PBDE LEVELS IN SOILS DOWNGRADIENT MUNICIPAL SOLID WASTE FACILITIES IN NORTHERN CANADA

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ABSTRACT
A field study was carried out to investigate the levels of polybrominated biphenyl ethers (PBDEs) around solid waste facilities in Northern Canada. The purpose was to achieve improved knowledge of the extent of PBDEs contamination in soils surrounding landfills, and the potential risk of contaminating nearby Arctic aquatic environments. Results were highly variable from one site to another, even at the same location. There was evidence of migration, both downwards and upwards, in soil samples. The relative concentrations of congeners also suggested breakdown of PBDEs over time in the environment.

RÉSUMÉ
Une étude a été effectuée pour étudier les niveaux des éthers diphényliques polybromés (ÉDPs) autour des équipements de déchets solides au Canada nordique. Le but était de réaliser la connaissance améliorée du taux de migration d'ÉDPs des remblais, de leur adsorption sur des sols et du risque potentiel de souiller les environnements aquatiques arctiques voisins. Les résultats étaient fortement variables d'un emplacement à l'autre, même au même endroit. Il y avait d'évidence de migration, en bas et vers le haut, dans des échantillons de sol. Les concentrations relatives des congeners ont également suggéré la panne de PBDEs avec le temps dans l'environnement.

1 INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are a group of brominated flame retardants used in many consumer products. There are 209 possible congeners classified by the number of bromine atoms in the chemical structure of the molecule. These congeners are commercially synthesized resulting in a product or fraction containing a mixture of PBDEs with various degrees of bromination (Rahman et al., 2001; D'Silva et al., 2004). Seven out of the 209 PBDE congeners, BDE-47, -85, -99, -153, -154, -183 and -209, are typically found within three commercial mixtures: pentaBDE (PeBDE), octaBDE (OcBDE) and DecaBDE (DeBDE), known as commercial products. Annual worldwide production of Pe-, Oc-, and DeBDE in 2001 were 7500, 3790, and 56100 metric tons respectively (Bromine Science and Environment Forum 2004).

The commercial BDE mixtures are incorporated in consumer products such as upholstered furniture, electrical goods (e.g. vacuum cleaners, plugs, sockets, computers), sealants and foams. PeBDE is used in polyurethane foams in furniture, carpet underlay and bedding. The most important congeners within PeBDE are BDE-47 and BDE-
99 (Hale et al., 2003). OcBDE is used in styrene, polycarbonates and thermosets (de Wit 2002; Alcock et al., 2003). The major congeners in OcPBDE are BDE-183, 153, unknown oct- and nona-BDEs (Hale et al., 2003). The DeBDE product is dominated by a single principal congener, BDE 209, and is predominantly used textiles and denser plastics such as housings for a variety of electrical products, in particular televisions and computers (D'Silva et al., 2004). Although, the EU and USA have banned, or are in the process of banning, some PBDEs, past production, current consumption and continuing disposal mean that even banned congeners will continue to persist in the environment for several decades at least. Moreover, the unbanned deca-BDE (-209) congener is still being produced and continues to be deployed as a flame retardant, with no early prospect of termination and replacement.

Environmental release of PBDEs may occur during their initial synthesis and incorporation into polymers, from related finished products or as a result of their ultimate disposal or recycling. Alcock et al. (2003) estimate that 80% of PBDEs are landfilled, with the remainder incinerated. The relatively low volatilities and aqueous solubilities of PBDEs suggest that the bulk of the environmental burden of these chemicals eventually resides in either sediments or soils (Palm et al., 2002; Hale et al., 2003). At present, there are no reported studies quantifying the loss of PBDEs via leaching from landfill sites. However, it has been found that sediments

in water surrounding landfill and sewage treatment facilities have elevated concentrations of tetra-bromodiphenyl ether (tetraBDE) and penta-pentabromodiphenyl ether (pentaBDE), ranging from 21×10^3 to 368×10^3 pg/g dw and 9.2×10^3 to 898×10^3 pg/g dw, respectively (Alcock et al., 2003; de Boer et al., 2003).

The levels of PBDEs have been rising at alarming rates globally, even in remote regions such as the Canadian Arctic. This has been attributed to transport from urban regions via atmospheric deposition, commonly known as long range atmospheric transport (LRAT). However, it is also important to look at the Northern communities, which use commercial products with PBDE additives, and also have solid waste disposal facilities which ultimately receive these products at the end of their life cycle. It is vital to examine these waste disposal facilities to determine the fate and transport of PBDEs and to study the nature and extent of PBDE contamination, their potential mobility, their bio-availability and their cumulative amount in vegetation, soil, and drainage water in such remote locations.

In July 2006 sampling was carried out in Northern Canada, specifically in Iqaluit, Nunavut and Yellowknife, Northwest Territories (NWT), to investigate PBDEs within Northern Canadian waste disposal facilities and related soils in the Canadian North. The objective of this project was to examine the mobility of PBDEs in soils by determining the lateral and vertical distribution of PBDEs in the surrounding soils. These results were compared to other published results from northern and southern locations. The work is intended to provide a better understanding of the mobility of PBDEs in soils and to collect information for models to predict PBDE migration in soil, thereby facilitating best management practices for waste disposal systems.

2 METHODS

Soil samples were collected from areas around two municipal solid waste facilities, in Iqaluit and Yellowknife.

2.1 Site Background

The first landfill, located south of the town of Iqaluit and Koojesse Inlet, (Figure 1), has been operating since 1995. Prior to 2002 the facility practiced open burning at its main dump site, and the ashes were dumped and covered at different locations in the surrounding area. Iqaluit currently compacts the solid waste it receives. The landfill receives approximately 6000 tonnes of waste annually, segregating it into different categories: metals and tires, garbage waste, shredded wood, organic compost, and sewage solid waste. A drainage system surrounds the landfill to collect leachate surface runoff, which drains into a nearby bay approximately 500 m south east of the landfill.

The second waste disposal facility is located north of the city of Yellowknife near Jackfish Lake, in the Northwest Territories. It has been in operation since 1970 and annually receives 10,000 tonnes of waste from the residential, commercial, industrial and construction sectors. The landfill is divided into designated areas for scrap metals, construction demolition, scrap vehicles, wood/carpet, tires, brush area, peat from the municipal sewage treatment lagoon, refrigerators, contaminated soils and from different oil spill sites in Yellowknife and waste bales (all waste received in garbage bags) (Figure 2). The received waste is processed, separated, baled and stacked in a designated area.

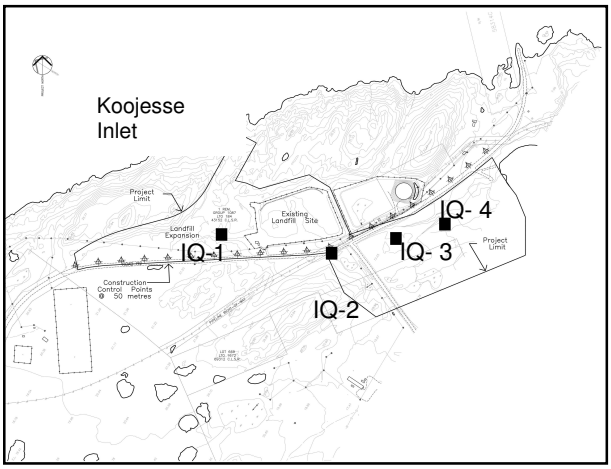

Figure 1. Iqaluit Solid Waste Disposal Facility site plan and the sampling locations.

Figure 2. Yellowknife Solid Waste Disposal Facility site plan and the sampling locations.

2.2 Soil Sampling

Soil samples were collected from a location upgradient of the Iqaluit landfill referred to as IQ-1, and at three locations downgradient from the landfill referred to as IQ-2, IQ-3 and IQ-4. These sites were along the drainage system, and towards the receiving bay. They were chosen to determine the effect of the disposal facility on PBDE contamination in the surrounding environment (Figure 1). Table 1 summarizes the GPS coordinates and elevations of all four locations. Due to the high water table levels, samples were only taken from 0-20 cm and 20-40 cm depths.

Samples from Yellowknife were collected from a background area (YELL-Bkgrd), approximately 500 m from the landfill, and from three areas downgradient and south of the landfill (Figure 2) referred to as YELL-1, YELL-2 and YELL-3 (Table 2). Due to the rocky nature of the region, it was only possible to collect samples from 0-15 cm and 15-30 cm depths in the Yellowknife case.

Table1: Sampling locations at City of Iqaluit Landfill Site and elevations (m).

Location	Elevation (m)	Coordinates
IQ-1	23	N 63° 93.821 W 68° 32.140
IQ-2	18	N 63° 43.891 W 68° 32.170
IQ-3	9	N 63° 43.736 W 68° 32.050
IQ-4	7	N 63° 43.680 W 68° 32.028

Table 2. City of Yellowknife Municipal Solid Waste Facility sampling locations.

Location	Elevation (m)	Coordinates
YELL-Bkgrd	186	N 62° 28.355 W 114° 22.396
YELL-1	199	N 62° 28.525 W 114° 23.145
YELL-2	195	N 62° 28.503 W 114° 23.143
YELL-3	194	N 62° 28.495 W 114° 23.146

The soil samples from each site were sent directly to the Institute of Ocean Sciences (IOS) in Sidney, BC and stored at -30°C until analysis. Extraction and clean-up of the samples were as described by (Ikonomou et al., 2001). Briefly, soil samples were extracted at high temperature (180°C) and pressure (2000 psi) with a mixture of toluene:acetone (80:20 v/v) using the accelerated solvent extraction method. The extracts were then subjected to an extensive cleaning process in preparation for analysis using a high resolution gas chromatograph /high resolution mass spectrometer (HRGC/HRMS).

3 RESULTS

3.1 Lateral Migration of PBDEs at Iqaluit Solid Waste Disposal Facility

The total concentrations of all 209 PBDE congeners (Σ PBDEs) at IQ-1, upgradient of the landfill, was almost 9 times greater than at locations IQ-2, IQ-3, and IQ-4 downgradient of the landfill (Figure 3), within the top 20 cm of the soil. Analysis of the distribution of single congeners within each sample, shows that BDE-47, -99, -85, -153, -154, and -209 congeners had the highest concentrations, consistent with the fact that they are the main constituents of the commercially produced PBDE mixtures. The concentrations for these 6 congeners are presented in Table 3. The levels in the top layer from the three locations downgradient of the facility are lower than the upgradient levels and independent of the distance downgradient of the landfill, except when ΣPBDEs are considered, concentrations decrease with distance from the landfill.

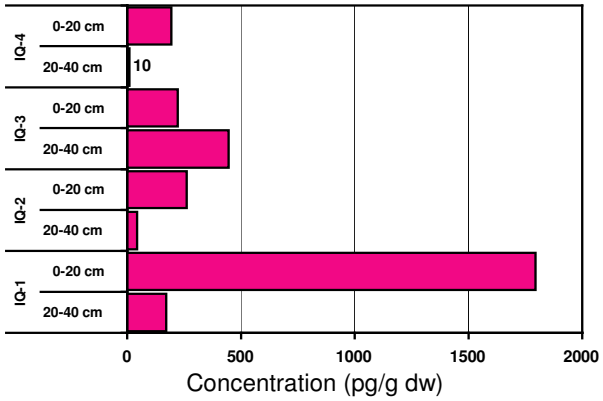

Figure 3. ΣPBDE across the 40 cm depth upgradient and downgradient Iqaluit Waste disposal facility.

In the 20-40 cm soil layer, the concentrations of the 209 congeners ranged from 10 to 45 pg/g dw (Figure 3) having BDE-47, -99, -85, -153, -154, and -209 mainly contributing to the cumulative PBDE total (Table 3). The highest total concentration in this layer occurred at IQ-3, due to elevated BDE-209 levels, while the other congeners levels at the same location and depth were relatively low or not detectable. The high BDE-209 levels reflect the ubiquitous presence of this congener, its dominant role in the commercial Deca product (still in commercial production) and possibly also the greater difficulty relative to other congeners in determining this congener accurately.

3.2 Vertical Migration of PBDEs at Iqaluit Solid Waste Disposal Facility

The results in Figure 3 and Table 3 show that PBDEs are present below a depth of 20 cm and downgradient of the source. Since BDE-209 is known to be the most ubiquitous congener, and is most difficult to analyze accurately, it is not possible to determine whether BDE-209 is mobile. As for the other congeners, lighter PBDEs such as BDE-47 and 85 are found further from the landfill (IQ-2) at higher concentrations than in the top layer. The results suggest that PBDEs can migrate downwards below the surface. However, it is necessary to continue monitoring PBDE levels to further confirm their mobility.

Table 3. Total concentrations of all 209 congeners (ΣPBDEs) and the concentrations of BDE-47, -99, -85, -154, -153 and -209 in pg/g dw.

		BDE-47	BDE-85	BDE-99	BDE-154	BDE-153	BDE-209	Σ PBDEs
Upper Layer (0-20 cm)	IQ-1	118	376	20	50	43	1,021	1,795
	IQ-2	4	2	ND	ND	ND	187	260
	IQ-3	ND	ND	ND	ND	ND	215	221
	IQ-4	37	6	ND	ND	ND	123	193
Lower Layer (20-40 cm)	IQ-1	ND	21	ND	3	3	120	171
	IQ-2	5	4	ND	1	1	25	44
	IQ-3	1	ND	ND	ND	ND	404	445
	IQ-4	ND	ND	ND	ND	ND	ND	10

ND: non-detected.

The ΣPBDE concentration and individual PBDE congeners varied inconsistently in both layers This inconsistency may be attributable to:

- External sources other than the landfill could contribute to PBDE contamination.
- Water infiltration and runoff due to melting snow, and seasonally fluctuating groundwater levels could lead to fluctuations in PBDE levels.
- Heterogeneity of the soil could cause the soil porosity to be different at various locations, allowing water to infiltrate at different rates, and thus PBDEs to be transported at different rates.
- High octanol-air partition coefficients (K_{OA}) of PBDEs, could lead to seasonally variable concentrations in the surface soil and air. In winter they could accumulate in the soil surface, trapped in the snow and ice, and then volatilize when temperatures rise in the spring and summer, escaping from the soil (Gouin et al., 2005).
- Photochemical decomposition of PBDEs, especially BDE-209 could occur at the surface, producing lower brominated congeners which could escape from the surface layer (de Wit 2002; Eriksson et al., 2004; Soderstrom et al., 2004).

3.3 Lateral Migration at Yellowknife Solid Waste Disposal Facility

The concentrations of each PBDE homologue group, and ΣPBDEs of all 209 congeners indicate that the background levels were 2 to 4 times higher than the downgradient levels at YELL-1, YEL-2, and YELL-3. This was mainly due to the BDE-209 concentration (Figure 4 and Table 4). If BDE-209 is excluded, locations remote from the landfill had higher concentrations of the other congeners. It is also observed that the YELL-3 concentrations, i.e. those furthest from the facility, recorded the highest concentrations, probably because this soil was from a soil cover on an area used for waste disposal until approximately 2000.

The percent contribution of PBDE congeners to the ΣPBDEs concentrations indicates that BDE-47, -99, -85, -153, -154 and -209 are dominant, in agreement with the samples from Iqaluit Solid Waste Disposal facility. The concentration of these congeners are summarized in Table 4.

In the 15-30 cm layer, concentrations of all 209 congeners were between 424 and 65,500 pg/g dw (Figure 5). The values for the dominant BDE congeners (BDE-47, -85, -99, -153, -154 and -209) are presented in Table 4. It is observed that PBDE levels at YELL-3 were the highest, again likely because the soil here was in direct contact with waste disposed in the past.

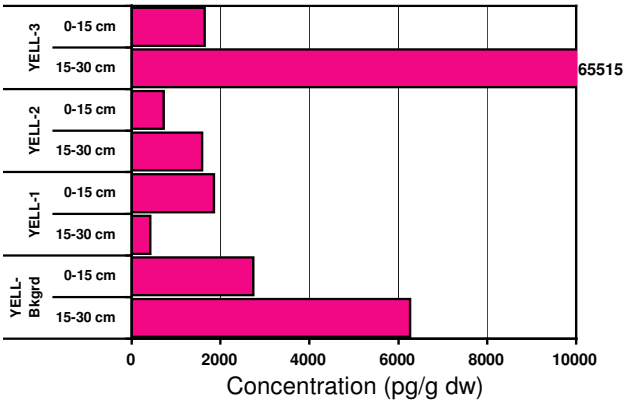

Figure 4. Comparison of sum of concentcrations of all 209 congeners surface layer (0-15 cm) and 15-30 cm level at the four locations at Yellowknife Solid Waste Disposal Facility.

3.4 Vertical Migration of PBDEs at Yellowknife Solid Waste Disposal Facility

Figure 4 indicates that PBDEs are mobile within the top 30 cm. Concentrations in the 15-30 cm layer from YELL-Bkgrd, and from YELL-2 and YELL-3, are greater than in the surface (0-15 cm) layer. For the background area, this is due to BDE-209, which presents analysis difficulties. As for YELL-2 and YELL-3, the elevated concentrations could be due to PBDEs migrating downwards from YELL-1, and also to their proximity to the former disposal area which contains buried municipal solid waste at depths beyond 30 cm. This suggests that PBDEs can also desorb and migrate upwards.

No specific trends can be unambiguously identified for the same reasons specified for the Iqaluit site in section 3-2. In addition, there is an apparent source of PBDEs away from the current disposal facility, probably originating from past waste, now covered by soil, which contributes mainly with congeners lighter than BDE-209 (Table 4).

4 DISCUSSION

4.1 Results Compared to Arctic Levels

A single study has reported PBDE levels in sediments from the Canadian North. Specifically this study pertains to a very remote site in the high Arctic, Devon Island (Stern *et al.*, 2005). Figure 5 compares the published data of total concentrations of all 209 PBDE congeners. The total concentration in the top layer (0-20 cm) of IQ-Location 1, and in the bottom 20-40 cm layer of IQ-Location 3 from Iqaluit, are 7 and 2 times more than what was recorded from the sediment samples on Devon Island. Total concentrations in the other layers from Iqaluit are 1.25 to 5 times less than their counterpart from Devon Island. The total concentrations of all PBDE congeners in the top layer (0-15 cm) at Yellowknife, are 3-11 times greater than the Devon Island total PBDEs concentration. The total concentrations in the 15-30 cm layer soil samples are 2-262 times higher, especially at YELL-Location 3.

4.2 Results Compared to North American and European Levels

Three published studies have reported PBDE levels in soils from North America and Europe due to a specific source of PBDE contamination. The studies investigated PBDE levels in biosolids amended-soils, from Kamloops, B.C. (Gorgy *et al.*, 2006), Ashcroft, B.C. (Gorgy *et al.*, 2005), and Sweden (Sellstrom *et al.*, 2005). Comparison of the total concentration of all 209 congeners from these areas with those obtained from Iqaluit and Yellowknife (Figure 5) indicate:

Table 4. Total concentrations of all 209 congeners (ΣPBDEs) and the concentrations of BDE-47, -99, -85, -154, -153 and -209 in pg/g dw in Yellowknife soil samples.

Depth and Location		BDE-47	BDE-85	BDE-99	BDE-154	BDE-153	Deca (BDE-209)	Total PBDE
Upper Layer (0-15 cm)	YELL-Bkgrd	20	63	4	8	11	2,528	2,743
	YELL-1	56	134	2	20	55	1,035	1,854
	YELL-2	9	4	ND	ND	ND	669	723
	YELL-3	454	428	19	27	32	552	1,642
Lower Layer (15-30 cm)	YELL-Bkgrd	34	50	3	3	6	5,908	6,270
	YELL-1	6	9	1	1	1	364	424
	YELL-2	39	46	1	3	3	1,431	1,586
	YELL-3	17,097	32,096	1,503	2,934	3,408	670	65,515

ND: non-detected.

- Total concentration in surface (0-20 cm) and lower (20-40 cm) soil layers from Iqaluit are similar or 1.25 to 30 times lower than for the Kamloops and Ashcroft sites.
- Total concentrations in Iqaluit soil samples are 1.5 to 4 times lower than their Swedish counterparts, except for the top soil layer at IQ-1, which is double the total Swedish concentration.
- Total concentrations of all PBDEs in surface (0-15cm) soil layers from Yellowknife are 2 to 5 times less than at Kamloops and Ashcroft. However, they are 2 to 3 times higher than the total PBDEs concentration in the surface soil from Sweden.
- Total concentrations in the 15-30 cm soil layers from Yellowknife are 1 to 125 times greater than total concentrations in the 20-40 cm soil layers from Kamloops. Again the greatest difference was at YELL-3.

A single study from the Russian Arctic recorded the summation of BDE-47, -99, and -100 in surface soil samples. The summation was in the range of 160 to 230 pg/g dw (de Wit *et al.*, 2006). The corresponding

summation from the 4 locations at Iqaluit were between ND and 574 pg/g dw, with the highest concentration occurring at the surface layer (0-20 cm) of IQ-1 (upgradient of the landfill), while the rest were almost 1 order of magnitude less. As for the Yellowknife samples, the summations of BDE-47, -99, and -100, were between ND and 51600 pg/g d.w.. with the highest concentration at location 3. Other locations were less or almost equal to their Russian counterpart.

From the above comparisons, the PBDE levels around the waste disposal site at Yellowknife, are of concern, since they sometimes exceeded Canadian and European Arctic levels. Of even greater concern, is that some concentrations were higher than those of biosolids-amended-soils, which are known to have elevated PBDE levels.

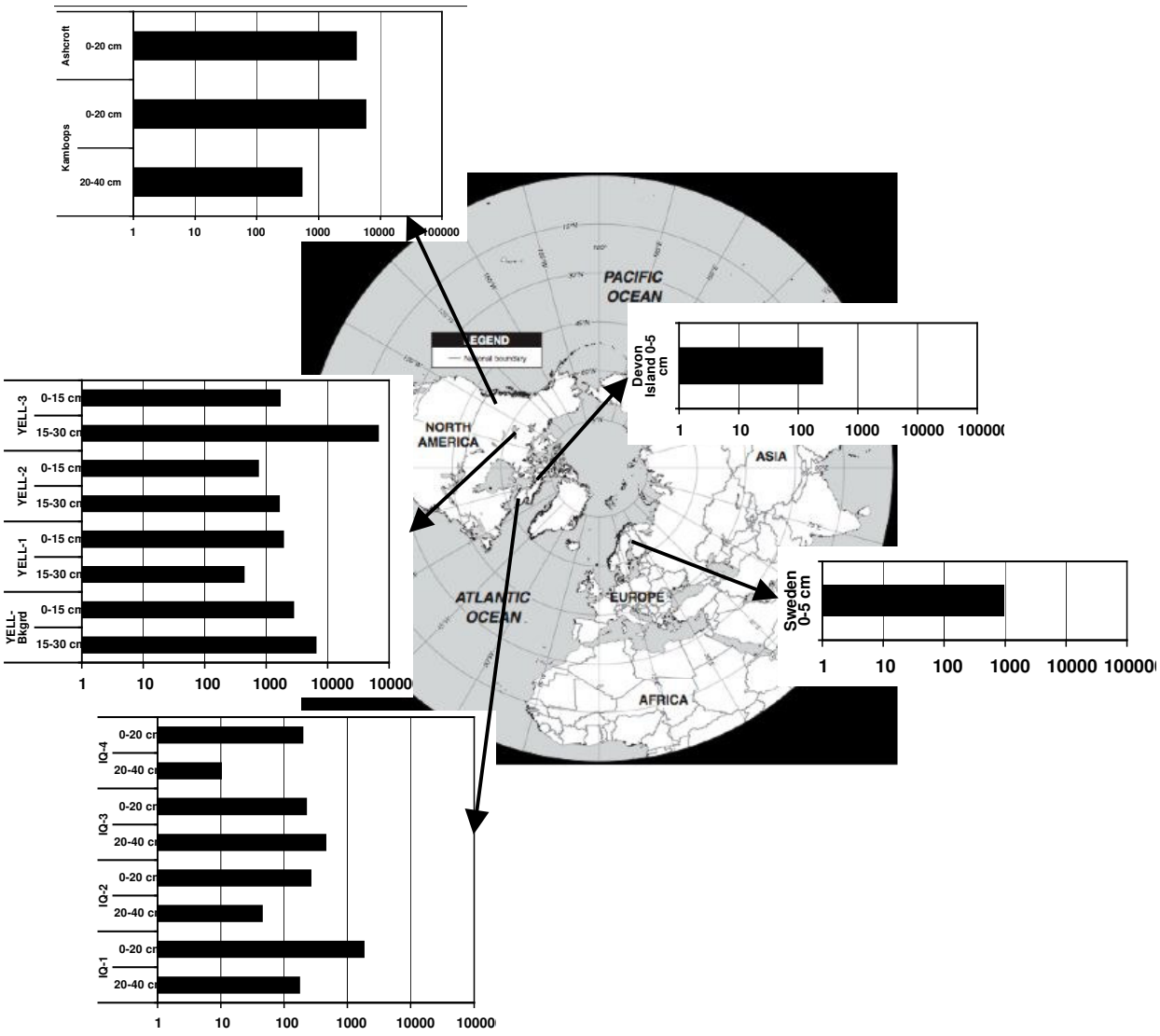

Figure 5. Comparison of the total concentrations of all 209 congeners from Iqaluit and Yellowknife with their counterparts from northern Canada, southern British Columbia and Sweden (Gorgy et al., 2005; Stern et al., 2005; de Wit et al., 2006; Gorgy et al., 2006). Concentrations are on the same logarithmic scale in pg/g dw.

5 CONCLUSION

The study indicates that PBDEs are mobile, migrating downwards below the surface soil. However it is difficult to establish a general trend for the lateral and vertical distribution of PBDEs in the soil, especially where there are seasonal variations contributing to the distribution of PBDEs. Uncontrolled factors caused by the melting snow, and fluctuating groundwater levels likely contribute to the variability.

PBDE levels in soil samples from Iqaluit and Yellowknife were higher than reported levels from the high Arctic and Russian north. The total PBDE concentrations in the surface soil layers from Iqaluit and Yellowknife were less than their southern Canadian counterparts from Kamloops and Ashcroft. However, some locations were higher than the Swedish counterparts.

One area downgradient of the city of Yellowknife Solid Waste Disposal Facility showed having total PBDEs concentration at 15-30 cm depths up to 125 times greater than total levels in biosolids-amended-soils in North America and Europe.

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