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Sources, gastrointestinal absorption and stereo-selective and tissue-specific accumulation of Dechlorane Plus (DP) in chicken

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HIGHLIGHTS

- Soil was the main DP source in chicken rather than food.
- No stereo-selective absorption was found for DP isomers in gastrointestinal tract.
- Lipid content and blood perfusion state affect DP tissue distribution.
- *Anti*-DP selectively accumulates in chicken especially in fat, brain, and liver.
- *Anti*-Cl_{11}-DP derived mainly from absorption rather than *in vivo* dechlorination.

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ABSTRACT

Dechlorane Plus (DP) isomers, along with two dechlorinated metabolites were measured in environmental matrices, chyme and digestive tract contents and tissues of chicken from an e-waste recycling site located in South China. Soil is proved to be the main source of DP in chicken rather than food because soil contributes more than 94% of total DP in chyme. In the gastrointestinal tract absorption processes, no selective absorption was observed for DP isomers during the ingestion processes. The tissue distribution of DP isomers in chicken exhibits complicated characteristics. The lipid contents in tissues are the main factors in the tissue distribution of DP, while the different blood perfusion state and the different tissue functions also seem to influence the tissue distribution of DP. The fat, brain, and liver exhibit higher f_{anti} values (0.65, 0.64, and 0.64) than the other tissues (0.54–0.59). The elevated f_{anti} values of DP from the contamination source (0.52 in soil) to chicken suggest stereoselective bioaccumulation of *anti*-DP in chicken. The similar ratios of *anti*-Cl_{11}-DP to *anti*-DP between soil and chicken imply that *anti*-Cl_{11}-DP mainly derives from the uptake from environment rather than *in vivo* dechlorination.

1. Introduction

Dechlorane Plus (DP) has been widely used as flame retardants in electrical wire, cable coatings, computer connectors and other polymeric systems for more than 40 years. DP has been categorized as a high-production-volume (HPV) chemical by the U.S. Environmental Protection Agency (US EPA, 2011). As a highly chlorinated compound, DP is considered to be very lipophilic (water solubility < 10 ng L^{-1}; log octanol–water partition coefficient >7) with low vapor pressure (less than 10^{-6} Pa) (Oxychem, 2011; US EPA, 2011). The technical DP mixtures consist of two isomers, namely the *syn*- and *anti*-DP. The isomer composition ratio is approximately 1:3 of *syn*- to *anti*-forms for all the three technical DP products (DP-25, DP-35 and DP-515). The occurrence of DP was first reported in air, soil and fish samples from the Great Lakes in 2006 (Hoh et al., 2006). Since then, DP has been ubiquitously detected in environment matrices, wild creatures and human beings (Sverko et al., 2011; Xian et al., 2011).

The isomer ratio of DP (f_{anti}) is defined as the fraction of *anti*-DP in the total DP (*anti*-DP and *syn*-DP). Several factors could influence the DP isomer compositions in the environmental matrices and organisms. A significant relationship exists between the f_{anti} and the distance from the manufacturing site in the particle phase samples from the Great Lakes region (Hites et al., 2010; Venier and Hites, 2008). Higher f_{anti} values detected in sediments compared to those of the commercial DP products indicate that *syn*-DP is more vulnerable to microorganism degradation (Sverko et al., 2010), which is also supported by other studies (Wang et al., 2010; Yu et al., 2010). Despite the numerous field studies in

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wildlife and numerous laboratory experiments, the isomer ratio data reveal complex features in biota samples (Sverko et al., 2011; Xian et al., 2011). A preference for *syn*-DP has been reported in various fish species in southern China (Wu et al., 2009), South Korea (Kang et al., 2010), and Lake Winnipeg and Lake Ontario (Tomy et al., 2007). This phenomenon has been confirmed by laboratory exposure experiments with juvenile rainbow trout exposed to DP (Tomy et al., 2008). Meanwhile, many studies reported similar *anti*-DP fraction in terrestrial creatures with the commercial DP products (f_{anti}: 0.65–0.75) (Tomy et al., 2007; Oxychem, 2011), including peregrine falcons eggs (0.77 in Spain) (Guerra et al., 2012), terrestrial passerine bird tissues (0.73–0.82) (Sun et al., 2012), and human hair (0.70) (Zheng et al., 2010). Relatively low f_{anti} values were also observed in terrestrial lives in previous studies. Peregrine falcons eggs exhibit a ratio of 0.58 in Canada (Guerra et al., 2012). Human hair (0.55) (Zheng et al., 2010) and serum (0.53) (Ren et al., 2009) from e-waste recycling sites also have lower f_{anti} values relative to those in the reference site (0.70 for hair and 0.63 for serum). For example, the fraction of *anti*-DP declined with the increasing trophic levels in both fish and bird species (Wu et al., 2009; Kang et al., 2010; Zhang et al., 2011a,b; Sun et al., 2012), suggesting that *syn*-DP is more bioaccumulative compared with *anti*-DP. The f_{anti} values can also be affected by the exposure dose to organisms. A stereo-selective accumulation of *syn*-DP was observed in rat and quail only when they were exposed to high dose of DP (Li et al., 2013a,b). The differences in DP structural configuration, binding affinity to protein, and the uptake and elimination ratios between the two isomers are suggested to play an important role in the bioaccumulation of DP (Li et al., 2013a,b). Moreover, the tissue specific stereo-selective distribution of DP was also reported in fish species (Zhang et al., 2011a,b; Pang et al., 2012), mainly determined by lipid partition. Since few studies simultaneously report the isomer composition of DP in organisms as well as their diet, it still remains unclear whether there is stereo-selective accumulation for DP in terrestrial birds. Additionally, few available studies concern the behavior of DP isomers in full bioaccumulation processes, such as dietary assimilation efficiency, the subsequent blood transportation, the whole body distribution, and the possible hepatic metabolism.

Soil and feed intake are both demonstrated as important exposure routes for chicken dietary exposure to the environmental chemicals (Eljarrat et al., 2008; Covaci et al., 2009; Waegeneers et al., 2009; Foster et al., 2011; Fournier et al., 2012). After the food (or soil) uptake processes, DP also exhibit tissue-specific deposition due to factors more than lipid partition efficiencies (Pang et al., 2012). Compartment is an important concept in pharmacokinetics and pharmacodynamics. Blood is believed to be the vehicle for the transportation of xenobiotic chemicals to different compartments (Rang, 2003; Savkovic). Pollutants are administrated to and distributed from the central compartment with abundant blood perfusion, such as kidney, heart, lung, and liver. After then the pollutants enter the peripheral compartment with poor blood perfusion such as the muscle, brain and fat (Rang, 2003). Only limited data is available about the DP toxicity. Crump et al. (2011) observed no significant changes in 11 target genes of chicken embryos. However, DP exposure was demonstrated to alter alkoxyresorufin O-dealkylase (AROD) activity in quail, and mRNA expression levels and CYP2B2 activities in rats (Li et al., 2013a,b). In the present study, we measured the levels and isomer profiles of DP in soil, chicken food and chyme to reveal the pollution routines, in chyme, intestinal contents and feces to study the gastrointestinal absorption process, in various tissues (liver, muscle, kidney, heart, gonad, brain, lung, and fat) of chicken to explore the blood transfer and tissue distribution of DP. The primary objective of the study was to reveal the different behaviors of DP isomers in dietary intake, gastrointestinal absorption processes and tissue distribution in chicken, so as to provide important information for insight knowledge of DP bioaccumulation parameters, isomer-specific pharmacokinetics, instructions for DP toxicokinetic model, and further risk assessment in birds or even terrestrial creatures.

2. Materials and methods

2.1. Sample collection

Chickens (*Gallus gallus domesticus*, $n = 12$; 1 cock, 11 hens) aging for several weeks were purchased from a local market in October 2011 and raised in a farmer's yard for seven months. The yard was located in an e-waste recycling site in Qingyuan county, Guangdong province and surrounded by e-waste recycling workshops. Atmospheric particles ($n = 10$), chicken food (mixture of rice, wheat and other types of grain, $n = 9$) and soil ($n = 10$) samples were collected during chicken breeding in the yard. After the chickens were sacrificed in May 2012, 8 kinds of tissues ($n = 12$ for liver, muscle, heart, gonad, brain, lung, and fat, and $n = 10$ for kidney), chyme and digestive tract contents ($n = 12$ for chyme, intestinal contents and feces) were collected. All the samples were weighted and stored at −20 °C until further analysis. Since there is only one cock, the gender difference in accumulation of DP was not discussed.

2.2. Sample preparation and analysis

Chicken tissues were extracted and cleaned according to the previously published method (Luo et al., 2009), with minor modification. Briefly, after being spiked with surrogate standards (BDEs 77, 181, 205), approximately 2 g of the lyophilized samples were extracted with 190 mL hexane/acetone (1/1, v/v) for 48 h. An aliquot of the extract was used to determine lipids (gravimetrical method); the rest of the extract was subjected to gel permeation chromatography column packed with 40 g of SX-3 Bio-Beads (Bio-Rad Laboratories, Hercules, CA) and further to a multilayer silica gel column. The extract was concentrated to near dryness under gentle nitrogen and reconstituted in 300 μL of iso-octane for analysis. Prior to instrumental analysis, the extracts were spiked with known amounts of internal standards BDE 118, BDE 128, 4-F-BDE 67, 3-F-BDE 153 for determination. More details were shown in Supplementary Materials.

2.3. Quality assurance/quality control

The method quality assurance (QA) and control (QC) was performed by regular analysis of procedural blanks, spiking blanks (DP mixture spiked in solvent blanks), and blind triplicate samples. Details were provided in Supplementary Materials.

2.4. Data analysis

Statistical calculations were performed by SPSS 16 for windows (SPSS). The level of significance was set at $p = 0.05$ throughout the present study. The differences in compound levels (logarithmic transformation) and f_{anti} values among the different groups (chyme, intestinal contents and feces; 8 kinds of chicken tissues) were analyzed using one-way analysis of variance (ANOVA). The calculation of DP contributions from food and soil to chyme, and f_{anti} values in whole chicken were provided in Supplementary Materials.