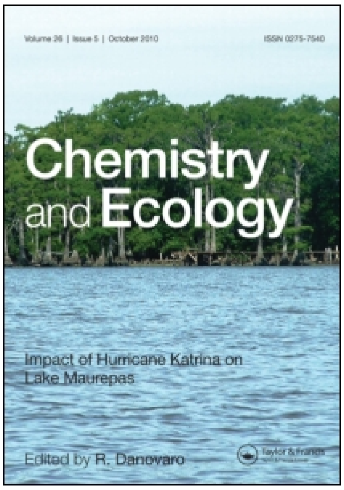

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Contaminants, benthic communities, and bioturbation: potential for PAH mobilisation in Arctic sediments

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Marine benthic fauna and biological mixing were studied in relation to sediment organic enrichment and polyaromatic hydrocarbons (PAHs) in bottom sediments of Svalbard. We investigated how organic enrichment may affect the fate and chemical composition of deposited contaminants by impacting biological reworking by faunal communities. Samples were collected near active coal mines at Barentsburg and at the mouth of Grønfjord. PAH sources in both areas were coal particles and pyrolytic compounds from coal-driven power stations. The results from a bioturbation experiment were consistent with the hypothesis that fauna enhance the vertical transport of PAHs within the sediment. Faunal community composition was similar at the two sites, with polychaete worms comprising $>$ 85% of the fauna. Abundances and taxon richness were eight and ten times higher in the organically enriched sediments near Barentsburg, and total PAH concentrations were up to three times higher in Barentsburg. Unlike expectations derived from models developed for temperate regions, organic enrichment in oligotrophic areas, such as this Arctic site, enhanced the biomass and bioturbation potential of benthic communities. Hence, new insights into the relationships among enrichment, benthic communities and the fate of contaminants must be considered in management and regulatory efforts in these areas.

Keywords: coal; eutrophication; fjord; sediment mixing; Svalbard

1. Introduction

Polyaromatic hydrocarbons (PAHs) from petroleum and coal sources, and from combustion processes, are a widely distributed contaminant group in marine sediments around the world. Sources and transport mechanisms are becoming better understood, and there has been considerable effort to document levels in organisms and within relevant matrices (e.g. water, fauna, sediments) [1]. PAHs have been described as having serious impact on fauna, where they act as mutagens, cause DNA damage and disrupt membrane function [2,3]. Tolerance in some organisms at lower trophic levels may result in these compounds being passed to higher trophic levels where

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their negative effects may endanger fish, birds and mammals [4]. The effects are often not only compound- (and taxon-) specific, but may also vary across environmental gradients.

Analysis of the circumpolar distribution of PAHs in bottom sediments has shown that total parent PAH concentrations can differ by three orders of magnitude from 3.9 to 3632 $ng \cdot g^{-1}$ dry weight. The highest levels of total parent PAHs (2144 $ng \cdot g^{-1}$ dry weight on average) are typical for Svalbard inshore bottom sediments [5]. Such extremely high PAH contamination is a result of rock erosion in coal-rich areas and the seepage of oil hydrocarbons [6]. The interrelation between Svalbard bottom sediments and possible contamination sources with coal-enriched formations of various geneses (clarain and sapropel coal) was shown on the basis of a comparative study of organic matter. The similarities in material and mineralogical structures, level of catagenetic transformation and the structure and distribution of *n*-alkanes and PAH composition have confirmed a general similarity in their geneses [7].

Inventories of PAHs and most contaminants alone, however, do not provide a comprehensive insight into their potential biological impacts. Highest concentrations often occur in sediments, but these environments are not a static sink and instead may be the location of intense recycling, transport and remobilisation both within sediments and to the overlying water. Some fauna tolerate PAHs, whereas others are capable of degrading at least some of the lower molecular mass compounds [2,8–10]. This may occur directly or via stimulation of microbial activity [11], and degradation in sediments populated by benthic fauna can be more than two times higher than in defaunated sediments [10,12]. This suggests that modelling impacts and the fate of PAHs should take into account the complex suite of mechanisms through which these compounds act and are influenced. These mechanisms are relatively poorly understood in most areas, but especially in oligotrophic systems such as open continental shelf habitats and high latitude regions.

Bioturbation, the mixing of sediment particles and solutes through the activities of infauna, is one of the most important factors controlling the fate of contaminants in sediments [12], because these activities enhance pore water oxygenation and keep contaminants in contact with active fauna [10,12]. In general, when PAHs and other contaminants are beneath the bioturbation layer, they are likely to enter long-term storage within sediments. Clearly, then, the fate of contaminants in sediments depends on the composition of the benthic community, and thus the type and size of the bioturbators present, as well as the sedimentation rate. These factors, which are affected by many environmental agents, particularly organic load, will control the speed at which compounds are recycled or enter the deeper layers of the sediment.

Although long-distance transport can be an important mechanism affecting the distribution of PAHs in sediments, inputs are generally concentrated in coastal areas with high industrial, agricultural and/or municipal discharges [13], and PAHs tend to remain close to sites of deposition [14]. This often results in high levels of organic enrichment accompanying contaminant discharge. Organic loading results in characteristic changes in chemical and biological properties. Along a gradient of increased organic loading, diverse communities including large bioturbating organisms, tend to become replaced by denser communities of smaller organisms, thereby reducing the depth of biological sediment reworking [15]. As the intensity of enrichment increases, enhanced microbial and faunal activity depletes oxygen levels in bottom waters with accompanying effects on sediment communities. In extreme cases, but cases that are occurring more frequently and across wider geographical areas, bottom-water anoxia leads to ‘dead zones’ where few organisms survive [16]. Coastal and shelf sediments in many tropical and temperate areas receive inputs of organic matter from a wide variety of natural and anthropogenic sources. Even moderate organic enrichment (eutrophication), therefore, can result in degradation of the benthic fauna, decreasing community bioturbation and their mixing of sediment and pore waters, and reduced cycling/remobilisation of contaminants.

However, oligotrophic areas with low sediment organic content, such as sandy shelf sediments, may respond differently to organic loading. When temperatures are low and microbial activity possibly reduced, such as in high latitude areas, organic loading may not necessarily lead to depletion of bottom-water oxygen content and associated faunal changes [17]. Arctic benthic communities are generally assumed to be food limited [18–20], and here we may expect that organic enrichment could, to a point, stimulate communities of infauna. Therefore, the cycling and fates of PAHs mediated by bioturbation, degradation and burial dynamics may be different in these areas compared with many other regions of the world's coastal ocean.

In this study, therefore, we ask the following questions: What are the down-core concentrations of PAHs in sediments within an Arctic fjord system varying in organic input? How does organic content affect benthic communities at two fjord locations? How does bioturbation by communities in areas of different enrichment vary? And finally, what are the consequences for interpreting inventories, burial rates and the ultimate mobility of PAHs, and, potentially, other sediment contaminants? This study provides insight into the potential implications of bioturbation at a high Arctic study site. The study evaluates the applicability to an oligotrophic Arctic region of a common ecological paradigm relating organic enrichment to benthic community structure and function developed for temperate systems with moderate to high organic loads.

2. Methods

Fieldwork was carried out in the Isfjorden complex, Svalbard, onboard R/V *Jan Mayen* in September 1998 for PAHs and September 2007 for benthic communities and bioturbation. Two stations were sampled in 2007, one in Grønfjord, close to the mining settlement of Barentsburg, and the other in Forlandsund, just outside the entrance to Isfjord. In this article we use both the geographical name and the corresponding station number from Cochrane et al. [21] (Figure 1). In 2007 we were unable to resample Forlandsund, and instead collected samples from Grønfjord and a station in the centre of Isfjord just outside the entrance to Grønfjord (Figure 1 and Table 1). Coal-fired power stations and coal terminals are located in Barentsburg and Longyearbyen, and both communities discharge sewage and industrial waste into the fjord. Of the stations sampled, the Grønfjord station receives considerably more organic material and contaminants than either the Isfjord or Forlandsund stations [21].

Samples for PAH determinations were collected using a four-tube multicorer with hydraulic penetration (Bowers & Connelly multicorer). Samples in tubes were preserved by freezing at -20°C until analysis. Analyses were performed from the following core intervals: 0–5, 5–10, 10–12, 12–18 and 25–37 cm; and 0–3, 3–10, 10–18, 18–23, 23–28 and 28–33 cm for both Forlandsund and Grønfjord. In these slices, the following PAHs were determined: fluoranthene (Fl), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b,j]fluoranthene (BbjF), benzo[a]pyrene (BaP), benzo[e]pyrene (BeP), indeno[1,2,3-cd]pyrene (IndP) and benzo[h,g,i]perylene (BghiP). Analyses were performed by the analytical laboratory of the All-Russia Research Institute for Geology and Mineral Resources of the World Ocean (VNIIOkeangeologia).

The analysis was performed using a GC/MS Hewlett Packard 5890/5972. Calibration of the instrument was carried out using a standard mixture of PAHs (CLPS-B, Protocol Analytical Supplies Inc). Instrument linearity was monitored by calibration with five points in the concentration range 50–5000 ng·mL^{-1}. After running a batch of samples, calibration was monitored by analysis of a mid-level standard solution. The results of the analyses were processed using the software package Chemstation [7]. The proportions of the less stable or kinetic PAH isomers relative to the more stable or thermodynamic isomers (PAH ratios) were used to identify PAH sources. The best potential to distinguish natural and anthropogenic sources is exhibited by ratios of the principal mass 202, 228, 252 and 276 parent PAHs [22].

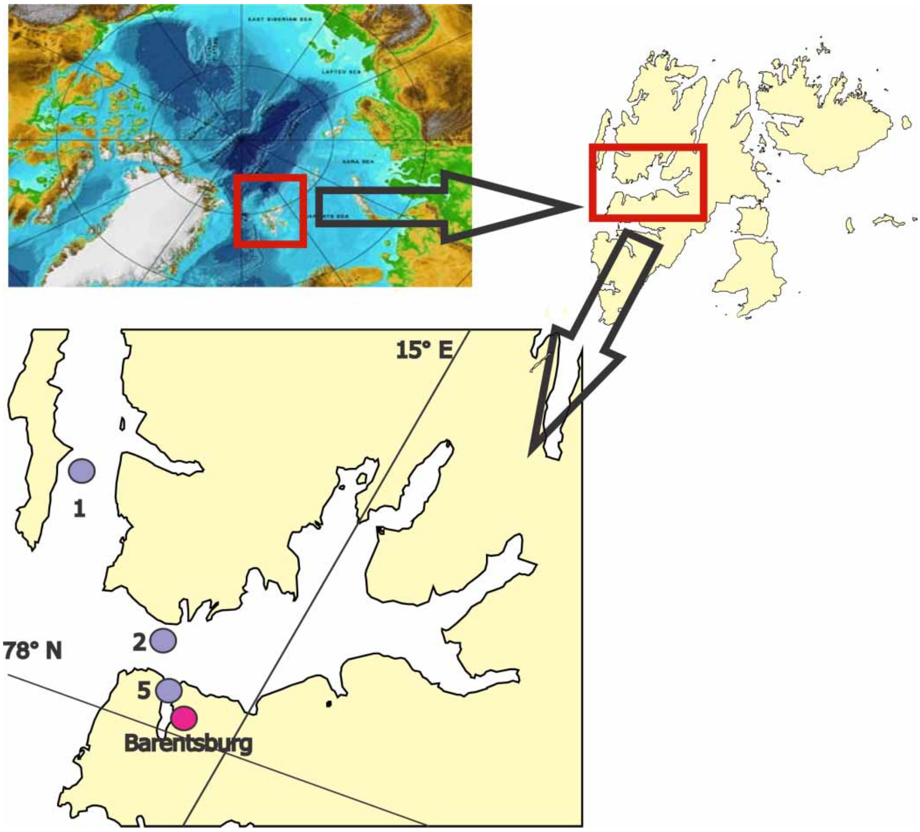

Figure 1. Map of the Isfjorden complex, including sampling stations (numbers) in Grønfjord (5), Isfjord (2) and Forlandsund (1).

Table 1. Positions, sampling details, and sediment information for cores collected in the Isfjorden complex for this study.

Station	Latitude (N)	Longitude (E)	Water depth	Organic carbon	Sed. rate	Bioturb. depth
Forlandsund*	78° 24.5′	12° 07.4′	242	1.7[a]	2.4[a]	9[a]
Isfjord	78° 09.5′	13° 49.4′	420	1.8[b]	(1.5[a])	(5[a])
Grønfjord	78° 04.1′	14° 09.2′	145	2.1 – 2.7[a,b]	5.8[a]	11[a]

Notes: Water depth values in m, organic carbon values are given in percent (w/w), sedimentation rate in mm·y^{-1} and bioturbation depth in cm. References for sediment data: [a]Cochrane et al. [21], [b]Killie et al. [23]. The sedimentation and bioturbation data for Isfjord are taken from a 300 m deep station from the centre of the fjord ∼ 25 km from this study's location. Data for Forlandsund were collected in 1998.

Benthic fauna were collected using a van Veen grab with a sampling area of 0.1 m^2, with three replicates taken at each station. Samples were washed through a 1 mm diameter round-pore sieve and fixed in a 10% formalin/seawater solution. Sampling and sample processing followed standard procedures (ISO 16665). The dominant taxa, Polychaeta and Bivalvia, were identified to the lowest possible taxonomic level, whereas the remaining taxa were identified at higher levels.

Material for the bioturbation experiment was collected using a box corer (0.25 m^2 sampling area). Subcores were collected at each location in plexiglass core tubes with an inner diameter of 12.5 cm and a penetration depth of 18–37 cm. The cores were aerated and kept refrigerated at 0 °C for 12 h after transfer to the laboratory to allow any sediment resuspended during transport to settle. Thereafter, 0.5 g glass beads of ∼ 0.01 mm diameter were added to the surface sediments in each core. Vertical penetration of the beads down the sediment profile was measured by sectioning the cores into 10 depth intervals (0–0.5, 0.5–1, 1–2, 2–3, 3–4, 4–5, 5–7, 7–10, 10–15 and 15–20 cm). Each sediment slice was homogenised and the number of beads within a subsample of 0.100 g wet sediment was quantified using light microscopy. Five such subsamples were analysed from 0 to 2 cm; at deeper intervals, three subsamples were analysed. One control core from each station was taken for analysis immediately after the introduction of glass beads, and two cores from each station were sectioned after 5 and 10 days, respectively.

3. Results

3.1. *PAH*

The total PAH concentration in sediment core slices from the two stations ranged from 629 to 1060 ng·g^{-1} dry weight (Forlandsund) and from 1781 to 2792 ng·g^{-1} dry weight (Grønfjord) (Table 2). In both cores there was evidence of a subsurface minimum in PAH around 5–10 cm deep in the core, with a subsequent decrease in concentration with depth below 15 cm (Figure 2). The highest PAH levels were found in the sampling site located close to the mining settlement of Barentsburg (Figure 1). In general, PAH ratios were low and consistent with depth to 20 cm (Figure 3).

3.2. *Fauna*

Faunal communities at the two stations were markedly distinct in their abundances and total taxonomic richness. In Grønfjord, a total of 1576 individuals were collected from the 0.3 m^2 sampled, comprising 58 benthic taxa. In Isfjorden, however, density and richness were considerably lower (262 individuals·m^{-2} in 30 taxa). Despite this, community composition at higher taxonomic levels was similar (Table 3), with communities at both sites dominated by polychaetes, both in terms of per cent abundance and taxonomic richness. Polychaetes accounted for 88.5%

Table 2. Concentrations (ng·g^{-1} dry weight) of higher molecular mass (g·mol^{-1}) polyaromatic hydrocarbons (PAHs), and PAH ratios, in slices from cores collected from Forlandsund (Station 1) and Grønfjord (Station 5).

		Core slices (cm)											
		Forlandsund						Grønfjord					
PAHs	Molecular mass	0–5	5–10	10–12	12–18	18–25	25–37	0–3	3–10	10–18	18–23	23–28	28–33
Fl	202	131	140	112	117	102	63	274	200	272	198	176	139
Pyr	202	107	109	89	87	82	78	354	270	330	264	267	223
BaA	228	22.4	26.1	18.2	20.4	18.5	13.8	97.6	76.2	88.1	63.1	61.3	51.4
Chr	228	248	256	206	200	188	166	589	422	535	435	431	376
BbjF	252	176	192	159	157	139	96	435	319	405	320	296	265
BeP	252	168	177	149	144	132	124	534	401	478	419	429	385
BaP	252	22.7	25.4	17.7	24.3	17.4	16.3	85.3	68.1	74.1	57.5	54.1	45.6
IndP	276	20.1	34.2	26.5	27.1	23.6	14.6	74.8	57.7	68.1	55.8	46.9	42.8
BghiP	276	88.9	100	87.7	75	68.2	56.8	348	280	301	263	282	252
Total PAH		983	1060	865	853	770	629	2792	2093	2551	2074	2042	1781
Fl/(Fl + Pyr)		0.55	0.56	0.56	0.57	0.56	0.45	0.44	0.43	0.45	0.43	0.40	0.38
BaA/(BaA + Chr)		0.08	0.09	0.08	0.09	0.09	0.08	0.14	0.15	0.14	0.13	0.12	0.12
BaP/(BaP + BeP)		0.12	0.13	0.11	0.14	0.12	0.12	0.14	0.15	0.13	0.12	0.11	0.11
Ind/(Ind + BghiP)		0.18	0.26	0.23	0.27	0.26	0.20	0.18	0.17	0.18	0.18	0.14	0.15

Notes: BaA, benz[a]anthracene; BaP, benzo[a]pyrene; BbjF, benzo[b,j]fluoranthene; BeP, benzo[e]pyrene; BghiP, benzo[h,g,i]perylene (BghiP); Chr, chrysene; Fl, fluoranthene; IndP, indeno[1,2,3-cd]pyrene; Pyr, pyrene.

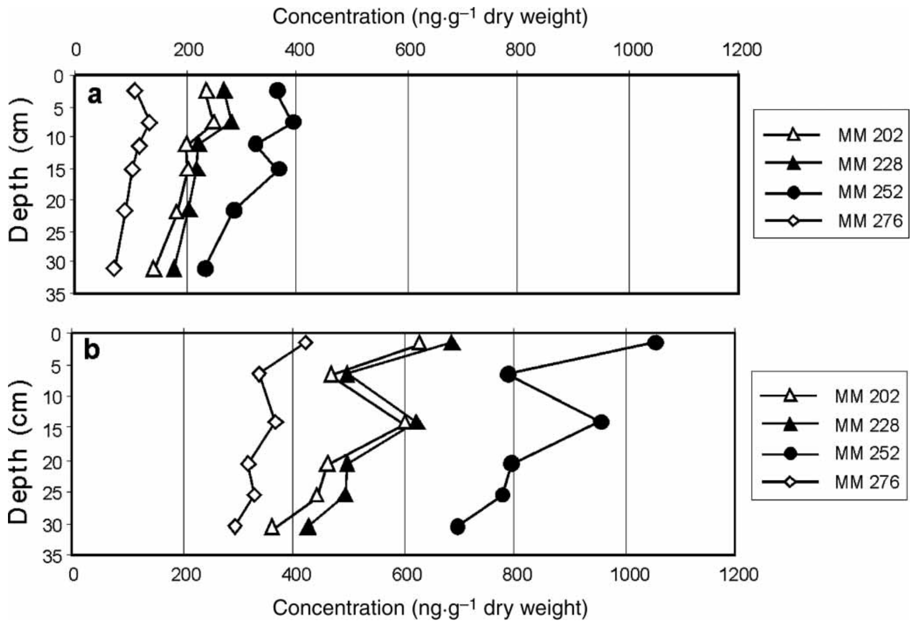

Figure 2. Plots of polyaromatic hydrocarbon (PAH) concentration by molecular mass group for Forlandsund (a) and Grønfjord (b).

of the total number of individuals in Isfjord, and 93.9% in Grønfjord. Molluscs were the second most numerous in Isfjord at 7.6%, whereas crustaceans were the second most abundant (3.4%) in Grønfjord (Table 3). Although abundances were considerably different between the two sites,

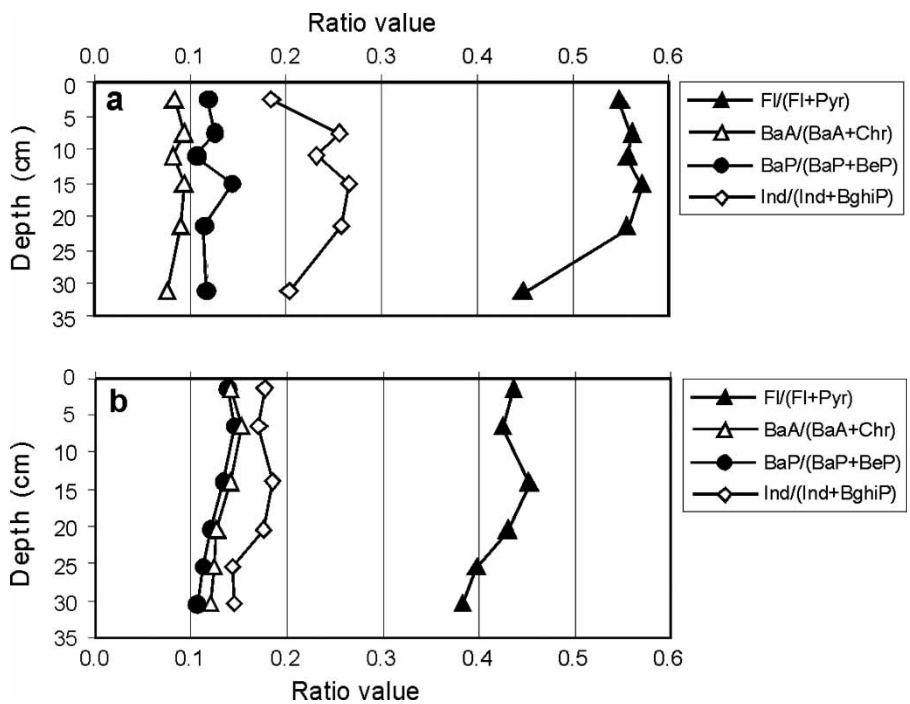

Figure 3. Plots of polyaromatic hydrocarbon (PAH) molecular mass group mass ratios for Forlandsund (a) and Grønfjord (b).

Table 3. Percent composition of faunal groups in terms of abundance and richness at the two stations sampled.

	Isfjord		Grønfjord	
Taxon	% abundance	% taxa	% abundance	% taxa
Crustacea	1.1	6.7	3.4	3.4
Mollusca	7.6	16.7	2.4	19.0
Polychaeta	88.5	70.0	93.9	74.1
Other	2.7	6.7	0.3	3.4

Table 4. Ten most abundant taxa (individuals·0.3 m^{-2}) at each of the two stations sampled in this study.

Isfjord (Station 2)			Grønfjord (Station 5)		
Taxon	Individuals · 0.3 m^{-2}	BIO	Taxon	Individuals · 0.3 m^{-2}	BIO
Chaetozone sp.	59	dm	*Chaetozone* sp.	504	dm
Scoloplos acutus	42	cb	*Aphelochaeta* sp.	274	sd
Capitella capitata	36	cb,sd	*Maldane sarsi*	170	cb
Scoletoma sp.	33	dm	*Galathowenia oculata*	128	cb
Maldane sarsi	25	cb	Cumacea g. sp.	71	sd,dm
Nuculana pernula	7	sd,dm	*Yoldiella nana*	69	sd,dm
Eteone cf. *longa/flava*	7	dm	*Rhodine gracilior*	44	cb
Aphelochaeta sp.	6	sd	*Lanassa venusta*	32	sd
Ophiuroida g. sp.	5	sd,dm	*Spio armata*	23	sd,dm
Galathowenia oculata	5	cb	*Lysippe labiata*	23	sd

Note: Bioturbatory activity (BIO) is indicated by the following abbreviations: dm, diffusive mixer; cb, conveyor-belt feeder; sd, surface-deposit feeder [36,37,38].

community composition and bioturbation mode were very similar. The five most abundant taxa at Station 5 were also among the ten most abundant taxa at Station 2 (Table 4).

3.3. *Bioturbation*

Glass bead distributions in bioturbation cores indicated particle mixing at both stations. At Isfjord, beads penetrated to only 2–3 cm after both 5 and 10 days, whereas in Grønfjord, beads were found at 5 cm after 5 days and 10 cm after 10 days of incubation (Figure 4). There is no obvious suggestion of subsurface peaks in bead distribution in any of the experimental cores. In control cores to which beads were added and the core immediately sectioned, 79–85% of the beads were found in the top 0.5 cm and $> 96\%$ in the top 1 cm. No beads were found below 2 cm in control cores.

3.4. *Sediment data*

Data on organic content, sedimentation rate and bioturbation depth contained in published reports from these same stations are provided in Table 1. Organic contents in Isfjord and Forlandsund are ~ 1.7–1.8% (dry w/w), and vary in Grønfjord from 2.1 to 2.7% [21,23]. Sedimentation rates are nearly four times higher in Grønfjord than in Isfjord, with Forlandsund having rates half that of Grønfjord. Bioturbation depth determined from radioisotope profiles are ~ 5 cm in Isfjord and twice that in the other two locations [21].

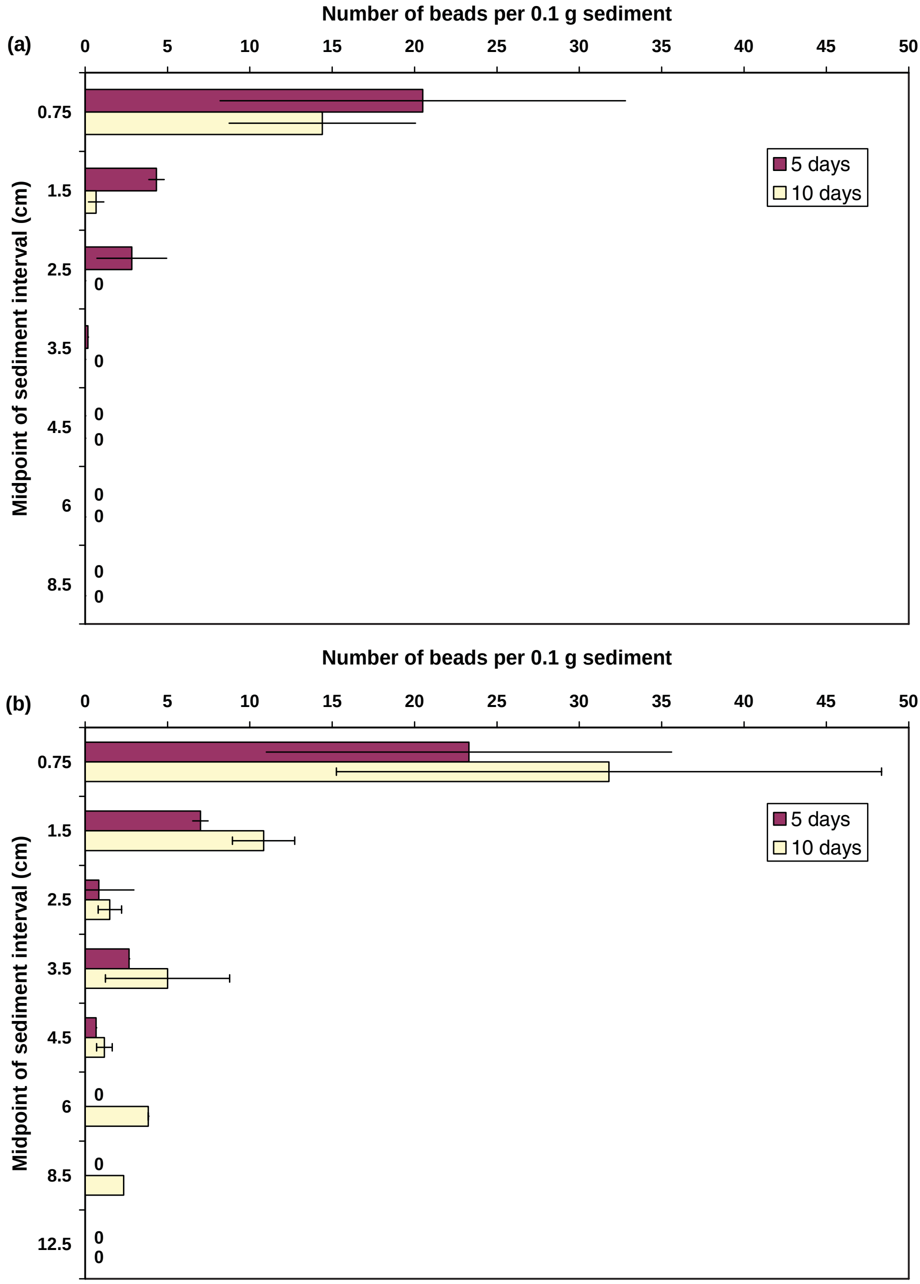

Figure 4. Depth profiles of the mean number of glass beads per 0.1 g sediment for 5- and 10-day incubations from Isfjord (a) and Grønfjord (b). Error bars represent ± 1 SD, no beads found is indicated by a 0.

4. Discussion

4.1. *Sources of PAHs*

Despite very high pyrogenic PAH concentrations, low values for three of the four PAH ratios at both stations (Table 2, Figure 3) suggested predomination of petroleum/coal sources of contamination

[22,24]. A relatively high Fl/(Fl + Pyr) ratio can, however, suggest the presence of combustion sources. It is possible that some pyrogenic signature was overwhelmed by the abundant coal particles in the sediment (D. Konovalov, pers. obs.). PAH ratios were similar to those measured from other locations in Svalbard fjords [25], including within the Isfjord complex. This suggests that deposition from coal-fired power stations located in Longyearbyen and Barentsburg are minor compared with petrogenic sources of heavier PAHs.

4.2. *Faunal communities in the Isfjord complex*

Faunal community composition and dominance by deposit-feeding taxa are similar to what has been found in other Svalbard fjord and shelf communities. The most abundant taxa found at the two locations in this study also dominate the outer fjord communities in nearby van Mijenfjord and Kongsfjord [26,27], although densities are somewhat lower in this study. Elevated faunal abundances at the more enriched site support the general paradigm that most Arctic benthic communities are food limited [18,19]. However, the extremely high proportion of the community that is made up of polychaetes ($>$ 90% of individuals) found at this location (this study and Cochrane et al. [21]) is somewhat surprising when compared with other typical fjordic environments in northern Norway [17] and Svalbard [26,28].

Sediment total organic carbon content reached well over 2%, which is not an uncommon finding for an Arctic location. However, such levels at lower latitudes often result in marked impacts on benthic communities that include a shift to 'opportunistic' taxa, high abundance/low biomass, and concentration of organisms in the top 2 cm, all as a result of decreasing oxygen supply and increases in toxic solute concentrations [15]. This is not the case in the Isfjorden complex. Rather, our findings support the conclusion of Holte et al. [17] that high latitude regions may be less vulnerable to depletion in bottom-water oxygen under moderate organic loadings because reduced water-column stratification leads to strong vertical mixing. Further, we suggest that colder waters allow for higher oxygen solubility and may reduce sediment microbial activity in these environments.

4.3. *Bioturbation in fjord sediments*

Sediment mixing depths determined from isotope profiles [21] and from our bioturbation experiments (Figure 4) fall within a similar range to that from another Arctic shelf (6–10 cm for the Chukchi Sea shelf) [29]. In that study, mixing depth is positively correlated with infaunal density, and in our study a five-fold higher faunal abundance results in a two- to three-fold deeper bioturbation layer.

Bioturbation was enhanced, not depressed, in the enriched sediment site (Figures 1 and 4). This enhancement was not due to different taxa dominating at the two sites, but instead to the greater abundance of organisms at the Grønfjord site (Tables 3 and 4). It is also possible that the fauna in Grønfjord were larger than in Isfjord (D. Konovalov, pers. obs.), or just exhibited higher burrowing activity in Grønfjord, but these hypotheses remain speculative. Arctic infauna is known to respond quickly to food inputs, and benthic communities exhibit higher oxygen consumption in areas of higher concentrations of purported food [30,31]. *Maldane sarsi*, a head-down, deposit-feeding polychaete, can inhabit vertically oriented tubes that extend 10–20 cm or more into the sediment, and is known to be efficient at particle mixing deep into sediments [32]. This species and several other conveyor-belt feeders were among the most abundant taxa in our samples (Stations 2 and 5, Table 4), and were likely to be responsible for the deepest sediment mixing. Their actions are capable of influencing the downcore profiles of PAHs in the sediment through bioturbation. The subsurface minima in PAH levels around the depth associated with tube length in these worms

(5–12 cm) suggests that bioturbation may have directly or indirectly enhanced PAH degradation. Whereas some polychaete worms are known to chemically modify PAH compounds (pyrene, fluoranthene) [2,10], the taxa abundant in our samples have not been examined.

Clearly, the time between sampling and different locations prohibit direct comparison of PAH data and faunal structure and function. The data are presented together to evaluate whether bioturbation may be consistent with PAH profiles from this (and other) studies conducted in the region. We do not suggest that we have conclusively identified the mechanism responsible for the PAH profiles, but instead that bioturbation is a potentially relevant process for influencing PAHs.

4.4. *Consequences of sediment fauna and organic content on contaminant cycling*

In Arctic fjords, the fate of contaminants such as PAHs is determined by both the level of organic matter resulting from sedimentation processes and anthropogenic inputs from industrial and municipal waste. In these oligotrophic, food-limited environments, moderate organic discharges may increase the biomass and abundance of benthic fauna, also affecting the depth of biological sediment mixing. Because organic effluents often also contain a contaminant load, and contaminant behaviour in sediments is affected by the activities of benthos, the background biological and physical processes are critical in determining the impacts of contaminants such as, but not limited to, PAHs. The correspondence between bioturbation depths and regions of the sediment column with high PAH levels further stresses the importance of the potential for biological processes to influence contaminant cycling.

Our results are distinct from many temperate and tropical sites where organic enrichment may lead to hypoxia, depleted benthic communities and a reduction in bioturbation depth. Holte et al. [17] argue that sub-Arctic fjords in northern Norway may respond to organic enrichment differently from boreal fjords. Owing to reduced stratification, greater vertical convection and a higher tidal range [33], northern fjord waters were less likely to have hypoxic/anoxic bottom waters, whereas in comparable (depth, sill, enrichment levels) fjords in southern Norway, similar organic inputs led to reduced bottom-water oxygen and a depleted benthic fauna dominated by small surface-dwelling taxa [34,35].

These results suggest that assessment of real or potential impacts of contaminant loading and human discharges in Arctic and other oligotrophic areas should not be based on the same criteria as used in the regulations and directives applied to, for example, European waters. Environmental impact assessments or monitoring carried out to regulate mineral or petroleum extraction and its associated infrastructure should pay particular attention to the biological and physical characteristics of Arctic ecosystems as a background for interpreting levels of contaminants.

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