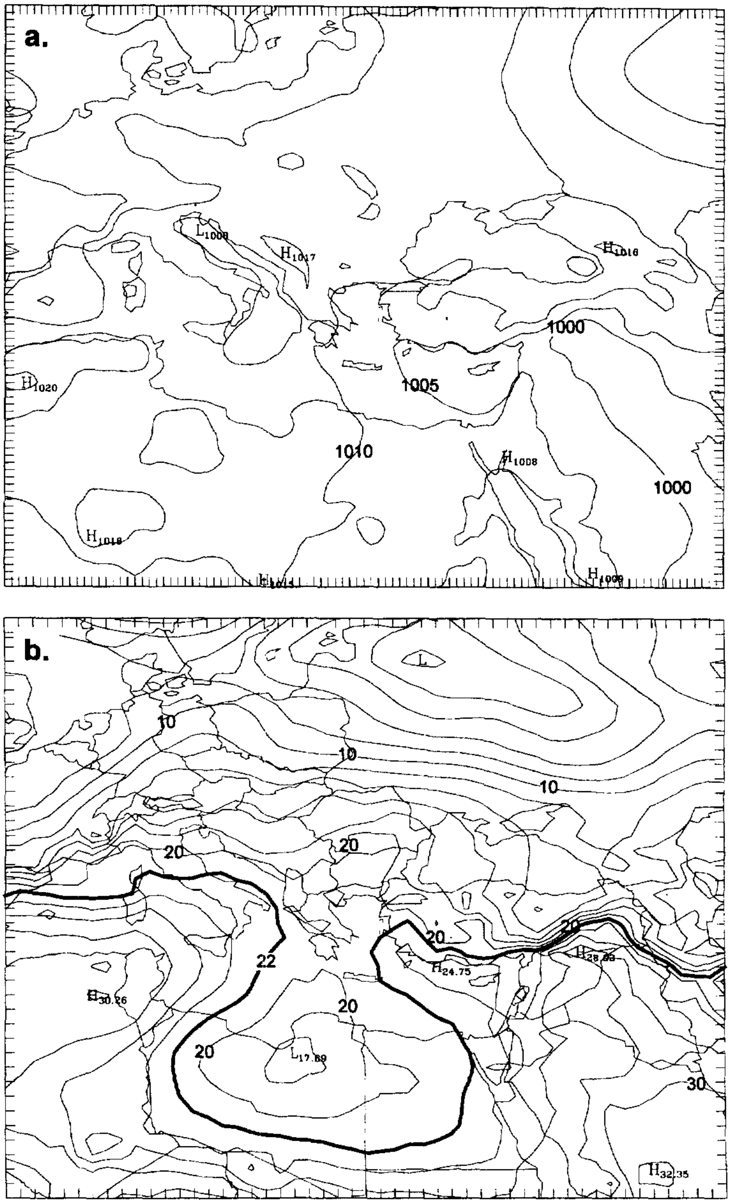

Fig. 2. Weather charts of: (a) surface pressure analysis at 0000 UTC, 7 July 1994, at 5 hPa interval; (b) isotherms at 850 hPa level, at 0000 UTC, 6 July 1994; (c) isotherms at 850 hPa level, at 0000 UTC, 7 July 1994. Isotherms are plotted every 2°C. The 22°C isotherm is given with a solid line.

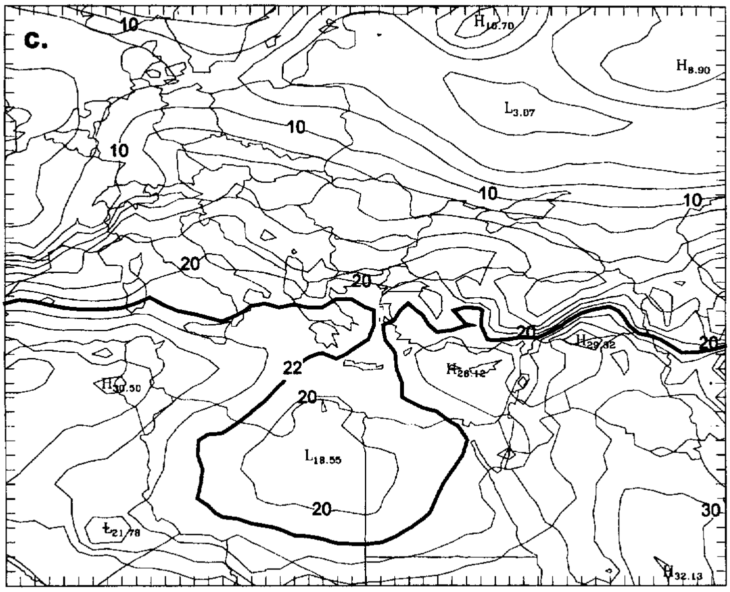

Fig. 2. (*continued*).

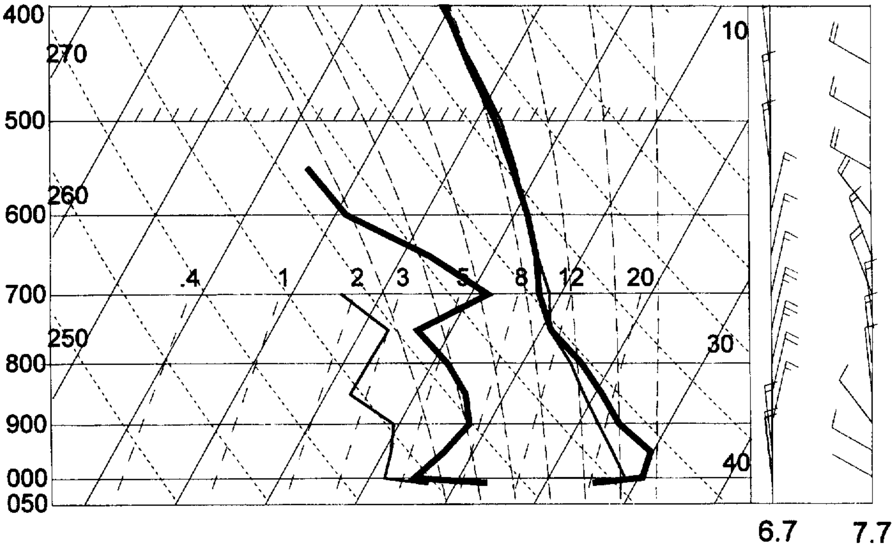

Fig. 3. Upper-air sounding at GMS upper-air station (see Fig. 1) at 0000 UTC 6 July 1994 (thin line) and 0000 UTC 7 July 1994 (solid line). One barb equals 5 m s^{-1}, one half-barb 2.5 m s^{-1}.

via an Avionics Instruments static inverter that converted the 24 VDC provided by the aircraft to 110/220 VAC. All of the data, collected every 30 s, were directed to a Campbell 21X datalogger which was connected to a portable PC.

3. WEATHER CONDITIONS

During the period of interest, between 6 and 8 July 1994, the Mediterranean region was characterized by stagnant conditions. As it can be seen in the surface analysis chart at 0000 UTC, 7 July 1994 (Fig. 2a), an extended anticyclonic regime dominated the greater part of the Mediterranean region and Europe, giving rise to light winds. This regime along with the lower pressures over the Anatolian Plateau resulted in a weak pressure gradient over Dardanelles and the Aegean Sea, and benign flow. This anticyclonic regime is also evident on the 850 and 700 hPa charts. As it can be seen in Figs 2b and c, there is a warming of about 2°C between 0000 UTC 6 July and 0000 UTC 7 July at the 850 hPa level. This warming is due to the transport of warm air masses from northern Africa towards the Greek Peninsula. The region over the Aegean Sea was characterized by a weak pressure gradient resulting in a light northerly flow at the

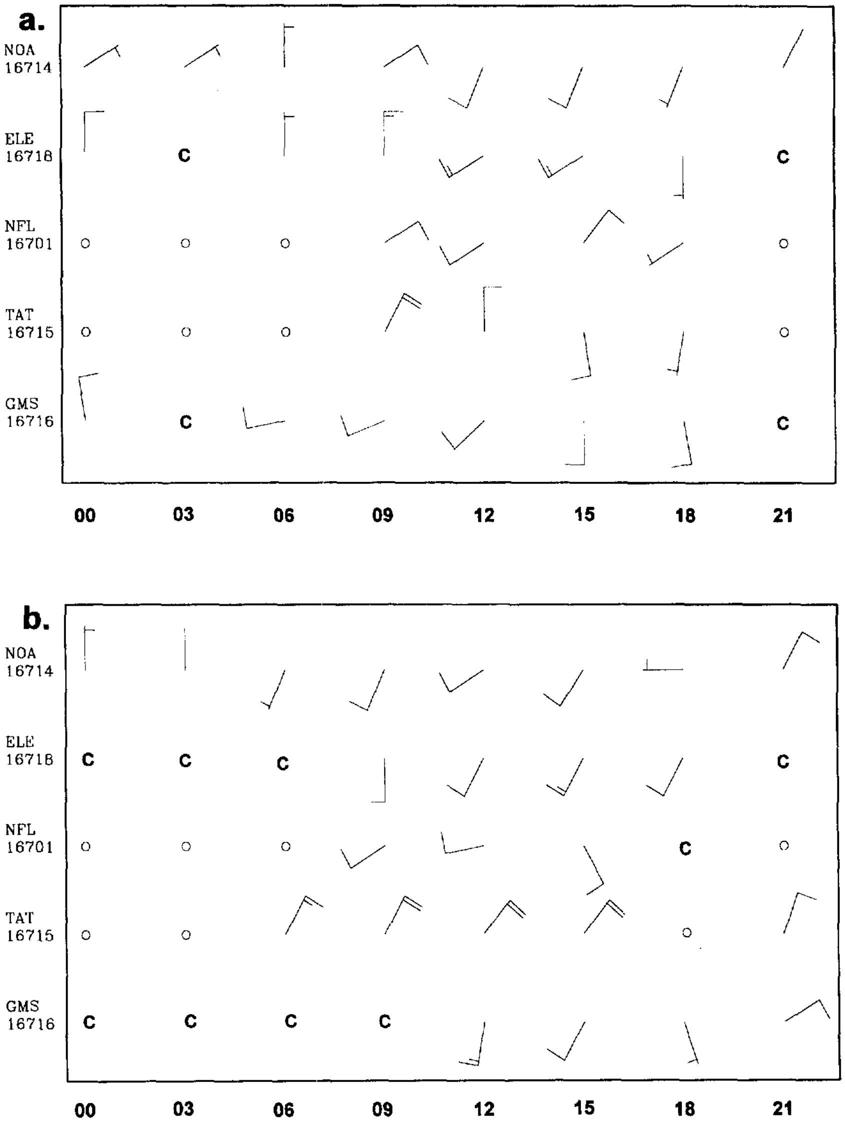

Fig. 4. Temporal variation of surface winds recorded by the surface meteorological network at the National Observatory of Athens (NOA), Eleusis (ELE), N. Filadelfia (NFL), Tatoi (TAT), and Greek Meteorological Service at Helinikon Airport (GMS) on (a) 6 July, (b) 7 July 1994. Letter c denotes calms, while open circles denote missing data. The hours in the time scale are in UTC (see Fig. 1 for the station location). One barb equals 4 m s^{-1}, one half-barb 2 m s^{-1}.

surface as well as aloft. This weak synoptic flow allows the development of mesoscale thermal circulations in the area of interest (e.g. sea/land breezes, upslope/downslope winds, drainage flows). This development was not associated with condensation and cloudiness in the region over and around Athens. These local-scale circulations influence the transport and dispersion of air pollutants over the GAA, forming, in general, poor dispersion conditions over the Attica Peninsula and Saronic Gulf (Kallos *et al.*, 1993).

Inspection of the sounding at 0000 UTC, 7 July 1994 (Fig. 3) shows the presence of a surface inversion of about 5°C over a layer of 500 m. Moreover, a warming of about 2°C is evident in the layer 900–800 hPa during the 24 h period between 0000 UTC, 6 July and 0000 UTC, 7 July 1994 (Fig. 3). The surface inversion is a common feature during summer due to lack of vegetation and rain. This surface inversion becomes deeper when the synoptic flow favors the transport of warm air masses in the lower-tropospheric layers. The anticyclonic regime prevailing on 6 June, favored the transport of warmer air masses from northern Africa over the Greek Peninsula, as observed in Fig. 2, resulting in the 2°C warming and the deepening of the inversion also evident in Fig. 3. These warm air masses were initially transported over the Central Mediterranean and Italy and then over Greece. This is a well-known path followed by warm air masses (Metaxas and Kallos, 1980). As shown in Fig. 4, the meteorological surface network in the GAA recorded light winds or calms in the early morning and late afternoon hours while during noon the development of sea breezes is evident in the coastal stations (note the southwesterly flow observed at the GMS and ELE stations at 1200 UTC). The aforementioned warming in the lower-tropospheric layers results in the suppression of the vertical development of the local thermal circulations and in the formation of a relatively shallow mixing layer. Under weak synoptic flow the local thermal circulations define the transport and dispersion of air pollutants over the GAA which are in general poor, at least for a few hours during the late morning and early afternoon. When the above situation is associated with warming at the lower-tropospheric layers, the dispersion conditions

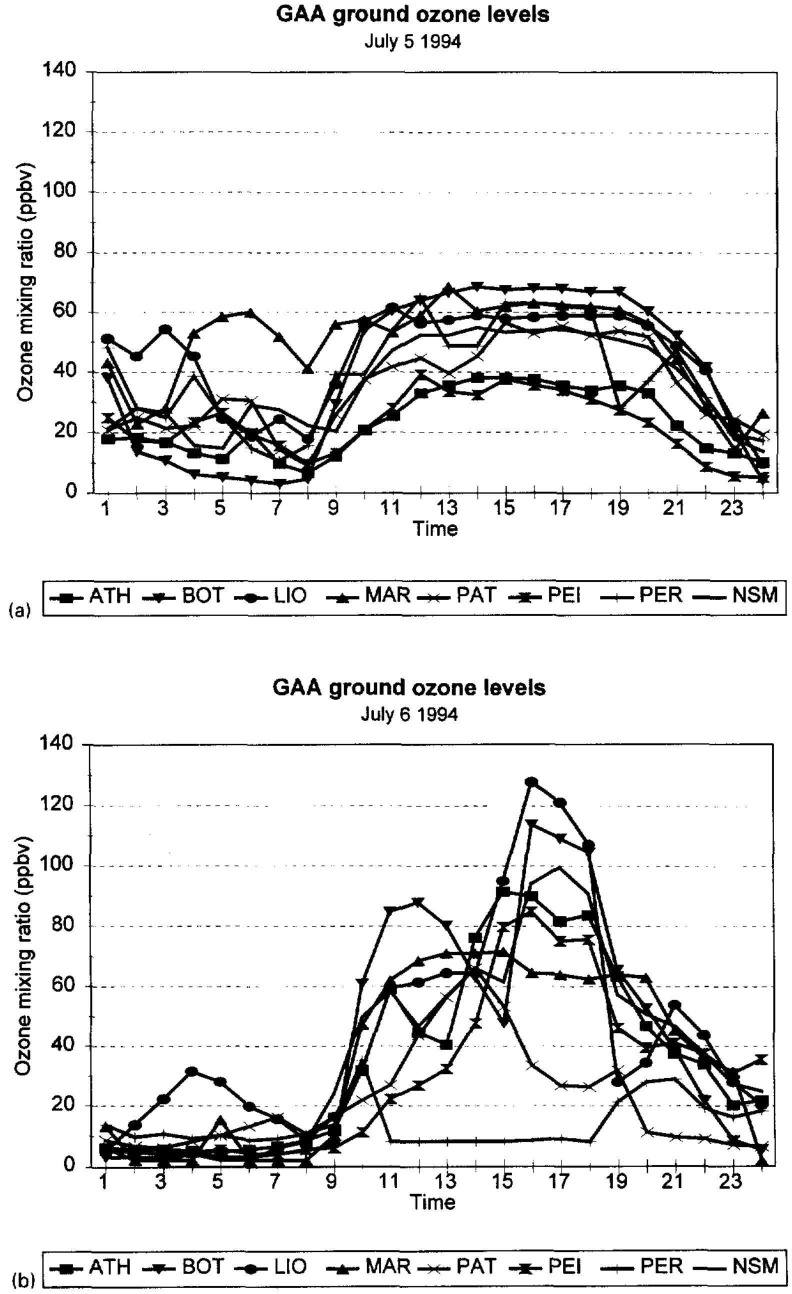

Fig. 5. Surface ozone levels as measured at the air-quality stations: Patisson (PAT); Athens (ATH); Geoponiki (BOT); Maroussi (MAR); Liossion (LIO); N. Smirini (NSM); Peristeri (PER) and Pireus (PEI), for (a) 5 July; (b) 6 July; (c) 7 July and (d) 8 July 1994. The hours in the time scale are in LT (see Fig. 1 for the station location).

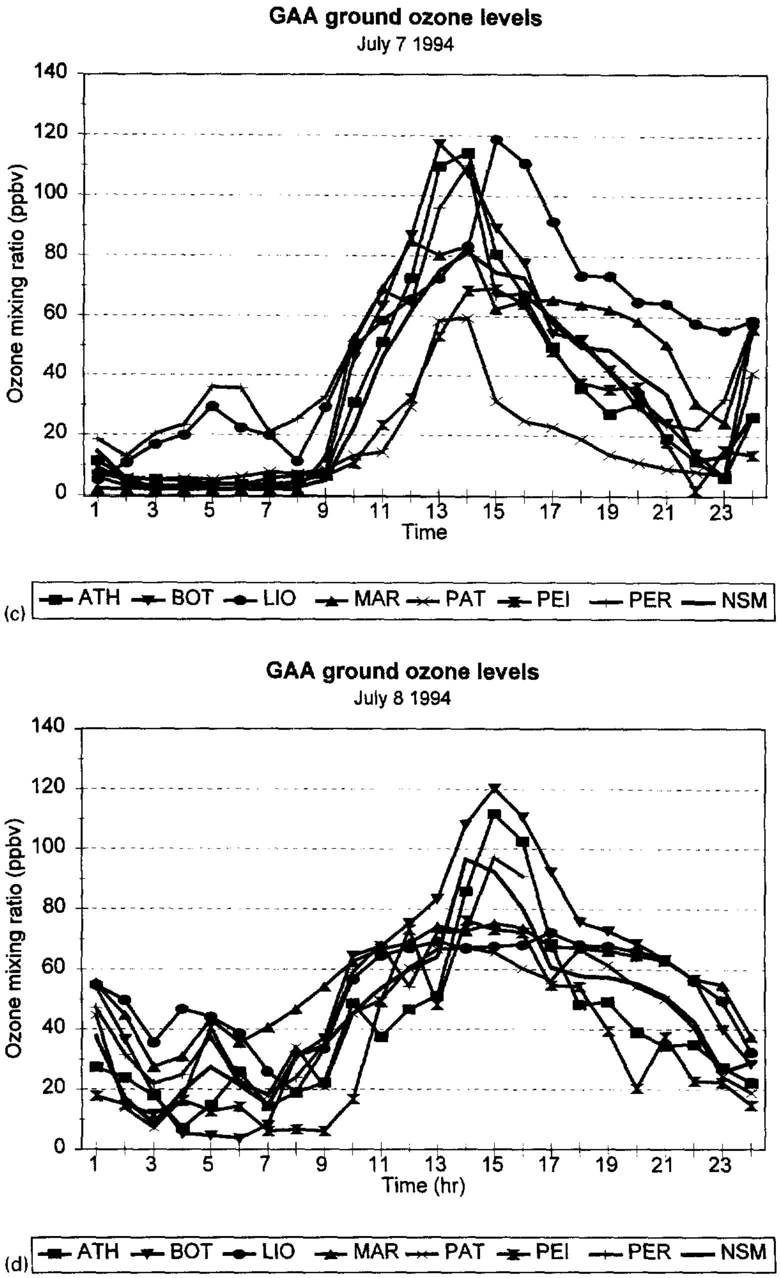

Fig. 5. (*continued*).

degrade causing air pollution episodes, which was the case between 6 and 8 July 1994.

4. AIR-QUALITY OBSERVATIONS

4.1. *Surface observations*

The ozone levels measured at the ground stations during the period 5 July through 8 July 1994 are shown in Fig. 5a–d (note that LT = UTC + 3 h). For the first day of the period (5 July), ozone concentrations were below 70 ppbv at all the measuring sites. The highest values were reported for the center of Athens and the lowest levels (35 ppbv) at the coastal site of Piraeus. Further, the levels at all of the sites changed little between 1000 and 1900 LT. From the above, it is apparent that for 5 July, the area was not under the influence of an ozone episode.

A sharp increase in the ozone levels was observed during the late afternoon of the following day (6 July), with levels above 120 ppbv. On 7 July, maximum ozone concentrations up to 120 ppbv were also encountered between noon and 1600 LT. Ozone levels of up to 120 ppbv were also observed on 8 July at about 1500 LT. For the next few days, the GAA continued to be affected by elevated ozone levels and only on 12

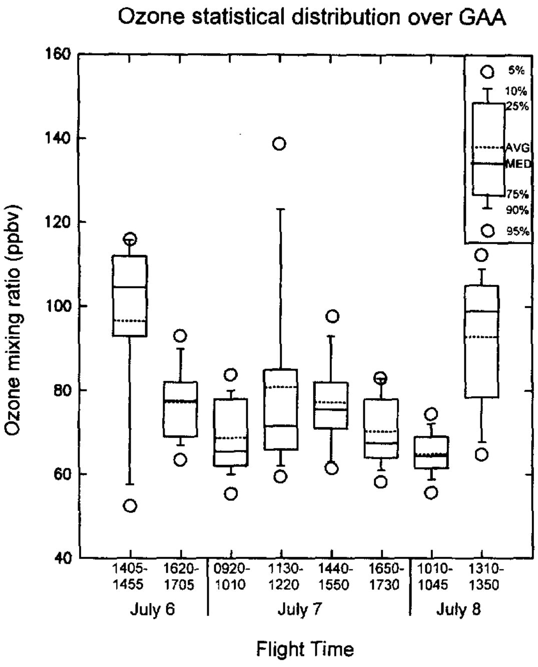

Fig. 6. Ozone statistical distribution levels measured during the eight flights over GAA from 6 to 8 July 1994 (see Table 1 for flight information). The inset explains the limits of the box and whiskers distribution levels.

Table 1. Summary of flights over GAA

Date	Time	Value	SO_2	NO_x	NO	O_3
6 July	1405–1455	Min.	5.7	9.2	0.4	52
		Max.	14.0	24.2	1.0	118
		Avg.	10.0	16.8	0.6	97
6 July	1620–1705	Min.	4.4	6.9	0.2	61
		Max.	6.3	17.6	0.7	90
		Avg.	5.3	10.0	0.4	74
7 July	0920–1010	Min.	7.4	5.4	0.06	49
		Max.	26.8	50.2	10.3	88
		Avg.	12.8	33.8	4.6	69
7 July	1130–1220	Min.	12.2	9.8	0.4	62
		Max.	22.0	42.6	1.9	134
		Avg.	17.6	18.2	0.9	85
7 July	1440–1550	Min.	4.8	3.5	0.2	55
		Max.	8.2	16.7	1.1	105
		Avg.	6.4	9.4	0.6	79
7 July	1650–1730	Min.	4.7	5.6	0.3	56
		Max.	7.0	14.5	0.8	84
		Avg.	5.5	9.3	0.5	70
8 July	1010–1045	Min.	4.1	4.9	0.5	56
		Max.	5.5	15.5	1.5	76
		Avg.	4.5	9.3	1.0	65
8 July	1310–1350	Min.	3.5	6.8	0.1	66
		Max.	7.3	19.9	0.6	118
		Avg.	5.5	11.7	0.3	93

July did the ozone concentration decrease to values below 80 ppbv.

4.2. *Airborne measurements*

From the noon of 6 July 1994 through noon 8 July 1994, eight research flights were performed over the GAA and continuing southwards over the Isle of Aegina and parallel to the eastern side of the Peloponnisos. Over the GAA the flight height was between 450 and 700 m (1400–2100 ft) above mean sea level (AMSL), although a number of measurements were performed up to a height of 2000 m. Over the sea the

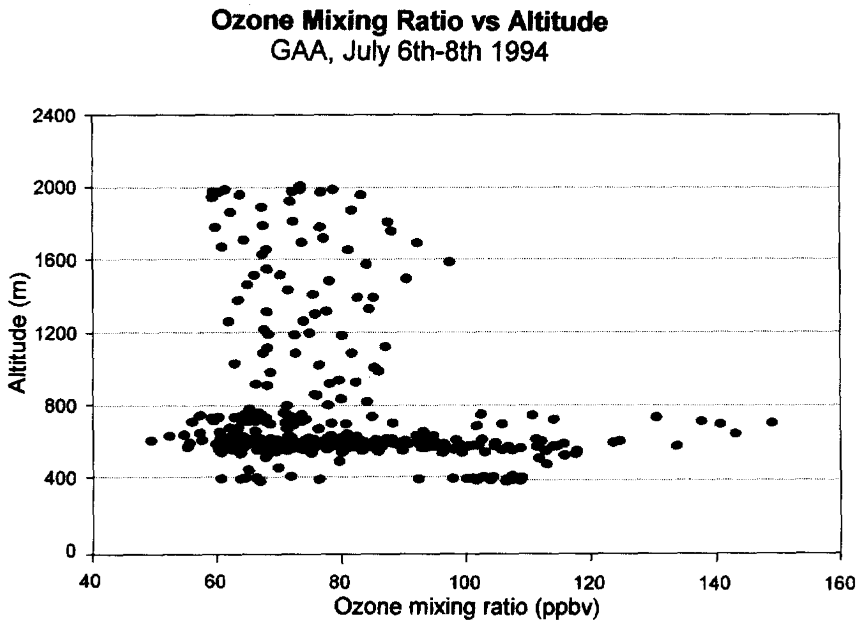

Fig. 7. Ozone levels variations versus altitude, between 400 and 2000 m, during the eight flights over GAA from 6 to 8 July 1994.

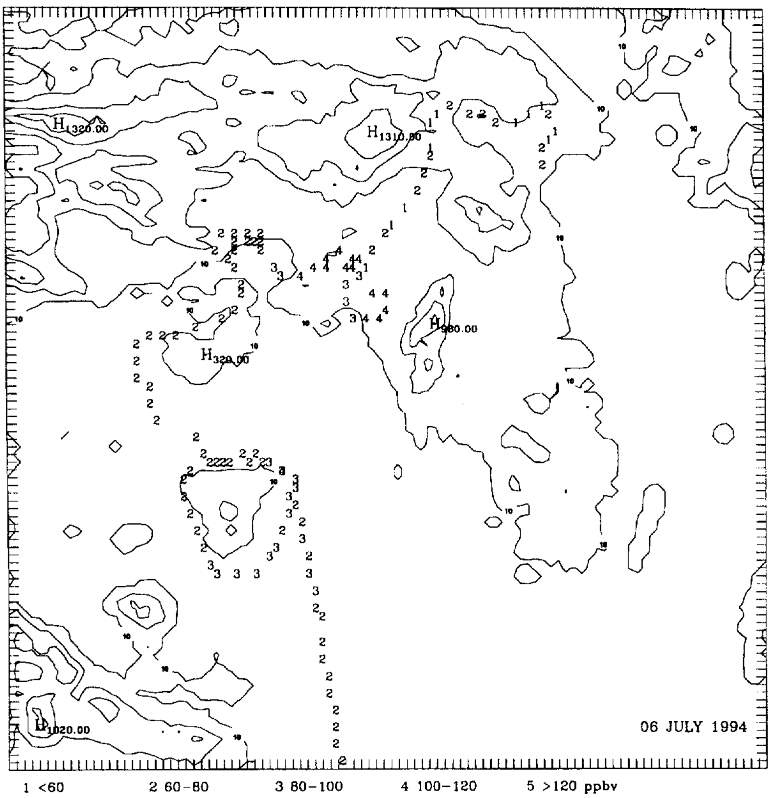

Fig. 8. Ozone concentrations measured by the research aircraft on 6 July 1994 for the time period 1400–1515 LT. (The numbers 1–5 indicate the ozone concentrations <60, 60–80, 80–100, 100–120 and >120, respectively.)

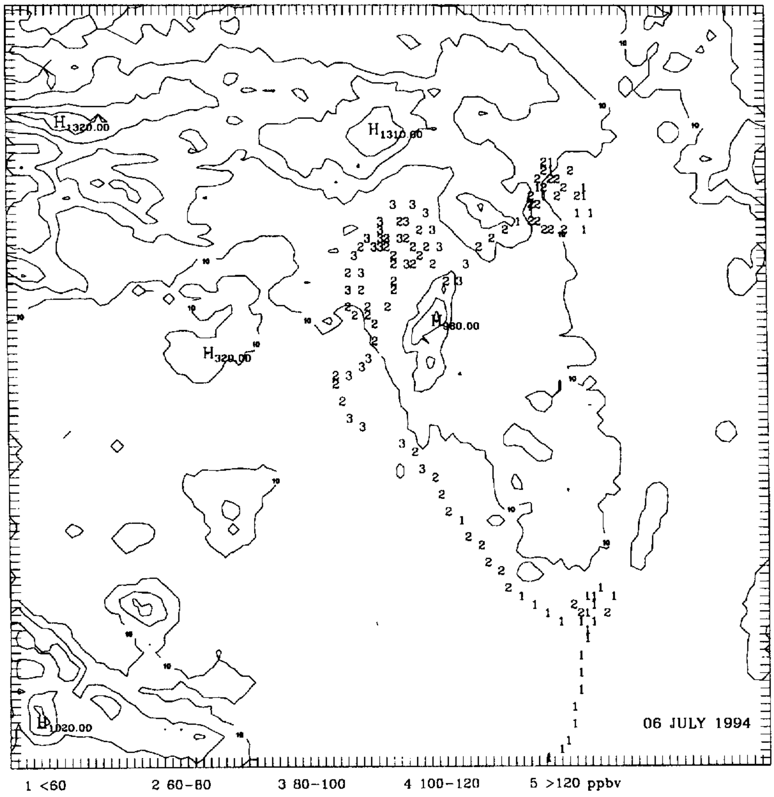

Fig. 9. Ozone concentrations measured by the research aircraft on 6 July 1994 for the time period 1600–1715 LT. (The numbers 1–5 indicate the ozone concentrations <60, 60–80, 80–100, 100–120 and >120, respectively.)

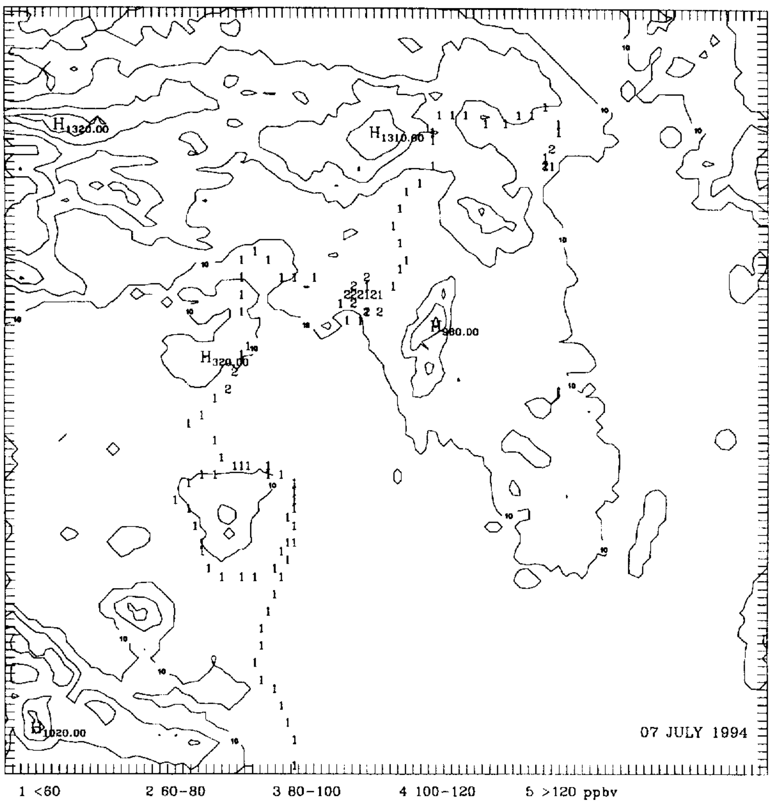

Fig. 10. Ozone concentrations measured by the research aircraft on 7 July 1994, for the time period 0900–1030 LT. (The numbers 1–5 indicate the ozone concentrations <60, 60–80, 80–100, 100–120 and >120, respectively.)

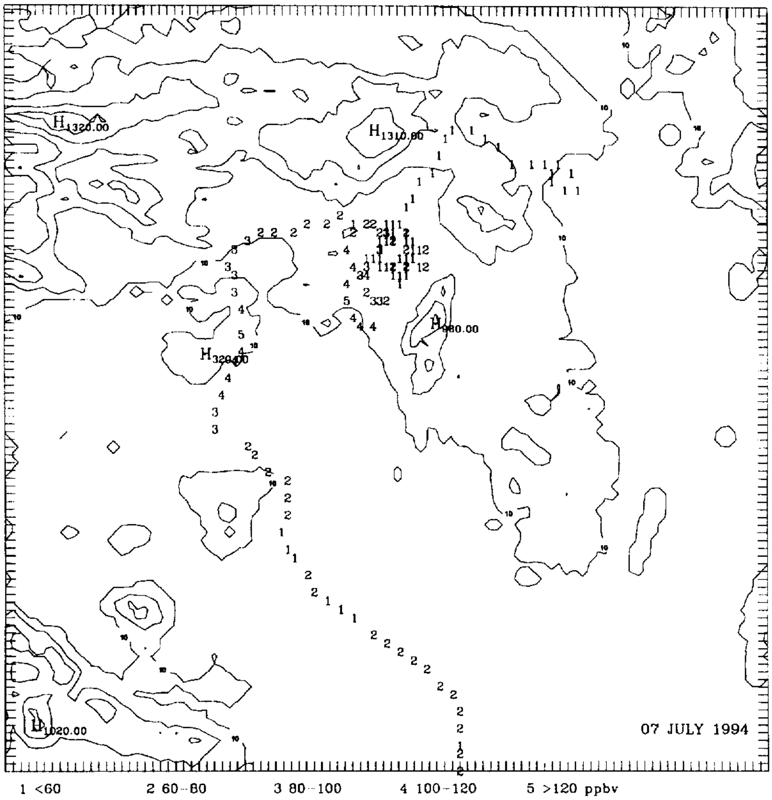

Fig. 11. Ozone concentrations measured by the research aircraft on 7 July 1994, for the time period 1100–1230 LT. (The numbers 1–5 indicate the ozone concentrations <60, 60–80, 80–100, 100–120 and >120, respectively.)

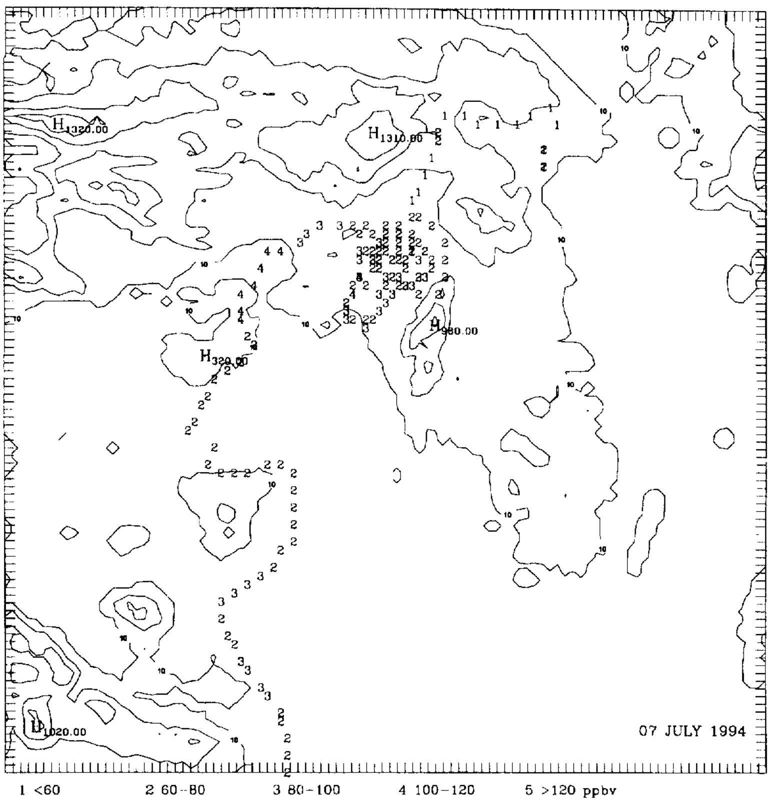

Fig. 12. Ozone concentrations measured by the research aircraft on 7 July 1994, for the time period 1430–1600 LT. (The numbers 1–5 indicate the ozone concentrations <60, 60–80, 80–100, 100–120 and >120, respectively.)

flight level was constant at around 300 m. A summary of the pollution levels for the eight flights limited over GAA is shown in Table 1. The area defined for the flight study as GAA stretched from Piraeus/Keratsini in the west to Hymettus in the east and from Glifada in the south to Kamatero/Metamorfossi in the north. This area covers approximately a circle of 25 km centered at downtown Athens (Zapio/Acropolis). Figure 6 shows the statistical distribution plot of the ozone levels measured during the eight flights over the area. The elevated SO_2 levels recorded during some of the flights (especially for 7 July in the morning) were due to entrapment of the local emission sources. The relatively high nitrogen oxide (NO_y) levels measured during the flights were caused by automobile emissions, especially those recorded during the morning flights which corresponded to peak traffic times. The low NO content measured indicates that most of the NO_y is already present as NO_2 indicating that photochemical activity is effective.

The effect of altitude on the ozone levels is demonstrated in Fig. 7 which shows the measured ozone concentrations at various elevations. While the highest concentrations are confined to the first 700 m AMSL (below the mixing layer), it is obvious that ozone reaches the upper layers (2000 m) at levels of 100 ppbv. This is due to the quick warming of the lowest atmospheric layers and transport from the mixing layer to the free troposphere.

A major question has been what happens to the urban pollution cloud produced over the GAA during an ozone episode. Inspection of Figs 8–15 reporting the airborne measurements of the flights between 6 to 8 July over GAA and continuing southwards over the Saronic Gulf and Aegean Sea can provide some clues to the above question. The afternoon flight on 6 July (1405–1455) showed ozone levels up to 120 ppbv over central Athens while in the late afternoon (1620–1705) the levels dropped below 100 ppbv—see Figs 8 and 9. The morning flight of 7 July (0920–1010) reported ozone levels of not more than 80 ppbv over Athens while for all the other areas the ozone levels were below 60 ppbv (Fig. 10). For the noontime flight (1130–1220), the ozone concentrations over both the western part of central Athens and Salamis exceeded 120 ppbv and over Aegina reached 80 ppbv (Fig. 11). The early afternoon flight (1440–1550) showed a decrease of O_3 to less than 100 ppbv over Athens, while around Salamis ozone levels reached 120 ppbv and even south of Aegina values of almost 100 ppbv were observed (see Fig. 12). The last flight for that day (1650–1730) showed ozone levels below 80 ppbv for the GAA while around and south of the Isle of Aegina ozone concentrations up to 100 ppbv were still observed (Fig. 13).

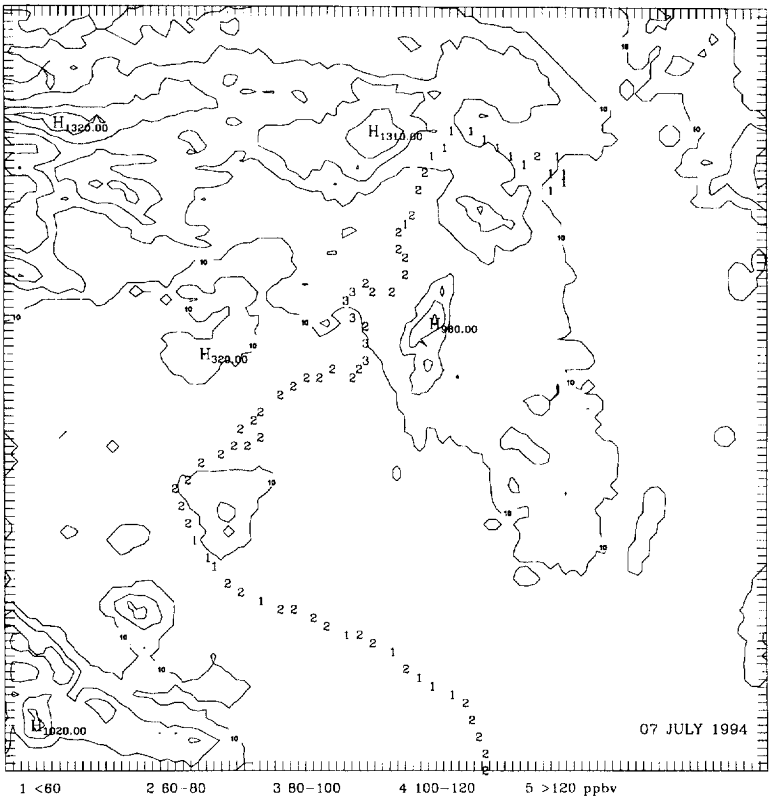

Fig. 13. Ozone concentrations measured by the research aircraft on 7 July 1994, for the time period 1630–1745 LT. (The numbers 1–5 indicate the ozone concentrations <60, 60–80, 80–100, 100–120 and >120, respectively.)

For the first flight on 8 July (1010–1045) ozone levels did not exceed 80 ppbv over GAA while around and south of Aegina values of up to 100 ppbv were measured (see Fig. 14). For the noontime flight (1310–1350), although ozone levels over the GAA reached 100 ppbv, high concentrations of up to 120 ppbv were recorded over the sea between Aegina and the main peninsula. South of Aegina values of up to 100 ppbv were still observed (see Fig. 15).

5. DISCUSSION

The ozone levels observed both from the surface and airborne measurements indicate that GAA was under the influence of an ozone air pollution episode during the period 6–8 July 1994. The airborne observations further indicate that the air masses released over the GAA can be traced out over the Saronic Gulf and southwards along the Southwestern Aegean, near the coast of east Peloponnisos. Since these air masses contain primary pollutants, photochemical activity continues to occur and elevated ozone levels were observed some 50–60 km, or 3–4 h travel time, from the pollutant sources, i.e. the Athens region. This delay in peak ozone formation is consistent with earlier observations indicating a 3.5–6 h delay between the release of the primary pollutants and peak ozone formation (Seinfeld, 1989; Luria *et al.*, 1992).

The photochemical processes responsible for ozone formation are the oxidation of reactive organic gases (ROG) in the presence of nitrogen oxides. While the ROG are consumed in these processes, the NO_y acts as a catalyst. During the summer and in urban regions, ozone production is limited primarily by the supply of NO_y (Trainer *et al.*, 1987, 1993; Jacob *et al.*, 1993). The correlation between ozone and total reactive nitrogen (NO_y) at times of high-photochemical activity has been used to determine the extent of photochemical activity and the ozone production rate efficiency (Fahey *et al.*, 1986; Trainer *et al.*, 1993).

Figure 16 shows the correlation between ozone and NO_y for the air mass over the sea between Aegina and the main peninsula for three different time periods during the flights on 8 July. For the first part of the flight during early morning, the path was over GAA and intensive photochemical activity had not as yet begun. During this period, the ozone levels did not rise above 80 ppbv, NO levels were as high as 2 ppbv and the ozone production rate was limited and close to unity. The correlation between ozone and NO_y was limited with $R^2 = 0.4$, as shown in Fig. 16. For the flight period over the open sea (1100–1330) no fresh

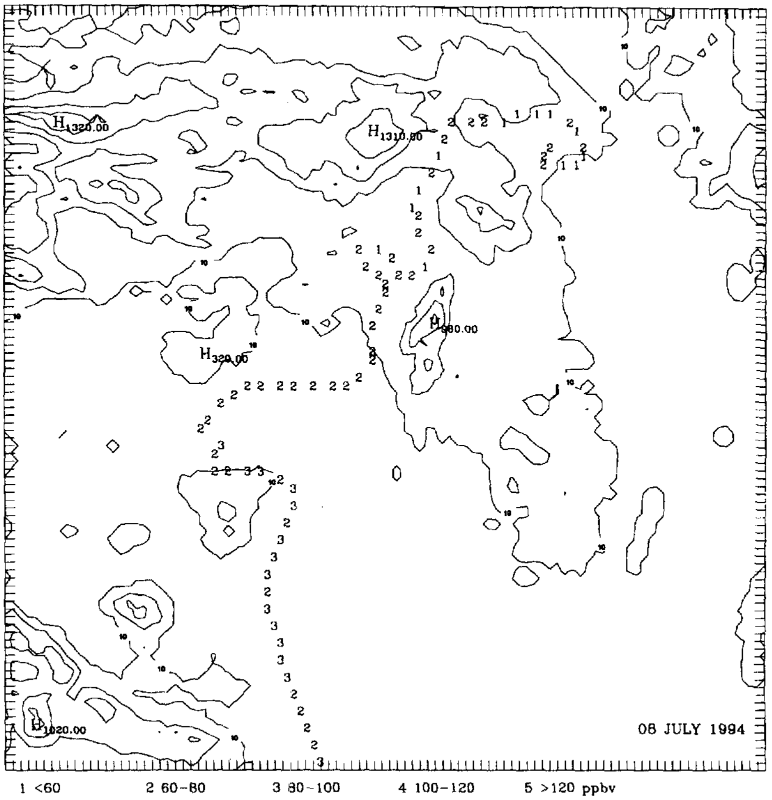

Fig. 14. Ozone concentrations measured by the research aircraft on 8 July 1994, for the time period 1000–1130 LT. (The numbers 1–5 indicate the ozone concentrations <60, 60–80, 80–100, 100–120 and >120, respectively.)

nitrogen oxides emissions were encountered and NO levels dropped to below 0.4 ppbv. Good correlation between NO_y and O_3 was observed with R^2 having a value of 0.8. The slope of the regression was almost six, indicating that six ozone molecules are formed for each NO_y molecule present. The observed intercept of almost 40 is close to previously reported baseline ozone mixing ratios (see Peleg *et al.*, 1994). The highest ozone levels were recorded during the return portion of the flight towards midday, when photochemical activity was enhanced and the air mass travel time was about 3 h. For the final part of the flight, between 1330 and 1400 over GAA, fresh pollution was entrapped in the air mass and correlation between O_3 and NO_y was once again limited ($R^2 = 0.4$), as observed in Fig. 16. Ozone production per NO_y molecule was now less than three.

The above results show good agreement with previous studies which measured elevated ozone levels over rural areas downwind of large urban areas as previously reported by a number of investigators (Olszyna *et al.*, 1994; Peleg *et al.*, 1994, 1995, among others). Studies by Imhoff *et al.* (1995) and Sillman (1995) have indicated that in an urban plume downwind from Atlanta, ozone increases consistently with increasing NO_y, as the NO_y levels varied between 4 and 12 ppbv and that O_3 remains constant or decreases when the NO_y is greater than 12 ppbv. The ozone production rate per NO_y present was about eight when the NO_y levels were below 12 ppbv but dropped to almost one when the NO_y values were higher.

The results obtained in the present investigation (see Fig. 16) indicate that the polluted air masses that originate over GAA remain photochemically active as they are swept south. Ozone continues to be produced during transportation giving rise to elevated O_3 levels tens of kilometers away from pollution emission sources. Further, since the highest ozone productivity yield observed during the flights was only six, it is obvious that the air masses were not photochemically exhausted. Thus, even higher ozone levels, than those measured in the present study, are to be expected as the air masses continue their travel.

Test simulations of this ozone episode have been performed using a meteorological model combined with a Lagrangian dispersion model. Preliminary results showed that the recirculation mechanism is partially responsible for this air-pollution episode. More specifically it was found that the urban plume of Athens of 6 July is dispersed all the way down the eastern coast of Peloponnisos, and a part of it returned back to Athens on 7 July. This is in good agreement with the airborne observations. This

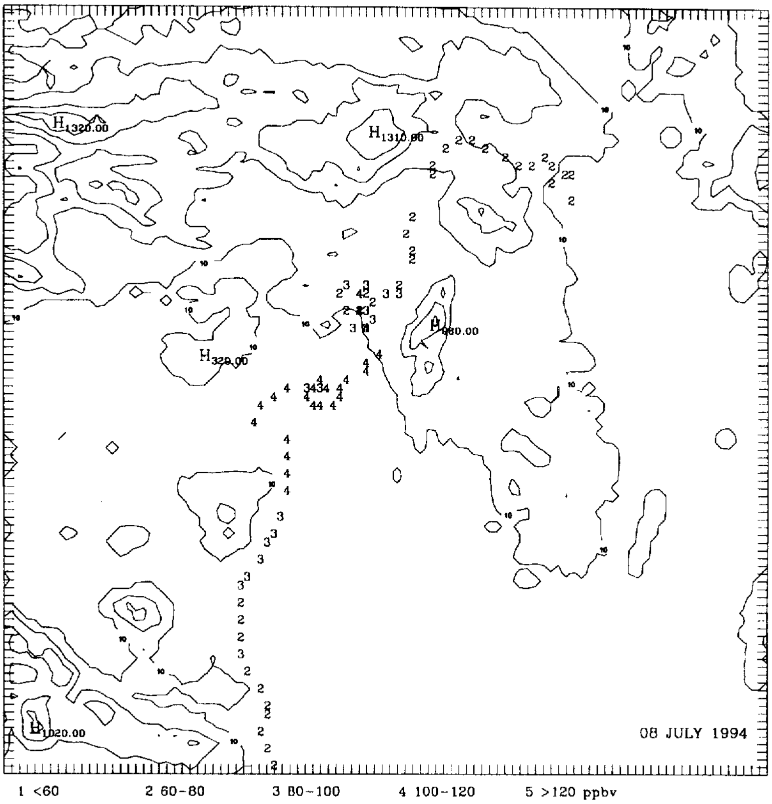

Fig. 15. Ozone concentrations measured by the research aircraft on 8 July 1994, for the time period 1230–1400 LT. (The numbers 1–5 indicate the ozone concentrations <60, 60–80, 80–100, 100–120 and >120, respectively.)

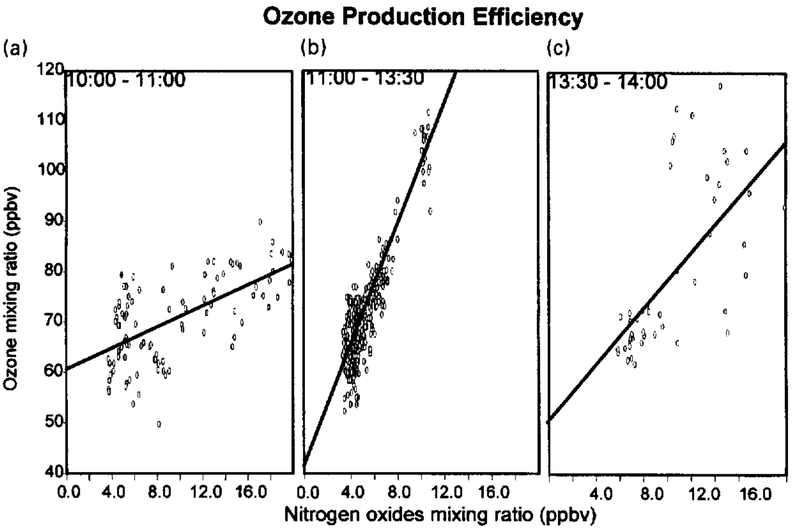

Figure 16. Correlation between ozone and nitrogen oxides over the Aegean Sea for three segments: (a) 1000–1100 LT; (b) 1100–1330 LT and (c) 1330–1400 LT for flight 8B on 8 July 1994.

modeling study is now under way and will be the subject of a future publication.

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