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Integrating population connectivity into pollution assessment: Overwintering mixing reveals flame retardant contamination in breeding areas in a migratory raptor

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A B S T R A C T

Determining the exposure and magnitude at which various pollutants are differentially assimilated at the breeding and non-breeding grounds of migratory wildlife is challenging. Here, the possibility of applying the migratory connectivity framework to understanding contamination in birds is illustrated by considering flame retardants in inviable eggs of a migratory raptor, the black kite (*Milvus migrans*). The occurrence and concentration of legacy and emerging compounds in eggs from the southeastern peri-urban area of Madrid city, central Spain, were compared with those from Doñana National Park in southern Spain. A much higher occurrence and concentration of multiple polybrominated diphenyl ethers and Dechlorane 602 were found in Madrid than Doñana, but the opposite patterns were found for Dechlorane Plus. Individuals from these and other breeding areas in western Europe showed a strong intermixing pattern over widespread wintering areas in Africa, as assessed by ringing recoveries and movements tracked by satellite devices. This diffuse migratory connectivity reveals breeding areas as the main contamination grounds. High contamination burdens sequestered in eggs point to rapid assimilation of these compounds before laying, associated with important emission sources in Madrid, especially landfills of partially incinerated urban refuse, and other anthropogenic operations. Diet composition regarding aquatic *vs.* terrestrial prey, and bioaccumulation and biomagnification processes are suggested to explain differential assimilation of some compounds, especially Dechlorane Plus in Doñana, although a local emission source polluting this area cannot be ruled out. Insight from the migratory connectivity framework can help to disentangle large-scale patterns of contaminant uptake and refocus attention on key regions and potential causes of chemical hazards in declining migratory species and human populations.

1. Introduction

Due to their multiple physiological effects on health, toxicants represent a significant threat to birds, with obvious implications for the conservation of declining species (Helander et al., 2002; Fernie and Letcher, 2010; Eng et al., 2017). Birds usually assimilate toxicants through dietary and other sources, and females can pass some toxicants through egg deposition (Gauthier and Letcher, 2009; Guigueno and Fernie, 2017). Eggs are laid in short time periods across the life-cycle of individual females, generally once a year, but they can sequester pollutants assimilated in previous periods of variable length depending on the species involved (Newton et al., 1981; Morrissey et al., 2004; Custer et al., 2014) and individual traits, such as the age of first breeding in species with delayed maturation (Rowe, 2008). This makes eggs useful targets for the integrated assessment of contamination levels over time and space, but it also hinders the determination of the specific timing and locations of contamination uptake (Yates et al., 2010; Bourgeon et al., 2013), especially in species performing large-scale movements between breeding and non-breeding areas (Runge et al., 2014).

Variable loads of pollutants contained in eggs are often linked to pollution exposure in particular areas, often inferred from expert knowledge of localized sources of contamination (Bustnes et al., 2006; Miller et al., 2014). However, there is scarce information on the timing of pollutant uptake, their accumulation in body tissues, and the physiological and life history determinants of their deposition in eggs of most avian species. In particular, it is generally unknown whether

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specific toxicants or cocktails of pollutants are differentially assimilated in the breeding and non-breeding grounds of migratory species (Elliott et al., 2007; Yates et al., 2010; Leat et al., 2013). To estimate the contribution of contamination in these separate ranges, it is critical to know the migratory schedules and connectivity through the annual cycle, which is often challenging for highly mobile species that travel over different nations and across continents (Lambertucci et al., 2014; Runge et al., 2014).

Knowledge on the timing and conditions associated with the occurrence of specific compounds may provide insights on contaminating sources, especially when the occurrence of distinct toxicants shows contrasting patterns between population nuclei. Among toxicants included on the list of global elimination compounds under the Stockholm Convention, polybrominated diphenyl ethers (PBDEs) added as flame retardants (FRs) to plastics, textiles and other materials have been recorded in multiple matrices (Eljarrat and Barceló, 2011). PBDEs were typically produced at three different degrees of bromination: Penta-BDE, Octa-BDE, and Deca-BDE commercial mixtures. Due to PBDE presence in the environment and its proven toxicity, Penta- and Octa-BDE commercial mixtures have been banned in the European Union (EU) and in some states of the USA since 2004, and also banned in Canada since 2006. In 2009, they were designated as new persistent organic pollutants and the Stockholm Convention decided to add the commercial mixtures with four, five, six and seven bromines to Annex A, to end their production and use (UNEP, 2015). In regards to the Deca-BDE commercial mixture, their use was strictly prohibited in 2008, only in the EU: Deca- BDE can no longer be used in electronics and electrical applications, but a maximum concentration value of 0.1% of total PBDEs by mass in homogeneous materials will be tolerated (ECD, 2009; Kemmlein et al., 2009).

In response to the constantly updated regulations, the priorities of the FR industry have been redirected to emerging halogenated FRs (HFRs) including halogenated norbornenes such as Dechloranes (Dec). These compounds are still unregulated, even though they have been found in several environmental and biological matrices, demonstrating their bioaccumulation capacity (Covaci et al., 2011; Feo et al., 2012). Previous studies of migratory birds showed contrasting effects of wintering grounds on pollutant burdens in eggs (Elliott et al., 2007; Yates et al., 2010). Among FRs, most research has focused on PBDEs because of their widespread commercial use, their persistence, ubiquity, ability to bioaccumulate and biomagnify, and their toxicological effects (Covaci et al., 2011; Eljarrat and Barceló, 2011). However, there is scarce information on the occurrence and levels of these pollutants in raptor eggs (Chen and Hale, 2010), despite the profound effects they may exert on reproduction (Guigueno and Fernie, 2017).

In this study, we investigated the occurrence of FRs in inviable eggs of a migratory raptor, the black kite (*Milvus migrans*), in the southeastern peri-urban area of Madrid city, central Spain. Several aspects make this species especially suitable for contaminant monitoring. First, kites are opportunistic predators and facultative scavengers that breed in a variety of open habitats, often near water, and frequently within or close to human settlements and rubbish dumps (Blanco, 1994; Kumar et al., 2014). Therefore, they are especially sensitive to contamination from toxic residues from agriculture, industry, urban settlements and landfills. Second, as long-lived top-predators and scavengers, they tend to accumulate persistent contaminants in adipose tissues and visceral organs as a result of bioaccumulation at the top of food webs and long-lasting exposure to air-borne contaminants (Merino et al., 2002; Blanco et al., 2003; Jiménez et al., 2004). Third, because kites are income breeders that can reproduce for the first time up to the age of seven years (Sergio et al., 2011), females may accumulate heavy burdens of contaminants, which are transferred to eggs, leading to potential breeding failure due to embryonic mortality and infertility (Merino et al., 2002; Blanco et al., 2003; Jiménez et al., 2004).

The study area has been highlighted in previous studies as highly contaminated with multiple pollutants (Jiménez et al., 1996, 2000). Estimates from this area were compared to those for another black kite population from a less anthropogenically altered area, Doñana National Park in southern Spain (Barón et al., 2014a). Given that the study species is a long-distance migrant, it is unknown whether the levels of particular pollutants detected in eggs of particular populations correspond to contamination mainly occurring in the European breeding areas or in the African wintering grounds. To unravel this, we evaluated whether distinct breeding populations in Europe share the wintering areas (overwintering mixing) or segregate spatially in these areas (overwintering segregation). Through this evaluation, we illustrate the possibility of applying the migratory connectivity framework to the understanding of uptake routes and degree of contamination in birds. Different breeding populations often overlap in their wintering quarters in a process of temporal and local intermixing leading to diffuse connectivity (Webster et al., 2002). In this case, the main source of contamination should be located at the breeding grounds when eggs show clear between-population differences in pollutant burdens. Alternatively, when individuals from different breeding nuclei segregate in the wintering quarters (strong connectivity), it is more difficult to assign contamination load in eggs to exposure in the wintering or breeding areas.

2. Material and methods

2.1. Model species, study area and field procedures

The black kite (hereafter “kite”) is a widespread medium-size raptor distributed throughout the temperate and tropical parts of the Old World and Australasia. In the Western Palearctic, this species is mainly present during the breeding season (March–September, Blanco, 1994). Western and southern European breeding populations cross the Gibraltar and Messina straits and undertake a long-distance migration through the western flyway to the wintering African quarters in the Sahel and Gulf of Guinea, while central and eastern European populations cross the Bosphorus through the eastern flyway to overwinter in eastern Sahel and the southern regions of Africa (Zalles and Bildstein, 2000).

We monitored kites in the Regional Park of southeastern Madrid, central Spain, an area devoted to irrigated, intensive agriculture (mainly maize and vegetables) and gravel extraction along the Manzanares and Jarama rivers. The area is polluted with organochlorines, PCBs, dioxins, heavy metals and pharmaceuticals from agricultural, industrial, urban and incineration activities (Fernández et al., 2000; Jiménez et al., 2000; Valcárcel et al., 2011; Escobar-Arnanz et al., 2018).

All breeding pairs of kites in the area were surveyed and their nests located and monitored to determine laying and breeding output. Briefly, laying date was determined from nests found during laying by backdating, using a laying interval of two days, and by combining observational data of the beginning of incubation with the estimated age of the chicks at banding. When chicks were feathered and before fledging, they were banded, weighed and measured for several morphological traits. Nests were monitored until nestlings fledged to determine breeding success and the number of fledglings (see details in Blanco et al., 2004, 2006). In this area, kites nest in riparian forests and feed mostly on European rabbits, *Oryctolagus cuniculus* (Blanco, 1997). Males usually arrive and settle in territories before females and clutches are laid by mid-April, about one month after the arrival of females in the territories. When chicks were feathered and before fledging (about 30–45 days of age from hatching), the nests were accessed to band the nestlings and collect failed eggs. Overall, we collected 28 eggs between 2007 and 2016. All failed eggs had remained in the nest for a similar amount of time, thus precluding biases due to differences in the evaporation of egg water content. All the eggs were frozen and sent to the laboratory in individual and protected containers. Data on reproductive parameters of black kites in the study areas can be found elsewhere