Organohalogen Compounds, Volume 61, Pages 203-207 (2003)

HUMAN HEALTH RISK ASSESSMENT FOR POLYBROMINATED DIPHENYL ETHERS IN BIOSOLIDS

Paul Chrostowski & Sarah Foster, CPF Associates, Inc. 7708 Takoma Avenue, Takoma Park MD USA

Introduction

Polybrominated diphenyl ethers (PBDEs) are a family of organic chemicals that are widely used as flame retardants. In general, PBDEs are used to fireproof a variety of plastics and fabrics. There are three main PBDEs in commerce – decabromodiphenyl oxide (DDBE), octabromodiphenyl oxide (ODBE), and pentabromodiphenyl oxide (PeDBE). Other PBDEs including tetrabromodiphenyl ether (TeBDE) and hexabromodiphenyl ether (HxBDE) may also be of environmental importance. All have different applications as fire retardants. PBDEs have recently been reported to be accumulating in human breast milk and have been associated with a variety of health effects including neurotoxicity.

PBDEs were reported in municipal sewage sludge in Sweden at concentrations ranging from 21 ng/g to 25 ng/g[1]. Another study from Sweden reported sewage sludge concentrations of TeBDE and PeBDE to be 15 and 19 ng/g, respectively.[2] The Swedish investigators concluded that PBDEs present in sewage sludge were from household and/or industrial discharge rather than atmospheric deposition. Palmquist confirmed the hypothesis that residential discharges were the source of PBDEs to wastewater treatment plants at a rate of 0.1 mg per person per year of HxBDE and 3.45 mg per person per year of TeBDE[3]. In 2001, Hale[4] analyzed 11 samples of biosolids from across the United States and found PBDEs in all the samples. The predominant form of PBDE was PeDBE which was found at an average concentration of 2.3 mg/kg in the biosolids samples. Smaller amounts of a DDBE were also found. These investigators reported PeDBEs in biosolids generated by a variety of methods including composted, lime-stabilized, heat treated and anaerobically digested materials. Due to the lack of standardized analytical techniques and chemical definitions for PBDEs in the environment, it is difficult to determine if the numerical difference between the Swedish and US samples is actual or artifactual. In a subsequent paper, Hale and LaGuardia[5] hypothesized that the presence of PBDEs in biosolids were a significant problem and called for risk assessment.

This paper presents the results of a focused human health risk assessment (HHRA) conducted to address the potential for adverse health effects raised by Hale and co-workers. This risk assessment was conducted according to general principles published by the U.S. Environmental Protection Agency (USEPA)[6] and the U.S. National Academy of Sciences[7]. This HHRA consists of a toxicological evaluation, exposure assessment, and risk characterization.

Toxicological Evaluation

USEPA and others[8] have conducted extensive reviews of the toxicology of the PBDEs. In general, the PBDEs are of low toxicity. They are generally not considered to be acutely toxic, irritating, skin sensitizing, mutagenic, or carcinogenic. USEPA has determined that, at low doses, PeDBE induces liver enzymes in laboratory animals. Although the relevance of liver enzyme induction to clinical human health effects has not been established, in the interest of health protectiveness, USEPA has determined that this is an adverse effect. USEPA has developed a reference dose of 2 µg/kg-day for PeBDE and a reference dose of 10 µg/kg-day for DBDE based on liver enlargement in laboratory animals. ATSDR has developed acute and intermediate maximum risk levels of 0.03 mg/kg-day and 0.007 mg/kg-day based on hepatic and thyroid effects, respectively[9]. Recent evidence suggests that PBDEs may be porphyrinogenic, immunotoxic, teratogenic, and neurotoxic. Neurotoxicity has been found at low doses (0.8 mg/kg) in behavioral tests in laboratory animals, however, the significance of this to humans is currently unknown[10].

Exposure Assessment

PBDEs are hydrophobic chemicals that tend to preferentially adsorb to sediment in the environment. When biosolids are applied to agricultural fields, there is not likely to be significant sediment runoff due to federal and state regulations and best management practices[11]. Because of their sorption potential, PBDEs in the soil are not likely to be transported to groundwater. Because of this behavior, exposure is likely to be limited to direct contact or the human food chain. Three exposure pathways were selected for evaluation –

direct incidental ingestion of soil, consumption of crops grown on biosolids-amended soil, and ingestion of beef from cattle fed with crops grown on biosolids-amended soil. Both child and adult receptors were considered in the HHRA. Two application scenarios were evaluated using agronomic application rates based on design parameters presented by USEPA[12]. The first scenario involved surface application of biosolids. This scenario was evaluated using average concentrations of 2.3 mg/kg for PeDBE and 1.0 mg/kg for DDBE. The second scenario assumed that the biosolids were tilled to a depth of 15 cm (6 inches) yielding incorporated concentrations of 0.08 mg/kg for PeBDE and 0.03 mg/kg for DBDE.

Exposures were calculated using mathematical fate and transport models and input assumptions identified by USEPA for multiple pathway HHRA[13]. The models and input assumptions collectively provide a conservative calculation of risk (i.e., they are expected to overestimate potential health risks). For example, exposures for all pathways, including incidental soil ingestion, were considered to occur 350 days/year every year over a period of 30 years for an adult and 6 years for a child. USEPA's methods further assume that 100% of the produce that a person ingests each year is grown only on biosolids-amended soil and that 100% of the beef consumed by a person each year is entirely comprised of their own home-grown beef. Moreover, all the feed ingested by beef cattle us assumed to be grown on biosolids-amended soil. The extent of uptake of PeDBE and DDBE into produce, feed crops, and beef tissue is calculated based on several regression equations that rely solely on a chemical's octanol:water partition coefficient (Kow). These equations are likely to overestimate uptake for chemicals with high Kow values such as the PBDEs.

Risk Characterization

Potential risks associated with the two land application scenarios were calculated by comparing exposure estimates for each pathway with the references doses for the PBDEs. The ratio of exposure to the reference dose is referred to as a hazard quotient. Hazard quotients were calculated for each chemical and each pathway and then summed across chemicals and exposure pathways. The sum of a number of individual hazard quotients is referred to as a hazard index and represents the potential risk associated with simultaneous exposure to both PBDEs through soil, produce, and beef ingestion. Adverse health effects are not expected to occur if the hazard quotients and hazard indices are below a value of one.

The hazard indices and hazard quotients found in this risk assessment are shown in Table 1 for biosolids tilled into the soil and in Table 2 for biosolids that are surface applied. These results are also summarized in Figure 1. As can be seen, all of these values are below the target level of one, indicating that human health effects are not expected to occur from exposure to PeDBE and DBDE through multiple pathways associated with land application of biosolids.

TABLE 1

POTENTIAL FOR NONCANCER HEALTH EFFECTS FROM PeDBE AND DDBE IN SOIL: BIOSOLIDS INCORPORATED INTO SOIL

Noncancer Hazard Quotients and Hazard Index Values for an Adult

Exposure pathway	PeDBE	DDBE	Total Hazard Index
Soil ingestion	5.5E-05	4.1E-06	6E-05
Produce ingestion	1.1E-03	1.1E-04	1E-03
Beef ingestion	1.3E-03	2.6E-05	1E-03
Total hazard index across pathways	2E-03	1E-04	2E-03

Noncancer Hazard Quotients and Hazard Index Values for a Child

Exposure pathway	PeDBE	DDBE	Total Hazard Index
Soil ingestion	5.1E-04	3.8E-05	5E-04
Produce ingestion	1.6E-03	1.7E-04	2E-03
Beef ingestion	5.8E-04	1.2E-05	6E-04
Total hazard index across pathways	3E-03	2E-04	3E-03

TABLE 2

POTENTIAL FOR NONCANCER HEALTH EFFECTS FROM PeDBE AND DDBE IN SOIL: BIOSOLIDS SURFACE APPLIED

Noncancer Hazard Quotients and Hazard Index Values for an Adult

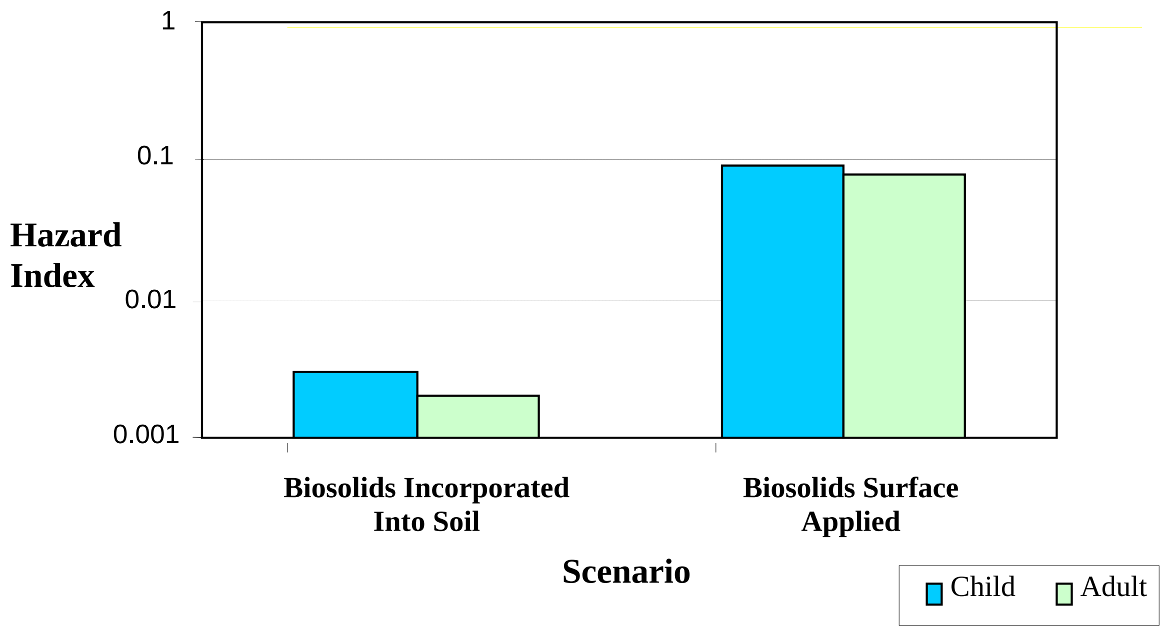
Exposure pathway	PeDBE	DDBE	Total Hazard Index
Soil ingestion	1.6E-03	1.4E-04	2E-03
Produce ingestion	3.1E-02	3.7E-03	3E-02
Beef ingestion	3.7E-02	8.7E-04	4E-02
Total hazard index across pathways	7E-02	5E-03	8E-02

Noncancer Hazard Quotients and Hazard Index Values for a Child

Exposure pathway	PeDBE	DDBE	Total Hazard Index
Soil ingestion	1.5E-02	1.3E-03	2E-02
Produce ingestion	4.6E-02	5.5E-03	5E-02
Beef ingestion	1.7E-02	3.9E-04	2E-02
Total hazard index across pathways	8E-02	7E-03	9E-02

FIGURE 1

PBDE RISK ASSESSMENT RESULTS



Discussion and Conclusions

The mere presence of a chemical in biosolids is not sufficient for concern regarding adverse health effects. In order for health effects to occur, both the conditions of exposure and the toxicology need to coincide to produce a level of significant risk. This HHRA has found that land application of biosolids containing PBDEs will not cause adverse human health effects. There is one significant uncertainty associated with this assessment that could lead to an overestimate of risk. Due to lack of definitive data, we assumed that PBDEs do not biodegrade in the environment. Carson has noted that microbial debromination of PBDEs may occur[14], but the rates have not been measured. Rapid reductive debromination of a dibromodiphenyl ether through anaerobic and photochemical degradation has recently been demonstrated[15]. The land application of biosolids has been shown to enhance biodegradation of other xenobiotics such as alkylphenols[16]. If this phenomenon also applies to PBDEs, the concentrations could be substantially diminished over the exposure period and the potential risks would be reduced proportionately to the degree of biodegradation. Toxicological uncertainty could lead to an underestimate of risk. Immunotoxicity, developmental toxicity, and endocrine effects of PBDEs have not been fully studied. It is possible that resolution of these questions could produce dose-response relationships that are numerically lower than USEPA's existing reference doses. Future exposure and toxicity studies should be performed on individual PBDE congeners to yield a better understanding of structure-activity-relationships.

Acknowledgements
Partial support for this research was provided by an unrestricted grant from Synagro Technologies Inc.

---

[1] Carson, B.L. (2001) Toxicology summary for selected polybrominated diphenyl ethers. National institute of Environmental Health Sciences, Research Triangle Park, NC.
[2] Sellstron (1993) cited in Carson, B.L. *op. cit.*
[3] Palmquist, H. (2001). Hazardous substances in wastewater systems: A delicate issue for wastewater management. Lulea Tekniska Universitat 2001:65.
[4] Hale,R.C., LaGuardia, M.J., Harvey, E.P. (2001). Nature 412:140 and supplementary material.
[5] Hale R.C. and LaGuardia, M.J. (2001). Directions in Science 1:10.
[6] USEPA (1989). Risk assessment guidance for superfund. Volume I: Human Health Evaluation Manual. EPA/540/1-89/002. *et seq*.
[7] National Academy of Sciences. (1983). Risk assessment in the federal government: managing the process. National Academy Press, Washington, DC.
[8] USEPA (2002). Integrated risk information system (IRIS); Hardy, M.L. (2002) Chemosphere 46:757; Carson, B.L. *op cit*., Darnerud, P.O., Eriksen,G.S., Johannesson, T., Larsen, P.B., Viluksela, M. (2001) Environ Health Perspect 109 Suppl 1:49.
[9] ATSDR 2002. Toxicological Profile for PBBs and PBDEs. Draft for Public Comment.
[10] Eriksson, P, Jakobsson, E, Fredricksson, A. 2001. Environ Health Perspect 109:903.
[11] USEPA (1995). Land application of sewage sludge and domestic septage. EPA/625/R-95/001.
[12] USEPA (1995). *Op. cit.*
[13] USEPA (1998) Human health risk assessment protocol for hazardous waste combustion facilities. EPA530-D-98-001.
[14] Carson B.L.(2001) *op cit.*
[15] Rayne, S., Ikonomou, M.G., Whale, M.D. (2003). Water Research 37:551.
[16] Environment Canada. (1998). Alkylphenol ethoxylate persistence in biosolids treated fields. Environment Canada And National Water Research Institute, Soil Project 71773