The Impact of Industrial Discharges on Metal Levels in Biota of the West Cumbrian Coast - 1990.

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Abstract

A suite of intertidal organisms (*Fucus vesiculosus*, *Littorina littorea*, *Nucella lapillus*, *Patella vulgata*, *Mytilus edulis*) have been used to determine the influence of industrial discharges on metal levels in biota from the Cumbrian Coast between Seascale and the Solway Firth. Sediments and infauna (*Scrobicularia plana & Nereis diversicolor*) were also sampled at a number of estuarine and harbour sites.

The most impacted shoreline was that between Whitehaven and St. Bees where exceptionally high levels of Cd, Cu, Zn, Ni, & Cr occur in tissues. Acute effects are restricted to a relatively small area; arguably however, chronic contamination for several elements is detectable over the entire survey area.

Localised inputs are indicated at other sites (eg. Harrington, [Pb]; Workington & Maryport, [Fe & Mn]), though often factors other than environmental levels can explain the relatively enhanced bioavailability at these sites.

Contents

Summary

Since there is no single species which fulfils the requirements of a "universal indicator" of metal contamination, it has been necessary to select a variety of organisms in order to assess the impact of industrial discharges along the Cumbrian coastline. Brown seaweeds (predominantly *Fucus vesiculosus*), gastropod molluscs (*Littorina littorea*, *Nucella lapillus*, *Patella vulgata*) and mussels (*Mytilus edulis*) were collected at coastal sites from Seascale northwards to Skinburness at the mouth of the Solway Firth (mainly rocky shores). In addition sediments, polychaetes (*Nereis diversicolor*) and deposit-feeding clams (*Scrobicularia plana*) were sampled at five locations within this range in order to evaluate contamination in estuarine/harbour environments.

Several important sources of metals are indicated from metal profiles in the "rocky shore" species, and for Cd,Cu,Zn,Ni and Cr at least, the most significant is the outfall from the 'Marchon' factory at Whitehaven. This plant discharges metal rich wastes, generated in the production of sodium tripolyphosphate from phosphate ore, onto the shore at Barrowmouth in Saltom Bay : For each of the above metals, previously recorded UK maximum tissue concentrations are exceeded in at least one of the species collected along this stretch of coastline. In the most notable example, Cr concentrations in *Littorina* from sites nearest to the outfall were found to be some 20 times higher than previous upper limits.

It is likely that other metals (Ag,As,Hg?) are discharged at this site though bioavailability may often appear higher some distance away due to modification/ inhibition of uptake in the presence of multiple contaminants. The presence of sewage, for example, which is also discharged into Saltom Bay, is thought to enhance the bioavailability of Ag: in contrast high levels of phosphate (also present in the Barrowmouth outfall) suppress the uptake of chemically similar arsenate. Interactions between other metals also seems likely (Cd-Zn, Ag-Cu, As-Fe).

For rocky shore species Fe and Mn bioavailability is highest in the Workington area, as is Sn (probably as tributyltin). Concentrations of a number of other metals appear to be elevated here, though not to the same extent as observed in Saltom Bay. Lead is unusual since data for several species indicate a source of the element at Harrington. The presence of high lead levels in *Fucus*, relative to the invertebrates sampled suggests dissolved (alkyllead?) species may be significant.

The impact of the Marchon factory discharge, in terms of enhanced metal burdens in tissues, is predominantly restricted to the shoreline some 5Km either side of the outfall. Arguably, however, the influence of this discharge may be detected over much larger distances, at least for the more soluble metals. For example, the minimum Cd concentrations in Cumbrian biota, usually those at the extremes of the current survey (Skinburness or Seascale), are still between 3 and 11 times higher than minimum reported values for the UK. It is possible therefore that, despite considerable dilution, the influence of the metal-rich effluent at Barrowmouth may be detectable over a distance of 50Km or more as a result of residual currents and wind generated surface water movements which may hold contaminated water close to the shoreline. Depending on the species, Ag, Cu, Cr and Zn may also be enhanced at the cleanest of Cumbrian sites (average enrichment relative to UK minimum concentrations=8, 3, 7 and 2 respectively).

For other metals (Fe,Hg,As), the least contaminated Cumbrian sites represent baseline values for the UK, highlighting the extremes in metal contamination found along this stretch of coastline. It is interesting to note, however, that Hg values at the southern limit of the present survey are

somewhat higher than the baseline values in the north -possibly indicating the influence of the more Hg-enriched waters of Liverpool Bay.

Metal concentrations in estuarine and harbour sediments are not exceptionally high, other than at Whitehaven (for Cd) and Maryport (for Mn), and this is generally reflected in the moderate tissue burdens determined for sediment dwelling organisms. Occasionally, however, factors other than sediment metal concentrations may influence body burdens, such as anoxia (increases bioavailability of Cu in clams), or high levels of competing ligands (Fe oxyhydroxide and organic coatings can reduce the uptake of As and Hg respectively). Since estuarine sampling did not include sites in the immediate vicinity of the Marchon outfall, it is perhaps not surprising that levels of metal accumulation in sediment-dwellers do not approach the exceptional concentrations found in some rocky shore species. Nevertheless it is noteworthy that baseline concentrations of Ag,Cd,Cu,Cr and Pb in clams and worms, like those in coastal organisms, are often higher than minimum values reported elsewhere in the UK, though the reasons for this enhancement are perhaps more difficult to attribute to specific sources.

Based on observations of metal bioavailability in resident flora and fauna, the most metal-impacted part of the Cumbrian coastline is clearly that section between Whitehaven harbour and St Bees Head. Biological effects are also most noticeable here, particularly over a distance of approximately 1km either side of the Barrowmouth outfall. Of the species studied, Fucoid seaweeds, gastropod molluscs and mussels have differential tolerance to the effluent discharged here. Although it is by no means certain that metals are the cause of these effects some contribution seems inevitable in view of the high concentrations measured.

Introduction

In June 1990 samples of biota from the littoral zone of the West Cumbrian Coast were collected for metal determination. The aims of this project were:

i) to establish baseline data about metal levels in selected areas of the North West coastline.

ii) to provide the necessary information as a precursor for evaluating the impact of metal emissions from certain discharges. (Essential to any review of consent conditions).

iii) to include some comparison of metal levels in West Cumbria with data for other estuaries in the UK.

Study Area

Sixteen sites were visited during the survey (Table 1., Fig. 1) covering the coastline between Seascale and Skinburness (Outer Solway). The majority of sites were, however, concentrated between St. Bee's and Workington since the major emissions of metals were thought to be situated along this part of the coastline.

Sampling

Unfiltered water samples were collected for analysis at several sites. However, since the object of this research was to judge the impact of industrial discharges on biota, the number of water samples was limited to 8.

The majority of intertidal sites visited provided substrates for typical "rocky shore" communities and, where present, representative dominant species were collected. These included macroalgae, *Fucus* spp.; mussels, *Mytilus edulis*; limpets, *Patella vulgata*; winkles, *Littorina littorea*; and dogwhelks, *Nucella lapillus*. As indicated in Table 1. and described below, not all species were found at some locations. At five sites it was possible to collect fine sediments and representative members of estuarine infaunal communities *Nereis diversicolor* and *Scrobicularia plana*.

Previous research at PML has demonstrated that it is necessary to choose such a wide selection of biological indicators, since no single species is likely to be suitable for all metals. The relative merits of the species listed above have, for example, been discussed by Bryan *et al* 1985.

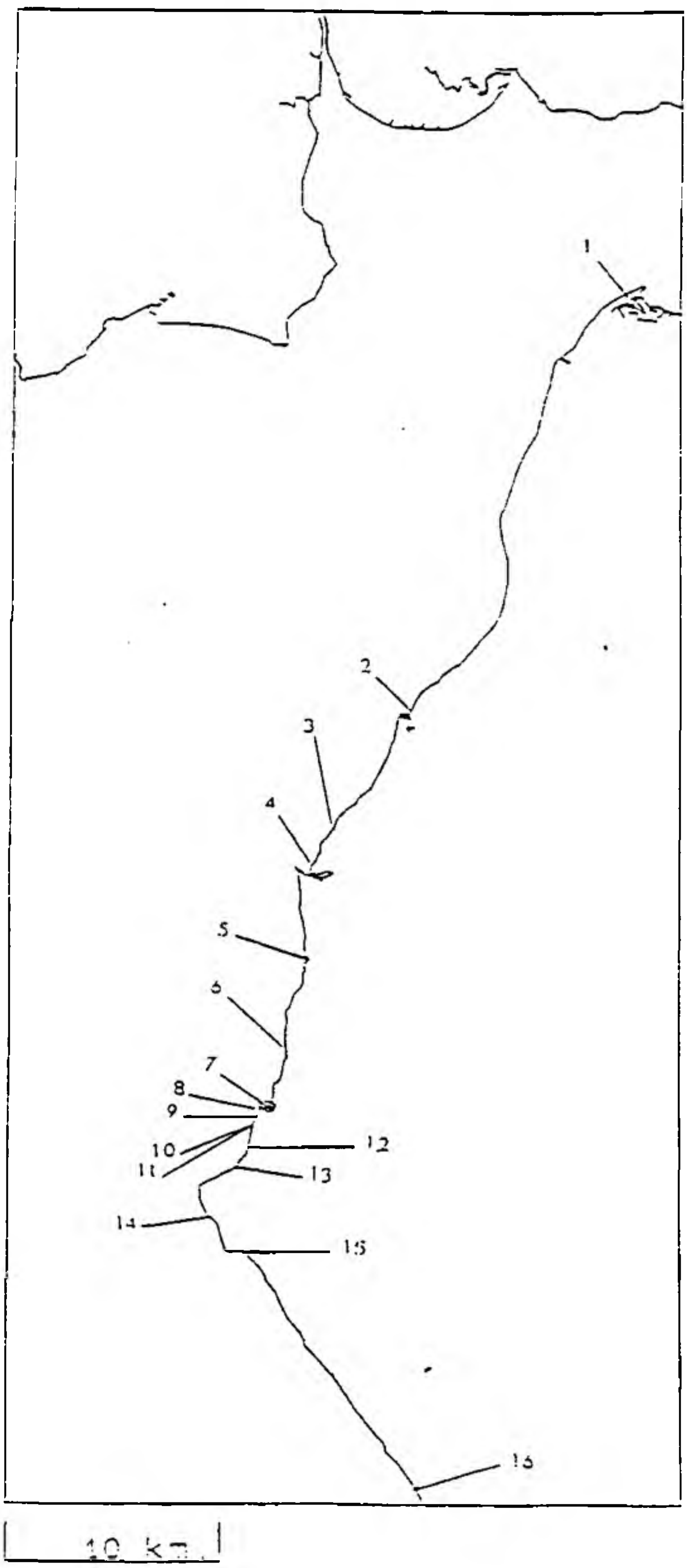
In addition, by selecting organisms of different feeding habit / ecological type (algae, grazers, deposit feeder, suspension feeder), it may be possible to evaluate the relative significance of contamination in different phases of the environment (dissolved, suspended solids, benthic sediments, diet etc.).

Because of their widespread distribution, and proximity to major discharges however, rocky shore species generally provide the most comprehensive information on spatial trends in contamination.

Sample Preparation and Metal Analysis

Following transport to the laboratory, organisms were maintained in clean seawater for upto 7 days in order to eliminate possible contamination from sediment particles present in the digestive system. Subsamples were digested by several methods and the analyses performed on the relevent digests for the metals silver, arsenic, cadmium, copper, chromium, iron, mercury, manganese, nickel, lead, tin and zinc. Total sediment metals (HNO_3 digest) as well as the 'bioavailable fraction' (1N HCl) were determined for the five surface sediment samples taken. Full details of these methods are discussed elsewhere (Bryan *et*

Fig. 1. Sampling Sites, West Cumbrian Coast 1990



	Site No.
Skinburness	1
Maryport	2
St. Helens	3
Workington	4
Harrington	5
Parton	6
Whitehaven	7
Tom Hurd's Rock	8
N. of Sewage Pipe	9
S. of Sewage Pipe	10
Whitehaven Mine	11
N. Limit of Outfall	12
S. Limit of Outfall	13
St. Bee's Head	14
St. Bee's	15
Seascale	16

Table 1. Sampling Sites and Occurence of Indicator Organisms, June 1990.

Site	Site No.	Map ref.	Km. from Skinburness	Species: Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	0.0	*	*		*		*	*
Maryport	2	NY034370	25.0	*	*	*	*	*	*	*
St. Helens	3	NX999317	32.0	*	*	*	*	*		
Workington shore	4a	NX989299	34.0	*	*	*	*	*		
Workington estuary	4b	NX994273	34.5	*					*	*
Harrington harbour	5a	NX989253	40.0	*	*				*	*
Harrington shore	5b	NX989253	40.0	*	*	*	*			
Parton	6	NX979211	46.0	*	*	*	*	*		
Whitehaven harbour	7	NX972183	48.0	*	*	*	*	*	*	*
Tom Hurd's Rock	8	NX965183	48.5		*	*		*		
N. of Sewage Pipe	9	NX965179	49.0		*					
S. of Sewage Pipe	10	NX964176	49.25		*	*	*	*		
Whitehaven Mine	11	NX964174	49.5	*	*			*		
N. Limit of Outfall	12	NX962165	50.5		*					
S. Limit of Outfall	13	NX955156	51.75	*	*	*		*		
St. Bee's Head	14	NX944133	55.0	*	*	*	*	*		
St. Bee's	15	NX958117	57.5	*	*	*		*		
Seascale	16	NY036007	71.0	*	*	*	*	*		

al, 1985; Langston, 1986). All results are expressed as $\mu g.g^{-1}$ (parts per million) on a dry weight basis.

Results

Because the sources and behavior of metals vary, results are described initially on a metal by metal basis. Where possible, concentrations are compared with similar data for other sites in UK estuaries and coastal waters. Since this data has been assembled in the PML laboratories, using identical methodology, the value of these comparisons is evident (no inter-laboratory variation). It should be stated however, that the data for UK sites in general has been accumulated over a period of at least 10 years and may not necessarily relate to current values, particularly where major changes in metal inputs have taken place recently.

Cadmium

Data for cadmium concentrations are presented in Appendix 1. Maps indicating the geographical extent of cadmium contamination, for each of the indicator species available, and for sediments, are also included (Figs. 2 - 9).

The most comprehensive information on spatial trends in cadmium contamination comes from data for the rocky-shore species (Figs. 2 - 6 and Fig. 10). All indicate maxima in the area between Whitehaven and St. Bee's Head (Saltom Bay), which coincides with the maxima for Cd in water (despite the limited data set). For *Littorina littorea* (Fig. 2) maximum concentrations (96 $\mu g.g^{-1}$) occur immediately to the north of the outfall of the "Marchon" chemical factory which is situated between stations 12 & 13, (Fig. 1), and decrease gradually in a northwards direction (by 50 - 75% at Whitehaven, 86% at Workington, and 95% at Skinburness). To the south of the outfall, concentrations decrease dramatically below St. Bee's Head which appears to act as a barrier in the movement of waterbourne cadmium in this region. The spread of cadmium contamination in a predominantly northerly direction from Saltom Bay is thus consistent with the residual currents along this part of the coast. Cadmium concentrations in other rocky-shore species; *Patella*, *Nucella*, *Mytilus* and *Fucus*, generally confirm these trends (Figs. 3 - 6).

For the current Cumbrian Survey, enrichment factors (Max/Min concentrations) for rocky shore species range from 5.4 for *Nucella* to 20.9 for *Littorina* (mean = 13).

As indicated in Table 1, *Littorina* was present at all sites but could not be found within 2-300m. of the Marchon outfall (Between sites 12 & 13, Northern and Southern limits of outfall). Indeed the shore was virtually devoid of life here. *Patella* and *Nucella* populations were affected over a slightly greater range (upto 400m. from the outfall, particularly on the Northern side, and the absence of these species was also apparent to the north of a sewage discharge into Saltom Bay (site 9). Mussels *Mytilus edulis* appear to be the species most affected along this part of the shoreline and were absent from all sites visited between Whitehaven Harbour and St. Bee's Head. The seaweed, *Fucus* spp. may also be impacted by the discharges since plants could only be found at two of the six sites in Saltom Bay.

Opportunities for sampling sediment habitats were much reduced along this stretch of coastline and were restricted to the sites shown in Figs. 7 - 9. Nevertheless, cadmium concentrations in sediment (Fig. 7) demonstrate a clear gradient with a maximum in Whitehaven Harbour declining northwards towards the Solway Firth by almost two orders of magnitude (Appendix 1.).

Cadmium concentrations in the ragworm, *Nereis diversicolor*; and clams *Scrobicularia plana* also mirror this contamination trend to some extent (Figs. 8 & 9), though it is clear that residues accumulated by these infaunal species

Fig. 2 Cadmium in Littorina littorea. Cumbria 1990.

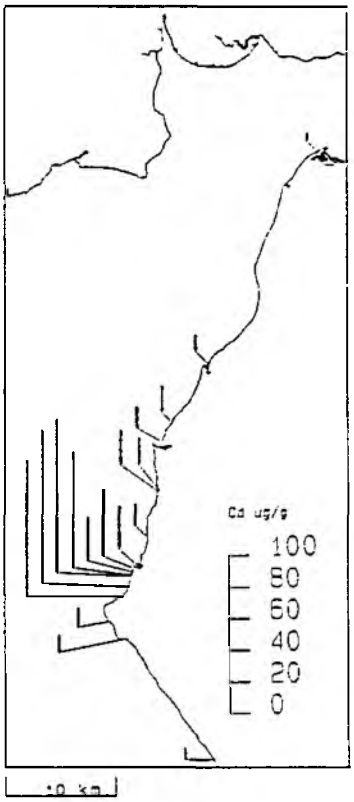


Fig. 3 Cadmium in Patella vulgata. Cumbria 1990.

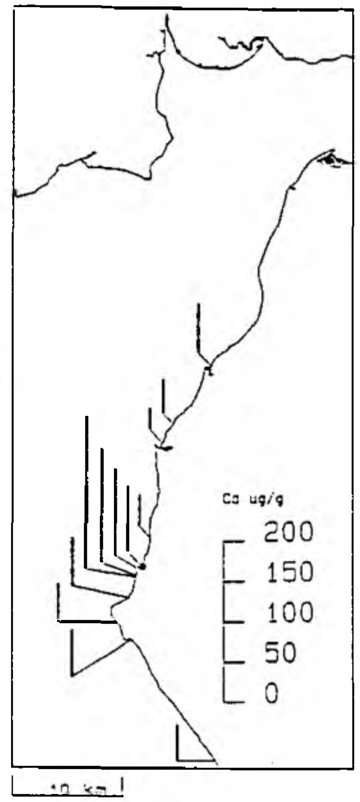


Fig. 4 Cadmium in Nucella lapillus. Cumbria 1990.

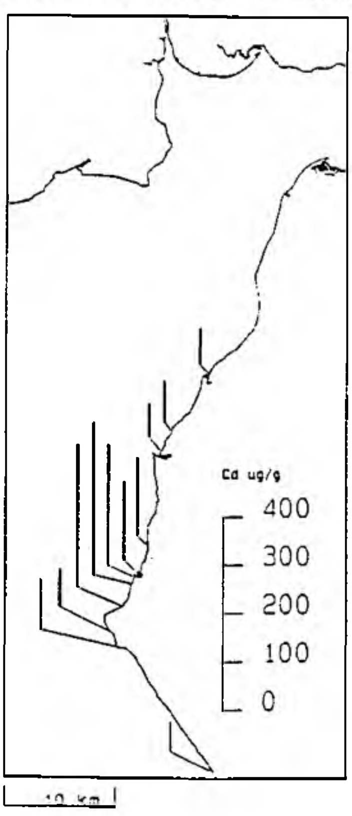


Fig. 5 Cadmium in Mytilus edulis. Cumbria 1990.

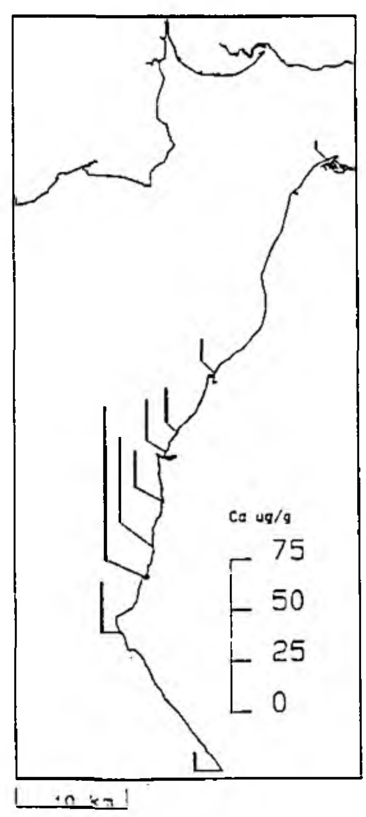


Fig. 6 Cadmium in Fucus vesiculosus. Cumbria 1990.

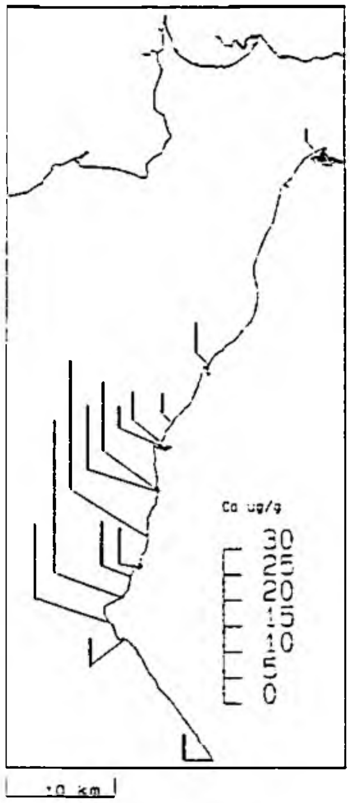


Fig. 7 Total Cadmium in Sediments. Cumbria 1990.

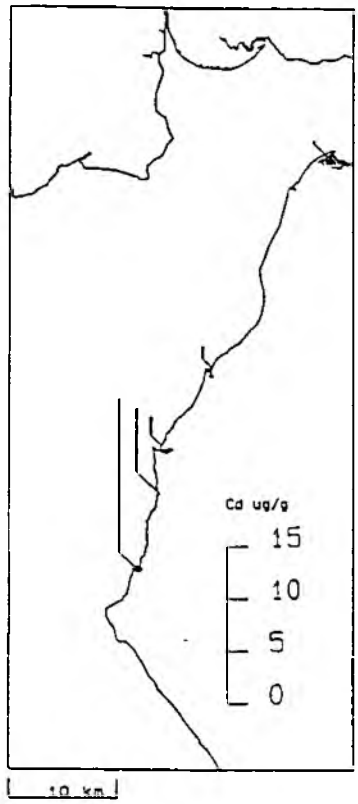


Fig. 8 Cadmium in Nereis diversicolor. Cumbria 1990.

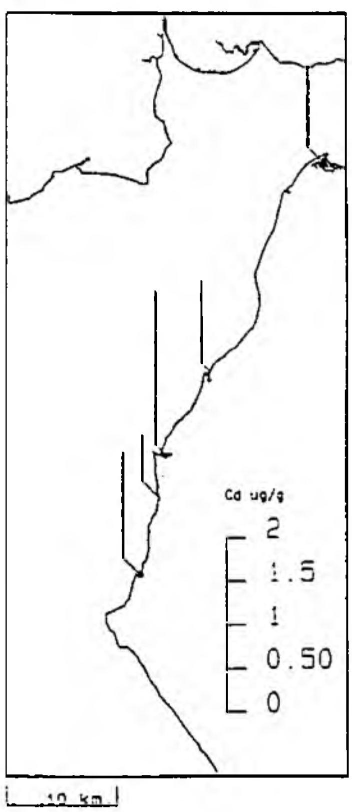


Fig. 9 Cadmium in Scrobicularia plana. Cumbria 1990.

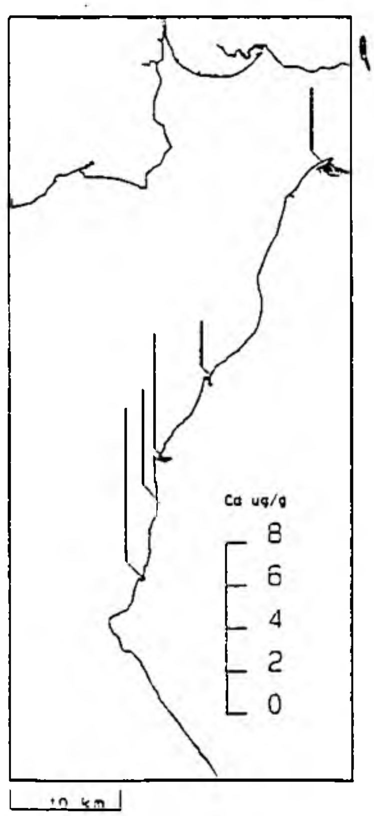
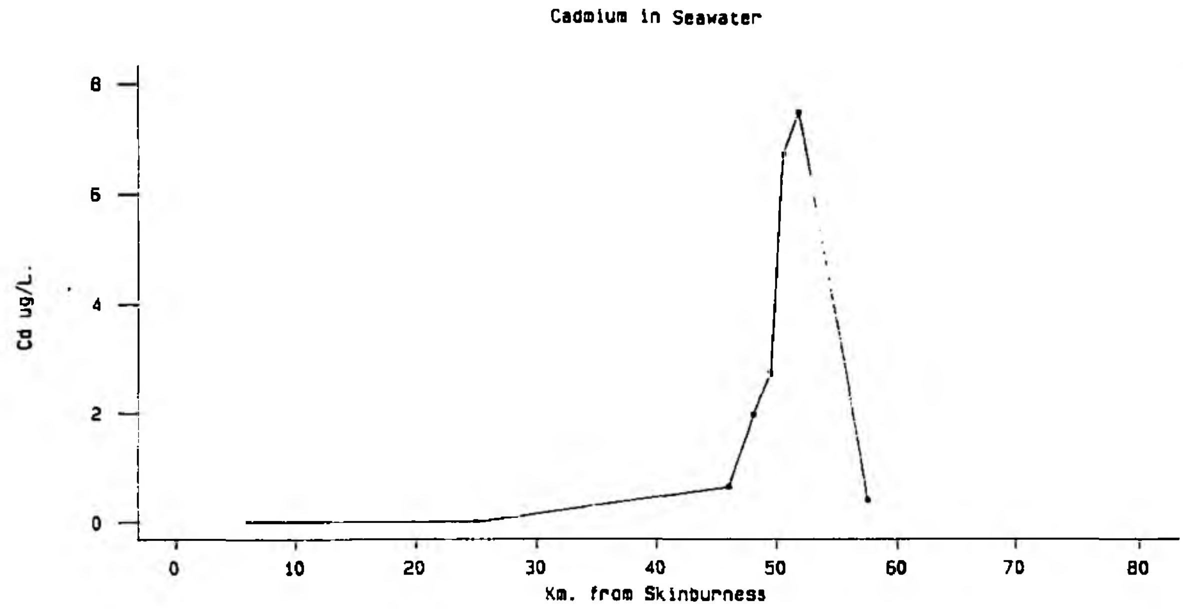
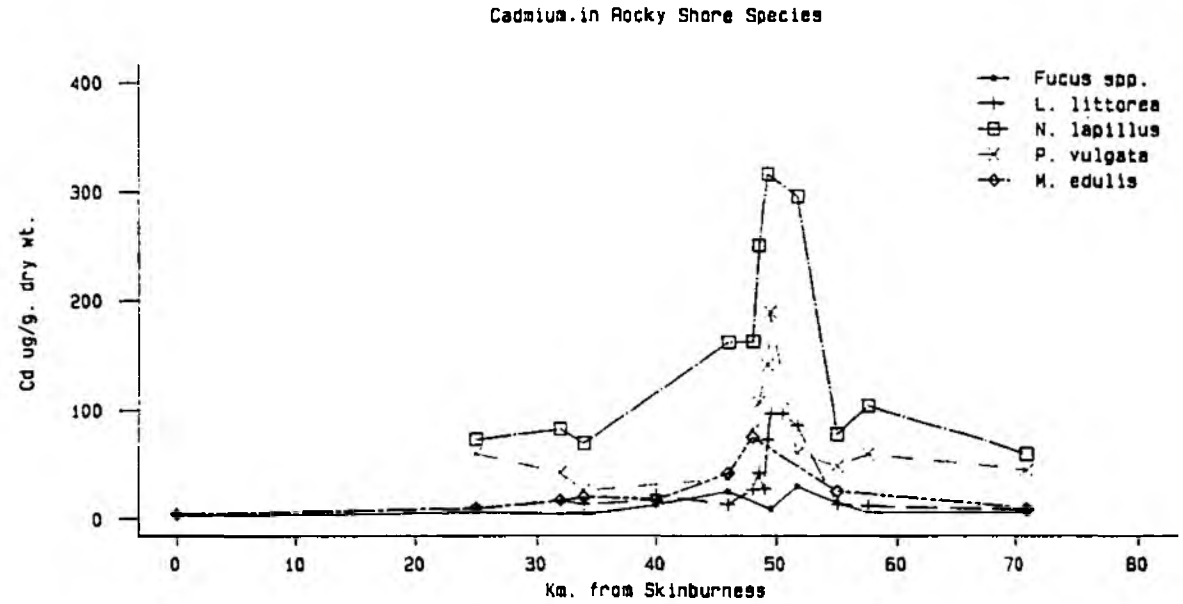
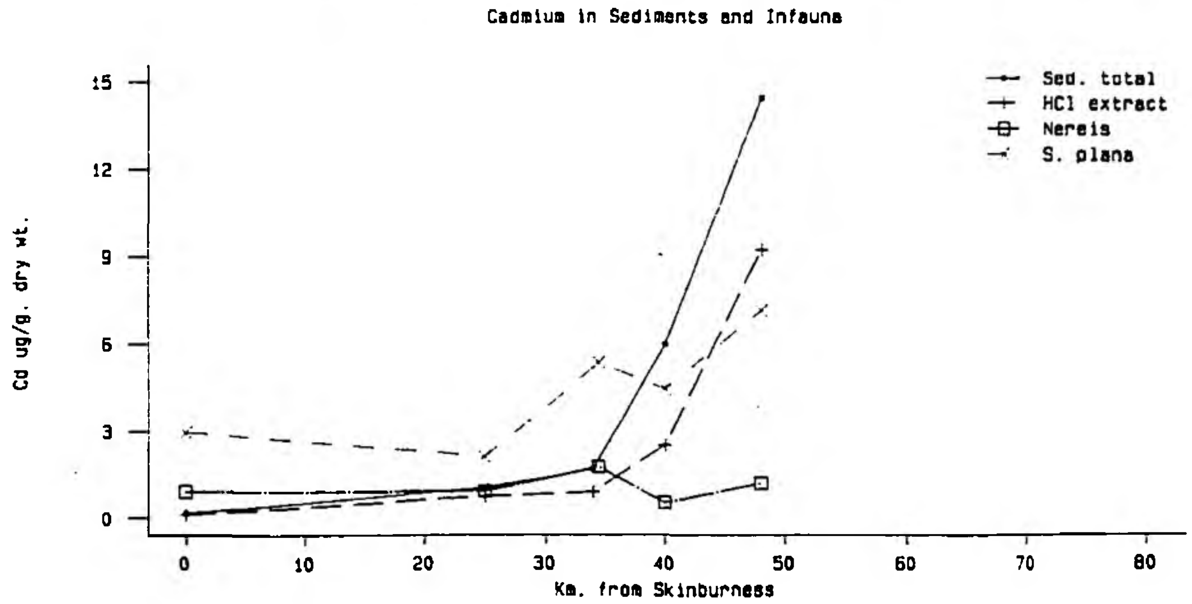


Fig. 10


Cadmium in Seawater
Cd ug/L.
8
6
4
2
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness


Cadmium.in Rocky Shore Species
Fucus spp.
L. littorea
N. lapillus
P. vulgata
M. edulis
Cd ug/g. dry wt.
400
300
200
100
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness


Cadmium in Sediments and Infauna
Sed. total
HCl extract
Nereis
S. plana
Cd ug/g. dry wt.
15
12
9
6
3
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

are relatively low, particularly in Nereis, and the gradients less steep (3 - 5 fold differences between maximum and minimum values) than for the rocky-shore species described previously. Even among the latter there is marked inter-specific variation in the amounts of cadmium accumulated and, allowing for the fact that not all species were collected at some of the sites, the "bioaccumulation potential" for cadmium declines in the sequence: *Nucella* > *Patella* > *Mytilus* > *Littorina* > *Fucus* > *Scrobicularia* > *Nereis*. The explanation for this ranking is probably related to the to the differences in quantity and quality of metal-binding ligands within each species and which influence the bioaccumulation process. Some of the ligands such as the metal-binding protein metallothionein are inducible and are thought to sequester, and thus fulfil a detoxifying role, for metals such as cadmium. It is tempting to speculate that variations in the ability to immobilise and hence detoxify metals may account for not only the differences in "bioaccumulation potential" mentioned above, but also the differences in tolerance observed between species along the shoreline in the vicinity of the Marchon outfall. Further research is urgently needed to address such problems. Similarly there is no certainty whether cadmium, or any other metal is solely responsible for the absence of organisms at any of the sites studied, though judging from the concentrations measured a contributary role seems inevitable.

A comparson of Cumbrian data with that of earlier surveys of UK estuaries (Appendix 1) shows that cadmium concentrations in Saltom Bay mussels and dogwhelks exceed upper limits, whilst those for limpets, winkles and seaweed, though slightly lower, are of a similar order to previously reported maxima.

It is interesting to note that although cadmium concentrations in Whitehaven sediments are exceptionally high, values for ragworms and clams are some 2 - 5 times lower than their corresponding UK maxima. It is, possible that the form of cadmium in Whitehaven sediments may be less bioavailable than at other locations. However, it may be relevant that both these sediment-dwelling species are less efficient accumulators of cadmium than for example; mussels, winkles, limpets and dogwhelks, and also that the sites where infaunal samples were taken are some distance from the Marchon outfall.

A final point of interest is that although cadmium concentrations decrease significantly either side of the Marchon outfall, (see Fig. 10 for summary) the data generally suggests that values may still be elevated at the extreme sites in the current survey. This evidence is supported by comparisons with UK minimum data (Appendix 1) and suggests that even at Skinburness, some 40km. north of the outfall, cadmium concentrations are 3 - 30 fold (depending on species) higher than values expected in the most pristine environment. A more extensive survey would be required to confirm these observations.

Arsenic

Data for As concentrations in biota and sediments are presented in Appendix 2.

Arsenic concentrations in water (Fig. 11) indicate an input of the metalloid in the area of the Marchon outfall, although within a relatively short distance, levels approach those considered to represent background values for UK waters (2 $\mu g.L^{-1}$).

A similar degree of As enhancement was observed in the brown seaweed, *Fucus vesiculosus*, and in the majority of rocky shore invertebrates studied (Fig. 11, Appendix 2), with maximum concentrations recorded between the outfall in Saltom Bay and Whitehaven harbour.

Based on the current Cumbrian survey, the Enrichment Factor (Max/Min), averaged for all rocky shore species is 3.8, with highest values in *Littorina littorea* (4.9), and lowest in *Mytilus edulis* (1.91). Lower values in the latter species are due to:

1) absence from the most contaminated sites, and;

2) the ability to partially regulate As bioaccumulation, thereby underestimating As contamination (Langston, 1984).

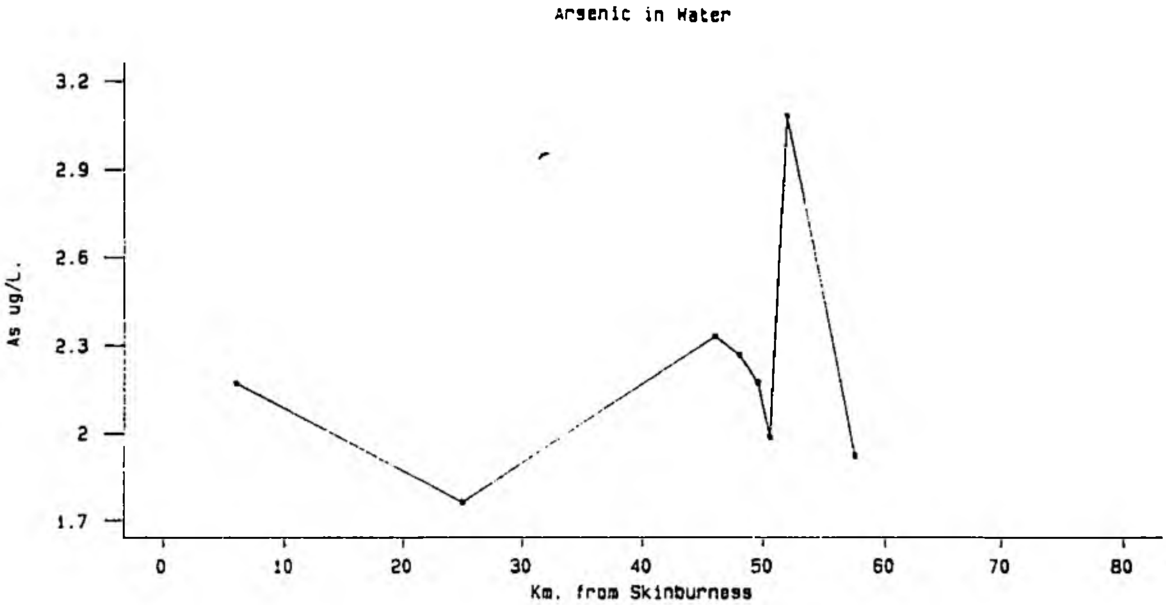
A map depicting As concentrations in *Littorina littorea* is presented in Fig. 12.

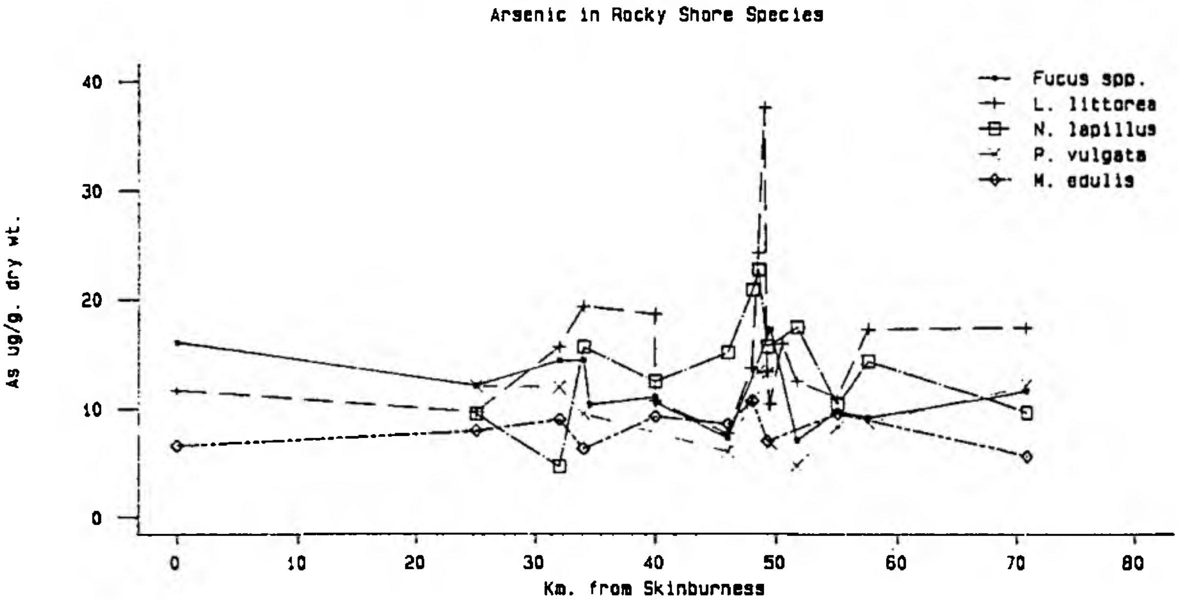
Arsenic enrichment along this part of the Cumbrian coastline therefore appears to be localised to a relatively small area of Saltom Bay. Since arsenic tends to be scavenged readily by precipitating Fe oxyhydroxides, especially where fresh and saltwater mix (Langston, 1983) removal to particulates may possibly be one explanation for the limited impact of this contaminant. In addition, competition for uptake between arsenate and phosphate is known to occur in a number of species. Since phosphate levels are likely to be high in this region they could be expected to influence the patterns of As contamination observed in biota. (See discussion for details).

Data for sediments and infaunal species are interesting in that, despite a seven-fold decline in total particulate As from Whitehaven, northwards, As concentrations in *Nereis* and *Scrobicularia* do not reflect this gradient, (Fig. 11). Tissue residues resemble more closely As concentrations found in 1N-HCl sediment extracts - often regarded as the best measure of biologically available sediment metal. However, lower sediment Fe concentrations at Skinburness may account for the relatively higher tissue burdens in clams and worms from this site (suggesting a reverse gradient almost) - previous studies have shown that As uptake is predictably reduced as Fe content in sediment increases (Langston, 1986). These results clearly demonstrate why it is often necessary to determine bioavailable, as opposed to total, metal in the environment, and in addition, why it is important to identify some of the major factors (other than the concentration of the contaminant) which might modify bioavailability.

Despite some evidence of inputs to Saltom Bay, the current results for the Cumbrian region suggest As concentrations in most species are among the lowest in the UK (possibly due to phosphate competition). Even at the most

Fig. 11


Arsenic in Water
As ug/L.
3.2
2.9
2.6
2.3
2
1.7
0
10
20
30
40
50
60
70
80
Km. from Skinburness


Arsenic in Rocky Shore Species
Fucus spp.
L. littorea
N. lapillus
P. vulgata
M. edulis
As ug/g. dry wt.
40
30
20
10
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

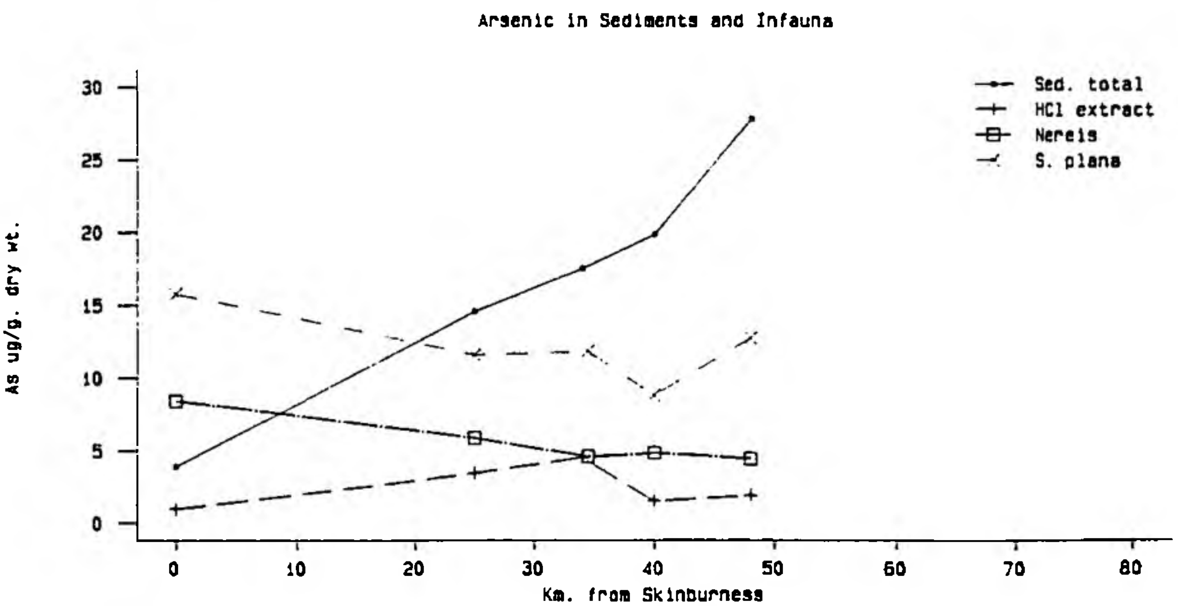
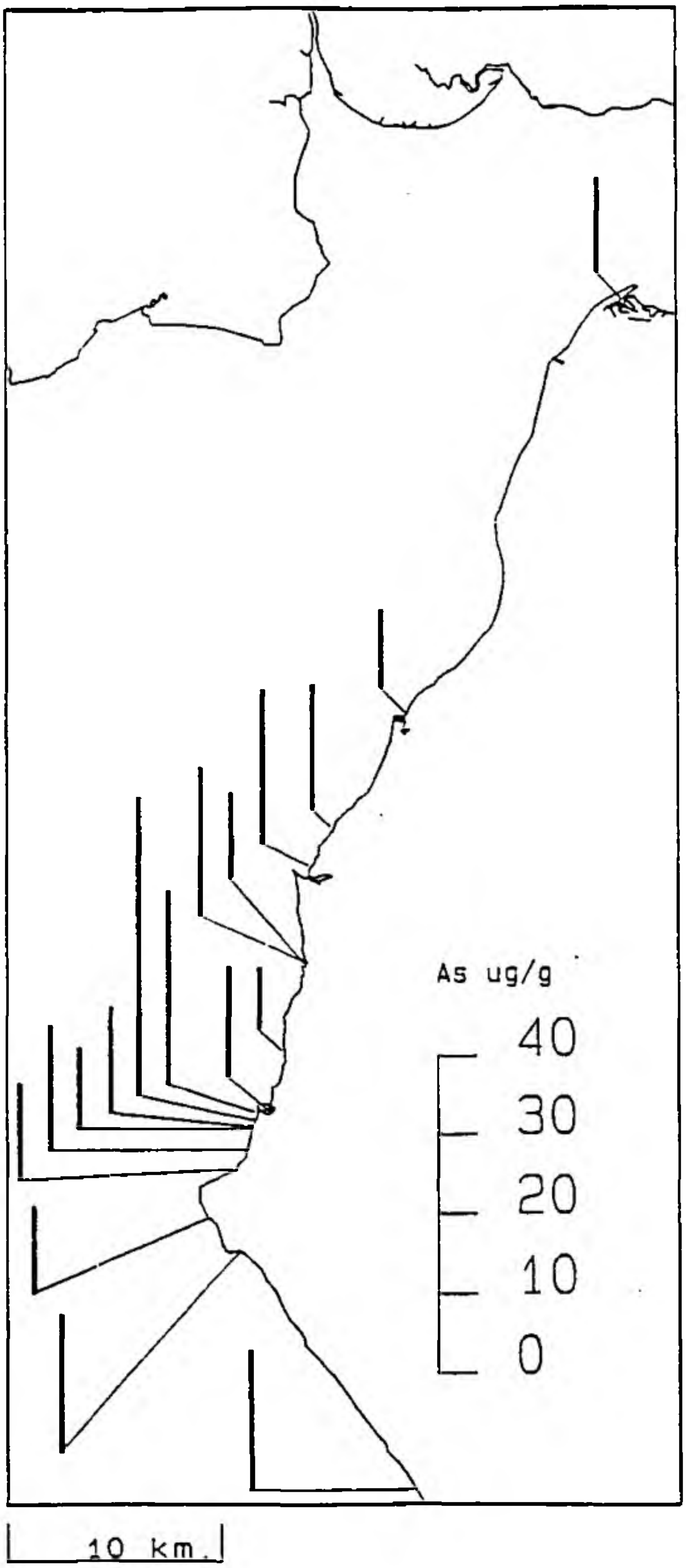

Arsenic in Sediments and Infauna
Sed. total
HCl extract
Nereis
S. plana
As ug/g. dry wt.
30
25
20
15
10
5
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

Fig. 12 Arsenic in Littorina littorea, Cumbria 1990.



contaminated sites, tissue burdens in *Fucus*, *Nereis* and *Scrobicularia* are usually at least an order of magnitude below UK maximum values (2 orders of magnitude for sediments).

Silver

Data for Ag concentrations in biota and sediments are presented in Appendix 3.

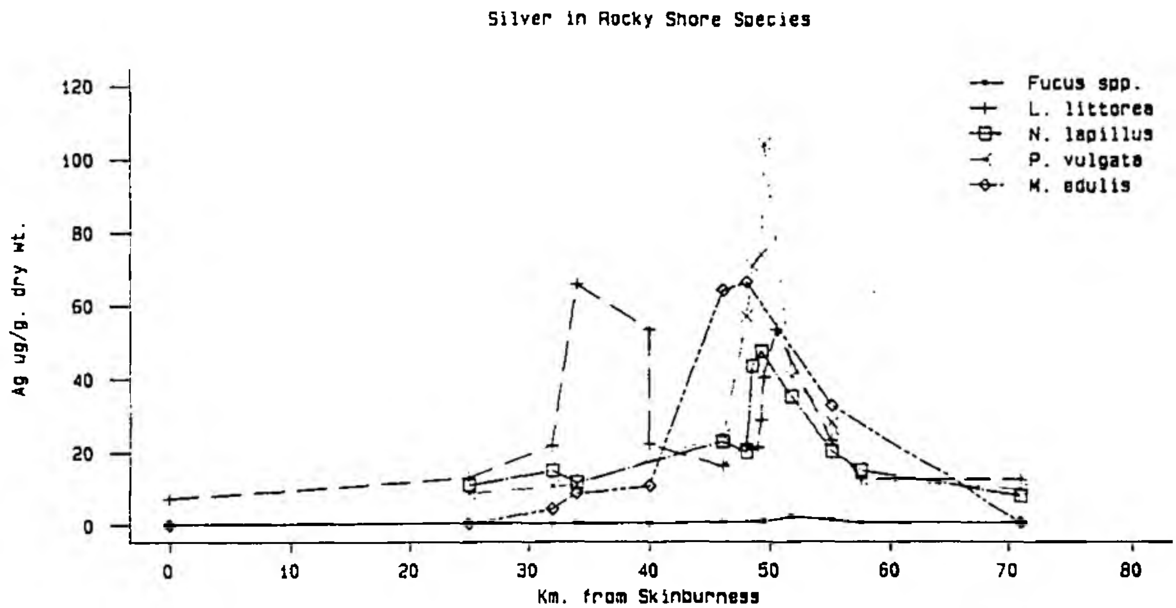
There are clear spatial gradients in Ag concentrations in all rocky shore species as summarised in Fig. 13. Although bioaccumulation of Ag in *Fucus* does not occur to the same scale as most invertebrates (hence relatively low levels in Fig. 13), mapping Ag in *Fucus* still establishes clear trends in contamination (Fig. 14), with peak values just to the south of the Marchon outfall. The majority of invertebrates indicate a sharp peak of Ag levels in the same area (Fig. 13). However, there are a number of species specific trends: Notably a second peak in Ag was detectable for *Littorina* at Workington and Harrington (Figs. 13 & 15). Mussels appear to be an exceptionally sensitive indicator for Ag since, despite their absence from the most contaminated area of shoreline, gradients in Ag can be detected over a wide area in the present survey (Fig. 13). This sensitivity is also reflected in the large range of values in mussels (Max/Min = Enrichment Factor = 761) compared to an average enrichment factor of 14 in other rocky shore species. [As mentionned earlier studies on metal binding components such as metallothioneins and granules should be conducted to explain these species differences in bioaccumulation potential].

Total Ag concentrations in sediments are highest at Whitehaven harbour (comparable to some of the highest UK values) and decrease northwards by a factor of 30, towards Skinburness (Fig. 13). This trend is reflected by Ag concentrations in clams, *Scrobicularia plana*, although the range of concentrations (5-fold) is less than for sediments, suggesting much of the total Ag in sediments may not be biologically available.

Silver in 1N-HCl sediment extracts and in worms, *Nereis diversicolor* displays a slightly different distribution (Fig. 13) with maximum values in the estuary at Workington. The reason for the apparent lack of correlation between Ag distributions in clams and worms is not clear but may indicate differential influences of dissolved and particulate metal between the two species, with *Nereis* preferentially accumulating Ag from overlying water (as observed for Cd in the Severn estuary, Bryan et al, 1985). Certainly, in comparison with clams, which probably absorb most metals from sediments, *Nereis* is not a particularly good accumulator of Ag as demonstrated by the concentrations shown in Fig. 13.

Much higher silver concentrations have been recorded in clams and worms from the Looe estuary which received effluents from film processing (see Appendix 3). For the majority of rocky shore species however, the peak silver values measured in samples from Saltom Bay generally exceed previously reported UK maxima. Only for *Littorina littorea* have comparable concentrations been determined elsewhere.

Fig. 13


Silver in Rocky Shore Species
Fucus spp.
L. littorea
N. lapillus
P. vulgata
M. edulis
Ag ug/g. dry wt.
120
100
80
60
40
20
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

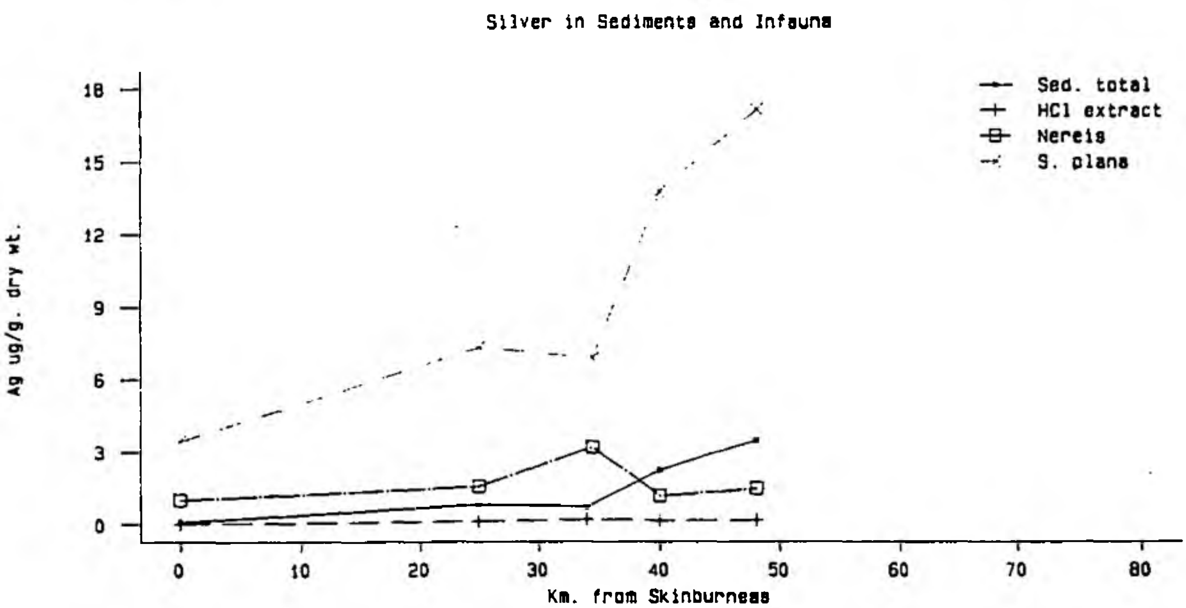

Silver in Sediments and Infauna
Sed. total
HCl extract
Nereis
S. plana
Ag ug/g. dry wt.
18
15
12
9
6
3
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

Fig. 14 Silver in Fucus vesiculosus. Cumbria 1990.

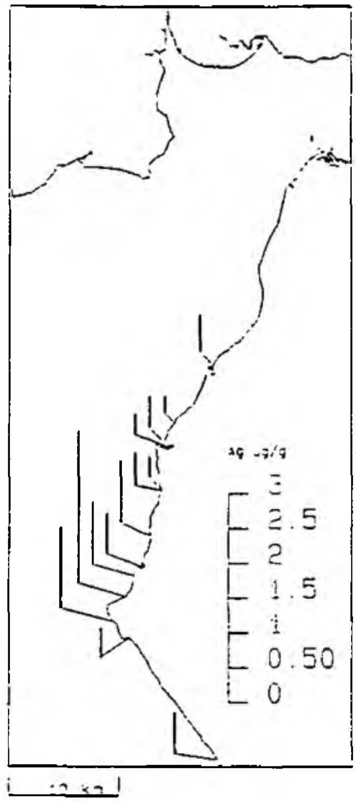
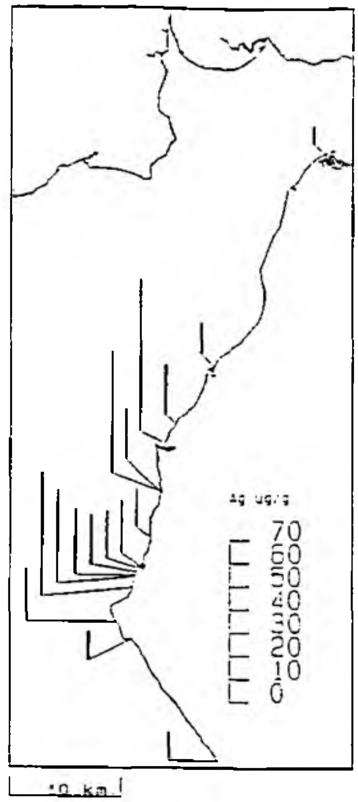


Fig. 15 Silver in Littorina littorea. Cumbria 1990.



Copper

Data for copper concentrations in sediments and biota are presented in Appendix 4.

The limited data for Cu in seawater clearly demonstrates a significant input in Saltom Bay close to the outfall from the "Marchon" plant. The effects of dilution and dispersion appear to restrict exceptionally high concentrations to within a few kilometers of the outfall. This pattern is reflected by all the invertebrates sampled from rocky shores (Fig. 16), though as is often the case with metals the amounts of copper accumulated vary considerably between species. This can partly be explained by the fact that Cu is an essential element and the organism may, under normal conditions (or slightly elevated Cu contamination) regulate body burdens to meet its requirements, which will clearly differ between species. At higher concentrations this regulation breaks down and Cu may be accumulated without control. Thus for *Nucella*, *Patella*, and *Littorina*, body concentrations appear constant, except for the area of shoreline between Whitehaven harbour and St. Bees Head (Fig. 16, Appendix 4 and map for *Littorina* (Fig.17)), where extremely high concentrations were measured. Accumulation of copper in seaweed *Fucus vesiculosus* is probably proportional to that in water (up to 100 $\mu g.L^{-1}$) and therefore residues in this species, though lower than invertebrates, are likely to be a true averaged reflection of gradients in Cu bioavailability in seawater (see map, Fig. 18).

It seems likely that Cu toxicity is a factor contributing to the absence of mussels, *Mytilus edulis* from a large part of the shoreline between Whitehaven and St. Bees Head. Lacking the Cu containing blood pigment haemocyanin, which is present in the gastropods sampled, it is unlikely that the Cu requirements of mussels is very high (hence the lower baseline values - Appendix 4). Thus although there is some evidence that mussels can regulate Cu to a limited degree (tissue residues considerably underestimate contamination with Cu) this species appears to be vulnerable to relatively low Cu concentrations. Experiments have shown effects on growth, larval development, and reproductive potential in the range 3 - 50 $\mu g.L^{-1}$ Cu, levels comparable with those measured in the current survey (Stromgren, 1982; Martin *et al*, 1981; Myint and Tyler, 1982). Though less widely affected, for reasons described above, the absence of other species from the most contaminated sites might also be ascribed partly to Cu toxicity.

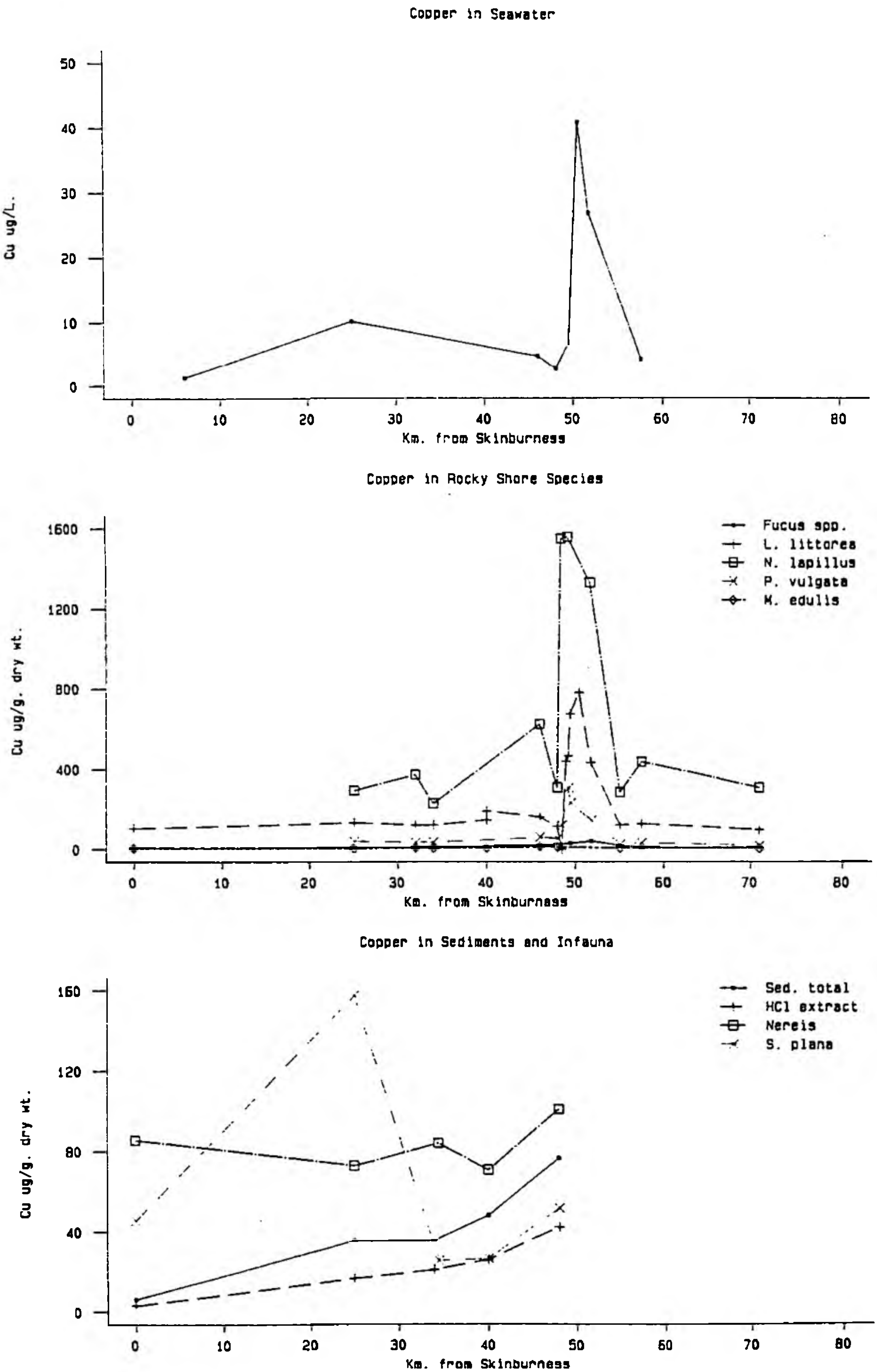
Enrichment factors (Max/Min) for Cumbrian rocky shore samples range from 5.2 for mussels to 20.5 for limpets (mean value = 9.9).

Nereis diversicolor is generally considered to be a good indicator of Cu contamination in particulates, and profiles for Cumbrian estuarine samples show that Cu gradients in sediments and worms (Total and 1N-HCl extracts) are significantly related (Fig. 16). The decrease in Cu concentration in worms northwards, away from Whitehaven, is however notably less pronounced than for sediments.

The behavior of *Scrobicularia plana*, with respect to sediment Cu, is not fully understood, since high tissue concentrations sometimes occur in clams at sites considered relatively uncontaminated with Cu (Bryan *et al*, 1985). It is suspected that very anoxic conditions may be responsible for such unusually high Cu burdens in clams, and this may be a possible explanation for the anomalously elevated Cu concentration in *S. plana* from Maryport harbour. At the time of sampling there was considerable disturbance of the harbour sediments, due to reconstruction of the quayside: remobilisation of Cu as a result of these operations may also have contributed to the high Cu concentration in clams.

Copper concentrations in sediments and infaunal species from Cumbrian

Fig. 16



Copper in Seawater
Cu ug/L.
50
40
30
20
10
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness
Copper in Rocky Shore Species
Fucus spp.
L. littorea
N. lapillus
P. vulgata
M. edulis
1600
1200
800
400
0
Cu ug/g. dry wt.
Km. from Skinburness
Copper in Sediments and Infauna
Sed. total
HCl extract
Nereis
S. plana
160
120
80
40
0
Cu ug/g. dry wt.
Km. from Skinburness

Fig. 17 Copper in Littorina littorea, Cumbria 1990.

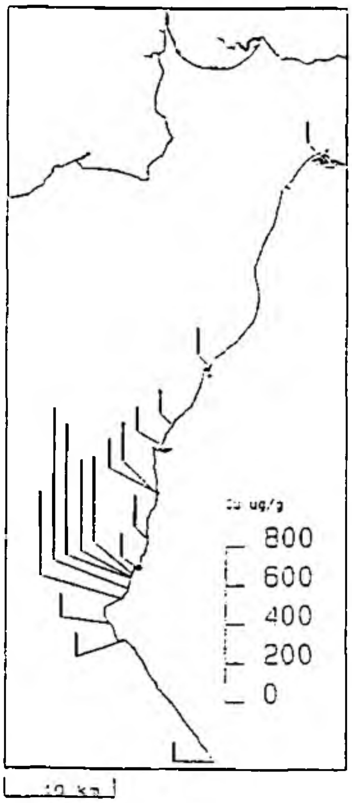
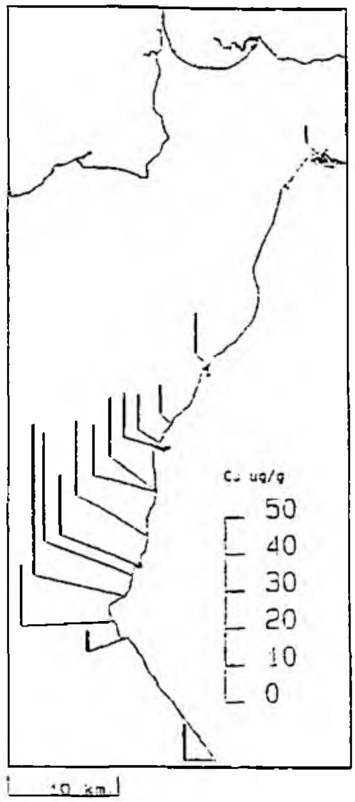


Fig. 18 Copper in Fucus vesiculosus, Cumbria 1990.



estuaries are, at worst, an order of magnitude lower than in samples from heavily polluted estuaries in south west England (Appendix 4). However, for a number of rocky shore gastropods collected at sites in Saltom Bay, Cu burdens are equivalent to some of the highest encountered in the UK. Even at the "cleaner" sites sampled in the present survey, Cu concentrations in gastropods (and worms and clams) are significantly higher than UK baseline values, suggesting that the influence of contamination is widespread along the coast, albeit at much lower levels than in Saltom Bay. The form of this contamination is uncertain however since, paradoxically, Cu levels in *Fucus* imply the approach of background levels for dissolved bioavailable Cu at the extreme sites in the current sampling range (Skinburness, Seascale, Appendix 4).

Chromium

Data for Cr concentrations in sediments and biota are presented in Appendix 5. No samples of *Patella vulgata* were analysed for this metal due to problems with acid digestion.

The highest Cr concentrations were measured in *Fucus* and *Littorina* collected closest to the outfall from the "Marchon" plant (Figs. 19 - 21). The relatively high concentrations in *Fucus* indicates the likely presence of dissolved chemical species of which hexavalent Cr(VI) is the most biologically available (and toxic) form.

Mytilus edulis has been suggested as a useful indicator for Cr(VI), based on experimental field trials using large enclosures (Schulz-Baldes *et al*, 1983). Despite their absence from the most contaminated sites, elevated levels of Cr were clearly evident in mussels from Saltom Bay between Whitehaven harbour and St. Bees Head (Fig. 19, 22). Levels declined sharply to the south of St.Bees and to the north of Whitehaven. However, a further increase in bioavailable Cr was detectable in mussels (and, to a lesser extent, *Fucus* and *Littorina*) from the Workington area (Fig. 19, 22).

The range of chromium values (Max/Min = Enrichment Factor) measured in Cumbrian samples is highly species dependent and, for the above species, varies between 13 for *Mytilus* and 64 for *Fucus*. The range of concentrations is lower in *Nucella* (6) which appears to be a relatively poor accumulator of Cr.

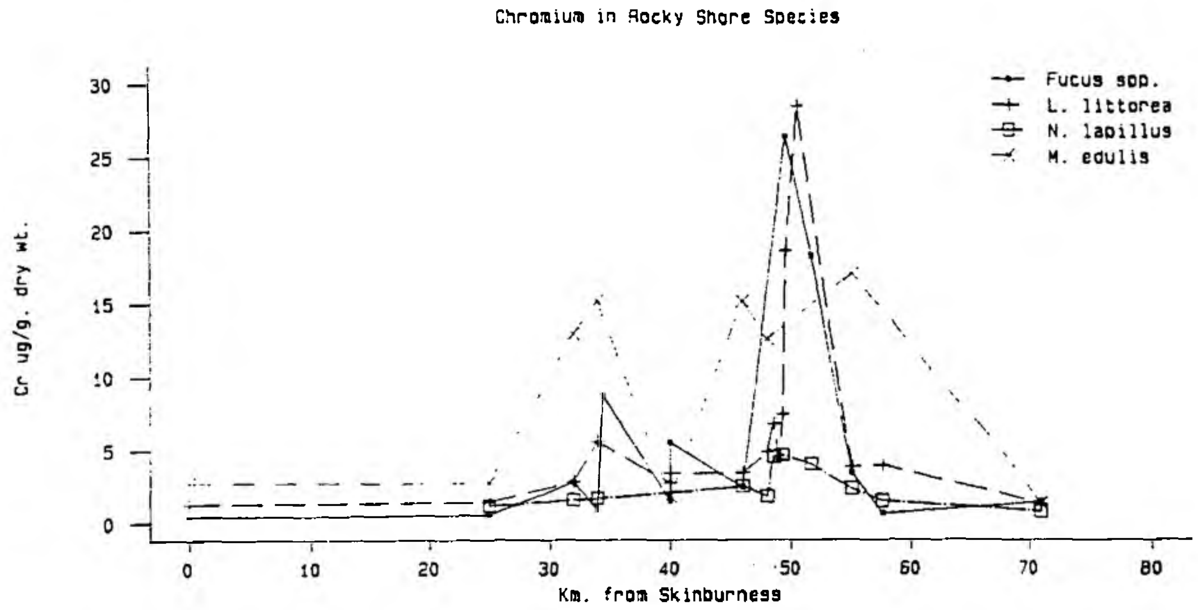
Zaroogian and Johnson (1983) demonstrated that mussels exposed to 10 $\mu g.L^{-1}$ dissolved Cr(vi) accumulated 9.4 $\mu g.g^{-1}$ Cr in their tissues after 3 months. Since Cr concentrations in mussels from St.Bees and Whitehaven harbour had accumulated 16.9 and 12.5 $\mu g.g^{-1}$ respectively it seems likely that dissolved Cr in Saltom Bay seawater may at certain sites exceed 10 $\mu g.g^{-1}$ by a considerable margin. A concentration of 50 $\mu g.L^{-1}$ Cr has been shown to impair oocyte development in mussels by 50% (Myint and Tyler, 1982): Therefore some contribution from Cr towards the elimination of mussels at heavily contaminated sites seems likely.

In UK estuarine sediments total Cr concentrations range from 15 - 800 $\mu g.g^{-1}$ (Appendix 5) with the highest contamination levels occuring near to effluents arising from the manufacture of tin plate. Sediments from Whitehaven harbour must therefore be considered as fairly contaminated, and the high percentage of easily (1N-HCl) extractable Cr (62%) is a good indication that much of this is anthropogenic in origin. The decline in total Cr in sediments, northwards, together with a reduction in the proportion of easily extractable Cr (11.1% at Skinburness) is consistent with a change towards more uncontaminated sediments.

Nereis diversicolor and *Scrobicularia plana* undoubtably accumulate some sedimentary Cr, although much of the metal in sediments is likely to be present as the less soluble, less bioavailable, trivalent Cr(III). Furthermore, because of the high sediment Cr loading (relative to tissue levels) at some sites, it seems likely that some of the body burden is due to contamination with fine particles of ingested sediment, despite efforts to clean up the animals for 1 week prior to analysis. Relationships between tissue levels (in clams and worms) and sediment concentrations are less obvious for Cr than a number of metals (Fig. 19).

An overall evaluation of Cr contamination is therefore complex: While Cr concentrations in sediment and infauna from the present survey may be somewhat lower than in estuaries impacted by tin-plating industry, the values measured in *Fucus*, *Littorina* and *Mytilus* between St. Bees and Whitehaven are the highest reported in UK waters to date. These results imply significant inputs of Cr in this region.

Fig. 19


Chromium in Rocky Shore Species
Fucus spp.
L. littorea
N. lapillus
M. edulis
Cr ug/g. dry wt.
0
5
10
15
20
25
30
0
10
20
30
40
50
60
70
80
Km. from Skinburness

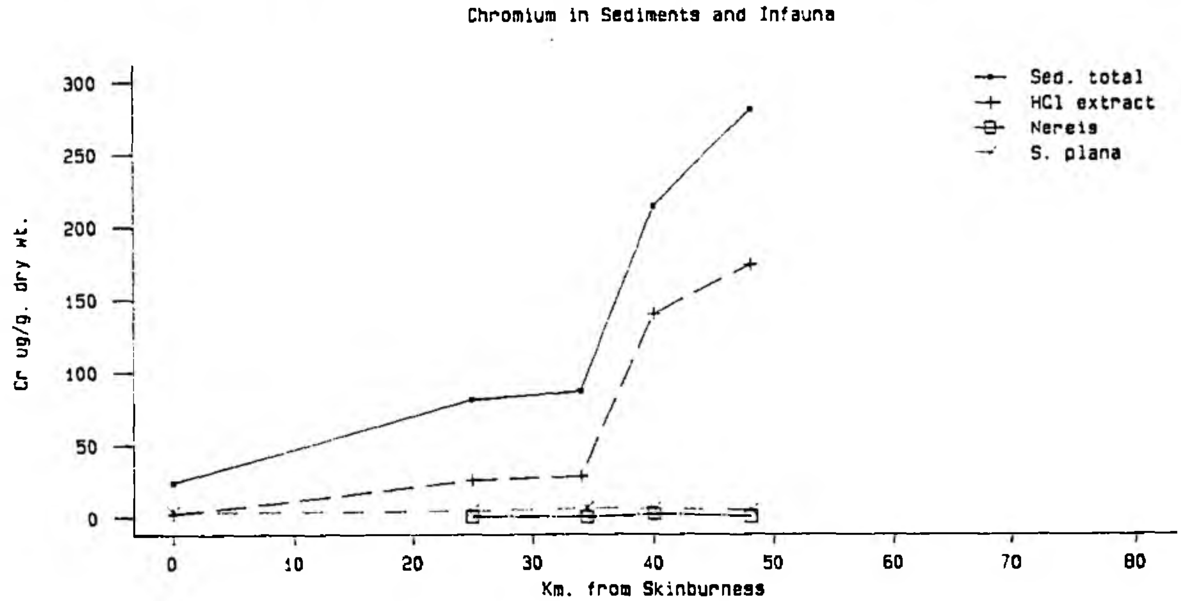

Chromium in Sediments and Infauna
Sed. total
HCl extract
Nereis
S. plana
Cr ug/g. dry wt.
0
50
100
150
200
250
300
0
10
20
30
40
50
60
70
80
Km. from Skinburness

Fig. 20 Chromium in *Fucus vesiculosus*, Cumbria 1990.

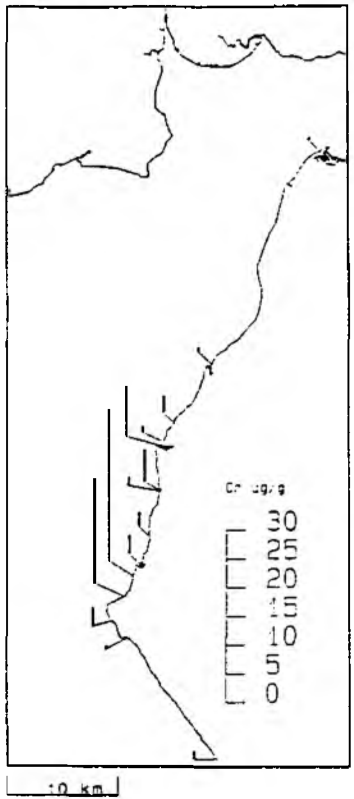


Fig. 21 Chromium in *Littorina littorea*, Cumbria 1990.

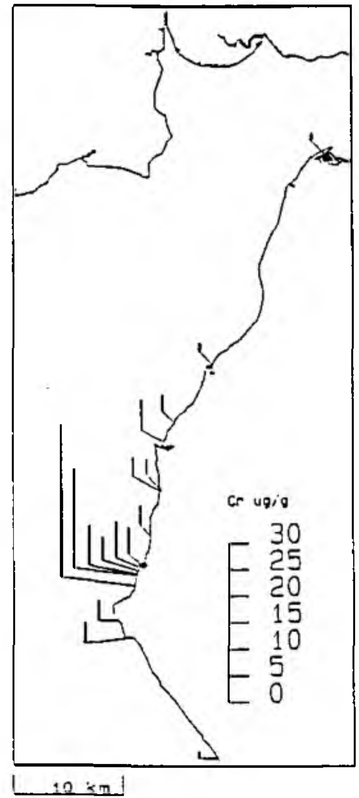
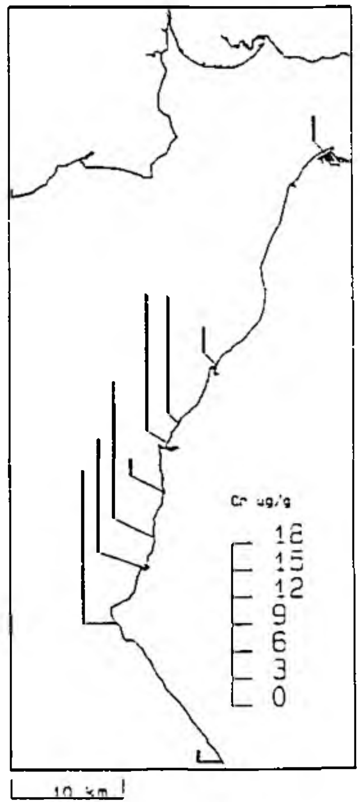


Fig. 22 Chromium in Mytilus edulis, Cumbria 1990.



Iron

Data for Fe concentrations in biota and sediments are presented in Appendix 6.

Iron is one of the most common elements in the earths crust, and sediments usually contain thousands, or even tens of thousands of ppm Fe. Since iron ore has been processed along this coastline for centuries, the occurence of relatively high levels (up to 40,882 $\mu g.g^{-1}$ at Workington) is not surprising. In contrast to total Fe, extractable (1N-HCl) Fe follows a well defined gradient decreasing from Whitehaven northward (Fig. 23).

Because it is a common element, Fe has been utilised as an essential metal by a variety of organisms - notably for example by *Patella vulgata* as a component of the radula (hence the high Fe concentrations in this species). As a result it is questionable whether any of these species is efficient at indicating Fe contamination. Also because Fe is so predominantly associated with particulates in marine systems, the presence of relatively small amounts of sediment-bound Fe (on the surface, or within the gut) can lead to an overestimation of tissue burdens. This is particularly important with *Fucus*, since despite attempts to clean fronds, fine particles of sediment may adhere to the surface, contributing to the total Fe burden, particularly where sediment Fe concentrations are high (eg. Workington). Bearing in mind these problems, a map of Fe in *Fucus* is shown in Fig. 24 with elevated levels occuring in the estuary at Workington and at sites 3, 5a, and 11 (St. Helens, Harrington Harbour, and Whitehaven Mine respectively).

The range of Fe concentrations (Max/Min) is relatively small for rocky shore invertebrates (3.1 in *Littorina* - 5.45 in *Patella*; mean = 4.7). Results for *Patella* and *Littorina* are similar to *Fucus*, indicating elevated levels to the south of Whitehaven and in the Workington/St. Helens region (Figs. 23, 25, 26).

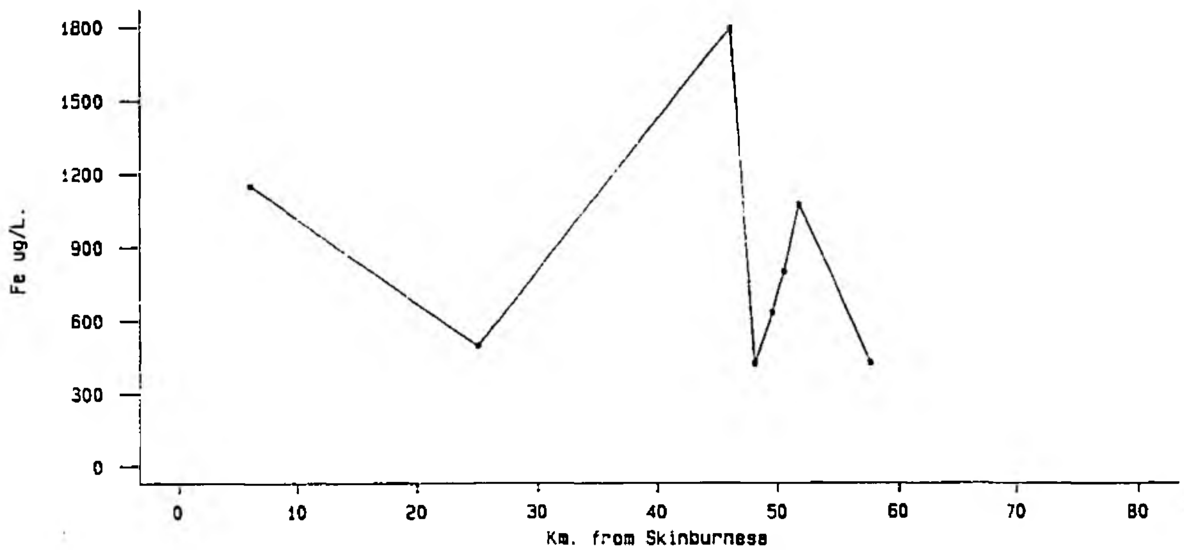
For *Fucus*, *Nucella* and *Patella* in particular, Fe levels are higher at some sites than previously reported UK maxima demonstrating a degree of Fe contamination in the area.

Overall, however, in view of possible regulation by organisms, coupled with the risk of contamination by particulates, it is difficult to determine with any certainty the extent and relative significance of industrial activities on Fe levels in Cumbrian biota, based on the evidence of the current survey. Further work would be necessary to confirm the apparent enhancement at sites summarised in Fig. 23.

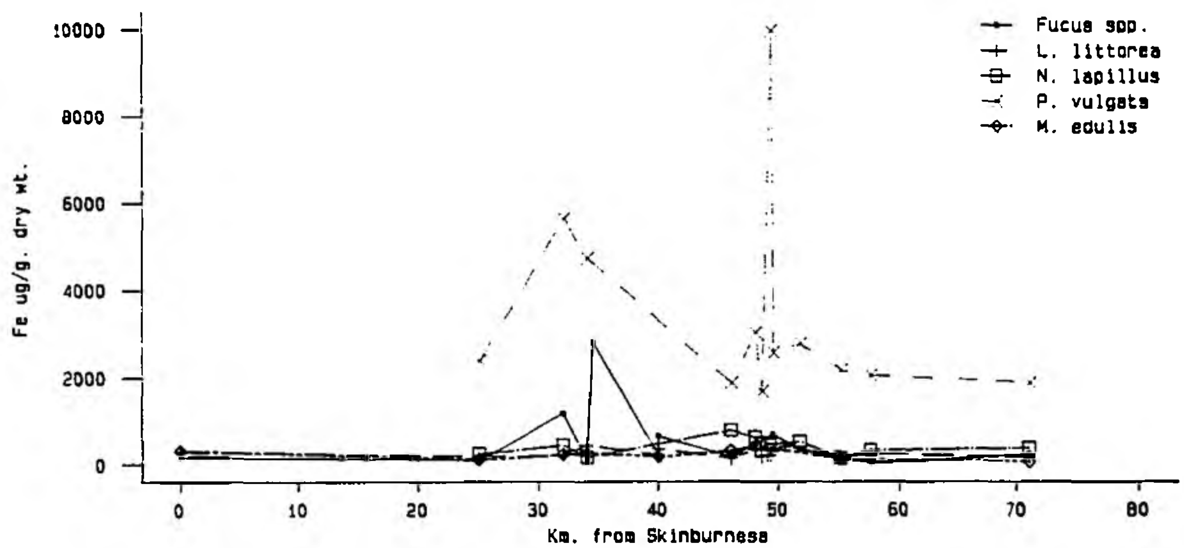
Although clams and worms are normally considered to be good indicators of sediment metal, their ability to monitor Fe contamination is very limited, for reasons described above. The Fe containing pigment haemoglobin is present in *Nereis diversicolor* reducing the value of this species as an Fe indicator still further.

Fig. 23

Iron in Seawater



Iron in Rocky Shore Species



Iron in Sediments and Infauna

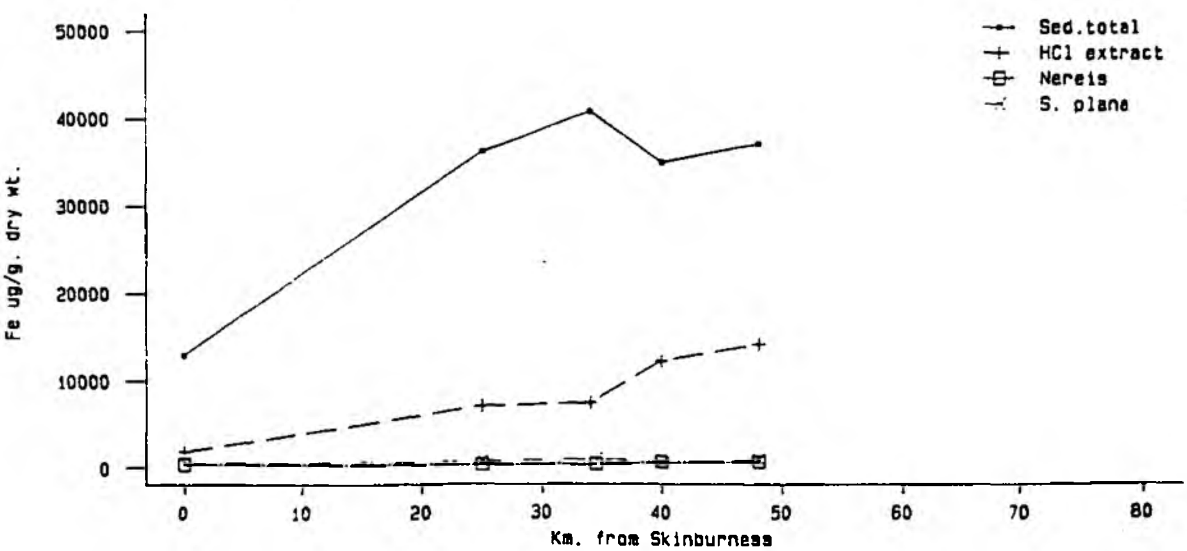


Fig. 26 Iron in *Littorina littorea*, Cumbria 1990.

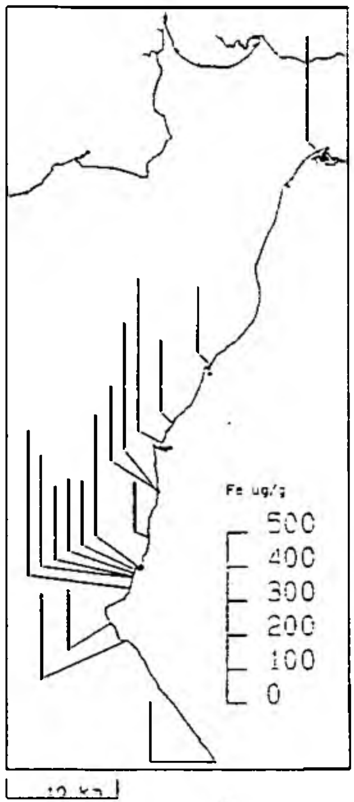


Fig. 24 Iron in Fucus vesiculosus. Cumbria 1990.

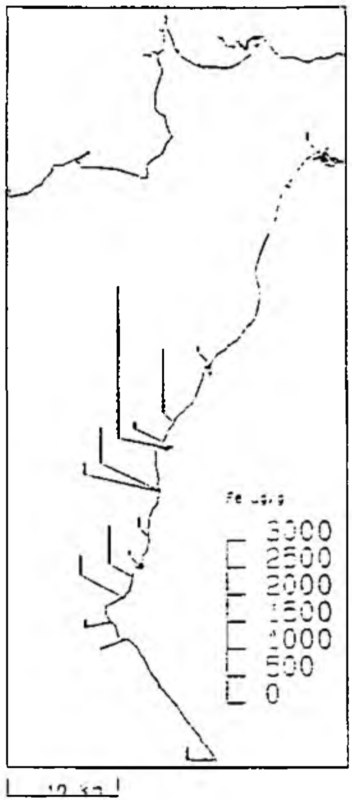
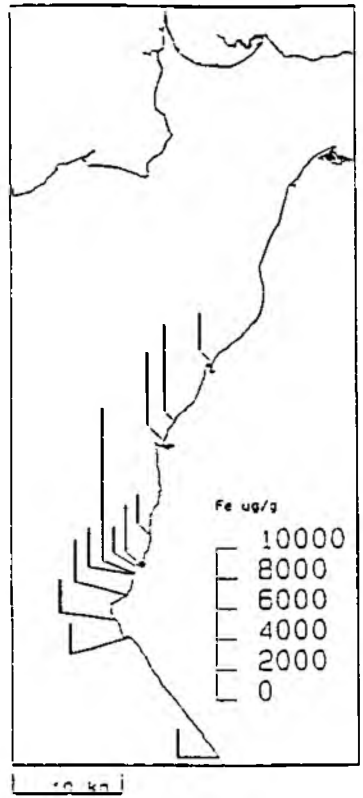


Fig. 25 Iron in Patella vulgata. Cumbria 1990.



Mercury

Data for Hg concentrations in sediments and biota are presented in Appendix 7.

There are clearly significant variations in Hg concentrations along the Cumbrian coast which are reflected in most of the rocky shore biota (Fig. 27). Enrichment factors (Max/Min concentrations) vary between 4.5 in *Fucus* and 10.6 in *Littorina* (mean = 6.0).

The highest Hg concentrations were encountered at sites in Saltom Bay, although the exact position of the maximum values varied between species. The most obvious peak in concentrations is observed in *Littorina*, since they are the only species collected relatively close to the "Marchon" outfall (see Figs. 27 and 28). Mercury gradients in most other species are less pronounced (Fig. 27, and demonstrated in map form for *Fucus* in Fig 29).

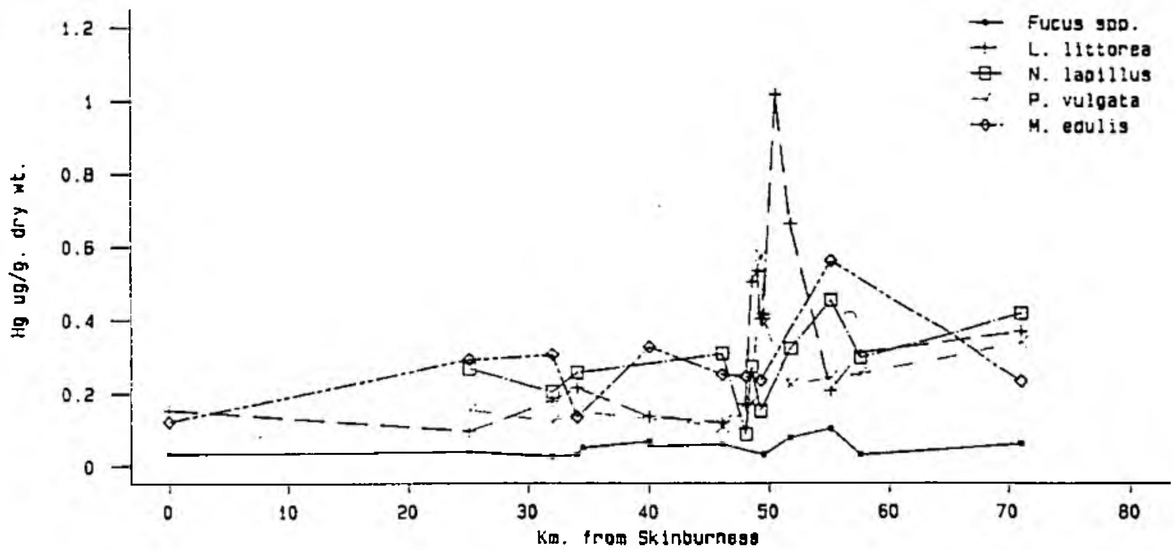
Although no other Hg inputs to the Cumbrian coast are identified in the current data, there is an underlying decreasing trend in Hg concentrations northwards (Fig. 27). The reason for this is not fully understood but it is interesting to speculate that the higher background concentrations in the south of the present survey area may reflect known sources of Hg enriched waters in this direction, notably Liverpool Bay (see for example MAFF, 1990). Thus, at Skinburness in the north, Hg concentrations in the majority of coastal species sampled are representitive of the most uncontaminated sites in the UK, but at Seascale/St. Bees (the southern limit of the present survey) concentrations tend to be higher by a factor of 2.

Despite the considerable enhancement of Hg concentrations in biota in Saltom Bay, tissue burdens in most species are generally lower (by about 50%) than worst-case UK values.

Apart from the enhanced levels found at Maryport mercury concentrations in sediments (Fig. 27) generally follow the expected south-north decline (by a factor of 18) observed for most other metals. Remarkably however, this trend is almost reversed in infaunal species *Nereis diversicolor* and *Scrobicularia plana* (Fig.27). One likely explanation for this apparent anomaly is that Hg availability to these organisms increases as the organic content of the sediment decreases (Langston, 1986): The low organic content of Skinburness sediments (1.2% weight loss on ignition) may effectively enhance Hg accumulation by infaunal organisms, whilst high organic loadings in, for example, Whitehaven sediments (6.7%) may suppress uptake. Thus, as is often the case with sediment-bound metals, it is not always the total amount of metal present which is biologically significant, but rather the physico-chemical form (speciation) or presence of competing agents which modify bioavailability.

Fig. 27

Mercury in Rocky Shore Species



Mercury in Sediments and Infauna

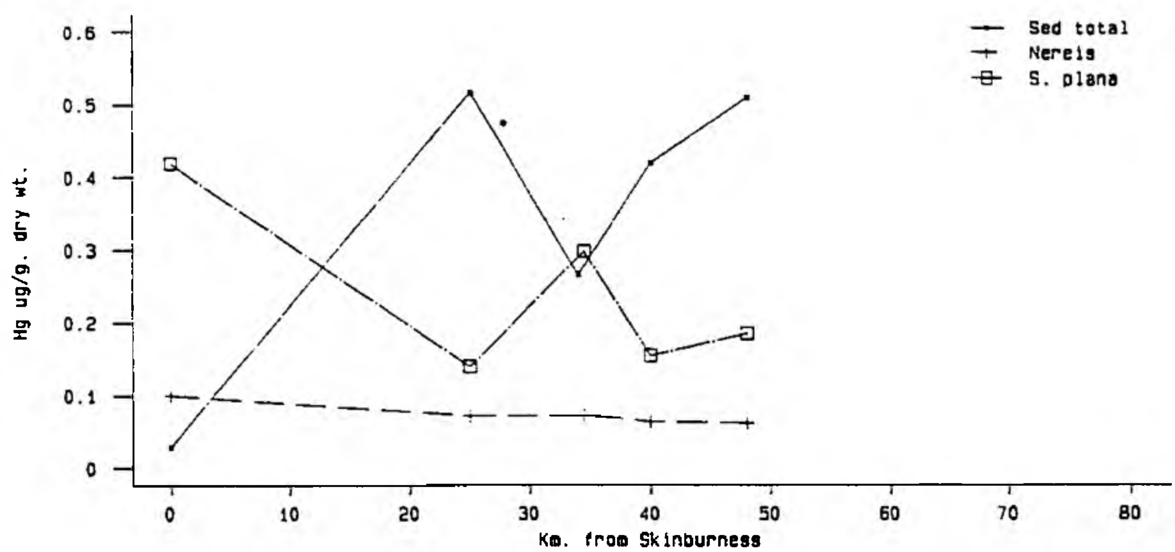


Fig. 28 Mercury in *Littorina littorea*, Cumbria 1990.

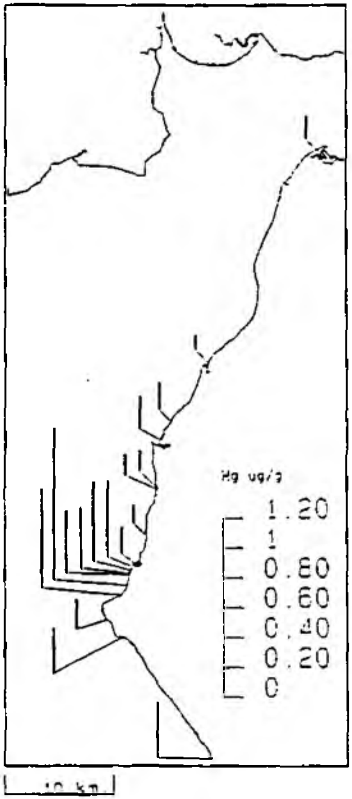
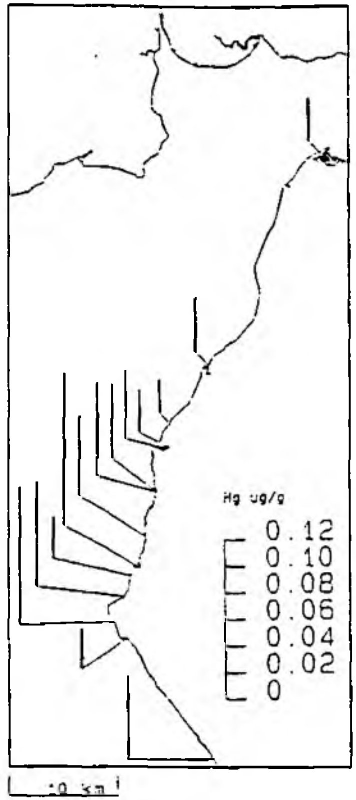


Fig. 29 Mercury in *Fucus vesiculosus*, Cumbria 1990.



Manganese

Data for Mn concentrations in sediments and biota is presented in Appendix 8.

Experimental evidence suggests that Mn in *Fucus* generally reflects environmental levels (Bryan *et al.*, 1985). Results of the present survey show that there is a major peak for Mn in *Fucus* in the estuary at Workington (Figs. 30 and 31), although the data should be viewed with some caution in view of possible sediment contamination of *Fucus* fronds (see section on Fe). Nevertheless gradients for Mn in *Patella* (Figs. 30, 32) and to some extent *Littorina* also indicate increased Mn levels along this part of the coastline. (Individuals of these two species were not found at Workington and peak concentrations occur further north).

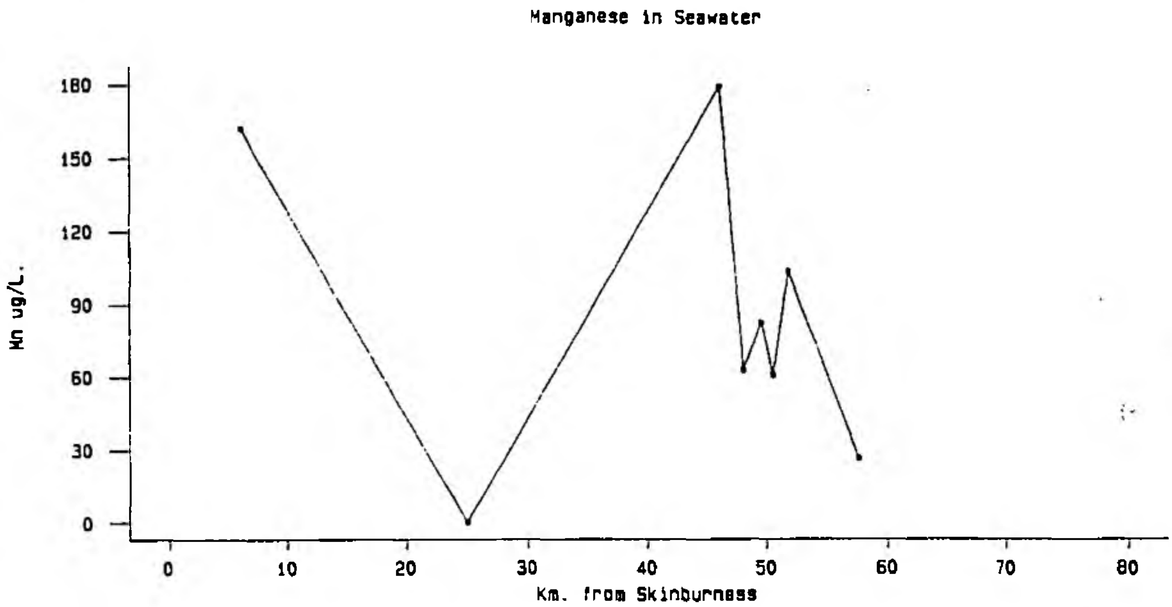
The ability to assess Mn contamination, using biological indicators, varies greatly depending on the selected organism. Thus, whilst *Fucus* and *Patella* respond noticeably, relationships between Mn in tissues and environmental levels are of only marginal significance for most other species. Consequently the enrichment factors (Max/Min concentration) measured in the present survey span a broad range, depending on the organism, from 1.65 in *Nucella* to 48.3 in *Patella*.

There is little evidence of Mn enrichment in biota from Saltom Bay, which is unusual in comparison with the other metals studied. The presence of Mn contamination cannot be ruled out here however, since high levels of other metals, notably Zn, are known to suppress bioaccumulation of Mn.

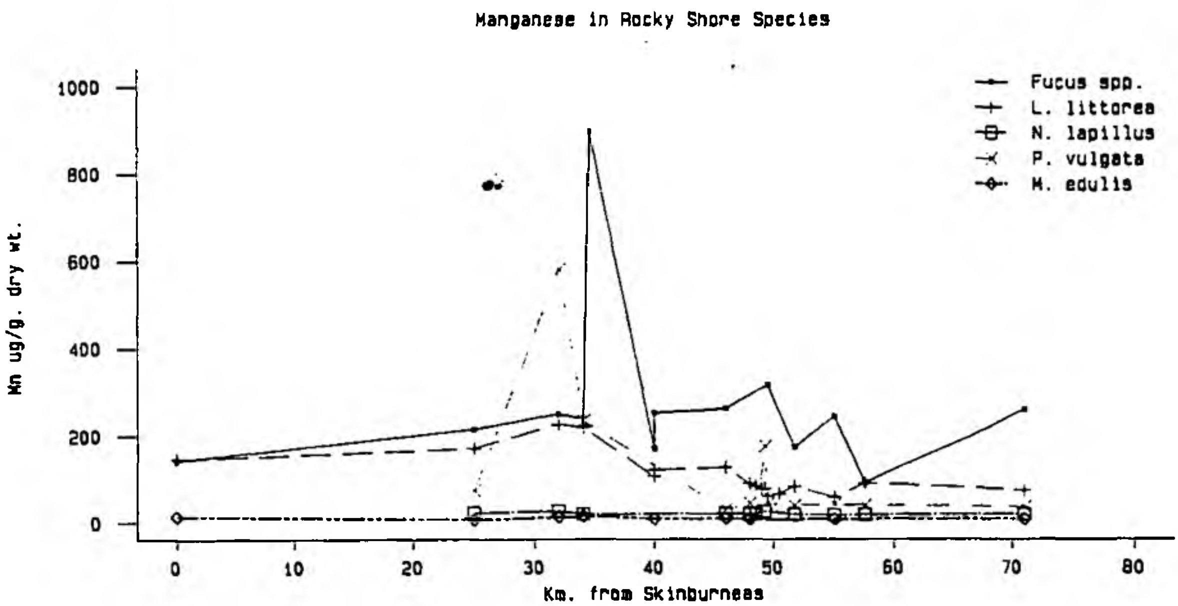
Sediment Mn concentrations at Workington and Maryport are amongst the highest recorded in UK estuaries. Most of the sediment-bound Mn is easily (lN-HCl) extractable (Appendix 8) suggesting the presence of anthropogenic inputs and/or a strong diagenetic influence: Thus as sediments become buried Mn is released from particulates under anoxic conditions. On migrating to the sediment surface, Mn is then re-oxidised and precipitated, often leading to high concentrations in surface sediments - even at sites considered relatively uncontaminated. Consequently even at such pristine sites organisms can sometimes be exposed to relatively high Mn concentrations, even at sites which do not receive Mn inputs.

For whatever reason Mn gradients in sediments (Fig. 30) are somewhat different to that observed for most other metals (with the highest concentrations occurring at Workington and Maryport, instead of Whitehaven). In general Mn concentrations in *Nereis* and *Scrobicularia* reflect this gradient (maximum concentrations at Maryport), though worms are clearly less efficient accumulators of Mn than clams. Particulate Mn concentrations thus appear to be a predominant feature governing tissue loadings in infaunal species, though other factors such as salinity and redox may well be of significance in modifying bioavailability.

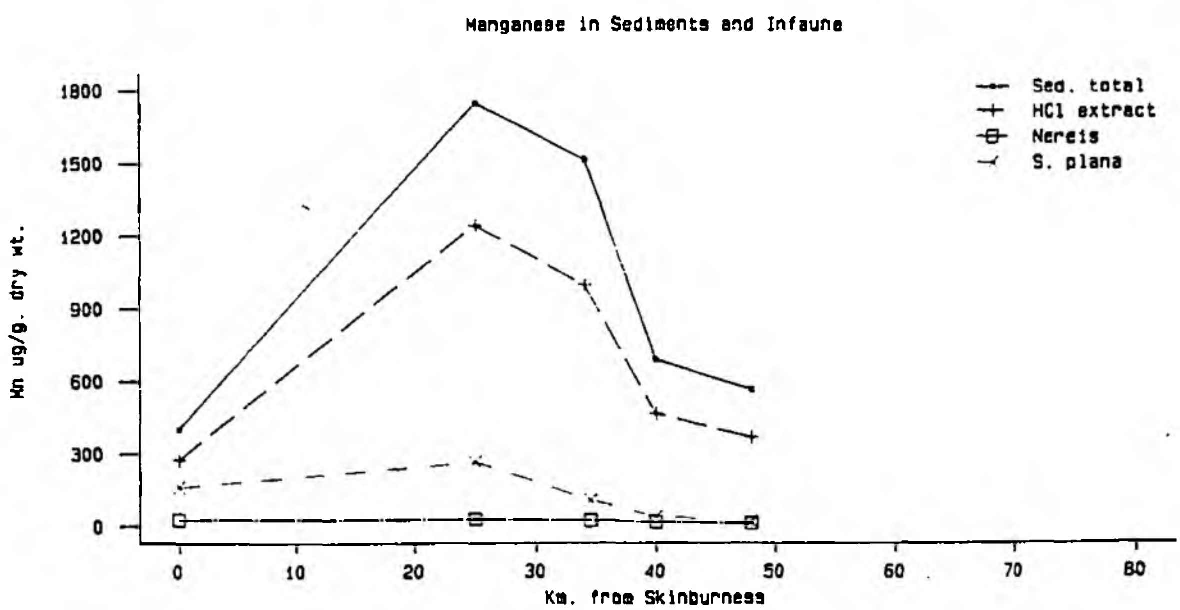
Fig. 30



Manganese in Seawater
Mn ug/L.
180
150
120
90
60
30
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness



Manganese in Rocky Shore Species
Fucus spp.
L. littorea
N. lapillus
P. vulgata
M. edulis
Mn ug/g. dry wt.
1000
800
600
400
200
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness



Manganese in Sediments and Infauna
Sed. total
HCl extract
Nereis
S. plana
Mn ug/g. dry wt.
1800
1500
1200
900
600
300
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

Fig. 31 Manganese in Fucus vesiculosus, Cumbria 1990.

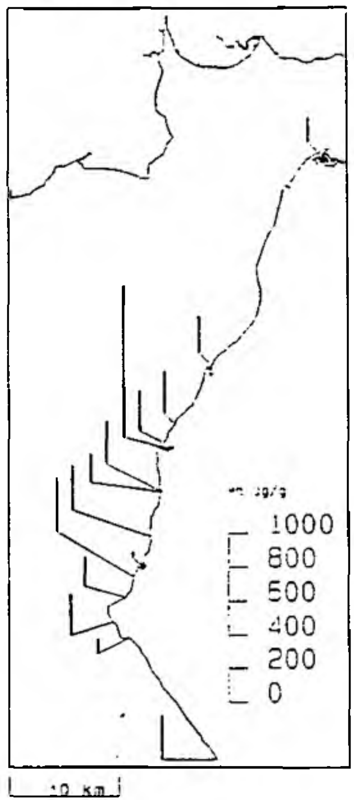
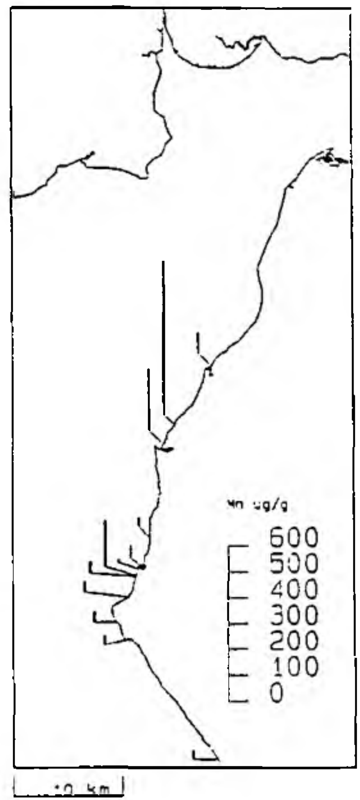


Fig. 32 Manganese in Patella vulgata, Cumbria 1990.



Nickel

Data for Ni concentrations in biota and sediments are presented in Appendix 9. No samples of *Patella vulgata* were analysed for this metal due to problems with acid digestion.

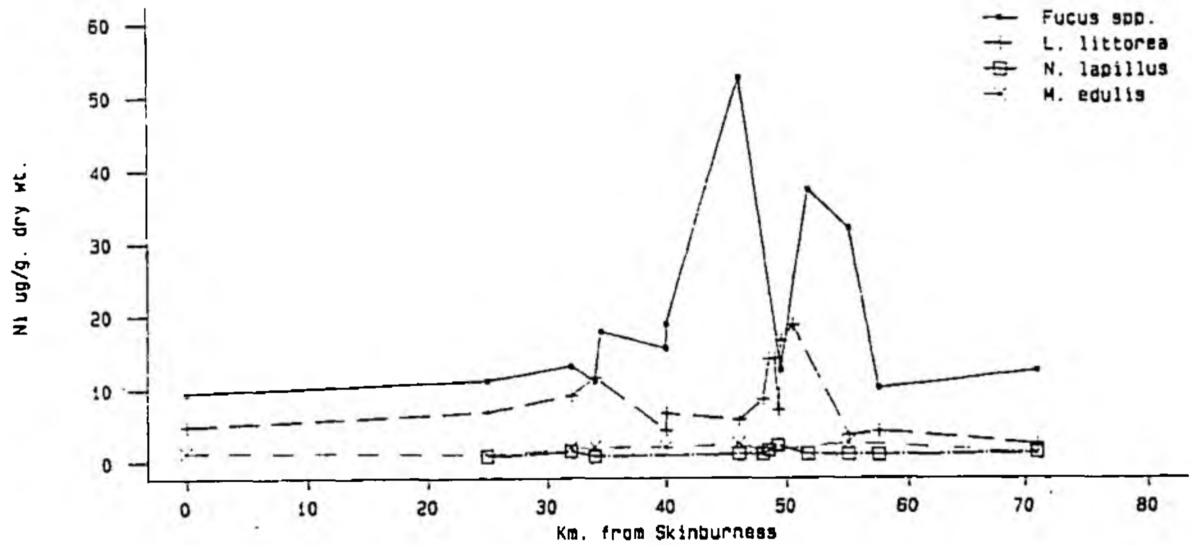
Nickel concentrations in biota from UK estuaries rarely exceed baseline concentrations by more than an order of magnitude (Appendix 9). At the least contaminated Cumbrian sites, Ni concentrations in most species are equivalent to, or are just above minimum values recorded in the UK, and in *Mytilus* and *Nucella* these tissue burdens increase only by a factor of 2-3 even at the most polluted sites. In contrast Ni concentrations in *Littorina* and *Fucus* at impacted Cumbrian sites are 8 and 17 - fold higher than baseline concentrations, and are among the highest recorded values for these species. Consequently gradients in Ni contamination are best portrayed using *Littorina* and *Fucus* as indicators (Figs. 33-35). Saltom Bay sites again appear to be most contaminated, with *Littorina* data suggesting the "Marchon" outfall maybe the major source. However there is clearly some variation between species regarding the location of maximum Ni concentrations. Studies on the interactions between metals in *Fucus* have shown that high levels of Zn (or Mn) in water can markedly suppress accumulation of Ni (and several other metals, Bryan *et al.*, 1985). It is possible that such interactions may be important in Saltom Bay, resulting in a more complex picture of contamination than would be predicted if inputs of single metals were occurring.

By UK standards, total Ni concentrations in sediments may be considered to be relatively high (appendix 9). However only 25% (or less) of the total Ni is readily available to infauna.

There is a general decrease in sediment Ni concentrations northwards towards Skinburness (Fig. 33), in common with most metals. Tissue residues in clams and worms do not mirror this gradient, however, suggesting that other, as yet unidentified factors may be modifying the bioavailability of sediment-bound Ni.

Fig. 33

Nickel in Rocky Shore Species



Nickel in Sediments and Infauna

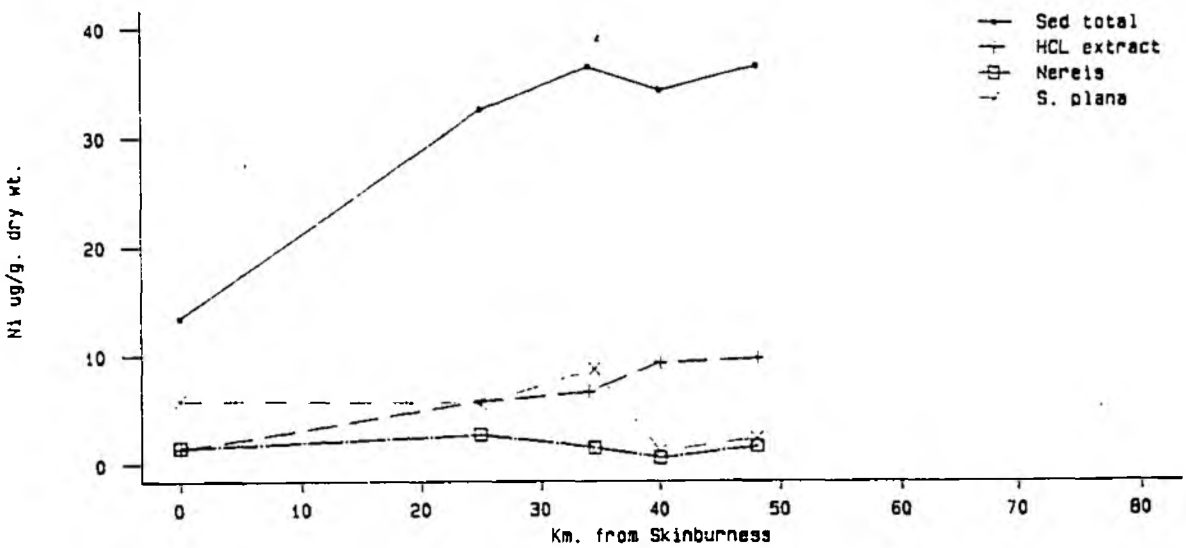


Fig. 34 Nickel in *Littorina littorea*, Cumbria 1990.

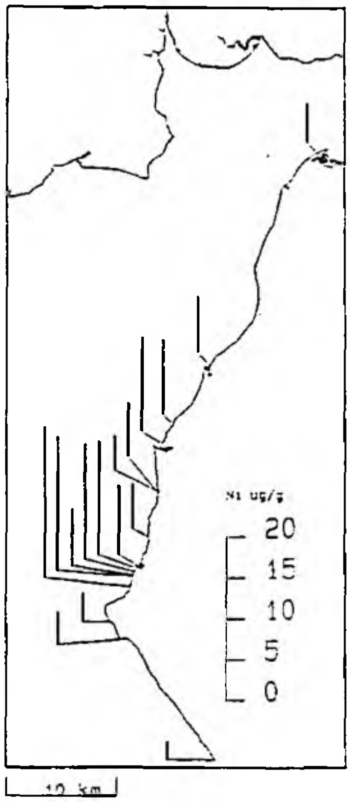
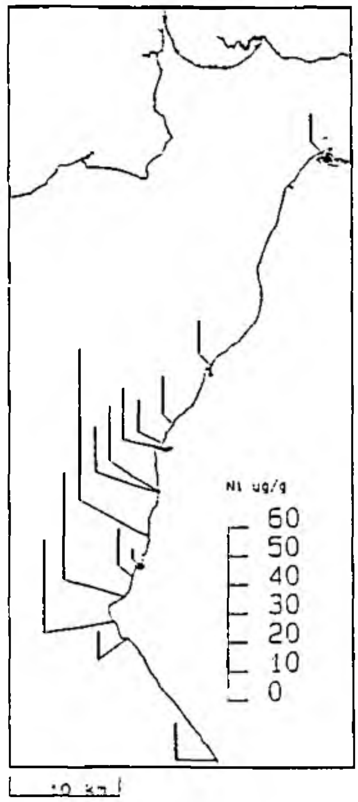


Fig. 35 Nickel in *Fucus vesiculosus*, Cumbria 1990.



Lead

Data for Pb concentrations in biota and sediments are presented in Appendix 10.

The affinity of *Fucus* for dissolved Pb is relatively high and concentrations in seaweed generally approach proportionality with Pb levels in water, making this species a good indicator of water-borne contamination. However inorganic Pb tends to be scavenged readily by particulates and it is rare for Pb levels in *Fucus* to reach equivalent concentrations to those found in many invertebrates. In the present survey unusually high Pb concentration (by UK standards, see Appendix 10) were found in *Fucus* collected at Harrington and in the estuary at Workington (Figs. 36, 37). Possible explanations for this are: i) some absorption of Pb from particulates by polyphenolic compounds present in *Fucus*. However, although this is thought to be a likely mechanism for Pb uptake at some estuarine sites (Luoma, *et al.*, 1982) it is difficult to explain why Pb concentrations differ so much in plants collected form Harrington and Whitehaven, where Pb concentrations in sediments are virtually identical. ii) Enhanced levels of bioavailable Pb in the water column. This seems a more probable explanation for the current results, although the precise form of Pb has not been measured. Previously however, anomalously high Pb levels in *Fucus* have been reported for the Mersey estuary which are almost certainly due to the presence of enhanced levels of hydrophilic alkyl-leads. It is possible that high levels of organolead compounds are present at the above sites, though this hypothesis together with determination of the origins (anthropogenic or natural) of such compounds requires further study.

Mussels, *Mytilus edulis* are likely to absorb metals from solution and suspended perticulates, and are generally considered to be useful indicators for Pb (Schultz-Baldes *et al.*, 1983), as are winkles, *Littorina littorea* (Bryan *et al.*, 1985). Profiles of Pb in both these molluscs closely resemble that observed in *Fucus*, although mussels appear to be better accumulators of Pb than winkles. Maximum values are again centred on Harrington (Figs. 36, 38, 39).

The other rocky shore species sampled, *Nucella* and *Patella* were not found at Harrington, or in the estuary at Workington and thus the range of Pb concentrations is relatively low (3.7 and 4.5 respectively). Both *Nucella* and *Patella* are nevertheless considered to be acceptable indicators for Pb.

There is a smaller peak in Pb concentrations in biota collected just south of Whitehaven (Fig. 36), although this peak does not coincide with the "Marchon" outfall, and may possibly be due to sewage inputs in this area.

Background Pb concentrations in nearly all species sampled are similar to UK baseline values. Even though Pb enrichment is striking at several sites, (described above), for most invertebrates these concentrations do not represent exceptionally high values, compared with sites in south-west England which are impacted by mine wastes (Appendix 10). The high Pb concentrations in *Fucus* at a number of Cumbrian sites are unusual, however, confirming that water soluble compounds, possibly hydrophilic alkyl leads, are of biological significance. Evaluation of Pb speciation in organisms, water and sediment would be of particular value in future surveys.

The distribution of Pb in estuarine sediments along the Cumbrian coast is similar to that for most other metals, and concentrations are lowest at Skinburness (Fig. 36). Inorganic Pb is readily scavenged by iron oxyhydroxide coatings on sediment particles - consequently much of the Pb (66-100^{o}/o) is readily extractable in IN HCl, and Pb and Fe concentrations correlate significantly in these extracts. However, compared with estuaries impacted with metal-mining wastes, Pb levels in Cumbrian sediments are low.

Previous studies have shown that Pb burdens in tissues of *Nereis* *diversicolor* are related to surface sediment loadings, and this is generally the case in the present study, even though the amounts of Pb accumulated are very low (Fig. 36).

Surface sediments are also thought to be the principal source of Pb in *S. plana*. However the best relationships are obtained by taking into account the influence of the major metal-binding component - namely Fe oxyhydroxide. Thus, Luoma and Bryan (1978) found that concentrations of Pb in clams were related not to total sediment - Pb levels, but to the Pb:Fe ratios in IN HCl extracts of surface sediment. High levels of Fe in sediments are thought to increase the binding capacity for Pb, reducing bioavailability (see also arsenic). Conversely low levels of extractable Fe in sediments may result in a relatively higher proportion of available lead. This may explain the anomalously high levels of Pb in Skinburness clams, relative to sediment Pb (Fig. 36).

Overall, the amounts of bioavailable Pb in Cumbrian sediments, as reflected in tissue burdens in clams and worms, is small compared with the most contaminated UK estuaries (Appendix 10).

Fig. 36

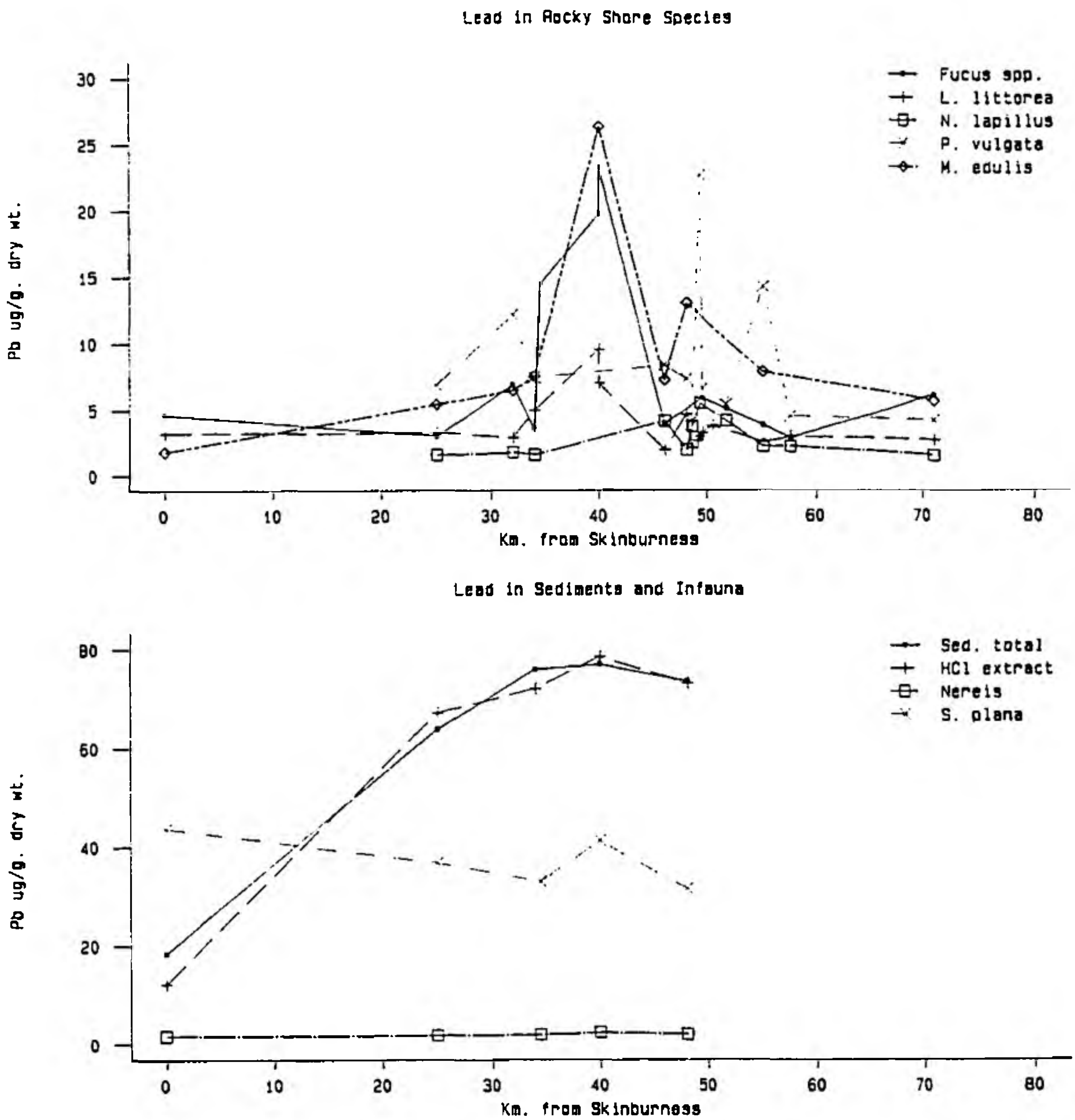

Lead in Rocky Shore Species
Fucus spp.
L. littorea
N. lapillus
P. vulgata
M. edulis
Pb ug/g. dry wt.
30
25
20
15
10
5
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness
Lead in Sediments and Infauna
Sed. total
HCl extract
Nereis
S. plana
Pb ug/g. dry wt.
80
60
40
20
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

Fig. 37 Lead in *Fucus vesiculosus*, Cumbria 1990.

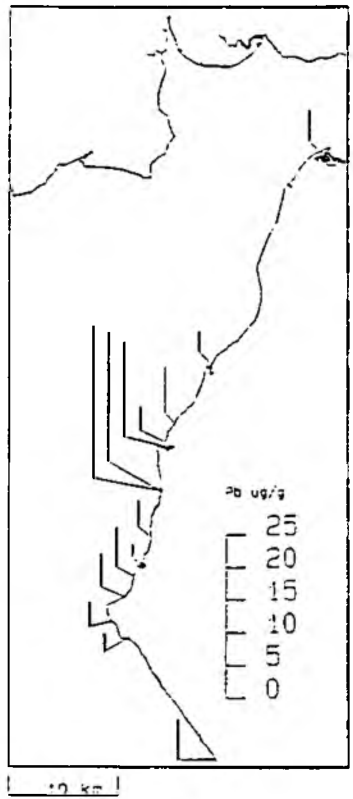


Fig. 38 Lead in *Mytilus edulis*, Cumbria 1990.

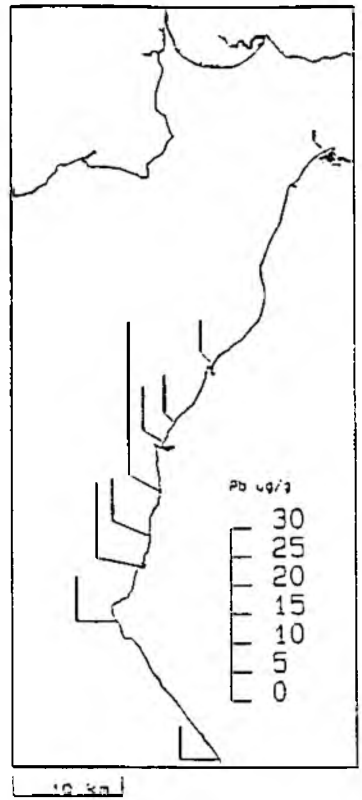
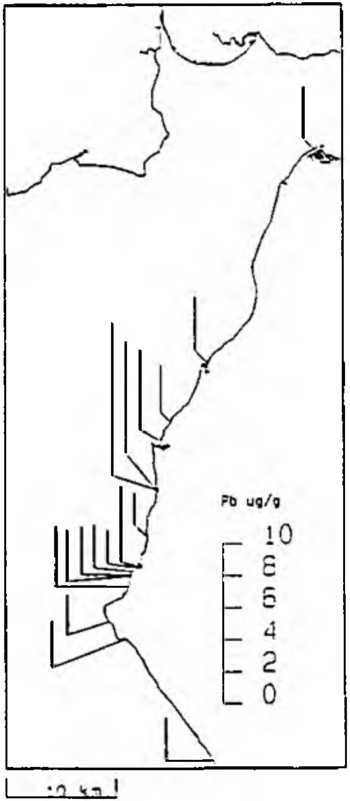


Fig. 39 Lead in Littorina littorea. Cumbria 1990.



Tin

Data for Sn concentrations in biota and sediments are presented in Appendix 11.

Inorganic tin in seawater is present as $SnO(OH_3)^-$ and as such is relatively insoluble. Bioaccumulation of this form of tin is therefore insignificant, except near to discharges from, for example, tin plate manufacture. In the present study very low Sn concentrations were measured in most species towards the northern and southern limits of collection. These tissue levels correspond to background values for the UK (Appendix 11). Much higher concentrations were measured at some sites in the central part of the study area, although the locations of maximum Sn concentrations were variable depending on the distribution of the species (Fig. 40).

Generally, highest Sn values were detected at harbour/estuarine sites (Appendix 11), indicating that the origin of contamination may be the relatively highly bioavailable organotins, including TBT (tributyltin), derived from antifouling paints used on boats. Tributyltin was widely used as such until 1987, when restrictions on usage (on vessels smaller than 25m) were introduced, following concern about toxicity to non-target organisms (see below)*

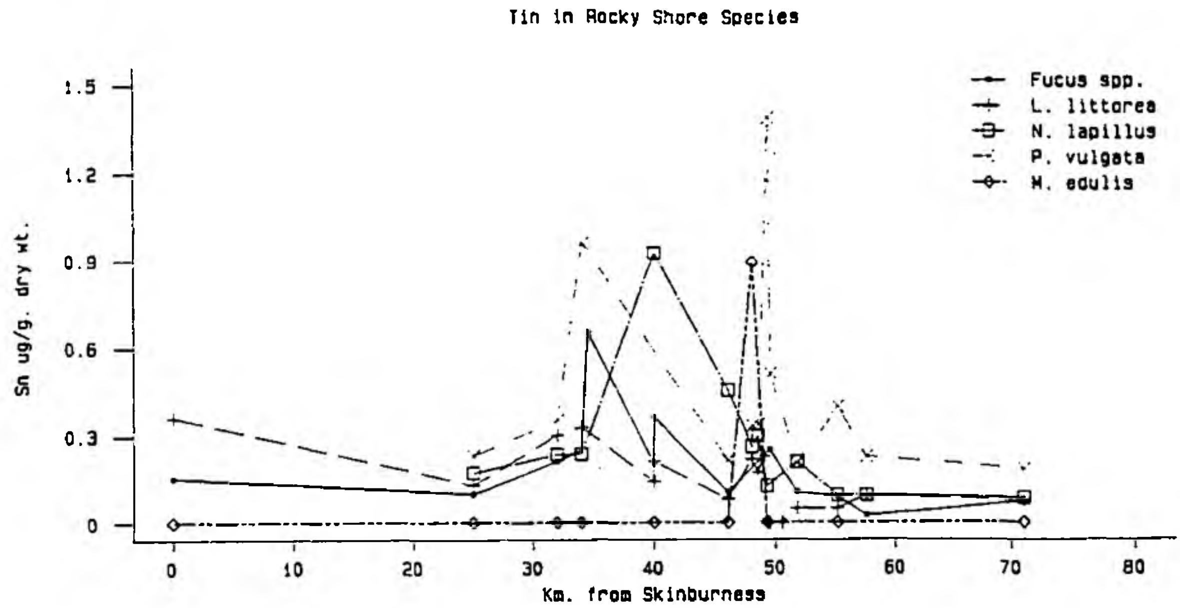
Despite the higher levels of Sn in harbours and estuaries along the Cumbrian coast, maximum Sn concentrations were generally well below corresponding UK maxima (Appendix 11). This is not surprising since the level of boating activity and the density of vessels is generally much lower than found, for example, at sites along the south coast of England, notably those bordering the Solent.

Sediment Sn concentrations, though displaying the expected north-south gradient (Fig. 40), were also low in comparison with cassiterite (SnO_2 contaminated estuaries in south west England (Appendix 11). Tin concentrations in clams and worms mirror the sediment gradient to some extent (Fig. 40), however, it is likely that tissue burdens are largely a reflection of TBT contamination. Thus, despite the low proportion of TBT:total tin in sediments the former chemical species is by far the most bioavailable form (Langston, 1990).

* TBT-induced reproductive abnormalities, (imposex) in *Nucella lapillus* provide an extremely specific and sensitive means of monitoring the biological impact of this contaminant in coastal waters (Bryan *et al* 1986; Langston *et al*, 1990). Recognition of such effects contributed significantly to the restrictions placed on TBT-based paints, in order to achieve Environmental Quality Targets of 2 $ng.L^{-1}$ (recommended by the Department of the Environment).

The intensities of imposex, expressed as relative penis size (RPS) indices (see Bryan *et al* 1986 for explanation of this index), were measured in most of the *Nucella* populations collected in the present survey. Results (Table 2) clearly indicate some impact at a number of sites, predominantly those close to harbours and estuaries. Put in perspective however, the RPS values along the Cumbrian coast, even at worst (37%) are much lower than values from sites such as Yarmouth on the Isle of Wight (91%). Indeed there are many sites in the Solent where dogwhelk populations have been eliminated by TBT contamination (Langston *et al*, 1990), whereas there is little evidence of such widespread decline in Cumbrian populations based on current results.

Fig. 40


Tin in Rocky Shore Species
Fucus spp.
L. littorea
N. lapillus
P. vulgata
M. edulis
Sn ug/g. dry wt.
1.5
1.2
0.9
0.6
0.3
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

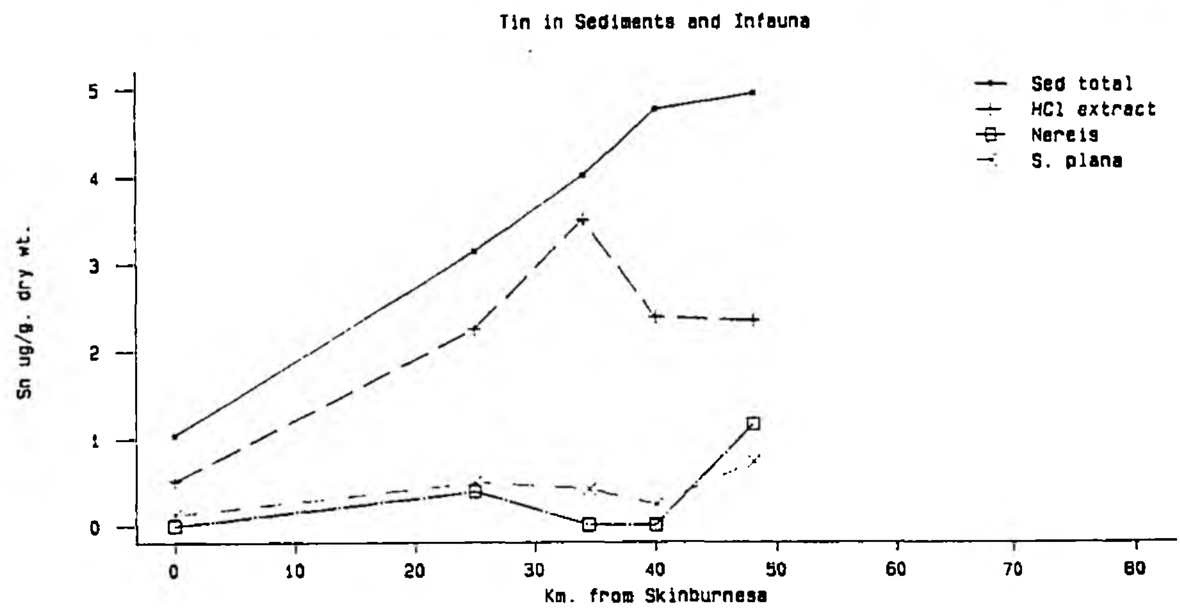

Tin in Sediments and Infauna
Sed total
HCl extract
Nereis
S. plana
Sn ug/g. dry wt.
5
4
3
2
1
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

Table 2 Incidence and degree of Imposex in *Nucella lapillus*, Cumbrian coast, 1990

Site	Number of Animals	% Females	RPS Index*
Maryport	19	57.1	12.8
St. Helens	10	60.0	36.8
Workington	19	73.7	21.8
Parton	20	60.0	3.76
Whitehaven Harbour	20	50.0	23.6
Tom Hurd's Rock	22	59.1	3.13
S. of Sewage Pipe	20	55.0	0.53
S. Limit of Outfall	20	40.0	0.53
St. Bees Head	20	35.0	2.13
St. Bees	20	55.0	1.24
Seascale	20	50.0	0.26

* RPS = $[(\text{mean female penis length})^3/(\text{mean male penis length})^3] \times 100$

Zinc

Data for Zn concentrations in biota and sediments are presented in Appendix 12.

Zinc concentrations in unfiltered seawater samples, collected along the Cumbrian coast, are dominated by a single input coinciding with the "Marchon" outfall (Fig. 41). Smaller inputs at other sites cannot be ruled out however, in view of the relatively small number of water samples taken.

Since the seaweed *Fucus vesiculosus* is a relatively good indicator of dissolved Zn, at least at low-moderate levels (concentration factors may be lower at high Zn concentrations), it is not surprising that Zn profiles in *Fucus* generally reflect those in water described above (Figs 41 & 42). Significantly elevated Zn concentrations were measured in *Fucus* at most sites between Workington and St.Bees Head, however (Fig. 42), suggesting fairly widespread contamination along this part of the coast. Possible sources of Zn are indicated at Parton, and in Workington estuary in addition to the major input to Saltom Bay. More intensive sampling would be required to confirm this feature.

Nucella lapillus appears to be an excellent accumulator of Zn relative to other species. Furthermore, concentrations of Zn in *Nucella* clearly mimic gradients observed for water, and seaweed, *Fucus vesiculosus* (Fig. 41).

The majority of rocky shore molluscs are thought to regulate Zn (to varying degrees) and hence tissue analyses probably underestimate high levels of Zn contamination. This is most obvious for Zn profiles in *Littorina*, *Mytilus* and *Patella* (Fig. 41), where slight increases in Zn burdens were observed only at sites closest to the major Zn sources. Consequently, the range of Zn concentrations (Max/Min) encountered in the current survey were 2.3, 2.3, & 3.1 for each of these species respectively. This compares with an equivalent enrichment factor of 16.1 in the more suitable indicator *Fucus*.

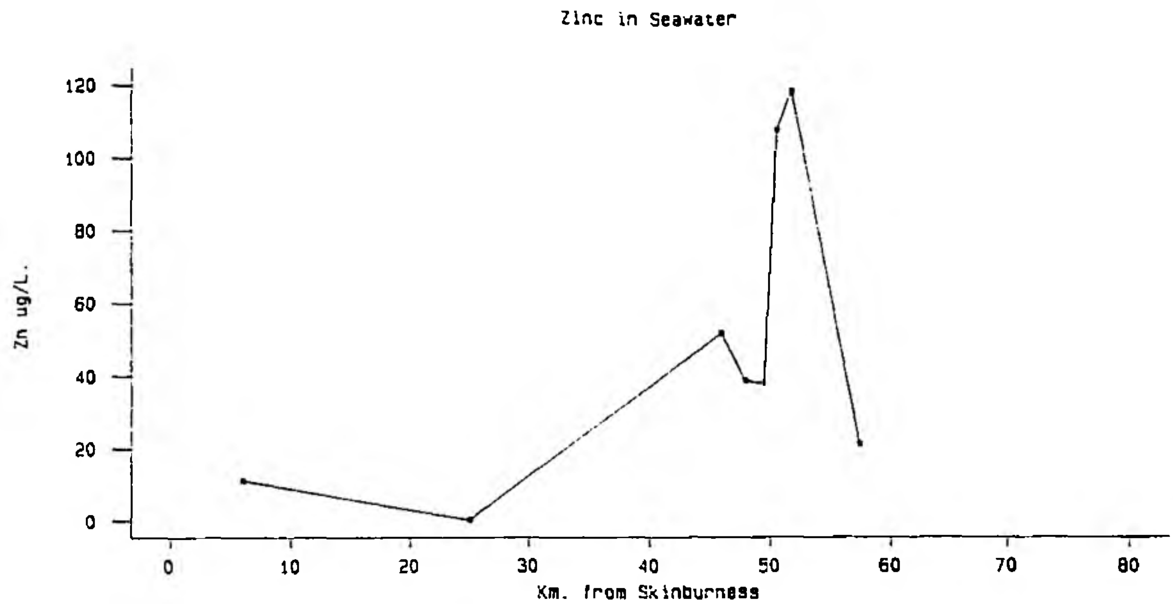
For the majority of species, Zn concentrations at the most heavily contaminated Cumbrian sites approach, though generally do not exceed UK maxima. *Nucella* is the only species in the current survey in which Zn concentrations are in excess of previously recorded upper limits, albeit by a small margin. It is perhaps relevent to note however, that Zn concentrations in individual dogwhelks may be highly variable and could be dependent on the type of diet (considered by Young (1977) to be the most important source of Zn in this carnivorous gastropod).

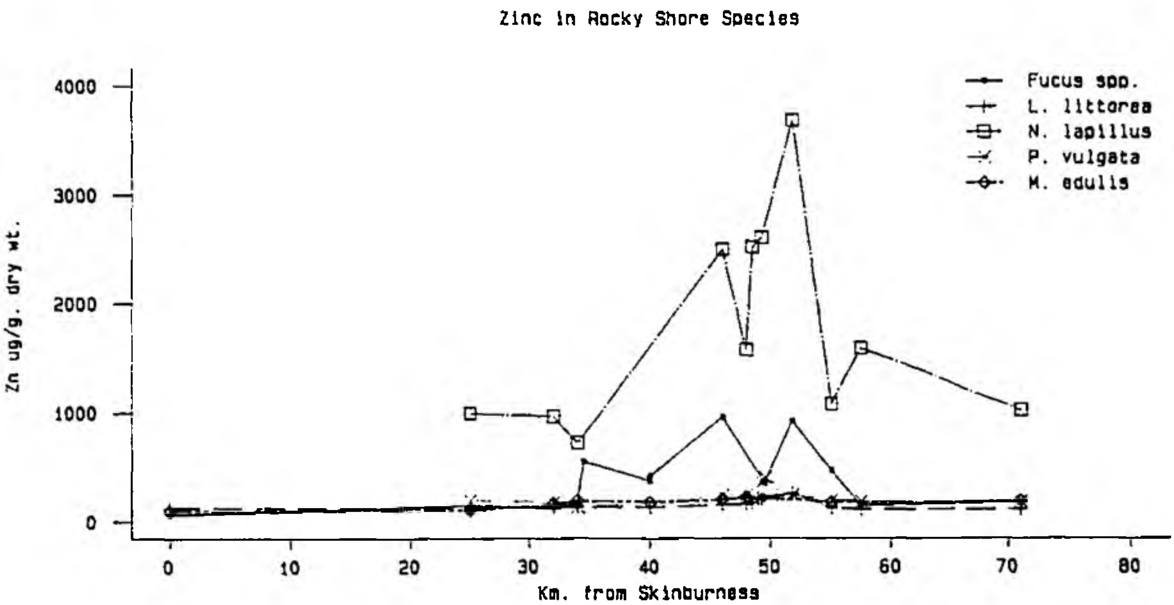
Zinc, like Cd has a greater tendency to remain in solution than many other metals. Hence sediment Zn concentrations at the sites sampled are generally low compared for example, to estuaries in south west England which are heavily impacted by metal-mining wastes (Appendix 12). Nevertheless, slight gradients are observed in the present survey (Fig. 41) which resemble the north-south trends described above for other metals. Usually more than 50% of the total Zn in Cumbrian sediments is readily extractable (1N-HCl) and therefore is probably anthropogenic in origin.

Zinc concentrations in *Nereis diversicolor* are relatively unaffected by environmental change (Bryan *et al*, 1985) and do not meaningfully indicate contamination gradients in the present study (Appendix 12, Fig. 41). Similarly it is likely that clams *Scrobicularia plana*, have some capacity for regulating Zn, although as with Cd it seems probable that some accumulation may occur from water as well as sediment. The present survey indicates elevated Zn levels in clams relative to UK baseline values, with maximum enhancement (2 - 3 fold) at Workington harbour, (see also *Fucus*).

Clearly the infaunal species analysed are much less valuable in detecting Zn inputs to Cumbrian coastal waters than *Fucus* or *Nucella*. The ability to regulate this metal, particularly in *Nereis diversicolor*, restricts their use,

Fig. 41


Zinc in Seawater
Zn ug/L.
120
100
80
60
40
20
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness


Zinc in Rocky Shore Species
Fucus spp.
L. littorea
N. lapillus
P. vulgata
M. edulis
Zn ug/g. dry wt.
4000
3000
2000
1000
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

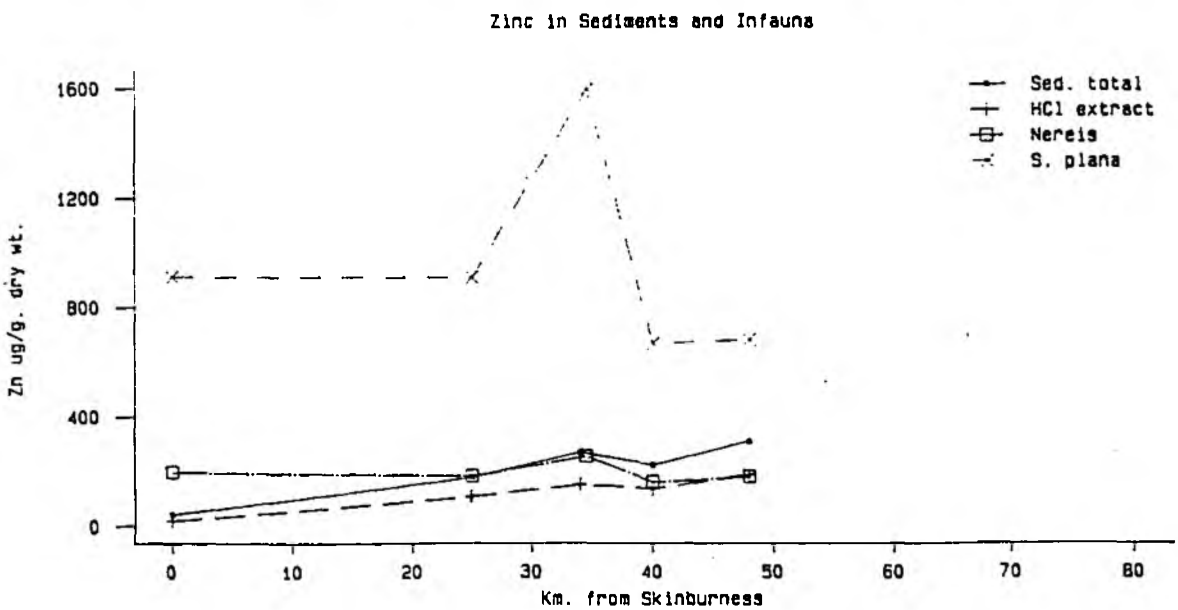
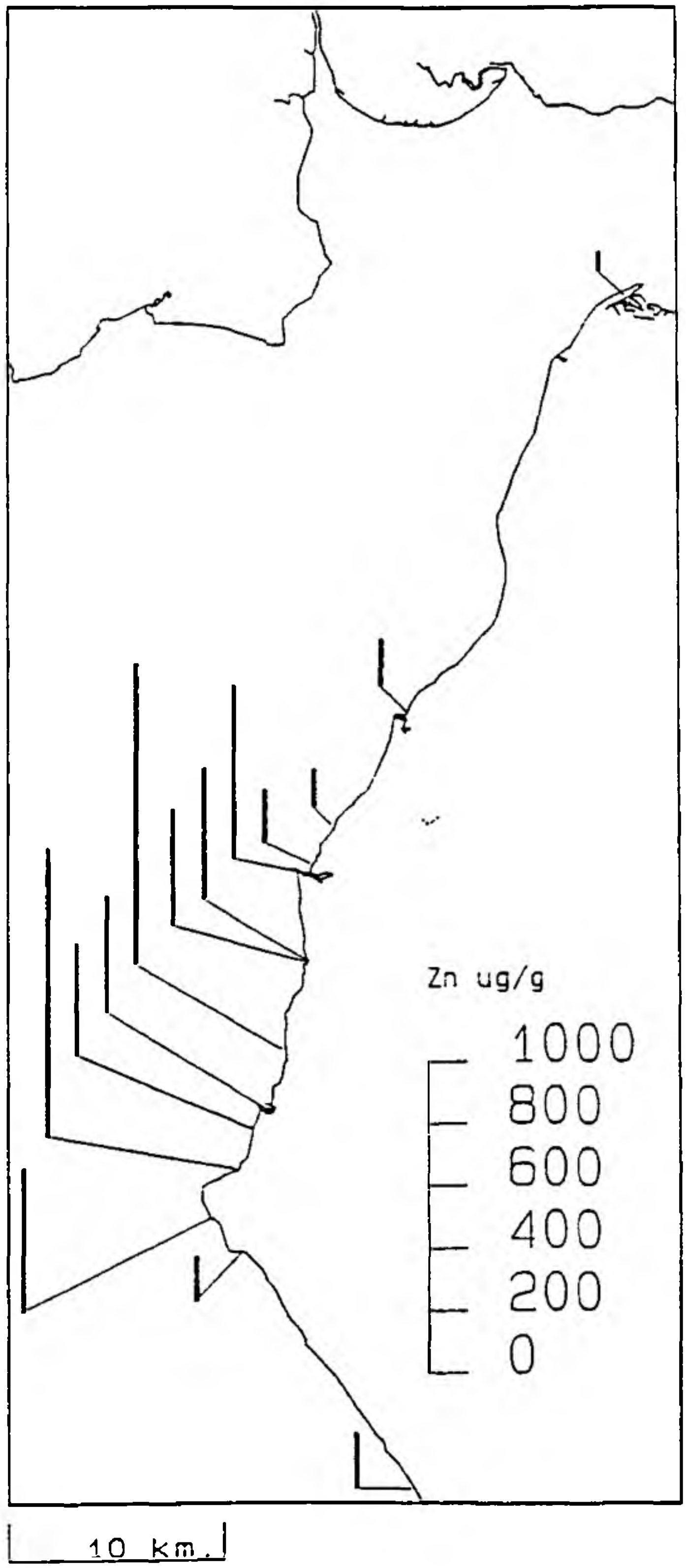

Zinc in Sediments and Infauna
Sed. total
HCl extract
Nereis
S. plana
Zn ug/g. dry wt.
1600
1200
800
400
0
0
10
20
30
40
50
60
70
80
Km. from Skinburness

Fig. 42 Zinc in Fucus vesiculosus, Cumbria 1990.



as indicators for Zn, to estuaries where there are major differences in contamination.

Discussion

The north west Cumbrian coastline may be considered as the outer part of the Solway Firth, a large coastal plain estuary, tributary to the north east Irish Sea. The Solway has little direct input from industry except in the portion of coastline lying between Maryport and St Bees Head (Perkins, 1977). During the Industrial Revolution the coastline was very important to the iron and steel industry, the evidence of which can still be seen as spoil heaps and other items of industrial waste, such as slagcrete (the remnants of blast furnace operation), dumped on the shore at certain sites. In addition there have in the past been localised inputs from acid mine-drainage and tar distilleries.

Recently, attention has focussed on the impact on water quality caused by industrial operations along the shoreline between Whitehaven and St Bees Head. Saltom Bay has, until the last few years, received considerable wastes from coal mining activities (initiated some 250 years ago): As recently as the last decade, much of the intertidal zone was covered with colliery wastes and washings, though the action of the sea has eroded and dispersed much of this material following the reduction in active mining. Sewage is also discharged from a pipe running over the shore approximately 1 km south of Whitehaven Harbour. However the major concern is the discharge from a chemical factory sited on the southern outskirts of Whitehaven.

The Marchon plant has over the last 30 years been involved with phosphoric acid production, for use in on-site fertilizer and detergent (sodium tripolyphosphate) manufacture. The production of phosphoric acid involves the reaction of calcium phosphate and sulphuric acid to produce orthophosphoric acid and the by-product calcium sulphate. The plant releases the majority of wastes from all its production lines through a single effluent pipe directly to the sea at Barrowmouth, although a much smaller input is known to occur some 750m to the south west of the main discharge. This is probably due to seepage of waste from older drainage sections of the plant (originally owned by the predecessors of the National Coal Board) and, ultimately, emergence on the shore in a small stream draining from a rockfault. In the earliest days of the plants operation, effluent spilled directly over the cliff tops resulting in a delta of gypsum extending for several hundred metres (Perkins, 1981). This phenomenon has now disappeared following the installation of the pipeline, which discharges in the littoral zone.

The quality of the effluent depends on the quality and source of the initial phosphate ore, together with the chosen end point of production. The principal components however are phosphogypsum (calcium sulphate), phosphoric and sulphuric acids, detergents, fluorides and heavy metals (most importantly Cd, Cr, Cu and Zn). The effluent has a low pH (2.3-4.7) and its toxicity, noted by the absence of biota around the outfall has often been attributed to this factor (Abbot and Perkins, 1979). However the levels of heavy metals are also of concern particularly as they may be accumulated by marine organisms - hence the reason for the present survey.

Water

A limited number of (unfiltered) water samples were analysed for metals and phosphate, which provide a "snapshot" of contaminant levels. The data are in general agreement with earlier water quality studies based largely on measurements of nutrients pH BOD etc. (Perkins, 1981). Of the elements measured, elevated concentrations of As, Cu, Cd and Zn were found predominantly between Parton and St Bees, covering a distance of 6.5 km, with highest levels centred around the Marchon outfall (Fig. 43). The direction of dispersion appears to have been slightly biased to the north at the time of the current study, coinciding with the general direction of longshore drift.

Confirmation of a common source for As, Cu, Cd and Zn - namely the effluent from the Marchon plant - is virtually established by the significant correlations between levels of these elements and phosphate concentrations in water (Table 3). Despite some evidence of Fe and Mn inputs from this source, concentrations in water are not so well correlated with other metals or phosphate. This may be partly due to a significant contribution from particulate material (relatively rich in Fe and Mn) in some of the unfiltered samples. The presence of other sources of Fe and Mn seems likely however, particularly from estuaries/rivers (including sediments) along the Cumbrian coastline. Thus, there are significant inverse correlations between concentrations of these two metals in water and corresponding salinity measurements ($r=0.77$, $P<0.05$ for Fe; $r=0.87$, $p<0.05$ for Mn). Peaks in Fe and Mn concentrations at Parton (46 km, Fig. 43), for example, are almost certainly due to the fresh water input from Distington Beck, together with its associated sewage content.

Table 3. Correlations between As, Cd, Cu, Zn and phosphate in water samples.

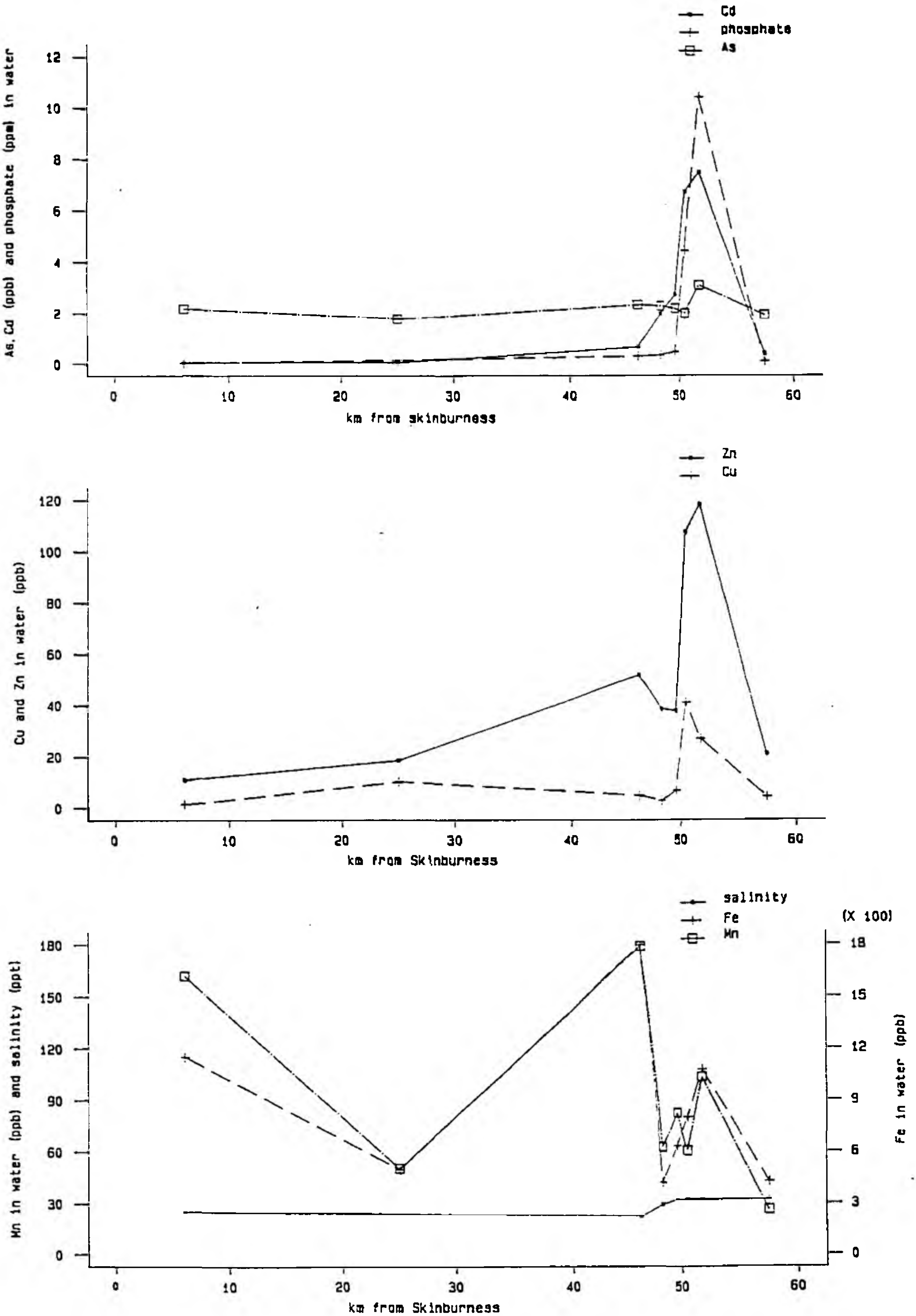
	PO_4	Cd	Cu	Zn	As
PO_4	-				
Cd	0.8946**	-			
Cu	0.7423*	0.9102**	-		
Zn	0.8915**	0.9519***	0.9056**	-	
As	0.7834*	0.5237	0.2163	0.5626	-

* significance level $P<0.05$, ** $P<0.01$, *** $P<0.001$

Rocky shore species - coastal sites

Fucoid seaweeds are good indicators of dissolved metals and have the advantage of integrating levels of contamination over time (days - weeks) compared with the more transitory signal obtained with water sampling and analysis. Copper, Cr and Ag concentrations in *Fucus*, collected along the Cumbrian coast, are clearly dominated by the single point source input, coinciding with the Marchon outfall (Fig. 44). To the south, concentrations of these metals decline sharply compared with those north of the outfall: residues in seaweed are still clearly elevated at Workington and arguably as far as Skinburness, suggesting a more gradual, though far-reaching dispersal of contaminants in this direction. Cadmium, Ni and Zn profiles in *Fucus* (Fig. 44) indicate a similar peak in concentrations in Saltom Bay, but also display a second peak at Parton. This suggests possible inputs of metals from Distington Beck, or that the low salinity water in this region may be enhancing the bioavailability of Cd, Ni and Zn originating from the Marchon outfall. Increased uptake of Cd and Zn at lower salinities has previously been demonstrated in experiments with *Fucus* (Bryan et al., 1985). Hg

Fig. 43 Metals and Phosphate in Seawater, Cumbria 1990.



concentrations in seaweeds were highest at Saltom Bay and Whitehaven harbour.

The profile for As concentrations in *Fucus* is interesting since, despite signs of a point source in Saltom Bay, the background trend is inverse to that observed for most metals (Fig. 44): There is a significant negative correlation between Cd and As, for example ($r=-0.69$, $P<0.01$). Arsenate uptake in algae is known to be inhibited by high levels of the chemically similar phosphate anion. It seems likely therefore that the overall inverse picture of As contamination relative to other metals, as depicted by *Fucus*, is due to the competitive effects between arsenate and phosphate.

Data for Fe, Mn, Pb and Sn in algae indicates that bioavailability of these metals is most significant at sites other than Saltom Bay. Pb and Sn are most enhanced at Harrington and Workington (estuary) respectively and may reflect sources of more biologically available organometallic species at these sites. Mn and Fe concentrations are also highest in *Fucus* from Workington though sediments may have a contributory role in these observations.

Metal concentration ranges in *Fucus* vary between 4.6 (Hg) and 120 (Fe). At the most impacted sites contamination with Ag, Fe, Cr, Ni, Mn and Pb is equivalent to current UK maxima, while at the other extreme, some of the lowest UK concentrations have been measured at Cumbrian sites (As, Ag, Cu, Fe, Cr, Hg, Mn and Sn), usually at the northern and southern limits of the collection range. Even at these sites however, Cd still appears to be threefold higher than baseline values for the UK; whether this is due to residual influences from the Whitehaven outfall or some other inputs is a subject for further study.

Metal profiles in *Littorina* are generally in good agreement with those described for *Fucus*, not surprisingly since the algal diet is probably a major source of metals for winkles. There are well defined peaks for most elements which (with the exception of Fe, Mn, Pb and Sn) coincide with inputs from the Marchon plant (Fig. 45). Profiles for Cr, Cu, Hg, Ni and Zn are all significantly correlated with that for Cd ($P<0.001$). For several metals (Ni, Cr, Sn, Hg and, most notably, Ag) there is a distinct peak in concentrations at Workington, while the Pb gradient is dominated by high concentrations around Harrington. Metal concentration ranges in *Littorina* vary, depending on the element, between 2.35 for Zn (which tends to be regulated) and 22.6 for Cr. At the most contaminated Cumbrian sites, Cr and Ni concentrations exceed UK maxima (by a factor of >20 for Cr), whilst at the cleanest sites, Cumbrian Littorinids have some of the lowest reported values for As, Fe, Hg and Sn. Significantly, however, Ag, Cr and Cd are considerably above UK baselines (by 18, 12 and 6-fold respectively) even at these 'pristine' sites.

The occurrence of *Patella vulgata*, *Nucella lapillus* and *Mytilus edulis* was found to be less extensive than that of *Fucus* or *Littorina*, particularly at the more contaminated sites between Whitehaven and St Bees. Nevertheless the profiles depicted in Figs. 46 and 47 clearly show the major impact of the Marchon outfall on tissue concentrations of Ag, Cd, Cu, Zn, Cr, Ni and to some extent As and Hg. Manganese, Fe Sn,Cr and Ni are elevated to varying degrees in mussels and limpets from Workington, whilst Pb concentrations in the former species confirm that bioavailability of this metal is most significant at Harrington.

At the most contaminated Cumbrian sites, concentrations of Ag (P,N,M*), Cd (N,M), Cu (P,N), Mn, Fe (P), Cr (M), and Zn (N) were equivalent to, or higher than (by upto 9 fold), previous uppermost values for these species. At the "cleanest" Cumbrian sites Ag, Cd (P,N,M), Cu (P,N), Cr and Zn (N,M) concentrations were also found to exceed UK minimum values by between 2 and 12 -fold.

Clearly, therefore, although metal accumulating ability varies appreciably, there are a number of reasonably consistent trends in metal

Fig. 44 Metals in Fucus, Cumbria 1990.

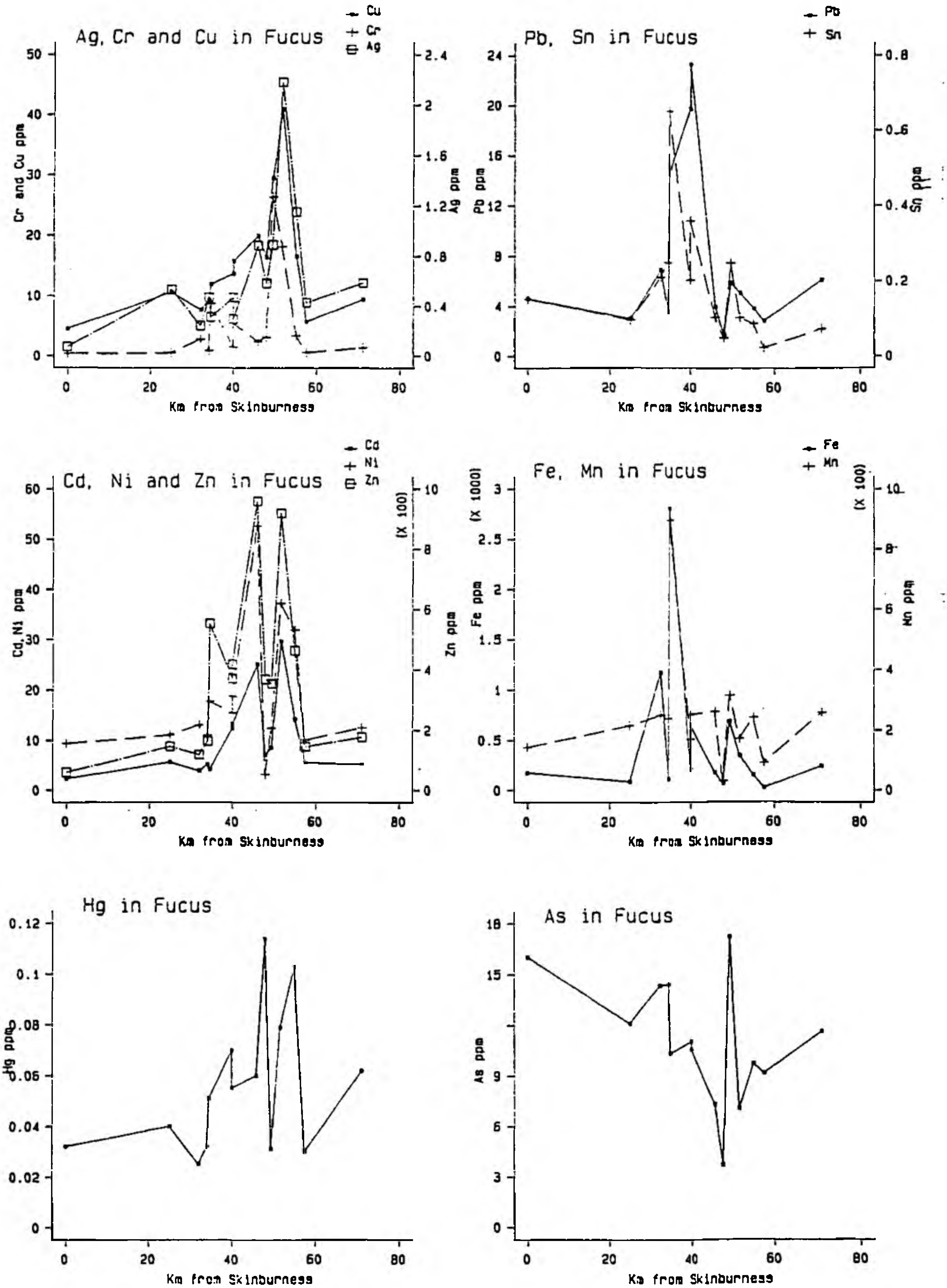


Fig. 45 Metals in Littorina, Cumbria 1990.

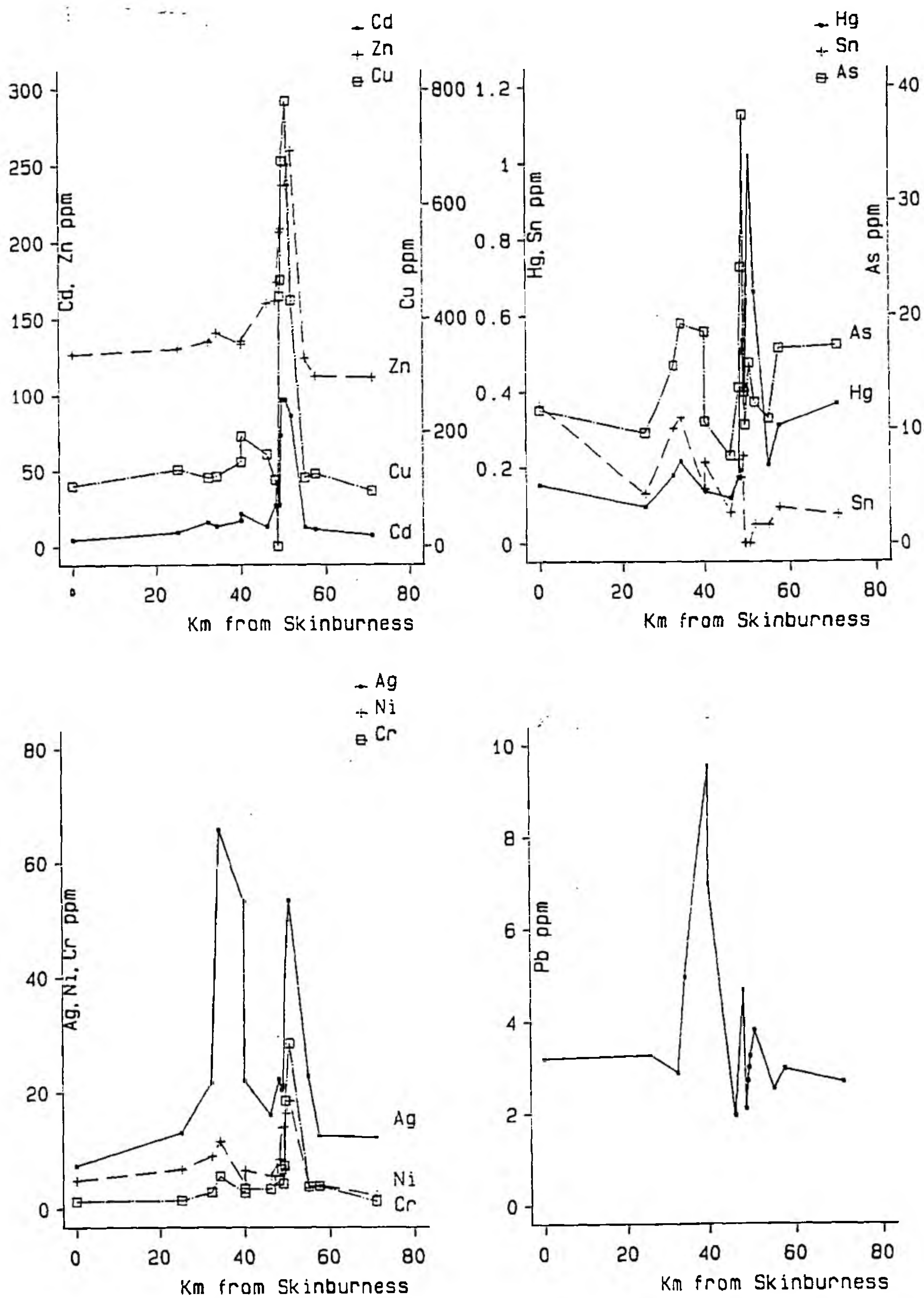


Fig. 46 Metals in Patella, Cumbria 1990.

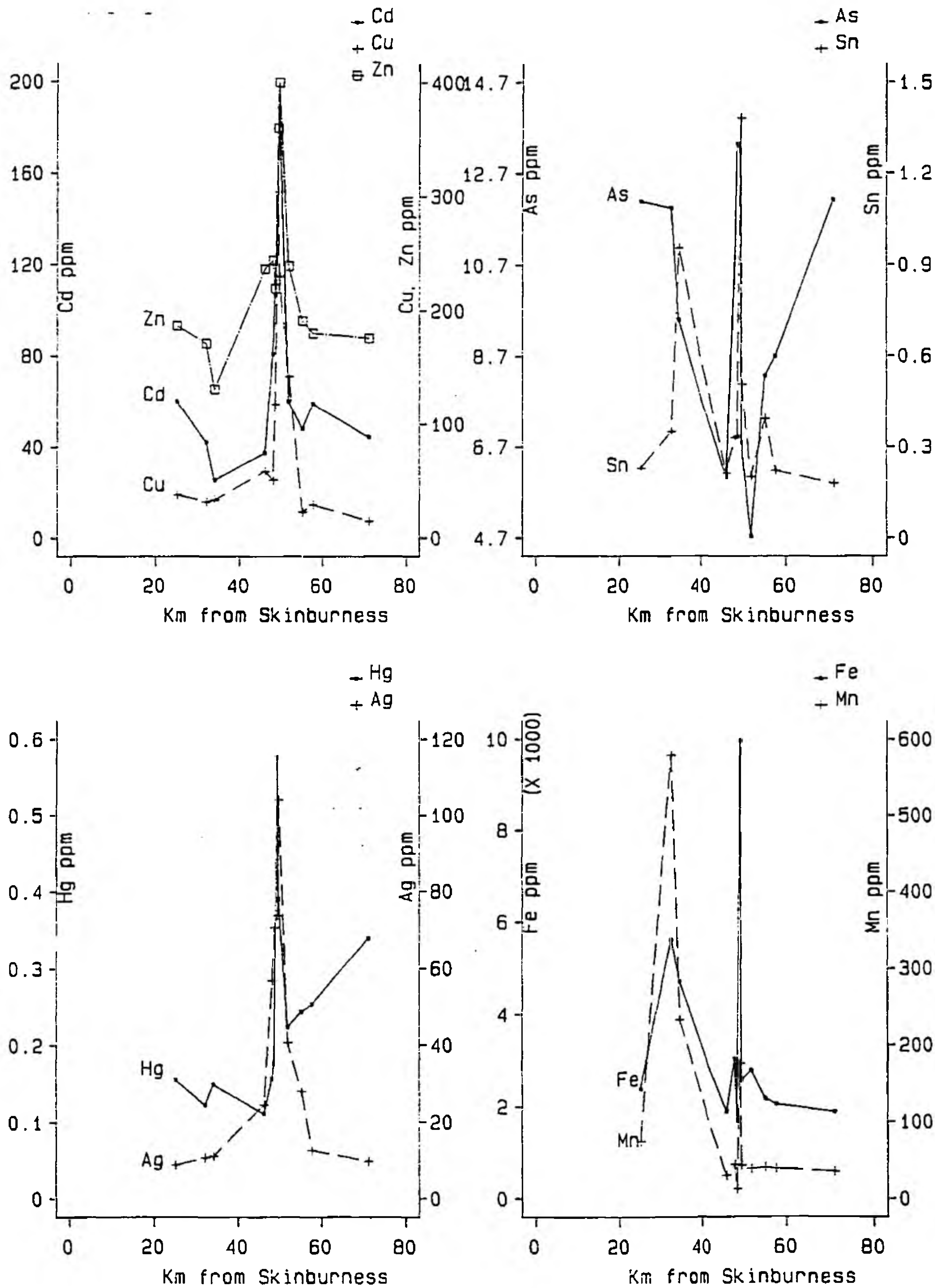
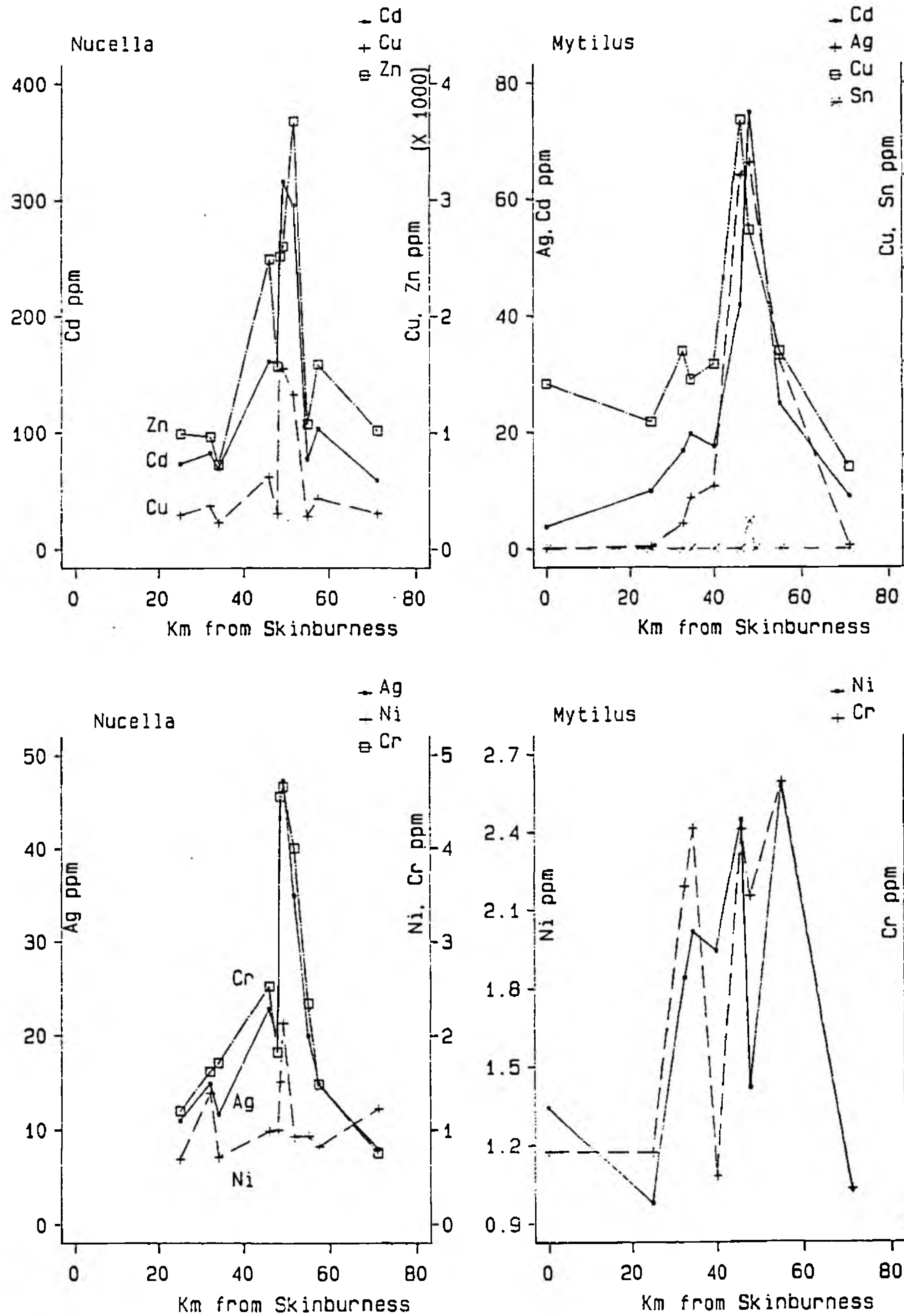


Fig. 47 Metals in Nucella and Mytilus, Cumbria 1990.



contamination patterns along the Cumbrian Coast as depicted by analyses of these rocky shore species.

* P - *Patella*, N - *Nucella*, M - *Mytilus*

Estuarine sediments and infauna

With the exception of Mn, metal concentrations in intertidal sediments decrease, overall, in a northerly direction for the five estuarine/harbour locations sampled between Whitehaven and Skinburness (Fig. 48). There are relatively few metals however, for which such a distinct distribution pattern satisfactorily accounts for the contamination levels in both clams and worms and clearly other factors significantly influence the bioavailability of metals in these sediment-dwelling species: For example Cu levels in clams are anomalously high at Maryport (anoxia?) and As and Hg relatively enhanced at Skinburness (due to the absence of competing ligands). For other metals highest tissue burdens are, generally found, in samples from either Whitehaven or Workington.

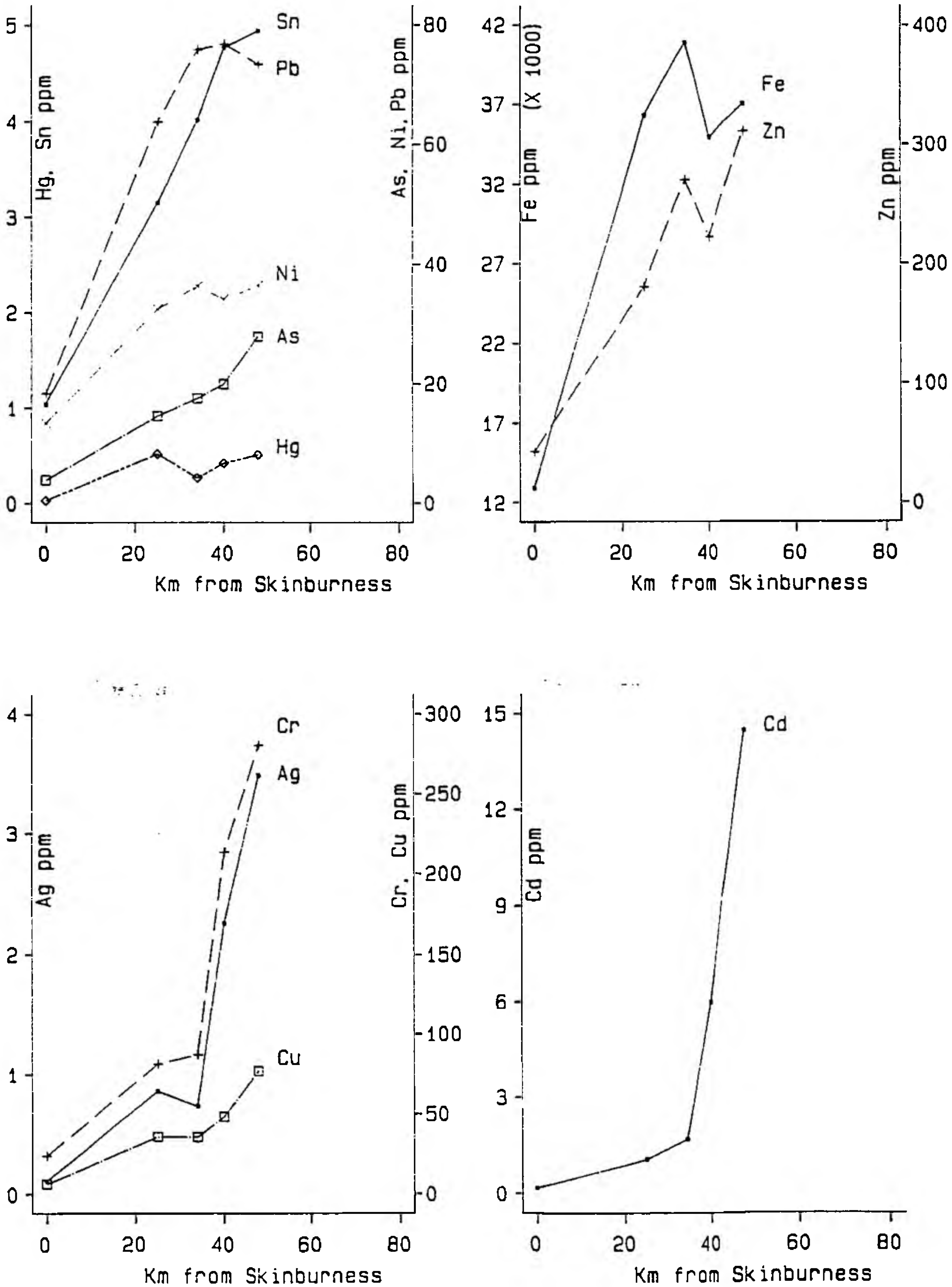
Manganese contamination in Maryport harbour represents the upper limit for UK sediments and even at Skinburness concentrations are five times higher than UK baselines, suggesting a high background level for Mn (together with Fe) along this part of the coastline. Cd concentrations in Whitehaven sediments are among the highest in the UK, whilst for other metals, particulate loadings are below, and in the case of As, Cu, Pb, Sn and Zn, considerably below, those present in contaminated estuaries in south west England. Minimum concentrations of As, Ag, Cu, Hg, Ni, Pb, Sn measured in sediments from the current survey are, indeed, some of the lowest encountered in UK estuaries.

Compared with rocky shore species, where extremely large ranges in tissue burdens are apparent along the Cumbrian coast, the maximum metal concentrations in *Nereis* and *Scrobicularia* are not exceptional and only for Fe and Mn do they approach (within 20%) upper limits for the UK. Nevertheless the lower values for Cd, Ag, Pb and Cu in the current *Nereis* samples appear to be considerably above UK baselines (by factors of 17,15,8 and 7 respectively). Similarily in *Scrobicularia*, the lowest Cumbrian values for Ag,Cr,Cd,Pb and Cu are 35,8,7,5 and 3 times equivalent UK baselines. Therefore, despite the absence of extremely high metal concentrations in sediment-dwellers , the evidence suggests, as with rocky shore indicators, that there may be fairly widespread chronic contamination, at least for some elements.

Possible biological effects

The major aim of the present study has been to determine the impact of industrial discharges on metal burdens in organisms along the Cumbrian coast. Although not intended as an ecological study, some observations on the ocurrence of rocky shore species are of relevance, particularly as distribution appears to be related to high contamination levels along the shoreline in Saltom Bay. Thus, *Fucus* spp. were virtually absent from 2Km north of the Barrowmouth outfall to 1Km to the south, while *Mytilus edulis* could not be found over over a similar stretch of the shore (2Km north - 1.5Km south). *Nucella lapillus* and *Patella vulgata* were absent between 1.7Km north and 700m south, and 1.1Km north and 500m south respectively. Limits for *Littorina littorea* were 500m north and 250m south of the outfall. These observations are very similar to results described by Perkins (1981), who concluded that species diversity was greatly reduced from Hurd Rocks to a site 500m south of the Marchon outfall during surveys conducted between 1970 - 77: No firm

Fig. 48 Metals in Sediments, Cumbria 1990.



conclusions were drawn, however, as to the relative significance of colliery or phosphate processing wastes as causitive agents. Evidence for the latter is now more positive since colliery wastes have been virtually eliminated from the littoral zone in the period between Perkins' studies and the present survey. It is possible that wave action may contribute to the absence of *Fucus* and *Mytilus*, although the symmetrical pattern of disappearance of the above species - either side of an axis centred on the Marchon outfall - suggests poor water quality is responsible for the lack of colonisation. Present results indicate that sensitivity increases in the following order: *Littorina* < *Patella* < *Nucella* <*Fucus*, *Mytilus* . Studies of the basic mechanisms of tolerance to contaminants (such as induction of detoxifying metal-binding proteins, granules, behavioural responses) would clearly be of value in helping to clarify the biological effects observed along this stretch of shore.

Acknowledgements

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Appendix 1. Cadmium in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Species Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	0.168	0.102	2.32	4.61		3.85		0.897	2.94
Maryport	2	NY034370	1.053	0.751	5.69	9.38	73.24	9.93	60.00	0.928	2.10
St. Helens	3	NX999317			3.87	16.06	82.41	16.83	41.99		
Workington shore	4a	NX989299			5.23	13.06	69.03	19.68	25.39		
Workington estuary	4b	NX994273	1.678	0.881	4.17					1.748	5.33
Harrington harbour	5a	NX989253	5.974	2.489	13.24	16.89				0.505	4.42
Harrington shore	5b	NX989253			12.42	21.39		17.53			
Parton	6	NX979211			25.03	12.69	161.5	41.82	37.37		
Whitehaven harbour	7	NX972183	14.459	9.226	7.05	26.64	162.20	74.92	80.78	1.186	7.13
Tom Hurd's Rock	8	NX965183				41.87	250.1		106.4		
N. of Sewage Pipe	9	NX965179				27.00					
S. of Sewage Pipe	10	NX964176				72.52	315.6		140.6		
Whitehaven Mine	11	NX964174			8.52	96.19			188.7		
N. Limit of Outfall	12	NX962165				96.13					
S. Limit of Outfall	13	NX955156			29.62	85.16	294.7		59.77		
St. Bee's Head	14	NX944133			14.17	12.57	77.11	24.88	47.86		
St. Bee's	15	NX958117			5.49	10.93	103.4		58.97		
Seascale	16	NY036007			5.15	7.19	58.96	8.99	44.13		
UK minimum*			<0.1		0.73	0.8	5.5	0.4	2.7	0.03	0.3
UK maximum*			3.6		75.0	146.0	114.0	65.4	289.0	5.8	39.7

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al 1985).

<u>Appendix 2. Arsenic in Cumbrian Biota and Sediments 1990</u>

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Species: Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	<u>3.863</u>	<u>0.965</u>	16.030	11.634		6.573		<u>8.374</u>	<u>15.765</u>
Maryport	2	NY034370	14.563	3.437	12.063	9.627	9.478	7.961	12.074	5.846	11.557
St. Helena	3	NX999317			14.331	15.601	<u>4.637</u>	8.961	11.922		
Workington shore	4a	NX989299			14.414	19.294	15.588	6.253	9.486		
Workington estuary	4b	NX994273	17.512	<u>4.522</u>	10.297					4.576	11.833
Harrington harbour	5a	NX989253	19.857	1.484	10.519	18.551				4.825	<u>8.778</u>
Harrington shore	5b	NX989253			11.010	10.607	12.428	9.252			
Parton	6	NX979211			7.248	<u>7.651</u>	15.067	8.509	6.025		
Whitehaven harbour	7	NX972183	<u>27.814</u>	1.900	<u>3.667</u>	13.689	20.818	<u>10.664</u>	10.856	<u>4.407</u>	12.742
Tom Hurd's Rock	8	NX965183				24.235	<u>22.699</u>		<u>13.346</u>		
N. of Sewage Pipe	9	NX965179				<u>37.561</u>					
S. of Sewage Pipe	10	NX964176				13.282	15.572	6.961	13.178		
Whitehaven Mine	11	NX964174			<u>17.201</u>	10.334			6.781		
N. Limit of Outfall	12	NX962165				15.875					
S. Limit of Outfall	13	NX955156			7.013	12.401	17.390		<u>4.725</u>		
St. Bee's Head	14	NX944133			9.676	10.924	10.301	9.481	8.268		
St. Bee's	15	NX958117			9.088	17.181	14.275		8.704		
Seascale	16	NY036007			11.579	17.375	9.576	<u>5.563</u>	12.122		
UK minimum*			3.4		11.0	9.0	38.0	7.0	11.0	5.6	5.0
UK maximum*			2520.0		382.0	70.5	48.0	28.5	33.0	87.0	191.0

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al 1985).

Appendix 3. Silver in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Species: Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	0.108	0.027	0.073	7.368		0.087		1.038	3.462
Maryport	2	NY034370	0.857	0.151	0.530	13.039	10.908	0.373	8.864	1.606	7.338
St. Helens	3	NX999317			0.241	21.727	14.857	4.356	10.726		
Workington shore	4a	NX989299			0.468	65.692	11.560	8.750	11.168		
Workington estuary	4b	NX994273	0.728	0.217	0.310					3.201	6.862
Harrington harbour	5a	NX989253	2.255	0.140	0.294	53.071				1.187	13.758
Harrington shore	5b	NX989253			0.468	22.049		10.713			
Parton	6	NX979211			0.883	16.106	22.820	64.063	24.487		
Whitehaven harbour	7	NX972183	3.485	0.183	0.581	22.307	19.865	66.257	56.860	1.493	17.134
Tom Hurd's Rock	8	NX965183				20.460	43.300		70.670		
N. of Sewage Pipe	9	NX965179				21.174					
S. of Sewage Pipe	10	NX964176				28.655	47.265		73.804		
Whitehaven Mine	11	NX964174			0.888	40.296			104.160		
N. Limit of Outfall	12	NX962165				53.266					
S. Limit of Outfall	13	NX955156			2.182		34.901		40.561		
St. Bee's Head	14	NX944133			1.145	22.910	19.939	32.448	27.864		
St. Bee's	15	NX958117			0.426	12.587	14.798		12.469		
Seascale	16	NY036007			0.585	12.324	7.899	0.573	9.707		
UK minimum*			<0.1		0.13	0.4	1.27	0.02	0.75	0.07	<0.1
UK maximum*			4.1		1.6	101.0	8.62	16.9	12.0	18.0	259.0

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al 1985).

Appendix 4. Copper in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Species: Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	6.19	3.2	4.52	106.9		5.31		85.48	45.39
Maryport	2	NY034370	35.7	17.0	10.61	134.7	293.7	4.07	37.98	72.74	157.7
St. Helens	3	NX999317			7.66	120.1	372.2	6.36	31.72		
Workington shore	4a	NX989299			9.26	122.5	226.5	5.44	33.53		
Workington estuary	4b	NX994273	35.45	21.10	11.94					84.10	25.39
Harrington harbour	5a	NX989253	48.12	26.2	15.76	147.5				70.63	26.46
Harrington shore	5b	NX989253			13.71	192.3		5.94			
Parton	6	NX979211			19.98	160.2	622.7	13.80	58.68		
Whitehaven harbour	7	NX972183	76.61	42.50	16.39	115.3	305.4	10.25	50.97	101.10	51.69
Tom Hurd's Rock	8	NX965183					1545.3		117.4		
N. of Sewage Pipe	9	NX965179				438.2					
S. of Sewage Pipe	10	NX964176				466.7	1555.0		303.7		
Whitehaven Mine	11	NX964174			29.50	674.4			229.3		
N. Limit of Outfall	12	NX962165				780.2					
S. Limit of Outfall	13	NX955156			40.96	430.6	1325.0		141.7		
St. Bee's Head	14	NX944133			16.57	119.9	281.9	6.37	22.79		
St. Bee's	15	NX958117			5.70	126.6	434.6		29.37		
Seascale	16	NY036007			9.43	96.6	304.9	2.63	14.79		
UK minimum*			16.0		7.3	47.0	28.0	6.0	5.1	10.0	9.0
UK maximum*			2540.0		302.0	1069.0	1377.0	262.0	225.0	1430.0	619.0

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al. 1985).

Appendix 5. Chromium in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

					Species						
	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	24.06	2.69	0.415	1.305		2.741			3.825
Maryport	2	NY034370	81.15	26.07	0.535	1.456	1.195	2.692		0.459	4.462
St. Helens	3	NX999317			2.811	2.880	1.613	12.900			
Workington shore	4a	NX989299			0.909	5.607	1.706	15.141			
Workington estuary	4b	NX994273	86.89	28.72	8.745					0.320	6.514
Harrington harbour	5a	NX989253	213.6	139.86	5.531	2.735				2.441	5.625
Harrington shore	5b	NX989253			1.493	3.438		1.787			
Parton	6	NX979211			2.450	3.436	2.527	15.096			
Whitehaven harbour	7	NX972183	280.5	173.88	3.196	4.917	1.818	12.500		0.349	4.812
Tom Hurd's Rock	8	NX965183				6.817	4.558				
N. of Sewage Pipe	9	NX965179				4.343					
S. of Sewage Pipe	10	NX964176				7.460	4.663				
Whitehaven Mine	11	NX964174			26.405	18.602					
N. Limit of Outfall	12	NX962165				28.535					
S. Limit of Outfall	13	NX955156			18.167		4.002				
St. Bee's Head	14	NX944133			3.406	3.829	2.338	16.922			
St. Bee's	15	NX958117			0.607	3.949	1.475				
Seascale	16	NY036007			1.441	1.260	0.747	1.301			
UK minimum*			15.0		0.15	<0.1	0.16	<0.2		0.1	0.5
UK maximum*			799.0		5.3	1.4	11.0	7.2		6.5	23.8

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al 1985).

Appendix 6. Iron in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Species: Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	12889.0	1805.0	174.2	309.3		320.0		313.7	635.0
Maryport	2	NY034370	36273.0	7092.0	82.3	189.9	257.5	105.0	2382.0	304.8	765.0
St. Helens	3	NX999317			1168.7	208.9	447.7	224.0	5622.0		
Workington shore	4a	NX989299			109.0	446.5	163.1	244.0	4714.0		
Workington estuary	4b	NX994273	40882.0	7444.0	2803.1					347.4	983.7
Harrington harbour	5a	NX989253	34901.0	12233.0	652.4	219.1				589.3	539.0
Harrington shore	5b	NX989253			204.5	367.7		172.0			
Parton	6	NX979211			172.0	142.8	786.4	314.0	1863.0		
Whitehaven harbour	7	NX972183	37064.0	14164.0	65.4	350.1	619.8	426.0	3018.0	631.3	915.0
Tom Hurd's Rock	8	NX965183				184.2	479.9		1671.0		
N. of Sewage Pipe	9	NX965179				207.0					
S. of Sewage Pipe	10	NX964176				213.4	455.7		9957.0		
Whitehaven Mine	11	NX964174			687.8	321.8			2559.0		
N. Limit of Outfall	12	NX962165				421.0					
S. Limit of Outfall	13	NX955156			343.3		532.2		2761.0		
St. Bee's Head	14	NX944133			149.6	176.8	150.8	125.0	2158.0		
St. Bee's	15	NX958117			23.2	250.5	334.2		2036.0		
Seascale	16	NY036007			241.1	175.4	388.2	56.1	1873.0		
UK minimum*			1100.0		104.0	218.0	184.0	64.0	1160.0	227.0	367.0
UK maximum*			63000.0		2081.0	926.0	474.0	669.0	2821.0	734.0	3420.0

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986,and Bryan et al 1985).

Appendix 7. Mercury in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Species: Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	0.028		0.032	0.154		0.122		0.099	0.419
Maryport	2	NY034370	0.516		0.040	0.096	0.267	0.293	0.155	0.071	0.139
St. Helens	3	NX999317			0.025	0.179	0.203	0.305	0.122		
Workington shore	4a	NX989299			0.032	0.215	0.257	0.134	0.149		
Workington estuary	4b	NX994273	0.265		0.051					0.072	0.298
Harrington harbour	5a	NX989253	0.419		0.055	0.137				0.063	0.154
Harrington shore	5b	NX989253			0.070	0.137		0.327			
Parton	6	NX979211			0.060	0.119	0.309	0.251	0.110		
Whitehaven harbour	7	NX972183	0.509		0.114	0.172	0.087	0.245	0.156	0.061	0.185
Tom Hurd's Rock	8	NX965183				0.505	0.273		0.176		
N. of Sewage Pipe	9	NX965179				0.534					
S. of Sewage Pipe	10	NX964176				0.402	0.150	0.235	0.576		
Whitehaven Mine	11	NX964174			0.031	0.415			0.390		
N. Limit of Outfall	12	NX962165				1.018					
S. Limit of Outfall	13	NX955156			0.079	0.661	0.322		0.223		
St. Bee's Head	14	NX944133			0.103	0.206	0.453	0.560	0.243		
St. Bee's	15	NX958117			0.030	0.310	0.295		0.253		
Seascale	16	NY036007			0.062	0.368	0.417	0.232	0.339		
UK minimum*			0.06		0.03	0.14	0.13	0.09	0.07	0.02	0.02
UK maximum*			1.2		0.24	1.69	0.44	2.2	1.19	2.5	1.3

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al 1985).

Appendix 8. Manganese in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Species: Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	399.0	270.6	141.9	144.2		13.17		24.73	164.0
Maryport	2	NY034370	1745.4	1239.3	214.3	170.4	21.36	6.41	74.94	29.03	267.7
St. Helens	3	NX999317			248.8	225.1	26.02	12.44	579.3		
Workington shore	4a	NX989299			237.4	217.5	18.86	14.66	233.3		
Workington estuary	4b	NX994273	1510.8	991.8	896.0					21.15	107.1
Harrington harbour	5a	NX989253	684.8	460.4	251.5	104.9				14.39	38.1
Harrington shore	5b	NX989253			167.8	120.5		7.64			
Parton	6	NX979211			261.2	126.5	19.39	8.57	29.41		
Whitehaven harbour	7	NX972183	560.4	362.7	29.8	87.15	17.71	5.83	43.93	9.46	17.6
Tom Hurd's Rock	8	NX965183				81.73	20.07		12.0		
N. of Sewage Pipe	9	NX965179				74.47					
S. of Sewage Pipe	10	NX964176				77.54	25.16		175.3		
Whitehaven Mine	11	NX964174			314.8	58.18			42.59		
N. Limit of Outfall	12	NX962165				64.92					
S. Limit of Outfall	13	NX955156			170.9	81.78	17.18		38.17		
St. Bee's Head	14	NX944133			242.4	56.24	15.68	7.18	41.31		
St. Bee's	15	NX958117			89.8	89.64	17.28		39.07		
Seascale	16	NY036007			258.5	72.09	17.88	6.71	35.49		
UK minimum*			77.0		51.0	24.0	10.4	4.0	3.4	5.6	10.0
UK maximum*			1160.0		573.0	309.0	36.8	35.0	102.0	34.3	333.0

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al 1985).

Appendix 9. Nickel in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Species Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	13.43	1.39	9.422	4.797		1.342		1.562	5.890
Maryport	2	NY034370	32.52	5.78	11.093	6.753	0.681	0.977		2.729	5.586
St. Helens	3	NX999317			13.071	9.009	1.382	1.840			
Workington shore	4a	NX989299			10.986	11.504	0.700	2.016			
Workington estuary	4b	NX994273	36.39	6.64						1.500	8.693
Harrington harbour	5a	NX989253	34.18	9.28	18.710	4.162				0.574	1.167
Harrington shore	5b	NX989253			15.373	6.555		1.942			
Parton	6	NX979211			52.553	5.589	0.978	2.446			
Whitehaven harbour	7	NX972183	36.43	9.66	3.142	8.490	0.990	1.417		1.669	2.350
Tom Hurd's Rock	8	NX965183				13.944	1.503				
N. of Sewage Pipe	9	NX965179				14.071					
S. of Sewage Pipe	10	NX964176				6.915	2.130				
Whitehaven Mine	11	NX964174			12.429	16.428					
N. Limit of Outfall	12	NX962165				18.567					
S. Limit of Outfall	13	NX955156			37.165		0.919				
St. Bee's Head	14	NX944133			31.831	3.510	0.923	2.578			
St. Bee's	15	NX958117			9.996	4.029	0.814				
Seascale	16	NY036007			12.500	2.225	1.216	1.027			
UK minimum*			18.0		2.6	1.0	0.61	0.5		1.5	1.2
UK maximum*			49.0		53.0	11.5	3.4	12.0		13.3	14.2

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al 1985).

Appendix 10. Lead in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Species: Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	18.32	12.2	4.59	3.20		1.81		1.66	43.62
Maryport	2	NY034370	63.85	67.2	2.97	3.27	1.59	5.42	6.87	1.83	36.63
St. Helens	3	NX999317			6.90	2.87	1.79	6.48	12.12		
Workington shore	4a	NX989299			3.50	4.96	1.59	7.50	7.40		
Workington estuary	4b	NX994273	75.94	72.1	14.40					1.97	32.82
Harrington harbour	5a	NX989253	76.91	78.5	23.26	9.55				2.38	41.08
Harrington shore	5b	NX989253			19.70	6.97		26.31			
Parton	6	NX979211			3.92	1.93	4.16	7.22	8.33		
Whitehaven harbour	7	NX972183	73.42	73.1	1.71	4.67	1.94	12.99	7.28	2.02	31.36
Tom Hurd's Rock	8	NX965183				2.08	3.75		4.18		
N. of Sewage Pipe	9	NX965179				2.69					
S. of Sewage Pipe	10	NX964176				2.98	5.51		22.62		
Whitehaven Mine	11	NX964174			5.89	3.23			6.53		
N. Limit of Outfall	12	NX962165				3.79					
S. Limit of Outfall	13	NX955156			5.06		4.17		5.38		
St. Bee's Head	14	NX944133			3.81	2.51	2.20	7.82	14.19		
St. Bee's	15	NX958117			2.83	2.95	2.20		4.45		
Seascale	16	NY036007			6.10	2.67	1.51	5.62	4.15		
UK minimum*			30.0		1.3	0.9	1.9	1.7	0.4	0.2	6.0
UK maximum*			2175.0		21.6	70.0	34.3	105.0	30.0	685.0	991.0

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al 1985).

Appendix 11. Tin in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Species: Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
Skinburness	1	NY136564	1.034	0.515	0.152	0.362		<0.02		<0.1	0.127
Maryport	2	NY034370	3.144	2.254	0.096	0.129	0.171	<0.02	0.229	0.394	0.493
St. Helena	3	NX999317			0.210	0.301	0.235	<0.02	0.350		
Workington shore	4a	NX989299			0.249	0.329	0.236	<0.02	0.955		
Workington estuary	4b	NX994273	4.008	3.501	0.650					<0.1	0.410
Harrington harbour	5a	NX989253	4.765	2.378	0.360	0.141				<0.1	0.233
Harrington shore	5b	NX989253			0.202	0.212	0.922	<0.02			
Parton	6	NX979211			0.103	0.080	0.451	<0.02	0.212		
Whitehaven harbour	7	NX972183	4.943	2.337	0.049	0.219	0.258	0.890	0.330	1.155	0.725
Tom Hurd's Rock	8	NX965183				0.172	0.296		0.335		
N. of Sewage Pipe	9	NX965179				0.229					
S. of Sewage Pipe	10	NX964176				<0.01	0.124	<0.02	1.383		
Whitehaven Mine	11	NX964174			0.249	<0.01			0.506		
N. Limit of Outfall	12	NX962165				<0.01					
S. Limit of Outfall	13	NX955156			0.103	0.050	0.207		0.201		
St. Bee's Head	14	NX944133			0.087	0.049	0.092	<0.02	0.394		
St. Bee's	15	NX958117			0.023	0.095	0.093		0.222		
Seascale	16	NY036007			0.073	0.075	0.084	<0.02	0.180		
UK minimum*			8.4#		0.04	0.01		0.02		<0.1	<0.1
UK maximum*			2672.0#		1.8	1.6		4.0		2.6	14.0

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al 1985; # - Bryan et al 1980 (tin fusions)).

Appendix 12. Zinc in Cumbrian Biota and Sediments 1990

(Comparison with data from other UK estuaries)

	Site No.	Map ref.	Sediment (total)	Sediment (HCl)	Fucus spp.	Littorina littorea	Nucella lapillus	Mytilus edulis	Patella vulgata	Nereis diversicolor	Scrobicularia plana
					Species						
Skinburness	1	NY136564	42.7	20.0	59.4	126.6		94.9		199.5	913.0
Maryport	2	NY034370	180.3	110.0	146.5	129.8	991.0	99.6	186.3	182.9	909.0
St. Helens	3	NX999317			118.4	134.5	963.0	163.5	170.6		
Workington shore	4a	NX989299			163.7	140.2	722.0	190.2	130.4		
Workington estuary	4b	NX994273	270.3	152.8	552.5					256.4	1596.0
Harrington harbour	5a	NX989253	221.9	135.3	417.2	132.6				160.5	666.0
Harrington shore	5b	NX989253			369.7	134.8		175.7			
Parton	6	NX979211			958.1	159.5	2495.0	203.3	236.1		
Whitehaven harbour	7	NX972183	311.2	190.8	366.9	160.9	1569.0	218.6	243.7	181.7	680.0
Tom Hurd's Rock	8	NX965183				173.3	2516.0		218.7		
N. of Sewage Pipe	9	NX965179				206.1					
S. of Sewage Pipe	10	NX964176				208.4	2603.0		359.7		
Whitehaven Mine	11	NX964174			352.6	236.9			400.0		
N. Limit of Outfall	12	NX962165				236.9					
S. Limit of Outfall	13	NX955156			918.1	259.6	3675.0		238.6		
St. Bee's Head	14	NX944133			461.5	123.4	1072.0	174.1	190.5		
St. Bee's	15	NX958117			144.6	111.3	1587.0		179.5		
Seascale	16	NY036007			177.0	110.8	1014.0	185.5	175.3		
UK minimum*			60.0		69.0	69.0	235.0	45.0	81.0	91.0	268.0
UK maximum*			3515.0		1739.0	956.0	3352.0	579.0	476.0	466.0	4920.0

- Underlined values refer to maximum and minimum values in the Cumbrian biota, 1990.

* - (values from Langston 1986, and Bryan et al 1985).