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Halogenated pollutants in terrestrial and aquatic bird eggs: Converging patterns of pollutant profiles, and impacts and risks from high levels

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ABSTRACT

We investigated the presence, levels, relationships, and risks of HCHs, DDTs, chlordanes, mirex, PCBs, and brominated flame retardants (BFRs) in terrestrial and aquatic bird eggs from an area in South Africa where DDT is used for malaria control. We found one of the highest ΣDDT levels reported this century; 13 000 ng/g wm (wet mass) in Grey Heron eggs which exceeds critical levels for reproductive success (3000 ng/g wm) calculated for Brown Pelicans, with a no-effect level estimated at 500 ng/g wm. Even higher ΣDDT levels at 16 000 ng/g wm were found in House Sparrow eggs (possibly the highest ever recorded for sparrows), with a maximum of 24 400 ng/g wm. Significant eggshell thinning in Cattle Egrets (33% between thickest and thinnest) was associated with increased levels of *p,p′*-DDT and *p,p′*-DDE. There were indications of unknown use of DDT and lindane. Relative to DDT, PCBs and BFRs levels were quite low. Ordinated data showed that different terrestrial pollutant profiles converged to a homogenised aquatic profile. Converging profiles, high levels of DDT in heron and sparrow eggs, and thinning eggs shells, indicate risk and impacts at release, in the aquatic environment, and in between. If characteristic life-strategies of birds in warm areas (e.g. longer-lived and fewer eggs per clutch) increases the risk compared with similar birds living in colder regions when both experience the same environmental pollutant levels, then malaria control using DDT probably has more significant impacts on biota than previously realised. Therefore, risk assessment and modelling without hard data may miss crucial impacts and risks, as the chemical use patterns and ecologies in Africa and elsewhere may differ from the conditions and assumptions of existing risk assessment and modelling parameters. Consideration of other findings associated with DDT from the same area (intersex in fish and urogental birth defects in baby boys), together with the findings of this study (high levels of DDT in bird eggs, eggshell thinning in the Cattle Egrets, and the apparent absence of breeding piscivore birds in the sprayed area) are strongly suggestive of negative impacts from DDT spraying for Malaria control. Our data presents strong arguments for an expedited process of replacing DDT with sustainable methods.

1. Introduction

Many reports about pollutants in birds concentrate on a single species (e.g. Elliott et al., 2012; Gentes et al., 2012; Gomez-Ramirez et al., 2012), compare related species (e.g. Hoffman et al., 1998; Morales et al., 2012), or consider the relationships of a specific compound between different species (e.g. Ramirez et al., 2011; Rudel et al., 2011). Only a few studies such as Gao et al. (2009) considered and compared different pollutants in disparate terrestrial and aquatic bird eggs from the same region. We collected and analysed aquatic and terrestrial bird eggs from a region in the Limpopo Province of South Africa with a mosaic of uses and releases of chlorinated and brominated compounds. Part of this region is controlled for malaria by indoor residual spraying (IRS) using DDT and other insecticides (Bouwman et al., 2012b). Additionally, chlorinated pesticides could occur in the region due to legacy as well as current use and through industrial chemicals occurring in imported products. The patterns and risks of DDT in breast milk from the region is reasonably well understood (Bornman et al., 2010; Bouwman et al., 2012b), there is good understanding of DDT in and around treated homesteads (Bornman et al., 2012; Van Dyk et al., 2010), and there is some understanding of the impacts of DDT in the aquatic environment (Barnhoorn et al., 2009, 2010).

The aim of this study was to investigate the presence, levels, relationships, impacts, and risks of HCHs, DDTs, chlordanes, mirex, PCBs, and brominated flame retardants (BFRs) in bird eggs in a complex rural source and release scenario, where it is assumed

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that the primary contaminant would be DDT from malaria control. It is predicted that, at least for DDT, the levels in eggs will increase from west to east, as this is the direction of increased DDT use, and thereby also the risk. With such knowledge, recommendations about protective interventions can be motivated.

2. Methods

2.1. Sampling sites and species

Fig. 1 shows the sample region for this study, located in the Limpopo Province of South Africa. Thohoyandou (the capital of the Province) is the largest city in this area with about 500 000 inhabitants. Towards the north is Zimbabwe, and towards the east is the northern part of the Kruger National Park (KNP). Many smaller villages and settlements are located here, interspersed with subsistence and small-scale agriculture. All rivers drain east to east towards the Indian Ocean. No large industries occur in the area, with the exception of commercial agriculture (fruit and forestry) that could contribute to legacy use of organochlorine pesticides. Electricity is readily available via substations in many of the villages, but transformers are mainly of recent origin as electrification of rural areas only became a priority after 1994. However, many households still cook over open fires. Small-scale motor vehicle repair and artisanal brick making does occur. Malaria is controlled with DDT and other insecticides using IRS from Thohoyandou eastwards, up to the KNP border, protecting about 1.84 million people. Very little if any malaria control has ever been done towards the west because of little or no malaria. In the 2009/10 malaria transmission season (when the eggs were collected), about 52 000 kg of 75% water wettable DDT powder, and 11 600 kg of α-cypermethrin was applied as IRS in South Africa. IRS insecticides are applied indoors on walls and outdoors under rafters, at 2 g/m^2. More on malaria control in this region can be gleaned from Bornman et al. (2010) and Bouwman et al. (2012b).

2.2. Egg collection and preparation

The project was approved by the ethics committee of the North-West University (NWU) (NWU-00055-07-S3). Permits for collection of bird eggs were obtained from the Department of Economic Development, Environment and Tourism of the Limpopo Provincial Government.

A local bird guide and interpreters were employed to assist in locating known and potential breeding colonies, and to communicate with the local communities. Sampling of heron and egret eggs was done by scaling stable trees using rock climbing techniques with a double-belay system. A team of two experienced rock climbers (including IMV) was used. Reachable nests were inspected and sampled. Where possible and safe, and depending on the number of eggs in a nest, either one or two eggs were collected, leaving the rest behind.

The abundant sparrow nest within the roofs of thatched houses drew our attention. We collected all the eggs (normally 4–6) from selected nests. Southern Masked Weavers eggs were also collected at Nandoni Dam from nests overhanging the water. All eggs were wrapped in pre-cleaned foil and marked. The eggs were then frozen. It must be noted that the nests in roof thatch are in very close proximity to the sprayed inner surfaces—only centimetres away. Since eggs were collected from the outside, no additional contamination of the eggs via handling is foreseen. Care was also taken not to contaminate the egg contents with the eggshells that may have extraneous DDT residues on the outside when removing the egg contents.

All equipment used for sample preparation was rigorously pre-cleaned using soap and water, rinsed three times with double distilled water, and washed three times with 96% ethanol. Eggs were measured and carefully broken open in the absence of direct light to protect light-sensitive compounds. The sparrow eggs from each nest were pooled as the small volumes of the eggs precluded individual analysis. The contents were homogenised using an ultrasonic homogeniser such that as little foam as possible was formed. Samples were shipped frozen and received so in Norway. The analyses were done at the Laboratory of Environmental Toxicology at the Norwegian School of Veterinary Science (NVH) in Oslo—it is an accredited analytical laboratory (NS-EN ISO/IEC 17025 (TEST 137)). The sample preparation, extraction and clean-up procedures were done while protecting the samples from UV-light to avoid the degradation of brominated compounds.

2.3. Extraction and analyses

Procedures and quality control for extraction and analyses (Brevik, 1978) are described and referenced in Bouwman et al. (2012a). Because we suspected very low levels of brominated compounds, only pooled samples from each species and site were analysed. We prefer reporting and discussion concentrations based on wet mass (wm) rather than lipid mass (lm), as embryonic metabolism affects lipid content of the egg (Romanoff, 1932). Wet mass is also the basis for determining risk. However, we do present summarised lipid-based data (Table 1), and when comparing with other studies (Table 2). Mean relative recoveries were 110% for organochlorines, and 99% for BFRs. The data were not corrected for recoveries. Detection limits are provided in Table 1, and calculated as 3 × noise level, except BDE-183, -207, -208 and -209 that was mean of blank value+2 × standard deviation due to problems with blanks. Compounds below quantification limits in all eggs were α-HCH (at LOD=0.006 ng/g wm), PBEB, (pentabromoethylbenzene), DPTE (2,3-dibromopropyl-2,4,6-tribromophenyl ether), and HBB (hexabromobenzene) (all at LOD=0.01 ng/g wm). BDE-206 and HBCD were analysed for, but because of

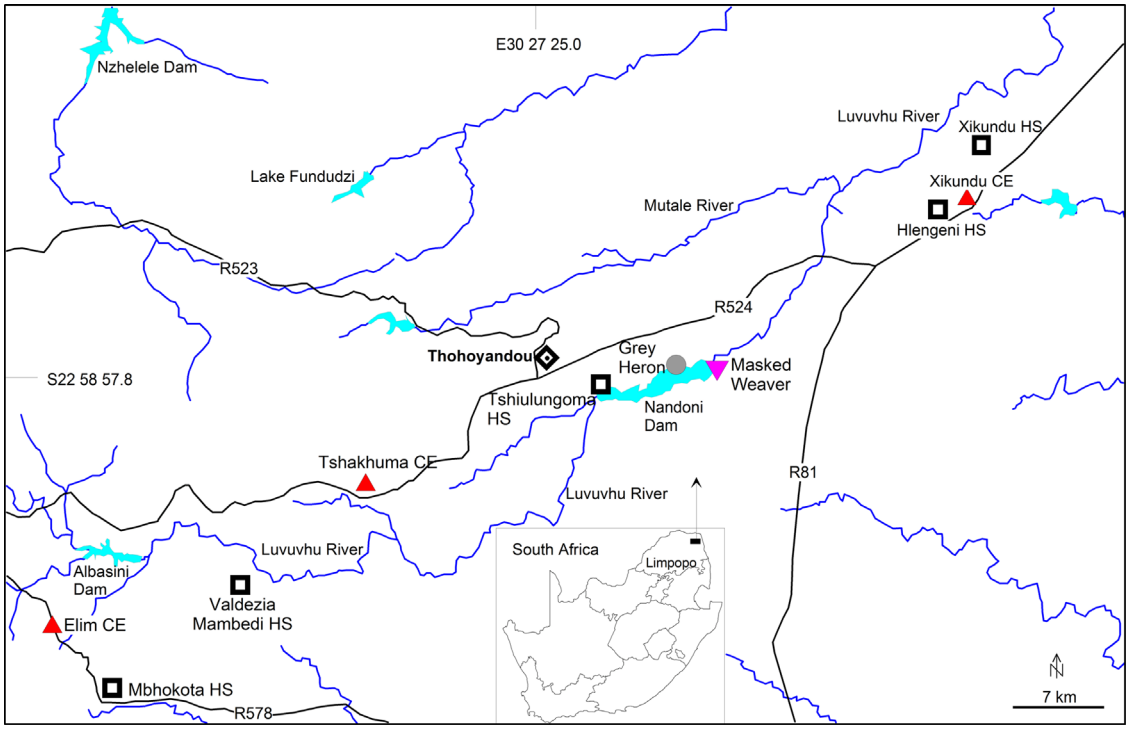

Fig. 1. Map of the region. The river flows west to east. HS—House Sparrow; CE—Cattle Egret.