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Review

Towards harmonisation of chemical monitoring using avian apex predators: Identification of key species for pan-European biomonitoring

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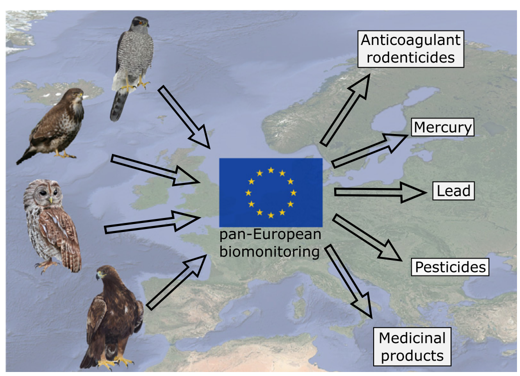
HIGHLIGHTS

- We identified key raptor and owl species for pan-European monitoring of pollutants.
- Selection was primarily on key ecological traits and distribution.
- Our focus was on Pb, Hg, rodenticides, pesticides and veterinary medicinal products.
- Common buzzard and tawny owl were the most suitable pan-European biomonitors.

GRAPHICAL ABSTRACT



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ABSTRACT

Biomonitoring in raptors can be used to study long-term and large-scale changes in environmental pollution. In Europe, such monitoring is needed to assess environmental risks and outcomes of chemicals regulation, which is harmonised across the European Union. To be effective, the most appropriate sentinels need to be monitored. Our aim was to identify which European raptor species are the likely most appropriate biomonitors when pollutant quantification is based on analysing tissues. Our current study was restricted to terrestrial exposure pathways and considered four priority pollutant groups: toxic metals (lead and mercury), anticoagulant rodenticides, pesticides and medicinal products. We evaluated information on the distribution and key ecological traits (food web, foraging trait, diet, preferred habitat, and migratory behaviour) of European raptors to identify the most appropriate sentinel species. Common buzzard (*Buteo buteo*) and/or tawny owl (*Strix aluco*) proved the most suitable candidates for many of the pollutants considered. Moreover, they are abundant in Europe, enhancing the likelihood that samples can be collected. However, other species may be better sentinels for certain pollutants, such as the golden eagle (*Aquila chrysaetos*) for lead, the northern goshawk (*Accipiter gentilis*) for mercury across areas including Northern Europe, and vultures (where they occur in Europe) are likely best suited for monitoring non-steroidal anti-inflammatory drugs (NSAIDs). Overall, however, we argue the selection of candidate species

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for widescale monitoring of a range of pollutants can be reduced to very few raptor species. We recommend that the common buzzard and tawny owl should be the initial focus of any pan-European raptor monitoring. The lack of previous widespread monitoring using these species suggests that their utility as sentinels for environmnetal pollution has not been widely recognised. Finally, although the current study focussed on Europe, our trait-based approach for identifying raptor biomonitors can be applied to other continents and contaminants.

Contents

1. Birds of prey as sentinels for pollution monitoring

The monitoring of environmental pollutants in raptors has a long history (Cade et al., 1971; Helander et al., 1982; Ratcliffe, 1967). Such monitoring was often initiated to understand the risks from pollutants to individual species of high conservation value, but it is now recognised that it also can provide insights into wider ecological health and a warning of potential human exposure and effects on health (García-Fernández et al., 2020). There are a number of characteristics that make predatory birds particularly suitable as sentinels, especially for compounds that bioaccumulate or biomagnify through food webs. These include foraging through both terrestrial and aquatic food webs, occupation of high trophic position (typically raptors are apex predators), a long history of ecotoxicological research and associated understanding of contamination in various species, and, where appropriate, the potential to obtain non-destructive samples (feathers, carcasses from accidents, deserted eggs, blood) for analysis (Espín et al., 2016; Gómez-Ramírez et al., 2014).

Monitoring in raptors can reveal spatio-temporal trends in environmental contaminant concentrations (Gómez-Ramírez et al., 2019; López-Perea and Mateo, 2018; Walker et al., 2012). It can therefore be a key tool for evaluating the outcomes of regulation and other mitigation measures designed to reduce environmental contamination over large spatial scales (García-Fernández, 2020; Shore and Taggart, 2019). Monitoring at national or smaller spatial scales across Europe has involved the use of a variety of species (Gómez-Ramírez et al., 2014) and sample types (Espín et al., 2016). However, chemical regulation in much of Europe is now harmonised and delivered through European Union (EU) directives and regulations, such as the Biocidal Product Regulation (EU 528/2012), regulation on Plant Protection Products (EC 107/2009) and REACH - Registration, Evaluation, Authorisation and Restriction of Chemicals (EC 1907/2006). Recently, the European Parliament and the Council on Veterinary Medicinal Products (VMPs) also repealed the previous Directive 2001/82/EC and replaced it with a stronger/harmonised regulation (García-Fernández, 2020). Therefore, monitoring to detect the outcomes of legislation applied to large spatial scales (such as EU legislation) needs to be at the same scale. This imperative has led to initiatives to develop pan-European monitoring capability, such as EURAPMON; www.eurapmon.net and its follow-up programme, the European Raptor Biomonitoring Facility ERBFacility; www.erbfacility.eu (Movalli et al., 2019). However, a key challenge for large-scale monitoring is to determine which species, or guild of species, are likely to be the most suitable sentinels for monitoring contaminants and how species selection may vary depending upon the contaminants of interest. This is a critical knowledge gap.

The current study aimed to address this gap and evaluate the relative merits and disadvantages of different species for harmonised biomonitoring within and across large-spatial scales such as Europe. We shortlisted candidate species based on their European distribution and on ecological traits relevant to exposure to priority environmental pollutants. Our initial analysis indicated that the distribution across Europe of raptors that utilise aquatic food-webs was largely limited and it is arguable that non-raptor and non-avian species, such as the Eurasian otter (*Lutra lutra*), gull species and pinnipeds, may prove more suitable for large-scale biomonitoring of pollutant transfer through freshwater and marine systems. Therefore, the present work focuses on terrestrial exposures to priority pollutants. Our work builds on previous research into monitoring schemes that were, or currently are, operative within Europe (Gómez-Ramírez et al., 2014) and the practicalities of what sample types are suitable for pollution studies (Espín et al., 2016).

2. Prioritised environmental pollutants

We focussed on addressing which species may be most suitable for monitoring a sub-set of priority compounds. The choice of compounds was agreed at a European workshop of 30 experts that was hosted by the ERBFacility in February 2019. Pollutants were selected on the basis that they remain a current environmental risk across Europe, particularly to vertebrate wildlife, and are typically also subject to regulation. Pollutants groups were prioritised using a ranking exercise that was conducted independently by three breakout groups and the average rankings calculated (Table SI-1). The selected priority pollutants were two toxic metals (lead (Pb) and mercury (Hg)), anticoagulant rodenticides (ARs), pesticides as a general group and medicinal products (MPs), and in particular veterinary medicinal products (VMPs).

Lead is a toxic non-essential trace metal that occurs naturally in parts of the earth crust, but anthropogenic uses such as mining and metal production have resulted in a ubiquitous environmental distribution (Abadin et al., 2007). Lead has been recently identified as a substance of very high concern by the European Chemicals Agency (ECHA) due to its reproductive toxicity and is therefore subject to authorisation within REACH (ECHA, 2018). Certain uses (such as in gasoline) have already been regulated or banned. However, Pb is still frequently used in hunting ammunition and fishing weights (Stroud, 2015), although the use of Pb shot and ammunition for hunting varies between EU Member States depending on their national/regional legislation (Mateo and Kanstrup, 2019). It is the dietary ingestion of Pb shot and ammunition fragments that poses the most serious threat for predators (Krone, 2018; Nadjafzadeh et al., 2015; Pain et al., 2019). Species that are exclusively scavengers (obligate scavengers) as well as species that scavenge and actively hunt (facultative scavenger) are at particular risk because they frequently feed on game mammals and waterfowl (García-Fernández et al., 2005; Krone et al., 2009; Mateo, 2009). For example, Pb intoxication has been identified as an important mortality factor for vultures and facultative scavengers across Europe (Berny et al., 2015; Helander et al., 2009; Krone et al., 2009). However, foraging on gunshot-injured but still living mammals and waterfowl can also result in significant exposure risk for non-scavengers (Gil-Sánchez et al., 2018; Mateo et al., 1999).

Mercury is also a highly toxic non-essential trace metal. It is naturally emitted through volcanic activities, sea salt spray and soil particles (Nriagu, 1989) but is released in greater quantities by industrial activities such as coal-combustion, refuse incineration and metal production (Amos et al., 2013; Nriagu and Pacyna, 1988). Due to its high toxicity, Hg is currently included within Regulation (EU) 2017/852, which regulates the import and use of Hg containing products. In the atmosphere, Hg occurs mainly in its elemental form (Hg^0), whereas it is predominantly in its organic form methylmercury (MeHg), in soil, sediments and surface waters. Mercury can biomagnify in both aquatic and terrestrial food webs (Cristol et al., 2008; Douglas et al., 2012; Lavoie et al., 2013), and elevated concentrations are accumulated in birds of prey and other predators (Badry et al., 2019; Sun et al., 2019). Biomagnification with increasing trophic level means that Hg can reach toxic concentrations in apex predators (Lavoie et al., 2013). In terrestrial environments, raptors can accumulate sufficient Hg such that reproduction is impaired and behavioural abnormalities are manifest (Burger and Gochfeld, 1997; Whitney and Cristol, 2018).

Anticoagulant rodenticides are widely used biocides commonly applied in agricultural and urban settings to control populations of rats, mice and, in some countries, voles (Geduhn et al., 2014; López-Perea and Mateo, 2018). Their use as biocides is regulated under the EU Biocides Directive but ARs are also used (and regulated for) as Plant Protection Products (PPPs) in some countries (e.g. bromadiolone in Italy, France, Netherlands, Romania; Regnery et al., 2019). Eight ARs are currently registered for use in Europe. These are the older first generation ARs (FGARs) - warfarin, coumatetralyl and chlorophacinone – and five second generation ARs (SGARs): difenacoum, bromadiolone, brodifacoum, flocoumafen and difethialone (Regnery et al., 2019). SGARs were developed in the 1970s due to increasing resistance of rodents against FGARs (Buckle et al., 1994; Eason et al., 2002) but they all broadly have a common mode of action, which is inactivation of the vitamin K epoxide reductase in hepatocytes and a consequent failure to synthesize clotting factors like prothrombin (Rattner et al., 2014). Because the clotting system is highly conserved in evolutionary terms, ARs affect all vertebrates.

SGARs are formulated mainly as coated wheat baits, wax baits and as gels and may be deployed in bait boxes, in burrows or may be buried underground in rodent galleries; application can be made throughout the year or targeted when rodent pests are most abundant (López-Perea and Mateo, 2018). Non-target small mammal species also take bait and individuals within 15 m of bait stations have been shown to accumulate the highest SGAR residues, although individuals can range widely in agricultural landscapes (Geduhn et al., 2014; Tosh et al., 2012). Predators are thought to typically be exposed secondarily to ARs, mainly as a result of preying on rodents and/or scavenging (Elliott et al., 2014; López-Perea and Mateo, 2018).

Pesticides are a diverse group of chemicals that are commonly classed as PPPs when their insecticidal, herbicidal or fungicidal properties are used to protect agricultural crops. However, the term pesticide can also be used to refer to the same active ingredient when it is used for other purposes, such as biocide to treat ectoparasites on livestock. The acute mortality caused by legacy plant protection products, such as the organochlorine insecticidal seed dressings dieldrin, in combination with poor reproduction caused by dichlorodiphenyltrichloroethane (DDT)-mediated eggshell thinning, was one of the first examples of pesticides causing population declines in raptors and other species (Newton, 1986; Ratcliffe, 1967). Such pesticides have been widely banned at national and European levels because of their toxic effects on humans as well as wildlife, but significant residues of legacy organochlorines are still detectable in raptors today (Gómez-Ramírez et al., 2019). Pesticides used in agriculture to protect crops are regulated in the EU as Plant Protection Products (EC 1107/2009), and those sold as biocides are regulated as biocidal products (EU 528/2012). A risk assessment represents the first step of the authorization of pesticides in the EU and requires that a predicted environmental exposure concentration is below a concentration that is considered to cause an effect in non-target organisms (Schäfer et al., 2019). However, empirical data on bioaccumulation in wildlife systems and on exposure of apex predators are scarce and it is argued that biomonitoring could contribute valuable information on the accumulation of pesticides within food webs (Movalli et al., 2019).

Medicinal products are widespread environmental pollutants that have been associated with threats to non-target wildlife such as raptors (Shore et al., 2014). Within Europe, medicinal products are classified and regulated as human medicinal products (HMPs) (2001/83/EC), as VMPs (Regulation (EU) 2019/6) or both (aus der Beek et al., 2016; García-Fernández, 2020). Environmental risks have been associated with hormones, anti-parasitics, antibiotics and anti-inflammatories used as HMPs and VMPs and with analgesics and antidepressants used as HMPs (aus der Beek et al., 2016; Mateo et al., 2015). Medicinal products can enter the environment via landfills, livestock production and through application of sewage sludge as fertilizer (Arnold et al., 2014; Shore et al., 2014). Potential wildlife exposure pathways in wildlife include intake via diet and contaminated water and inhalation of dust in areas of intensive animal feeding operations (Shore et al., 2014). Even though the environmental half-lives of medicinal products are generally lower than those of many persistent organic pollutants (POPs), environmental emissions can exceed removal rates and so they are considered pseudo-persistent pollutants (Daughton and Ternes, 1999; Lazarus et al., 2015). Some medicinal products are predicted to accumulate along aquatic food chains (Connors et al., 2013; Lazarus et al., 2015), thereby potentially reaching toxic concentrations. The environmental life cycle for most medicinal products as well as their accumulation and metabolism in non-target wildlife species remains poorly understood (Shore et al., 2014), but these products can have devastating impacts, as demonstrated by the impact of diclofenac on *Gyps* vultures (Oaks et al., 2004).

3. Methods of selection of candidate species based on ecological traits

The 2019 ERBFacility workshop identified a putative "long-list" of candidate species (Table SI-2) that were considered suitable European species for monitoring the priority pollutants that are the focus of the present work.

The ERBFacility workshop also discussed what type of monitoring would be feasible if a pan-European monitoring programme was to be established. The consensus was that, while active monitoring, for example sampling nestling blood, might offer a structured monitoring programme, it would be difficult to develop a sustainable programme with adequate geographical coverage. This is because such monitoring requires ethical permits, trained volunteer or professional personnel and is expensive. Although shed feathers or failed eggs could be collected from nests instead of blood, this would not overcome the likely geographical patchiness of sampling from nest sites and such samples, particularly feathers, are of limited use toxicologically. Espín et al. (2016) discussed in detail the advantages and disadvantages of different sample matrixes for contaminant monitoring in raptors and concluded that liver [and blood] were the most effective matrices for most analytes. Liver samples can be obtained from the carcasses of raptors found dead. Current monitoring schemes have demonstrated the feasibility of using interested members of the public to report and collect the carcasses of raptors that they find (Gómez-Ramírez et al., 2014; Jager et al., 1996; Naccari et al., 2009; Walker et al., 2008a); such collections can be across a broad geographical scale. The selection of candidate species for biomonitoring for the present study was therefore predicated on the assumption that pollutant characterisation would involve analysis of tissue samples obtained from the carcasses of birds that died from a variety of causes but particularly traffic accidents, other trauma and starvation (Jager et al., 1996; Naccari et al., 2009; Walker et al., 2008a).

After the conclusion of the ERBF workshop, we reduced the species long-list using an objective logical framework that first considered the geographical distribution of the species and then evaluated whether their trait characteristics were suitable for biomonitoring. Our first category, widespread distribution within Europe, was deemed the most important selection criterion given the aim for any biomonitoring was to track changes across Europe (Table SI-3). We considered Europe (here defined as EU countries together with Norway, Switzerland, United Kingdom (UK) and Iceland; Fig. 1) to consist of four regions (eastern, northern, southern and western Europe) based on the United Nations Geoscheme (United Nations Statistics Division, 1999). We classed a species as widely distributed if it was present in three or more countries in at least three of those four regions. Species distributions were taken from BirdLife International (2019). The requirement for widespread distribution reduced the species "long-list" down to 19 species that feed mainly on terrestrial species (Table SI-3). None of the raptors feeding on aquatic prey (Table SI-4) nor the vultures (Table SI-5) met the criteria for widespread distribution. The present work therefore subsequently focussed only on terrestrial exposure to our selected priority pollutants.

We then considered the main traits likely to influence exposure to our priority compounds; these were predominant feeding trait (scavenging, active hunting), diet, and type of habitat utilised (Table SI-3). Although we focused on terrestrial species, we also considered which was the predominant food web (terrestrial, freshwater, marine) when considering species that were mixed feeders as contaminant levels in birds of prey can be affected by their respective food webs (Eulaers et al., 2011; Jaspers et al., 2006). We extensively searched existing published information to describe the

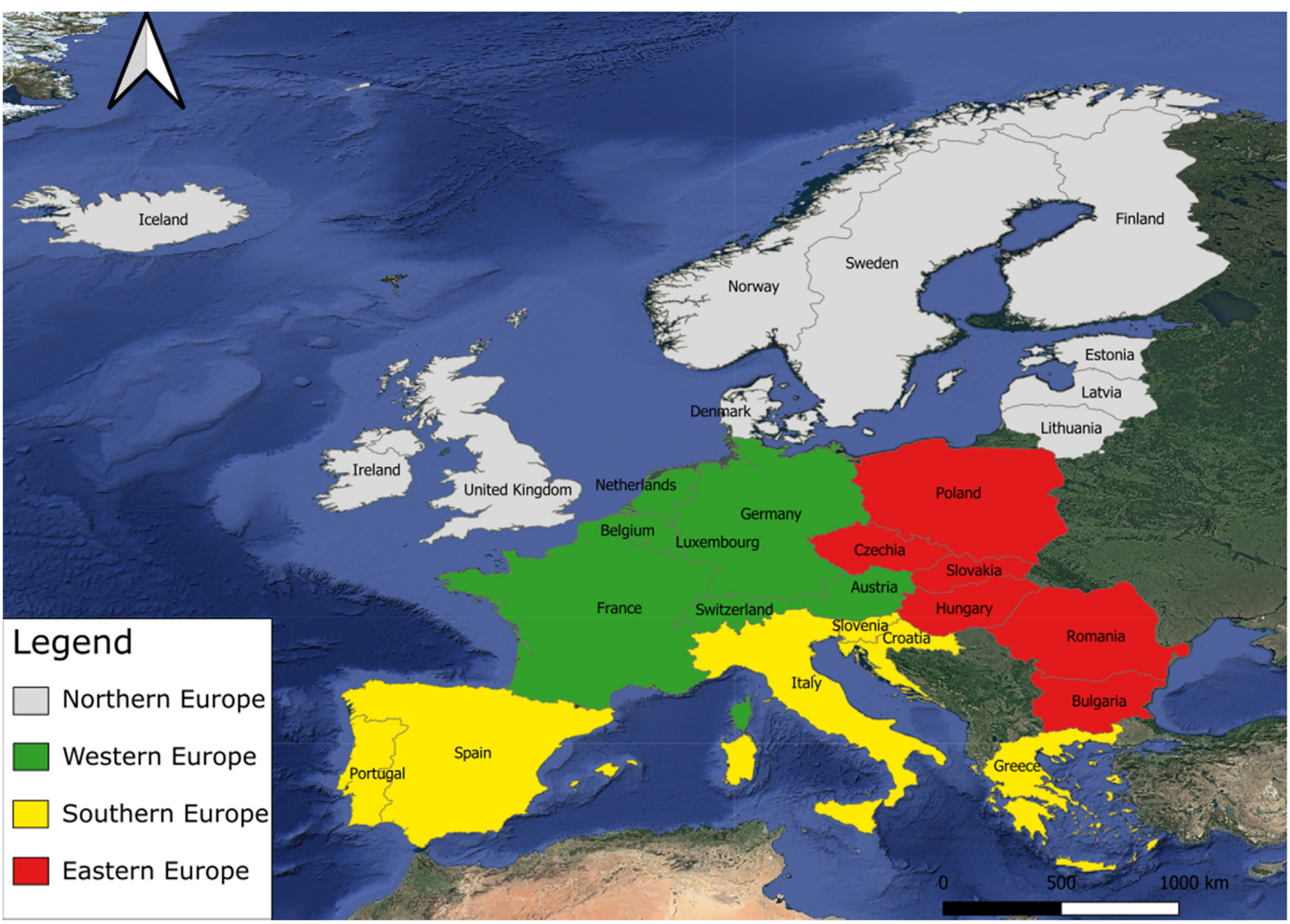

Fig. 1. Main regions of Europe based on the United Nations Geoscheme (United Nations Statistics Division, 1999). Considered countries include European Union countries together with Norway, Switzerland, UK and Iceland.

characteristics of each trait for every raptor in the reduced long-list. These are given in Table SI-3.

As pollutant characterisation was assumed to be based on tissue analysis, we included migration as a key trait. This was because exposure to and assimilation of a contaminant in a tissue could occur at location[s] distant from where the bird later died and was collected for analysis. This is particularly salient for contaminants that are only slowly metabolised in tissues, and potentially for contaminants accumulated in fat depots; body-lipids are remobilized during migration which can elevate liver concentrations of lipophilic compounds (Henriksen et al., 1996). Although migration may not affect residue magnitude for all contaminant classes/matrices (Elliott et al., 2007; Leat et al., 2019) and previous contaminant studies have involved migratory raptors, the origin of contaminant exposure can be difficult to interpret (Goutner et al., 2011; Lavoie et al., 2010). This adds uncertainty, compared to the use of non-migrants, when the aim is to use spatial and temporal variation in raptor contamination to inform chemicals management. This uncertainty may be particularly acute when using long-distant migrants as exposure may occur outside the jurisdiction of regulatory authorities, even when jurisdictions are continental in size. We categorised raptors as resident, partial-migrants and long-distance migrants (Table SI-3).

We classified different characteristics of each trait with respect to their suitability for pan-European monitoring of the compound of interest. Traits characteristics were categorised as advantageous (AD), limiting (LI) or excluding (EX). Advantageous characteristics were those likely to result in, and potentially maximise, exposure to the pollutant of interest. Residency and widespread distribution were also classified as advantageous. Limiting criteria were trait characteristics likely to lead to pollutant uptake through routes not considered the most important exposure pathway. Traits characteristics, such as partial migration, that somewhat compromised the spatial integrity of biomonitoring were also considered limiting, as was the absence of a species in three or more countries in one of the main regions of Europe. Exclusion criteria for pan-European biomonitoring were traits characteristics that were likely to markedly limit or prevent exposure. Long-term migration was also considered an excluding factor. We concluded that the species with the highest number of advantageous traits and no exclusion criteria were the most suitable for pan-European monitoring of the specific contaminant of interest. The trait categorisations for Pb, Hg, ARs, pesticides and MPs are given in Tables SI-7, SI-9, SI-12, SI-14 and SI-16, respectively.

We then examined how the trait characteristics described for each raptor species (Table SI-3) corresponded against our defined AD, LI and EXC criteria. In this way, we assigned an AD, LI or EXC category to each trait for each raptor. We then used this information to compile a short list of candidate species for each priority pollutant. Species were only included in these short-list on the basis that they had no excluding traits. The species short list for each priority pollutant, and their categorised trait characteristics, are given in Tables 1-5. There was typically more than one species in the short-list and the relative merits and demerits of short-listed species, in terms of their use as biomonitors, is the focal point of discussion in the current paper (Section 4). Where possible, this discussion reduced the short-list further to just one or two species that were argued to be the most suitable for biomonitoring at a pan-European scale. This included taking into account species abundance as a secondary or contextual criterion. The number of raptor carcasses found and submitted for contaminant analysis tends to be positively correlated with relative abundance (Newton et al., 1999).

After one or two species were identified as the most suitable candidates for pan-European monitoring, we conducted a web-based literature research, using specific key words and Boolean operators (Table SI-6), to ascertain whether it had been used for monitoring the contaminant of interest. Evidence of such monitoring provides some proof that generation of contaminant data in that species is actually possible.

4. Candidate species for biomonitoring of prioritised environmental pollutants within Europe

4.1. Trace metals

4.1.1. Lead (Pb)

After widespread geographical distribution, feeding ecology was considered to be the critical trait for selecting a sentinel for pan-European Pb monitoring. This was because predators and scavengers that feed on game species generally accumulate the highest Pb burdens and suffer incidents of Pb-related mortality (García-Fernández et al., 2005; Krone, 2018; Mateo et al., 2003). Scavengers and active predators of game species were therefore considered candidate species (Table 1). Of those, species that undertake partial migration were deemed less suitable for monitoring. This was because ability to examine spatial variation in exposure is likely to be important for Pb as regulations on hunting and use of Pb shot varies between countries and regions within Europe (Mateo and Kanstrup, 2019). Hence, the use of partial migrants as well as species feeding on migratory prey was considered limiting due to the uncertainty as to whether accumulated residues reflected local or pre-migration exposure. Habitat was considered a less important trait for selecting candidate species since foraging on game and waterfowl occurs across a broad range of different habitats (Table SI-7). By applying the aforementioned criteria, we compiled a short-list of just two candidate species, the common buzzard (*Buteo buteo*) and the golden eagle (*Aquila chrysaetos*) (Table 1).

The common buzzard is widely distributed across Europe, although it is a partial migrant in northern areas, as birds migrate to avoid unfavourable weather conditions (BirdLife International, 2019; Holte et al., 2017). Restricting sampling to birds found dead in the breeding season would largely avoid exposure biases resulting from migration as Pb tissue half-lives are relatively short (1–-3 months; Krone, 2018) and tissue residues in the breeding season can likely reflect exposure at that time. However, restricting sampling in this way might induce a temporal bias if exposure is maximal during the hunting season but this does not coincide with the buzzard breeding season. Furthermore, the common buzzard predominantly forages on non-game species, such as rodents, when such prey is highly abundant (Table SI-3 and references therein). This is likely to limit potential exposure to Pb-shot in injured prey and it is notable that liver Pb concentrations in common buzzard are generally lower than those in species, such as golden eagles, that are thought to forage more frequently and consistently on game species (Table SI-8). Nevertheless, the widespread distribution of common buzzards, together with their relative abundance (and associated high likelihood of carcass availability) are favourable characteristics and they have been used for measuring Pb contamination previously (Jager et al., 1996; Naccari et al., 2009; Walker et al., 2008a).

Golden eagles forage predominantly on terrestrial prey, mainly medium-sized mammals, including game species (Table SI-3). They scavenge carrion in the winter (Halley and Gjershaug, 1998) which makes them highly susceptible to Pb exposure and toxicosis (Ecke et al., 2017; Madry et al., 2015). Golden eagles are also non-migratory and territorial, which enhances their suitability for detecting regional differences in Pb exposure, although there can be long-range dispersal for sub-adults in Scandinavia and Estonia (Nebel et al., 2019). Golden eagles have been used previously for Pb monitoring studies (Ecke et al., 2017; Madry et al., 2015; Mateo et al., 2003) and have been widely used as a sentinel of environmental pollutants generally within Europe (Gómez-Ramírez et al., 2014), indicating that sampling of this species is feasible. However, golden eagles are not evenly distributed within Europe, mainly as a result of human persecution (BirdLife International, 2019; Watson and Whitfield, 2002), and are restricted to remote and wilderness habitats like montane/alpine regions in western Europe and forest landscapes

Table 1
Key traits of shortlisted candidate species for pan-European monitoring of Pb. A complete list of traits and species with associated references can be found in Table SI-3 and the assessment of the criteria as suitable for Pb monitoring is indicated in Table SI-7. Overall suitability is indicated for each criterion except for distribution where individual suitability is given in the superscript of each main region. AD = advantageous criterion, LI = limiting criterion for pan-European Pb monitoring.

Species	Distribution	Food web	Feeding trait	Diet	Migration
Common buzzard (*Buteo buteo*)	• Eastern Europe**AD** • Northern Europe**AD** (except Iceland) • Southern Europe**AD** • Western Europe**AD**	• Terrestrial ➔ **AD**	• Active hunter • Facultative scavenger ➔ **AD**	• Mainly small mammals • Insects • Birds • Reptiles ➔ **LI**	• Partial migration in autumn and winter to southern Europe (depending on weather conditions) ➔ **LI**
Golden eagle (*Aquila chrysaetos*)	• Eastern Europe**AD** • Northern Europe**AD** • Southern Europe**AD** • Western Europe**AD** (only alpine)	• Mainly terrestrial ➔ **AD**	• Active hunter • Facultative scavenger (enhanced during autumn/-winter) • **AD**	• Mainly medium-sized (game-) mammals • Livestock and large game carcasses ➔ **AD**	• Resident (but sub-adults might show dispersal in Northern Europe) ➔ **AD**

in north-east Europe. This limits their suitability for pan-European monitoring.

Although primarily a species that feeds through aquatic food webs, the white-tailed sea eagle (*Haliaeetus albicilla*) also preys on and scavenges game species (Table SI-4). It has been widely used in ecotoxicological studies across Europe (Gómez-Ramírez et al., 2014) and, like golden eagles, suffer from Pb intoxication through ingestion of Pb ammunition in game species (Helander et al., 2009; Krone et al., 2009; Nadjafzadeh et al., 2013). White-tailed sea eagles are mainly distributed in northern and eastern Europe, but are absent in large parts of Europe (BirdLife International, 2019). Thus, they did not meet our selection criteria for widespread distribution and were not included per se in our candidate short-list (Table 1). However, it could perhaps be used in combination with the golden eagle. This would have the benefit of increasing likely sample availability in areas where golden eagles are absent in western (Germany and non-alpine habitats in Austria), eastern (Czech Republic and parts in Poland and Hungary) and northern (Iceland) Europe (BirdLife International, 2019). However, neither species is present in southern parts of the UK, Ireland, Benelux and non-montane regions of France (BirdLife International, 2019).

One difficulty in using a combined golden eagle/white-tailed sea eagle approach for monitoring Pb is that exposure and accumulation is not necessarily directly comparable across the two species. White-tailed sea eagles are mixed food web feeders, predominantly forage on fish, and compared with golden eagles, take more avian game such as waterfowl (Tables SI-3 and SI-4). Liver Pb concentrations were found to be lower in white-tailed eagles than golden eagles from the same area in Norway (Table SI-8). Such inter-species differences in exposure might be minimised by only sampling those individuals that die in winter, when both species frequently scavenge game animals (Halley and Gjershaug, 1998; Nadjafzadeh et al., 2016). In addition, stable isotope signatures such as $\delta^{13}C$ and $\delta^{34}S$ which can be used to determine the likely habitat (aquatic vs. terrestrial) from which prey are taken (Eulaers et al., 2014; Kelly, 2000), could be used to screen samples so that only individuals feeding predominantly on terrestrial prey were included in any monitoring programme.

In summary, pan-European monitoring for Pb using raptors is likely best served using either the common buzzard or the golden eagle (alone or in combination with the white-tailed sea eagle). Use of either species has advantages and disadvantages that need to be weighed against the primary aims of the monitoring programme. For instance, use of the common buzzard may be most suitable where the primary aim is to track temporal changes in Pb contamination at a European scale. The high abundance of this species and its widespread distribution throughout Europe would help ensure the availability of adequate samples. However, use of the golden eagle would perhaps be better where the aim is to identify spatial differences in exposure or identify the likelihood of toxic effects – the foraging behaviour, territoriality and accumulation of high residues in golden eagles are all beneficial traits for such monitoring.

4.1.2. Mercury (Hg)

The main exposure route to Hg for vertebrate birds and mammals in aquatic and terrestrial food webs is dietary exposure (Kidd et al., 2012). We focused on the terrestrial exposure of Hg within this analysis and selected only species that predominantly feed on terrestrial food webs. We considered even partial migration and a preference for natural/montane habitats as exclusion criteria (Table SI-9). This was to ensure that monitoring could identify local anthropogenic emissions within countries, which can elevate Hg burdens in raptors (Badry et al., 2019). Since Hg has shown to biomagnify in food webs, correction of trophic level using $\delta^{15}N$ (Jardine et al., 2006; Kelly, 2000) may be needed to company residue analysis so as to untangle the effects on exposure of intra-species differences in foraging. By coupling these criteria to those for distribution and applying then to the species listed in Table SI-3, we compiled a candidate species shortlist of one raptor and four owl species: northern goshawk (*Accipiter gentilis*), tawny owl (*Strix aluco*), Eurasian eagle owl (*Bubo bubo*), barn owl (*Tyto alba*) and little owl (*Athene noctua*) (Table 2). Each of these species has traits that impact their suitability for pan-European monitoring of Hg in the terrestrial environment.

The northern goshawk forages mainly on avian prey, including other raptors, and on small mammals (Table SI-3). It generally favours forest as breeding habitat but hunts in farmland and has also started to breed in urban areas (Table SI-3). Northern goshawks are generally considered resident but some individuals, such as juveniles in Fennoscandia, disperse (Table SI-3). Nevertheless, due to their widespread distribution, sedentary behaviour and well-known ecology, northern goshawks are considered by others as suitable sentinels of environmental pollution in terrestrial ecosystems within Europe (Dolan et al., 2017; Eulaers et al., 2013; Martínez et al., 2012).

The tawny owl mainly forages on small mammals, in particular small rodents, as well as on birds and hunts over a wide-range of habitats including farmland, forest patches and urban areas (Table SI-3). They are widely distributed within Europe although absent in northern parts of Fennoscandia and Iceland (BirdLife International, 2019). Due to their territoriality, residency, abundance and the fact that non-destructive samples are easily obtained from individuals in nest boxes, they have been frequently used as sentinels for metal and trace element contamination, even at their most northern distribution range (Bustnes et al., 2013; Carneiro et al., 2015; García-Seoane et al., 2017).

The suitability of the other owl species for pan-European monitoring is more limited, largely because of restricted distribution or migration. The Eurasian eagle owl takes the largest prey (Comay and Dayan, 2018), mainly mammals but also birds including raptors (Lourenço et al., 2015). It inhabits forest patches and agricultural habitats across Europe (Table SI-3). However, its distribution is irregular and it is absent in the UK, Ireland, the Netherlands and Iceland as well as in parts of France, Poland and Hungary (BirdLife International, 2019). This limits its capacity to act as sentinel for pan-European monitoring. Nevertheless, this species has been used as a sentinel for regional pollution

Table 2
Key traits of shortlisted candidate species for pan-European monitoring of terrestrial Hg. A complete list of traits and species with associated references can be found in Table SI-3 and the assessment of the criteria as suitable for terrestrial Hg monitoring is indicated in Table SI-9. Overall suitability is indicated for each criterion except for distribution where individual suitability is given in the superscript of each main region. AD = advantageous criterion and LI = limiting criterion for pan-European Hg monitoring.

Species	Distribution	Habitat	Migration
Northern Goshawk (*Accipiter gentilis*)	• Eastern Europe[AD] • Northern Europe[AD] (except Ireland, Iceland) • Southern Europe[AD] • Western Europe[AD]	• Forest habitats • Forest patches • Rarely urban habitats **➔ AD**	• Resident (but juvenile dispersal might occur in Fennoscandia) **➔ AD**
Tawny owl (*Strix aluco*)	• Eastern Europe[AD] • Northern Europe[AD] (except Ireland, Iceland) • Southern Europe[AD] • Western Europe[AD]	• Wide-habitat niche • Urban habitats • Farmland with patched forest • Forest habitats **➔ AD**	• Resident **➔ AD**
Eurasian eagle owl (*Bubo bubo*)	• Eastern Europe[AD] • Northern Europe[LI] (except UK, Ireland and Iceland) • Southern Europe[AD] • Western Europe[AD]	• Forest patches • Agricultural habitats • Open habitats **➔ AD**	• Resident **➔ AD**
Barn owl (*Tyto alba*)	• Eastern Europe[AD] • Northern Europe[LI] (except Fennoscandia and Estonia) • Southern Europe[AD] • Western Europe[AD]	• Farmland habitats • Urban habitats **➔ AD**	• Resident **➔ AD**
Little owl (*Athene noctua*)	• Eastern Europe[AD] • Northern Europe[LI] (except Fennoscandia, Ireland, Estonia) • Southern Europe[AD] • Western Europe[AD] (except alpine regions)	• Open farmland habitats **➔ AD**	• Resident **➔ AD**

studies (Gómez-Ramírez et al., 2019; Langford et al., 2013) and is known to bioaccumulate Hg (Broo and Odsjö, 1981; Espín et al., 2014). Barn owls predominantly feed on rodents in farmland habitats and are considered resident once they start breeding (Table SI-3). Although widely distributed, they are absent in Iceland and Fennoscandia, Estonia, alpine regions, and in parts of Romania and Bulgaria (BirdLife International, 2019). Finally, the little owl is more insectivorous than the other candidate owls (Comay and Dayan, 2018) but predominantly eats small mammals and birds (Table SI-3). The little owl is resident and prefers open agricultural landscapes, but, like the barn owl, is absent from Fennoscandia, Iceland, Estonia, alpine regions and Ireland (Table SI-3; BirdLife International, 2019).

Overall, the species listed in Table 2 generally meet key criteria for pan-European monitoring of Hg in the terrestrial environment. On the basis of selecting widespread species that do not migrate, tawny owl and northern goshawk may be the most suitable sentinels but there are two major advantages of the tawny owl. The first is that tawny owls occupy a large variety of different habitats, thereby facilitating assessment of habitat influences on Hg exposure. The second is that tawny owls are far more abundant with 535,000–939,000 breeding pairs in Europe compared with 166,000–220,000 for northern goshawks (BirdLife International, 2017). Northern goshawk might be the species of choice for monitoring particularly in areas of northern Fennoscandia due to its broader distribution in this region compared with that of tawny owls (BirdLife International, 2019). Interestingly however, although liver Hg concentrations were higher in northern goshawks than in tawny owls in Belgium, liver Hg concentrations in birds from Norway and Spain (Table SI-10) and feather Hg concentrations in individuals from Germany, Sweden and Spain were generally comparable in the two species (Table SI-11). Models for Hg deposition have reported highest deposition rates to be in central Europe and in localised regions in the UK (Lee et al., 2001). This is consistent with differences in Hg levels for both species for individuals from Germany, Belgium and UK compared with birds from Spain and Norway (Tables SI-10 and SI-11), which underlines their suitability for Hg biomonitoring. However, more studies using higher sampling numbers are needed to confirm this pattern, especially since local effects, such as the past use of alkyl-Hg in agriculture, might have resulted in elevated Hg levels in tawny owls from Sweden (Table SI-11).

4.2. Anticoagulant rodenticides (ARs)

A main factor associated with secondary exposure to ARs are a specialisation on rodent prey (López-Perea and Mateo, 2018), and so species that frequently forage on small mammals were shortlisted as the best candidate sentinel species. Facultative scavenging, because of increased likelihood of feeding on acutely poisoned prey was considered an advantageous trait and therefore obligate predation (species not known to scavenge) was considered a limiting factor. Use of habitats where ARs are commonly applied is associated with higher levels of exposure (López-Perea and Mateo, 2018) and so utilisation of anthropogenic land uses (habitats where ARs are most likely to be used) was also considered an advantageous trait. Limited distribution, lack of preference for mammalian prey, restricted habitat utilisation and partial migration were all considered traits that limited the suitability of the species for pan-European AR monitoring (Table SI-12). By applying these criteria, the list of all potential candidate species (Table SI-3) was reduced to the common buzzard, common kestrel, tawny owl, barn owl, Eurasian eagle owl, little owl and long-eared owl (Table 3). While all of these species have traits that make them suitable for pan-European monitoring of ARs, they also each have traits that limit their usefulness.

Although a generalist, the common buzzard predominantly forages on rodents when they are abundant and also scavenges rodents and other small mammals (Table SI-3 and references therein). These characteristics predispose this species to ingest sub-lethal AR concentrations in live prey and likely higher residues in poisoned rodents (López-Perea and Mateo, 2018). Common buzzards have been used in Europe to monitor both rodenticide exposure and poisoning (Coeurdassier et al., 2014; López-Perea and Mateo, 2018; Shore et al., 2006) but their partial migration in northern Europe limits their suitability for spatially-resolved pan-European monitoring of AR exposure. Although, the red kite (*Milvus milvus*), another scavenger, is particularly at risk of secondary AR exposure and

Table 3
Key traits of shortlisted candidate species for pan-European monitoring of ARs. A complete list of traits and species with associated references can be found in Table SI-3 and the assessment of the criteria as suitable for monitoring ARs is indicated in Table SI-12. Overall suitability is indicated for each criterion except for distribution where individual suitability is given in the superscript of each main region. AD = advantageous criterion, LI = limiting criterion for pan-European AR monitoring.

Species	Distribution	Foraging trait	Diet	Habitat	Migration
Tawny owl (*Strix aluco*)	• Eastern Europe**AD** • Northern Europe**AD** (except Ireland, Iceland) • Southern Europe**AD** • Western Europe**AD**	• Active hunter ➔ **LI**	• Small mammals Insects • Small birds ➔ **AD**	• Wide-habitat niche • Urban habitats • Farmland with patched forest ➔ **AD**	• Resident ➔ **AD**
Common buzzard (*Buteo buteo*)	• Eastern Europe**AD** • Northern Europe**AD** (except Iceland) • Southern Europe**AD** • Western Europe**AD**	• Active hunter • Facultative scavenger ➔ **AD**	• Mainly small mammals • Insects, Reptiles, Birds ➔ **AD**	• Agricultural habitats • Forest mosaics • Rarely urban habitats ➔ **AD**	• Partial migration ➔ **LI**
Common kestrel (*Falco tinnunculus*)	• Eastern Europe**AD** • Northern Europe**AD** (except Iceland) • Southern Europe**AD** • Western Europe**AD**	• Active hunter ➔ **LI**	• Mainly rodents • Avian prey • Invertebrates ➔ **AD**	• Agricultural habitats • Urban habitats ➔ **AD**	• Partial migration (mainly to SE but also to northern Africa) ➔ **LI**
Eurasian eagle owl (*Bubo bubo*)	• Eastern Europe**AD** • Northern Europe**LI** (except UK, Ireland and Iceland) • Southern Europe**AD** • Western Europe**AD**	• Active hunter ➔ **LI**	• Mainly mammals • Avian prey ➔ **AD**	• Forest patches • Agricultural habitats • Open habitats ➔ **AD**	• Resident ➔ **AD**
Barn owl (*Tyto alba*)	• Eastern Europe**AD** • Northern Europe**LI** (except Fennoscandia and Estonia) • Southern Europe**AD** • Western Europe**AD**	• Active hunter ➔ **LI**	• Mainly rodents ➔ **AD**	• Farmland habitats • Urban habitats ➔ **AD**	• Resident ➔ **AD**
Little owl (*Athene noctua*)	• Eastern Europe**AD** • Northern Europe**LI** (except Fennoscandia, Ireland, Estonia) • Southern Europe**AD** • Western Europe**AD** (except alpine regions)	• Active hunter ➔ **LI**	• Small mammals • Invertebrates ➔ **AD**	➔ Open farmland habitats ➔ **AD**	➔ Resident ➔ **AD**
Long-eared owl (*Asio otus*)	• Eastern Europe**AD** • Northern Europe**AD** (except Iceland) • Southern Europe**AD** • Western Europe**AD**	• Active hunter ➔ **LI**	• Mainly small mammals • Birds ➔ **AD**	• Forest patches • Agroforestry ➔ **AD**	• Partial migration in Fennoscandia ➔ **LI**

poisoning (Berny and Gaillet, 2008; Coeurdassier et al., 2012; Molenaar et al., 2017; Walker et al., 2018), it was not included as a candidate species for pan European monitoring because it is mostly absent in northern Europe and is migratory.

The common kestrel preys largely on small mammals but is not thought to scavenge extensively (Table SI-3), has a wide European distribution across agricultural and urban landscapes where ARs are widely used, and is known to be exposed to ARs (López-Perea and Mateo, 2018). Common kestrels are therefore likely to be generally suitable for monitoring exposure to ARs but they partially migrate to southern Europe (Holte et al., 2016), numbers are declining (BirdLife International, 2017) and they are less abundant than common buzzards (estimated European population of 409,000–603,000 pairs compared with 814,000-1,390,000 pairs of common buzzards; Birdlife International, 2017).

Of the owls, the tawny owl and barn owl have both been used for short and long-term monitoring of AR exposure in Europe (Geduhn et al., 2016; López-Perea and Mateo, 2018; Shore et al., 2019). They are abundant and often killed in traffic collisions, so carcasses are readily available for collection and subsequent analysis (Walker et al., 2008b). The barn owl however is more restricted than the tawny owl in habitat use, tending to be found primarily in agricultural landscapes, and is absent from parts of Europe. The Eurasian eagle owl and little owl can also be exposed to ARs (López-Perea and Mateo, 2018), but like the barn owl, both species are absent from areas of Europe (Table 3). The long-eared owl is similar to the barn owl in that it is a rodent specialist but is restricted in its habitat use, favouring agroforestry habitats (Table SI-3); it has been less widely monitored for ARs across European countries compared with, for example, common buzzards or tawny owls (López-Perea and Mateo, 2018).

On the basis of our proscribed methodology, the likely best candidate species for AR exposure monitoring at a European scale were the common buzzard and the tawny owl (Table 3). Another factor that may be important is whether sample mass for chemical analyses is a critical factor – the average mass of livers in non-starved individuals found dead in the UK between 2002 and 2019 were greater in common buzzard than tawny owls (mean ± SD of 16.9 ± 5.0 g ($n = 284$) vs 10.7 ± 2.9 g ($n = 392$); *Shore-pers. comm.*). Although common buzzards, as facultative scavengers, might be expected to accumulate higher liver AR residues than tawny owls, average residues in the two species, where measured, appear to be broadly similar (Table SI-13). Thus, facultative scavenging per se may in fact not be a more advantageous trait than active hunting when selecting a sentinel for monitoring AR exposure at a European scale. Furthermore, the partial migration of common buzzards in central and northern Europe (Table SI-3) is likely to be a significant issue if a key aim of monitoring is to examine spatial variation in exposure. This cannot be overcome, as suggested for Pb, by restricting carcass selection to the breeding season because liver half-lives can be months for some ARs (Vandenbroucke et al., 2008), longer than for Pb. Although the tawny owl may not be as widely or heavily exposed to ARs as some other species in Europe (López-Perea and Mateo, 2018; Walker et al., 2008b), its traits of feeding widely on rodents, residency and utilisation of multiple habitats, coupled with widespread distribution, abundance and availability/accessibility of carcasses, make it the most suitable species for monitoring pan-European spatio-temporal trends in AR exposure.

4.3. Pesticides

There are a large number of legacy and current-use pesticides that may potentially be of interest for pan-European monitoring. This diversity makes it difficult to select a single or small number of sentinel species that are best suited for monitoring this group of compounds as a whole. The current work focusses on selecting candidate species to evaluate outcomes of chemical, including PPP, regulation. However, raptors are unlikely to be first choice sentinels for monitoring trends in pesticides that do not bioaccumulate/biomagnify through food webs, as widespread significant exposure at high trophic levels is unlikely. However, birds of prey have been widely used to monitor environmental trends in legacy pesticides, such as organochlorine insecticides (Helander et al., 1982; Newton, 1986; Ratcliffe, 1967).

Exposure to pesticides is mainly related to foraging within agricultural settings such as farmlands, agroforestry and orchards. Active foraging is also likely to be an advantageous trait over facultative scavenging since exposure in scavengers may include individuals subject to deliberate and illegal persecution through the use of poison baits (Table SI-14). Although information on such persecution is important for forensic and toxico-surveillance studies, it is outside the scope of the current study. Finally, we also selected species that predominantly feed on rodents. Although feeding on avian prey is not a trait that prevents or limits bioaccumulation of pesticides (Herzke et al., 2002; Jaspers et al., 2006; Newton, 1986), feeding on relatively sedentary rodents and invertebrates means that exposure of prey and predator will broadly be co-located. The presence of birds migrating along the African-Eurasian flyway in the diet of sedentary raptors in Europe can introduce some uncertainty in the origin of contaminants. This can be avoided if the monitored raptors feed on sedentary prey. We used these traits and our European distribution as selection criteria to reduce the candidate species list for pesticide monitoring to common buzzard, common kestrel, Eurasian eagle owl, barn owl, little owl, long-eared owl and tawny owl. With the exception of favouring active hunting over facultative scavenging, the selection of species was the same as for ARs, reflecting that exposure to both pesticides and ARs is effectively largely influenced by the same ecological traits.

Given the shortlist of species was the same for pesticides as for ARs, we used the same logic as for ARs to eliminate Eurasian eagle owl, barn owl and little owl on the grounds of irregular species distribution, and common buzzard, common kestrel and long-eared owl because of partial migration. This left the tawny owl as the only species that met all the outlined criteria necessary for a sentinel suited for assessing spatio-temporal trends in exposure to pesticides (Table 4).

Although our selection criteria identified the tawny owl as potentially the most suitable raptor for biomonitoring PPPs, selection of a single species is problematic. This is because of the diversity of PPPs compounds and their varied environmental behaviour. Even if just bioaccumulative compounds such as the legacy organochlorine insecticides are considered, the tawny owl was used to monitor these compounds (for example; Table SI-15) but so were a wide range of other raptor species, and eggs were often analysed as well as tissues (Blus, 2011; Elliott and Bishop, 2011). As far as we are aware, there has been no over-arching evaluation of the relative sensitivities of different raptor species for monitoring temporal and spatial trends in OC insecticides. Such an analysis may provide a clearer picture of which raptor species may prove the most effective for monitoring trends of bioaccumulative pesticides. In terms of more current pesticides such as neonicotinoids, we found few studies that reported residues in raptors (Byholm et al., 2018; Taliansky-Chamudis et al., 2017). This likely reflects the move towards preventing registration of PPPs with high bioaccumulation potential and lower-trophic species may prove more useful sentinels for tracking changes in wildlife exposure (Bonneris et al., 2019; Bro et al., 2015). However, raptors that nest on the ground in agricultural habitats, such as Montagu's harrier (*Circus pygargus*) and western marsh harrier (*Circus aeruginosus*), may be useful indicators of risk from direct exposure (Cardador et al., 2012; Espín et al., 2018). Both are long-distance migrants but analysis of blood from nestlings or addled eggs might still provide important information for regional exposure within agricultural areas.

4.4. Medicinal products (MPs)

Terrestrial environmental emissions of MPs have been related to losses from human and animal manure fertilizers in arable and pasture areas, from livestock/poultry production units and from landfills (Arnold et al., 2014; Sarmah et al., 2006; Shore et al., 2014). Scavenging on treated livestock and other medicated animals, as exemplified by the effects of non-steroidal anti-inflammatory drugs (NSAIDs) on Asian vultures (Oaks et al., 2004), is also a key direct point of entry of VMPs into wildlife food webs (Blanco et al., 2017; Cuthbert et al., 2014; Margalida et al., 2014). Facultative scavenging and utilisation of agricultural habitats were therefore regarded as key traits facilitating exposure to MPs but, unlike with previous contaminant groups, long-distance and partial migration were not considered reasons to exclude species as candidates for pan-European monitoring (Table SI-16). This is because MPs typically have short half-lives in tissues of hours to days (Hutchinson et al., 2014) and so detection of residues in carcasses is likely to reflect recent exposure. Using these criteria, candidate species for monitoring MPs at a pan-European scale included common buzzard, common kestrel, Eurasian eagle owl, barn owl, little owl, long-eared owl and tawny owl. This is the same short-list that was derived for anticoagulant rodenticides (Table 3) and pesticides.

Of these species, the common buzzard meets the highest number of advantageous criteria for pan-European monitoring of MPs (Table 5). It is potentially exposed to MPs, particularly VMPs, through multiple routes because it actively forages in agricultural settings and is a facultative scavenger of livestock carcasses (Table 5). Other species that similarly scavenge include the red kite and also the black kite (*Milvus migrans*) which forages near freshwater habitats as well as dump sites (Table SI-3). However, both red and black kites are mainly absent in Fennoscandia (BirdLife International, 2019). Thus, these species may be suitable for monitoring MPs over large spatial ranges but their absence from the northern Europe region excluded them from the short-list of candidate pan-European biomonitors of MPs.

Table 4
Candidate species for pan-European monitoring of pesticides. A complete list of traits and species with associated references can be found in Table SI-3 and the assessment of the criteria as suitable for monitoring pesticides is indicated in Table SI-14. Overall suitability is indicated for each criterion except for distribution where individual suitability is given in the superscript of each main region. AD = advantageous criterion and LI = limiting criterion for pan-European pesticide monitoring.

Species	Distribution	Foraging trait	Habitat	Migration
Tawny owl (*Strix aluco*)	• Eastern Europe[AD] • Northern Europe[AD] (except Ireland, Iceland) • Southern Europe[AD] • Western Europe[AD]	• Active hunter ➔ **AD**	• Wide-habitat niche • Urban habitats • Farmland with patched forest • Forest habitats ➔ **AD**	• Resident ➔ **AD**

Table 5
Candidate species for pan-European monitoring of MPs. A complete list of traits and species with associated references can be found in Table SI-3 and the assessment of the criteria as suitable for monitoring of MPs is indicated in Table SI-16. Overall suitability is indicated for each criterion except for distribution where individual suitability is given in the superscript of each main region. AD = advantageous criterion and LI = limiting criterion for pan-European MP monitoring.

Species	Distribution	Foraging trait	Habitat	Migration
Common buzzard (*Buteo buteo*)	• Eastern Europe[AD] • Northern Europe[AD] (except Iceland) • Southern Europe[AD] • Western Europe[AD]	• Active hunter • Facultative scavenger ➔ **AD**	• Agricultural habitats • Forest mosaics • Rarely urban habitats ➔ **AD**	• Partial migration in autumn and winter to southern Europe (depending on weather conditions) ➔ **AD**
Common kestrel (*Falco tinnunculus*)	• Eastern Europe[AD] • Northern Europe[AD] (except Iceland) • Southern Europe[AD] • Western Europe[AD]	• Active hunter ➔ **LI**	• Agricultural habitats • Urban habitats ➔ **AD**	• Partial migration (mainly to SE but also to northern Africa) ➔ **AD**
Tawny owl (*Strix aluco*)	• Eastern Europe[AD] • Northern Europe[AD] (except Ireland, Iceland) • Southern Europe[AD] • Western Europe[AD]	• Active hunter ➔ **LI**	• Wide-habitat niche • Urban habitats • Farmland with patched forest ➔ **AD**	• Resident ➔ **AD**

Of the non-scavenging species, tawny owls and common kestrels had a similar number of favourable traits for MP monitoring as did the common buzzard, if exposure from scavenging livestock carcasses was not a focal interest (Table 5). However tawny owls may be preferred because of their greater abundance and the current stability of their numbers (BirdLife International, 2017). Other non-scavengers such as the Eurasian eagle owl, barn owl, little owl, and long-eared owl had similar favourable traits to tawny owls for monitoring MP but, as with other compounds, were excluded as pan-European biomonitors because of their limited species distribution. Further non-scavenging species like Montagu's harrier and western marsh harrier both breed in agricultural habitats and might therefore be directly exposed to MPs from manure fertilization (Table SI-3). However, as discussed previously for pesticides, major limitations for using harriers include their absence in most parts of northern Europe. Furthermore, their abundance is low in comparison with species such as the common buzzard (BirdLife International, 2017).

According to our proscribed methodology, the common buzzard appears to be the key sentinel species meeting the highest number of advantageous criteria, although tawny owl and common kestrel may also be suitable if exposure from scavenging was not a focal interest. However, we found no studies that reported MP residues in these species. This might be associated with low detection rates due to rapid metabolism of residues in tissues, although our knowledge of metabolic pathways in non-mammalian species is poor (Hutchinson et al., 2014), and may also reflect that screening of feathers for rapidly-metabolised pharmaceuticals (Whitlock et al., 2019), may not be commonplace. Furthermore, many wildlife studies on MPs have focussed on NSAIDs and concentrated on those species most at risk.

NSAIDs are poorly metabolised within their target organisms (Cuthbert et al., 2014; Sarmah et al., 2006) and this leads to direct exposure in scavengers that forage on dead livestock and other medicated species (Margalida et al., 2017). Monitoring of NSAIDs in vultures in Europe is of prime interest because exposure to these compounds, principally diclofenac, has led to massive population declines in *Gyps* vulture populations elsewhere (Margalida et al., 2014; Oaks et al., 2004). Such monitoring provides important information that underpins and informs risk management. There are four vulture species resident within Europe, namely the bearded vulture (*Gypaetus barbatus*), cinereous vulture (*Aegypius monachus*), Egyptian vulture (*Neophron pernopterus*) and Eurasian griffon vulture (*Gyps fulvus*). All four forage on livestock within montane regions, and this is likely to be their main route of exposure to NSAIDs and to VMPs generally (Cuthbert et al., 2014), although the Egyptian vulture also frequently feeds on landfills (Table SI-5) which can contain HMPs. These vulture species do not have widespread European distributions and are mainly restricted to montane/alpine regions (Table SI-5), and so are not suitable monitors for pan-European monitoring of MPs. However, monitoring of NSAIDs in vultures across all the areas of Europe where they occur is merited for conservation and risk management purposes.

5. Conclusions

Our study has focussed on biomonitoring the European terrestrial environment for a set of priority pollutant groups through measurement of residues in tissues obtained from the carcasses of raptors found dead. Traits that we argue may be key are widespread distribution across the monitoring area, feeding ecology and habitat selection. Overall, for the geographical region, compound groups and monitoring techniques that we considered, the common buzzard and tawny owl were amongst, if not the most, suitable species. Both are abundant and widely distributed across Europe (BirdLife International, 2017). Although used in contaminant studies in Europe (Gómez-Ramírez et al., 2014), they are by no means the most intensively monitored species. While choice of one over the other of these two species may depend upon how important scavenging is considered as an exposure pathway and whether partial migration (common buzzard) is likely to compromise the aims of any programme, the lack of widespread contaminant monitoring in these two species suggests that their utility as sentinels for environmnetal pollutaion is not generally recognised. Furthermore, although we focussed here on monitoring across Europe, the trait-based approach that we used to identify the most suitable species could be easily applied to other continents and contaminants. Such analyses may equally reveal species in those regions that have been under-appreciated as biomonitors of pollution.

Our study, not surprisingly, does not suggest that a single species is ideal for monitoring exposure to all different groups of priority pollutants. For example, we concluded that the golden eagle, perhaps in combination with the white-tailed sea eagle, may be better than common buzzard for monitoring exposure to, and particularly toxic effects from, Pb in ammunition. However, such combined monitoring is potentially complex as confounding factors include interspecific differences in pharmacokinetics or seasonal variation in scavenging. We also suggest that when northern Fenno-Scandinavian habitats may be an important component of biomonitoring, the northern goshawk could be a better monitor than tawny owl for terrestrial Hg, reflecting the lack of tawny owls in those northern areas. However, there are always likely to be trade-offs in terms of balancing completeness of spatial coverage against likely widespread availability of carcasses for monitoring. Even when a single species appears the most suitable biomonitoring candidate, there may be significant intra-specific variation in contaminant exposure because individuals have different diets (Palma et al., 2005). Such effects may be marked when individuals feed at different trophic levels (Badry et al., 2019; Elliott et al., 2009). Stable isotopes values of nitrogen ($\delta^{15}N$), carbon ($\delta^{13}C$) and sulphur ($\delta^{34}S$) can be used as proxies to control for dietary plasticity and trophic position in raptors (Eulaers et al., 2014; Eulaers et al., 2013) and we recommend that they are routinely measured along with the contaminants. This would help refine interpretation of apparent spatial and temporal trends in contaminants, particularly if accompanied by information on the isotopic signatures of common prey species.

In conclusion, the present study suggests that the selection of candidate species for continental-scale monitoring of exposure to a range of contaminants can be reduced to very few raptor species. This may be true for many regions of the world, not just Europe, and a trait-based evaluation (as used here) of the suitability of raptors as biomonitors across other continents may prove worthwhile. In our study, the common buzzard and tawny owl appear to be the two most suitable species for a range of contaminant groups. A logical conclusion from our study is that, if common buzzard and tawny owl are both broadly suitable for monitoring pan-European spatio-temporal trends in exposure, then any such trends will be similar in both species. We are not aware of datasets which we can use to test this prediction or assess the relative power of monitoring in both species to detect such trends. This is a key data-gap. Pilot monitoring studies involving harmonised sampling across Europe in these two species are merited.

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Declaration of competing interest

The authors declare no conflicts of interest.

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Appendix A. Supplementary data

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