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Brominated flame retardants and organochlorines in the European environment using great tit eggs as a biomonitoring tool

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ABSTRACT

Large-scale studies are essential to assess the emission patterns and spatial distribution of organohalogenated pollutants (OHPs) in the environment. Bird eggs have several advantages compared to other environmental media which have previously been used to map the distribution of OHPs. In this study, large-scale geographical variation in the occurrence of OHPs, such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and organochlorine pesticides (OCPs), was investigated throughout Europe using eggs of a terrestrial residential passerine species, the great tit (*Parus major*). Great tit eggs from 22 sampling sites, involving urban, rural and remote areas, in 14 European countries were collected and analysed (5–8 eggs per sampling site). The environmentally most important congeners/compounds of the analysed pollutants were detectable in all sampling locations. For PCBs, PBDEs and OCPs, no clear geographical contamination pattern was found. Sum PCB levels ranged from 143 ng/g lipid weight (lw) to 3660 ng/g lw. As expected, PCB concentrations were significantly higher in the sampled urban compared to the remote locations. However, the urban locations did not show significantly higher concentrations compared to the rural locations. Sum PBDEs ranged from 4.0 ng/g lw to 136 ng/g lw. PBDEs were significantly higher in the urbanized sampling locations compared to the other locations. The significant, positive correlation between PCB and PBDE concentrations suggests similar spatial exposure and/or mechanisms of accumulation. Significantly higher levels of OCPs (sum OCPs ranging from 191 ng/g lw to 7830 ng/g lw) were

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detected in rural sampling locations. Contamination profiles of PCBs, PBDEs and OCPs differed also among the sampling locations, which may be due to local usage and contamination sources. The higher variance among sampling locations for the PCBs and OCPs, suggests that local contamination sources are more important for the PCBs and OCPs compared to the PBDEs. To our knowledge, this is the first study in which bird eggs were used as a monitoring tool for OHPs on such a large geographical scale.

1. Introduction

The presence of organohalogenated pollutants (OHPs) in the environment has been a great cause of concern, because of their persistent character, bioaccumulative potential and adverse effects on both humans and wildlife (Vos et al., 2000). Different OHPs have, for example, been shown to cause effects on reproduction in birds through different mechanisms, such as eggshell thinning, embryotoxicity and effects on reproductive behaviour (Elliott and Martin, 1994; Fernie et al., 2008; Gilbertson et al., 1991; McCarty and Secord, 1999). However, sensitivity to the effects of OHPs varies among bird species, as for example passerine species have been shown to be less sensitive to the effects of DDE on eggshell quality than other groups of birds (Gill et al., 2003). There is also evidence of long-range transport of these substances to regions where they have never been used or produced. As a consequence these pollutants have been distributed worldwide and even remote locations, such as the polar regions, have been reached by these pollutants (Braune, 2007). Due to regulatory controls on the use of these compounds, there seems to be a decreasing temporal trend of polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) in biota (Jones and de Voogt, 1999). Nevertheless, concentrations in the environment are still prominent and may exert a potential health risk. Recently, the presence of polybrominated diphenyl ethers (PBDEs) in the environment has received much attention (Law et al., 2006). PBDEs are a group of chemicals that are widely used in different materials, such as plastics, textiles and foams, because of their flame retarding properties. Large-scale production and use have led to their ubiquity in the environment and in biota, in which PBDE levels have increased rapidly (Elliott et al., 2005; Hites, 2004; Norstrom et al., 2002). The production and use of the Penta- and Octa-BDE mixtures have recently been banned in Europe (Directive EEC, 2003). Currently, the Deca-BDE product is the only PBDE commercial mixture which can still be used in the EU. There is increasing evidence that BDE 209 in the Deca-BDE mixture may debrominate in the environment and in biota to form less-brominated BDE congeners which are more bioaccumulative and toxic than BDE 209 itself (Söderström et al., 2004; Stapleton et al., 2004; Van den Steen et al., 2007a).

Monitoring studies are essential to assess the current levels and risks of different OHPs in the environment. Birds' eggs have been used successfully to monitor OHPs in numerous studies (Donaldson et al., 1999; Elliott et al., 2005; Jaspers et al., 2005; Norstrom et al., 2002; Van den Steen et al., 2006; 2008). Eggs of most bird species can easily be collected and the removal of a single egg from a clutch is expected to have only a minor effect on the population level (Furness, 1993; Henny and Kaiser, 1996, Henny et al., 2004). Because eggs can readily be sampled from the same location each year, long-term monitoring studies using eggs are also feasible. Moreover, the study of widespread bird species enables monitoring on a larger spatial scale. Large-scale geographical studies are very valuable to obtain more information about the local usage, emission patterns and spatial distribution of OHPs. However, to date, few studies have monitored concentrations of OHPs in birds' eggs on a broad geographic scale. For example, double crested cormorant (*Phalacrocorax auritus*) and herring gull (*Larus argentatus*) eggs have been used to assess the spatial distribution of OHPs in the Great Lakes (Hebert et al., 1994; Ryckman et al., 1998). Other media that have been used to map the spatial distribution of OHPs are passive air samplers (Gioia et al., 2006; Jaward et al., 2004), pine needles (Jensen et al., 1992), tree bark (Simonich and Hites, 1995) and butter (Kalantzi et al., 2001). In contrast with these environmental media, birds are generally exposed to OHPs through the same routes of exposure (i.e. food) as humans. Another advantage of the use of birds is that they can be used as indicators of exposure and effects at the same time. Potential effects on reproduction and survival can be monitored and linked to the observed pollutants. Residential passerine bird species are particularly useful for monitoring local contamination with OHPs. In contrast with birds of prey and other species, residues in eggs of residential passerine species are expected to reflect local contamination much better because of their small home ranges, territories and foraging areas (Moore, 1966; Dauwe et al., 2006).

The great tit (*Parus major*) is a small insectivorous songbird species with a small foraging area close to the nest site (Cramp and Perrins, 1993). During the breeding season, they feed primarily on caterpillars, spiders and beetles (Cramp and Perrins, 1993). Great tits have several characteristics which make them very useful as a biomonitor for pollutants (Van den Steen et al., 2006). The chief asset of great tits as biomonitors is their ubiquity, which permits sampling in almost all the wooded areas of Europe (Eens et al., 1999). Because great tits are cavity-nesting birds, nest sites are often a limiting resource and they will readily nest in man-made nest boxes. Thus, breeding populations can rapidly be established and monitored, and eggs can easily be collected. Therefore, great tits are widely used as a study species in both behavioural and ecotoxicological research. They have, for example, been proven useful as long-term monitors of climate changes in Europe (Sanz, 2002; Visser et al., 2003). Great tits have large clutch sizes (up to 12 eggs; Cramp and Perrins, 1993) and one randomly collected egg has been shown to represent the contamination levels of PCBs, PBDEs and DDTs of the whole clutch (Van den Steen et al., 2006). In a previous study with great tit eggs, we investigated the small-scale geographical variation of OHPs in Flanders (Belgium; Van den Steen et al., 2008). In that study, levels of PCBs and PBDEs were highest in the industrialized sampling locations, whereas OCPs were highest in the rural locations. Previous studies with other monitoring tools, such as air samplers, have also shown that the levels of PCBs and PBDEs are linked to the degree of urbanization and industrial development and that OCP levels are highest in rural areas (Harner et al., 2006; Jaward et al., 2004). The objective of the present study was to investigate the geographical variation in the occurrence of different OHPs (PCBs, PBDEs and OCPs) in the European environment using the eggs of great tits. This was done to gather more information about background levels in Europe, to provide a baseline for long-term biomonitoring and to identify potential contamination sources. In addition, we investigated the influence of the type of sampling location on the presence of OHPs. To achieve this, we analysed great tit eggs from 22 sampling locations, involving urban, rural and remote locations, from 14 countries in Europe. Based on previous studies, urban sampling locations were expected to have the highest levels of PCBs and PBDEs, while OCPs were expected to be highest in the rural sampling locations.

2. Materials and methods

In the breeding season of 2006 (April–May), researchers from 14 European countries (Fig. 1) collected great tit eggs in 22 existing nest box populations. In agreement, one random egg per clutch (5–8 eggs per sampling site, see Table 1) was gathered. The eggs were collected before incubation, labeled individually and stored in a freezer (−20 °C) until transported to the laboratory. Eggs were transported on dry ice and stored frozen until further analysis. In total, 145 great tit eggs were analysed for PCBs, PBDEs and OCPs. A questionnaire was sent to the collectors in order