Environmental fate of hexabromocyclododecane from a new Canadian electronic recycling facility

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ABSTRACT

An electronics recycling facility began operation at the municipal landfill site for the City of Edmonton, Canada in March 2008 with the goal of processing 30,000 tonnes of electronic wastes per year. Of the many by-products from the process, brominated fire retardants such as hexabromocyclododecane (HBCD) can evolve off of e-wastes and be released into the environmental media. HBCD has been identified by many countries and international bodies as a chemical of concern because of its ability to bioaccumulate in the ecosystem. An evaluation of the potential emission of HBCD indicates that up to 500 kg per year may be released from a landfill and recycling facility such as that operating in Edmonton. A multimedia fugacity model was used to evaluate the dispersion and fate of atmospherically emitted HBCD traveling into surrounding agricultural land and forested parkland. The model indicates that the three isomers of HBCD partitioned into environmental media similarly. Much of the HBCD is lost through atmospheric advection, but it is also found in soil and sediment. Modeled air concentrations are similar to those measured at locations with a history of e-waste recycling. Since HBCD has been shown to bioaccumulate, the HBCD released from this source has the long-term potential to affect agricultural food crops and the park ecosystem.

1. Introduction

The City of Edmonton in Alberta, Canada, began operation of its new Global Electric and Electronic Processing Inc. (GEEP) recycling facility at the Edmonton Waste Management Centre (EWMC) in March, 2008. This state-of-the-art facility is the first facility to be built by GEEP in western Canada and was designed with the goal of processing 30,000 tonnes of electronic waste (or e-waste) per year (EWMC, 2011). Edmonton is located at 53° 34′ N latitude and 113° 31′ W longitude; the greater metropolitan area has a population of approximately 1.034 million people (Statistics Canada, 2007).

Landfill and recycling processing methods are possible emission sources of hexabromocyclododecane (HBCD; CAS registry number: 25637-99-4). HBCD is a cyclic, aliphatic, brominated fire retardant produced in the third largest (Younglai et al., 2007; Zegers et al., 2005). HBCD is used for fire safety in expanded polystyrene foams, textiles, and electronics (Peck et al., 2008). Therefore, many consumer products contain HBCD including, of particular interest to this study, electrical appliances and computer equipment. HBCD has no known natural sources. Therefore, recycling of e-wastes may produce by-products that can become airborne (Covaci et al., 2006). Of the many by-products, brominated fire retardants can evolve off of the wastes and be released into the environmental media. While recycling e-waste is positive from an environmental standpoint, given the great increase in the disposal of obsolete equipment (Macauley et al., 2003), there have been observed increases in airborne brominated compounds near other electronic processing facilities (Remberger et al., 2004).

Several national and international reviews of HBCD are underway for its persistence in the environment and toxicity (UNECE, 2010; Environment Canada, 2010). HBCD has the propensity to bioaccumulate in the food chain once released into the environment (de Wit, 2002) as the octanol–water partitioning coefficient (log K_{ow}) has been estimated as 5.6 (CMA, 1997). In a recent review, Covaci et al. (2006) highlighted that, unlike many other brominated fire retardants, the environmental toxicology of HBCD is largely unknown. However, there are low acute toxic effects and evidence that HBCD may induce cancer through non-mutagenic means. HBCD can damage the thyroid hormone system and can have neurological effects in laboratory animals. Concentrations become elevated in top

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predators such as dolphins, falcons, and marine mammals. Not surprisingly, HBCD has also been detected in human breast milk and blood serum (Restrepo et al., 2008). Yet, there appear to be no studies of HBCD in agricultural products (Marvin et al., 2011).

In 2001, worldwide production of HBCD was 16,700 tonnes and had grown to 22,000 tonnes by 2005 (Isobe et al., 2007). In the United States and Europe, release of HBCD had not been regulated (Zegers et al., 2005). Therefore, more detailed information regarding emissions of HBCD is not available despite recent identification of HBCD as a persistent organic chemical by the United Nations Economic Commission for Europe (2010) and further as a chemical for elimination by 2015 (UNECE, 2011). The largest sources of HBCD are identified as municipal landfill sites, locations that process wastes, and production facilities. The most notable releases have been from industries that rely heavily on production of flame retarded plastics and the textile industry (Remberger et al., 2004). While much work has focused on the landfill leaching of toxic chemicals, the atmospheric route to exposure is less well characterized (Moy et al., 2008). Local atmospheric concentrations of up to 1070 ng/m^3 have been observed near these facilities. Landfill sites have been implicated where air samples concentrations are in the range of 0.013–0.18 ng/m^3. By comparison, remote locations report less than 0.001–0.028 ng/m^3 (Covaci et al., 2006; Remberger et al., 2004).

2. Model parameterization and application

The objectives of this risk assessment are to characterize the potential release of HBCD from an electronic recycling facility operating at the magnitude proposed by EWMC and, further, to assess potential for atmospheric deposition on productive farmland and national parkland downwind of the facility. Most specifically, the goal is to estimate potential for short-range transport of airborne products into Elk Island National Park, downwind, and then the partitioning HBCD into other environmental media. This is a unique ecosystem that encompasses a transitional zone between aspen parkland and the boreal forest and is important as a migratory bird refuge (Parks Canada, 2011). The Canadian Centre for Environmental Modeling and Chemistry (CEMC) multimedia fugacity models Level 1 and 2 (CEMC, 2004) were employed to estimate HBCD concentration, distribution and fate in the natural environment.

2.1. Physical environmental parameters

Physical environmental properties required for the CEMC model to describe the region are summarized in Table 1. The total land area of 1014 km^2 is contained within major highways east of the City of Edmonton and surrounds the 194 km^2 Elk Island National Park with a minimum distance of 10 km. The region incorporates both land and water masses; hence, a large enough area was selected to include agricultural lands and natural habitat for fish predators. To assess exposure concentration of HBCD to agricultural crops, a soil volume was determined by multiplying the total land area by a one meter depth of soil with an organic fraction of 3.5% as estimated for crop production (Alberta Agriculture, Food and Rural Development, 2010). Soil density in prairie regions of Alberta is approximately 1.25 g/cm^3 (Lemke et al., 1998).

Volumes for major local lakes were estimated by multiplying published lake surface areas with associated depths (University of Alberta, 2005). The total model water volume in Table 1 is the sum of these volumes; no river volumes were included. Using the total lake surface area as the basis, the suspended sediment volume was estimated using a depth of 0.25 m while the sediment volume was calculated using a depth of one meter (University of Alberta, 2005). Sediment and suspended sediment organic carbon fraction is 18–20% for lakes in central Alberta, Canada (Campbell et al., 2000). The corresponding density factors are not likely critical to interactions and reactions of HBCD (Heeb et al., 2005; Law et al., 2006; Remberger et al., 2004), but a value of 1.5 g/cm^3 has been estimated for the region (Redding and Devito, 2006). The total volume of fish is also determined using total lake water volume. The most common and abundant lake fish of the region is Northern Pike (*Esox lucius*) with a moderate, yet realistic, number density of 0.0043 kg/m^3 determined from an estimate of 500 adult fish per hectare (Nasmith et al., 2010). Wild Northern Pike have a 2.3% fat content (Jankowska and Zakes, 2008).

Focusing on the spring and summer season when water bodies would be free of ice, a typical midday ambient temperature of 20 °C and atmospheric pressure of 93.5 kPa were estimated from the 1971–2000 Canadian Climate Normals for Edmonton (Environment Canada, 2011). A summer average midday atmospheric mixing height was estimated at 2000 m using data from Raddatz and Noonan (2004). Multiplication of land area by atmospheric mixing height established the air volume. Since organic chemicals are likely to adhere to fine particulate matter,

Table 1
Regionally specific parameters to describe the physical environment.

Media	Volume (m^3)	Density (kg/m^3)	Organic carbon or lipid fraction (g/g)	Reaction half lives (days)[a]	Advective flow residence times (hours)
Air	2.03×10^{12}	1.185	N/A	5.2	5.42
Aerosol	11	1600[f]	N/A	Negligible	N/A
Water	1.65×10^8	1000	N/A	130	43,800
Soil	1.014×10^{9c}	1250[h]	0.035[c]	6.9	N/A
Sediment	4.34×10^{6b}	1500[g]	0.18[d]	128	43,800
Suspended sediment	1.64×10^{6b}	1500[g]	0.18[d]	128	N/A
Fish	390	0.0043[i]	0.023[e]	5110[j]	N/A

N/A: not applicable.
[a] Marvin et al., 2011.
[b] University of Alberta, 2005.
[c] Alberta Agriculture, Food and Rural Development, 2010.
[d] Campbell et al., 2000.
[e] Jankowska and Zakes, 2008.
[f] Stein et al., 1994.
[g] Redding and Devito, 2006.
[h] Lemke et al., 1998.
[i] Nasmith et al., 2010.
[j] Ismail et al., 2009.

PM$_{2.5}$ regional mass concentration was used to estimate aerosol mass as a proportion of the total air volume. The five-year (1998–2003) mass concentration average for the Edmonton region was 8.4 μg/m^3 (CASA, 2006).

2.2. HBCD chemical parameters and emissions estimate

HBCD can be obtained commercially from independent manufacturers around the world as isomeric mixtures. From the fabrication process, six stereogenic centers are formed leading to 16 possible stereoisomers with six pairs of enantiomers and four meso forms (Heeb et al., 2005). Several studies conclude that mixtures usually contain 75–89% α-HBCD, 10–13% β-HBCD and 1–12% γ-HBCD. As well, these three stereoisomers are typically isolated in animal lipids (Heeb et al., 2005; Peck et al., 2008; Zegers et al., 2005). HBCD can rearrange when temperatures exceed 160 °C changing the mixture of stereoisomers. After heat exposure, the relative amounts become 78% α-HBCD, 13% β-HBCD and 9% γ-HBCD (Covaci et al., 2006). Solubilities vary between α, β and γ-HBCD forms, and are 48.8, 14.7, and 2.1 μg/L, respectively (Covaci et al., 2006). Stereoisomers labeled − and -HBCD were reported; however, concentrations are extremely low and have never been reported in the environment to date (Covaci et al., 2006; Peck et al., 2008; Zegers et al., 2005).

Literature-derived reaction half-lives are found in Table 1. Due to rapid atmospheric oxidation, the half-life in air is on the order of only 2 days. The UNECE (2009) has identified aerosol media as a likely long-range transport mechanism for HBCD due to its low vapor pressure of 6.27 × 10^{-5} Pa at 21 °C (CMA, 1997). However, for near distances of concern here and given the low atmospheric aerosol concentration in the model area, errors in equating air and aerosol parameters has little effect on model outcomes for the local region (Covaci et al., 2006; Isobe et al., 2007). Water hydrolysis is estimated by Environment Canada (2010) using the HYDROWIN model from the United States Environmental Protection Agency (USEPA, 2000) at 520 years; however, the estimated half-life in aquatic systems is 182 days. The reaction half-life for soil is estimated as one-half year, while that for sediment is one year (Environment Canada, 2010). Since the suspended sediment processes are primarily anaerobic (Davis et al., 2005), the value for sediment was used. The reaction half-life for fish is estimated as 14 years from that for lake trout in Canada (Ismail et al., 2009).

The annual emitted mass of HBCD from the facility was estimated using the maximum proposed tonnage of recycled electronic waste at 30,000 tonnes per year (EWMC, 2011). The proportion of HBCD in e-waste has been estimated as 17 mg per kg (Morf et al., 2005). Therefore, the input mass of HBCD used for the Edmonton scenario was 500 kg as a conservative value for the first year of operation. It is assumed that all of this mass becomes airborne and that this amount is the only input of HBCD into the model.

3. Model results

As HBCD exists as a mixture of three isomers, separate CEMC Level 1 models were first run for each of the α-HBCD, β-HBCD and γ-HBCD forms to assess any differences in partitioning to environmental media. The Level 1 models assume that the chemical is conserved and that partitioning is possible into all media (Mackay, 1998). The model indicates high soil partitioning consistent with the literature (Environment Canada, 2010). The difference in behavior of the three forms was 0.01% between air and soil, most likely due to differing solubilities; therefore, further applications used only α-HBCD because of its greater abundance and its greater tendency to bioaccumulate in organisms (Covaci et al., 2006).

Table 2
Resulting phase properties for HBCD dispersion in environmental media.

Phase	Annual mass (kg)	Proportion (%)	Concentration
Air	0.298	44.6	0.147 ng/m^3
Aerosol	7.84 × 10^{-13}	1.17 × 10^{-10}	4.45 × 10^{-8} ng/g
Water	7.35 × 10^{-6}	1.10 × 10^{-3}	4.45 × 10^{-5} ng/L
Soil	0.359	53.7	2.54 × 10^{-4} ng/g
Sediment	8.52 × 10^{-3}	1.27	1.31 × 10^{-3} ng/g
Suspended sediment	3.22 × 10^{-3}	0.481	1.31 × 10^{-3} ng/g
Fish	3.65 × 10^{-11}	5.45 × 10^{-9}	9.35 × 10^{-8} ng/g

The Level 2 CEMC model was employed to characterize environmental concentrations under steady state conditions. When all major environmental media compartments to and from which HBCD may move are included in its box model parameterization, the CEMC model is able to account for the fate of the emitted chemical through the resulting mass balance. Migration of the chemical is dependent on chemical properties as well as characteristics of the environmental media. The results include estimates of accumulation and concentration in environmental media compartments as well as insights into reactions of the chemical and losses through advection (Mackay, 1998).

The only source of α-HBCD is air-borne and 96.9% of the chemical is lost through atmospheric advection from the area to be distributed more broadly. However, in the region, any remaining HBCD is predicted to be found in air (45%) and soil (54%) as shown in Table 2. Therefore, within the distance between the waste treatment facility and the agricultural or national park lands, the model indicates that HBCD does have the potential to be removed from air by deposition to soil.

Air sampling near landfill sites have resulted in concentrations up to 0.18 ng/m^3 (Remberger et al., 2004). The modeled air concentration of 0.15 ng/m^3 could be a reasonable value for this region. The soil concentration predicted by the model (0.254 pg/g) is low when compared to sampling results from areas with a history of waste processing. For instance, in Sweden, HBCD in soil samples ranged from 111 to 23,200 ng/g (Covaci et al., 2006). However, present modeling did not take into account long-term accumulation and may better represent an initial concentration with the potential to increase annually.

4. Conclusions

Fugacity modeling predicts HBCD deposition is possible in the model area east of the waste management facility. Model air concentrations are similar to those measured at sites with a history of electronics recycling. Therefore, it may be predicted that soil deposition will occur on an annual basis. With the continuous operation of the facility at 30,000 tonnes per year of e-waste recycling, Edmonton may surpass other areas of the world in HBCD deposition in surrounding environments.

Within the distance between the waste treatment center and the agricultural lands, the model indicates that HBCD has potential to be deposited to local soils. This raises implications about the potential for HBCD to be taken up by the agricultural and find its way into the food supply. Currently, there is very little information on HBCD in plant tissues or plant uptake from contaminated soils which should be an area of further research. Increasing concentrations of HBCD have been linked with declining earthworm populations (Law et al., 2006) affecting the health and productivity of the soil environment.

The presence of a protected national park in the model region raises additional concerns about ecosystem accumulation of brominated fire retardants, in general. It is important that HBCD and other brominated fire retardants be monitored in areas

surrounding EWMC to determine deposition of this compound on agricultural lands and natural areas. Since HBCD has been shown to bioaccumulate, the long-term consequences of e-waste recycling could have unexpectedly negative results. If environmental monitoring were to be implemented at the beginning of the facility's operation, initial values for the sampling history would be available to understand potential changes to this and other areas that consider employing e-waste recycling practices aiding in evaluation of risk of exposure to brominated chemicals.

Increased use of electronics results in a corresponding increase in waste as technology rapidly changes (Macauley et al., 2003). E-waste recycling becomes viable for communities who need to dispose of waste, but it also becomes financially favorable as resources diminish (Duan et al., 2011). However noble the concept of recycling becomes, it is important to consider that most processes have a negative outcome that needs to be balanced with the positive. Ecologically-minded communities will engage in environmental monitoring to understand changes that may be taking place and continuous improvement in practices to reduce the potential for negative emissions.

Acknowledgments

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvman.2012.10.024.

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