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An Approach to the Assessment of Local Trace Metal Pollution in the Mediterranean Marine Atmosphere

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Assessing the extent of local trace metal pollution in marine atmospheric particulates presents a number of difficulties, and one potential approach to the problem has been evaluated by reference to a series of aerosols from the lower troposphere over the Eastern Mediterranean. This region, which is less remote from the continents than open-ocean areas, has two significantly different atmospheric particulate catchments: being bordered in the north by nations having industrialized, semi-industrialized and rural economies, and in the south by the North African desert belt. The elemental chemistry of the particulates is illustrated in terms of the distributions of Fe and Pb, which are presented in the form of enrichment factor diagrams, and is shown to be essentially controlled by the dilution of a 'European' background material, common to 'remote' regions of the latitudinal belt, with crust-derived, desert components. Major local perturbations in the dilution relationship can occur when either crust-derived or pollutant components strongly dominate the total particulate population. The extent of these local perturbations can be tentatively assessed using appropriate enrichment factor diagrams, which offer a framework within which to interpret the elemental chemistry of marine atmospheric particulates providing sufficient data is available for their construction. By using such an EF diagram it is suggested that the Eastern Mediterranean atmospheric particulates have not suffered local pollution on a gross scale with respect to Pb.

Pollution of the marine atmosphere has received increasing attention over the last decade. Particulates from the lower troposphere have been collected at various continental and marine sites, including those adjacent to centres of urban and industrial activity (see e.g. Pierson *et al.*, 1974; Cambray *et al.*, 1975) and those from 'remote' areas (see e.g. Duce *et al.*, 1975; Maenhat *et al.*, 1979). Data resulting from collections such as these have shown that over certain regions of the World Ocean the atmospheric population is dominated by crust-derived components. One such region is that underlying the Atlantic north-east trades which receives a large input of material from the North African desert belt (Chester *et al.*, 1972; Prospero & Carlson, 1972), the chemical composition of which is largely controlled by that of the crust-derived solids raised into the atmosphere from the reservoir of loose arid soils (Chester & Stoner, 1974). However, in remote regions which do not receive a large input of crust-derived solids certain elements in the particulates are considerably enriched with respect to crustal material. Further, it has been suggested (see e.g. Duce *et al.*, 1975) that these enrichments are of a similar magnitude in remote regions of both the northern and southern hemispheres, and a number of attempts have been made to assess the elemental composition of unpolluted 'background' air and to establish whether or not it differs from that in non-remote regions.

Problems in establishing the causes of trace metal enrichments in global atmospheric particulates make it difficult to assess the effects of local sources of the metals in specific oceanic regions. However, in certain regions the catchments, which supply the material forming atmospheric particulates, are reasonably well-defined and by investigating the elemental compositions of particulates from such regions it should be possible to provide a general framework within which to interpret any local compositional variations which occur. In the present paper, such a framework is suggested for atmospheric particulates from the Eastern Mediterranean, and it is proposed that it could be applied to other areas of this sea for which aerosol trace metal pollution is suspected.

In recent years the Mediterranean Sea has been the subject of a great deal of scientific attention with respect to marine pollution, and was singled out by the United Nations for special regional consideration (Anon, 1975). From the point of view of atmospheric trace metal chemistry, the

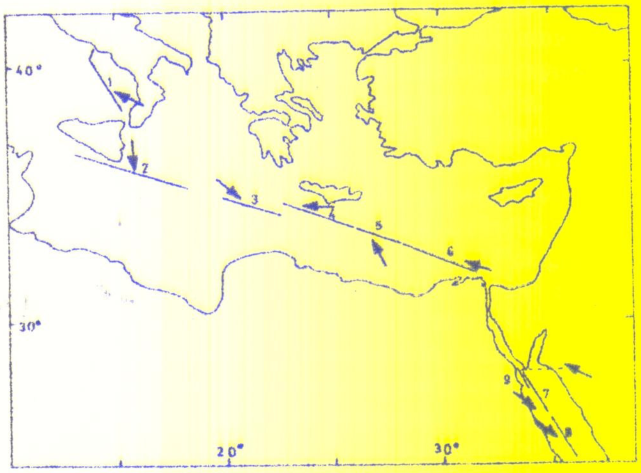

Fig. 1 Collection of atmospheric particulates. The solid lines indicate the ship's track during the collection of each sample, and the arrows the prevailing wind direction.

lediterranean provides an excellent example of a marine egion which has a number of contrasting catchments. It is ordered in the north by nations having a variety of conomies, ranging from industrial and semi-industrial to gricultural, and in the south by the North African desert elt. There is also volcanic activity in the region, and in ddition the Mediterranean is one of the most crowded eaways in the World Ocean.

Chester *et al.* (1977) were able to show that soil-sized tmospheric particulates in the Eastern Mediterranean onsist of at least two individual populations, one riginating on the European mainland and one in the deserts of North Africa and the Middle East. Because of the resence of these two distinct populations it was decided to arry out further particulate collections from the lower tmosphere over the Eastern Mediterranean and Red Sea, sing a high volume filtration system which samples all article sizes. By this means it was hoped that the effects of he different catchments on the elemental chemistry of the articulates could be evaluated, and so be extrapolated to rovide a framework within which to identify the presence of local atmospheric pollutant components.

Atmospheric Particulate Samples

The locations of the samples, which were collected on board *R.R.S. Shackleton* during the period September 979-February 1980, are illustrated in Fig. 1, together with he prevailing wind directions. With the exception of No. 9, all samples were collected using a high volume filter system. This system, which was positioned ahead of the ship's bows, incorporated Whatman 41 filters. Sample 9 was collected sing a mesh technique, but is included here because it was aken in dust storm conditions in the Red Sea and represents he best available example of a 'pure' population from the North African desert belt catchment. Meshes collect a size-fractionated atmospheric particulate sample (Goldberg, 971; Parkin *et al.*, 1972) under most conditions, but when arge-sized crustal components dominate the population hey sample essentially the same material as filters (Chester & Stoner, 1974).

Analytical Procedures

Whatman 41 filters were dissolved in redistilled HNO_3 and Aristar HF in PTFE beakers and the digests stored in polystyrene bottles. The mesh-collected sample was washed from the mesh in redistilled water and treated in the same manner as the filter samples. A series of elements was then determined in the digests using either flame or flameless atomic absorption.

Results and Discussion

The elements Al, Fe, Mn, Cr, Ni, Cu, Zn and Pb were included in the analyses, and these are used to establish certain characteristics within the Eastern Mediterranean particulates. However, for the purpose of defining a framework within which to describe the chemistry of the particulates, attention in the present paper is focused on two elements which have contrasting distribution patterns, i.e. Fe and Pb, and Al which is used as an indicator element.

TABLE 1

Elemental concentrations and enrichment factors in the Eastern Mediterranean atmospheric particulates.

Sample No.	Al		Fe		Pb		Prevailing wind direction	Rainfall ‡
	Conc.*	EF †	Conc.	EF	Conc.	EF		
2	60	1.0	55	1.3	25	2778	NNW	+
3	113	1.0	76	1.0	20	1180	NW	+
1	216	1.0	128	1.5	9.0	227	SE	×
8	416	1.0	294	1.0	7.0	112	NW	×
7	874	1.0	647	1.0	16	122	NW	×
4	1030	1.0	645	0.9	16	101	E	×
5	1150	1.0	665	0.9	2.3	13	SE	×
6	1620	1.0	751	0.7	9.0	37	ESE	×
9	4348	1.0	3756	1.3	2.3	3.5	ESE	×

*Conc. in ngm^{-3} of air.

† For explanation see text.

‡ + indicates rain in the general area.

× indicates the absence of rain in the general area.

The atmospheric concentrations of Al, Fe and Pb are listed in Table 1, in which they are ranked on the basis of increasing Al concentration. It is apparent that there are considerable variations in the concentrations of Al, and that these variations are strongly dependent on the prevailing meteorological conditions such as wind direction and rainfall. Because total particulate loadings vary to such an extent, both temporally and spatially, various authors have used an enrichment factor (EF) to estimate the origin of the elements in atmospheric solids. The most commonly used EF employs Al as a reference element for crust-derived material and is calculated according to the equation: $EF_X = (C_{XP}/C_{AlP})/(C_{Xc}/C_{Alc})$, in which C_{XP} and C_{AlP} are the concentrations of an element X and Al respectively in the particulate, and C_{Xc} and C_{Alc} are their concentrations in average crustal material. Enrichment factors close to unity are taken as an indication that an element has a mainly crustal origin, and those $\gtrsim 10$ are considered to indicate that a substantial portion of an element has a non-crustal origin. The EFs for Fe and Pb are included in Table 1.

It is evident from the data in Table 1 that although the atmospheric concentrations of Fe vary over three orders of magnitude the EFs are all close to unity, i.e. Fe is not enriched in any of the Eastern Mediterranean particulates relative to the crust. In contrast, the EFs for Pb vary over four orders of magnitude in the sample set, and in many of the particulates this element is very considerably enriched relative to the crust. It can be seen from Table 1 that over the entire region the variations in the EFs of Pb are related to variations in the concentrations of Al in the atmosphere and, following Rahn (1976), this relationship is expressed in the form of an enrichment factor diagram in Fig. 2, which also includes Fe. The EF diagram for Fe has a form which is characteristic of non-enriched atmospheric elements (Rahn, 1976), i.e. the EFs plot in a broad horizontal field, which has only a relatively small vertical range, and are independent of variations in the concentration of Al. The EF diagram for Pb, however, is characteristic of those of enriched atmospheric elements, i.e. the EFs plot as part of a triangular field with the hypotenuse along a line of constant concentration which forms an apparent lower limit of concentration (Rahn, 1976). Anthropogenic sources strongly affect the concentrations of Pb in the marine atmosphere (see e.g. Buat-Menard & Chesselet, 1979), and this element may therefore be used to illustrate the parameters which control the atmospheric particulate chemistry of enriched elements in a region which has inputs from both crustal and

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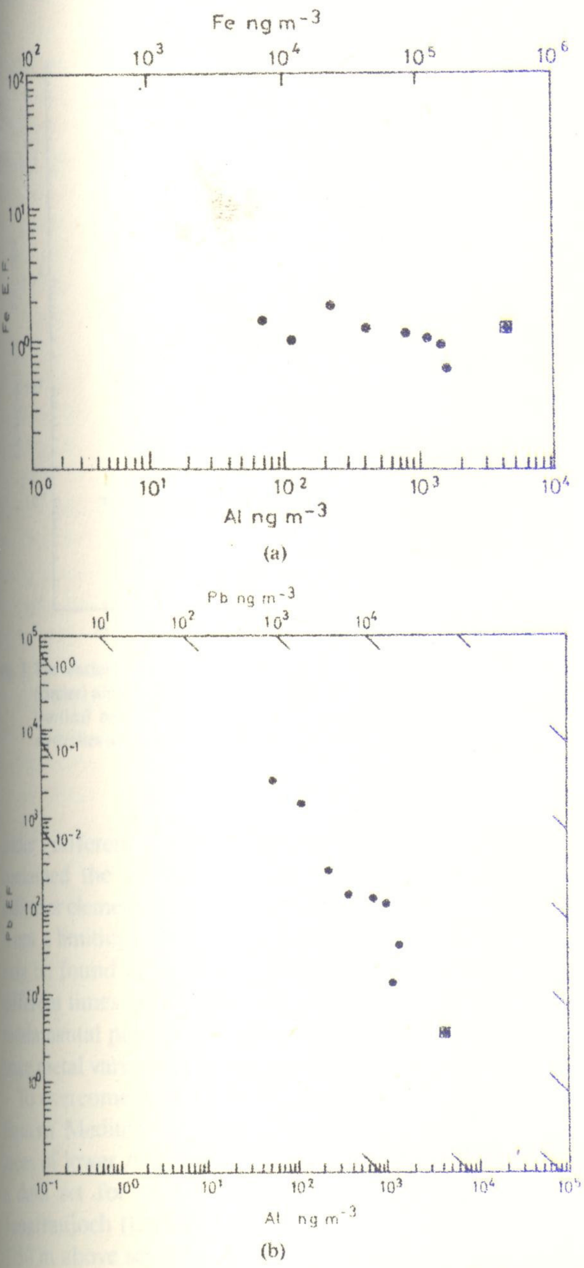

Fig. 2 Enrichment factor diagrams for the Eastern Mediterranean particulates. Data points for samples 1–8 are plotted as solid circles, and that for sample 9 (the desert-derived 'end-member') as a solid circle within a square. Fig. 2a, Fe; Fig. 2b, Pb.

non-crustal, or 'background', sources. In the lower atmosphere over the Eastern Mediterranean it is apparent from the present data that there is a background component, which contains relatively high concentrations of Pb (and other enriched elements) and presumably has a small particle size (Duce *et al.*, 1976). This background will itself contain anthropogenic Pb, and in the Eastern Mediterranean its composition is diluted by an Al-rich crust-derived component which contains low concentrations of the enriched elements and has most of its particles in the soil-sized range (Chester *et al.*, 1972). The mutual proportions of the two components, which depend on the prevailing meteorological conditions, control the overall elemental chemistry of the present particulates, and it is therefore of interest to examine the EFs of these two 'end members' from the Eastern Mediterranean atmosphere.

TABLE 2

Elemental enrichment factors in soil-sized atmospheric particulates from the desert belt of North Africa

	Enrichment factor*		
	Atlantic north east trades, off the coast of West Africa		
Element	Data from Chester *et al.* (1979)	Data from Cross (1979)	Sample 9, present work; Red Sea
Al	1.0	1.0	1.0
Fe	0.8	1.0	1.3
Mn	0.5	1.2	0.8
Cr	1.0	0.7	1.4
Ni	0.9	0.7	1.3
Cu	0.9	1.4	0.9
Zr	2.0	4.0	2.0
Pb	7.7	11	3.5

*For explanation see text.

From the present data the best estimates of the compositions of the two 'end members' are given by samples 2 and 9. Sample 9, which was collected in dust storm conditions in the Red Sea, may be assumed to be representative of the crust-derived desert component transported to the Eastern Mediterranean. The EFs for a number of elements in sample 9 are listed in Table 2, together with those for a series of samples collected in the Atlantic north-east trades, i.e. from a catchment area at the western end of the North African desert belt. It can be seen from this table that the EFs of all the elements are reasonably similar, i.e. $\lesssim 10$, in both the Eastern Mediterranean and the Atlantic north east trade samples. It may be concluded, therefore, that sample 9 provides an acceptable estimate of the elemental composition of the crust-derived material raised into the atmosphere from the North African desert belt. Sample 2 has the lowest Al concentration and so offers the best example from the present data set of the 'background' material which is diluted by the desert component. Duce *et al.* (1975) provided data on the range of EFs for a number of elements in particulates collected from the lower troposphere over the North Atlantic north of 30°N and compared them graphically with those of samples taken at the South Pole. In Fig. 3 a similar graphical comparison is made between the range of EFs of the North Atlantic samples and those of sample 2 and it is apparent that, with the exception of Cu, the EFs of the Eastern Mediterranean sample all lie within the ranges found over the North Atlantic. The tentative conclusion which can be drawn from the data comparison made in this way is that the elements in the background components from the two regions have similar sources.

There are difficulties, however, in using EFs alone to compare different sample sets. For example, Adams *et al.* (1980) have suggested that EFs should only be used to characterize aged, or well-mixed, aerosols of nearly uniform composition. One of their objections to the use of EFs for aerosol characterization was that there can be large temporal variations in them at a given site. This objection is at least equally valid for the Eastern Mediterranean samples which were collected at different sites since it has been shown that the EFs of the enriched element Pb vary considerably, being dependent, among other factors, on the amount of desert component present. That is, the kind of variations in sample composition found along the present sample transect are similar to those that could occur at a single site

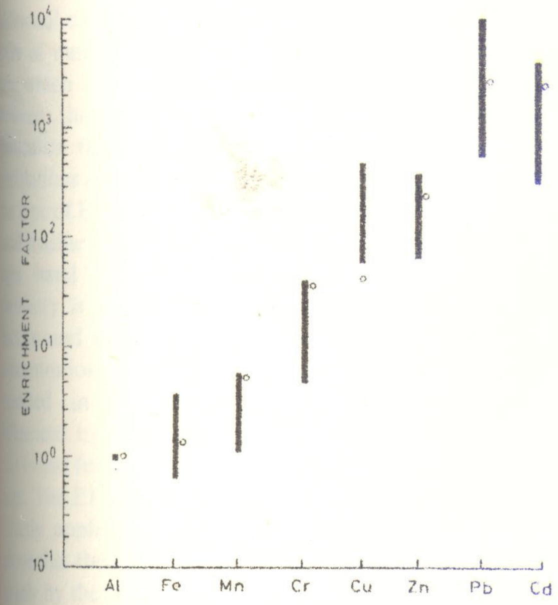

Fig. 3 The enrichment factors for sample 2, Eastern Mediterranean (circles) and the enrichment factor geometric standard deviations (vertical bars) for particulates collected in the North Atlantic westerlies north of 40° N (after Duce *et al.*, 1975).

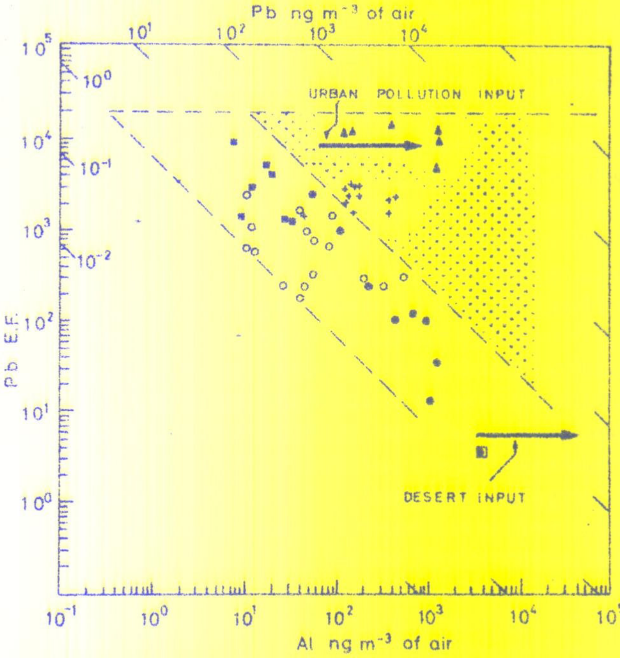

Fig. 4 Composite enrichment factor diagram for Pb. This diagram includes the Eastern Mediterranean samples (solid circles, sample 9 in square), samples collected at a remote mountain station in Switzerland (open circles; data read from figure), samples collected at a series of UK non-urban sites (crosses), samples collected over the North Atlantic north of 40°N (solid squares) and polluted samples collected at Ghent, Belgium (solid triangles) – for references, see text. The 45° broken lines enclose the field in which the elemental chemistry of the particulates is controlled largely by the dilution of the 'European' background material by crustal solids. The arrow at the lower right hand corner of the diagram indicates an input of soil-sized, desert-derived, crustal solids, and when these predominate the EFs will be generally constant for all elements and the diagram will be extended horizontally to the right under the constraints of low EFs at high Al concentrations. The arrow at the upper middle section of the diagram indicates an input of pollutant-rich urban particulates, and when these predominate the diagram will be extended horizontally to the right under the constraints of high EFs at high Al concentrations. This is a much more simplistic Pb EF diagram than that given by Rahn (1976), which included data points from many regions outside the Mediterranean latitudinal belt. On the present diagram some would fall to the left of the broken lines, indicating a lower minimum Pb concentration in certain aerosols, e.g. that at the South Pole. Others would fall to the right of the broken lines and cover a wide range of EFs in the field lying between the urban and desert input arrows, i.e. in the stippled field. Comprehensive EF diagrams, therefore, exhibit a wide scatter of data points. However, the purpose of the present investigation is to attempt to use such diagrams to provide a tentative framework within which to assess the elemental compositions of atmospheric particulates from specific regions, here the Mediterranean Sea, by identifying within the plots the general field in which the particulate compositions are controlled by the dilution of non-local background material by crustal solids. For example, samples from the Mediterranean atmosphere having Pb EFs $\gtrsim$ 10 which plot in the stippled field should be suspected of containing at least one local component in addition to those in background and crustal solids. Further, the degree to which the additional local component(s), which may have a pollution source, affects the elemental composition of the sample will be reflected, to a first approximation, by the position in which it plots, i.e. by the magnitude of the EF relative to the Al concentration.

under different meteorological conditions. Sample 2 contained the smallest concentration of Al, the crustal indicator element, but it is important to understand that this is not a limiting concentration and that still smaller values may be found at other sampling sites in the region or at different times at the same site. This, therefore, presents a fundamental problem in assessing the importance of local trace metal variations in any region.

To overcome this problem it is necessary to extend the Eastern Mediterranean EF diagrams to the left, i.e. to areas of lower Al concentration. In order to illustrate this, a data set for Pb in atmospheric samples collected at Jungfraujoch (Dams & De Jonge, 1976), a mountain site 3750 m above sea level in Switzerland, has been added to the EF diagram for this element for the Eastern Mediterranean samples (see Fig. 4). It can be seen that there is a generally good fit between the two data sets and that, relative to Al, the background material which is diluted by crust-derived components at the Swiss mountain site has a reasonably similar composition, at least with respect to Pb, to that over the Eastern Mediterranean. The EF diagram for Pb has been further extended to the left by the inclusion of 7 samples taken over the remote North Atlantic north of ~40°N and reported by Duce *et al.* (1973). This data set also fits the overall relationship found for the Eastern Mediterranean, i.e. the dilution of a background material by crust-derived solids. This background material is apparently common to remote areas lying in the latitudinal belt which includes most of Western Europe, and may be referred to as the 'European' background material. In order to assess the effects of various urban and rural atmospheric components on this background material two further Western European data sets have been included in Fig. 4. One consists of samples collected at Ghent, Belgium (Demnynck *et al.*, 1976), for which the bulk of the elements were shown to have an anthropogenic origin. These data points plot well to the right of those for the Eastern Mediterranean, Swiss and North Atlantic populations, i.e. they have relatively high Pb EFs even at high Al concentrations. The second data set is for a series of non-urban UK sites (Cawse, 1978, 1980). These are less 'remote' than the elevated Swiss mountain station and the data points generally fall to the right of the field in which the elemental chemistry of the particulates is largely controlled by the dilution of the 'European' background material with crustal solids, but below the urban polluted sample grouping. These additional data serve to show that although there is a ~ 2 order of magnitude range of EFs for each Al concentration, which results in sample overlap, there is a gradation between the 'remote', non-urban and urban-polluted particulate fields. Further, the separation of the various populations appears to be sufficient enough to suggest that this type of EF diagram has potential for the assessment of local marine atmospheric pollution. In Fig. 4 much of the right hand (stippled) field, in which the particulate chemistry is not controlled only by the dilution of the background material by crust-derived solids, falls at Al concentrations $\gtrsim 10^2$ ng m^{-3} of air. These values are to be expected in non-remote marine regions which are influenced by the input of continental solids, and are in fact found for most of the Eastern Mediterranean particulates. The EF diagram shown in Fig. 4 can therefore be directly applied to these particulates. It was suggested above that their elemental composition could be explained largely by the dilution of a background material by crust-derived solids. From Fig. 4 it can also be shown that, for Pb, this dilution process is similar to that operative over other remote marine and continental regions within the same latitudinal belt. Atmospheric metal loadings over the Eastern Mediterranean may be higher (i.e. $\gtrsim 10^2$ ng m^{-3} of air) than those over open-ocean areas of the North Atlantic, due to the relative proximity of the Mediterranean to continental land masses, and so metal fluxes to the sea surface will be higher. However, the important conclusion which may be drawn from the present investigation is that, for Pb, the general particulate population over the Eastern Mediterranean is no more 'polluted' than are those over remote marine and continental sites in the same latitudinal belt. That is, any anthropogenic Pb in them has been mainly transported from outside the immediate area, and they have not suffered local pollution on a gross scale.

From the EF diagram in Fig. 4, it can be seen that over the Eastern Mediterranean perturbations in the concentrations of Pb in the 'European' background material arise mainly from the injection of crust-derived solids into the atmosphere. The extent of the dilution caused by these crust-derived solids is extremely dependent on prevailing meteorological conditions (see also Demnynck *et al.*, 1976). It may be that atmospheric particulates from other parts of the Mediterranean will also be free from local pollution, and EF diagrams of the form shown in Fig. 4 will be useful in establishing this to the extent that they will reveal whether or not their elemental compositions are controlled by the dilution of the 'European' background material, transported from outside the immediate area, by crust-derived solids.

Conclusions

Enrichment factor diagrams of the form shown in Fig. 4 offer a potential framework within which the elemental chemistry of Mediterranean atmospheric particulates can be described, especially with regard to the degree of local pollution they have suffered. This has been illustrated in terms of the enriched element Pb for which it has been shown that atmospheric particulates from the Eastern Mediterranean have their compositions controlled largely by the dilution of a 'European' background material by crustal solids originating in adjacent desert belts. These particulates have not apparently suffered local pollution on a gross scale, but have had any pollutant Pb mainly transported from outside the immediate region.

For the Eastern Mediterranean atmospheric particulates, the identification of the 'European' background material is critical to the interpretation of the EF diagram. However, it must be further stressed that this background material itself will include pollutant components and that background aerosols from different regions are probably best defined by minimum concentrations as first suggested by Rahn (1976) and later by Adams *et al.* (1980). In order to determine these minimum concentrations more data is required on sample sets from other marine regions in both the northern and southern hemispheres. From such data it may be possible to establish whether or not the 'European' background material has a similar elemental composition to those from other localities. Nonetheless, for the Mediterranean Sea itself EF diagrams having the general form shown in Fig. 4, i.e. using the Eastern Mediterranean as a 'baseline', can be used as an aid for the interpretation of the elemental chemistry of atmospheric particulates in terms of local pollution.

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