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Atmospheric sulfur flux rates to and from Israel

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Abstract

Both field measurements and model simulation studies have shown that Israel is the recipient of long range transported air pollutants that originated over various parts of Europe. The present paper presents results of aircraft measurements aimed at quantitizing the sulfur flux arriving at Israel's western coast from Europe and the Israeli pollution contribution to the air masses leaving its eastern borders towards Jordan. During the research flights, measurements of sulfur dioxide and sulfate particulates and meteorological data were recorded. Two different legs were performed for each research flight: one over the Mediterranean Sea, west of the coast and the second along the Jordan Valley. All flights were carried out at a height of approximately 300 m above ground level. A total of 14 research flights were performed covering the summer and autumn seasons. The results indicate that the influx of sulfur arriving at the Israeli coast from Europe varied in the range of 1–30 mg S/h, depending on the measuring season. The particulate sulfate level in the incoming LRT air masses was at least 50% of the total sulfur content. The contribution of the local pollutant sources to the outgoing easterly fluxes also varies strongly according to season. During the early and late summer, the Israeli sources contributed an average of 25 mg S/h to the total pollution flux as compared to only approximately 9 mg S/h during the autumn period. Synoptic analysis indicates that conditions during the summer in Israel favor the accumulation of pollution species above the Mediterranean basin from upwind European sources. This season features a shallow mixed layer and weak zonal flow leads to poor ventilation rates, inhibiting an efficient dispersion of these pollutants while being transported eastward. Under these conditions, in flux, local contribution and the total out-flux of these pollutants are elevated as opposed to during other seasons. During the fall, the eastern Mediterranean region is usually subjected to weak easterly winds, interrupted at times by strong westerly wind flows inducing higher ventilation rates. These meteorological conditions and the lack of major emitting sources eastwards of Israel result in lower sulfur budgets to and from Israel for this season. An estimate of the yearly flux showed that approximately 0.06 tg S arrived at the Israeli coast from the west. This is approximately 15% of the estimated pollution leaving Europe towards the eastern edge of the Mediterranean basin. The local contribution to the out-flux towards Jordan was calculated to be 0.13 tg S per year, almost all of the sulfur air pollutants emitted in Israel.

Keywords: Sulfur; Flux rates; Israel; Synoptic conditions; Ventilation rates

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1. Introduction

Long-range transport (LRT) of air pollutants within both North America and Europe has been widely studied while almost no attention has been paid to the Eastern Mediterranean region, and especially, the region between Greece, Turkey and Israel. Al-Mormani and Aygun (1998) and Gullu et al. (1998) have recently reported measurements of particulate sulfate for a site on the Mediterranean coast of Turkey. A number of studies have suggested that the eastern part of the Mediterranean Sea may be influenced by relatively high levels of particulate sulfate (Luria et al., 1989, 1996; Benkovitz et al., 1994). An observational and modeling study has confirmed that Israel is indeed affected by the long-range transport of air pollutants originating over Europe (Wanger et al., 2000). A study by Luria et al. (1996) has shown that these long-range transported air masses arriving in Israel from the west have high sulfate content levels. While variations in wind direction are observed on a diurnal and seasonal basis depending on the synoptic conditions affecting the region, the predominant wind flows over Israel are from the west towards the east. This implies that air pollutants emitted to the west of the eastern Mediterranean will reach Israel from upwind sources and will be added to those emitted locally. Both contributions will then be advected eastward.

The local pollution sources will be limited due to the fact that Israel is a very small country although possessing a densely populated central region. Israel is situated at the eastern edge of the Mediterranean Sea with a total area less than 22 000 km^2 with almost all the large urban regions and industrial and commercial activity (electricity, industry, etc.) limited to central Israel which has an area of only 7000–8000 km^2. The total anthropogenic annual emissions for Israel for 1996 were estimated as 0.2 tg sulfur corresponding to emission rates of approximately 21 ton per km^2 for central Israel (Gabbay, 1998). The Israeli contribution to total world emissions for sulfur is of the order of 0.1%.

The present study reports an attempt at quantitizing the amount of sulfur (SO_2 and SO_4^{2-}) compounds reaching Israel from upwind European sources as well as the amount exiting Israel along its eastern boarder towards Jordan.

2. Experimental

Aircraft research flights were performed in an attempt to characterize air masses entering and leaving Israel. The instrument package installed in the aircraft consisted of a high sensitivity SO_2 analyzer (TEII 43S, pulsed fluorescence method, ± 0.1 ppbv sensitivity), a semi-automatic sulfate aerosol sampler, together with temperature, relative humidity and pressure-altitude sensors. The aerosol sampler consisted of a five-filter holder system, facing backwards and fitted outside the aircraft so that the air sampled came directly in contact with the filters without interference of contaminating materials. During each flight, a dynamic blank was taken in addition to the actual aerosol samples taken during the different flight legs. The filter used for collecting the sulfate particulate was a Teflon filter (Millipore FALP 047, 1 μm pore size). The process of sulfate particle collection, even below micrometer size, is not solely by sieving but includes other mechanisms such as static forces, impaction, caking, etc. The sampling flow rates were continuously monitored and recorded using a mass flow meter. After aerosol extraction from the filters, sulfate analysis was performed using an ion chromatographic technique (Baltensperger and Hertz, 1985). The zero levels of the monitor were verified both on the ground and in the air using a PbO scrubber for SO_2. Span calibrations were performed on the ground before take off. A GPS (Global Positioning System) was used to continuously monitor the position of the aircraft during the research flight. The GPS data, together with data available from the aircraft instruments, were used to calculate wind speed and direction at various positions along the flight path. The data were recorded every 10 s and stored on a datalogger (Campbell Scientific Inc. model 21X).

Two paths were performed during each research flight: one approximately 70 km offshore, parallel and west of the Israeli coastline with Tel Aviv in

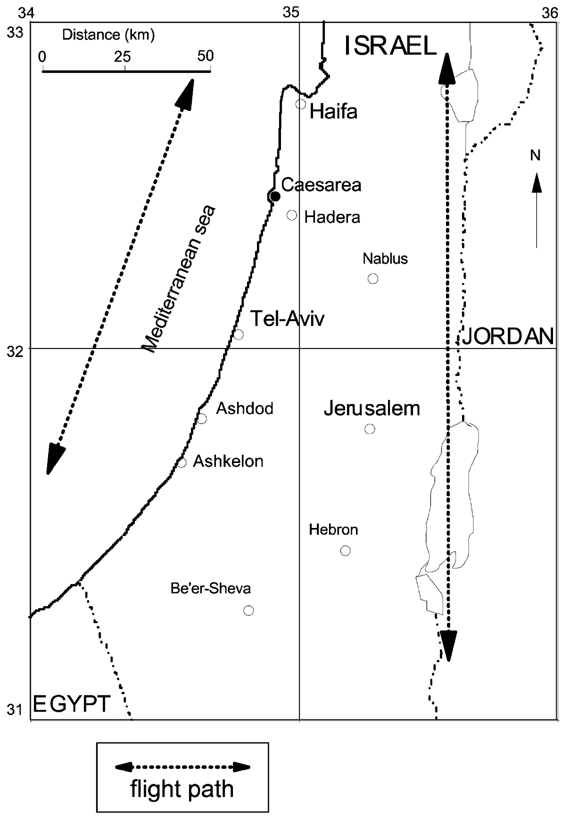

Fig. 1. Map of Central Israel showing flight paths.

the center, and the other from the Dead Sea continuing along the Jordan Valley up to the Lake of Galilee (see Fig. 1). Each leg was between 180 and 200 km in length and allowed for approximately 1 h filter sampling time. All flights were performed at midday and at altitudes of approximately 300 m above ground level, well within the boundary-mixing layer. The mixing layer height was determined for the western and eastern flight legs from measurements of temperature and relative humidity performed while climbing up to a height of approximately 1500 m and compared to radiosonde data measured routinely, twice a day, over the central coast of Israel. Measurements were performed during two different seasons, early (27 May–4 June, 1998, six flights) and late (3–10 September 1996, four flights) summer and for autumn (27–30 November, 1995, four flights).

3. Results

Results of airborne measurements for 29 May, 1998 are shown in Fig. 2 and are typical for the early summer period in Israel. The results are superimposed upon a map of central Israel. The figures show the pollution concentration levels for SO_2 as measured to the west of the Israeli coast and over the eastern border with Jordan. Fig. 3 shows upper airflow patterns as wind vectors for the three different aircraft measuring campaigns. As is observed in Fig. 3a for 29 May, 1998, the wind flow was mainly westerly over all of central Israel. A similar pattern was observed for all the other days of the early summer measuring period. Typical results representing the late summer period are shown in Fig. 3b for 3 September, 1996. During this research period the wind flows were also, in general, from the west and northwest towards the east and southeast. Fig. 3c shows the airflow patterns as recorded along the flight legs for 29 November, 1995, representing the fall research period. As is observed, the wind flows were from the east or northeast as opposed to the predominant westerly flows typified by summer.

In order to assess the atmospheric sulfur budgets to and from Israel, the pollution flux at the west and east borders of central Israel was calculated based on the flight measurements using the following equation:

$$F = C*V*L*H*3.6/1000 \tag{1}$$

where

- F — sulfur flux in mass per unit time (mg/h);
- C — concentration of sulfur (g/m^3);
- V — vector component of wind speed (m/s) in eastern direction;
- L — length of each flight leg (m); and
- H — mixing layer height (m).

The concentration for the sulfur compounds includes the sum of sulfur dioxide and sulfate presented as total sulfur. The factor 3.6/1000 converts the flux from units of g/s to mg/h. The results of the flux rates for the sulfur compounds on the west and east borders of Israel are summarized in Table 1 for the different research

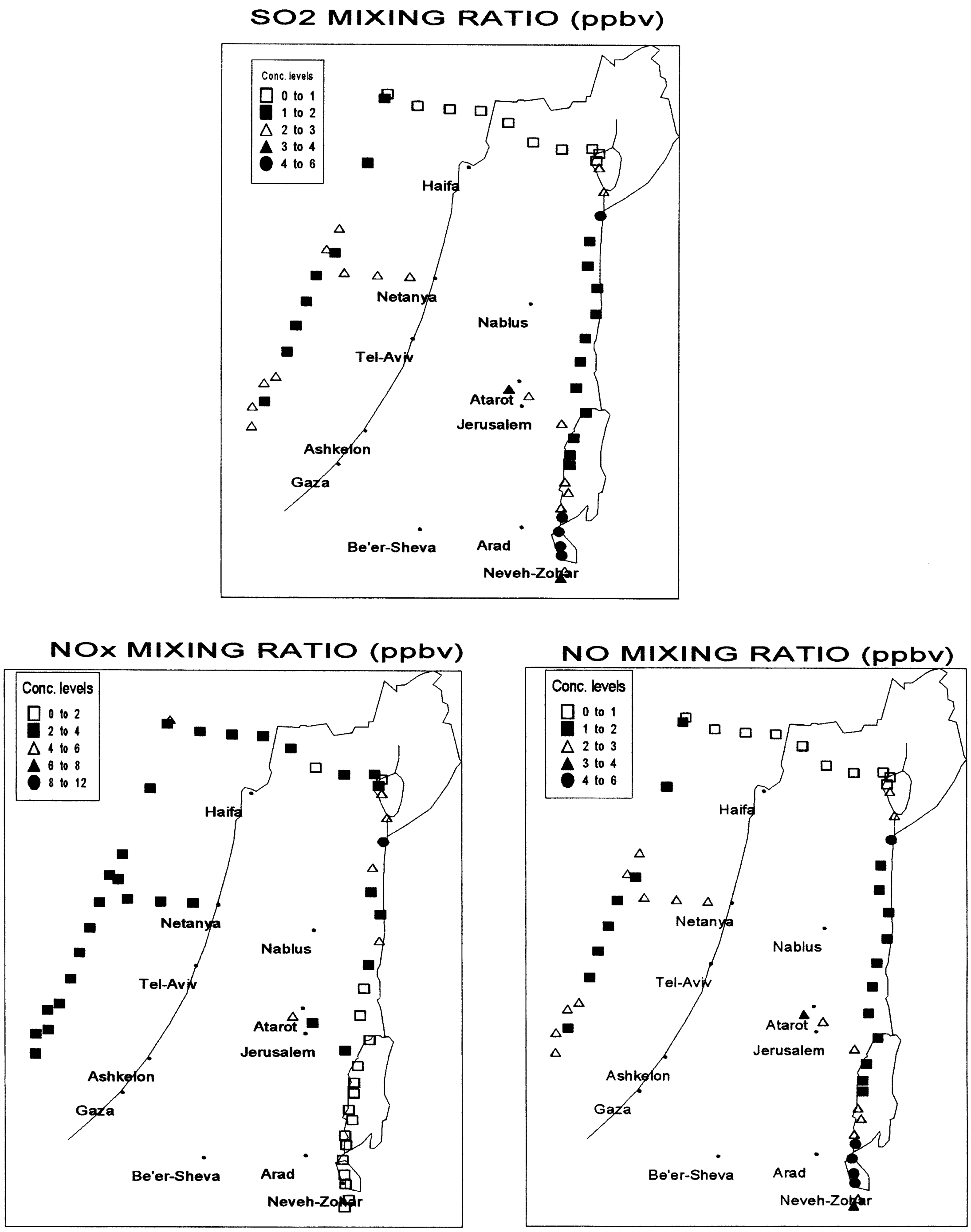

Fig. 2. Sulfur dioxide pollution measurement map for flight performed on 29 May, 1998.

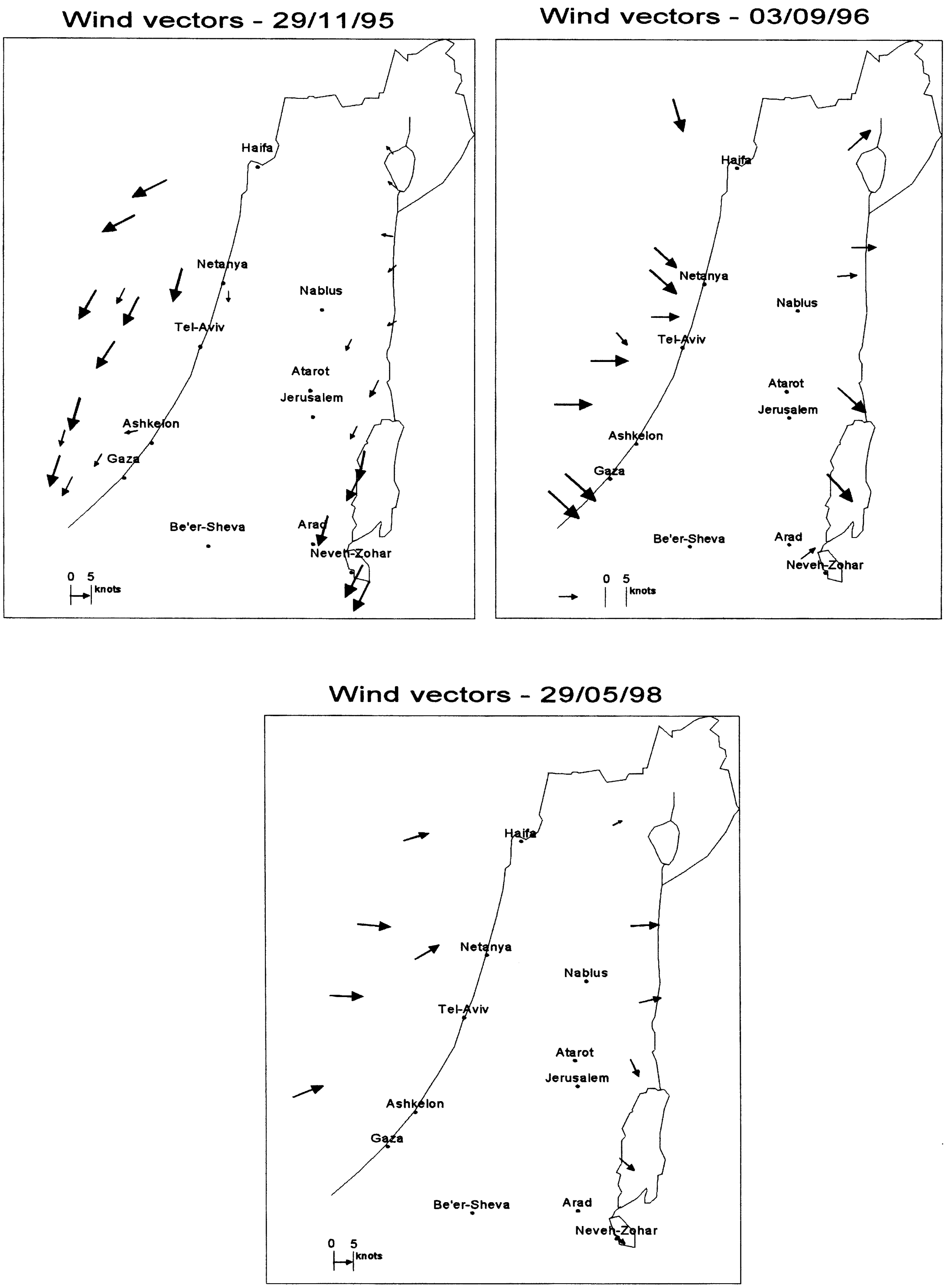

Fig. 3. Wind vector measurements for the three measuring periods: early summer, late summer and fall.

Table 1
Sulfur fluxes over Israel for early and late summer and fall (mg/h)

Date	West	East	Contribution
Early summer			
27.5.98	30.2	43.7	13.5
28.5.98	9.3	60.3	51.0
29.5.98	29.6	45.7	16.1
1.6.98	14.8	47.2	32.4
3.6.98	16.8	38.1	21.1
4.6.98	9.8	23.7	13.9
Average	**18.4**	**43.1**	**24.7**
Late summer			
3.9.96	13.0	48.6	35.4
5.9.96	0.9	13.9	12.9
8.9.96	5.1	38.9	33.8
10.9.96	2.0	17.7	15.7
Average	**5.3**	**29.8**	**24.5**
Fall			
27.11.95	1.2	10.6	9.4
28.11.95	2.2	9.6	7.4
29.11.95	7.2	19.0	11.8
30.11.95	4.9	12.5	7.6
Average	**3.9**	**12.9**	**9.1**

seasons. An examination of Table 1 shows that the highest fluxes from the west were for the early summer time, averaging 18.4 mg S/h. During this season, the levels were more than three times higher the averages for late summer (5.3 mg S/h) and almost five times the average levels measured for the fall (3.9 mg S/h). The local pollution contribution during the early and late summer periods were similar (close to 25 mg S/h), and almost triple the autumn sulfur level, 9.1 mg S/h.

4. Discussion

Examination of the sulfate contribution in the incoming air masses (west) shows that it varied between 50 and 90% of the total sulfur level. For the outgoing air masses, the sulfate content dropped to approximately only 25%.

4.1. Sulfur species

The relatively high sulfate content in the incoming air masses and the lower levels of sulfate at Israel's eastern border can be explained by referring to the sulfate formation mechanism. During the summer, homogeneous conversion of SO_2 to particulate sulfate can be assumed to be the main oxidation mechanism under the conditions generally prevailing over the eastern Mediterranean region. The major path for oxidation is via HO radicals according to the multi-stage process proposed by Stockwell and Calvert (1983):

$$SO_2 + HO + (H_2O + O_2) \rightarrow H_2SO_4 + HO_2 \qquad (2)$$

Conversion rates generally appear to be higher in the summer than in the winter, and higher at noon compared to night-time (Meagher et al., 1983). Most field studies have indicated an upper limit of 0.07 h^{-1} at noon and 0.02 h^{-1} for the daily average under summer conditions (Eatough et al., 1994). Due to this slow conversion rate, particulate SO_4^{2-} will be formed only hours after the release of the SO_2 into the atmosphere. Because of the slow oxidation rate of SO_2 coupled with the slow dry deposition velocity of 0.01–0.4 cm s^{-1} for particulate sulfate, the atmospheric lifetime of particulate sulfate was estimated as being up to 6 days in the absence of precipitation (Chin et al., 1996). Thus, particulate sulfate can be transported thousands of kilometers before significant removal occurs. It is, therefore, to be expected that the sulfate content of the long range transported air masses entering Israel from the west will have a relatively high sulfate content. Conversely, due to the entrapment of fresh SO_2 as the air masses travel eastwards over Israel and the slow sulfate conversion rate, it is only to be expected that the contribution of sulfate to the total sulfur content of the outgoing air masses will be lower than for the entering air masses.

4.2. Synoptic and dispersion conditions for each aircraft measurement period

The rate of potential transport of contaminants in the atmosphere is strongly influenced by the intensity of wind flow within the mixed layer and its depth. Further along the Eastern Mediterranean coastal zone, both coastal-breeze circulation and large-scale synoptic situation modulate the mean

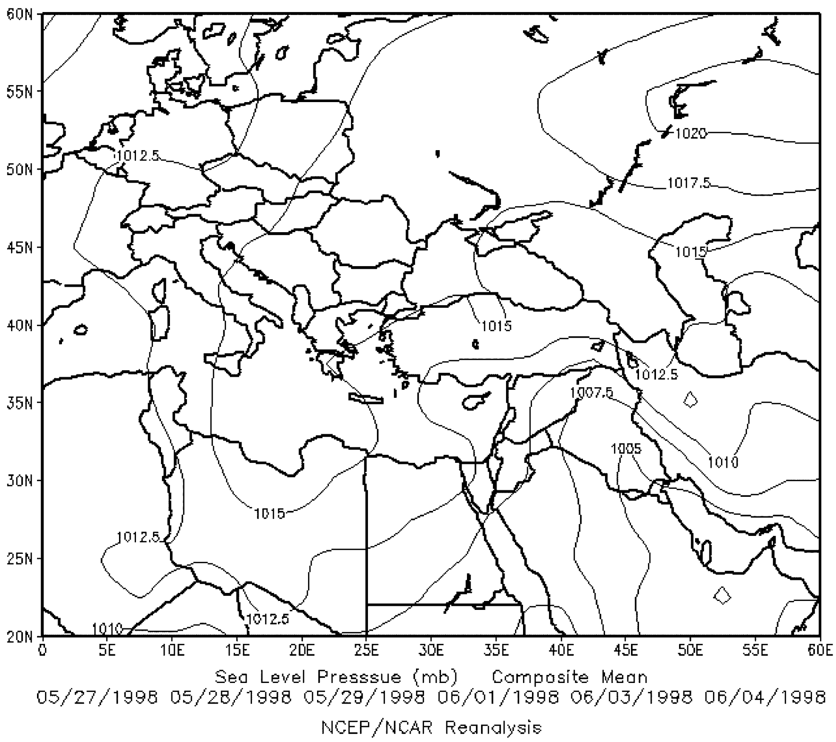

Fig. 4. Mean sea level pressure chart for early summer (28 May–4 June, 1998).

mixed layer depth. Ventilation, defined as the mixed layer depth multiplied by the mean wind speed in the layer, could, therefore, serve as a valuable indicator for atmospheric efficiency to transport contaminants to and from Israel. In order to characterize the flow intensity, a synoptic analysis for the region, as well as the general conditions featuring each flight research campaign were analyzed.

The dominant synoptic condition prevailing during the summer (mid-May to mid-September) over the eastern Mediterranean is influenced by two major systems surrounding the region, resulting in overall monotonic weather conditions. From the east, mid- and south Asia, land warming during the summer leads to the development of the Monsoon Low, resulting in predominant north winds over the eastern Mediterranean. One of its barometric cores is located in the Persian Gulf, forming the so-called 'Persian trough' which extends from the Persian Gulf towards the southern shores of Turkey, and dominates the Mediterranean in the summer. This synoptic configuration generates northwest winds, influencing the north and the central regions of Israel. Westerly to the region, the Azorean high influences the area as part of the subtropics belts of highs. The Azorean high is centralized at the Atlantic Ocean, near the Azorean islands, and dictates northwest air motions. The combined synoptic winds coincide with the sea breeze reinforcing it during daytime and opposing it during the night, when land breeze takes over.

Early and late summer are featured by the overwhelming influence of the sub-tropical high-pressure system characterized by frequent subsidence inversions aloft accompanied by a shallow pressure gradient on the surface. Moreover, solar heating creating large land-water contrasts in the surface temperature is not in its maximum intensity. All these thermodynamic and synoptic conditions create weak zonal winds above ground, and consequently, poor ventilation conditions as manifested by rising air pollution concentrations within the relatively shallow mixed layer (Dayan and Rodnizki, 1999).

As opposed to the summer season, in which an almost exclusive surface synoptic pattern persists, the average distribution of sea level pressure in the eastern Mediterranean for the transitional and the winter seasons can be divided into three most frequent pressure systems, which are not semi permanent. The Siberian High, a typical winter monsoon system (i.e. which forms because of the intense cooling of the continent), leading to a dry, cold and strong northerly flow (Saaroni et al., 1996). The Red Sea Trough, a pronounced trough of low barometric pressure extending northward from Equatorial Africa, crosses over the Red Sea and the eastern Mediterranean countries. This synoptic pattern brings southeasterly winds to the region, and the associated weather is usually hot and dry (Saaroni et al., 1998).

Cyclonic activity accompanied with precipitation and strong westerly winds occur between the above mentioned dry spells.

Table 2
Potential rate of atmospheric transport of contaminants in the Eastern Mediterranean

Date	Mixing depth (m)	Zonal wind ($m\ s^{-1}$)	Ventilation rate ($m^2\ s^{-1}$)
27.5. 1998	400	1.0	400
28.5.1998	350	5.0	1750
29.5.1998	500	4.0	2000
1.6.1998	825	3.5	2888
3.6.1998	600	2.0	1200
4.6.1998	600	1.5	900
Mean First Period	**545**	**2.8**	**1526**
3.9.1996	660	5.0	3300
5.9.1996	500	3.0	1500
8.9.1996	1000	4.0	4000
10.9.1996	1150	4.0	4600
Mean Second Period	**828**	**4.0**	**3312**
27.11.1995	1500	2.0*	3000
28.11.1995	1000	4.0*	4000
29.11.1995	1000	5.0*	5000
30.11.1995	1000	6.0*	6000
Mean Third Period	**1125**	**4.25**	**4500**

Based on data as measured by the Israeli Meteorological Service and represents the Central coastal region.
* Zonal winds were easterly winds.

4.2.1. Early summer period

The sea level pressure chart for the first flight period demonstrates the 'Persian trough' characterized by a shallow pressure gradient over Israel (Fig. 4) leading to a mean zonal flow not exceeding 3 $m\ s^{-1}$. The predominant wind flows during this period are from western and central Europe, areas containing a large number of pollution-emitting sources. At higher atmospheric levels, the quasi-permanent sub-tropical high system forms a subsidence inversion capping the mixed layer and restricting its depth to approximately 550 m (Table 2). Under such reduced ventilation rates (average of 1500 $m^2\ s^{-1}$), the influx of sulfur compounds arriving at the Israeli coast from the high upwind sources located in Europe are relatively elevated, with an average flux of 18 mg S/h. The contribution of the local pollutant sources to the outgoing easterly flux is also pronounced, averaging 25 mg S/h, leading to an average outflow of 43 mg S/h (see Table 1).

4.2.2. Late summer

Towards the end of summer, the 'Persian trough' begins its retreat towards the east while weakening as part of the whole Monsoon Low withdrawal (as shown in the mean sea level pressure chart — Fig. 5). Accordingly the surface winds veer to a more northerly component. Concurrently, aloft, the strength of subsidence diminishes due to a south-east migration of the Sub-Tropic High. These developments result in a mean deeper mixed layer, stronger winds, and hence, higher ventilation rates as registered during the second aircraft measurement period (Table 2). The pollutant in-flux rates advected within this north-westerly flow towards Israel is, on average, much lower (5.3 mg S/h; see Table 1) for two reasons: the upwind sources for sulfur are lower than those located over the western and central parts of Europe and the higher ventilation rates disperse the pollutants more efficiently. Local contributions are similar to the early summer period (Table 1). The reduction in the out-flux towards Jordan results, therefore, from the lower in-flux levels.

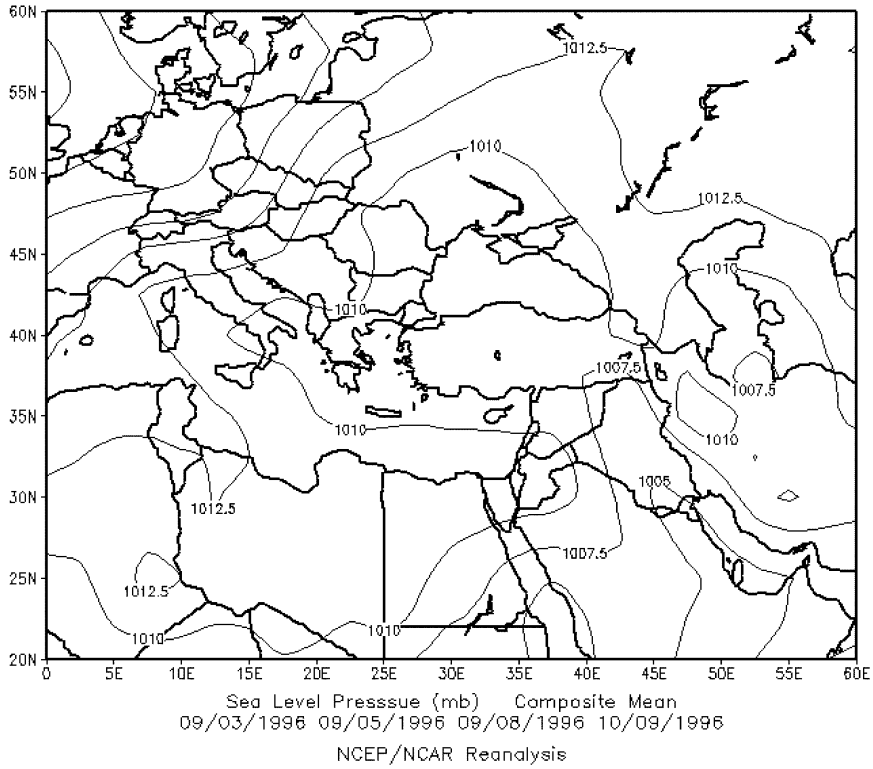

Fig. 5. Mean sea level pressure chart for late summer (3 September–8 September, 1996).

4.2.3. Autumn

The third flight legs were executed over 4 consecutive days at the end of November. The mean sea level surface synoptic chart shows a pronounced trough of low barometric pressure extending northward from equatorial Africa, over the Red Sea and the eastern Mediterranean countries, i.e. the 'Red Sea trough' (Fig. 6). In this synoptic situation, the eastern Mediterranean region is usually subjected to easterly winds accompanied by a well-developed mixed layer. The resulting mean ventilation rate for this period was 4500 $m^2\ s^{-1}$, three times that of early summer (Table 2). Under these increased dispersion conditions, the local contribution of sulfur was, as expected, much lower than the summer values (see Table 1). Since there are practically no pollution sources east of Israel, the pollution levels measured on the eastern flight legs must represent the imported pollution from the west, together with the local contribution in the re-circulated air masses. Similarly, the pollution levels measured west of the Israeli coast represent re-circulated imported air masses. However, due to the elevated ventilation rates, the pollution levels in these air masses are also very low (see Table 1). Re-circulation effects in Israel have previously been reported by Robinsohn et al. (1992) and Alper-Siman Tov et al. (1997). The pollution levels measured in the re-circulated air masses, therefore, represent the maximum pollution levels arriving and leaving Israel under the above synoptic conditions. Dayan (1986) has shown that the above synoptic conditions, accompanied by winds from the east, are especially predominant during the fall and spring for between one-third and half of the time.

During the winter period, no flights were performed due to disturbed weather conditions and to the fact that very low pollution levels were to be expected. As previously mentioned, the three main typical surface flows (i.e. northerly, easterly and westerly flows) resulting from the three synoptic

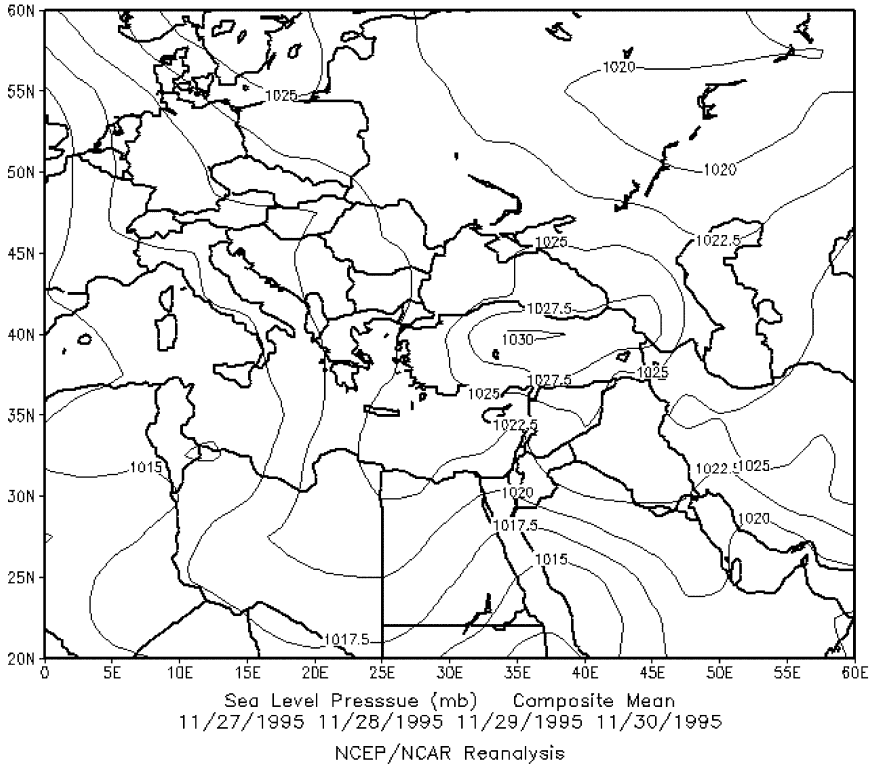

Fig. 6. Mean sea level pressure chart for autumn (27 November–30 November, 1995).

types are all accompanied by an elevated mixed layer leading to high ventilation rates. Furthermore, high precipitation over central Europe during winter causes pollution wash out. The above conditions indicate that long-range transport of pollution towards Israel during the winter will be almost zero.

4.3. Yearly budget

Based on the data available from the three research campaigns, an attempt was made to evaluate a yearly budget of the input and output of sulfur compounds over Israel. As shown above, the ventilation rate strongly affects the pollution flux over Israel. The mean and standard deviation (S.D.) of the mixing depth and wind speed were calculated for a 51-year period, and from this data, the long-term range of ventilation rates. A summary of the results is shown in Table 3. An examination of the above table shows that it is possible to divide the ventilation rates into two groups: the fall and winter months (October through March) which have elevated ventilation rates (reaching more than 20 000 m^2 s^{-1}), and the remaining period (spring–summer) with rates below 10 000 m^2 s^{-1} (only for April the values are slightly higher). Generally speaking, it appears that for 50% of the time, the region is influenced by high ventilation rates, the other half of the time by relatively low rates.

Assuming an average of the data obtained from the two summer measuring periods as representing the entire spring–summer, an average of the fall flux value as representing the fall period, and a zero flux rate for the wintertime as representing the winter–fall period, it is then possible to evaluate a yearly flux budget. Based on the above, the yearly in-flow to Israel was calculated to be 0.06 tg total sulfur. Katsoulis and Whelpdale (1990)

Table 3
Long term (51 year) mean (LTM) and standard deviation (S.D.) of the mixing depth, wind speed and range of ventilation rates

Month	LTM Mixing depth (m)	S.D. of LTM Mixing depth (m)	LTM of Wind speed (m s^{-1})	S.D. of Wind speed (m s^{-1})	Range of LTM Ventilation rates (m^2 s^{-1})
January	1700	950	7.0	4.75	1688–31 138
February	1830	1135	7.5	5.00	1738–37 063
March	1790	860	7.0	4.75	2093–31 138
April	850	660	5.5	3.75	333–13 968
May	750	525	4.5	3.25	280–9880
June	810	470	5.5	2.25	1105–9920
July	870	450	5.0	1.65	1365–8780
August	820	395	4.5	1.50	1275–7290
September	930	510	5.0	2.10	1218–10 224
October	1650	910	4.5	3.50	740–20 480
November	1500	940	5.5	4.50	560–24 400
December	1700	990	6.0	4.50	1065–28 245

LTM and S.D. are long term mean and standard deviation for mixing depth values for the years: 1955–1968 (Rindsberger, 1974), 1981–1984 (Dayan et al., 1988), 1987–1989 (Dayan and Rodnizki, 1999). LTM and S.D. of wind speeds are from the NCEP/NCAR Reanalysis Project (NOAA-CIRES Climate Diagnostic Center): 51-year data record: 1948–1999.

estimated the output from southeast Europe (Greece) to the east to be of the order of 1.2 tg S/year. A comparison of our estimates for the yearly input into Israel with those of Katsoulis and Whelpdale (1990), based on mass/length, shows that only a small portion, less than 10% of the yearly pollution output from southeast Europe finally reaches Israel. The yearly Israeli contribution to the output flux towards Jordan was estimated to be 0.13 tg total sulfur. Considering the uncertainties in the above estimations, this value is in reasonable agreement with the anthropogenic annual emissions estimated by the Israeli Ministry of the Environment (based on fuel consumption) to be 0.2 tg S (Gabbay, 1998). This would indicate that almost all of the pollution emitted in Israel finally makes it way out of Israel towards its eastern neighbors. The total sulfur out-flow per year was calculated to be of the order of 0.19 tg S.

Preliminary measurements for nitrogen compounds flux showed similar trends as for the sulfur compounds. On a yearly average, approximately 10% of the total nitrogen leaving southeast Europe (estimated by Katsoulis and Whelpdale (1990) as being between 0.7 and 2.1 tg N/year) arrived at the Israeli coast, while approximately 80% of the nitrogen pollution emitted in Israel [estimated by Gabbay (1998) at 0.12 tg N] is emitted to the east.

5. Conclusions

Based on the limited number of research flights and by extrapolating the measurements using climatological data, the flux of sulfur compounds to and from Israel was estimated. While long-range transported sulfur flux entering Israel consisted of between 50% and 90% particulate sulfate, this decreased to an average of approximately 25% in the exiting air masses. The above is consistent with the relatively slow formation and deposition rate of particulate sulfate and the fact that the incoming pollution air parcel originated thousands of kilometers upwind. Due to the narrow (approx. 75 km) geographical width of Israel, between the Mediterranean and the Jordan Valley, most of the added pollution load remains in the form of gaseous SO_2. The limited travel time available allows only a small fraction of the local pollution to be converted to the sulfate before the air masses exit Israel. The present data set indicates that approximately 15% of the sulfur pollution emitted over Europe finally reach the Israeli Mediterranean coastline. Experimental observations, when extrapolated to estimate annual averages, show that practically the entire sulfur compounds released into the air from sources within Israel are transported eastward into Jordan.

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