The use of mineral magnetic measurements to study the transport and sedimentation of particles in the Gulf of Lions (NW Mediterranean)

Magnetic parameters
Sediments
Anthropogenic
Gulf of Lions
Saharan dust

Paramètres magnétiques
Sédiments
Anthropogénique
Golfe du Lion
Poussières sahariennes

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ABSTRACT

Mineral magnetic and chemical studies indicate the impact and great importance of atmospheric *versus* riverine input of particles in the sediments of the Gulf of Lions and provide some strong indications about the presence of anthropogenic particles in offshore deep sediments of this region. The data indicate that Saharan dust and anthropogenic particles coexist at several depth intervals in the sediment, supporting the hypothesis that Saharan dust episodes could play an important role in facilitating deposition and coprecipitation of anthropogenic particles from the atmosphere and the water column.

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RÉSUMÉ

Étude de transport et sédimentation des particules du Golfe du Lion utilisant les mesures des paramètres magnétiques

Les mesures des paramètres magnétiques et l'analyse élémentaire des échantillons de sédiments prélevés dans le Golfe du Lion indiquent que l'apport atmosphérique peut avoir un impact aussi important que l'apport tellurique. Ces mesures fournissent aussi des indications importantes concernant la présence des particules d'origine anthropogénique dans les sédiments profonds prélevés en pleine mer. Nos données montrent que des particules, tant d'origine saharienne qu'anthropogénique, existent aux différentes profondeurs dans les sédiments. Ceci peut signifier que les épisodes de transport des poussières sahariennes peuvent jouer un rôle important dans le dépôt et la coprécipitation des particules anthropogéniques par voie atmosphérique dans la colonne d' eau.

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INTRODUCTION

In this paper, by using a somewhat rarely employed technique, namely mineral magnetic measurements, in combination with conventional chemical analyses, we shall attempt to increase our understanding of the coexistence of anthropogenic particles in offshore deep sediments of the NW Mediterranean, their origin and the most probable major mechanism of their transport and deposition to the sea bottom.

It has been shown that the magnetic properties of particulate matter and sediments can provide valuable information concerning a number of processes. Scoullos *et al.* (1979) and Scoullos and Oldfield (1986) have used magnetic measurements in the marine environment in order to study the fate, transport and deposition of particles through major pathways.

Up to now, mineral magnetic measurements have been applied in several systems, including lakes, estuaries, enclosed gulfs and atmospheric samples, with the aim of increasing our knowledge about erosion or pollution mechanisms (Hilton, 1987; Oldfield *et al.*, 1981; Oldfield *et al.*, 1985 *a*; Hunt, 1986; Thompson *et al.*, 1980). The present paper reports on their application in the framework of the European River Ocean System "EROS-2000" project which, to the best of our knowledge, constitutes the first attempt to employ them in an open, deep sea system.

SAMPLING AND METHODS

The area studied [Gulf of Lions (Fig. 1)] was sampled in July 1987 from R/V *Tyro* and in May 1990 from R/V *Marion Dufresne*. Sediment samples were collected from twenty stations by means of a box-corer, specially designed at the NIOZ laboratories. Subcore undisturbed samples were then obtained in perspex tubes and pre-measured on board for their magnetic susceptibility (x) (volume specific) employing a Bartington susceptibility meter MS1 with a whole core loop sensor. Upon completion of these measurements the cores were carefully sliced, into sub-samples 1 to 2 cm thick, using plastic tools, down to 10 cm depth for the *Tyro* samples and 20 cm depth for the *Marion Dufresne* samples, placed in plastic bags and stored at 4°C. Upon arrival in the laboratory, the sediments were wet sieved and air-dried at < 40°C and the < 61 µm fraction was measured for a wide range of magnetic parameters, while a series of parallel chemical analyses was conducted in our laboratory and in the laboratories of other participants in the EROS-2000 programme. For comparison reasons, this paper only discusses the profiles of the first ten centimetres of the sediment cores, in combination with the chemical findings on trace metals, Pb^{210} and PAHs made by other independently working groups (Nolting and Helder, 1991; Zuo *et al.*, 1991; Lipiatou and Saliot, 1991).

The dried and powdered sediments were weighed into 10 ml polystyrene pots that had been acid-washed and pre-measured for their minimum magnetic contamination. It should be noted here that the method is non-destructive and the samples are left intact.

The magnetic parameters measured were the following:

- Magnetic susceptibility *x* ($10^{-8} m^3/kg$), represents the ease with which a material can be magnetized. The x is measured in weak alternating fields (< 0.1 mT) at low (1 kHz) and high (10 kHz) frequencies, using a Bartington susceptibility meter MS1. The x low (x_l) is an estimate of the concentration of the magnetic minerals present in a sample;

- Frequency dependent susceptibility *x fd%*, defined by the ratio $[(x_l - x_h)/x_l] * 100$ and very helpful in identifying very fine grains with dimensions close to the superparamagnetic /stable single domain boundary (0.03 µm), that are difficult to recognize otherwise. Their x_h is low due to relaxation phenomena;

- Saturation Isothermal Remanent Magnetization (SIRM): mAm^2/kg. This parameter, which is chiefly a measure of magnetite content, is measured in a fluxgate magnetometer (Minispin, Molspin Ltd) after placing the sample in a strong ("saturation") uniform d.c. magnetic field (1 000 mT) at a given temperature (24°C).

- Anhysteretic Remanent Magnetization (ARM): $mA\ m^2/kg$. This parameter is also measured in a fluxgate magnetometer (Minispin, Molspin Ltd) after magnetizing the sample in a alternating field of 100 mT in the presence of a weak

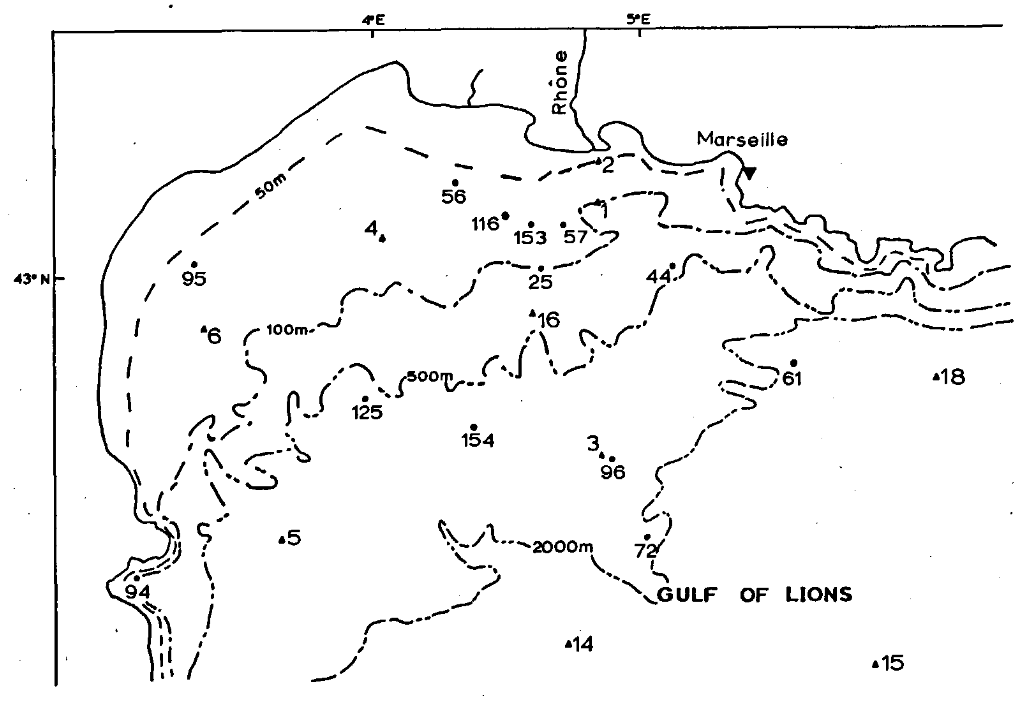

Figure 1

Area studied and stations sampled.

steady field of 0.4 mT (AF demagnetizer, Molspin Ltd). ARM is very sensitive to the small grain size fraction (< 1 μm) present in the sample (Gillingham and Stacey, 1971)

RESULTS AND DISCUSSION

Table 1 summarizes the results concerning the magnetic parameters measured in the top layer (surface 1 cm) of the sediments of the Gulf of Lions. From these results one can immediately distinguish the nearshore stations, with x values lower than 20 (10^{-8} m^3/kg), SIRM values lower than 2 000 (mA m^2/kg) and ARM values lower than 60 (mA m^2/kg), from the offshore ones which have considerably higher values for x, SIRM and ARM.

The aforementioned classification of stations implies that at stations 56, 57, 116, 153, 95, 1, 2, 4, 6, 25 and 16 with depths less than 1 000 m (group 1), where all the magnetic parameters measured have low values, there is high accumulation of coarse (> 1 mm), not strongly ferrimagnetic grains, apparently of riverine and terrestrial origin. This is supported by the relatively low values of the magnetic parameters ($13.4 < x < 18.8$ $10^{-8}m^3$/kg, $21.2 < ARM < 52.8$ mAm^2/kg) measured on five surface sediments collected from the Rhône estuary, which fall within the range of values of the near-shore sediments.

At the deeper stations 3, 5, 14, 15, 18, 154, 96, 72 and 61 (group 2), the x values are higher, ranging between 21 and 26 ($10^{-8}m^3$/kg), whereas at station 96 the x value is extremely high (50 * $10^{-8}m^3$/kg). Similarly the SIRM values range between 2 157 and 3 200 (μAm^2/kg); and at station 96 SIRM is 6 329 (μAm^2/kg), and ARM values range between 65 and 90 (μAm^2/kg). These magnetic readings indicate the presence of high concentrations of ferrimagnetic material at the offshore stations. The exceptionally high values of SIRM at station 96, and to a lesser extent at stations 61 and 18, followed by high values of ARM, reflect considerable accumulation of very fine (< 1 mm) particles rich in ferrimagnetic material, particularly in the surface sediments of these stations.

The x fd % values in most stations of the studied system exceed 2 %, indicating the presence of ultra-fine (< 0.03 μm) particles, which, with the exception of few stations (57, 116, 95), cannot be otherwise easily identified throughout the study area. At stations 1, 2, 6, 16, 3, 14 and 15, x fd % values exceed even 8 %, a percentage usually found in surface soils. The absence or very low x fd % values could indicate a negligible contribution from erosion and runoff and higher contribution from anthropogenic sources, particularly if the low x fd % is paralleled by high x and SIRM values (Scoullos *et al.*, 1979).

The geographical distribution of the two above-mentioned groups of stations coincides with the interaction boundary between the water mass influenced by the Rhône river and the open sea, which has been identified in the work of Guieu *et al.* (1991) and follows approximately the ~ 2 000 m depth isocontour.

The observed distribution of the magnetic characteristics could be partly attributed to the cyclonic circulation prevailing in the area (Millot, 1987; Millot and Monaco, 1984). As the riverine particulate matter is discharged into the Gulf of Lions, the coarser grains, which are magnetically less active, are deposited rapidly outside the river mouth in the area situated on the prolongation of the river axis, including stations 1, 2, 56, 57, 116, 153, 16 and 25, and to the west of it (stations 4, 6 and 95), whereas only the fine grains reach the outer stations 3, 5, 154, 14, 96 and 72.

If we consider that the Rhône river provides the major riverine input of particles in the area and that ~ 90 % of these particles are deposited within a distance of 6 km from the river mouth, as reported by Martin *et al.* (1989), then only some 10 % of the load discharged by the river could contribute to the intense magnetic behaviour of the offshore surface sediments. It should be clarified, however, that in the magnetism could also contribute particles, smaller than 0.4 μm, which are not included in the aforementioned balance, since suspended matter concentrations are usually determined by filtration of a 0.4 μm pore-size filter.

Study of the downcore profiles of the magnetic parameters reveals interesting features which facilitate our understanding of the evolution and variation of the sedimentation patterns.

The stations located far from the river mouth (96, 61, 72, 3, 14, 15 and 18), at depths > 1 000 m, have low sedimentation rates (~ 0.05 cm/yr) according to detailed Pb^{210} investigations (Zuo *et al.*, 1991) and exhibit maximum values of magnetic parameters in the surface layers of the sediment column, indicating a differentiation in nature and texture

Table 1

Magnetic parameters of surface sediments in the Gulf of Lions.

Stations	x[a]	x_{fd}%	SIRM[b]	ARM[b]
56	16.67	2.89	1876	58.8
57	15.16		1767	50.22
116	15.94		1792	57.55
153	14.29	5.36	1538	47.08
95	15.66		1780	64.71
1	18.70	17.8	1611	
2	18.00	11.4	1900	
4	16.40	6.90	1690	
6	17.90	11.3	1851	
25	14.89	3.88	1417	37.78
16	20.00	11.9	1497	
154	20.91	5.26	2249	66.86
96	49.96	2.68	6329	88.92
72	20.47	4.88	2263	65.32
61	26.08	4.16	3692	75.05
5	22.50	5.20	2556	
3	21.30	8.90	2492	
14	19.30	9.60	2154	
15	21.50	13.0	2157	
18	26.30	4.30	3200	

[a] *In* 10^{-8} m^3/kg
[b] *In* μA m^3/kg.

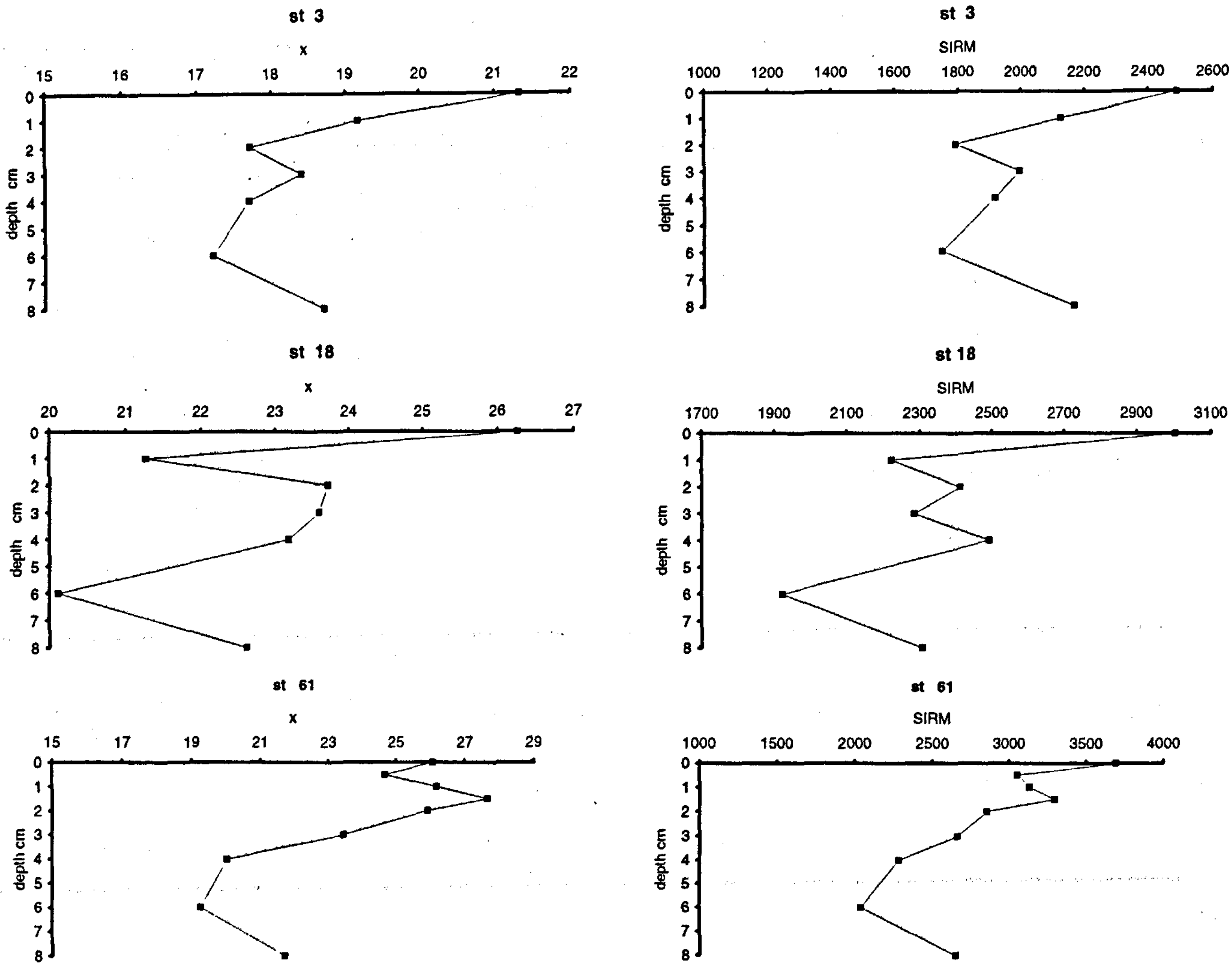

Figure 2

a) Depth profiles of magnetic susceptibility (x) (10^{-8} m^3/kg) for cores collected at the outer part of the Gulf;.b) Depth profiles of SIRM (μA m^2/kg), for the same cores. The increased values at the top first centimetres are obvious.

between surface sediments deposited during the last thirty years and the subsurface ones. Such significant differences between surface and subsurface layers are not observed on the profiles of the nearshore stations, where higher sedimentation and mixing rates occur. Figure 2 presents the variations of x and SIRM for typical offshore stations (18, 3 and 61), while Figure 3 presents the same parameters for nearshore stations (116, 56 and 2). In Figure 4 the ARM downcore distributions are given for the deep stations 61 and 72. It may be observed that ARM reaches considerably high values downcore (up to 100 μA m^2/kg).

On the basis of the x and SIRM values, we could predict that the surface sediments of these stations will have a high content of fine (< 1 μm) strongly ferrimagnetic particles. However, the profiles of Figure 4 indicate the presence of higher portions of similar and even smaller particles in the subsurface layers. This could be related to the known diminishing loads of particulate matter discharged by the Rhône in recent years (due to the construction of dams upstream) in relation to particles contributed by other sources (*e. g.* biogenic, *etc.*).

The alteration in the relative ratio of particulate populations is also paralleled by the increase of the ARM/x ratio, with a simultaneous decrease of the SIRM/ARM ratio downcore (*e. g.* stations 61, 72), a feature that is not observed at the nearshore stations [*e. g.* stations 153 (Fig. 5)]. The ARM/x ratio also provides a useful estimate of the grain sizes present in the sample, since ARM is more sensitive to the finer grain size fraction and x to the coarser one (Banerjee *et al.*, 1981; King *et al.*, 1982). The SIRM/ARM ratio could also be used.

In an endeavour to explain our experimental findings, and particularly the enrichment of magnetically active particles in the surface layer of the offshore stations, three working hypotheses could be advanced:

1) The river has, over recent decades, contributed higher percentages of magnetically active particles, due to alterations in its catchment area; and/or there is a higher contribution of fine material of anthropogenic origin carried by the river.

2) The peak is mainly due to atmospherically-transported ferrimagnetic particles discharged at an increasing rate into

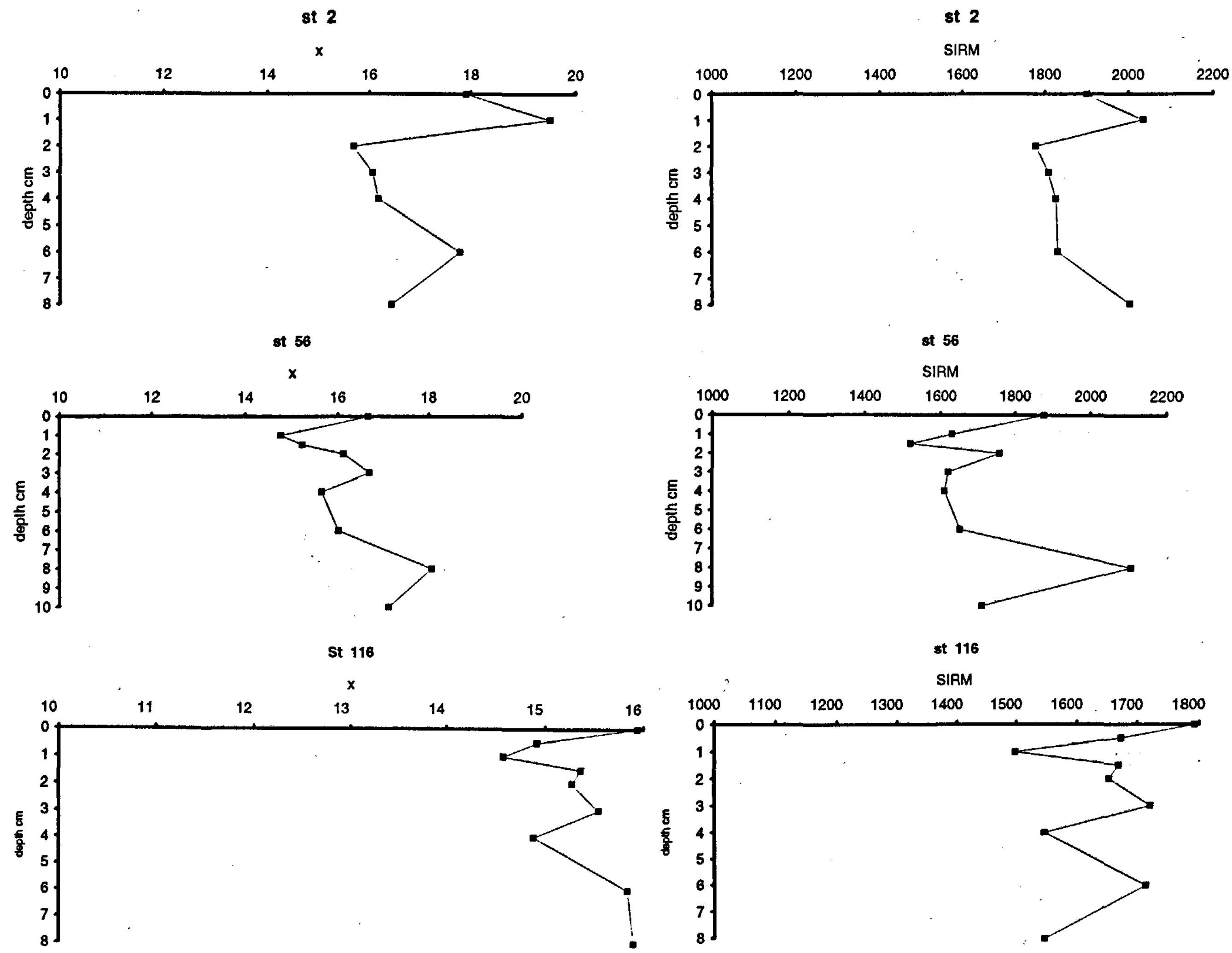

Figure 3

a) Depth profiles of magnetic susceptibility (x) (10^{-8} m^3/kg), for the nearshore stations 2, 56 and 116. b) Depth profiles of SIRM (μA m^2/kg), for the same stations.

the atmosphere over the last few decades as a consequence of rapidly increasing combustion of fossil fuels. This increase becomes more obvious at offshore sites, due to the lower contribution of magnetically "inert" riverine material there. The mechanism whereby these particles are deposited could be combined with the deposition of Saharan dust.

3) At offshore sites, which also have relatively low bioturbation, diagenetic changes within the sediments lead to the "consumption" of ferrimagnetic material downcore through the reduction and solubilization of magnetically active iron oxides. This is a rather rare phenomenon observed in haemipelagic and lacustrine sediments (Karlin and Levi, 1983; Anderson and Rippey, 1988). A combination of the aforementioned hypotheses cannot be excluded, although it is essential to identify the most probable major mechanisms.

The fact that on the basis of ARM readings, the percentage of fine (< 1 μm) particles increases downcore in offshore stations, in combination with the fact that the river load does not contribute to a major extent at the offshore sites (Martin *et al.*, 1989) minimizes the possibility that the river constitutes the major carrier of magnetically active mate-

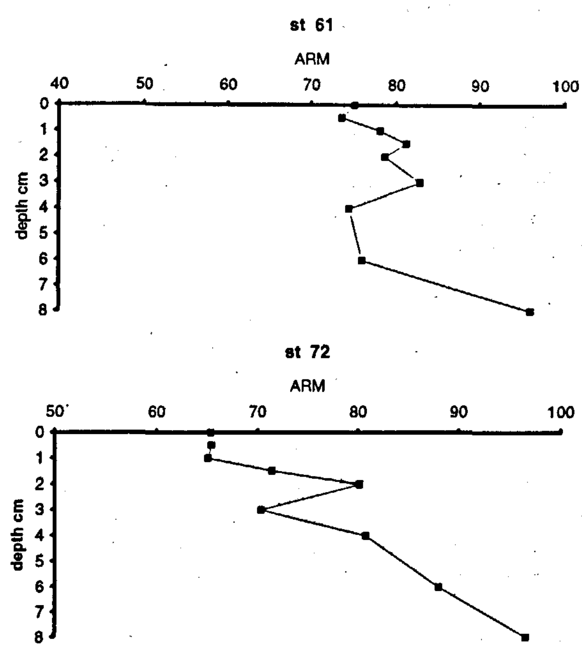

Figure 4

ARM (μA m^2/kg) vertical distribution for stations 61 and 72.

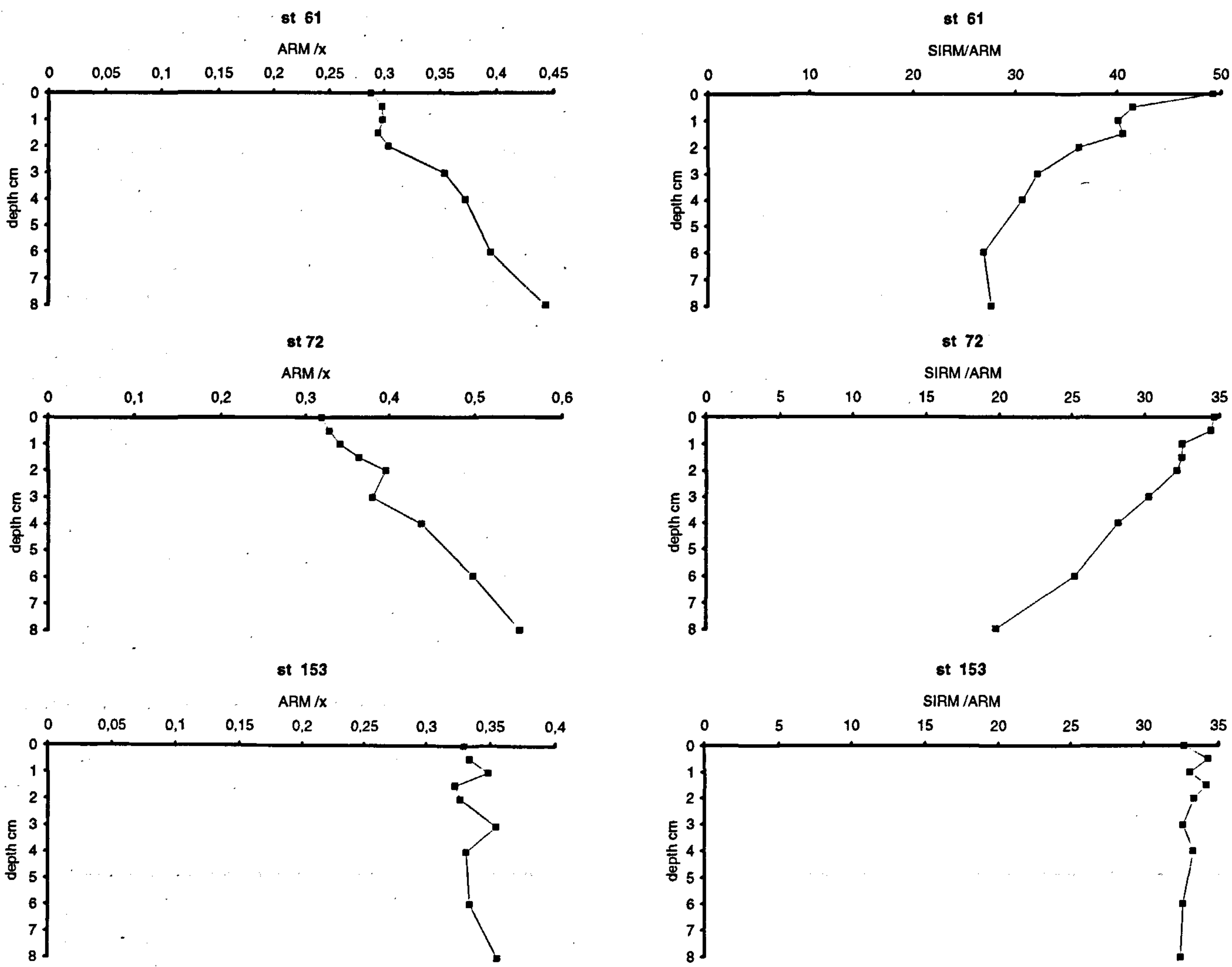

Figure 5

a) ARM/x (kA/m) ratio for the sediment cores collected at the deep parts of the gulf, stations 61 and 72, and near the Rhone mouth, station 153. b) SIRM/ARM ratio for the same sediment cores.

rial of natural or anthropogenic origin in areas far removed from the river mouth. The picture in the delta area and in a zone immediately affected by the river plume could be different. The importance of particle sorting as an enrichment mechanism for magnetically active material in the surface layers of the sediment column is not excluded, but cannot provide a satisfactory explanation of the important peaks observed offshore.

From the magnetic parameters alone, it is virtually impossible to exclude one or other of the two remaining hypotheses. However, the combination of the magnetic with the available chemical data could provide an answer. Oxygen penetration was measured by Helder (1989) in the deep, offshore sediments of the area and it was found that the penetration depth is larger than 30 cm. Furthermore, the dissolved manganese concentrations in the pore waters of the sediments of station 15 reported by van Hoogstraten and Nolting (1991), and of stations 61 and 72 reported by Nolting (1989), have relatively low values (< 5 μM) and show no increase downcore, thereby, excluding the reduction and diagenetic dissolution of iron oxides at these sites. Similarly, the Fe/Al ratios given by Nolting and van Hoogstraten (1992) indicate clearly that any diagenetic processes in the sediments are restricted to the vicinity of the river mouth and do not affect the offshore sites. Therefore we should conclude that the peaks observed in the surface samples studied are principally due to the atmospheric transport of particles into the deep marine environment of the NW Mediterranean. In fact this conclusion should not be considered as "exotic", since atmospheric inputs to the deep sediments appear to prevail (4.10^6 $t.y^{-1}$) over those of terrestrial and riverine origin [1.10^6 $t.y^{-1}$ (Martin *et al.*, 1989)].

That anthropogenic particles deriving from the European coast find their way by atmospheric long-range transport into the deep-sea sediments of the NW Mediterranean is supported by the findings of Nolting and Helder (1991) and Lipiatou and Saliot (1990). These authors, working independently, have found high concentrations of lead, zinc and PAH's at the same depth intervals and in the same samples where extremely high x values have been recorded

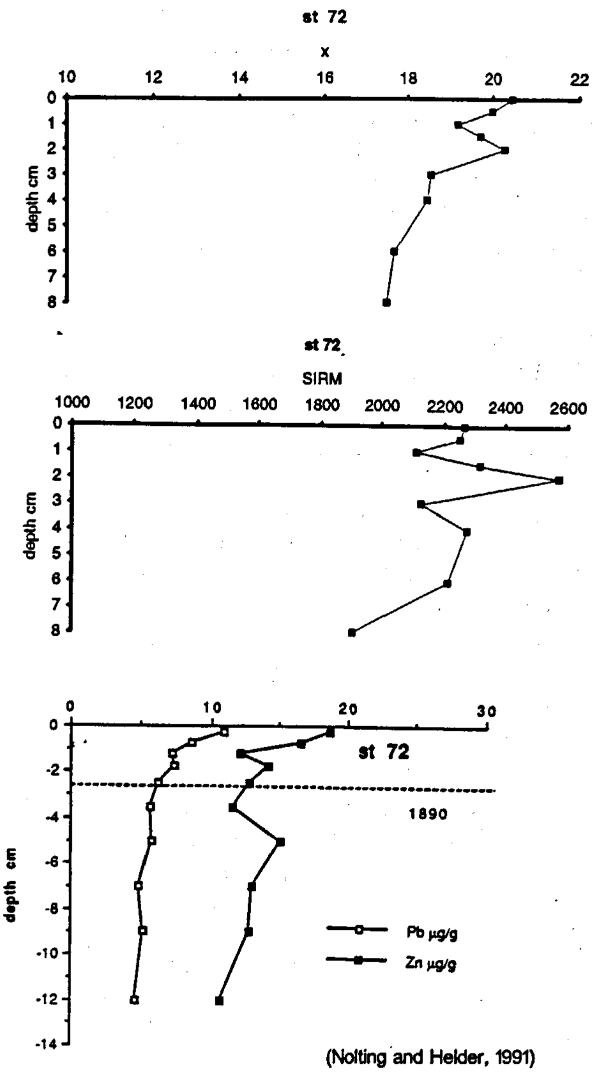

Figure 6

Comparison of magnetic with chemical data (Nolting and Helder, 1991) for station 72 at the outer part of the gulf. It is interesting to note the similarity of the profiles of x ($10^{-8}m^3/kg$) and SIRM (µA m^2/kg) with lead and zinc concentrations (µg/g).

[Scoullos *et al.*, 1990 (Fig. 6)], providing powerful evidence of a coexistence of PAHs, lead and ferrimagnetic cubic iron oxides (deriving most probably from the combustion of fossil fuels) in atmospheric aerosol particles of < 1 µm. The preferential association of heavy metals with such aerosol particles in the Mediterranean atmosphere has been demonstrated by Seghaier (1984), Remoundaki *et al.* (1991), *etc.*

A further question concerns the mechanism whereby such small particles are deposited at great depths, sometimes so rapidly that they retain their characteristics and even the PAHs which they have collected by adsorption. From the data in our possession, we believe that dry deposition and particularly the particle fluxes known to occur, mainly during the Saharan dust episodes, could play an important role in facilitating the deposition and coprecipitation of fine anthropogenic particles from the atmosphere and the water column to the sediments. The coexistence in the same samples of high x values which cannot be attributed to the natural mineral sources of the area, with high x_{fd}% values indicative of eroded surface soils such as those of Sahara, is in conformity with the aforementioned hypothesis.

Magnetic characterization of atmospheric dusts of N. African origin (Prospero *et al.*, 1981 and Parkin *et al.*, 1970), has been performed by Oldfield *et al.* (1985 *b*) and Hunt (1986) on samples collected from Barbados and the Northern Atlantic. Two aerosol samples collected in Corsica have been studied by Scoullos and Zeri (1992) and provide the characteristics of atmospheric fluxes of Saharan origin, possibly influenced by anthropogenic inputs. A comparison between the magnetic properties of these samples and those of surface sediments of the NW Mediterranean is given in Table 2. It is clear from this table that the values of x, SIRM and ARM for the deep, offshore stations are comparable to those of the Saharan dusts collected in Barbados, the Northern Atlantic "B" and Corsica, whereas the nearshore sediments of NW Mediterranean have much lower values, due to the higher contribution of magnetically inert terrestrial and fluvial particles in the area immediately affected by the Rhône plume. Atmospheric dusts originating elsewhere (*e. g.* Sea of Japan, Northern Atlantic dusts "A") and influenced by urban/industrial activities, included in the table for comparison reasons, have significantly higher magnetic signals. This means that their slightest presence in an offshore site could provide a relatively strong signal, and explains the high values (x > 20 and SIRM > 2 000) at stations 3, 5, 14, 15, 18, 61, 72, 96 and 154. In a few of these stations, *i. e.* 96, 61 and 18, the high x and SIRM values are coupled with relatively lower x_{fd}% values ($2.68 < x_{fd}\% < 4.3$) in the upper layer (Fig. 7), but all other stations have high x_{fd}% values (> 8 %). These features provide strong

Table 2

Comparison of magnetic parameters of atmospheric particles with those of the surface sediments of the Gulf of Lions.

Sample	x$^{\alpha}$	SIRM$^{\beta}$	ARM$^{\beta}$	
Barbados dusts*	51-83	4 000-7 000	40-80	(Saharan)
N. Atlantic dusts*				
A	130-2 600	21 000-166 000	100-600	
B	67-83	5 000-10 000	67-300	(Saharan)
Corsica dusts+				
A	79	4 835	119	(Saharan)
B	131	8 706	449	(Saharan)
Sea of Japan dusts*	> 500	> 50 000	> 100	
Gulf of Lions surface sediments				
Nearshore	14-20	1 400-2 000	37-65	
Offshore	20-50	2 000-6 000	65-90	

* *from Hunt (1986) and Oldfield* et al. *(1985* b*).*
\+ *from Scoullos and Zeri (1992).*
$^{\alpha}$ *In $10^{-8}m^3/kg$.*
$^{\beta}$ *In µA m^3/kg.*

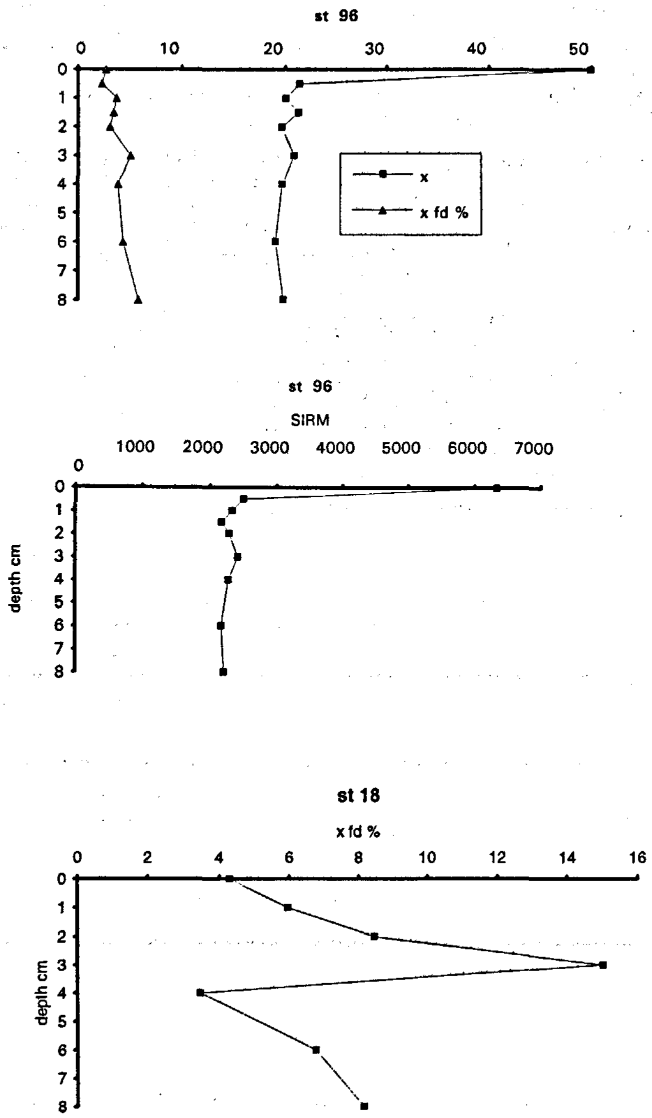

Figure 7

At stations 96 and 18 the increased x ($10^{-8} m^3/kg$) and SIRM ($\mu A\ m^2/Kg$) values at the surface layer are coupled with relatively low x_{fd}% values. See *also Figure 2.*

indications that particles of Saharan origin may act as carriers of small quantities of magnetically highly active matter of anthropogenic origin existing in the atmosphere or already deposited in the surface layers of the water column. The fact that Saharan dust particles were not detected in the area by Wegrzynek *et al.* (1992) employing electron probe X-ray micro analysis (EPXMA) could be attributed to the second explanation offered by the authors, according to which the bulk of the Saharan dust particles are smaller than about 0.5 μm. Our own measurements indicated clearly that the Saharan dusts have high concentrations of particles in the range of < 0.03-0.4 μm.

The increase of x, SIRM and SIRM/ARM in the uppermost layer of the sediment column, corresponding to a time span of less than 100 years, which is paralleled by an increase in heavy metals and other pollutants, provides an indication of the significance of the anthropogenic contribution in offshore Mediterranean sediments, whether direct (greater pollution), or indirect (retention of magnetically inert, fluvial particles) due to the construction of dams.

A definite answer concerning the role of the Saharan dust as a potential carrier of anthropogenic particles and the relative importance of the various sources of particulate inputs in the Gulf of Lions requires further research on magnetic and chemical parameters, this is already under way.

Aknowledgements

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