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Intake, distribution, and metabolism of decabromodiphenyl ether and its main metabolites in chickens and implications for human dietary exposure☆

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ABSTRACT

Diet is considered as the most important human exposure pathway for polybrominated diphenyl ethers (PBDEs). Metabolism and accumulation patterns of PBDEs in different growth periods of chickens are helpful for evaluating human dietary exposure, but such information is scarce. In this study, female chickens were fed with food spiked with BDE-209 at 85 mg kg^{-1}, and the intake, accumulation, and excretion of BDE-209 and its main metabolites in various tissues were examined. Concentrations of BDE-209 in chicken tissues increased over time in a tissue-specific manner; they were the greatest in liver and generally the lowest in breast meat during the entire exposure period. The kinetic patterns were dependent on both growth-dilution effects and accumulated concentrations of BDE-209. Tissue concentrations of $\sum_8$PBDE (sum of BDE-28, 47, 99, 100, 153, 154, 183, and 209) followed the sequence of liver > blood > skin > intestine > stomach > leg meat > breast meat. Different tissue partition coefficients and perfusion rates for blood may have resulted in different PBDE concentrations in tissues. The absorption efficiency of BDE-209 in chicken tissues followed the sequence of liver (0.15 ± 0.032%) > skin (0.14 ± 0.038%) > intestine (0.071 ± 0.021%) > breast meat (0.062 ± 0.020%) > leg meat (0.059 ± 0.016%) > stomach (0.021 ± 0.0095%), likely due in part to facilitated absorption of BDE-209 by transport proteins (P-glycoproteins). On average, 9.3 ± 1.7% of BDE-209 was excreted in feces. Estimated human average dietary intake via the consumption of chicken tissues of $\sum_8$PBDE for adults and children was 319 and 1380 ng day^{-1} for liver, 211 and 632 ng day^{-1} for leg meat, and 104 and 311 ng day^{-1} for breast meat from the contaminated group. Liver clearly poses the highest exposure risk for human consumption, particularly if chickens are fed with contaminated feed.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are widely used as flame retardants (Alonso et al., 2012; Pirard and De Pauw, 2007). Penta-, octa-, and deca-BDE commercial mixtures have been gradually banned from manufacture and use by the European Union (EU) since 2004 (EU, 2004, 2008), because PBDEs could result in thyroid toxicity, developmental neurotoxicity, and endocrine disruption on animals and humans (Bellés et al., 2010; Kim et al., 2013; Reverte et al., 2014). Despite these bans and restrictions on them, PBDEs can still be easily released to the environment as they are physically bonded in products (Chen et al., 2007; de Wit, 2002; Domingo, 2012). They biomagnify through the food web (Losada et al., 2009; Mizukawa et al., 2009), resulting in higher PBDE concentrations in humans through consumption of food (Ohta et al., 2002).

Therefore, reducing the concentrations of PBDE in food is

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significant for the protection of human health, which in turn relies on informed knowledge about the bioaccumulation and metabolism of PBDEs in animals.

Previous studies have focused on the concentrations of PBDEs in chicken tissues (Zhao et al., 2016), as well as those of PBDEs, hexabromocyclododecanes, polychlorinated dibenzo-p-dioxins, tetrabromobisphenol, polychlorinated biphenyls, and polychlorinated dibenzofurans in chicken eggs (Piskorska-Pliszczynska et al., 2016; Squadrone et al., 2015; Zeng et al., 2016). The pharmacokinetics of low-brominated PBDEs in chicken (*Gallus gallus domesticus*), a major food item worldwide, were also carefully investigated (McKernan et al., 2010). However data about bioaccumulation kinetics of highly brominated BDE congeners in chicken have remained scarce (Covaci et al., 2011). Pirard and De Pauw (2007) found that concentrations of ∑PBDE (sum of BDE-47, 99, 100, 153, 154, and 183) were higher in adipose than in liver and eggs of laying hens. Voorspoels et al. (2006) indicated that the concentrations of ∑PBDE (sum of BDE-28, 47, 99, 100, 153, 154, and 183) in the tissues of common buzzards (26–130 ng g^{-1} lipid weight) were an order of magnitude lower than those of sparrow hawks (360–1900 ng g^{-1} lipid weight); BDE-209 was found in liver and serum only. Understanding the pharmacokinetics of a wide range of PBDEs in chicken tissues is significant for quantifying the mechanisms of PBDE bioaccumulation in chicken, a major source of proteins for the general population. This information is critical for gauging the magnitude of human health risk through consumption of chickens, because diet is considered as the most important human exposure pathway for PBDEs (Kang et al., 2010; Ni et al., 2012) and BDE-209 is the prominent congener in most animal-derived food products (EFSA, 2011).

Dietary habits also vary by country; e.g., viscera organs such as liver and intestines are widely consumed in some countries such as Korea, France, and China (Hoffmeister et al., 2007; Xing et al., 2010), but are relatively uncommon in Japan and North America. Because chicken offal is often used to feed other livestock, this practice would eventually cause exposure risk to humans. Therefore, distribution patterns of PBDEs in chicken tissues at different growth stages are of great significance for assessing human health effects. Distribution patterns of PBDEs in chicken visceral organs, and particularly in liver, would provide insights into metabolism, migration, and accumulation of PBDEs in chickens. A previous study on rats suggested that lipophilic tissues such as skin, adipose, and gastrointestinal tract were the major reservoirs of PBDEs (Orn and Klasson-Wehler, 1998). Currently, no data are available on the accumulation and transformation patterns of PBDEs in poultry of different growth stages.

To address the above-mentioned knowledge gap, we carried out a series of studies on the bioaccumulation kinetics of BDE-209 in chicken domesticated in a farmland. The objectives of the present study were to (1) obtain concentrations of BDE-209 in chicken tissues of different growth stages; (2) compare the distributions of BDE-209 and its main metabolites accumulated in different tissues of chicken; and (3) estimate the dietary intakes of PBDEs for children and adults via the consumption of chicken muscle and liver and related potential health risk.

2. Materials and methods

2.1. Materials

Eight target PBDE standard solutions, including BDE-28, 47, 99, 100, 153, 154, 183, and 209), sum of which is designated as $\sum_8$PBDE, three internal standards, i.e., BDE-69, 3-fluoro-2,2′,4,4′,5,6-hexabromodiphenyl ether (F-BDE-139), and 4′,6-difluoro-2,2′,3,3′,4,5,5′,6′-octabromodiphenyl ether (F-BDE-201), and three surrogate standards, including BDE-51, 115, and 4′-fluoro-2,2′,3,3′,4,5,5′,6,6′-nonabromodiphenyl ether (F-BDE-208), were all purchased from AccuStandard (New Haven, CT, USA). Sulfuric acid was obtained from Damao (Tianjin, China). Dichloromethane and *n*-hexane of HPLC grade were obtained from Oceanpak (Gothenburg, Sweden), and acetone of analytical reagent grade was obtained from GongDong Hing Wah (Guangdong, China). Bio-Beads SX-3 was obtained from AnPel (Shanghai, China). Acetone was redistilled in an all-glass system before use, and the Bio-Beads were soaked in dichloromethane until use.

Sunflower oil (Hebei, China) was spiked with BDE-209 and mixed with 8.5 kg of commercially available feed under constant agitation (150 rpm at 20 °C) to prepare contaminated feed with final nominal concentrations of 85 mg kg^{-1} for BDE-209, with impurities of 0.04 and 4.4 mg kg^{-1} for BDE-47 and 99, respectively. The concentration of BDE-209 prepared in contaminated feed was similar to that in soil (mean: 58.7 mg kg^{-1}; range: 17–146 mg kg^{-1}) near a manufacturing plant (Li et al., 2015). The control feed was treated in an analogous manner with no BDE-209 added.

2.2. Accumulation of BDE-209 in chicken

Two groups of 30-day old female chickens, each with 15 individuals, were raised indoors under controlled conditions in individual hencoops in a chicken farm at Luogang (Guangzhou, China), equally separated as contaminated and control groups and fed with contaminated and non-contaminated feeds, respectively. After a 10-day period of adaptation, each individual chicken was reared with 90 g of feed per day. At each 10-day exposure time, three chickens from contaminated and control groups, respectively, were slaughtered. During the entire exposure period of 50 days, the mass of the chickens was weighed and chicken feces were collected.

2.3. Sample preparation and extraction

For each batch of slaughtered chickens, blood was collected in polytetrafluoroethylene tubes and agitated at 3500 rpm at 20 °C. Serum was collected and stored at −80 °C. Tissue samples (liver, intestine, stomach, skin, leg meat, and breast meat) and feces samples were excised and wrapped by aluminum-foil, vacuumed, and stored at −80 °C until processing. At that time, each freeze-dried tissue sample was ground into fine homogeneous powders using a grinding miller, wrapped by aluminum-foil, vacuumed, and stored at −80 °C until extraction. The contaminated and control groups were processed separately to prevent cross contamination.

Each sample of 5 g for the control group or 0.5 g for the contaminated group chicken was spiked with surrogate standards and sonicated three times with a mixture of hexane: dichloromethane: acetone (2:2:1 in volume). Twenty percent of the combined extract was used to determine lipid content. Sulfuric acid was added to the remaining extract to remove organics. The supernatant of the sulfuric acid extract was concentrated to 2 g, then fractionated with 30 mL of dichloromethane: hexane (1:1 in volume) through a gel permeation chromatographic column containing 6 g of Bio-Beads SX-3. The fraction eluted at 15–30 mL was reduced to 100 μL under a gentle nitrogen stream. Finally, internal standards were spiked before instrumental analysis.

2.4. Instrumental analysis

Extracts were analyzed with an Agilent 7890A gas chromatograph coupled to a 5975C mass spectrometer in the negative chemical ionization mode. A DB-5HT capillary column (15 m × 0.25 mm i.d; 0.1 μm film thickness) was used to separate