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Seasonal chemical and mineralogical variability of atmospheric particles in the coastal region of the Northeast Mediterranean

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Abstract

A time-serial analysis of atmospheric particles collected since 1990 at a coastal site bordering the northeastern Mediterranean Sea shows that the collected samples display seasonal patterns in both concentration and color. The 1990–91 and 1992 collections yield an annualised average of around 0.5 g/day dust loading reducing to around 0.3 g/day when the sea-salt contribution is excluded. The highest concentrations of atmospheric particles of continental origin for both natural (e.g. Fe, Mn) and anthropogenic (e.g. Cd, Pb) materials are observed between April and October and this variability is inversely related to the frequency and the amount of rainfall. Sea-salt aerosol (Na) contents do not follow this pattern but are related to local wind speeds.

Trace elements in the dusts also display seasonal variations in enrichment factors (EF), with respect to average elemental abundances in crustal rocks. The non-enriched (crustal) elements appear to be indicators of long-distance supply while the strongly enriched (anthropogenic) and moderately enriched suites of elements represent inputs from local or "anomalous" sources.

The similar morphology of quartz and calcite grains and the uniformity of the clay mineral assemblages in samples collected over a two-year period strongly indicate a common source for the bulk of the atmospheric particles. The dominant clay types (palygorskite and smectite) document a desertic source areas.

Keywords: Northeast Mediterranean; atmospheric particles; trace elements; clay minerals; dust trajectory

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1. Introduction

Atmospheric transport is an important route by which materials (natural and anthropogenic) may be delivered to the sea and land. The Mediterranean basin, bordered to the north by countries with economies which range from industrial (such as France and Italy) to semi-industrial (e.g. Greece and Turkey) and limited to the south by the desert belt of north Africa, is an example of a region with a number of contrasting supply areas. The typical Mediterranean aerosol can, thus, be considered to be a mixture of material of European origin and of dust from the Sahara (Chester et al., 1981, 1984).

Past aerosol studies have been largely restricted to the western Mediterranean and little is known about the eastern part (Nihlen and Solyom, 1986; Nihlen, 1990; Pye, 1992; Tomadin and Lenaz, 1989). The overall amount of dust reaching the Mediterranean basin is large compared to other regions, and plays a significant role in the geochemistry of the basin (Chester et al., 1984; Guerzoni et al., 1989a,b). In a detailed study of atmospheric particles reaching and settling in Israel, Yaalon and Ganor (1979) found that on an annual basis approximately 25 million tonnes of dust settle on the eastern Mediterranean Sea. The computed trajectories of dust particles showed that most of the dust originates from the Libyan, Egyptian, Sinai and Negev deserts.

The respective importance of river and atmospheric inputs to the western Mediterranean sea was extensively studied within the context of EROS-2000 (European River

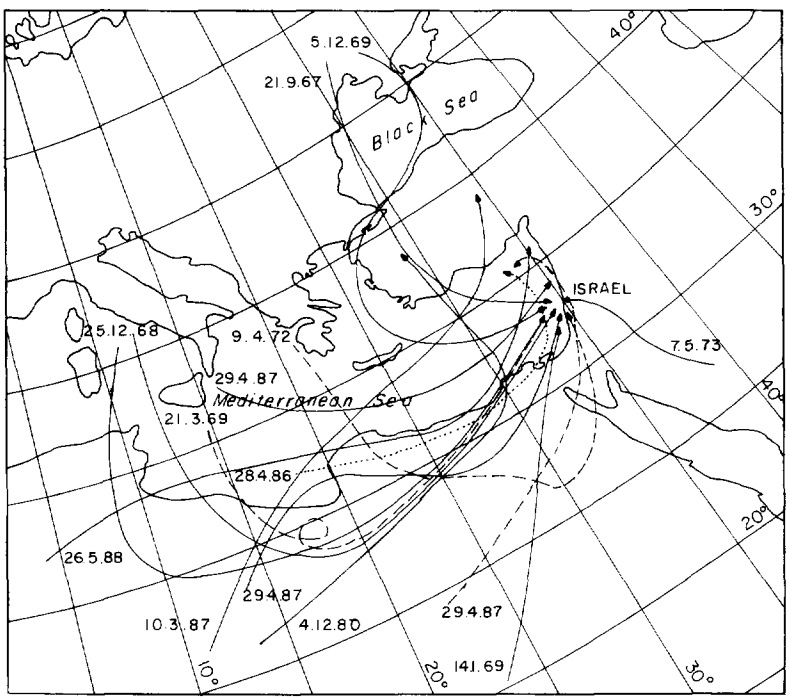

Fig. 1. The 1967–1988 dust trajectories to Israel (compiled from Dayan, 1986; Dayan, unpubl. data).

Ocean Systems). It has been reported that, in the western Mediterranean basin, the influx of Saharan dust is of the same order of magnitude as the annual influx of particles discharged by the rivers (Martin et al., 1989).

Atmospheric deposition of N, P, Fe and input of toxic materials over the oceans results in enhancement of marine photosynthetic activity and assimilation, and thus affects primary productivity (Bergametti et al., 1992). In the eastern part of the Mediterranean, the terrestrial dust may remove PO_4, which is the limiting nutrient for this well-known oligotrophic water mass (Krom et al., 1991). Thus, aeolian materials in the Mediterranean may play the role of either source or sink. Studies conducted by Ganor and Mamane (1982), Ganor (1990), Dayan et al. (1990) and Ganor et al. (1992), following the pioneering works of Singer (1967), Dan and Yaalon (1968), Yaalon and Lomas (1970) and Jackson et al. (1971, 1972), on the aeolian materials of the eastern Mediterranean and based on clay and mineral analyses, together with data on elemental compositions, have showed a significant effect on soils.

The study reported here represents an initial contribution to a planned wider programme (by an international team representing a range of circum-Mediterranean dust

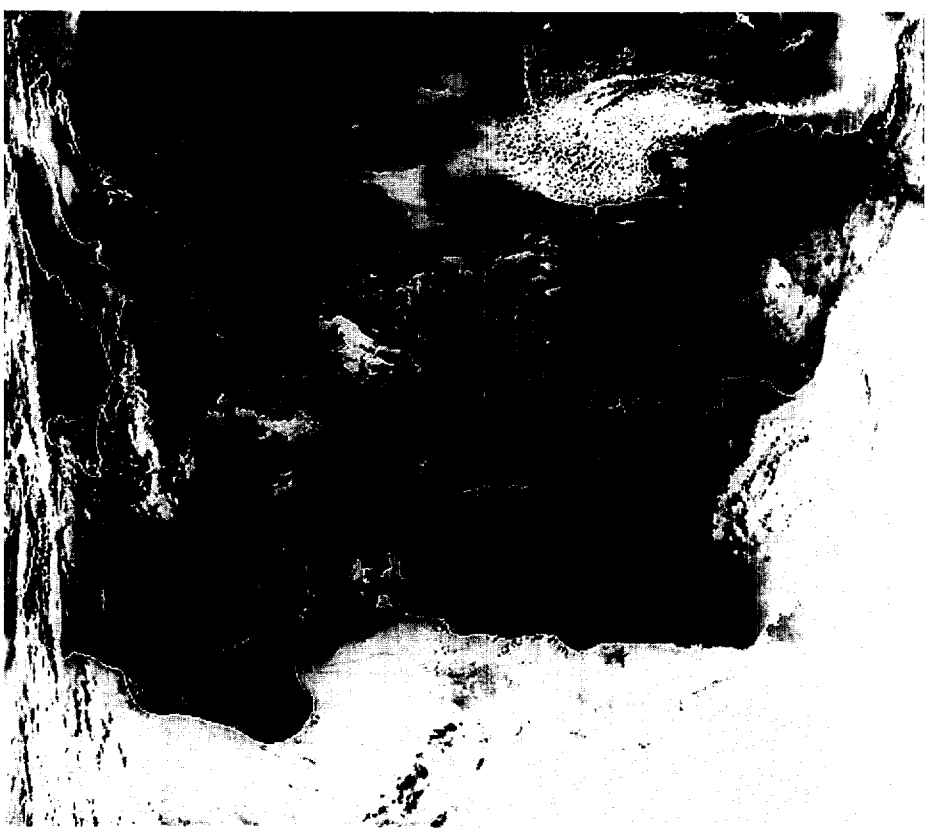

Fig. 2. A satellite image (NOAA 10, Visible) of the heavy dust storm (DS) of 17 October 1990, which transported material from Saudi Arabia.

sources and depositional sites) and reports our findings concerning the temporal variations in dust loading and their chemical and mineralogical composition, as measured by mesh-collecting techniques at one coastal site.

1.1. Meteorological conditions

In order to understand the transport of aeolian materials to the area from either anthropogenic or natural sources the meteorology of the basin must be well understood.

Atmospheric circulation over the Mediterranean Sea is variable: westerlies, easterlies and the coastal land–sea breezes are the common wind systems in summer and autumn while local winds, *poyraz* (a local Turkish name for a cold and northwesterly continental wind) and *sirocco* occur in winter and spring (Özsoy, 1981). Coastline topography of the northeastern Mediterranean provides both barriers to, and channels for, airflows that bring at times extremely different air masses to the coastal region.

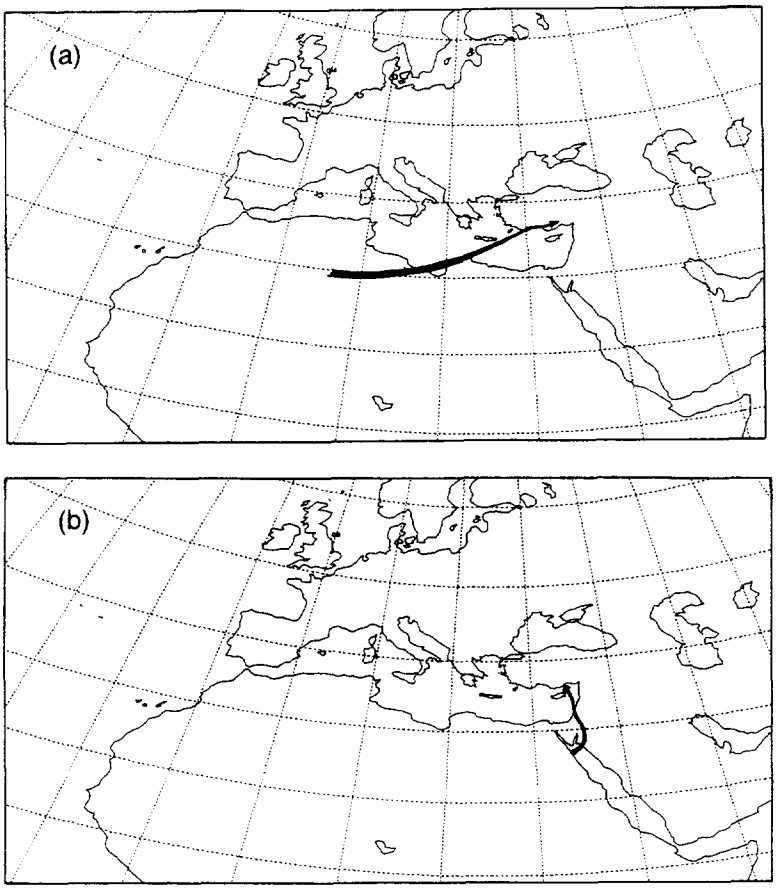

Fig. 3. Back trajectories of the 14 December 1990 (a) and 17 October 1990 (b) heavy dust storms from the Sahara (north Africa) and the Arabian Peninsula.

Dayan (1986), summarizing five years of atmospheric trajectories for the eastern Mediterranean, provided a better understanding of wind patterns. The major trajectories identified for Israel are shown in Fig. 1 (Dayan, 1986 and Dayan, unpub. data). The trajectory directions were divided into three distinctive classes and can be generalized for the northeast Mediterranean, since both Israel and Turkey lie downstream on the air patterns crossing the Mediterranean basin.

1. Long-fetch maritime air masses from northwest Europe crossing the Mediterranean Sea, occur uniformly throughout the year.

2. Northeast continental flow that originates in eastern Europe and is most frequent during the summer season.

3. Flows originating from the African–Arabian continental region and active throughout the year, as follows: A. The southeast flow from the Arabian peninsula, which is infrequent and occurs mainly during the fall. B. The southwest flow along the north African coast which is most frequent during late winter and spring. C. The south-southwest flow from inland north Africa which is most frequent during the winter and spring.

It is also possible to utilize remote sensing to observe the propagation of air masses and associated fronts. A typical Advanced Very High Resolution Radiometer (AVHRR) photo representing an air mass originating from the Arabian peninsula during 17 October 1990 is shown in Fig. 2. A more precise way of identifying the source of the moving air masses is to use 3-day back-trajectory analysis. Representative examples are the 14 Dec. 1990 and 17 Oct. 1990 heavy dust storms from the Sahara and Arabian Peninsula respectively (Fig. 3).

2. Materials and methods

An atmospheric sampling tower has been constructed at the harbor jetty of the Institute of Marine Sciences, Middle East Technical University, located near Erdemli (39°33′ N, 34°15′ E), on the southeastern coast of Turkey. The tower was originally constructed from frames of two commercial cargo-containers welded on top of each other and secured with steel wires to decrease wind resistance. Side walls of the top container were removed. During 1991 the height of the tower was further raised by 4 meters by adding another container frame. The sampling platform is now 18 meters above sea level and since 1991 incorporates a Hi-Vol cascade and a Hi-Vol sampler, collecting samples on Whatman 41 filter papers, together with an automatic wet and dry deposition meter. The tower is also equipped with an Aandrrea meteorological station with direction sensors for automatic measurement of temperature, pressure and wind speed. The three dimensional (3D) air mass back-trajectory analyses are performed by utilizing ECMWF (European Centre for Medium Range Weather Forecasts) wind fields and models (Kallberg, 1984). Calculations of the trajectories are done for 3-days backward and at 700 mbar levels, at mid-time of the sampling period.

During the initial phase of this study, which covered the period 1990–91, the mesh samplers were mounted on wooden holders 3 m above the top of the tower enabling us to sample 21 meters above the sea level. The meteorological data used during the initial period were obtained from Erdemli, which is located 8 km east of the tower.

A total of 97 samples were collected during 1990, 65 in 1991, and 71 in 1992. Total trace element analyses were conducted on these samples. Thirty-four samples (along with material collected during the heavy storm of 16 October 1990) were selected for mineralogical analyses, using color and dust loading as contrasting variables to represent the time and spatial distributions. Collections were made on two terylene meshes joined down the middle in a cruciform shape. The terylene mesh has square apertures of 300 μm. When fully extended, these provide a filter-area of 1 m^2 facing the wind coming from all directions. Immediately after exposure the filters were sealed in polythene bags, the dust extracted by washing and agitation with Milli-Q water and then the samples were dried in an oven at 40°C. The dried samples were then weighed and ground in an agate mortar to ensure uniformity and avoid contamination. The samples obtained from this type of mesh cannot represent the full spectrum of atmospheric particles and associated elements. Nevertheless, such data are useful for describing the elemental composition of the atmospheric particles on a nonquantitative basis. The material collected on the mesh, in addition land-derived components, also retains sea salts and smaller particles which are embedded in the larger grains. It is believed that humidity also helps the retention of sub-micron particles.

The color of the collected samples was classified according to the Munsell Soil Color Chart. Organic carbon (%) was determined by the modified Lichterfelder method (Schlichting and Blume, 1966). The $CaCO_3$ equivalent (%) was determined volumetrically using 10% HCl in a Scheibler calcimeter (Black, 1965). Clay mineral types, quartz and calcite were determined by X-ray diffraction analyses of the < 2 μm clay fraction and ground powder samples respectively. Semiquantitative data for the minerals was determined by their peak heights (abundancy) in the diffractograms.

Approximately 0.2 g of each mesh sample was digested in hot concentrated $HF–HNO_3$ mixture and the residue dissolved in 0.1 M HNO_3. The solutions were analyzed by a Varian Techtron model AA 6 atomic absorption spectrophotometer equipped with a deuterium background corrector. Aluminum, Fe, Mn, Cr, Ni, Cu, Pb and Na were measured by flame atomic absorption spectrophotometry, and Cd by flameless AA spectrophotometry.

3. Results and discussion

The analytical results show that the aerosol composition displays a seasonal pattern both in concentration and color (Figs. 4, 5, 8 and 9).

Average monthly dust loadings during 1990, 1991 and 1992 are shown in Fig. 4. During the 1990 sampling period the observed fluctuations were much greater than those encountered in the 1991–1992 sampling period. This difference is attributed to the cumulative effect of sampling height and treatment techniques used during 1990. During the 1990 sampling period the supernatant liquid with the sea salt on the meshes was not siphoned off. Since 1991 the sea salt has been removed from the samples and the overall height of the sampling position was raised to 21 meters, which helped to reduce the erratic fluctuations.

Excluding the 1990 samples, the average dust loadings for 1991–1992 for the 136 samples is 0.2–0.3 g/day, broadly similar to loadings recorded from Crete by Pye

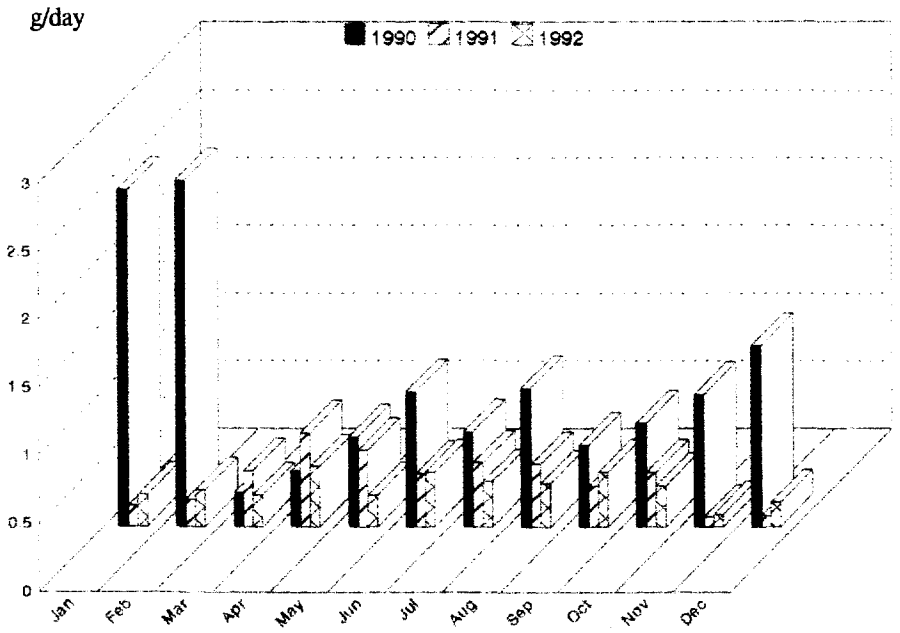

Fig. 4. Monthly averages of dust load (g/day).

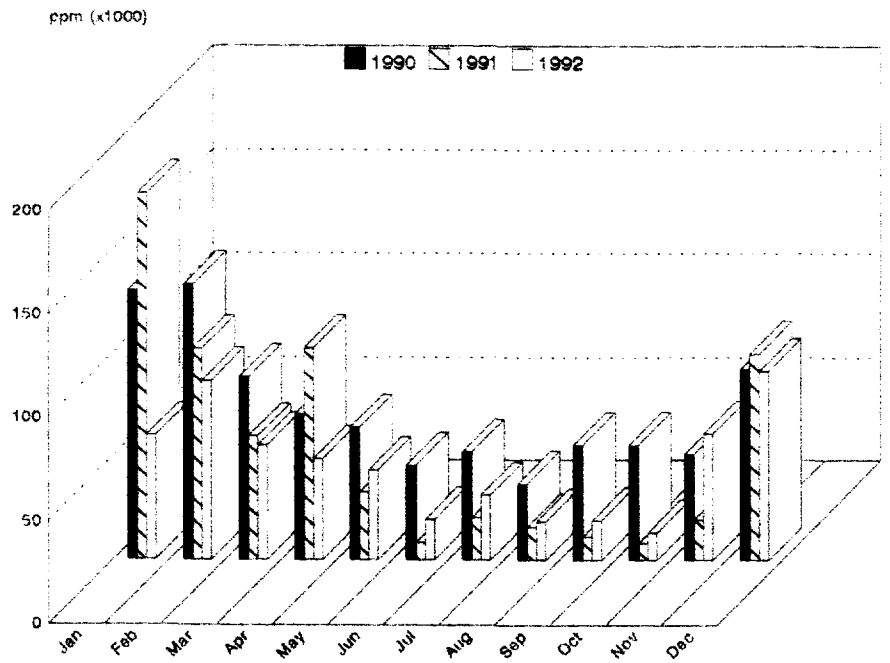

Fig. 5. Monthly averages of Na (ppm).

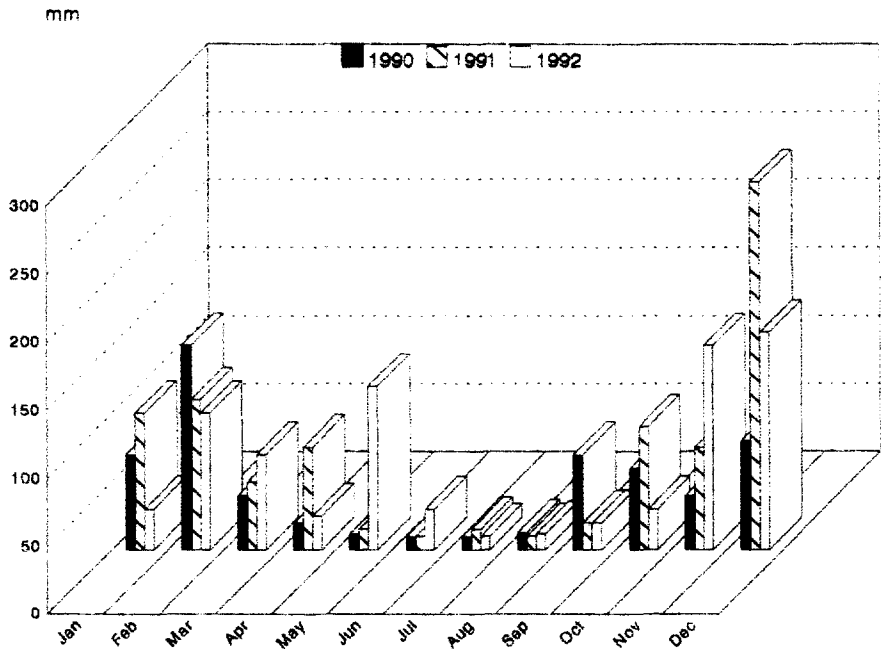

Fig. 6. Monthly averages of rain (mm).

(1992). Maximum levels were reached during April and the late summer season and minimum levels during November and December. Monthly annual Na and rain values are shown in Figs. 5 and 6 for the 90–91–92 sampling periods. During spring, early-summer and autumn dust loading is at maximum, whereas the Na contribution is relatively small. The high summer levels in dust loadings decrease markedly over the winter. As expected, seasonal changes of atmospheric dust loading correlate inversely with the Na concentrations which correlate well with the precipitation rate. The high concentration of Na corresponds to the wet season of the NE Mediterranean. This may result from rapid reloading of the atmosphere with sea salt particles immediately after rains. During the wet season this is to be expected since, precipitation events are always coupled with fronts associated with strong synoptic-scale meteorological conditions. The observed minimum dust loadings during the wet season is attributed to another deficiency of the mesh sampling technique. The loss of the soluble part of sample during the rain event possibly results in underestimation of the dust loadings.

The most conspicuous feature observed during examination of the samples is the change in the color of material collected on the mesh. The summer samples are mainly light brown in color and become grey during the winter season. This conspicuous change has led us to use this property as a means of general classification. Thus, color is the initial criterion for classifying the dust materials. Color classification over a three year sampling period, as well as examination of selected samples, reveal the two main classes associated with dust particles, indicating the strong seasonal diversity of source areas. The dominant colors of the samples from January to March were black and

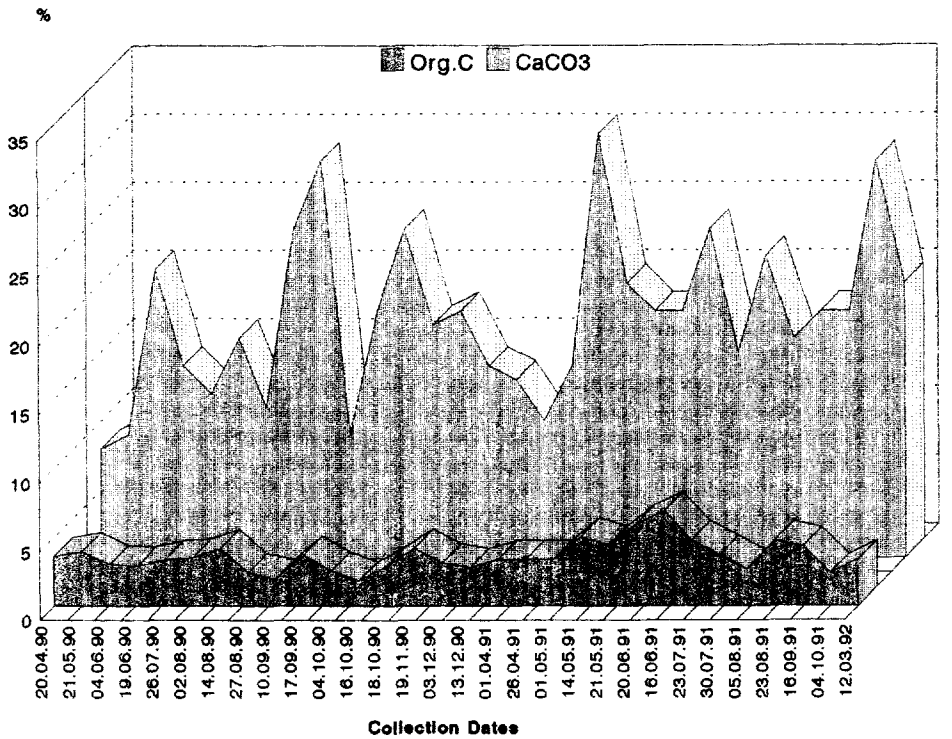

Fig. 7. Distribution of organic carbon and $CaCO_3$ in the 1990–1992 period.

brownish black (10 YR 2/1 and 10 YR 3/2). During April the samples were yellowish brown (10 YR 4/3) which was also the dominant color from June to November. In November the color of the samples again became darker. Seasonal changes in color are interpreted primarily as indications of the diversity in source areas, which in turn is controlled by seasonal changes in the prevailing wind directions.

Organic carbon content of the samples ranged from 2 to 6%. A few samples with lower amounts of organic carbon (~ 2%) were collected in September and October 1990 and March 1992. Higher amounts of organic carbon (expected to darken the colors) were generally present in May and from August to November in 1990 with an exception in October and from May to September in 1991 (Fig. 7). This is contrary to the lighter colors (yellowish brown — 10 YR 6/2, 6/3, 6/4, 7/3) observed during almost the same period indicating that there is some other factor that masks the expected dark colors of samples rich in organic carbon (> 3%) (Fig. 7). The content of iron, which is the red colorant for soil material, shows an increase from April to August in 1990, and in April as well as July to October in 1991 and 1992 (Fig. 8). However the general levels of iron for 1991 remain relatively more uniform and higher throughout the year compared with 1990 or 1992. Thus, the high amounts of the total iron seem to be the reason for the yellowish color during these periods. Total manganese may be responsible for the brown tints as indicated by the increasing amounts from June to August in 1990

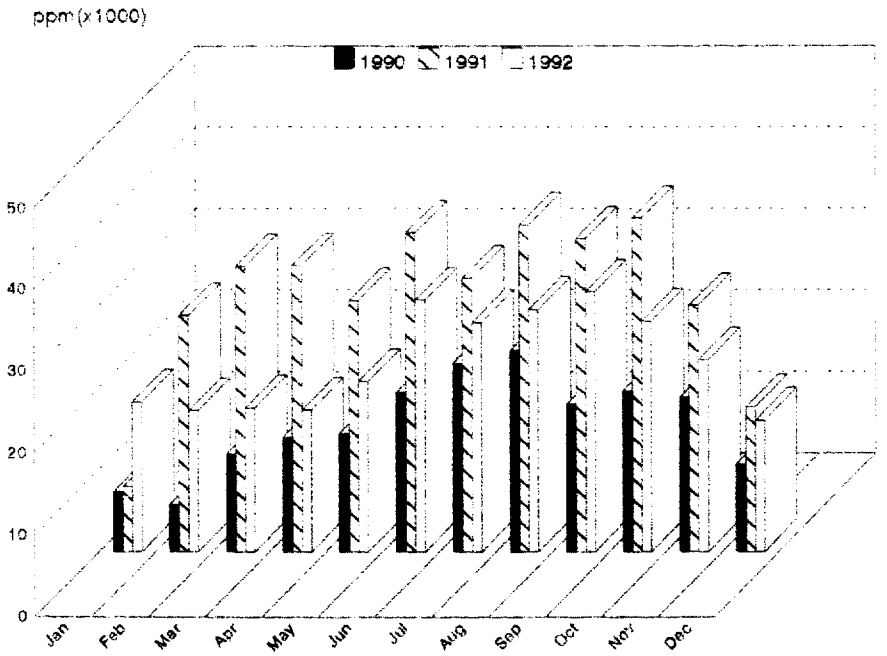

Fig. 8. Monthly averages of Fe (ppm).

and from May to October in 1992 (Fig. 9). The Mn values of 1991 are more uniform throughout the year when compared to 1990 and 1992, as is shown by the iron values. Thus, the enhanced values of Fe and Mn are probably responsible for the masking of the dark colors, which would normally be associated with the high amounts of organic carbon determined in the 1990 and especially in the 1991 summer samples.

Total carbonate content of the atmospheric particles by dry weight ranged from 8% to 31% $CaCO_3$ (Fig. 7). The carbonate content of the atmospheric particles normally originates from terrigenous carbonates and not from anthropogenic sources. Terrigenous carbonates are derived mainly from, limestone, dolomite, and calcretes (Yetis, 1988; Kapur et al., 1993). The similarity between the clay mineral types of 1990 and 1991 samples indicates transportation of the aeolian material from the same source, most probably from north Africa (Sahara) (Mermut et al., 1978; Kapur et al., unpub. data; Kapur et al., 1987; Ganor, 1990; and Ganor et al., 1992; Kubilay et al., 1993). The 14 Dec. 1990 heavy dust storm material from north Africa contains equal amounts of smectite and palygorskite as well as diatom frustules, most probably derived from basins with brackish paleo/recent lakes in north Africa (M. Senol, pers. commun.; Tasman-Rabnikar, 1975; see Pye, 1992).

All samples contained abundant quartz and calcite (Fig. 10). The quartz grains (commonly with etched surfaces) and subordinate feldspars probably originate from soils and sandy deserts of North Africa.

The average concentrations and the ranges of each element measured during the sampling period are summarized in Table 1. For comparison, the results from other

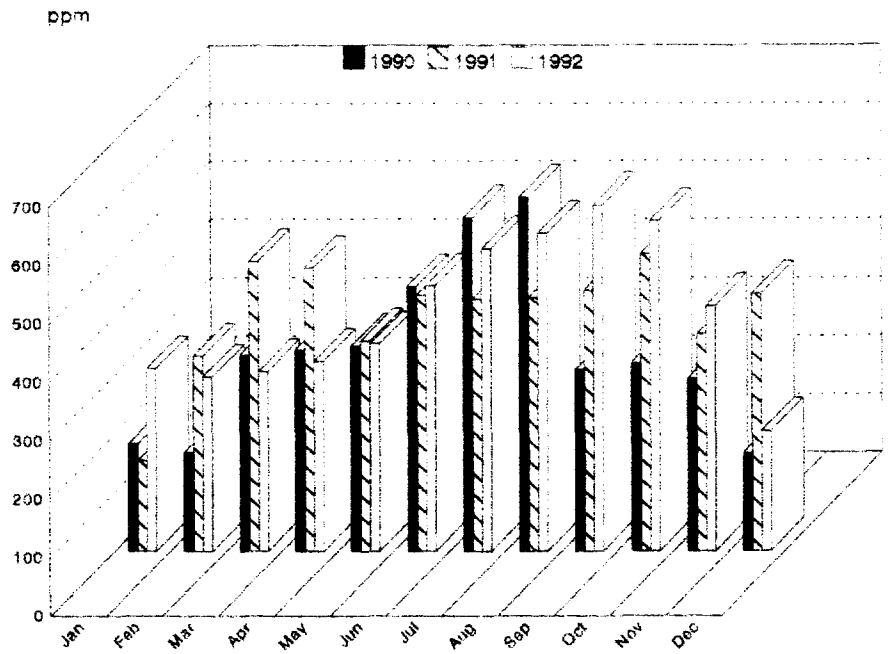

Fig. 9. Monthly averages of Mn (ppm).

studies made by filter-mesh techniques and crustal averages also are included in Table 2. Data published by Chester et al. (1984) and Saydam (1981) were obtained from samples collected on ships, whereas the results of Ganor (1990), like our study, were obtained from samples collected at a land-based station. Our results represent continuous sampling during 1990, 1991 and 1992, whereas Ganor (1990) collected samples during 23 heavy dust storm episodes between 1968 and 1987. Rahn's (1976) values represent trace elements in urban aerosols collected from 29 different circum-Mediterranean cities.

Table 1
Elemental analyses of atmospheric particles collected during the present study; all values are in ppm

	1990			1991			1992		
	Average	Min	Max	Average	Min	Max	Average	Min	Max
Al	23528	1738	68701	32969	1625	51375	28403	2125	58917
Fe	16387	1471	43674	33263	3597	61722	24434	2723	37938
Mn	341	45	1403	414	96	708	437	47	455
Cr	120	7	493	212	25	598	358	67	1234
Cu	30	4	165	38	10	69	55	13	314
Ni	86	11	154	234	28	1612	165	32	353
Zn	163	16	825	247	60	629	238	44	551
Pb	79	13	370	137	32	387	166	26	1043
Cd	1	0.07	4.61	1.07	0.02	12.55	0.21	0.03	0.69
Na	68734	1051	221107	42108	3095	238985	39398	4193	154567

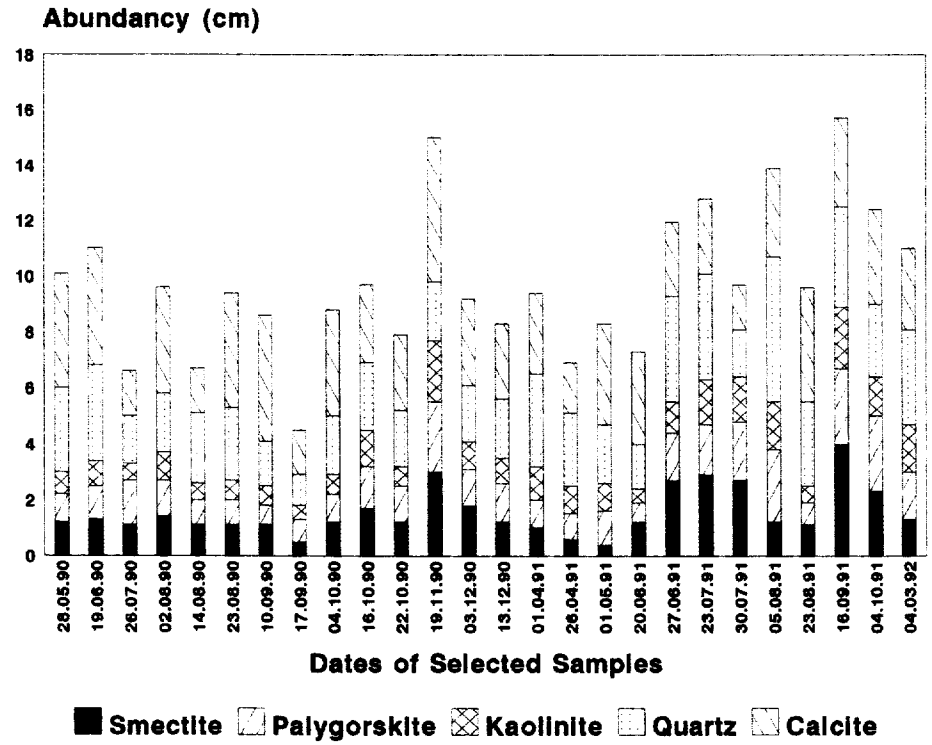

Fig. 10. Mineralogical composition of selected samples (height of the XRD peak in cm).

As seen in Tables 1 and 2, the elemental concentrations reported in the present study appear to be intermediate between urban aerosols and Ganor's (1990) results where local aerosols and long range transported particles affect the observed concentrations. It has been found that the majority of the measured metal concentrations in dust samples show marked similarity in composition to the average elemental composition of crustal rocks. Exceptions do occur, however, such as the concentrations of Ni and Cr which are relatively high compared to the average composition of crustal rocks. The reason for this is believed to be the presence of ophiolitic minerals transported by wind action from the Taurus ranges.

In theory, the heavy metal data for atmospheric particles should be broadly comparable in composition to average sedimentary rocks. Regional exceptions to these global trends reflect unusual geology in the source areas and to some extent, anthropogenic effects. Therefore, atmospheric particles collected from the lower troposphere contain trace metals having both "natural" and "anthropogenic" sources. For trace metals, the most important natural source is weathering of the earth's crust.

A common method of identifying exceptions from the crustal background (Rahn, 1976), is by calculating the enrichment factor (EF):

$$EF = \frac{(X/Al)A}{(X/Al)C}$$

Table 2
Elemental analyses of transported materials compared to data from other studies: 1 — Chester et al., 1984; 2 — Rahn, 1976; 3 —Saydam, 1981; 4 — Ganor, 1990; 5 — average composition of crustal rocks. Numbers in parentheses indicate average values; all values are in ppm

	1	2	3	4	5
Al	–	20000	58000–80496 (72033)	26000–62000 (41600)	81500
Fe	29000–99000 (52000)	27000	41000–63111 (50867)	17000–39000 (29300)	56300
Mn	310–3720 (1338)	750	372–606 (530)	275–900 (524)	950 (552)
Cr	21–850 (119)	150 (	57–188 128)	32–169 (89)	100
Ni	10–720 (96)	200	41–680 (105)	35–75 (53)	75
Cu	23–490 (168)	2000	35–3268 (490)	23–120 (42)	55
Zn	135–3100 (780)	5200	126–695 (411)	104–255 (165)	70
Pb	59–2750 (588)	12000	320–11362 (2224)	40–360 (105)	12.5
Cd	– (0.35)	46	–	0.1–1.7	0.2
Co	3–36 (10)	28	–	5–24 (9.25)	25

– Data not available.

where the numerator is the ratio of the concentration of the element to Al in the atmospheric particles (A) and the denominator is the ratio of the same element to Al in the average crustal material (C). The crustal levels of elements used to calculate enrichment factors in this work are those published by Taylor (1964). The averages and ranges of EF for each element are presented in Table 3. These can be grouped into (1) *non-enriched* elements usually having EF < 1 (Fe, Mn), (2) *moderately enriched* with 1 < 10 (Ni, Cu, Cr), and (3) the *enriched* ones EF > 10 (Cd, Pb, Zn), These EF's should

Table 3
Averages and ranges of EF of the main metallic elements

	1990			1991			1992		
	Average	Min	Max	Average	Min	Max	Average	Min	Max
Fe	1.1	0.6	1.8	1.6	0.9	5.5	1.4	0.3	2.1
Mn	1.3	0.6	3.1	1.2	0.4	6.4	1.4	0.6	4.0
Cr	5.0	1.2	31	6.4	0.9	26	14	1.5	88
Cu	2.7	0.9	14	2.4	0.6	11	4.1	0.7	28
Ni	5.0	1.8	16	18	3.0	540	7.9	1.4	21
Zn	11	0.8	56	11	2.6	56	12	2.1	33
Pb	31	4.4	160	36	8.6	201	50	6.0	379
Cd	39	4.2	224	46	0.4	969	8.9	0.9	27
Na	31	0.1	343	18	0.2	425	13	0.4	245

be viewed as a first order approximation of the crustal component of the 1990–91–92 atmospheric particles and only the order of magnitude variations in the factor should be considered to be significant (Table 3). Although Ni is identified as a moderately enriched element in 1990 and 1992, its 1991 average was significantly higher than during the other years. This is due to the pollution caused by the Ni-rich black rains from Kuwait and northern Iraq (Evliya, 1992; Evliya et al., 1993), during the Gulf war.

4. Conclusions

The results reported here represent a preliminary contribution to the complex problems of atmospheric transport of particles in the eastern Mediterranean. Nevertheless, the simple collection techniques utilised in this study have yielded significant new data from a land-based site that can be logically related to our current knowledge of the patterns of atmospheric transport and provenance in this region.

Thus, the observed variations in monthly-average dust loadings can be related to seasonal changes in the strength and direction of prevailing winds and in the scavenging efficiency of such winds, together with the variations in meteorological conditions that influence the formation of marine aerosols across the eastern Mediterranean basin.

Conspicuous changes in color of the samples collected on the samples are also seasonally influenced and the paler colors of the late spring and summer samples appear to be caused by enhanced levels of Fe and Mn, the oxidised forms of which are responsible for masking the "normal" darker hues associated with the less variable content of organic carbon. In general, the levels of Al and Fe recorded in this study are somewhat lower than both average crustal values and the contents recorded by Yaalon and Ganor (1979). These differences are attributed to the different sampling strategies adopted in these two long-term observations. The average values quoted here are derived from the total range of samples collected at regular intervals throughout the year and the relative contribution of crustal material in these samples, therefore, fluctuates considerably. In contrast, the crustal factor in the samples of Yaalon and Ganor (1979), collected during large storm-events, is likely to be more constant and less influenced by non-crustal inputs.

The dominance of rounded and etched quartz grains and rounded calcite particles in the dusts is consistent with (but does not necessarily prove) ultimate derivation from arid source-terrains marked by intense physical abrasion. Despite the fluctuations in total clay mineral abundance the relative proportions of the three main clay species remain remarkably constant, demonstrating that the clays probably are derived from the same ultimate source. The prevalence of smectite and palygorskite in the clay fraction attests to the present and past arid conditions in the aeolian source-areas (Sahara–Arabia), while the persistent occurrence of subordinate kaolinite is attributable to more humid conditions, perhaps reflecting past (Quaternary) cycles but some contribution from existing deltaic and lacustrine settings cannot be excluded.

The chemical components of the dust samples can be assigned to crustal and anthropogenic categories and in this context the use of the enrichment factor (EF) of Rahn (1976) for heavy metal contents is revealing. Thus, the color influencing seasonal

variations in Fe and Mn (non-enriched or crustal elements) reflect major changes in the long-distance supply of aeolian materials while the fluctuations in both enriched (anthropogenic) elements (e.g. Zn, Pb, and Cd) and moderately enriched components (e.g. Ni, Cu and Cr) result from more local or "abnormal" supply.

Thus, even within the limitations of the mesh-sampling techniques used in this study it is possible to ascribe most of the mineralogical and chemical properties of the collected dusts to relatively regular changes in the long-range transport of aeolian materials from desert areas surrounding the eastern Mediterranean basin, coupled with episodic supply from pene-marine, local and anthropogenic sources. It is recognised, however, that confirmation and better quantification of the particulate fluxes identified in this preliminary study require the deployment of more sophisticated sampling systems (including Hi-Vol samplers) and a more extensive network of sampling stations. This further work is now underway.

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