Multivariate Approach to Distribution Patterns and Fate of Polycyclic Aromatic Hydrocarbons in Sediments from Smelter-Affected Norwegian Fjords and Coastal Waters

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The environmental significance of polycyclic aromatic hydrocarbons (PAHs) generated by the production of aluminum and manganese alloys is an important issue in numerous Norwegian fjords. The fate of these compounds have been elucidated by analyzing surface and subsurface sediment samples collected over a 15-year period from seven smelter-affected fjords applying principal component analysis (PCA). Consistent patterns within and between fjords and between production types suggested causal relationships and formed the basis for formulating hypotheses regarding the fate of the compounds. The PCA was able to differentiate between various combustion origins at receptor locations, so far hidden in the traditional treatment of the data restricted to individual fjords. This differentiation reflected industrial processes and most probably cleaning arrangements at the different plants. The source-specific signals were discernible over considerable distances and down to concentrations 3−4 times background levels. Compound-specific transformation reactions occurring during transport and incorporation into the sediments contributed little to the total variance and did not suppress the source-specific signals. This may imply a rather low availability to the bottom fauna and raise questions as to the potential harm inflicted by smelter-generated PAHs.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous contaminants of coastal marine sediments. PAHs arise from incomplete combustion of organic matter in flames, engines, and industrial processes and from emissions of noncombustion-derived petrogenic oils. PAHs also occur naturally from events such as forest fires and volcanic eruptions (*1*, *2*). The relative composition of the compounds, the PAH profile, is dependent on the origin, but the compounds may undergo specific biotic and abiotic transformation reactions that may lead to changes in the profiles. The environmental concern about PAHs is due to their potential to form carcinogenic and mutagenic diols and epoxides (*3*, *4*).

The metallurgical industry may constitute important sources of combustion-derived PAHs. These PAHs appear to be strongly particle associated. Compared to petroleum-derived PAHs, they are less available to desorption processes and presumably also less bioavailable (*5*, *6*). The microbial degradation of PAHs with three or less condensed rings is well documented. Less is known about microorganisms capable of utilizing higher molecular weight PAHs as a sole source of carbon and energy, although recent research has reported that this may apply for pyrene (*7*, *8*). In general, PAHs of higher molecular weight may be subject to co-metabolic transformations.

In Norway, aluminum and manganese alloy smelters discharge PAHs in effluent wastewater to marine recipients. In the Al plants, the PAH originates mainly from seawater scrubbing of ventilation air and pot fumes from production processes based on the Søderberg technology and, in some cases, also from the production of anodes. The PAH problem is mainly related to the use of coal tar pitch as a binder in the production of ramming paste and anode paste and the fabrication of anode and cathode blocks. In the Mn alloy smelters, the scrubbing of pot fumes is the main source of PAHs, and in these plants, coke used as a reduction agent is probably more important than pitch. The PAH is introduced to recipient waters through pipelines or from settling basins separated from seawater by sand and gravel dikes. Annual discharges have been estimated to be at least 1−10 t of PAH per smelter, but the figures have been reduced drastically in recent years. However, discharge data are uncertain due to the lack of standardized methods for sampling and analysis.

PAH contamination has been monitored in Norwegian recipient waters since the mid-1970s. In the sediments, parent PAH compounds, in particular fluoranthene, chrysene/triphenylene, benzoflouranthenes, benzopyrenes, and benzo[*ghi*]perylene, are quantitatively the most important (*9*). Generally, the monitoring has addressed total concentrations and occurrences of selected compounds, while little attention has been paid to patterns of distribution, although Bjørseth et al. (*10*) revealed distance-related profile changes in one fjord.

In the present study, monitoring data of surface and subsurface sediments collected over a 15-year period from a number of smelter-affected Norwegian fjords have been assembled and analyzed using principal component analysis (PCA) in a search for common patterns and trends. Particular emphasis has been placed on fjord comparisons. Consistent patterns suggest causal relationships and form the basis for formulating hypotheses regarding the fate of the compounds. Another important aspect of the analyses has been to discern and render visible minor variation patterns that are usually overlooked or viewed as noise in routine statistics. The use of PCA in evaluating environmental chemistry data, e.g., for source recognition and transport pathways, has recently been encouraged (*11*, *12*).

Sampling, Chemical Analysis, and Data

Data from seven smelter-influenced fjords and coastal areas (four Al, three Mn alloy) were selected for the present study (Figure 1, Table 1). Most Norwegian smelters are located near the heads of fjords or on semi-enclosed coastal waters. In total, the data comprise 199 surface and 41 subsurface stations, covering water depths from 8 to 570 m and spanning distances from immediate proximity to 46 km from the effluent outfall points. The sediment particle composition at the

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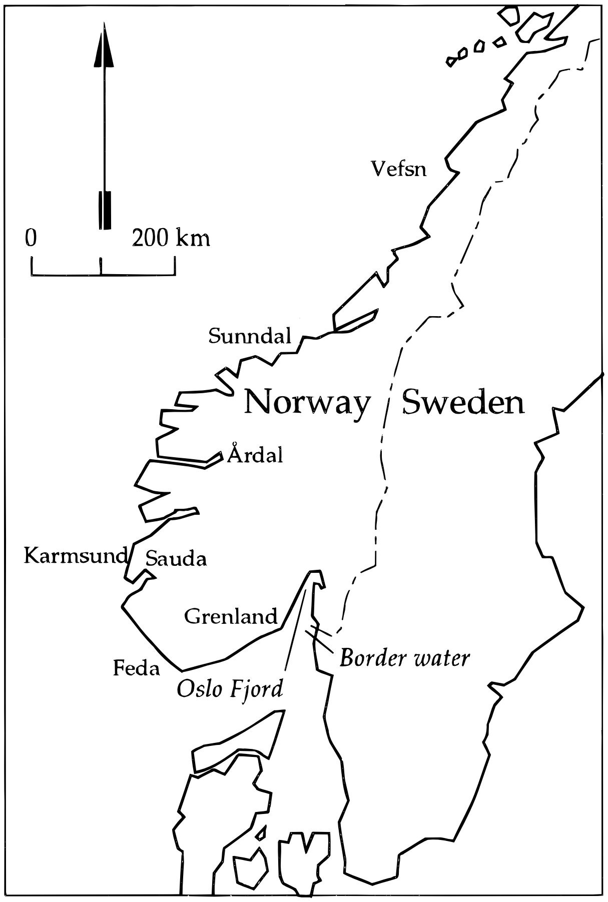

FIGURE 1. Smelter locations and sampling sites. The Oslo Fjord and border waters between Norway and Sweden are unaffected by smelter point sources.

stations ranged from sand to almost pure silt and clay; however, the grain size variations were minimized by sampling the fine-grained sediments in the deeper parts of the fjords. Most samples were taken using a 5 cm i.d. gravity corer. The cores were sliced into 1−2 cm sections, and three−eight replicate cores were pooled from each station to ensure enough material.

PAH concentrations were determined by GC−FID or GC−MSD equipped with a split/splitless injector and a 25 m × 0.25 mm i.d. DB-5 fused silica (FID) or a 12 m × 0.2 mm i.d. HP-1 column (MSD) after Soxhlet extraction of freeze-dried sediment with cyclohexane. The GC was calibrated using the Supelco standard containing 16 individual PAHs. The relative accuracies were checked against SRM 1941 from NIST (U.S. National Institute of Standards and Technology). The detection limits depend on sample amounts, final extract volumes, and instrument conditions but were normally 0.5 ng g^{-1} dry weight for individual PAHs. Details of the methods are given by Næs et al. (*9*).

The number of quantified PAHs varied from fjord to fjord in the original monitoring programs. The present analyses were performed on 13 compounds that had been measured regularly and dominated the sedimentary profiles: phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benz[*a*]anthracene (BaA), chrysene/triphenylene (Chr), benzo[*b*+*j*+*k*]fluoranthene (BFs), benzo[*e*]pyrene (BeP), benzo[*a*]pyrene (BaP), perylene (Per), indeno[1,2,3-*cd*]pyrene (Ind), dibenz[*a,c*/*a,h*]anthracene (DBA), and benzo[*ghi*]perylene (BP). However, occasionally a few values were missing (not analyzed, below the detection limit) in which cases the compounds concerned had to be omitted from analysis. Prior to the final PCAs, preliminary analyses were performed to unveil possible chemical analysis errors and to identify outliers. The data span a wide range of concentrations from background to nearly 800 000 ng g^{-1} dw of PAH_{13} (= sum of the 13 compounds) (Table 1). A value of 300 ng g^{-1} dw is set by the Norwegian sediment quality criteria (*13*) as the upper background limit for fine-grained fjord sediments not influenced by point sources. An overview of the occurrence of PAH in sediments and organisms in Norwegian marine waters affected by Al smelters may be found in Næs et al. (*9*).

PCA [CANOCO version 3.10 software (*14*)] were used to apportion the variance in the data sets. The analyses were performed as "ordinary PCA" (variable-centered) and "double-centered PCA" (variable- and sample-centered) on log-transformed data. The latter option is comparable to standardized PCA by eliminating the effects of scale, thus focusing on relative patterns. The PCA results are presented in combined plots (biplots) [CanoDraw 3.0 software (*15*)] of variables and stations. The stations are shown as points, while the variables are represented by arrows running from the origin of the plot to the position of the variable scores (loadings). The direction of an arrow represents the direction of increasing variable concentration across the stations, while the length represents the strength of the increase. The angle between pairs of arrows indicates the approximate correlation between variables.

Results

Surface Sediments. An obvious trend in all fjord data sets is a decline in total concentrations with distance from the effluent outfall. In ordinary PCAs for the individual fjords, this appears as a very strong first axis (PC1) capturing more than 80% of the total variance (Table 2). The successive axes (PC2, PC3) consequently represent minor components of the total variation. These variance components, however, may be interpreted as profile patterns superimposed on the concentration gradient and may thus reflect source signals and processes transforming individual PAHs in the recipient waters.

In order to focus on the profile patterns, the effects of concentrations were deducted from subsequent analyses, which were performed double centered. Figure 2 shows analyses of surface sediments in fjords influenced from Al and Mn alloy smelters, respectively. The Al fjord data (Årdal, Sunndal, Karmsund, Vefsn) comprised more than a 100 stations and also some replicate samples (Figure 2A). One sample series (Vefsn 1989) was omitted because it showed large and apparently unsystematic variations that suppressed the patterns among other samples. In the plot, the samples formed groups for fjords, to some extent (e.g., Årdal) also for sample series from different years. This clustering of samples according to constituent data sets indicates that each fjord had a typical PAH profile, in some cases even each sampling. Looking at the samples in relation to the variable pattern (PAHs) allows a characterization of the profiles, showing for instance that Karmsund, Vefsn (1978), and Årdal (1983) had a relative dominance of lighter weight compounds, while the second series from Årdal (1989) had a higher representation of heavy weight compounds. These characteristic profiles must actually be discernible along the entire PAH concentration gradient in the fjords. There was a tendency, however, for stations occupying the area near the mouth of the fjords to be displaced to the right in the plot (not illustrated), indicating a relative increase of lighter PAHs as compared to stations near the source.

The individual PAH compounds were arranged in a sequence that roughly followed their molecular weights from light to heavy. This arrangement implies that compounds with similar molecular weights had similar distributions across the stations (high correlations), while compounds of different molecular weights varied proportionate to the size of the difference. The strongest trend (PC1) separated the light and

TABLE 1. Summary Data for Stations Sampled in Different Fjord Areas

fjord area	receiving water	sampling year	no. of stations (core stations)	distance from source, km^a	water depth, m^a	sediment section (depth), cm^{a,b}	$\text{PAH}_{13}^{a,c}$ ng g^{-1} dw
Årdal	narrow fjord	1983	9 (2)	0.6−15	84−570	0−1 (20)	3 401−784 296
		1989	21	0.6−3.5	48−165	0−1	
Sunndal	open fjord	1986	21 (1)	0.1−44	16−345	0−1 (14)	341−772 506
		1990	10	0.5−3.2	82−123	0−2	
Karmsund	coastal sound	1988	16 (2)	1.1−11	13−93	0−1 (18)	8 500−88 700
Vefsn	open fjord	1978	6	2.5−20	159−485	0−2	
		1984	8 (8)	1.8−12	39−458	0−1 (3)	684−173 590
		1989	10 (2)	1.8−20	35−487	0−1 (10)	
Feda	narrow fjord	1984	13 (1)	0.1−19	23−384	0−2 (10)	357−144 858
Sauda	open fjord	1976	6 (3)	0.3−20	80−500	0−2 (8)	
		1981	11 (1)	0.3−20	40−375	0−1 (20)	158−499 401
		1986	18 (2)	0.3−20	84−322	0−1 (5)	
		1990	10	0.4−2.5	55−178	0−2	
Grenland	narrow fjord	1986	14	2.3−20	22−113	0−2	342−60 572
		1989	26 (19)	1.1−46	8−217	0−2 (18)	

[a] Distance from source, water depth of stations, and sum of 13 PAHs are given as ranges. [b] Sediment section refers to the thickness of the analyzed sediment slice, depth (in parentheses) to the subbottom depth of the deepest slice. [c] See text for definition of PAH_{13}.

TABLE 2. Data for Ordinary PCAs of PAHs in Separate Fjord Areas

fjord area	variable excluded	no. of stations included	variance explained (%)		
			PC1	PC2	PC3
Årdal		26	87	10	2
Sunndal	Per	26	95	3	1
Karmsund	Ant, DBA	16	88	5	4
Vefsn	Ant, Per, DBA	20	83	9	4
Feda	BeP	9	97	2	0.5
Sauda	Per	39	89	6	3
Grenland	Per, DBA	32	87	8	2

heavy compounds, which appeared to show a negative correlation and diverged on a relative scale. Most medium weight compounds deviated from the pattern and were fitted on PC2, which was most distinct for triphenylene/chrysene (Table 3).

The Mn alloy fjord data (Feda, Sauda, Grenland) comprised nearly 100 stations (Figure 2B). Some stations in Grenland located near petrochemical industry were omitted. These stations were markedly dominated by lighter compounds that suppressed the dissolution in the plot. The PAHs were arranged in a pattern similar to that found in the Al fjords, but the samples were less clearly grouped for fjords, although Sauda and Grenland tended to be separated. The latter showed a relative preponderance of light and medium weight PAHs.

Figure 2C shows an analysis for Al and Mn alloy fjord data put together. Most samples were grouped according to type of production. The Al sites tended to have more light and medium weight PAHs, while Mn alloy sites tended to have more heavy PAHs. The groups may be almost separated by projecting the site points on an axis running from the lower left to the upper right in the direction of the arrows for indeno-[1,2,3-*cd*]pyrene and benzo[*ghi*]perylene on the one side and pyrene and benz[*a*]anthracene on the other. The results suggest that for the present data selection these compounds were particularly typical of the type of smelter and that their relative amounts may be used to distinguish between profiles.

Considering that source-related profiles were detectable for the fjords, further comparisons were made with areas that lacked point sources. Figure 3 shows smelter fjord profiles compared to profiles from the Oslo Fjord (3 stations) and the open coast border waters between Norway and Sweden (9 stations). Both areas represent environments that are moderately influenced by general human activity (mean PAH_{13} < 2000 ng g^{-1}). The most strongly contaminated fjord stations (>5000 ng g^{-1} PAH_{13}) were omitted to restrict the analysis to samples with comparable PAH concentrations. It appeared that the smelter-affected fjords may generally be distinguished from the diffusely affected areas. The samples from Vefsn, the Oslo Fjord, and the border waters formed clear groups, while the Mn alloy fjords (Sauda, Feda, Grenland) formed less distinct groups and partly overlapped with the border waters. The difference between the Oslo Fjord and the border waters suggests that diffusely affected areas may also have characteristic profile patterns.

Downcore Profiles. The data for downcore profiles comprised 68 samples (core sections) from 18 stations in six fjords (Årdal, Feda, Karmsund, Sauda, Sunndal, Vefsn). Most samples were grouped according to fjords (Figure 4). Further, within fjords there was a clear tendency to group sections according to stations and sampling years. As in the analyses of surface sediments, PC1 mainly fitted the lower and higher molecular weight compounds, while PC2 fitted the intermediate molecular weight compounds. Benzo[*a*]pyrene deviated somewhat and correlated most strongly with PC3. The Mn alloy fjords (Feda, Sauda) were associated with the heavier PAHs.

Discussion

The amount and distribution of PAHs in the fjord sediments depend on the effluents involved, transport, and sedimentation as well as on possible transformation reactions. The present study demonstrates that most fjords had a characteristic PAH profile or "fingerprint". The fingerprints were particularly clear for the Al smelter-influenced fjords, where also different samplings could be distinguished. Aluminum production could be distinctly separated from manganese alloy production. The patterns were also evident downcore in the sediments in the fjords. Comparison with areas without smelter point sources indicated that the profile patterns were discernible down to 3−4 times background levels.

The systematic nature of the fingerprint patterns suggests that these reflect source characteristics. There is no indication that the patterns could be artifacts from analytical performance, considering also that all chemical analyses were performed by the same laboratory. Verification of the source characteristics can hardly be made, however, because discharge data for the smelters are scarce and mainly represent total PAH levels. Yet, some information may be deduced on the basis of knowledge about the production processes and cleaning arrangements. In the Al smelters there are various installations for cleaning of emissions, e.g., scrubbing of ventilation air and pot fume, dry cleaning with alumina, and electrostatic precipitation (*16*). Generally, scrubbers are efficient for retaining large- and medium-sized particles and

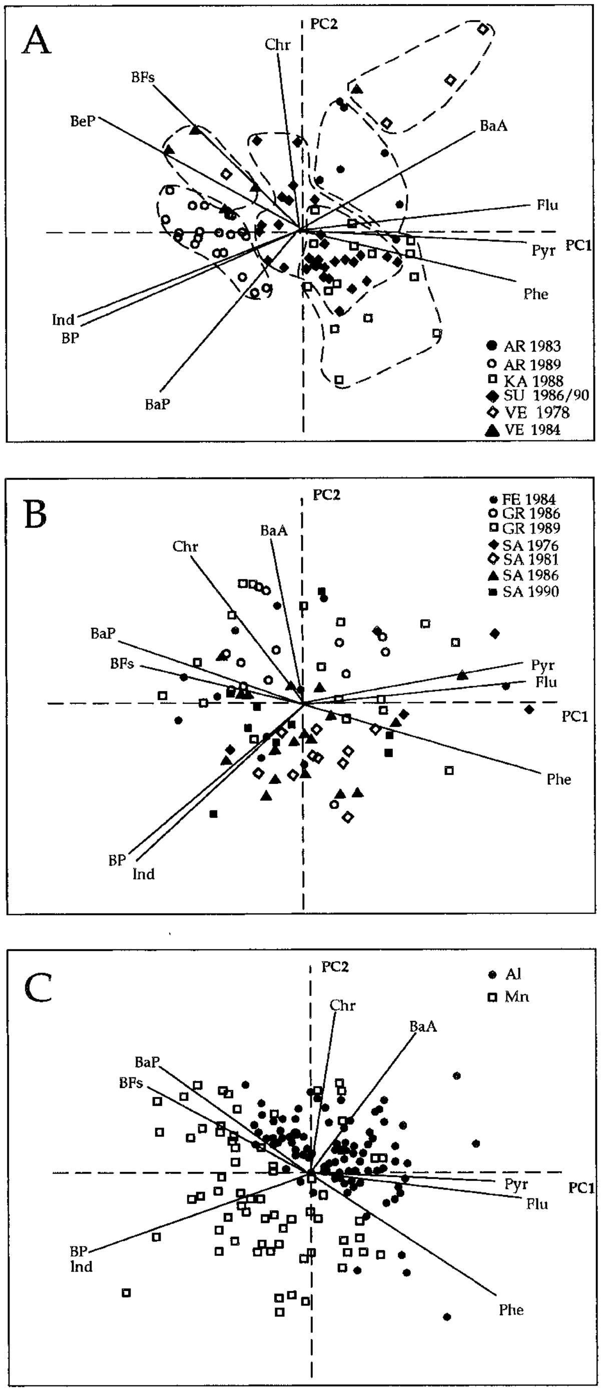

FIGURE 2. Double-centered PCA for inter-fjord comparison. (A) Al smelter-influenced stations, number of samples = 82, PC1 and PC2 account for 51 and 16% of variance, respectively. (B) Mn alloy smelter-influenced stations (*N* = 83, PC1 = 48%, PC2 = 21). (C) Combined Al and Mn alloy smelter-influenced stations (*N* = 159, PC1 = 49%, PC2 = 19). Abbreviations: AR, Årdal; KA, Karmsund; SU, Sunndal; VE, Vefsn; FE, Feda; GR, Grenland; SA, Sauda.

inorganic compounds, while electrostatic precipitation is efficient for the removal of small particles and organic compounds (*17*). Retention of individual PAHs therefore may be supposed to vary, essentially more or less in relation to molecular weight as the heavier PAHs generally have the strongest affinity for particles. The efficiency may also gradually change with time. For instance, in Årdal the efficiency of the electrostatic precipitation declined during the 1980s (information from the plant), which may explain the transition to profiles dominated by higher molecular weights from 1983 to 1989. Profile differences could also stem from the PAH source material (coal tar pitch). Differences in pitch type and softening point influence the PAH profile (*18*), but there is no evidence that this caused systematic differences in the effluent PAH.

Fjords influenced by Mn alloy smelters were less clearly differentiated than the Al fjords, although most Grenland samples were distinguishable from Sauda samples. Mn alloy smelters have similar cleaning systems, which translates into similar routes of PAH entry into the recipient waters. The smelters also have the same central supplier of anodes. Accordingly, there are no particular source characteristics among the smelters that would be expected to cause different effluent profiles. The differences in the Grenland samples

TABLE 3. Cumulative Variance Fit for Individual PAHs in PCA for Al Smelter-Influenced Fjords

fraction fitted	PC1	PC2	PC3	PC4
variable	0.51	0.16	0.09	0.08
Phe	0.75	0.81	0.98	0.98
Flu	0.77	0.78	0.83	0.88
Pyr	0.62	0.62	0.79	0.89
BaA	0.12	0.17	0.23	0.60
Chr	0.01	0.50	0.56	0.56
BFs	0.23	0.54	0.77	0.77
BeP	0.49	0.72	0.73	0.73
BaP	0.20	0.53	0.60	0.84
Ind	0.62	0.79	0.79	0.81
BP	0.67	0.82	0.83	0.93

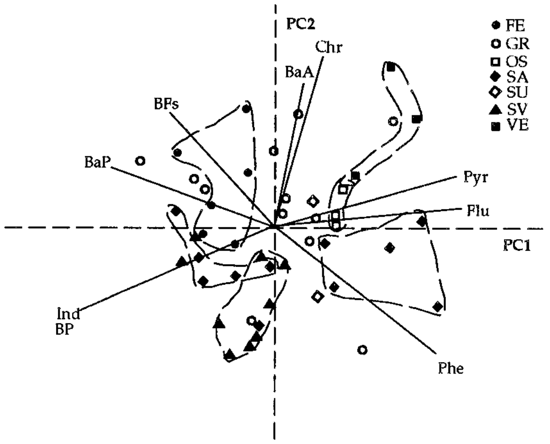

FIGURE 3. Double-centered PCAs of smelter-influenced stations with PAH_{13} concentrations of less than 5000 ng g^{-1} combined with stations unaffected by point sources (*N* = 47, PC1 = 54%, PC2 = 19%). OS, Oslo fjord; SV, border waters Norway/Sweden.

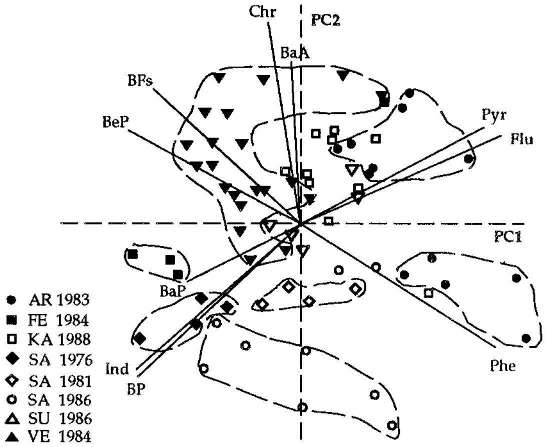

FIGURE 4. Double-centered PCA for downcore samples (*N* = 68, PC1 = 30%, PC2 = 30%).

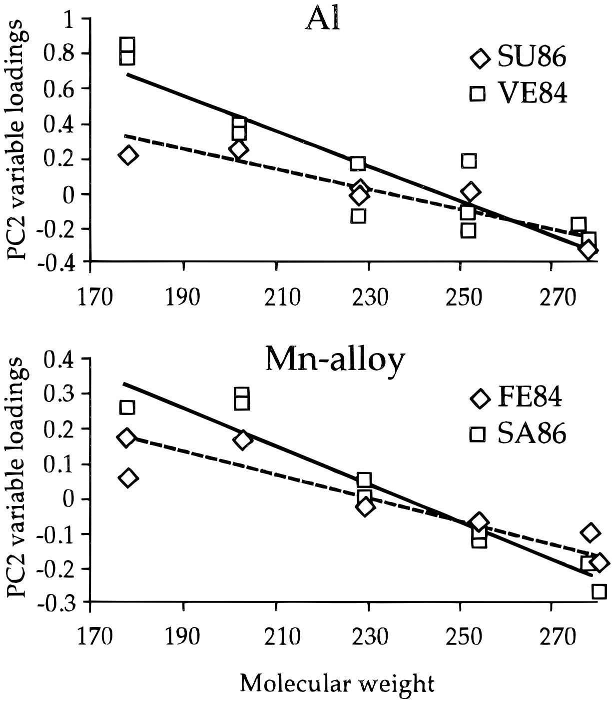

FIGURE 5. Plot of molecular weights of the individual PAH compounds against PC2 coordinates based on variable-centered PCA on individual fjord data sets. Fjords affected by aluminum production (Al) include Vefsn sampled in 1984 and Sunndal in 1986. Fjords affected by manganese alloy production (Mn) include Feda sampled in 1984 and Sauda in 1986. All regression lines are significant at $p < 0.001$.

probably reflect influences from other sources in the area, in particular PAHs of petrogenic origin dominated by low molecular weight PAHs which is quite typical of fossil fuel sources (*19*). Grenland is the most heavily industrialized area of Norway, receiving numerous anthropogenic inputs.

Comparisons of the fjords with different production types indicated that the Mn alloy industry generates a PAH profile dominated by higher molecular weights. Presumably, this is due to temperature differences in the production processes. In Mn alloy smelters, PAHs originate in high temperature pot rooms (1200−1500 °C), while in Al smelters, the PAH generating processes (evaporation from anode, anode production) operate at far lower temperatures. Higher temperatures will facilitate the evaporation of PAHs of higher molecular weights, thereby shifting the profile toward heavier compounds (*20*). It is also possible that the pH of the effluent waters (approximately 7 in Al production as compared to 10 in Mn alloy) affects the profiles as PAH degradation and sorption to particles can be pH dependent (*21*). However, nothing is known about the significance of such processes in effluent waters.

The arrangement of the variables (PAHs) in the analyses reflects the variation of the individual PAHs in the recipient waters. All analyses depicted the PAHs in a consistent pattern that can be related to molecular weight. Generally, molecular weight may be taken as a descriptor of structure−property and structure−activity relationships (*22*). Most probably, the variable patterns therefore represent processes operating on the PAHs in the sea. In the present analyses combining data set from different fjords and samplings, however, source differences may confound the individual variation, but the variable pattern also emerges in analyses of single sample series. This is demonstrated in Figure 5 showing variable scores (loadings) related to the molecular weight of the compounds. The illustrated results are from ordinary PCAs (see Table 2) in which PC2 captures the pattern appearing on PC1/PC2 in the double-centered PCAs. The compound-specific processes may operate during transport, during sedimentation, and after incorporation in the sediments. Various studies have shown that PAHs in seawater may be transported as dissolved (truly and/or apparent), sorbed to natural organic matter such as humics, or associated with different-sized particles (*23*, *24*). Thrane (*17*) and Bjørseth (*20*) showed that in the Al smelters the PAHs were present in both the gaseous and particulate phases of ventilation air. Hence, in the smelter effluents, the PAHs may occur both dissolved and associated with particles, which suggest subsequent selective transport in the fjord. Correspondingly, this could account for the general seaward shift toward a lighter profile in the fjords. Similar findings are supported by studies elsewhere (*10*, *25*, *26*).

In the sediments, microorganisms may degrade PAHs and alter the profile (*7*). The analyses of downcore profiles, however, demonstrated also that subsurface samples retained a profile related to source characteristics. These results contrast with experiments, mostly in the laboratory, showing that PAH may be biodegraded after incorporation into the sediments (*8*, *21*, *27*, *28*). However, disagreement in laboratory-predicted half-lives and field observation data may be a consequence of differences in environmental factors (*29*, *30*). Also, the binding type of the PAH molecule and its associated matrix is of fundamental importance. The PAHs in the sediments are probably to a great extend bound to oxides and black carbon-containing particles (soot). The latter association is known to render the PAHs resistant to degradation and also to imply that desorption and repartitioning into the porewater is strongly restricted (*5*, *6*, *31*).

The PCAs were able to differentiate between various combustion origins at receptor locations, so far hidden in the traditional treatment of the monitoring data restricted to individual fjords. In the litterature, various approaches have been proposed for PAH source recognition, including alkyl homologue series, binary PAH ratios, and parent PAH profiles (*19*, *20*, *32*). The results from the present data show that parent PAH profiles analyzed with multivariate data processing tools such as PCA are powerful means of source resolution and could also facilitate the early detection of sediment responses to changes in input. The consistency of the source-specific signals shows that the PAHs were resistant to or have a severely restricted potential for natural post-depositional transformation reactions. This may imply a rather low availability to the bottom fauna and raise questions as to the potential harm inflicted by smelter-generated PAHs. Studies of benthic communities in the fjords have indicated that the ecological effects are small (*33*), but further studies regarding PAH uptake and their metabolization by benthic organisms are needed.

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