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Interstitial water iodine enrichments in sediments from the eastern Pacific

by S. J. Wakefield¹ and H. Elderfield²

ABSTRACT

The vertical distribution of iodine has been examined in interstitial waters of sediment cores from the Peru Basin, Bauer Basin, Guatemala Basin and the East Pacific Rise and the sediments have been analyzed for iodine and organic carbon. Interstitial water profiles show a strong enrichment of iodine at the sediment surface relative to overlying sea water. This enrichment is much greater than would have been expected had all the iodine originated solely from the breakdown of newly sedimented plankton. This implies recycling of iodine in the region of the water-sediment interface with most of the upward diffusing iodine being rescavenged and a small fraction escaping to the deep sea. A flux equivalent to 1–5% of the diffusive flux from interstitial waters can account for an excess iodine anomaly previously described for deep Pacific waters.

1. Introduction

Recent marine sediments can contain high concentrations of iodine (I) — up to 0.2% in some highly productive shelf areas (Price and Calvert, 1973). The iodine concentration within the sediments does not correlate with the mineralogy or major element composition but co-varies only with organic carbon (C) content (Bojanowski and Paslawska, 1970; Price *et al.*, 1970; Price and Calvert, 1973). Nevertheless the iodine contents of sediments under oxygenated waters are much higher than can be accounted for by direct, unmodified planktonic input. This has been explained in terms of the recycling of iodine during early diagenesis. It is envisaged that iodine released from sediments during diagenesis, and known to be enriched in interstitial waters (see the summary in Pedersen and Price, 1980, Table 2), is transported by diffusion across the water-sediment interface where in oxic areas it is scavenged by suspended matter.

The studies of I geochemistry have largely been confined to shelf, marginal sea, and hemipelagic deposits and little is known of the behavior of iodine in deep-sea sediments. Some limited data have been presented by Shishkina and Pavlova (1965), Bennett and Manuel (1968) and Pavlova and Shishkina (1973) and the then available results led Price and Calvert (1973) to suggest that the I/C ratio of surface pelagic

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sediments may be similar to those found in surface oxic sediments in shallower environments.

We report here some data on the distribution of I in deep-sea sediments and associated interstitial waters from the Pacific Ocean, and the behavior of iodine during shallow-burial diagenesis will be discussed. It is of additional interest that the sites sampled include those where the water column distributions of iodine species have been measured and which led to the observation that a deep-water dissolved iodine anomaly exists which might be maintained by diffusion of iodine from bottom sediments (Elderfield and Truesdale, 1980). This suggestion will be tested using the interstitial I data.

2. Materials and methods

The main part of the work was based on five gravity cores collected on R.R.S. *Shackleton* cruise 2/76 from sites on the N.W. Nazca Plate, S.E. Pacific Ocean (Table 1). The sites were chosen to represent different environments of deep-sea sedimentation in general: Peru Basin (LD1), the Bauer Basin above the carbonate compensation depth (LD2) and below it (LD4), and the East Pacific Rise flank (LD5) and crest (LD7). The lithologies, and major element compositions of the surface sediments are summarized in Wakefield (1982). Full details of the chemistry and mineralogy of the cores are given in Wakefield (1981).

Interstitial waters were removed from extruded sections of the cores on board ship at *in situ* temperature using a water-jacketed PVC-PTFE hydraulic squeezer (Kalil and Goldhaber, 1973). The expressed waters were filtered through 0.4 μm Nuclepore filters, stored, unacidified, in acid-cleaned polypropylene vials and refrigerated. The squeezed sediment cakes were stored frozen.

To supplement these samples, interstitial water samples were obtained from two cores collected in 1977 during Leg 1 of the INDOMED expedition of R.V. *Melville*. The cores (9BC27B3 and 9BC33B8) were collected from "Site H," 6°33'N 92°48'W, water depth 3520–3590 m, in the Guatemala Basin. The samples were kindly donated by Dr. G. Klinkhammer and are aliquots from a set used in a broad-based study of interstitial water chemistry (Emerson *et al.*, 1980; Klinkhammer, 1980). Details of sample extraction and handling can be obtained from these reports.

Total dissolved iodine was determined automatically using the method of Truesdale (1978). A 100 μl sample was diluted to 1 ml with iodine-free artificial sea water and analyzed using a Technicon autoanalyzer. Analytical reproducibility was very high with a coefficient of variation of 0.6%.

Sediment samples were oven dried at 50°C and finely powdered. Total iodine was determined by X-ray spectrometry on a Philips PW1212 instrument using a W anode, a LiF 2200 diffracting crystal and a scintillation detector, and operating at 80 kV and 18 mA. Calibration was made using synthetic standards of potassium iodide spiked into clay (kaolinite) and carbonate (Analar CaCO_3) matrix. Tests showed no

Table 1. R.R.S. *Shackleton* core locations and characteristics.

	Station		SH 1582/LD2		SH 1587/LD4		SH 1588/LD5		SH 1589/LD7	
	No/	Core								
Position	11°28'S		8°40'S	9°27'S	7°51'S	7°00'S				
	85°52'W		101°29'W	103°18'W	105°30'W	107°02'W				
Water depth (m)	4443		4150	5293	3660	3155				
Core Length (cm)	66		130	132	186	206				
Chemistry of surface sediment										
I (ppm)	25		19	24	13	20				
C (%)	0.6		0.46	0.73	0.28	0.31				
I/C × 10 ⁻⁴ (weight)	42		41	33	46	65				
(molar)*	3.9		3.9	3.1	4.4	6.1				

*All I/C ratios discussed in text are molar unless otherwise stated.

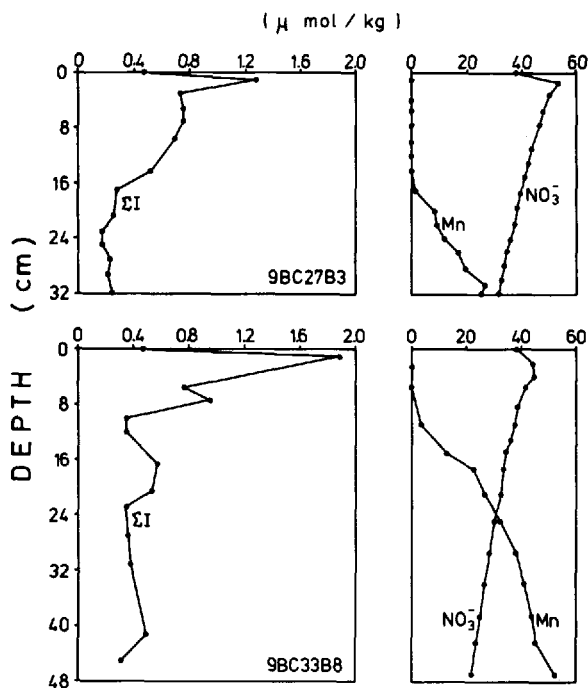


Figure 1. Distributions of total dissolved iodine (ΣI), manganese and nitrate in INDOMED cores.

systematic variations in I content for clay/carbonate mixtures over the full compositional range. The analytical precision (coefficient of variation) was better than 20% at both the 20 ppm and 10 ppm levels. A test of relative accuracy was made by analyzing twelve samples by the method of Fuge *et al.* (1978); the results showed excellent agreement ($r = 0.93$) with the XRF method.

Organic carbon contents were determined by a method similar to that used by Pedersen and Price (1980) whereby the samples are leached with acid to eliminate CaCO_3 , ensuring that any acid-soluble organic carbon is retained with the decalcified solids for analysis. The leached and dried sediment was then analyzed on a Leco carbon analyzer using standard conditions (see also Lee, 1980). Standardization was achieved using Leco metallic standards and by spiking sediments with glucose. All samples were run in duplicate and the analyses accepted if the two results were within 10% of the mean; otherwise, additional replicates were run. Analytical precision was $\pm 9\%$ (2σ) at the 0.5% C level.

3. Results and discussion

The results of the interstitial water analyses for total iodine from the INDOMED 1 and *Shackleton* cores are illustrated in Figures 1 and 2. Figure 1 also shows the concentration of nitrate and manganese reported by Emerson *et al.* (1980).

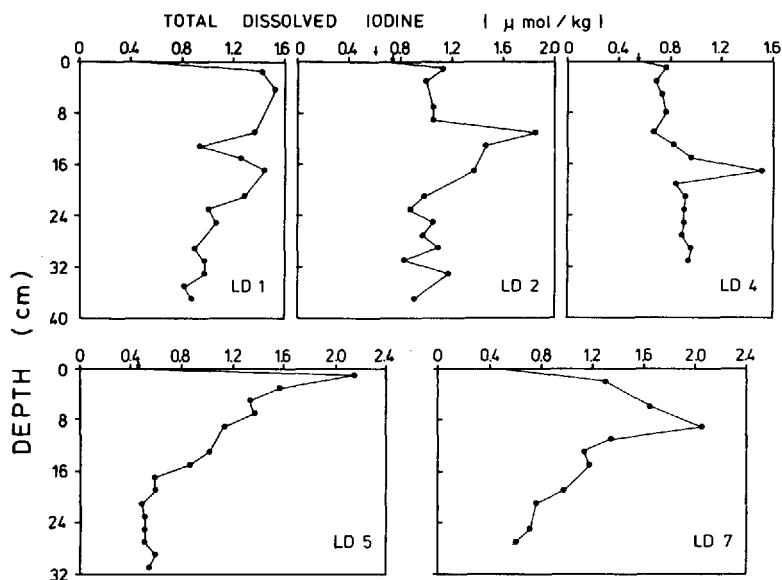


Figure 2. Distributions of total dissolved iodine (ΣI) in *Shackleton* cores. Arrows indicate supernatant water values where measured.

The total iodine contents of Pacific deep waters normally fall within the range 0.47–0.49 $\mu\text{mol}/\text{kg}$ (Elderfield and Truesdale, 1980). The interstitial water profiles shown here all are characterized by an interstitial iodine maximum in the uppermost sample analyzed, i.e., there is a significant enrichment in iodine at the sediment surface relative to the overlying seawater, by a factor of about 1.7 to 4.7. Below this the concentrations either decrease sharply with depth and then remain constant (cores LD5, LD7, 9BC27, 9BC33) or decrease less significantly and more gradually (LD1, LD2, LD4). This overall pattern is different from that in organic-rich sediments which are usually characterized by a reasonably regular increase in iodine concentration with depth (e.g., Pedersen and Price, 1980; Ullman and Aller, 1980; Elderfield *et al.*, 1981; Ullman and Aller, 1983).

The enrichment of iodine in interstitial waters from the uppermost section of the sediment column is indicative of diagenetic recycling of iodine at the sediment-water interface. Furthermore, given a number of reasonable assumptions outlined below, quantification of the iodine enrichment compared with that of nitrate (Fig. 1) shows that I release is in excess of planktonic input. The reasoning is as follows. We assume that decomposing organic matter is the only source of nitrogen species and iodine to the interstitial waters and that all the organic nitrogen is oxidized to NO_3^- during O_2 reduction. (Emerson *et al.* (1980) report small (1–2 $\mu\text{mol}/\text{kg}$) concentrations of NH_4^+ indicating incomplete oxidation of organic N but these values are substantially less than the concomitant increase in nitrate (5–14 $\mu\text{mol}/\text{kg}$) and so do not alter our conclusions.) Provided that the total iodine is dominantly iodide (as opposed to iodate)

then the diffusion coefficients of the iodine species and nitrate are virtually identical (Li and Gregory, 1974). Such an assumption of iodine speciation is supported by recent studies (H. Kennedy, *pers. comm.*). Also, given the nature of the chemical species under consideration, it is reasonable to neglect adsorption in these slowly accumulating deep-sea sediments (*cf.* Ullman and Aller, 1983; Bender and Heggie, 1984). Finally, we need to define a C/N ratio for the decomposing organic matter. Although the "Redfield ratio" of 106:16 (6.6) has been shown to be lower than when decomposing organic matter stoichiometry is calculated from pore water profiles or sediment chemistry considerations (e.g., Grundmanis and Murray (1982) give values of 8.1–13.7) we use the traditional stoichiometry of 106:16 here since our conclusions are not affected by even a 50% error in this figure. Thus, given the above (which may combine to produce an uncertainty of $\pm 15\%$), the ratios of the sea water excesses of total iodine and nitrate, $\Delta I/\Delta\text{NO}_3^-$, in the near surface interstitial waters when divided by a C/N ratio of 106:16 lead to I/C molar regeneration ratios in the sediment of about 80×10^{-4} for core 9BC27B3 and 340×10^{-4} for core 9BC33B8, vastly in excess of the I/C ratio of $\approx 1 \times 10^{-4}$ in planktonic material and deduced from stoichiometric relationships in sea waters (Elderfield and Truesdale, 1980). Moreover these regeneration ratios are significantly greater than the ratio of 16×10^{-4} calculated by Ullman and Aller (1983) using SO_4^- reduction and I production rates from incubation experiments, and measured I/C ratios found in debris caught in sediment traps. (Spencer *et al.* (1978) report I/C ratios of 3.1×10^{-4} for "green" fecal pellets and 1.6×10^{-4} for "red" fecal pellets. Brewer *et al.* (1980) noted a strong correlation between iodine and organic matter in their traps which converts to an I/C ratio of 3.4×10^{-4} .)

Hence despite the uncertainty introduced as a consequence of the assumptions made above there is no doubt that the enrichment of iodine in surface pore waters is much greater than can be expected from the decomposition of typical marine organic matter. It is of further interest that I/C sediment regeneration ratios also are higher than the highest I/C ratios recorded in marine sediments of $\approx 38 \times 10^{-4}$ (see Pedersen and Price, 1980). Overall, this suggests that whereas a significant amount of the iodine released from normal sedimentary organic matter is scavenged at or near the water-sediment interface, some proportion may be lost to the overlying water column (see later discussion).

Surface sediment values for I and C are given in Table 1 along with I/C ratios expressed on both a weight and atom basis. Depth profiles for I and C are shown in Figure 3 and Figure 4 respectively. Both elements are enriched in the surface sediments of all cores relative to the underlying samples.

The majority of the sediment samples have I contents in the range $5\text{--}30 \mu\text{gg}^{-1}$. These values are similar to those previously reported for deep-sea sediments (Shishkina and Pavlova, 1965; Bennett and Manuel, 1968; Pavlova and Shishkina, 1973) and contrast with the normally high I contents reported for nearshore and hemipelagic areas (e.g.,

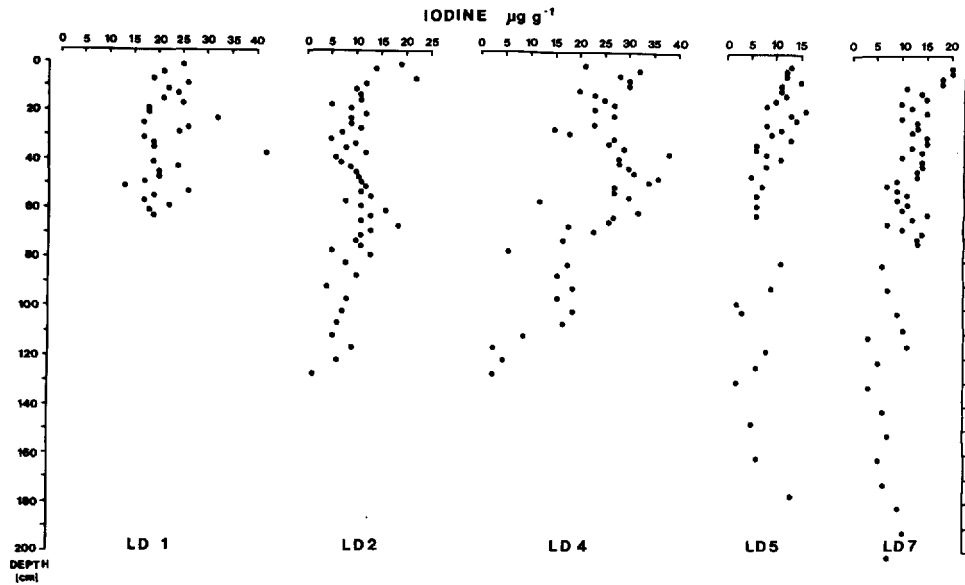


Figure 3. Vertical profiles of iodine in *Shackleton* cores. Results are on a total sediment basis.

Price and Calvert, 1973; Malcolm and Price, 1984). Organic carbon contents are low (0.08%–0.73%, mean of 0.27%, $n = 206$) and typical of the deep-sea environment (e.g., Premuzic *et al.*, 1982; Emerson *et al.*, 1985). Four of the five cores display a rapid decrease in organic carbon content within the top 10 cm tending to near constant values at depth. This diagenetic loss of sedimented carbon amounts to 35%–67% of the surface concentration. These figures are comparable to those found by Muller and Mangini (1980) in the northern Pacific and Reimers and Suess (1983) in the vicinity of the Pacific-Antarctic Ridge.

Price and Calvert (1973), in their work on the surface sediments of the S.W. African continental shelf, have shown that the I/C ratios range from 17 to 253 ($\times 10^{-4}$) on a weight basis ($1.6 \times 23.9 (\times 10^{-4})$ on an atom basis). The lowest values are found in the organic rich nearshore sediments underlying oxygen depleted waters whereas the higher ratios are found in the more oxidizing environment of the outer shelf. Additionally, atomic ratios as high as 36×10^{-4} have been recorded by Price *et al.* (1970) in the Barents Sea and Pedersen and Price (1980) in the Panama Basin. On the basis of the very limited data then available in the literature, Price and Calvert (1973) suggested that the I/C ratios of surface pelagic sediments may be similar to those found in surface oxidized sediments from shallower environments. This contention is not borne out by the present work on sediments from the deep sea. Indeed, the I/C surface sediment ratios for these southeast Pacific samples are closer to values for anoxic sediments than to the high values recorded for oxic sediments from shelf and

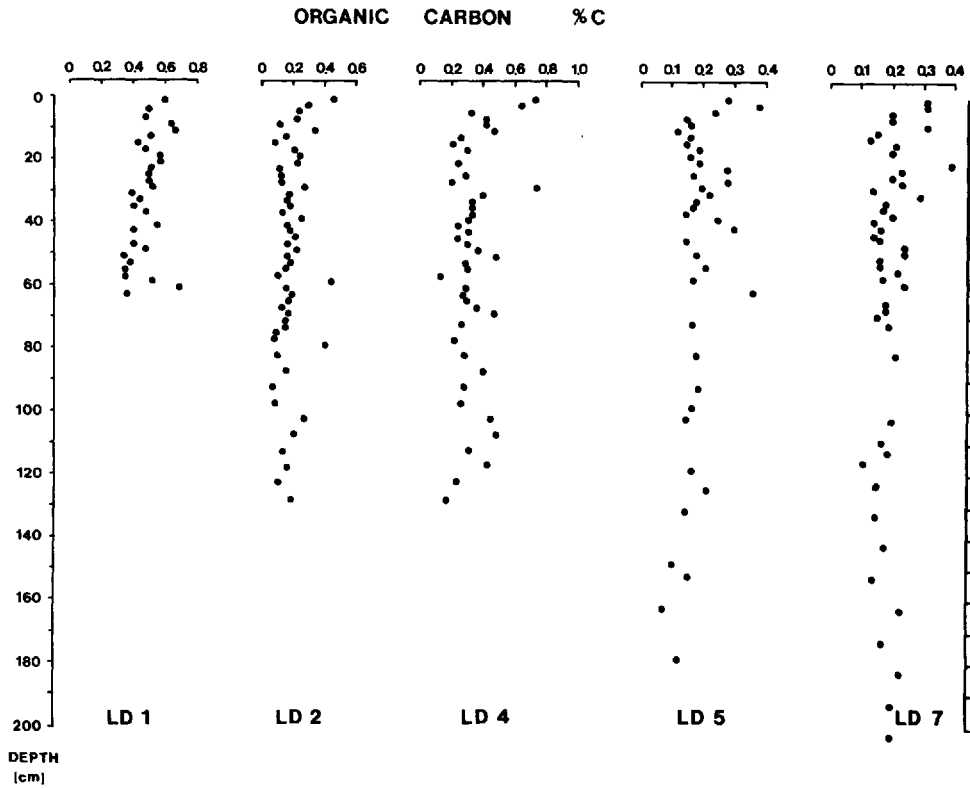


Figure 4. Vertical profiles of organic carbon in *Shackleton* cores. Results are on a total sediment basis.

hemipelagic environments and also are comparable to the value found in Long Island Sound (4.7×10^{-4} , Ullman and Aller, 1983).

It seems clear from the earlier discussion on iodine in interstitial waters that iodine is released by the decomposition of organic matter during very early diagenesis and, in oxic sediments, is subsequently scavenged. This leads to the trapping of iodine in association with sediment particles with high I/C ratios and its continual recycling at or close to the water-sediment interface. Clearly this process is more prevalent nearshore where there is an abundance of reactive organic matter than in the deep sea where the organic content of the sediments is much less. This quasi-closed cycle for I at the sediment-water interface underlying oxygenated waters has also been recognized by Ullman and Aller (1980). Hence the surface sediment I/C ratios reflect the extent of iodine scavenging that has taken place subsequent to sedimentation. Since this ill-defined chemical mechanism would appear to require oxidizing conditions (Shaw, 1959; Harvey, 1980; Malcolm and Price, 1984) the ratio found is dependent upon these two parameters, namely the conditions at the water-sediment interface and the

availability of scavenging material. Thus nearshore sediments underlying oxygenated waters have the highest ratios (optimal scavenging conditions), deep-sea sediments have distinctly lower ratios (still an oxidizing environment but significantly less reactive organic material) and sediments underlying anoxic waters reflect only primary I/C input (planktonic ratio) since any further scavenging (I enrichment) is inhibited.

The fact that all seven profiles of this study show iodine enrichment in their surface interstitial water sample relative to the overlying sea water implies a substantial flux of iodine from the sediment column out into this water. This possibility was recognized earlier by Elderfield and Truesdale (1980). From measurements of the vertical distributions of iodine in the S.E. Pacific water column overlying the *Shackleton* sites they identified an excess of iodine in deep ocean water for which diffusion from interstitial waters was suggested as one possible source. From the magnitude of the anomaly the required benthic flux was estimated at $\approx 2 \text{ nmol cm}^{-2} \text{ yr}^{-1}$. It is now possible to compare this figure with the diffusive I flux implied by the data presented in Figures 1 and 2. The simplest approach is to adopt Fick's first law and multiply the near surface iodine interstitial water concentration gradient by a transport term, effectively a modified diffusion coefficient (see, for example, Berner, 1980). Thus:

$$J = \frac{-\phi D \, dc}{\theta^2 \, dx}$$

where J = flux in $\text{nmol cm}^{-2} \text{ yr}^{-1}$; D = diffusion coefficient of iodide in sea water at 0°C ($10.3 \times 10^{-6} \text{ cm}^2 \text{ s}^{-1}$, Li and Gregory, 1974); ϕ = porosity (0.86); θ = tortuosity ($\theta^2 = 1.8$, Li and Gregory, 1974) and dc/dx = concentration gradient (nmol cm^{-4}). The concentration gradients measured in this study range from $0.30 \text{ nmol cm}^{-4}$ (LD4) to $1.68 \text{ nmol cm}^{-4}$ (LD5). Hence the calculated diffusive fluxes range from $41\text{--}200 \text{ nmol cm}^{-2} \text{ yr}^{-1}$ (mean of the seven sites: $104 \text{ nmol cm}^{-2} \text{ yr}^{-1}$). Thus even allowing for the simplicity of approach it is clear that only about 1–5% of this diffusive flux is required to account for the deep Pacific iodine excess. As a corollary, the absence of a much larger deep water excess adds further support to the scavenging mechanism, suggesting that more than 90% of the upward diffusing iodine is trapped at or near the water-sediment interface. Such a scenario has also been postulated by Ullman and Aller (1980) who showed that in a nearshore environment fluxes calculated from either pore water gradients or production rate estimates were higher than those directly measured in incubated box cores. Additionally a diffusive input of iodine from the sediment pile has also been inferred by Chapman (1983) from hydrographic data within the Benguela Current upwelling system.

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