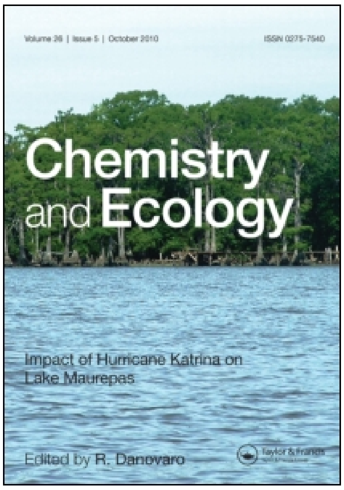

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STABILIZED COAL ASH ARTIFICIAL REEF STUDIES

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In 1989, an experimental stabilized coal ash reef was deployed in Poole Bay off the southern coast of the UK. Three different mixtures of pulverised fuel ash, gypsum, flue gas desulphurisation sludge and cement were used along with concrete controls. The aim was to study the environmental compatibility of the reef materials through heavy metal analyses of the blocks to determine if any leaching or changes were occurring. At the same time, reef encrusting and associated biota have been analyzed together with material from concrete control and natural reefs to determine if there was any evidence for excess bioaccumulation.

This paper presents the results from studies of sectioned reef blocks immersed for 3 and 4 years. No significant change in levels of heavy metals (Cd, Cr, Cu, Mn, Ni, Pb, Zn) has been detected. Surface loss of calcium, presumably associated with the gypsum component, is confirmed with evidence of replacement by magnesium. Trends in data suggest initial surface changes which slow with time.

Epibiota have been analyzed for heavy metal content. The variety of reef-associated organisms has been extended to include mobile, resident species which are higher in the food chain: molluscs, crustaceans and territorial fish. No consistent evidence of bioaccumulation of a range of heavy metals (above those levels found in organisms associated with control reefs) has been detected. Monitoring of the encrusting species range and densities shows little difference between colonisation of the concrete and ash mixture blocks.

KEY WORDS: coal ash, artificial reef, heavy metals, bioaccumulation

INTRODUCTION

Artificial reefs for the promotion of fisheries and aquaculture have been constructed from a wide range of materials. Waste products such as building rubble, tyres and car bodies provide an economical source of reef building materials. The stabilization of unconsolidated waste coal ash for artificial reef construction was pioneered by the Coal Waste Artificial Reef Program (CWARP) in the USA (Woodhead *et al.*, 1985; 1986). Investigations into the use of coal or pulverised fuel ash (PFA) for oyster settlement have been undertaken by Price *et al.* (1988). Large fishery reefs have been constructed from coal ash concrete in Japan (Suzuki, 1985, 1994) and Taiwan (Kuo *et al.*, 1994). In Italy the environmental compatibility of stabilized coal ash blocks has been studied by Relini *et al.* (1994).

Stabilized oil ash (Metz and Trefry, 1988; Nelson *et al.*, 1988) and incinerator ash (Breslin *et al.*, 1988; Roethel and Breslin, 1994) are also being investigated as potential reef construction materials.

Most UK electricity is generated by coal-fired power stations. Less than half the ash produced is used by the construction industry, leaving the rest for disposal. The bulk of this goes to landfill, although two power stations currently dispose of ash by marine dumping. In response to a European Community directive to reduce sulphur dioxide emissions to 60% of the 1982 level by the year 2003, it was proposed to fit flue gas desulphurisation (FGD) plants to major coal-fired power stations. The favoured option was to use limestone slurry sprayed though the flue gases producing gypsum. With the adoption of this process, the volume of gypsum produced would far exceed the requirements of the UK construction industry, providing a further disposal problem. This study, investigating the possibility of stabilizing PFA and FGD gypsum with cement to produce artificial reefs for fishery enhancement, is the first of its type in the UK. Whilst other studies have provided information on the environmental impact of stabilized coal ash, this study used UK sourced coal ash and gypsum in different mixes.

An experimental structure was deployed in Poole Bay, off the central south coast of England, in June 1989. Three different mixtures of PFA, FGD gypsum, cement and gravel were made up in 40 × 20 × 20 cm blocks at a commercial block-making plant (Collins *et al.*, 1990; 1991; 1992). Heavy metals naturally present in coal are concentrated after combustion in the resultant ash. The purpose of stabilizing the ash as blocks is twofold:

(1) to immobilize heavy metals (or other components)

(2) to provide hard substratum for the attachment of organisms.

Fifty tonnes of blocks were formed into eight conical units each 1 m high by 4 m diameter. The eight units duplicate three different PFA/gypsum mixtures and one concrete control. The reef structure is 10 m below MLWS (tidal range 2 m) on a flat sandy sea-bed.

The Poole Bay artificial reef is the focus of an integrated study to examine the effectiveness of this stabilization and hence the environmental compatibility of the block materials. The biological development has been described by Jensen *et al.* (1994a). Results of chemical studies during the first two years of immersion have been described previously (Collins *et al.*, 1990, 1991, 1992). This paper concentrates on two aspects of the chemical work:

(1) examination of the heavy metal concentrations in reef block sections to determine if there is evidence of leaching;

(2) analysis of the epibiota to determine if there is evidence for bioaccumulation of these metals above background levels.

METHODS

Reef blocks from each of the 3 mixtures were raised, 3 and 4 years after development. A 5 × 5 × 10 cm deep core was cut from the centre of each block using a diamond saw. The cores were sectioned 7, 20, 33, 60 and 100 mm from the surface. Sections were dried in an oven at 90°C for several days to constant weight and any epifauna was carefully scraped off. The block section samples were placed inside a polyethylene bag and crushed with a hammer, then passed through a 500 μm sieve to remove the inert gravel fraction.

Epibiota and reef-associated fauna (algae, hydroids, bryozoans, sponges, ascidians, molluscs and crustaceas) were removed by divers from each control site and reef unit. Fish were caught in small traps set on each reef unit. Organisms were stored, frozen, in numbered polyethylene bags. Any adhering reef block material on the epibiota samples was removed by scraping/brushing and finally rinsing the organisms in a small amount of distilled water. Larger species (molluscs, crabs and fish) were dissected into different tissue types. The samples and tissues were placed in plastic weighing boats and dried in the same way as the block samples.

Block and epibiota samples were analyzed for cadmium, chromium, copper, lead, manganese and zinc. In addition, the concentrations of calcium and magnesium were measured in the block samples. Samples were digested at 80°C for 24 hours in concentrated nitric acid before flame atomic absorbtion spectrophotometry using a Pye Unicam SP9 AAS. BDH 'Spectrosol' standards were used. Additionally, standard reference material (TORT-1 lobster Hepatopancreas, National Research Council, Canada, Marine Analytical Chemistry Standard) was analyzed to check the accuracy of the standards. Results for the range of elements described in this paper were within one standard deviation of the certified values. One blank was included for every 10 samples. All samples from immersed blocks were analyzed alongside pre-deployment block samples for reference and standardization. The standard deviation of determinations was within ±1%, although the reproducibility of sampling due to the non-uniformity of block material increased this to ±5%.

The nitric acid digestion of PFA does not completely mobilise the heavy metals. To determine the fraction remaining after nitric acid digestion, representative block samples were also totally digested in boiling hydrofluoric acid. After evaporation to dryness, addition of perchloric acid and evaporation to dryness, distilled water was added and the digests analyzed as above.

RESULTS AND DISCUSSION

Block Analysis

The differences in heavy metal composition of the 3 block mixtures are not significant (Collins *et al.*, 1990; 1991) and results presented have been averaged across them. Figure 1 shows the average concentration of heavy metals against depth within the ash reef blocks (2 of each of 3 mixtures) after 4 years' immersion. There is no significant change (beyond one standard deviation) in any of the metal concentrations through the block section. Very similar results were obtained after 3 years' immersion.

Two possible trends, surface loss of cadmium and surface enrichment of chromium and manganese, were noted in previous reports (Collins *et al.*, 1992, 1994). These trends for cadmium and chromium are indicated in Figure 1 but are within the variability of the data. No surface enrichment of manganese is suggested by this data set.

The concentrated nitric acid digestion of PFA gives an underestimate of the metal component because that contained within the glassy matrix is not dissolved. However, the fraction which is not mobilised by boiling concentrated nitric acid is unlikely to be environmentally available. A comparison of metal concentrations calculated from hydrofluoric (total digestion) and nitric acid digestion of ash block material is shown in Table I.

Changes in the concentrations of calcium and magnesium were evident, showing loss of calcium and increase in magnesium at the surface. The concentrations of these two elements are plotted against each other in Figure 2 to show this inverse

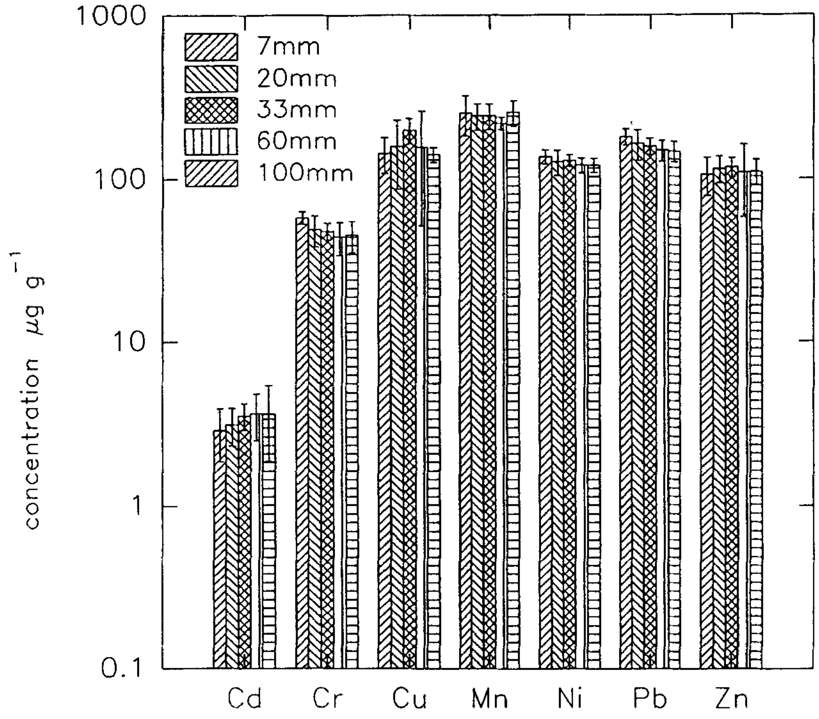

Figure 1 Average concentration (±1 standard deviation) of heavy metals at different depths in an ash reef block section.

Table I Comparison of heavy metal concentrations (μg g^{-1}) in ash reef block samples using hydrofluoric and nitric acid digestion methods

	hydrofluoric acid average	hydrofluoric acid std. dev.	nitric acid average	nitric acid std. dev.	recovery %
Cd	4.33	0.36	3.76	0.11	87
Cr	99.4	5.6	37.1	0.8	37
Cu	138	10	91	4	65
Mn	214.1	21.4	199.5	5.1	93
Ni	165.7	11.7	85.8	0.7	52
Pb	138.7	13.8	108.9	2.3	79
Zn	196	19	125	4	64

relationship. Sectioning the blocks reveals a lighter coloured outer zone approximately 10 mm deep, which presumably represents the main area of interaction with the sea water. The depth of this visible zone has remained constant in sections cut from blocks sampled after 1, 2, 3 and 4 years' immersion, suggesting that changes occurred rapidly after deployment and have slowed subsequently. Increasing compressive strength of the ash blocks with time immersed has already been reported (Collins *et al.*, 1992). This has also been noted by Carleton and Muratore (1985) Roethel and Oakley (1985) and Suzuki (1994).

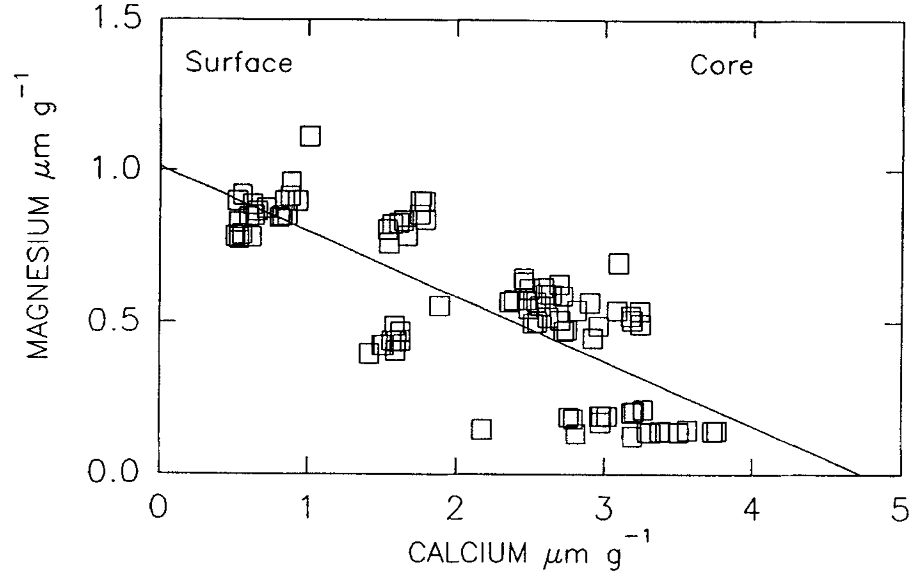

Figure 2 Plot of calcium against magnesium concentrations in ash reef block sections with a first order linear regression line to indicate the trend in the data.

Roethel and Oakley (1985) reported the loss of calcium, largely due to the solution of gypsum, from coal waste/FGD sludge blocks during the initial stages of the Coal Waste Artificial Reef Programme (CWARP). Replacement of calcium by magnesium in the same materials was noted by Labotka *et al.* (1985). Hockley and van der Sloot (1991) examined mineralogical and chemical changes in a CWARP block (made from coal ash and FGD gypsum) after 8 years' immersion. The depth of the region of change in composition was between 1 and 2 cm from the surface. The most notable feature of this zone was the exchange of magnesium for calcium. Minerals derived from the FGD gypsum (gypsum and calcium sulphite) and cement/PFA (ettringite and portlandite) were replaced by calcite.

In a study of leaching from coal-ash and coal-ash products, Van der Sloot *et al.* (1985) demonstrated considerably reduced availability of metals from stabilized products. The metal concentration profiles of a coal waste block from the CWARP experiment, immersed for 1.5 years, were also examined. They showed no significant uptake or loss of iron, cobalt, chromium or vanadium, a slight loss of antimony, zinc and copper and, due to large variation in the data, no clear pattern for arsenic and cadmium. Molybdenum and lead were identified as being significantly lost from the surface layer. Some uptake of manganese was found and an inward flux rate was estimated. Roethel and Oakley (1985), working with similar blocks after 500 days' immersion, also note an apparent enrichment by manganese and proposed a number of mechanisms for this. Scavenging of trace metals by coal ash blocks in tank studies has been reported by Seligman and Duedall (1979) and Heaton *et al.* (1982). Hockley and van der Sloot (1991) observed a CWARP block immersed for 8 years, reporting no loss of zinc, copper or manganese. The limited surface loss of arsenic observed was similar in magnitude to the trend for cadmium suggested in Figure 1.

EPIBIOTA ANALYSIS

Both the concrete and ash reef blocks were rapidly colonised by a wide variety of epibiota, fish and Crustacea (Collins *et al.*, 1991, 1992; Collins and Jensen, 1991; Jensen *et al.*, 1994a and b). Epifauna and algae from the surface of the reef blocks have been analyzed routinely for heavy metals to determine if there has been any excess uptake relative to that of organisms growing on control surfaces: concrete reef blocks, sea-bed stones and local natural reefs. The principal species or genera included within the taxonomic groups used are:

Red algae	*Calliblepharis ciliata*, *Bonnemaisonia* sp., *Chondria* sp.
Brown algae	*Dictyota dichotoma*, *Dictyopteris membranacea*, *Desmarestia* spp.
Hydroids	*Halecium halecinum*, *Sertularella* sp., *Tubularia* sp. *Aglaophenia* sp., *Plumularia* sp.
Bryozoans	*Bugula* spp., *Vesicularia spinosa*, *Bicellariella* sp.
Sponges	*Scypha ciliata*, *Amphilectus* sp., *Halichondria* spp.
Ascidians	*Ascidia mentula*, *Styela clava*

Results for ascidians were largely based on samples of *Ascidia mentula*. This was a pioneering species and continues to flourish on the underside of blocks and in crevices. Specimens were large and likely to be of a similar age to the reef (4 years). If there had been significant release of metals from reef blocks, this species, a filter feeder, would be well placed to indicate this. Hydroids, bryozoans and red algae showed annual decline and growth cycles (Jensen *et al.*, 1994.). Sponges settled on the reef blocks after 1 year, giving a miximum age for the samples analyzed of 3 years.

The results of analyses from samples of these organisms taken in summer 1993 (3 years after deployment) are summarised in Table II and those taken in 1993 (4 years after deployment) shown in Figure 3. In each case at least 10 individuals or colonies of each organism from ash and control reefs were analysed. Both sets of data show no significant differences between the heavy metal content of organisms growing on the ash reef blocks compared to those on the control surfaces.

REEF PREDATORS

The possibility of transfer and concentration of excess heavy metals by predatory fauna higher in the food chain has been considered. Detailed examination of the reef by divers through the life of the reef has shown that some species are resident and actively browse on the reef epibota. Representative organisms sampled were gastropod molluscs (*Buccinum undatum*), crabs (*Liocarcinus puber*) and fish (*Labridae*). The concentrations of heavy metals in another predatory gastropod mollusc, *Oceanebra erinacea*, have been reported previously (Collins *et al.*, 1994). There are larger, commercially important, crustaceans resident on the artificial reef: lobsters (*Homarus gammarus*) and crabs (*Cancer pagurus*) (Collins and Jensen, 1991). However, these are generally nocturnal and have not been observed feeding. By contrast the smaller crab, *Liocarcinus puber*, has been seen both feeding and present in larger numbers, allowing more representative sampling. The most numerous fish

Table II Comparison of heavy metal concentrations ($\mu g\ g^{-1}$) in reef epibiota (red algae, hydroids, sponges, bryozoans and ascidians)sampled in 1992, 3 year after reef deployment

		Cd		Cr		Cu		Mn		Ni		Pb		Zn	
		avg	sd	avg	ad	avg	ad	avg	sd	avg	sd	avg	sd	avg	sd
brown algae	ash	2.15	0.46	3.1	1.8	11.2	3.3	122	93	11.5	1.6	22.6	7.0	71	36
	control	3.06	0.34	4.2	1.9	11.0	2.7	150	56	14.4	1.6	24.6	7.6	55	7
red algae	ash	2.69	1.84	3.9	2.2	13.5	4.6	153	60	13.45	1.8	24.2	6.5	77	18
	control	2.19	0.86	3.9	1.6	11.6	1.7	139	80	14.8	3.2	28.1	11.8	84	23
bryozoa	ash	4.03	1.23	4.3	1.7	14.2	3.0	116	56	25.4	4.3	29.9	10.4	124	36
	control	4.55	2.38	3.7	1.2	13.6	3.0	141	52	25.5	7.9	34.0	14.2	157	157
hydroids	ash	4.67	2.83	3.5	1.8	22.1	6.9	149	70	25.4	16.3	56.8	60.8	110	54
	control	2.16	0.30	3.9	1.9	18.8	1.2	181	83	30.6	20.3	91.7	87.4	131	52
sponges	ash	2.50	0.79	2.6	1.0	14.9	6.8	85	31	12.6	4.9	21.3	6.9	68	41
	control	2.56	1.06	2.3	1.3	14.7	9.0	114	81	12.7	3.6	24.9	13.3	105	185
ascidians	ash	2.12	1.03	2.6	1.3	10.6	2.5	103	90	13.8	4.4	19.8	10.2	72	14
	control	2.04	0.83	2.7	0.8	10.4	2.5	88	28	13.8	5.0	19.8	7.0	77	33

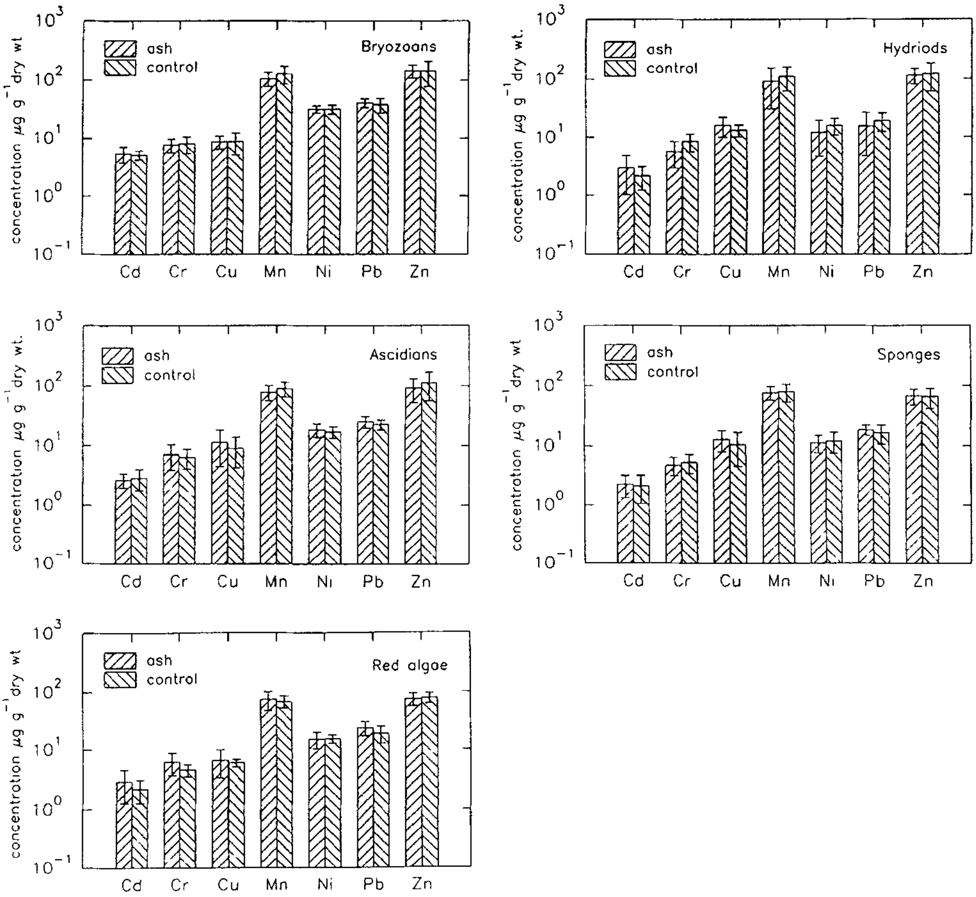

Figure 3 Comparison of heavy metal concentrations (±1 standard deviation) in reef epibiota (red algae, hydroids, sponges, bryozoans and ascidians) sampled in 1993, 4 years after deployment.

on the reef are pout (*Trisopterus luscus*) which form shoals of several hundred around each unit during the day and disperse at night. Stomach content analysis indicates that this species feeds on small crustaceans from the surrounding sea-bed. Wrasse species (Labridae: corkwing, *Crenilabrus melops* and goldsinny, *Ctenolabrus rupestris*) are resident and territorial, staying close to and within a specific reef unit. It is therefore assumed that the diet must be largely derived from that reef unit.

Table III shows the results of analyses of the different tissues (approximately 10 samples in each case) from animals living on ash reefs and those collected from the control reefs (pooled results of the concrete reef units and natural reefs). As with the epibiota, all concentration differences are within the variability of the data, suggesting that there are no significant differences between the ash and control populations.

Previous work on the Poole Bay artificial reef showed no appreciable difference between epibiotic colonisation of the different PFA/gypsum mixtures or the control surfaces (Jensen *et al.*, 1994.). Similarly, epibiota heavy metal concentrations (Collins *et al.*, 1991, 1992 and current work) are comparable. Similar conclusions were drawn

Table III Comparison of heavy metal concentrations ($\mu g\ g^{-1}$) in different tissues of reef predators (gastropod, *Buccinum undatum*; crab, *Liocarcinus puber* and fish, Labridae) from ash and control reefs

			Cd		Cr		Cu		Mn		Ni		Pb		Zn	
			avg	sd	avg	sd	avg	sd	avg	sd	avg	sd	avg	sd	avg	sd
Labridae	flesh	ash	o.5	0.1			2.4	0.8	2.5	0.9	3.7	1.1	5.5	1.2	14.2	2.4
		control	0.5	0.1			2.0	0.8	3.1	1.6	4.1	1.5	6.4	2.1	13.1	4.2
	liver	ash	0.6	0.3			4.9	1.2	5.5	1.0	4.6	2.3	9.8	2.3	47.3	15.8
		control	0.9	0.3			5.3	2.9	5.5	2.4	4.9	3.4	12.0	4.2	37.0	18.4
Liocarcinus puber	gill	ash	35.3	13.6	0.3	0.5	230	86	8.1	2.5	8.7	2.6	138	52		
		control	29.1	8.2	0.2	0.4	139	79	23.6	26.0	9.6	1.0	11.7	1.8	115	157
	hepatopancreas	ash	25.1	13.8	0.5	0.6	315	121	17.2	7.2	13.7	9.1	12.4	5.1	141	39
		control	14.0	3.8	0.4	0.6	216	185	32.3	28.3	10.5	1.5	11.1	1.6	99	63
	muscle	ash	4.6	1.0	0.1	0.3	58	20	3.8	2.9	6.2	1.9	6.8	1.4	258	51
		control	3.7	0.9	0.1	0.3	32	15	9.3	11.2	6.9	1.1	8.5	1.7	118	77
Buccinum undatum	gonad	ash	28	45	1.4	3.7	96	92	24.1	3.9	7.4	7.3	10.3	10.2	396	380
		control	316	137	1.4	1.4	944	802	10.2	2.3	12.8	2.4	16.4	4.2	1541	427
	higher digestive gland	ash	9	12	0.2	0.6	63	145	4.9	2.7	5.4	3.5	7.7	3.0	63	42
		control	52	26	1.2	1.1	132	61	9.6	2.6	19.7	3.5	19.4	5.0	185	173
	lower digestive gland	ash	104	38	1.6	1.1	448	157	13.8	4.9	13.9	5.0	18.7	6.4	618	426
		control	288	114	1.1	1.9	717	539	10.5	3.6	15.8	6.0	24.4	7.5	1439	299
	muscle	ash	17	5	1.4	1.3	115	63	6.1	2.2	11.4	8.5	11.6	5.4	186	129
		control	156	206	0.7	1.0	126	121	6.0	2.0	10.7	2.3	10.8	3.0	529	465
	foot	ash	6	2	0.0	0.0	16	10	15.9	2.8	6.9	0.1	10.9	0.9	35	2
		control	15	14	0.9	2.2	37	14	4.7	1.7	11.6	6.9	10.0	3.4	143	92
	shell	ash	20	4	8.1	0.7	11	6	12.5	1.7	31.8	3.7	36.5	5.7	21	20
		control	20	2	6.4	3.0	9	1	12.4	1.1	27.5	9.6	34.5	6.2	10	3

from the CWARP study (Woodhead *et al.*, 1986) and from Italian studies by Relini *et al.* (1994).

CONCLUSIONS

There have been surface changes in the PFA/gypsum block material which have been described by other workers, principally the loss of calcium and its replacement by magnesium. A lighter coloured, visibly affected, surface layer (10 mm deep) is seen in block cross section. The depth of this appears to have remained constant over 1, 2, 3 and 4 years. Block section studies of heavy metal (Cd, Cr, Cu, Mn, Ni, Pb, Zn) concentrations have remained at similar levels through this period with no significant loss. These observations suggest attenuation of diffusion processes with time. When combined with evidence for increasing block compressive strength this would suggest long-term stability of the material.

No evidence has been found for excess bioaccumulation of heavy metals by reef bioata and reef predators associated with the ash block reefs compared to those from natural and concrete control reefs.

The results from four years' study of the chemical stability of the coal-ash material and reef colonisation by a diverse epibiota are encouraging. They suggest that stabilized coal-ash may be used for the construction of artificial habitats for fisheries. Monitoring of heavy metal concentrations in the Poole Bay artificial reef blocks and reef associated biota is continuing in order to gain an understanding of the long-term behaviour of the stabilized coal ash mixtures in the marine environment.

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