

Fluxes Trace elements Deep ocean sediments Tropical Atlantic Ocean

Flux Éléments-traces Sédiments profonds Océan Atlantique tropical

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ABSTRACT Fluxes of 16 major and trace elements into a deep-ocean sediment trap in the Western tropical Atlantic are reported. Fluxes are temporally variable and this may be the result of the influence of South American rivers on this area. Despite this variability, fluxes of aluminium and other trace elements associated with abiogenic and biogenic material, correlate with organic carbon, suggesting that biogenic and abiogenic material are transported by the same mechanisms as in central ocean regions. A small compositional difference is seen between the samples collected during periods of high and low total flux. This appears to result from dilution of three of the main components (clay, calcium carbonate and organic matter) by silica. Lead fluxes are higher than expected. This may be a result of enhanced removal of lead due to boundary scavenging effects.

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RÉSUMÉ

Variabilité des flux de quelques éléments dans l'ouest de l'Océan Atlantique tropical

Les flux de seize constituants majeurs ou à l'état de traces sont étudiés à l'aide d'un piège à sédiments profonds dans l'ouest de l'Océan Atlantique tropical. Ces flux varient avec le temps, probablement sous l'effet des fleuves de l'Amérique du Sud. Malgré la variabilité, une corrélation existe entre la concentration en carbone organique et les flux d'aluminium et d'autres éléments à l'état de traces associés à des matières biogéniques et abiogéniques; le transport de ces matières serait donc régi par les mêmes mécanismes qu'au centre de l'océan. Les échantillons présentent une petite différence de composition selon qu'ils ont été prélevés pendant une période de flux total élevé ou faible. La différence est attribuée à la dilution de trois des constituants majeurs (argile, carbonate de calcium et matière organique) par la silice. Les flux de plomb sont plus élevés que prévu, peut-être par suite d'une extraction accrue du plomb à proximité des marges océaniques.

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INTRODUCTION

In recent years the use of deep-sea sediment traps has considerably advanced our understanding of deep-sea sedimentation processes (e.g. Honjo, 1978; 1980; Brewer et al., 1980; Deuser and Ross, 1980; Wefer et al., 1982; Honjo, 1984; Martin and Knauer, 1985; Deuser, 1986; Honjo et al., 1988). These processes can in some cases be related to variation in the productivity of the overlying surface waters (Deuser, 1986). Efficient mechanisms clearly exist to transport the material to deep water. Among them is the packaging of particulate matter in surface water by zooplankton into faecal pellets which can fall rapidly to the sea floor (McCave, 1975; Honjo, 1976; Deuser *et al.*, 1983; Fowler and Knauer, 1986) though the significance of this mechanism has been questioned (Pilskaln and Honjo, 1987). The sedimenting material contains abiogenic material, including clays, anthropogenically derived trace metals, and radionuclides formed within the water column (Deuser, 1984). It is apparent that the same mechanisms that remove biological material from surface water can also remove abiogenic particles from the surface and deep water (Bacon *et al.*, 1985). Investigations near ocean margins demonstrate the increased complexity of sedimentation processes in these areas (Martin and Knauer, 1985). Here we report the results of chemical analyses of sediment trap samples from a site 190 km east of Barbados and compare them with results from the Sargasso Sea (Fig. 1). A detailed comparison of the total fluxes at the two sites has already been reported (Deuser *et al.*, 1988).



Figure 1

Map showing location of sediment traps in the equatorial Atlantic and the Sargasso Sea.

METHODS

The sediment trap used in this study had the same geometry as that used in earlier studies (Honjo *et al.*, 1980; Deuser *et al.*, 1981), but was equipped with a

Table 1

Sediment trap samples collected during this study.

cup rotator for sequential collection of six samples in a pre-set sampling cycle. The trap was deployed at 13°13'N, 57°41'W, 3200 m below the surface, in an area where the water depth is 4250 m. No preservative was used in the collection cups to avoid compromising some of the subsequent analyses. We recognise that the absence of poisons may have affected the fluxes calculated from the accumulated sample weights but believe this effect to be relatively small at the ambient temperature (2.6°C) based on preliminary results of the analyses of supernatants from sediment traps in the Sargasso Sea (Jickells and Deuser, unpublished). Equipment problems unfortunately resulted in discontinuous sample coverage and uneven sampling duration over a 25 month period, beginning 11 June 1981 and ending 9 July 1983 (Tab. 1).

Sample processing was similar to that reported previously (Deuser et al., 1981) and the results reported here apply to the less-than-37-µm fraction which resulted from wet sieving of the samples through a set of stainless-steel sieves shortly after recovery. With the exception of samples 4 and 5, the $< 37 \,\mu m$ fraction constituted between 62 and 79% of the total samples (Tab. 1). Samples were analysed by instrumental neutron activation analysis (INAA methods), similar to that previously described (Deuser et al., 1981) for the elements Al, Ca, Mg, Ba, Ti, Sr, Mn, V and I, and by wet chemical methods followed by atomic absorption (AAS) or colourimetry as described by Jickells et al. (1984) for Fe, P, Co, Ni, Cu, Zn and Pb. The only difference was the inclusion of a hydrofluoric acid digestion step to ensure that the relatively high proportion of alumino-silicate material in these samples was completely dissolved.

RESULTS

Comparison of Ca and Al measured by both atomic absorption and neutron activation methods suggests good agreement and efficient chemical digestion $(Al_{INAA}/Al_{AAS} 1.12 \pm 0.04; Ca_{INAA}/Ca_{AAS} 1.07 \pm 0.09,$ n=4 for both). Results are presented in Table 2. Blanks for the whole digestion and analysis procedure were

ample	Collection dates	Duration (days)	Total dry weight (g)	Flux (mg m ⁻² d ⁻¹)	Percentages of total in $< 37 \ \mu m$ fraction
1	6/11/81-7/11/81	30	2.235	48.37	76
2	7/11/81-8/10/81	30	1.338	28.96	63
3	8/10/81-9/9/81	30	3.986	86.28	67
4	9/9/81-10/9/81	30	0.952	20.60	13
5	10/9/81-11/8/81	30	1.388	30.05	26
7	1/16/82-2/27/82	42	4.070	62.92	76
8	2/27/82-4/10/82	42	2.675	41.35	63
9	4/10/82-5/22/82	42	8.725	134.90	78
10	5/22/82-7/3/82	42	6.112	94.49	79
12	11/4/82-12/3/82	29	8.146	182.40	67
13	5/19/82-5/31/83	12.5	1.240	64.44	70
14	5/31/83-6/13/83	13	3.073	153.51	65
15	6/13/83-6/26/83	13	1,112	55.53	62
16	6/26/83-7/9/83	13	2.456	122,67	73

Table 2

Major and trace element concentrations in the $< 37 \ \mu m$ material from the sediment trap.

	Organic	Al	Ca	Fe	Mg	Ba	Ti	Sr	Mn	P	V	Co	Ni	Cu	Zn	I	РЬ
Sample	carbon (%)				(mg/	g)						_	(µg/g)			
1	4.75	52	124	23	14	0.75	2.75	1.04	350	700	75	8	40	83	196	189	133
2	4.91	58	123	23	18	.74	3.60	1.55	355	750	82	7.5	54	43	302	253	108
3	5.05	47	118	22	16	0.555	2.38	1.16	330	660	66	7.5	62	57	189	217	132
4	5.89	71	103	23	17	0.71	2.83	1.47	880	700	73	10	64	91	306	208	161
5	5.32	58	114	22	18	0.75	2.62	1.47	420	795	78	10	56	79	295	174	164
7	4.42	55	107	25.5	18	0.66	2.84	0.85	302	790	82	14	64	58	194	171	106
8	5.32	49	112	23	14	0.77	2.28	0.86	221	761	72	14	56	60	232	189	120
9	4.24	44	108	24	12	0.71	2.13	1.01	299	540	71	12	67	55	135	162	89
10	3.67	42	112	24	14	0.69	2.25	1.06	662	570	71	12	73	54	110	196	77
12	4.21	41	115	22	14	0.76	2.41	1.00	734	605	68	(28)	65	58	(500)	153	55
13	3.59	43.5	93	-	11	0.54	2.24	0.46	735	-	66	- '	-			166	_
14	2.90	45.5	97	25.5	15	0.68	2.54	0.80	585	740	76	13	60	55	140	179	100
15	2.10	43	81	-	16	0.69	2.41	0.64	727	-	72	-	-	-	-	179	
16	2.27	34	71	24	11	0.41	2.07	0.36	570	655	53	11	42	53	116	118	88

Bracketed data are suspect and excluded from subsequent correlations.

< 10% of measured concentrations for all elements, except lead (< 15%). Average daily fluxes (Tab. 3) were calculated by multiplying the fluxes for each sampling interval by the sampling interval, summing these and dividing by the total sampling time. In addition to the full digestion, two samples were also soaked in 1% (v/v) nitric acid for several days to determine an opera-

Table 3

Weighted average fluxes of major and trace elements in $<37~\mu m$ fraction (June 1981-July 1983).

Total flux	53.6	$mg m^{-2} d^{-1}$
Organic carbon	2.2	$mg m^{-2} d^{-1}$
CaCO ₃	5	$mg m^{-2} d^{-1}$
Al	2.4	$mg m^{-2} d^{-1}$
Ca	5.8	$mg m^{-2} d^{-1}$
Fe	1.3	$mg m^{-2} d^{-1}$
Mg	0.76	$mg m^{-2} d^{-1}$
Ba	0.036	$mg m^{-2} d^{-1}$
Ti	0.13	$mg m^{-2} d^{-1}$
Sr	0.051	$mg m^{-2} d^{-1}$
Mn	26	$\mu g m^{-2} d^{-1}$
P	35	$\mu g m^{-2} d^{-1}$
V	3.8	$\mu g m^{-2} d^{-1}$
Co	0.56	$\mu g m^{-2} d^{-1}$
Ni	3.4	$\mu g m^{-2} d^{-1}$
Cu	3.1	$\mu g m^{-2} d^{-1}$
Zn	7.8	$\mu g m^{-2} d^{-1}$
I	9.5	$\mu g m^{-2} d^{-1}$
Pb	5.1	$\mu g m^{-2} d^{-1}$

tionally defined labile fraction for these elements measured by AAS and colourimetry (Tab. 4). We believe this simple leaching technique should liberate trace elements bound relatively weakly to solid phases. This

Table 4

Percentage of each element that is soluble in 1% nitric acid.

	Sample Number *				
Element	10	14			
Al	22	21			
Ca	105	92			
Fe	18	18			
Mn	82	77			
P	22	24			
Co	58	54			
Ni	29	42			
Cu	46	62			
Zn	39	35			

* See Table 1.

should include elements associated with adsorbed, organic, Fe Mn oxide and calcium carbonate phases. Although not a rigorous speciation method (e. g. Tessier *et al.*, 1979) this technique has the advantage of simplicity and comparability with our previous studies (Jickells *et al.*, 1984).

DISCUSSION

There are few other data sets with which to compare the results reported here. The only other sediment trap deployment in this area was that reported by Brewer *et al.* (1980). In view of the temporal variability shown by our data we do not compare our results with the data of these authors in detail since they were based on single 98-day deployment. However, a comparison of the earlier results with those reported here shows a general similarity, though during the earlier deployment aluminium concentrations were lower and calcium concentrations higher.

Mechanisms of sedimentation

In contrast to the situation described for the Sargasso Sea (Deuser, 1986), there is little evidence of strong seasonality in the fluxes measured at our site in the tropical Atlantic, though the discontinuous nature of the sampling might have obscured it to some extent. However, based on the zonal climatology, a much damped seasonal amplitude in the equatorial Atlantic relative to the Sargasso Sea had been expected. Daily fluxes (average value 77 mg m⁻² d⁻¹) were similar to those in the Sargasso Sea (20-120 mg m⁻² d⁻¹; Deuser, 1986) and underwent considerable temporal variations (21-182 mg m⁻² d⁻¹; Tab. 1) possibly related to the passage of patches of water with varying nutrient content through the area (Deuser et al., 1988). The fluxes of organic carbon and total mass are closely correlated as are those of organic carbon and aluminium (Fig. 2, Tab. 5). We assume, as previously (Deuser et al., 1983), that aluminium is predominantly associated with clays. Such correlations have previously been interpreted in terms of biological control of the removal



Organic Carbon Flux (>37 μ m) mg/m²/day

of abiogenic material from the water column (Deuser et al., 1983).

While the mechanism of transport of the aluminosilicate debris does appear to be similar at this site and in the Sargasso Sea, the Al/organic carbon ratio is quite different, being about 2.5 times higher at this site. This indicates that the biota can cope with the higher clay concentrations in this much more dynamic environment by the same or similar removal mechanism to those described in the Sargasso Sea (Deuser et al., 1983).

Bulk composition of sedimenting material

The major components in the $< 37 \,\mu m$ fraction of the collected material can be estimated from the average Table 5

Correlations of elemental fluxes with organic carbon for $< 37 \,\mu m$ fraction.

Element	n	Correlation coefficients
Total flux (< 37 µm)	14	0.92
Al	14	0.94
Ca	14	0.97
Fe	12	0.89
Mg	14	0.91
Ba	14	0.94
Ti	14	0.91
Sr	14	0.96
Mn	14	0.73
P	12	0.86
v	14	0.92
Co	11	0.84
Ni	12	0.95
Cu	12	0.93
Zn	11	0.93
I	14	0.92
РЬ *	11	0.90

* 1 Pb point excluded.

concentrations of organic carbon, aluminium and calcium corrected for that associated with clays (Turekian and Wedepohl, 1961) in Table 2 (4, 4.5 and 25% respectively). If the aluminium content of clays is 10 then 45% of the material in the $< 37 \,\mu m$ fraction is clay and a further 25% is calcium carbonate (e.g. coccoliths). The average organic carbon content is 4%, giving an organic matter content of about 8% (Deuser et al., 1981); the remaining 22% is presumed to be silica.

The presence of silica is confirmed by SEM-EDS studies in which quartz silica particles can be readily identified and by X-ray diffraction studies which reveal a strong detrital quartz silica peak.

The clay content of the samples collected at this site is about 2.5 times that of the samples from the Sargasso Sea (Jickells et al., 1984) while the calcium carbonate content is about half of that in the Sargaso Sea. Although the calcium carbonate concentration is lower in the Barbados trap material, the flux is similar to that in the Sargasso Sea (Deuser et al., 1981). The lower concentration simply reflects dilution by the higher clay flux. The source of the high aluminium flux could be aeolian input (since this sample collection site lies down wind of North Africa), fluvial transport from the river systems of South America (Deuser et al., 1988) or lateral transport of clays within the ocean. Several estimates of atmospheric aluminium fluxes in this area are available (Prospero et al., 1987; GESAMP, 1990; Lim and Jickells, submitted). All these studies note the considerable variability of atmospheric fluxes due to the influence of Saharan dust storms. However, in all cases the estimated atmospheric fluxes are significantly less than the deep sea fluxes

reported here. This suggests that fluvial and/or lateral ocean transport play important roles, though we cannot distinguish between them on the basis of available data. Deuser *et al.* (1988) have discussed the extreme variability of total flux at this site and suggested this is related to patches separated from the Amazon and Orinoco plumes moving through this area and increasing primary productivity. We can now consider the effect this variability has on the bulk composition of the sedimenting material. Such variability is clearly small since the coefficient of variation for most elements is < 30%. The elemental flux plots (Fig. 2) do not provide such information because small variations in concentrations are overwhelmed by large changes in total flux.

Plots of Ca and Al concentrations against percentages of organic carbon (Fig. 3a, b) show that all three elements are positively correlated. Furthermore there is a clear distinction between the low flux period (sample 1-



8) when concentrations of all three components are high and the high flux period when concentrations of all three components are low. This implies dilution of the clay, calcium carbonate and organic carbon phases by a fourth component. This must be the component tentatively identified as silica. This is estimated by difference and hence is rather uncertain but it does clearly increase in the high flux periods as organic carbon declines (Fig. 3c). The origin of this silica phase is uncertain. It probably is not biogenic opal as this would be expected to be positively correlated to organic carbon. More likely it is detrital quartz carried to the area in the river plumes.

Elemental composition of sedimenting material

In a previous report (Jickells et al., 1984) we have examined trace element contamination problems for the type of sediment trap used. We believe that for most elements there is no serious problem. The only exception appears to be cadmium which appeared to be a contaminant in this and the previous study. In previous studies in the Sargasso Sea (Deuser et al., 1981; Jickells et al., 1984) we have demonstrated that the fluxes of all elements to the sediment trap are closely correlated to the organic carbon flux. The same appears to be the case for the samples from the tropical Atlantic (Tab. 5), suggesting that the biological mechanism is efficient at removing these trace elements as well as aluminium. This is not surprising for those elements, (e.g. I, Sr, Ba, Mg and Ca) which are associated with biogenic phases, but rather surprising for the other elements. The elemental ratios of Ti, Mn, V, Fe, Ni, Cu and Co to Al in the sediment trap samples are similar to those of shale and deep-sea clay (Turekian and Wedepohl, 1961) which suggests that these elements are largely contained within clay lattices, and hence their correlations only reflect the correlations of aluminium and organic carbon.

The fraction of an element in the $< 37 \,\mu\text{m}$ material which is soluble in 1% nitric acid varies from about 20% for iron and aluminium to 100% for calcium, reflecting the relative solubilities of clays and calcium carbonate. Comparing the results from this site with those from the Sargasso Sea (Jickells *et al.*, 1984), it is apparent that the order of relative solubility is rather similar in both cases though the labile proportion of the metals is much smaller at this site. This is consistent with the earlier conclusion that most of the trace elements in these samples are bound within clay lattices, at sites that are relatively resistant to attack by dilute acid.

For Pb and Zn there appears to be a modest enrichment in concentrations over crustal values (Tab. 6). In the Sargasso Sea larger enrichments were observed for these two elements and copper was also enriched (Jickells *et al.*, 1984). It is possible that a moderate enrichment of zinc over its abundance in shale could result from its involvement in biological cycling in the oceans (Bruland and Franks, 1983), but that effect should be similar at both sites. The smaller enrichment of zinc in the tropical Atlantic compared to the Sargasso Sea, which is directly downwind of large North American

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Table 6

Enrichment factors with respect to average shale (Turekian and Wedepohl, 1961) of some elements in sediment trap material collected in Sargasso Sea and tropical Atlantic oceans.

	Sargasso Sea*	Tropical Atlantic
Fe	0.85	0.90
Ti	-	0.93
Mn	5.7	1.0
V	1.4	1.0
Ca		0.96
Ni	2.9	1.6
Cu	9.6	2.3
Zn	8.4	2.7
Pb	14.8	8.4

* Sargasso Sea data from Jickells et al. (1984).

pollutant sources, could argue that the zinc enrichment in the Sargasso Sea arises from pollution. However, the average zinc flux at the two sites in excess of that associated with clays are very similar, in the Sargasso Sea $(5 \ \mu g \ m^{-2} \ d^{-1})$ and in the tropical Atlantic $(6 \ \mu g \ m^{-2} \ d^{-1})$, suggesting either that anthropogenic effects are minor or of the same magnitude in both areas.

Lead is enriched over its crustal abundance at the tropical Atlantic site though less so than in the Sargasso Sea (8-fold against 15-fold). However, if the lead fluxes at this site and Sargasso Sea site which are in excess of that associated with the clays are calculated, the excess lead flux at this site is greater than that in the Sargasso Sea (4.5 vs. $2.2 \ \mu g \ m^{-2} \ d^{-1}$). This result is unexpected since the Sargasso Sea site is downwind of major lead sources in North America, while the site under discussion is in the trade wind belt, under the influence of air masses originating over North Africa.

However, such considerations do not take account of the evidence for enhanced removal of lead-210 and other particle reactive radionuclides at ocean margins (Carpenter *et al.*, 1981; Bacon *et al.*, 1985). We suggest therefore that the higher excess lead fluxes at this site are caused by enhanced scavenging of lead from the water column due to higher suspended particulate matter concentration at this site than in central ocean areas. The higher particulate matter could arise from ocean margin processes or due to the elevated primary production at this site.

The possibility of enhanced lead removal at ocean margins is supported by the results of earlier trap deployments in this area and in the Sargasso Sea (Brewer et al., 1980) which included analyses for lead-210 in their trap material. These authors reported that in the Sargasso Sea the atmospheric lead-210 flux exceeded that into the sediment trap by a factor of about two and Jickells et al. (1987) similarly found that stable-lead fluxes from the atmosphere exceed those to a sediment trap in the Sargasso Sea. In contrast, Brewer et al. (1980) estimated that in the equatorial Atlantic the flux of lead-210 to the trap exceeded that from the atmosphere by more than 50%. Recent preliminary estimates of atmospheric lead deposition in this area of the equatorial Atlantic (Lim and Jickells, submitted) suggest atmospheric lead inputs to be an order of magnitude less than the flux to the trap. Thus for both lead-210 and stable lead there seems to be clear evidence that in the Sargasso Sea atmospheric inputs exceed sedimentation and that in this area of the equatorial Atlantic the reverse situation occurs. This pattern is consistent with the suggestion of enhanced fluxes to the sea bed in areas of higher suspended matter concentrations (Carpenter *et al.*, 1981; Bacon *et al.*, 1985).

One other element is worthy of note. Although the average manganese concentration is consistent with the deep sea clay Mn/Al ratio, and similar to the results of Brewer *et al.* (1980), the manganese concentrations in the samples are much more variable than those of any other element, and the correlation with organic carbon is rather weak. The cause of this variation is uncertain; it may reflect complex manganese geochemistry in a boundary region or possibly manganese mobilisation in the collection cups (Martin and Knauer, 1985 and references therein).

As with bulk composition, it is of interest to consider if the trace element composition of the sedimentary material changes between the high and low flux periods. By analogy to the bulk composition we can do this by plotting concentration of the trace element against percentages of organic carbon. A positive correlation indicates higher trace element concentration associated with higher organic carbon contents and hence with lower fluxes. This, as with Al and Ca, implies dilution of the trace element flux under conditions of high flux. Five elements are significantly correlated (90% level) with the percentage of organic carbon; Ba(r 0.49), Sr (r 0.75), Zn (r 0.79), I (r 0.53) and Pb (r 0.68). The weaker correlations for the other elements may imply that these elements have a significant source in the "silica" phase or it could simply reflect analytical uncertainties. Ba, Zn and I have well documented biological associations in surface waters (e.g. Bruland and Franks, 1983; Bernstein et al., 1987; Bishop, 1988; Jickells et al., 1988) but the behaviour of Pb appears to be dominated by abiogenic scavenging (Schaule and Patterson, 1983; Shen and Boyle, 1988). The strong correlation between Pb and organic carbon concentrations and Pb and Al (r 0.77) but not Pb and Ca (r 0.22) implies that the scavenging of Pb occurs onto organic carbon or clay phases, a suggestion consistent with evidence from other studies (Schaule and Patterson, 1983; Shen and Boyle, 1988).

CONCLUSION

The transport of material to the sediments in the Atlantic off the Lesser Antilles appears to be controlled by biological processes in the overlying water. We suggest that this process involves packaging of biogenic and abiogenic particulate matter into faecal pellets and other large aggregates which rapidly fall through the deep ocean. In comparison with the results from the Sargasso Sea, this site has a much higher proportion of clay and silica in the sedimenting material. This higher abiogenic loading does not seem to overwhelm the biological mechanism, but simply increases the total flux, dilutes the biogenic components and thereby produces small, but significant, changes in the bulk and trace element chemistry of the sedimenting material. The increased total particle flux may increase the scavenging of particle-reactive elements such as lead.

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