

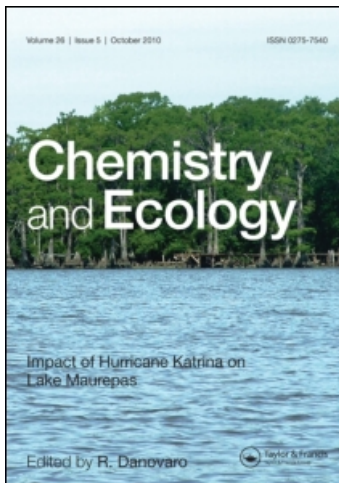
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Disposal of Coal Fly Ash at A Deep Water Site in the Eastern Mediterranean Off Israel - Six Years of Monitoring

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DISPOSAL OF COAL FLY ASH AT A DEEP WATER SITE IN THE EASTERN MEDITERRANEAN OFF ISRAEL – SIX YEARS OF MONITORING

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Coal fly ash (CFA) is dumped at a deep sea disposal site (1,500 m water depth) in the eastern Mediterranean, ca. 70 km off the Israeli shore. Since 1989, about one million tons of CFA were dumped at the 200 km² allocated area. Six years of monitoring at the dump-site shows that the CFA is heterogeneously distributed; there are areas where CFA covers about 1.3 cm depth of the sea floor while at others no CFA is found. CFA is present as a fine powder, small aggregates and even as large blocks both in the dump-site as well as at its peripheries. Cadmium, copper and zinc concentrations in the CFA decreased as a result of the prolonged contact with sea water at *in situ* conditions while inconclusive changes in mercury, iron and manganese were detected. No changes were observed for lead, iron and aluminium concentrations. A controlled long term field experiment, now in progress at the site, is expected to clarify further chemical changes occurring in the CFA.

Keywords: Coal fly ash; ocean dumping; monitoring; heavy metals; eastern Mediterranean

INTRODUCTION

The Israeli Electric Cooperation (IEC) operates two coal fired power plants, both located at the Mediterranean coast: Orot Rabin in Hadera (since 1981) and Ruthenberg in Ashkelon (since 1991). During 1982–1995, out of the 6.1 million tons of CFA generated, 65% were

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utilized for cement production and 18% served to construct embankments around the Hadera power station. The remaining 17% (more than 1 million tons CFA) were disposed at sea (Metzger, 1996), with authorization from the Israeli Ministry of the Environment. The marine disposal option was chosen due to lack of sites available for land disposal, the threat of contamination to ground water, a scarce and valuable resource in Israel, and the proximity of the power plants to the sea.

The potential availability of elements to the environment is a major ecological concern in the utilization or disposal of CFA inland or at sea. Leaching behaviour of different CFA and CFA products in contact with sea water was characterized in a number of studies (Hjelmar, 1983; Crecelius, 1985; Rose *et al.*, 1985; van der Sloot and Nieuwendijk, 1985; van der Sloot *et al.*, 1985; Kress, 1993). These studies found that CFA constituents leached to sea water, mainly those elements forming anionic species in sea water as arsenic, chromium, selenium. The extent of the leaching depends on the type of ash, the contact time and the ratio of CFA to sea water. Ocean disposal of coal wastes was evaluated by Crecelius (1985); Rose *et al.* (1985); Merifield *et al.* (1985) and Harvey (1989), while the effects of CFA dumping on the near shore environment was reviewed by Bamber (1980, 1984) and Norton (1985, 1989). The main findings of these studies were: (1) CFA sinks rapidly to the ocean floor, (2) the predicted leaching of metals during deep sea dumping complies with toxicological and bioaccumulation related criteria, (3) bioassays show bioaccumulation of elements in the target organisms, (4) severe depletion of the benthos is found in near shore dump areas, both in terms of species and abundance, (5) fine CFA particles are dispersed by currents and with the bedload in the near-shore area.

Considering these findings, a dump-site was designated for the disposal of excess CFA generated by the IEC. The dump-site comprises an area of 210 km² located 70 km off Hadera, at 1,400–1,500 m water depth (Fig. 1). An *in situ* experiment performed prior to the commencement of dumping operations found that CFA descended rapidly to the bottom and within four hours the water column was virtually free of CFA (Kress *et al.*, 1993). Similar results were recorded by Rose *et al.* (1985) during an experimental dump at the Deepwater Dump-site 106, 240 km east of Cape Henlopen, Delaware (USA).

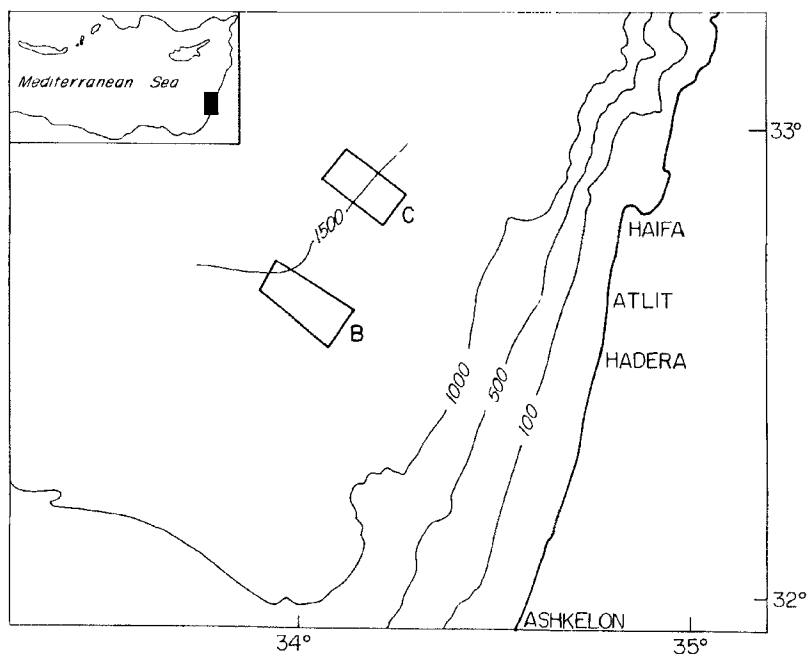


FIGURE 1 Map of sampling areas. B—Hadera dump-site, and C—Control area off Atlit. Contours represent water depth (m).

Consequently, the monitoring studies performed at the CFA dump-site and at a control area off Atlit (Fig. 1) emphasize the benthic compartment: delimitation of the affected area, physical and chemical effects on the sediment, benthic community structure (species diversity and abundance) and bioaccumulation of trace metals in benthic fauna and in deep sea sharks. Initial findings of the monitoring programme showed severe impoverishment of the benthos in the disposal area compared to a control site (Kress *et al.*, 1993). No bioaccumulation of metals was found in deep sea sharks or in benthic fauna collected at the dump-site compared to the control site (Hornung *et al.*, 1993; Kress *et al.*, accepted).

It is the aim of this paper to describe the spatial distribution of CFA in the area after six years of monitoring and to examine the changes in heavy metals composition in the CFA after prolonged contact with sea water in natural, *in situ* conditions.

METHODS

Sampling, Sample Preparation and Chemical Analysis

Sediment and CFA samples were collected annually inside the dump-site and at its periphery during 1990–1995. Figure 1 depicts the location of the dump-site (site B) and Figure 2 the sediment sampling points (P) and the trawl lines (L). Sediment samples were collected also at a control site (Fig. 1, site C) once every two years.

Sediments were sampled at twelve stations during each survey with an Ocean Instruments BX 700 A1 box corer with an area of 0.062 m². Stations P1–P9 (Fig. 2) were sampled during at least four out of the six surveys. Undisturbed sub-cores were taken manually from the box corer using clear Plexiglas hollow cylinders. CFA aggregates were collected by a beam trawl (2 m mouth), and during the benthic survey trawls with a Marinovich trawl with 8 m mouth. The samples were stored frozen until the laboratory analysis. In the laboratory, cores were extruded and sliced, and the upper 1 cm taken for analysis. The CFA aggregates were rinsed with deionized water to remove sediment

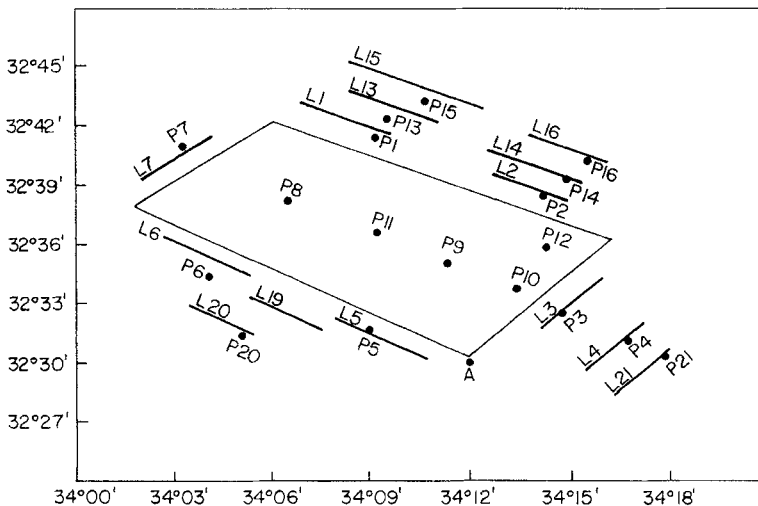


FIGURE 2 Map of the dump-site with sediment sampling stations (P) and trawl lines (L). Point A is the location of the controlled field experiment.

particles that adhered to their surface. Small aggregates (1–100 g) were crushed and homogenized. The large aggregates (1–8 kg) were split and the outer and inner parts of each aggregate scraped using a plastic knife. The sediment and CFA samples were lyophilized for 48 hr and then totally digested with a mixture of hydrofluoric acid and aqua-regia as described in ASTM (1983). A separate digestion was performed in concentrated nitric acid (Hornung *et al.*, 1989) for the determination of mercury.

The digest solutions were analyzed for cadmium, lead, copper, zinc, manganese, iron and aluminium by atomic absorption spectrophotometry using an IL-951 or a Perkin–Elmer 1100 B spectrophotometer equipped with flame and graphite furnace modules. Mercury was analyzed by cold vapour atomic absorption spectrometry on a Coleman Mercury Analyzer MAS-50. Quality control and quality assurance of the results were made with standard reference materials from the National Institute of Standards and Technology (NIST–Coal Fly Ash-1633a and Estuarine Sediment-1646). The standards were digested and analyzed in the same manner as the samples, with each analytical run. All metals gave results within 5% of the certified values. Only for cadmium, the recovery in NIST's Coal Fly Ash-1633 ranged between 71 and 88% thus the results were corrected to 100% recovery. Lead, copper and zinc in the box corer samples from 1991 to 1992, at the beginning of the monitoring studies, were analyzed after digestion with nitric acid. The concentrations in the samples that contained a large proportion of ash (marked by an asterisk in Tab. I) were corrected for incomplete recovery, based in the results of the standard reference material.

The concentrations in representative, six-monthly fly ash composite samples collected at the power stations were taken from the reports presented to the Ministry of the Environment by the IEC, as part of the dumping license requirements. Representative samples from the Orot Rabin and Ruthenberg power stations are denoted as HFA and RFA, respectively. Aluminium and iron concentrations in the representative samples were determined by us, by the same methods used for the CFA sampled from the sea.

The procedures used in the statistical analyses were the *t*-test, and the Mann–Whitney *a*-parametric test, conducted under the assumption of 95% confidence level.

TABLE I Metals concentration in sediments from the dump-site, the control area and in representative coal fly-ash

Sample	date	Hg	Cd	Cu	Zn	Pb	Mn	Fe	Al	% ash	layer
				$\mu\text{g g}^{-1}$				wt%		cm	
P9	1991	na	0.04*	95.0*	71.1*	72.5*	731	3.38	11.3	77	0.5
P9	1992	0.290	0.23*	82.0*	81.3*	73.3*	711	3.73	12.6	91	1.3
P9	1993	0.178	0.17*	101	84.6	74.5	834	4.53	12.4	84	1
P9	1994	0.251	0.32*	53.9	154	79.7	574	3.85	12.3	93	1
P9	1995	0.118	0.16*	68.5	84.2	53.4	966	4.65	11.6	73	0.7
P1	1991	na	0.10	67.1	90.4	48.4	2047	5.95	7.60	0	
P1	1992	0.047	0.09	76.4	88.8	47.9	1861	5.43	7.61	7	
P1	1993	0.050	0.03	66.9	100	45.9	1793	5.32	7.62	9	
P1	1994	0.092	0.07	78.2	91.0	48.3	1608	5.32	7.39	13	
P1	1995	0.071	0.09	71.8	89.7	33.7	1728	5.42	7.49	10	
Natural sediment #											
n = 9	Avg	0.035	0.13	72.5	116	47.5	1992	5.84	7.63		
	std	0.012	0.05	6.2	15	8.5	152	0.54	0.25		
HFA	Avg	0.22	0.69	80	100	76	414	3.59	14.3		
	std	0.06	0.12	7	14	15	29	0.29	0.4		
	n	8	13	13	13	12	9	4	4		

na-not analyzed, Avg-average, std-standard deviation, n-number of samples. # Kress *et al.*, 1995.

* Corrected for incomplete recovery, see method. HFA-Orot Rabin Power station representative coal fly-ash.

RESULTS AND DISCUSSION

Distribution of CFA in the Dumping Area

CFA has a distinctive silver grey colour that contrasts with the brown natural sediment in the area, enabling visual examination of sediment cores and measurement of the CFA layer that accumulated on top of the sediment. The distribution of CFA in the dump site was very heterogeneous. Station P9 had CFA layers of 0.5–1.3 cm (Tab. I) while an adjacent site such as station P11 had only traces of CFA. In 1993, a station sampled just 1 km away from station P9 had a layer of only 0.3 cm (Kress *et al.*, 1994). Significant layers of CFA (0.3–1.0 cm) were found also at stations P8, P10 and P12. No measurable layers of CFA were found at stations located at the periphery of the site (P1–P7).

CFA collected with the box corer was always in a powdery state, sometimes with the presence of small (up to 1 cm diameter) aggregates, while the trawl hauls landed CFA aggregates of varying sizes and shapes, from round aggregates of 1–2 cm diameter to blocks of

undetermined shapes up to 80 cm in length. It is known (Bamber, 1983) that CFA exhibits pozzolanic activity and is expected to aggregate in sea water. We assume that the pozzolanic activity caused the formation of the smaller aggregates found. However, it is reasonable to assume that the larger aggregates were dumped as such. Most of the CFA dumped at the site (ca. 0.85 million tons) was piled up for 2–6 years next to the Orot Rabin power plant, exposed to rain and weathering. It solidified and the pile was broken down before the CFA was loaded on the ship and then dumped at sea, (IEC-personal communication).

CFA aggregates were found inside the dump-site as well as at its periphery, up to a distance of 4–6 km. Trawling inside the dump-site was discontinued after 1992 because the net overfilled with CFA and was almost impossible to haul it aboard the ship. CFA was present since 1990 toward the northeast (L1 and L2) and to the southeast (L3 and L4) of the site while no CFA was found to the northwest (L7) (Fig. 2). The southwestern area (L5–L6, Fig. 2) was free of CFA up to 1994, prior to the commencement of ash dumping from the Ruthenberg power plant. Since then, CFA was found even at L20 (Fig. 2), 5 km from the limits of the dump-site.

Sediment Composition

The levels of mercury, cadmium, lead, copper, zinc, manganese, iron and aluminium and the calculated percentage of CFA in the top 1 cm layer of the sediment cores are given in Table I. We chose to depict one station influenced by CFA, P9, where a layer of at least 0.5 cm of CFA on top of the sediment was measured during all surveys; and one station, P1, where no CFA was detected visually. The percentage of CFA in the samples was calculated from their aluminium and manganese contents using a simple mixing diagram between natural deep sea sediments and HFA as end members, as described by Kress *et al.* (1993). Table I includes also the natural metal levels found in the sediments at the control site (Kress *et al.*, 1995) as well as the levels in representative samples of CFA from the Orot Rabin (HFA). The levels of mercury, cadmium, lead and aluminium in natural sediments were significantly lower than the levels found in HFA while manganese and iron were higher in the sediments. Copper and zinc concentrations in the natural sediments were similar to the concentrations found in

HFA. The samples from station P9 has higher levels of mercury, lead and aluminium than the natural sediments and lower manganese and iron, due to the presence of CFA (Tab. I). We expected to find also higher cadmium levels. However, cadmium concentrations in the P9 samples were not significantly different from the natural background concentration in the sediments. Cadmium is one of the elements that solubilize from the CFA while in contact with sea water (Crecelius, 1985; van der Sloot and Nieuwendijk, 1985; Kress, 1993). Cadmium leaching was also observed when CFA, recovered from the dump-site, was placed in contact with running sea water for one year, showing cadmium leaching to be a long term process (Shoham–Frider, 1997). Therefore, the lower than expected cadmium values in the P9 samples was attributed to leaching. Calculation based on the mean concentration of cadmium in HFA and P9-1994 sample (with 93% CFA) estimates 54% leaching, higher than ca.30% leaching found in experiments performed with HFA and Mediterranean sea water (Kress, 1993; Shoham–Frider, 1997). The concentrations of copper, and to a lesser degree zinc, in P9 showed a high degree of variation but were mostly similar to the natural concentrations.

The concentrations found in the sediments at station P1 were natural for all metals studied (Tab. I). Mercury was higher than the natural levels in sediments at P1-1994 and P1-1995 when the calculated percentages of CFA in the sediments were 13% and 10%, respectively. This fact indicates that mercury concentration is a very sensitive parameter that can be used to detect CFA presence in the sediments, in addition to aluminium and manganese concentrations.

Changes in Composition of CFA

The presence of CFA in prolonged contact with sea water under natural conditions, the ability to retrieve it from the sea floor and the questions raised concerning changes in the cadmium concentrations in the box corer samples prompted us to add to the monitoring programme the analysis of CFA aggregates recovered from the sea. The purpose was to follow temporal changes in CFA composition and determine its leaching behaviour. Particular emphasis was given to the leaching of cadmium, which is addressed specifically in the Israeli legislation for the prevention of marine pollution by dumping.

CFA was analyzed for mercury, cadmium, lead, copper, zinc, manganese, iron and aluminium. The results covered a wide range of concentrations : Hg 0.032–0.562 $\mu\text{g g}^{-1}$, Cd 0.06–1.09 $\mu\text{g g}^{-1}$, Pb 41.4–112 $\mu\text{g g}^{-1}$, Cu 31.3–113 $\mu\text{g g}^{-1}$, Zn 23.9–168 $\mu\text{g g}^{-1}$, Mn 370–600 $\mu\text{g g}^{-1}$, Fe 1.85–6.02 wt% and Al 8.30–17.0 wt%. Average values and standard deviations for each survey are summarized in Table II and the distribution of the data for cadmium, copper and aluminium is presented in box-whisker plots (Fig. 3). *T*-test and Mann–Whitney comparisons between the mean composition of CFA during each survey and the mean composition of HFA showed that for the 1993 samples, mercury has higher and cadmium and iron were lower than in the HFA. In 1994, mercury and manganese were higher and cadmium and copper were lower in the CFA samples while in 1995, manganese was higher and copper was lower in CFA.

It is known that the composition of CFA changes with contact with sea water (Hjelmar, 1983; Crecelius, 1985; Rose *et al.*, 1985; van der Sloot and Nieuwendijk, 1985; van der Sloot *et al.*, 1985; Kress, 1993).

TABLE II Metals concentration in representative fly-ash samples and in fly-ash collected from the dump-site

Sample	Date		Hg	Cd*	Cu	Zn	Pb	Mn	Fe	Al
			$\mu\text{g g}^{-1}$						wt%	
HFA		Avg	0.22	0.69	80	100	76	414	3.59	14.3
		std	0.06	0.12	7	14	15	29	0.29	0.4
		n	8	13	13	13	12	9	4	4
RFA <i>n</i> = 9		Avg	0.20	0.90	82	133	84	351	na	na
		std	0.12	0.26	11	34	16	57		
CFA	1993 <i>n</i> = 16	Avg	0.496	0.55	86.6	91.9	76.3	423	2.56	14.3
		std	0.194	0.17	14.5	20.6	8.2	34	0.40	1.2
	1994 <i>n</i> = 18	Avg	0.369	0.48	63.6	87.0	80.6	467	3.68	12.9
		std	0.152	0.34	11.2	34.1	21.3	59	0.91	2.2
	1995 <i>n</i> = 17	Avg	0.219	0.61	70.8	99.8	78.4	458	4.30	14.6
		std	0.075	0.24	6.6	33.9	18.5	40	0.89	1.9
A-CFA	A-1994 <i>n</i> = 8	Avg	0.223	0.29	68.5	63.4	96.7	371	3.84	15.7
		std	0.138	0.19	9.9	17.5	24.7	67	0.50	1.7
	A-1995 <i>n</i> = 5	Avg	0.246	0.42	74.3	77.2	93.5	477	3.79	16.2
		std	0.114	0.16	13.1	15.6	20.3	47	0.69	0.8
SFA <i>n</i> = 17		Avg	0.114	0.56	93.6	113	67.8	411	4.24	13.8
		std	0.049	0.12	26.5	22	8.5	45	0.52	1.4

na-not analyzed, Avg-average, std-standard deviation, *n*-number of samples.

* Corrected for incomplete recovery, see methods. HFA and RFA-Orot Rabin and Ruthenberg Power stations representative coal fly-ash, respectively; CFA-coal fly-ash from the sea; A CFA-CFA from station A; SFA fly-ash sampled from the ship, prior to dumping during the controlled field.

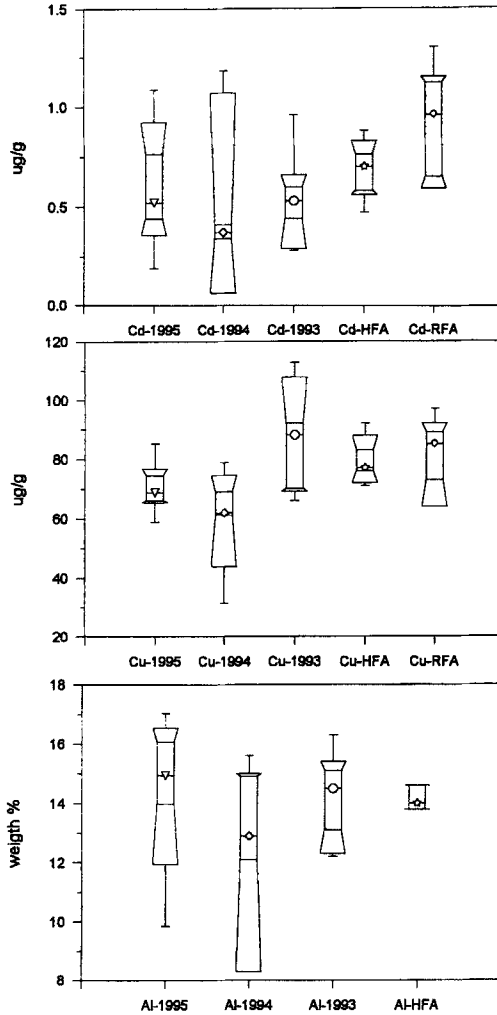


FIGURE 3 Box-Whisker plots of cadmium, copper and aluminium concentrations in coal fly-ash from the dump-site and in the representative coal fly-ash samples. The horizontal lines mark the minimum and maximum and the 10th, 25th, 50th, 75th and 90th percentile points.

In addition, a limited preliminary study performed on four 1993 CFA samples showed that calcium and silicon levels decreased while magnesium increased in the ash (Kress *et al.*, 1994). However, the results from the CFA collected from the sea were inconclusive.

Cadmium concentrations in samples from 1993 and 1994 were lower than in HFA (by 20% and 30%) while for the samples in 1995, the cadmium concentrations were not significantly different from HFA. The samples analyzed in 1995 were all collected in the area located to the southwest of the site (L5, L6, L19 and L20), the area we suspected had received CFA from the Ruthenberg power plant. Comparison between the cadmium concentrations in HFA and in representative samples from the Ruthenberg power plant (RFA) (Fig. 3, Tab. II) found the latter to be significantly higher. This comparison, together with the higher cadmium concentrations found in the 1995 samples, seems to support the assumption that the ash found since 1994 toward the southwest from the dump site originated from the Ruthenberg power plant. Consequently, comparison between the 1995 samples and RFA showed that cadmium, zinc and copper were lower (by 32%, 14% and 25%, respectively) and manganese higher (by 30%) in the 1995 samples than in RFA. Iron and aluminium concentrations are not available for RFA, and therefore were not compared.

This analysis had two drawbacks. The initial composition of the ash prior to dumping was not determined, and we used the mean concentration of HFA as the best available approximation of the initial concentration. However, most of the ash was exposed to rain and weather prior to dumping and it is reasonable to assume that some change in its composition occurred while still on land, although it was shown experimentally that further changes occur after contact with sea water (Kress, 1993; Shoham-Frider, 1997). Moreover, the exact elapsed time between dumping of CFA, that started in 1989, and the time of sampling is unknown and could range between < 1–6 years. In order to characterize better the changes occurring in the CFA, a controlled long term field experiment was initiated, where the initial composition of the CFA and the time of dumping were known.

Controlled Field Experiment

Site A in the dump-site (Fig. 2) was identified as having no CFA in the sediments, based on the dumping events location map (Kress *et al.*, 1993). During the month of October 1993, a total of 26,000 tons of CFA from the Ruthenberg power plant was dumped at site A, in 17 separate loads. CFA was sampled from the ship (referred here as SFA)

prior to each dumping and characterized (Tab. II). During 1994 and 1995, CFA aggregates were samples from site A and the concentrations of mercury, cadmium, lead, copper, zinc, manganese, iron and aluminium measured (Tab. II). *T*-test comparison between the A-1994 and SFA mean values showed that cadmium, zinc and copper were lower in the samples from the sea by 48%, 44% and 27%, respectively, while aluminium was higher by 14%. Except for aluminium, the changes that occurred in the ash from site A were similar to the changes observed in the CFA collected before, from different areas at the dump site. In 1995, only five samples were analyzed, a sample size that is too small for significant statistical analysis due to the high variations in concentrations found in the ash. The controlled field experiment will be expanded as an integral part of the monitoring programme.

CONCLUSION

CFA was found at the dump site as a layer of fine powder, accumulated on top of the sediment, and as small and large aggregates. It was distributed heterogeneously in the site and found also outside the designated dumping area. Apart from aluminium and manganese, mercury concentrations were found to be a possible indicator to the presence of CFA in the sediments. CFA composition changed in the sea; cadmium, copper and zinc concentrations in the ash decreased as a result of the prolonged contact with sea water at *in situ* conditions while inconclusive changes in mercury, iron and manganese were detected. A controlled field experiment now in progress is expected to clarify the questions that raised during this research.

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