



Brominated flame retardants and toxic elements in the meat and liver of red deer (*Cervus elaphus*), wild boar (*Sus scrofa*), and moose (*Alces alces*) from Latvian wildlife

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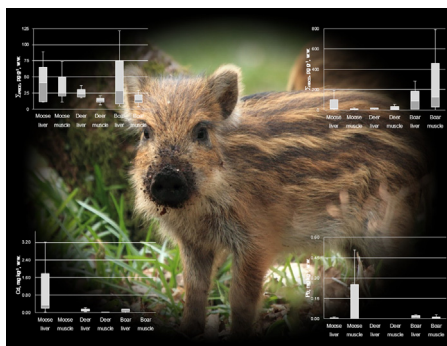
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HIGHLIGHTS

- Occurrence of BFRs and heavy metals in wild game samples from Latvia was evaluated.
- Liver showed higher levels of contaminants in comparison with the musculature.
- Levels of selected BFRs in Latvian terrestrial biota are lower than in other regions.

GRAPHICAL ABSTRACT



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ABSTRACT

In order to evaluate the contamination status of terrestrial biota in Latvia, muscle and liver tissues of red deer (*Cervus elaphus*), wild boar (*Sus scrofa*), and moose (*Alces alces*) were analyzed for the content of polybrominated diphenyl ethers (PBDE), hexabromocyclododecane (HBCD), tetrabromobisphenol A (TBBPA), as well as cadmium and lead. The highest mean concentrations of PBDEs (46.6 pg g^{-1} wet weight (w.w.)), cadmium (0.95 mg kg^{-1} w.w.), and lead (0.22 mg kg^{-1} w.w.) were observed in the tissues of moose, while the wild boar samples contained the highest levels of HBCD, with the mean concentration equal to 264 pg g^{-1} w.w. in muscle tissues. Generally low mean concentrations of TBBPA from 0.52 to 4.54 pg g^{-1} w.w. were observed. The liver tissue of all analyzed specimens was found to contain higher concentrations of contaminants, compared to muscle tissue. The congener profile of PBDEs in the analyzed tissues indicated that the recently used “penta-BDE” formulation was a probable source, while components of HBCD, “octa-BDE”, and “deca-BDE” technical mixtures are likely to undergo congener-specific or diastereomer-specific bioaccumulation or metabolic degradation. Considering the reports from other regions, it can be concluded that the terrestrial biota in Latvia is less affected by the studied contaminants.

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1. Introduction

Brominated flame retardants (BFRs) comprise a diverse group of anthropogenic chemicals that are used to prevent fire incidents. BFRs lower the flammability of a wide array of products, such as textiles,

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plastics, building materials, and electronic equipment (D'Silva et al., 2004). Despite the clear benefits provided by the use of BFRs, the ubiquitous presence of these chemicals has resulted in their diffusion into the environment during the manufacturing, use, and disposal. Due to their persistency and lipophilicity, BFRs enter the terrestrial and aquatic food chains (D'Silva et al., 2004). Some of the most important contaminants in this family are polybrominated diphenyl ethers (PBDEs) with the estimated worldwide production up to 67,000 tons per year in the early 2000s, and extensive use over several decades (U.S. EPA, 2010). However, considering that these compounds are persistent organic pollutants (POPs) and taking into account their potential for long-range transport (Hites, 2004), certain restrictions were imposed on their commercial availability (European Union, 2003). Therefore, in order to meet the market demand for FRs, several alternative BFRs were developed, among which the most consumed were hexabromocyclododecane (HBCD) and tetrabromobisphenol A (TBBPA). Similarly to PBDEs, these alternative BFRs also showed POP-like properties and their use followed by disposal led to environmental contamination (D'Silva et al., 2004; Abdallah, 2016). While HBCD was classified by the European Commission (EC) as a bioaccumulative and toxic compound because of its high persistence, low water solubility, and high log K_{ow} value (European Chemicals Agency, 2008), there is no consensus on the risks due to TBBPA. Older references report that TBBPA seems to have a comparatively low toxicity compared to the other BFR groups (Damerud, 2003). Nevertheless, the most recent studies reflect toxicological concerns regarding the environmental presence of TBBPA (Lai et al., 2015), therefore attracting scientific interest to the occurrence of this chemical in the environment.

Atmospheric transport delivers significant quantities of BFRs from contaminated areas into other aquatic and terrestrial environments where these POPs are readily bioaccumulated through food chains (Law et al., 2014). Considering the fact that terrestrial animals are generally known to be less susceptible to the bioaccumulation of POPs compared to aquatic animals, because aquatic ecosystems are affected by POPs through additional pathways (e.g., via TOC-rich sediments and particulate matter suspended in water and by further bioaccumulation and biomagnification of POPs) (Law et al., 2003; Law et al., 2006), there has been less scientific interest towards the investigation of BFRs in terrestrial wildlife and only a handful of reports are available on this topic, with a focus on such carnivore species as fox and bobcat (Corsolini et al., 2000; Boyles et al., 2017; Fuglei et al., 2007). Less information is available on the occurrence of BFRs in herbivores and omnivores (Suutari et al., 2009; Mariussen et al., 2008; Christensen et al., 2005), even though such data would provide a better understanding of environmental transport, deposition patterns, and the ultimate fate of BFRs.

Another important group of toxicants is heavy metals, among which cadmium and lead are of major concern (Flora and Agrawal, 2017). Urbanization, industrialization, intensive agriculture and aquaculture can affect the environment with heavy metal emissions. Such factors as metal smelting and reclamation, fossil fuel combustion, overuse of mineral fertilizers or improper waste disposal are considered to be the most important sources of undesirable metallic elements in the global environment (Falandysz et al., 2005). Despite the substantial efforts to minimize anthropogenic environmental pollutants since the 1970s, the levels of heavy metal contamination in wildlife still may be significant (Cooper et al., 2017).

Red deer (*Cervus elaphus*) and moose (*Alces alces*), being the two most dominant species (population of ~52,000 and ~21,000 animals in 2014, respectively) after roe deer (*Capreolus capreolus*, population of ~130,000 in year 2014), are the largest herbivores among Latvian wildlife (NeoGeolv, 2014). Red deer and moose have the longest life expectancy and thus are the most susceptible to bioaccumulation of BFRs and heavy metals, and can be considered to be the most sensitive herbivore bioindicators of environmental contamination status. While the diet of red deer and moose consists generally of plants and represents

the air/soil – plant – herbivore system, wild boar (population of ~55,000 in Latvia in 2014 (NeoGeolv, 2014)) is known to be an omnivore consuming a wide variety of plants and insects, as well as carrion, fish, and mollusks that provide an additional uptake of contaminants. It has been shown that the accumulation of heavy metals in plants and soil may increase the risk of transfer to herbivorous wild mammals, including game animals (Falandysz et al., 2005; Bilandzic et al., 2012). In contrast to pure herbivores, wild boars have a highly adaptable diet and are exposed to contaminants via the multiple pathways. Therefore, the differences in nutritional habits of herbivores (red deer and moose) and an omnivore (wild boar) could provide an insight into the exposure routes and bioaccumulation of BFRs and heavy metals.

In this study, we carried out a broad analysis of the most commonly used chemicals from the BFR family, namely, PBDEs, HBCDs, and TBBPA, as well as two heavy metals – lead and cadmium. Considering the bioaccumulation of the selected contaminants, as well as previous recommendations for field studies (Lazarus et al., 2014; Neila et al., 2017; Van den Brink et al., 2016), meat and liver were selected for the analysis within the framework of this study. In order to assess the geographical differences in the distribution of contaminants, the specimens were sourced from different districts of Latvia.

2. Materials and methods

2.1. Samples

A total of twenty-four wild animals were analyzed in the current study, including seven specimens of moose, eight specimens of red deer, and nine wild boars. The individual specimens were killed by local hunters in different districts of Latvia (Fig. 1), according to the relevant Latvian legislation for game hunting during the period from September to December 2016. The animals were shot by hunting rifle and their age and sex were determined. The dissection of the animal carcasses and sampling of the musculature and liver was carried out in the field. Animal tissue samples were packed in polyethylene bags with ice, uniquely coded and delivered to the laboratory within 24 h. In order to minimize the influence of possible contamination of the samples during transportation, approximately one centimeter thick layer was removed from the surfaces of samples after receiving at the laboratory, the samples were homogenized and stored at -18°C until analysis. The appropriate sample amounts (5–20 g) were analyzed for the content of twenty-four PBDE congeners, three HBCD diastereomers, TBBPA, lead, and cadmium. Detailed information about the analyzed specimens is presented in Table 1.

2.2. Chemicals and materials

All of the solvents used were at least of pesticide purity grade. Silica gel for column chromatography, nitric acid, hydrogen peroxide, and sulfuric acid were purchased from Sigma-Aldrich Chemie GmbH (Buchs, Switzerland) or from Acros Organics (Morris Plains, NJ, USA). The native and isotopically labeled standards for the analyzed BFRs were purchased either from Cambridge Isotope Laboratories (Tewksbury, MA, USA) or from AccuStandard (New Haven, CT, USA). Standard solutions for lead and cadmium were obtained from Merck (Darmstadt, Germany).

Along with the analysis of lead and cadmium, the following groups of brominated flame retardants were determined (for compounds given in bold, $^{13}\text{C}_{12}$ -labeled surrogates were available and were used as internal or recovery standards):

- 1) Di- through decabrominated diphenyl ethers (PBDEs): IUPAC numbers 7, 15, 17, **28**, **47**, 49, 66, 71, 77, 85, **99**, **100**, 119, 126, 138, **139**, **153**, **154**, 155, 166, 181, **183**, 190, **209**.
- 2) Tetrabromobisphenol A and hexabromocyclododecane diastereomers: **TBBPA**, α -HBCD, β -HBCD, and γ -HBCD

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