A review on contaminants of emerging concern in European raptors (2002-2020)

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Abstract

Raptors (birds of prey and owls) have been widely used as suitable bioindicators of environmental pollution. They occupy the highest trophic positions in their food chains and are documented to bioaccumulate high concentrations of persistent pollutants such as toxic metals and legacy persistent organic pollutants (POPs).Whereas raptors played a critical role in developing awareness of and policy for chemical pollution, they have thus far played a much smaller role in current research on contaminants of emerging concern (CECs). Given the critical knowledge obtained from monitoring 'legacy contaminants' in raptors, more information on the levels and effects of CECs on raptors is urgently needed. This study critically reviews studies on raptors from Europe reporting the occurrence of CECs with focus on the investigated species, the sampled matrices, and the bioanalytical methods applied. Based on this, we aimed to identify future needs for monitoring CECs in Europe. Perfluoroalkyl substances (PFASs), novel flame retardants (NFRs), and to a lesser extent UV-filters, neonicotinoids, chlorinated paraffins, parabens and bisphenols have been reported in European raptors.White-tailed Eagle *(Haliaeetus albicilla)*, Peregrine falcon *(Falco peregrinus)* and Northern goshawk *(Accipiter gentilis)* were the most frequently studied raptor species. Among matrices, eggs, feathers and plasma were the most widely employed, although the potential role of the preen gland as an excretory organ for CECs has recently been proposed. This review highlights the following research priorities for pollution research on raptors in Europe: 1) studies covering all the main classes of CECs; 2) research in other European regions (mainly East Europe); 3) identification of the most suitable matrices and species for the analysis of different CECs; and 4) the application of alternative sample treatment strategies (e.g. QuEChERS or pressurized liquid extraction) is still limited and conventional solvent-extraction is the preferred choice.

Keywords: birds of prey; emerging organic contaminants; perfluoroalkyl substances; novel flame retardants; bisphenols; neonicotinoids.

1. Introduction

Biomonitoring practices are very important for studying the environmental impact of chemical pollution **(Woodruff, 2011)**. The direct analysis of contaminants in organisms provides information on bioavailability but also information about the intake by biota or potential exposure pathways for humans. Among all the organisms that can be considered sentinels of environmental contamination, raptors (members of *Falconiformes, Strigiformes* or *Accipitriforme*) are especially suitable for monitoring persistent, bio-accumulative and toxic (PBT) substances **(Movalli et al. 2019, Espín et al. 2016, Sergio et al. 2005, 2006)**. As top predators, they are likely to have particularly high levels of environmental contaminants and therefore individual- and population-level effects are most likely measureable in raptors prior to other taxa **(Kovács et al. 2008, Helander et al. 2008)**. Furthermore, they are widespread and long-lived organisms, thus allowing the assessment of spatial and temporal trends of PBT chemicals and their associated effects on populations **(Furness, 1993)**. Another advantage of using these avian species for monitoring is that non-destructive sampling approaches can be applied (e.g. blood, feather, preen gland oil) **(Espín et al. 2016, EURAPMON, 2014).** Indeed, raptors have been used as bioindicators in many studies worldwide **(Elliott et al. 2009, Grove et al. 2009, Behrooz et al. 2014, Martínez-**

López et al. 2015, Elliott et al. 2015, Rattner et al. 2018, Manning et al. 2018, Hong et al. 2019, Aver et al. 2020).

In Europe, there has been a focus with several biomonitoring programs on raptors to track the success of mitigation strategies for reducing the exposure to contaminants **(Gómez-Ramírez et al. 2014)**. However, these schemes have mainly been performed in western European countries and have hitherto focused on the monitoring of toxic elements, such as mercury (Hg) and lead (Pb), and of legacy persistent organic pollutants (POPs), mainly organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), dioxins, furans and anticoagulant rodenticides **(Gómez-Ramírez et al. 2014)**. Among the POPs, OCPs, PCBs and PBDEs are widespread in wildlife **(Blanco et al. 2018, Monclús et al. 2018)** and their use is currently regulated by the Stockholm Convention **(United Nations Evironment Programme (UNEP, 1972))**. Although a great effort is currently being made to coordinate and integrate these multiple independent monitoring programmes on raptors in Europe **(Gomez-Ramirez et al. 2014, European Raptor Biomonitoring Facility-ERBFacility 2017, Badry et al. 2020)**, there is still a greater need to identify current trends at the broader European spatial scale **(Espin et al. 2016, Movalli et al. 2019)**. The current European Raptor Biomonitoring Facility (ERBFacility, COST Action CA16224) is active in this respect, working towards coordinated Europe-wide monitoring of contaminants in raptors, with a view to support the implementation of EU chemicals regulations and reducing chemical risks to the environment and to human health (https://erbfacility.eu/). Further, the LIFE APEX project is working to demonstrate the use of apex predators, including raptors, for the monitoring of contaminants of emerging concern, with a view to better chemicals management in Europe (www.lifeapex.eu). New challenges are emerging due to the

restrictions and bans on the use of POPs, which has led to the emergence of other chemical classes of contaminants in recent years, also known as "contaminants of emerging concern" (CECs) **(Thomaidis et al. 2012, Sauvé & Desrosiers 2014)**. These chemicals, for which currently no or very limited regulations are established, have received considerable attention due to their widespread occurence in the environment and their potential toxicity **(Blum et al. 2015)**. According to the network of reference laboratories for monitoring of emerging environmental contaminants (NORMAN), CECs can be defined as substances that have been detected in the environment, but which are currently not included in routine monitoring programmes and whose fate, behaviour and (eco) toxicological effects are not well understood. More than 700 emerging contaminants have been identified in water bodies up to now, such as novel flame retardants (NFRs), perfluoroalkyl substances (PFASs), bisphenols, ultraviolet-filters (UV filters), chlorinated parafins and parabens **(NORMAN List of Emerging Substances, 2016)**. Despite their current concern, otherchemical categories like rodenticides or PBDEs are not considered CECs and are not included in this review.

It is essential to improve the effectiveness of evaluation, risk assessment and early warning in relation to the regulation of CECs **(Cousins et al. 2020)**. Until now, only limited biomonitoring schemes for PFASs in raptors are established in Europe at a national scale even though they have been identified as Persistent Bioaccumulative and Toxic (PBT) compounds **(Espín et al. 2016)**. For the remaining CECs, there is still limited data regarding their bioaccumulation in biota. However, as mentioned above, efforts are currently underway through ERBFacility and LIFE APEX to assess occurrence in raptors **(Movalli et al 2019; ERBFacility, 2017; LIFE APEX)**.

With this as background, the main aim of the present study was to 1) review current studies of CECs in raptors to determine what species, matrices and analytical

methods are used and their frequency, and 2) to determine any knowledge gaps and areas for future research highlighted by the literature review.

2. Method of literature review

Only international peer-reviewed studies related to the analysis of CECs in raptors from Europe were collected by the search engines of Web of Science, Science Direct and Google Scholar. Grey or other unpublished studies (such as theses, conference proceedings, book chapters or reports) were not included. Initially, a total of 1,742 articles related to the analysis of contaminants in birds were found when using "contaminants" and "birds" as keywords in Web of Science. This search was then filtered by area (Europe), species (raptors) and type of contaminants (PFASs, NFRs and other CECs). For that purpose, we used different combinations of contaminant names (PFASs, NFRs, bisphenols, paraffins, neonicotinoids, benzophenones, UV-filters, parabens) and European countries as keywords along with "Europe", "raptors", "birds of prey", "owls" "contamination", "predators" "scavengers" and species (white-tailed eagle, Northern gowshak, peregrine falcon, Eurasian sparrowhawk *(Accipiter nisus)*, common kestrel *(Falco tinnunculus)*, red kite *(Milvus milvus)*, long-eared owl *(Asio otus)*, barn owl *(Tyto alba)*, tawny owl *(Strix aluco)*, common buzzard *(Buteo buteo)*, and others) and/or matrices names (eggs, fat, feathers, blood, plasma, liver, internal tissues, bones). References from the collected publications were also reviewed to ensure that all relevant studies were included. Although a limited period of time was not initially set, all the peer-reviewed studies of our interest were published in the period 2002-2020, which was finally considered. All studies with solid methods and quality assurance/quality control were included, even though they had low sample sizes due to

151 the scarce literature on contaminants of emerging concern in raptors. (n=37) were
152 included in the review.

153 For each study, concentrations of contaminants, analytical methods employed, sample
154 types and discussion on temporal trends and comparison of levels with legacy
155 compounds were collected. Contaminants were classified in three major classes:
156 perfluoroalkyl substances (PFASs), novel flame retardants (NFRs) and other
157 miscellaneous CECs, namely, bisphenols, UV-filters, neonicotinoids, paraffins and
158 parabens. Concentrations in the text are provided as ranges, means or medians of single
159 contaminants or contaminant classes (total sum of the concentrations of all the
160 contaminants within a class) and expressed in ng g^{-1} or ng mL^{-1}. Contaminant
161 abbreviations used throughout the manuscript are listed in **Table 1.**

162 **(Insert Table 1)**

163

164 **3. Results and discussion**

165 To provide a general overview, two tables (**Tables 2 and 3)** were constructed
166 with the collected data. **Table 2** contains data related to the raptor species, including
167 number of analyzed individuals (sample population) and year of collection, countries
168 where the studies were performed, analyzed matrices and measured concentrations.
169 Sample treatment and analytical technique/s employed in each case are presented in
170 **Table 3**. The most relevant results derived from the studies collected in Tables 2 and 3
171 are presented below.

172 **(Insert Tables 2 and 3)**

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174

175 **3.1 Occurrence of PFASs, NBRs and other CECs in raptors across Europe**

176 Most of the studies on raptors from Europe focus on POPs; we only found 19
177 studies on PFASs, 12 on NFRs and 6 on other miscellaneous CECs. An overview of the
178 European countries in which the different classes of contaminants have been assessed in
179 the respective studies is presented in **Figure 1**.

180

181 **(Insert Figure 1)**

182

183 As it can be observed, most of the analyses were perfomed in Western Europe,
184 mainly in Norway and Spain. Raptors from Norway (11 out of 19 studies) were the
185 most collected for the analysis of PFASs and those from Spain for NFRs (7 studies out
186 12).

187

188 The compounds that were most frequently determined within each group were
189 carboxylates (PFCAs) and sulfonates (PFSAs), especially perfluorooctane sulfonic acid
190 (PFOS), for PFASs, and dechloranes (Dec), dechlorane plus (DP; derivative of
191 dechlorane class), and phosphorus flame retardants (PFRs) for NFRs.

192 Regarding the analysis of the other miscellaneous CECs, only six studies were
193 found in the literature with samples from different European countries: two on
194 neonicotinoids in Finland **(Byholm et al. 2018)** and in Spain **(Taliansky-Chamudis et**
195 **al. 2017)**, one on UV-filters in Spain **(Molins-Delgado et al. 2017)**, one on parabens in
196 Germany and Polish coastal areas of the Baltic Sea **(Xue & Kannan 2016),** one on

paraffins in Sweden and Denmark **(Yuan et al. 2019)** and one on bisphenols and benzophenone-type UV-filters in Greenland and France **(González-Rubio et al. 2020).**

3.1.1 Perfluoroalkyl substances (PFASs)

PFASs are a group of chemicals of anthropogenic origin that have been in use since the 1940s in many consumer products like cookware, food packaging, and stain repellants (**Giesy & Kannan 2002; Begley et al. 2005; Gewurtz et al. 2009)**. Most of them exhibit similar structures consisting of a partially or fully fluorinated alkyl chain with a particular functional head group leading to compounds with unique physicochemical properties. PFASs are ubiquitous environmental contaminants and, although they exhibit favorable properties in manufacturing, they have proved to be toxic, very resistant to degradation and long-term bioaccumulative **(Cousins et al. 2019, Hekster et al. 2003)**. However, so far, only perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) have been restricted in production and use from 2009 and 2020, respectively, according to Annex B and A of the Stockholm Convention on Persistent Organic Pollutants **(UNEP 2009a, UNEP 2020).** Others have been only recommended for voluntary phase-out, or still unrestricted in their use. The concern related to the toxicity of PFASs has considerably increased, the number of studies related to these compounds over the past decade **(Ghisi et al. 2019, Galatius et al. 2019).** Among PFASs, PFCAs and PFSAs are the most studied and bioaccumulate in marine food chains, with highest concentrations found in organisms at higher trophic levels **(Kannan et al. 2005)**. While the toxic effects of legacy compounds have frequently been studied in birds of prey **(Bustnes et al. 2011, Mora et al. 2011, Crosse et al. 2013, Elliot et al. 2015),** the knowledge about the potential effects of emerging

compounds such as several PFASs is scarce. In this section, the most relevant results from studies on PFAS from raptor samples collected in Europe have been reported.

3.1.1.1. PFASs concentrationsin liver samples

An increasing interest has emerged on the presence of PFASs in raptors in Europe since in 2002 **Kannan et al.** reported for the first time the occurrence of PFOS and related PFASs in liver samples of white-tailed eagles collected from 1979 to 1999 in Germany and Polish coastal areas of the Baltic Sea.They only found quantificable concentrations of PFOS, ranging from <3.9 to 127 ng g^{-1} wet weigh (ww). Higher concentrations of PFOS were detected in further studies in liver samples collected from Belgium: a concentration range from 47.6 to 775 ng g^{-1}(ww) was reported in Eurasian sparrowhawks **(Meyer et al. 2009)** and from 42 to 992 ng g^{-1} (ww) in Barn owls **(Jaspers et al. 2013).** These higher concentrations were potentially due to that individuals from both studies were sampled from an area in the close vicinity of the city of Antwerp (Belgium), where a major PFAS chemical plant is located. Other PFASs were detected in both studies but at lower concentrations (3.2-40.6 ng g^{-1} (ww) in **Meyer et al. 2009** and of 6-116 ng g^{-1} (ww) in **Jaspers et al. 2013**.

3.1.1.2. PFASs concentrationsand temporal trends in eggs

Many studies determined PFASs in eggs (6 out of 19). Eggs were found suitable to study temporal trends of PFASs concentrations **(Arehns et al. 2011, Bustnes et al. 2014, Faxneld et al. 2016)**. However, it is difficult to compare the results of different studies due to the fact that concentrations (although reported as ng g^{-1}) were expressed in different ways (in wet, dry or lipid weight, in arithmetic or geometric means or

medians). This highlights the need to harmonize the units for reporting levels in environmental analysis and to have additional data with the publications on wet weight and lipid percentage regardless of the units used in the publication.

Although a decline of PFOS should be expected as a result of the phase-out and regulatory interventions, most of them have reported it as the most abundant PFAS being presentat high concentrations that kept constant during the whole examined time period. This may be due to the high persistence and long half live of this compound or due to a short course of time between the phase-out and the sample analysis.

In 2010, **Holmström et al.** established for the first-time temporal trends for PFASs in terrestrial biota with the analysis of Swedish peregrine falcon (*Falco peregrinus*) eggs collected from 1974 to 2007. Authors showed that the overall PFAS profile was dominated by PFOS (83 ng g^{-1} ww mean concentration in samples from 2006) and changes in concentrations were not significant between 1984 and 2006 (mean concentrations: 80-90 ng g^{-1} (ww)). In contrast, increasing temporal trends of long-chain PFCAs were recorded in many studies. PFCA bioaccumulation potential increases with increasing chain length and they seem to accumulate to a higher extent in lipid-rich tissue, including eggs, than PFASs with shorter chains **(Braune & Letcher, 2013). Holmströmet al (2010)** reported that PFCA concentrations increased exponentially over the studied time period. Similar results were observed by **Eriksson et al. (2016)** and **Vorkamp et al. (2019).** Significant differences in PFOS concentrations in Swedish osprey (*Pandion haliaetus*) eggs within the periods 1997-2001 (103 ng g^{-1} ww), 2008-2009 (64 ng g^{-1} ww) and 2013 (70 ng g^{-1}ww) were determined in **Eriksson et al. (2016).** Furthermore, a significant increase was observed for long chain PFCAs (C_{10}-C_{14}) in years 1997-2001 and 2008-2009. Concentrations of PFASs in the eggs of peregrine falcons from Greenland collected between 1986 and 2014 (100-303 ng g^{-1}dw;

ΣPFAS median range) were also reported in **Vorkamp et al. 2019**. In this case, PFOS accounted for 94% on average of all PFSAs, but did not show a significant time trend. The long-chain PFCAs C_9, C_{12} and C_{14} showed increasing concentrations, with an annual increase of 0.8%, 2.9% and 5.6%, respectively **(Vorkamp et al. 2019)**. A longer-term study reported temporal and spatial trends of PFASs in eggs from white-tailed sea eagles from two fresh water and two marine areas in Sweden collected from 1966 to 2010 **(Faxneld et al. 2016).** Most PFASs showed a general increase (from the 1960s/1980s to 2010) and significant decreasing concentrations were not observed during the most recent years, including PFOS, which is in the same line with the above-mentioned recent studies.

Some studies have shown decreasing concentrations of PFOS, for example in tawny owl (*Strix aluco*) eggs from Norway (1986-2009) with an annual decrease of 1.6% (**Ahrens et al. 2011).** However, C_{10}-C_{13} PFCA concentrations increased significantly with an annual increase of 4.2-12%, as observed by other authors **(Eriksson et al. 2016, Holmström et al. 2010)**.The results of **Ahrens et al. (2011)** suggested that individual traits had very little impact on the concentrations of PFASs in the eggs while there was an important influence of annual variation in environmental conditions such as the North Atlantic Oscillation, temperature, snow or food availability.

3.1.1.3. PFASs concentrations in plasma

PFASs have also frequently been determined in blood plasma (9 out of 19 studies). In fact, half of the studies analyzing PFASs in raptors have used plasma as preferred matrix due to their protein affinity. Thus, these compounds are primarily retained in blood plasma and internal organs due to their physicochemical properties. The negative impact of the presence of organohalogen compounds in the clinical-

297 chemical parameters of blood plasma was first investigated in 2010 by **Sonne et al.** in
298 chicks of three raptor species from Norway (2008-2009). Furthermore, PFASs
299 concentrations in this study (21-55 ng mL^{-1}ww; ∑PFAS, median) were higher than
300 those reported in further ones (median, ng mL^{-1}ww): 42 **(Sletten el al. 2016),** 17-46
301 **(Gómez-Ramírez et al. 2017),** 9-32 **(Løseth et al. 2019b),** 5-17.5 **(Briels et al. 2019).**
302 In 2012, **Sonne et al.** carried out a larger study (Norwegian raptor nestlings,
303 2008-2010). Concentrations in this study (12-151 ng mL^{-1} ww; ∑PFAS mean range)
304 were the highest reported in plasma from European raptors so far.
305 Other studies in plasma reported concentrations of a single PFAS either because
306 authors focused the investigation on one of them **(**PFOS median range: 6.2-13 ng mL^{-1}
307 ww, **Bustnes et al. 2013)** or because only one compound was detected (PFUnA median
308 range: 1.15-3.9 ng mL^{-1} ww, **Løseth et al. 2019a and b).**
309
310 ***3.1.1.4. PFASs concentrations and temporal trends in feathers***
311 **Sun et al. (2019)** reported long-term (1968-2015) spatio-temporal trends of
312 PFAS by using archived body feathers of white tailed-eagles from the West Greenland,
313 Norwegian and Central Swedish Baltic coasts. They observed a significant decline of
314 PFOS since the mid-1990s to 2000 in the Greenland and Norwegian subpopulations,
315 consistent with the phase-out of PFOS-based compounds. However, increasing PFOS
316 trends were observed in the Swedish subpopulation, which is probably due to the high
317 remaining contamination in the Baltic Sea. PFOS concentrations were significantly
318 higher in Sweden (median range: 3-20.1 ng g^{-1}ww) than in Norway (median range: 1.6-
319 9.5 ng g^{-1} ww) and Greenland (median range: 1.6-9.5 ng g^{-1} ww) populations. FOSA
320 (median ranges in ng g^{-1} ww; Sweden: 0.3-1.5, Norway: 0.7-3.5 Greenland: 1.1-8.3) and
321 ∑PFCA concentrations (median ranges in ng g^{-1} ww; Sweden: 0.1-9, Norway: 2-6.1

Greenland: 3.1-8.2) were similar among the subpopulations. ΣPFCA concentrations significantly increased in all three subpopulations throughout the study periods. These results indicated that spatial differences in exposure may influence time trends.

Some studies investigated the associations between PFASs in blood plasma and body feathers to find out if feathers could be a non-invasive strategy to monitor PFASs exposure in raptors. **Gómez-Ramírez et al. 2017** determined PFAS concentrations in blood plasma (45 ng mL^{-1}ww; ∑PFASs mean) and body feathers (13 ng g^{-1} ww; ∑PFASs mean) from white-tailed eagle nestlings and found significant correlations between them. However, in **Løseth et al. 2019a**, only PFUnA was quantified in over 50% of both plasma (1.15-3.59 ng mL^{-1}; PFUnA median range) and feather (0.15-0.58 ng g^{-1}; PFUnA median range) samples, yet their correlation was poor and not significant. Therefore, the authors concluded that the use of feathers for monitoring PFASs may be limited, which is also supported by **Jaspers et al. (2019)**. Correlations between plasma (4.7-17.5 ng mL^{-1}ww; ∑PFASs median range) and feathers (1.8-6.2 ng g^{-1} ww; ∑PFASs median range) were also under discussion in another study, since strong and significant associations were found for some PFAS compounds and no correlation was found for others **(Briels et al. 2019).** Finally, it is also worth mentioning that the lack of correlations between plasma and feathers in some cases may be due to the fact that the latter ones reflect atmospheric contamination and/or different exposure during their time of growth, which may or may not coincide with plasma collection.

3.1.1.5 PFASs concentrations in other matrices

Other matrices have also sporadically been analysed to determine PFASs such as muscle, preen oil and fat of barn owls from Belgium **(Jaspers et al. 2013).** Concentrations reported in this latter study were especially high in the preen oil (22-431

ng g^{-1} ww; PFAS median range). Furthermore, a recent study **(González-Rubio et al. 2020)** highlighted the potential role of the preen glad from white-tailed eagles from Greenland as a major excretory organ for a group of CECs (bisphenols and benzophenones), which may be also worth to investigate for PFASs. Therefore, the investigation of PFASs concentrations in excretory matrices (such as preen gland and oil) in future research it is highly recommended.

Finally, on the basis of the studies collected on PFASs in raptors in Europe, eggs and blood are the most used matrices. Furthermore, in plasma, PFASs present the higher detection frequencies for all the compounds in the higher concentrations **(Briels et al. 2019, Løseth et al. 2019a**) which suggest that the birds are recently and continuosly exposed to these emerging contaminants, likely through dietary input. For the study of temporal trends, eggs are the most used matrix **(Ahrens et al. 2011, Faxneld et al. 2016)**. Thus, depending on the type of study that is going to be carried out, either plasma or eggs are recommended. Feathers may not be that suitable, as suggested in **Briels et al. 2019** and **Løseth et al. 2019a,** and other matrices need to be further investigated, with particular attention to excretory ones.

3.1.1.6 Comparison of PFASs and PBDEs.

Several studies simultaneously reported concentrations of PFASs and PBDEs (legacy organic contaminant group) in raptors. **Table S1** in the Supporting Information provides a comparison between them. PBDEs are organobromine compounds widely used as flame retardants in building materials, electronics, plastics, textiles, and other materials **(McGrath et al. 2017)**. Because of their toxicity and persistence, some PBDEs were phased-out in 2009 under the Stockholm Convention on persistent organic pollutants **(UNEP, 2009b and c)**. Thus, lower concentrations of these compounds are

372 expected to be bioaccumulated compared to PFAS ones, for which only PFOS and
373 PFOA have been restricted so far too. **Table S1** shows PFAS and PBDE concentrations
374 in plasma and feathers from six different studies. In two studies, plasma and feathers
375 were collected from northern goshawks and white-tailed eagles from Norway between
376 2015 and 2016 **(Briels et al. 2019, Løseth et al. 2019a).** Other three studies analyzed
377 the plasma of the same species and of golden eagle *(Aquila chrysaetos)* individuals from
378 Norway in the period 2008-2010 **(Sonne et al. 2010, Sonne et al. 2012**, **Løseth et al.**
379 **2019b)**. The other study analyzed these contaminants in plasma from Egyptian vultures
380 *(Neophron percnopterus)* from Spain from 2010 to 2016 **(Ortiz-Santaliestra et al.**
381 **2019)**. PFASs concentrations in these studies in plasma were higher than those of
382 PBDEs, as expected. However, when comparing PFASs and PBDEs concentrations in
383 feathers (**Table S1**) differences were less consistent than those measured for plasma.
384 For example, in **Løseth et al. (2019a)**, the median range of BDE-47 (0.3-1.73 ng g^{-1}) in
385 feathers of Norwegian white-tailed eagles was higher than the median range of PFUnA
386 (0.15-0.58 ng g^{-1}). This could be due to a lower PFAS detection frequency rate in
387 feathers, which has been demonstrated in previous studies (**Gómez-Ramirez et al.**
388 **2017**). The rest of studies reported greater concentrations of PFASs than of PBDEs.
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391 ***3.1.2. Novel flame retardants (NFRs)***
392 Restrictions on the use of legacy flame retardants, such as PBDEs or
393 hexabromocyclododecanes (HBCDs), have resulted in an increased use of novel flame
394 retardants (NFRs) in recent years as alternative to meet flammability standards.
395 Consequently, environmental concentrations of NFRs have been on the rise worldwide.
396 Some of them have already been detected in humans **(Ma et al. 2017, Pirard &**

Charlier 2018), as well as in a few captive and wild birds **(Gentes et al. 2012, Guigueno & Fernie 2017)**, marine mammals **(Lewis et al. 2020)**, and fish species **(Sapozhnikova & Lehotay 2013)**. Similar to PBDEs, NFRs are highly hydrophobic and display relatively low volatility. However, differences in their molecular structure result in specific differences in physicochemical properties and hence, in diverse behaviour in environmental and biological systems. The most common NFRs replacing PBDEs are organophosphorus flame retardants (OPFRs), dechlorane plus (DPs) and "novel" brominated flame retardants (NBFRs) such as decabromodiphenyl ethane (DBDPE), 1,2-*bis*(2,4,6-tribromophenoxy) ethane (BTBPE), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (TBB or EHTBB) and bis(2-ethylhexyl)-3,4,5,6-tetrabromo-phthalate (TBPH or BEHTBP).

Interest regarding the analysis of NFRs in European raptors has grown in recent years. Furthermore, further investigation of these contaminants in raptors is needed since these species seem to be more sensitive to flame retardant toxicity than domesticated species and songbirds **(Guigueno & Fernie 2017).** The first study to report the accumulation of dechlorane plus (DP) and dechloranes (Dec) in European biota was carried out by **Guerra et al. (2011)** by the analysis of eggs in peregrine falcons from Spain. ΣDec means ranged from 0.2 to 8 ng g^{-1} lipid weight (lw) and a ΣDP mean value of 2 ng g^{-1}(lw) was reported. In 2014, higher concentrations of Dec were detected in bird eggs of several raptor and non-raptor species from Doñana (a national natural park located in South-western Spain) as well as its surroundings, where small urban areas with intensive agriculture could be found **(Barón et al. 2014)**. ΣDec means ranged from 7 to 46 ng g^{-1} (lw). In 2015, a temporal trend study was performed in the same geographical area **(Barón et al. 2015)**, which reported Σ(Dec, DP) of 17-19

ng g^{-1} (lw, median range) in eggs of black kites (*Milvus migrans*) between 1993 and 2013. However, clear temporal trends were not observed for DP, and the authors highlighted the importance of continuous ecotoxicological monitoring of local species. Recently, **Eljarrat et al. 2019** determined ∑(Dec, DP) in one egg of a western marsh harrier individual from central Spain at a mean concentration of 10 ng g^{-1}lw.

Regarding NBFRs,**Vetter et al. (2017)** found concentrations of several NBFRs (HBDC; 2,3-Dibromopropyl-2,4,6-tribromophenyl ether (DPTE); 2-Bromoallyl-2,4,6-tribromophenyl ether, (BATE); Allyl-2,4,6-tribromophenyl ether, (ATE); Hexabromo benzene, (HBB); 2-4-6-Tribromoanisole, (2-4-6-TBA); 2,4,6-Tribromophenol, (2-4-6 TBP); Pentabromoethylbenzene, (PBEB); Pentabromotoluene, (PeBT); mean range: 1-33 ng g^{-1}lw) in the eggs of peregrine falcons collected in 2014 from Southern Germany. Higher NBFR concentrations (mean range: n.d-353 ng g^{-1} lw) were reported in eggs of black kites collected from an area in Madrid (Spain) that was considered as highly contaminated **(Blanco et al. 2018)**. In 2017, **Vorkamp et al.** studied temporal trends of legacy and NFRs (TBB, BTBPE, DPTE, DBDPE, DP) in eggs of peregrine falcons from Greenland collected between 1986 and 2014. NBFRs had low median concentrations (ranging from 0.5-4.7 ng g^{-1} lw) and, as reported by **Barón et al. (2015),** DP did not show a significant increase.

OPFRs (Tris (phenyl) phosphate (TPhP) ;tris (1-chloro-2-propyl) phosphate, (TCiPP) ;Tris-(2-chloroisopropyl)phosphate, (TCPP) ;Tris (chloroethyl) phosphate, (TCEP); Tri-(2-butoxyethyl)-phosphate (TBOEP); 2-Ethylhexyl diphenylphosphate, (EHDPP)) were determined in feathers and plasma of white-tailed eagle nestlings from Norway **(Eulaers et al. 2014)**. Concentrations in feathers (4-110 ng g^{-1} ww, median ranges) were much higher than those detected in plasma (<LOQ-0.22 ng mL^{-1}) and were not significantly correlated. The authors pointed to atmospheric depositions of OPFRs

447 on the feathers as a potential source of contamination **(Eulaers et al. 2014)**. In another
448 study, OPFRs (TCEP, TCiPP, TPhP, Tris-(2,3-dichloropropyl)-phosphate, TDCiPP)
449 were determined in down and contour feathers of Spanish cinereous vulture *(Aegypius*
450 *monachus)* nestlings and contaminants concentrations were compared between both
451 types of feathers from the same individual **(Monclús et al. 2018)**. They found that
452 concentrations in contour (∑OPFRs median range: 0.4-10.5 ng g^{-1} dry weight dw) and
453 down feathers (∑OPFRs median range: 0.4-13.6 ng g^{-1}dw) were similar. On the basis of
454 these results, a further study on the relationship between corticosterone and OPFR
455 concentrations was assessed and no associations were found **(Monclús et al. 2019).**

456 **Briels et al. 2019** determined concentrations of legacy and NFRs (NBFR:
457 BTBPE, TBB, TBPH; OPFRs: TCEP, TBOEP, TPHP, EHDPHP, TCiPP, TDCiPP; DP)
458 in three non-destructive samples (preen oil, feathers and plasma) of northern goshawks.
459 From all the NFRs analyzed in this study, OPFRs (25.5-206 ng g^{-1}ww; ∑OPFRs median
460 range) were the dominant compounds in feathers and were thought to originate mainly
461 from external deposition, as they were not detected in the other two matrices. Other
462 NFRs (NBFRs and DPs) were generally not detected in the nestlings, suggesting low
463 presence of these emerging contaminants in their environmentand/or low absorption.

464 ***3.1.2.1 Comparison of NFRs and PBDEs.***

465 **Table S2** shows the concentrations of NFRs and PBDEs derived from different
466 studies. Most studies of egg samples collected before 2010, showed higher
467 concentrations of PBDEs than of NFRs. For example, **Vorkampet al. 2017** reported
468 ∑PBDEs and ∑NFRs median ranges of 6-1900 ng g^{-1} and of 0.5-4.7 ng g^{-1}, respectively,
469 in the eggs of peregrine falcons collected from 1986 to 2004 in Greenland. Likewise,
470 **Barón et al. 2015** reported median ranges of ∑PBDEs and ∑NFRs of 20-55 and 17-19
471 ng g^{-1}, respectively) in the eggs of black kites from Spain (1999-2013). In contrast, most

studies based on samples collected after 2010 reported greater concentrations of NFRs than of PBDEs, this being in agreement with the increasing phase out and restrictions imposed on PBDEs **(Briels et al. 2019, Løseth et al. 2019, Blanco et al. 2018, Monclús et al. 2018, Eulaers et al. 2014, Barón et al. 2014).** However, due to their high persistence and bioaccumulation capacity, high PBDE concentrations are still dectected in some European regions. For instance, **Eljarrat et al. 2019** reported ∑NFRs and ∑PBDEs means of 10 and 645 ng g^{-1} lw, respectively, from an unhatched western marsh harrier egg from central Spain. Although only one egg was determined, a high number of samples from other non-raptor species (collected from 2010 to 2017) were analyzed and PBDEs were detected in all of them at high concentration values. Nevertheless, around 50%decline in concentrations of PBDEs was determined **(Eljarrat et al. 2019)**. High concentrations of PBDEs were also reported by **Vetter et al. 2017** in the eggs of peregrine falcons from Germany collected in 2014 (∑PBDEs mean range: 145-320 ng g^{-1}). These concentrations were higher than the NFRs ones (∑NFR mean 1-33 ng g^{-1}).

3.1.3. Other contaminants of emerging concern

Due to the low number of studies (*n*=6) dealing with other emerging contaminant classes in raptors, a thorough analysis of their presence and occurrence trends in Europe was not feasible. Nevertheless, most of these articles were published in the last five years, suggesting a growing interest in their analysis and effects in raptors. Studies of these compounds on non-raptors species are also included for discussion.

Neonicotinoids are insecticides that are widely used for pest control and concerns about negative impacts on non-target organisms have raised in the last years **(Thompson et al. 2020)**. In 2018, the European Commission banned the outdoor use of

clothianidin **(Commission Regulation, 2018/784)**, imidacloprid (IMC) **(Commission Regulation, 2018/783)**, and thiamethoxam **(Commission Regulation 2018/785)** but no regulation have been established for the others yet. Although many studies have reported neonicotinoid bioaccumulation in birds from Europe **(Humann-Guilleminot et al. 2019, Lennon et al. 2019)**, little is known about the accumulation of these contaminants in raptors.In fact, no studies on analysis of neonicotinoids in these species were found beyond Europe.The first evalutation of the exposure of neonicotinoids in raptors was carried out in **Taliansky-Chamudis et al. 2017**. IMC was the only chemical found in the blood from Eurasian eagle owls (*Bubo bubo*) from Spain at a concentration of 3.3 ng mL^{-1}. In a recent study, concentrations of three neonicotinoids were determined in the blood of European honey buzzards (*Pernis apivorus*), but they were found at relatively low concentrations (0.008-0.04 ng mL^{-1}) **(Byholm et al. 2018)**. This low bioaccumulation potential may be related to the fact that raptors are non-insectivorous species and may therefore be less exposed to neonicotinoids. For example, a recent large-scale study on neonicotinoids on avian species in the United States, found a major decline in diversity of insectivore birds feeding in agricultural areas in comparison with non-grassland and non-insectivorous birds **(Li et al. 2020)**. The decline found in this study indicates that birds more exposed to neonicotinoids (as songbirds) might die and therefore, raptors feeding on birds could be feeding on the less contaminated ones. A rapid metabolism or excretion of neonicotinoids in raptors can be another question to consider. However, although insectivorous birds are more exposed to pesticides, another study **(Mineau & Whiteside 2013)** highlighted the possible impact of pesticides on raptors in North America and linked population decline of grassland birds (including some owls and raptors) with lethality of pesticides used.

UV-filters, including benzophenones (BzPs), are a class of CECs that are frequently used in sunscreen products and personal care products **(Asimakopoulos et al. 2014, 2016)**. They are PBT compounds and can contaminate drinking water and migrate from food packaging into food **(Mao et al. 2019)**. Furthermore, they are linked to cancer, endocrine disruption, and organ system toxicity **(Kinnberg et al. 2015, Watanabe et al. 2015).** As a result of this, the use of benzophenone 3 (BzP-3), which is the most represented BzP analogue, is restricted in Europe in cosmetics since 2017 **(Commission Regulation 2017/238)**. Many studies have analyzed BzPs in soil, water, sediments and food **(Tarazona et al. 2010, Sánchez-Brunete et al. 2011, Li et al. 2012)**. However, studies on adverse effects and bioaccumulation of these compounds in wildlife are still scarce. The first study on UV-filters in European raptors was reported by **Molins-Delgado et al. (2017)**. Seven UV-filters (Benzophenone-1 (BzP-1); BzP-3; 4-Hydroxybenzophenone (4-OH-BzP); 4,4′-dihydroxybenzophenone (4-DHB) ;octocrylene (OC); 2-ethylhexyl 4-(dimethylamino)benzoate (ODPABA); 2-(2H-benzotriazole-2-yl)-4-methyl-6-(2-propyl)-phenol (AllylBZP)) were determined in unhatched eggs from Western marsh harriers *(Circus aeruginosus)* and common kestrels from a Spanish preserved area. Mean concentrations ranged between 1.5-210 ng g^{-1}d.w. for common kestrels and between 2.7-895 ng g^{-1}d.w for Western marsh harriers. Furthermore, high detection rates from 90 to 100% were reported for all contaminants, thus indicating bioaccumulation. The rank order of mean concentrations for benzophenone-type UV-filters found in the eggs from the assessed raptor species was 4-OH-BzP> BzP-1 > BzP-3, which was in agreement to the rank found in a recent study on raptor tissues **(González-Rubio et al. 2020).** In the latter study, concentrations on benzophenone-type UV-filters and bisphenols in tissues (kidney, liver, preen gland,

545 brain, muscle and fat) from white-tailed eagles, Eurasian sparrowhawks and long-eared
546 owls from Greenland and France were reported for the first time.
547 Bisphenols (BPs) are another class of CECs that are used in the manufacture of plastic
548 articles **(Asimakopoulos et al. 2016)**. Among BPs, bisphenol A (BPA) is one of the
549 highest production volume chemicals worldwide. However, its production and usage
550 has been regulated due to a raised concern over widespread human exposure and
551 associated health effects **(Bonefeld-Jørgensen et al. 2007)**. The European Union
552 prohibited its use in infant feeding bottles since 2011 **(Commission Regulation**
553 **10/2011).** As a result of this, several BP analogues (such as bisphenol F (BPF);
554 bisphenols S (BPS); and bisphenol AF, (BPAF)) are nowadays introduced onto the
555 market as substitutes **(Chen et al. 2016)**. The occurrence and/or effects of BPs in
556 wildlife have been reported **(Flint et al. 2012, Bhandari et al. 2015)**. Regarding birds,
557 many studies have analyzed these contaminants in domestic (particularly broiler
558 chickens) avian species **(Singh et al. 2016, Shilov et al. 2020)** but few ones have
559 investigated their accumulation on wild birds or raptors. A recent paper is the only peer-
560 reviewed study to document BPA in raptors in Europe and multiresidues of other seven
561 BPs in tissues from white-tailed eagles, long-eared owls and Eurasian sparrowhawks
562 **(González-Rubio et al. 2020)**. BPA and BPF were the dominant contaminant analogues
563 based on the concentrations found in tissues. On the contrary, BPAF and bisphenol Z
564 (BPZ) were not detected in any sample. BPA concentrations have been previously
565 reported in raptor species from outside Europe (**Elliott et al. 2019)** and in non-raptor
566 species (the herring gull, *Larus Argentatus)* in Europe (**Stanizewska et al. 2014,**
567 **Nehring et al. 2017).**
568
569 **Stanizewska et al. (2014)** reported mean concentrations of BPA in the muscle, liver

570 and guano of herring gulls from The Gulf of Gdansk (Southern Baltic) of 62.9, 11 and
571 996 ng g^{-1}d.w., respectively. **Nehring et al. (2017)** also reported BPA concentrations in
572 feathers from the herring gull (ranging on average from 132 to 153 ng g^{-1}d.w.).
573 Regarding analyses carried out in raptors, **Elliott et al. (2019)** reported a median BPA
574 concentration of 1.05 ng/mL in bald eagle (*Haliaeetus leucocephalus*) nestling
575 plasma samples (n=381) from the U.S. In Europe, BPA was detected in preen gland
576 of raptors at concentrations ranging 64.1-149 ng g^{-1}w.w.**(González-Rubio et al., 2020)**.
577 Parabens are a group of substances of emerging concern commonly employed as
578 preservatives, mainly in personal care products, pharmaceuticals and food **(Błędzka et**
579 **al. 2014).** Parabens are believed to exert endocrine disrupting properties **(Vitku et al.**
580 **2018).** They have been linked to breast cancer, skin cancer, and decreased sperm count
581 **(Charles & Darbre, 2013)**. Because of these risks, five different parabens
582 (propylparaben (PrP); butylparaben (BuP); phenylparaben (PhP); benzylparaben, (BnP)
583 and pentylparaben (PeP)) were banned in cosmetic products in the European Union in
584 2014 **(Commission Regulation 358/2014)**, while others are strictly regulated.
585 Methylparaben (MeP) and PrP are the most commonly used parabens and are often used
586 in combination. Although several studies have reported the ubiquitous occurrence of
587 parabens in humans and the environment **(Jiménez-Díaz et al. 2011, Asimakopoulos et**
588 **al. 2014)**, little is known about the accumulation of these chemicals in birds. The only
589 study on parabens that has been carried out in raptors found MeP, ethylparaben (EtP),
590 BuP and heptyl paraben (HeP) concentrations in 20 liver samples of white-tailed sea
591 eagles from the Baltic Sea coast **(Xue & Kannan, 2016)**. MeP was the most abundant
592 compound, as expected, with a detection rate of 75%, at concentrations that ranged from
593 < 8.01 to 657 ng g^{-1}. Furthermore, a very high concentration of 4-hydroxybenzoate (4-
594 HB; 68,600 ng g^{-1}ww) was reported in the liver of one individual, and was detected in

91% of the tissue samples **(Xue & Kannan 2016)**. A major bioaccumulation of 4-HB might be expected, as it is considered to be the final metabolite of parabens in biota **(**Aubert et al., 2012**)**.

Chlorinated paraffins (CPs) are other industrial chemicals of concern with an extensive global use as metal-cutting fluids, flame retardants, plasticizers, and sealants **(Bayen et al. 2006)**. They are highly lipophilic and bioaccumulative in humans and wildlife. In 2012, short carbon chain paraffins (SCCPs) were banned in EU but no regulations for medium carbon chain paraffins (MCCPs) or long carbon chain paraffins (LCCPs) are yet established **(Commission Regulation 519/2012)**. Regarding their accumulation, very little has been studied in raptors so far. A recent study reported concentrations of CPs in the muscle and eggs of white-tailed eagles, eagle owls, golden eagles *(Aquila chrysaetos)*, peregrine falcons, common kestrels, and tawny owls from Sweden, Denmark and the Baltic Sea **(Yuan et al. 2019)**. LCCPs concentrations were highest and predominated (55% of total CPs) in the muscle of peregrine falcons (LCCP median range: 210-1200 ng g^{-1} lw). Another recent study outside Europe **(Fernie et al. 2020)**, described for first time the exposure-related toxicity and potential effects of SCCPs in American kestrels (*Falco sparverius).* Authors observed changes in thyroid function. The potential impact of these changes on thyroid-mediated growth and survival in wild birds requires further investigation.

These results highlight the lack of data on CECs other than NFRs and PFASs. Due to the high concentrations reported for some of these CEC classes and their potential toxicity, further studies are urgently needed to assess contamination concentrations and evaluate the environmental threat they may pose.

3.2 Suitability of sample matrices for the determination of CECs in raptors.

As reviewed above, different sample matrices (e.g., eggs, feathers, preen oil, blood, plasma, serum, and tissues) have been widely collected from different raptor species across Europe to analyse and monitor different types of contaminants.There is not an optimal all-purpose tissue for monitoring contaminants and the scientific relevance of each matrix depends on the aims and objectives of each study and the compound(s) of interest ("fit-for-purpose"). Previous surveys have attempted to show the most suitable matrices for determining major contaminant classes. **Espin et al. (2016)** published a study in which 249 papers were collected to provide information on sample types most widely used in Europe for the determination of certain contaminant groups, such as POPs, PFASs, metals and anticoagulant rodenticides. They concluded that there is no optimal all-purpose tissue and that the scientific relevance of each sample type depends on the aims and objectives of any study and the compound(s) of interest. Although **Espin et al. (2016)** provided useful information regarding the matrices that can be used for POPs, data on raptor matrices used to determine CECs have not been reported yet.

Figure 2 shows the matrices that were predominantly used to measure PFASs, NFRs and other CECs in raptors during the period 2002-2020.

(Insert Figure 2)

Data on other miscellaneous CECs (neonicotinoids, parabens, paraffins, bisphenols and UV-filters) was not representative due to the low number of reported studies (n=6). Plasma, feathers and eggs were the most common sample types used for determining PFASs and NFRs in raptors, as can be observed.

These results are in accordance with previous studies performed on raptors **(Gómez-Ramírez et al. 2014, Espín et al. 2016)** and suggest that eggs and feathers are still the most popular matrices for pollution biomonitoring since their collection and

storage effortis relatively easy (**Espin et al. 2020**). In addition, their use in biomonitoring offers several advantages as will be discussed below.

3.2.1 Eggs

Many organic contaminants are sequestered in eggs during their formation **(Espín et al. 2016)**. For instance, critical concentrations of PFASs have been found in different studies in the last years **(Vorkamp et al. 2019, Holmström et al, 2010)**. PFASs, NFRs and other organic contaminants are transfered to the yolk depending on the amount of lipids invested in a clutch of eggs, which is different depending on the species **(Herzke et al. 2005)**. Another important aspect is that contamination concentrations in eggs are directly related to the concentrations in female raptors and reflect the exposure in this segment of the population. Furthermore, eggs are useful for the assessment of contaminants concentrations over time and space, since they can accumulate contaminants that were assimilated in previous time periods of variable duration by breeding females **(Bourgeon et al. 2013).**

3.2.2 Feathers

Feathers are non-destructive samples that have typically been used for monitoring heavy metal concentrations **(Burger 1993)**. During the last decades their use for biomonitoring organic contaminants is also increasing and has been successfully validated for monitoring internal concentrations of legacy POPs **(Jaspers et al. 2006, 2019)**. Nevertheless, a recent study **(Løseth et al. 2019a)** indicated that the use of feathers for monitoring CECs may be limited. The study of **Løseth et al. (2019a)** was the first that presented a wide investigation of the concentrations of both legacy and CECs (specifically PFRs, NBFRs, PFASs and DPs) in feathers and preen oil in relation to plasma of raptor species from Norway. The authors found good correlations for the

legacy POPs but not for PFASs, NFRs and DPs. It is also worth mentioning that **Eulaers et al. (2014)** found that atmospheric depositions of NFRs on the feathers could cause external contamination. As indicated by **Jaspers et al. 2019**, no studies have been carried out thus far regarding the suitability of feathers for the analysis of other CECs in raptors, such as BPs or UV-filters. However, previous studies have reported concentrations of BPA and alkylphenols in the feathers of a non-raptor species (the herring gull*)* from the Baltic Sea **(Stanizewska et al. 2014; Nehring et al. 2017)**. Feathers were selected as the matrix of study because, although BPs and alklyl phenols are generally accumulated by living beings through the dietary intake **(Xiao et al. 2006, American Chemistry Council 2016)**, their corresponding elimination from the organisms can be achieved through their incorporation into feathers and also laying eggs **(Grajewska et al. 2015)**. The results showed that the contamination in the herring gull feathers depended on the inhabited area and, consequently on the diet of this species. A diet rich in fish manifested elevated BPA concentrations in covert feathers (29-512 ng g^{-1}).

3.2.3 Blood

There is a limited number of articles that use whole blood to determine the target contaminants **(Byholm et al. 2018, Taliansky-Chamudis et al. 2017)**. Although blood is a non-lethal sampling matrix, sample volume is sometimes limited by body size. Furthermore, contaminant half-lives are relatively short in blood and contaminant concentrations are generally lower and more variable than in other matrices. For lipophilic substances, such as POPs, low concentrations are expected in blood since the lipid content in this biological fluid is relatively low compared to other commonly

694 sampled tissues such as eggs. Studies in which analysis were carried out in whole blood
695 were only found for neonicotinoids, which are polar insecticides, but not for PFASs or
696 NFRs **(Figure 2)**. In all cases, small volumes of bird blood were used (~100-500 µL)
697 and low limits of quantification were obtained (at the pg ml^{-1} range). Most of the studies
698 analyzing PFASs in raptors have used plasma as preferred matrix due to their protein
699 affinity. Concentrations of studied PFASs in whole blood are approximately half those
700 in plasma **(Ehresman et al. 2007)**. This difference is attributed to volume displacement
701 by red blood cells (and that fluorochemicals are potentially not present intracellularly
702 **(Ereshman et al. 2007**). Apart from PFASs, plasma has also been used for the
703 determination of NFRs (**Løseth et al. 2019a, Eulaers et al. 2011)**, because most lipid
704 soluble contaminants are attached to the triglycerides present in this matrix. Another
705 thing to consider is that although lipophilic contaminants do not build up in blood, they
706 still circulate in the blood, and these concentrations may be more representative of
707 recent exposure, either via diet or recent mobilization from lipid stores if a bird is in a
708 fasted state for any reason.
709
710 **3.2.4 Internal tissues**
711 Studies on internal organs and tissues are less common and limited to PFASs
712 and other CECs analysis (see **Figure 2**), since most raptors are protected species. Thus,
713 the collection of these matrices for biomonitoring purposes is based mainly on
714 carcasses. Liver and fat were the most analyzed matrices since non-polar compounds
715 are mainly accumulated in matrices that contain a high proportion of triglycerides
716 **(Jaspers et al. 2013, Eulaers et al. 2014)**.
717 **González-Rubio et al. (2020)** reported that the liver, kidney and preen gland
718 tissues from raptors were suitable for the biomonitoring of BPs and BzP contaminants.

719 Furthermore, a greater bioaccumulation of these chemicals in preen gland was found
720 and a need for further investigation of the role of this matrix as a potential excretory
721 organ for CECs was highlighted.
722
723 **3.3 Selection of raptor species for biomonitoring CECs**
724
725 Most studies on CECs in raptors in Europe have been performed on eagles, owls
726 and *Accipiters* which are at the top of the food chain and therefore, may be most
727 susceptible to effects. Depending on the species, raptors may be migratory and therefore
728 it is often difficult to know where they incorporated contaminants during the annual
729 cycle **(Badry et al. 2020)**. Furthermore, the choice of species and associated traits (such
730 as foraging in terrestrial or freshwater habitats) need to be matched to the fate pathways
731 of the compounds of interest. Thus, the accumulation of different POPs and CECs in
732 different raptor species may vary. The selection of species for previous biomonitoring
733 studies was mainly performed based on the abundance, geographical distribution and
734 conservation status. Recently **Badry et al. (2020)** proposed a selection for raptor
735 species for monitoring priority pollutants in Europe (mercury, lead, anticoagulant
736 rodenticides, pesticides and medicinal products) based on information on the
737 distribution and key ecological traits (food web, foraging trait, diet, preferred habitat,
738 and migratory behaviour). They identified the common buzzard (*Buteo buteo*) and
739 tawny owl (*Strix aluco*) as good biomonitoring species for these pollutant classes. Other
740 factors affect this selection, as the difficulty to capture them alive for blood sampling or
741 the restriction on handling protected raptors (for which only addled eggs or those found
742 dead can be used). Studies are usually based on samples that have been previously
743 collected for other purposes.

744

745 The raptor species collected for analyzing PFASs, NFRs and other CECs are
746 demonstrated in **Figure 3**.

747

748 **(Insert Figure 3)**

749

750 Amongst raptors, the white-tailed eagle, the northern goshawk and the peregrine
751 falcon, were the most frequently studied. The white-tailed eagle was also the most
752 analyzed species for PFASs determination. Amongst owls, the tawny owl was the most
753 studied for the determination of PFASs and other CECs. No data on common buzzard
754 are yet available for CECs in Europe, and therefore it would be interesting to investigate
755 the suitability of this species as it was identified as a good European wide biomonitor
756 for the priority compounds proposed by **Badry et al. (2020)**.

757

758 **3.4. Preferred bioanalytical methods for the analysis of CECs in raptors**

759 The development of efficient and sensitive analytical methods is crucial for
760 correctly assessing the exposure to organic contaminants in wildlife. The analytical
761 methodologies for the determination of emerging contaminants in the reviewed studies
762 are mainly based on sample preparation step (extraction and clean-up with specific
763 solvents or adsorbents) and subsequent analysis commonly based on liquid (LC) or gas
764 chromatography (GC) coupled to mass spectrometry (MS) of low and high resolution.
765 The frequency of use of different sample pretreatments (extraction and/or clean-up) per
766 contaminant class per matrix, respectively, is presented in **Figures 4 and 5**.

767

768 **(Insert Figures 4 and 5)**

Conventional solvent extraction (thus including liquid-liquid and solid-liquid extraction, depending on the sample analyzed) was preferred for the determination of NFRs and was the only method employed for PFASs. Due to the non-polar nature of NFRs, the most common organic solvents were mixtures of hexane and dichloromethane **(Briels et al. 2019, Monclús et al. 2018)**. For PFASs (more polar substances), methanol or acetonitrile were the preferred choice **(Løseth et al. 2019, Eriksson et al. 2016)**. Conventional solvent extraction was predominantly applied to eggs and it was the only method applied in plasma, feathers, tissues and other matrices such as fat or preen oil. Other extraction techniques were also reported namely pressurized liquid extraction (PLE) **(Barón et al. 2014)**, soxhlet extraction **(Vorkamp et al. 2017)** and QuEChERS (salting-out extraction and simultaneous dispersive SPE clean-up) for the determination of neonicotinoids in blood samples **(Byholm et al. 2018, Taliansky-Chamudis et al. 2017)**.

As it can be observed in **Table 3**, the application of classical extraction techniques for the determination of other CECs in eggs, such as Soxhlet, has declined since 2012 in favor of more modern approaches, e.g. PLE, since lower solvent consumption and rapid and simpler procedures are obtained. In general, Soxhlet was applied using dichloromethane **(Guerra et al. 2011)** and hexane:acetone (4:1, v/v) (**Vorkamp et al. 2019, Vorkamp et al. 2017)** as extraction solvents, while PLE was carried out with hexane:dichloromethane (1:1, v/v) (**Blanco et al. 2018)**. QuEChERS extraction step,which is usually based on a salting-out process with acetonitrile and that is widely employed for enhancing the extraction of polar and medium polar compounds,

793 was applied to determine neonicotinoids in blood **(Taliansky-Chamudis et al. 2017,**
794 **Byholm et al. 2018)**.

795 The amount of extracted sample was highly variable and depended on the
796 method and the matrix. For solid samples, these amounts varied from 0.1 g (dw) **(Vetter**
797 **et al 2017)** to 10 g (lw) of egg media **(Yuan et al. 2019)** and, for liquid samples from
798 0.1 mL of blood **(Byholm et al. 2018)** to 1 mL of plasma **(Eulaers et al. 2014)**.

799 Further clean-up after extraction was usually necessary. For PFASs, clean-up
800 consisted always of dispersive-SPE using acidified ENVI-Carb as sorbent. On the
801 contrary, different clean-up procedures were developed for the determination of PBDEs
802 and NFRs, such as SPE using ENVI-Florisil (especially in recent years) or acidified
803 alumina cartridges **(Monclús et al. 2019, Blanco et al. 2018)**. One study also proposed
804 gel permeation chromatography on a Bio Beads SX-3 (packed with neutral, porous
805 styrene divinyl benzene copolymer beads) **(Guerra et al. 2011)**. Regarding other CECs,
806 dispersive-SPE clean-up (by using magnesium sulphate, primary secondary amine
807 (PSA) and polymerically bonded trifunctional C_{18} silica sorbent) was applied for
808 neonicotinoids while a QuEChERS method (based on a mixture of PSA, C_{18} and
809 graphitized carbon black (GCB)) and SPE (C_{18} or Oasis HLB cartridges) was used for
810 UV-filters and bisphenols. PSA can eliminate organic and fatty acids, C_{18} removes fats,
811 sterols and other nonpolar interferences and GCB eliminates pigments and sterols.

812 Regarding the instrumental analysis for NFRs, GC-MS on a DB-5 capillary
813 column was the most reported technique, with limits of quantification (LOQs) ranging
814 from 0.002 to 32.2 ng g^{-1}(or mL^{-1}). Regarding the analysis of PFASs, LC-MS-MS and
815 LC-time-of-flight (TOF)-high resolution MS with a C_{18} column were the most applied
816 techniques, with LOQs ranging from 0.004 to 0.4 ng g^{-1} or ng ml^{-1}. LOQs were very
817 low in the case of UV-filters (0.5-20 pg g^{-1}) **(Molins-Delgado et al. 2017)** and

neonicotinoids (0.6-30 pg mL^{-1}) **(Byholm et al. 2018)**, potentially due to the combination of selective and sensitive sample treatment and analysis, namely PLE coupled to LC-QTRAP-MS-MS or QuEChERS coupled to LC-MS-MS, respectively. The development of new analytical methods for CECs is desirable to decrease the limits of detection and quantification since they are usually found in trace concentrations, and for expanding the applicability in a variety of matrices.

Conclusions

In this study, a comprehensive and critical review concerning the presence and analysis of PFASs, NFRs and other CECs in European raptors (birds of prey and owls) was presented. It was concluded that in Europe, most of the studies on CECs were perfomed in Western Europe, mainly in Norway and Spain.Most temporal trend studies on PFASs in European raptor eggs reported increasing concentrations of PFCAs over time and inconclusive results for PFOS. Furthermore, concentrations of PFASs in plasma and feathers were greater than for legacy PBDEs in most cases. Regarding the occurrence of NFRs, most studies in which raptor samples (eggs, feathers and plasma) were collected after 2010 reported greater concentrations than PBDEs, this being in accordance with the phase out and restrictions imposed on these persistent organic pollutants. Likewise, the detection of other miscellaneous CECs (including neonicotinoids, UV-filters, chlorinated paraffins and bisphenols) in raptors and the low data available in the literature suggests an urgent need to collect more information on these compounds in order to evaluate the environmental threat they may pose.

White-tailed Eagle, Peregrine falcon, Northern goshawk and tawny owl were the most frequently studied raptor species in Europe. However, studies on CECs in common buzzard are currently lacking and could be recommended for investigation as this species, along with tawny owl, has also been proposed as a European wide biomonitoring for priority contaminants in a recent study **(Badry et al. 2020)**. Regarding the matrices, eggs, and blood plasma can be recommended for PFASs analysis in raptors, while studies on different matrices for other CECs are limited.There is not an optimal all-purpose tissue for monitoring contaminants in raptors and the selection of the matrix depends on the objectives of each study, the conservation status of the species regarding sampling permits and the compound(s) of interest. Feathers are a popular non-destructive sampling matrix for raptors, but as highlighted by recent studies feathers may not be suitable for some CECs and specific attention is needed to potential external contamination on the feather surface. A recent study highlighted the potential role of the preen gland as a major excretory organ for CECs, which can be further investigated as a potential matrix for contaminants in raptors. Regarding sample treatment and analysis, conventional solvent extraction prior to both GC and LC coupled to mass spectrometry were the most used sample treatment and instrumental technique(s), respectively. Alternative sample treatments (e.g. QuECheRS or pressurized liquid extraction) have been only scarcely investigated in this field. Further analytical developments are expected in the future to increase the scope of the analysis and to improve the analytical features of the methods, such as extraction efficiency and clean-up, key factors in these complex and diverse matrices.The use of high-resolution MS will probably also be an increasing trend, since it opens the possibility of non target screening and the identification of unknown substances or untargeted species by retrospective screening.

The results of this study highlight the need for further research on CECs in other European regions (mainly Eastern Europe), with the aim to improve the assessment of temporal and spatial trends. It is also advisable to further examine suitable matrices and other species and to develop new and sensitive analytical strategies for their analysis (especially for CECs) in raptors. Finally, the toxicological effects and ecological impact of the exposure to CECs on birds of prey and owls in Europe have not received much attention thus far and this would be essential to guide which CECs should be investigated in future research

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Figure captions

Figure 1. Maps showing the locations of the studies performed on PFASs, NFRs and other CECs on raptors (birds of prey and owls) in Europe (2002-2020).

Figure 2. Number of studies on PFASs, NFRs and other CECs on different raptor matricesin Europe (2002-2020). Preen oil and adipose tissue were referred to as “other”.

Figure 3. Number of studies on PFASs, NFRs and other CECs on different raptor species in Europe (2002-2020).

Figure 4. Number of studies on PFASs, NFRs and other CECs in Europe (2002-2020) according to the sample treatment strategy.

Figure 5. Number of studies on PFASs, NFRs and other CECs on raptors in Europe (2002-2020) using different sample treatment strategies according to the sample matrix. Preen oil and adipose tissue were referred to as “other”.

Table 1. Contaminant abbreviations used and ordered alphabetically.

Abbreviation	Contaminant
ACE	Acetamiprid
AllylBZT	2-(2H-benzotriazole-2-yl)-4-methyl-6-(2-propyl)-phenol
ATE	Allyl-2,4,6-tribromophenyl ether
BATE	2-Bromoallyl-2,4,6-tribromophenyl ether
BnP	Benzyl paraben
BP	Bisphenol
BPA	Bisphenol A
BPAF	Bisphenol AF
BPAP	Bisphenol AP
BPB	Bisphenol B
BPF	Bisphenol F
BPM	Bisphenol M
BPP	Bisphenol P
BPS	Bisphenol S
BPZ	Bisphenol Z
Br-PFOS	Branchedperfluorooctanesulfonate
BTBPE	1,2 Bis(2,4,6-tribromophenoxy)ethane
BuP	Butyl paraben
BzP	Benzophenone
BzP-1	Benzophenone-1
BzP-2	Benzophenone-2
BzP-3	Benzophenone-3
BzP-8	Benzophenone-8
DBDPE	Decabromodiphenylethan
Dec	Dechlorane
3,4-DHB	3,4-dihydroxybenzoic acid
4-DHB	4,4-Dihydroxybenzophenone
DP	Dechlorane Plus
DPTE	2,3-Dibromopropyl-2,4,6-tribromophenyl ether
EHDPP	2-Ethylhexyl diphenylphosphate
EtP	Ethylparaben
4-HB	4-Hydroxy benzoate
HBB	Hexabromobenzene
HBCD	Hexabromocyclododecane
HeP	Heptylparaben
IMC	Imidacloprid
LCCP	Long chainchlorinatedparaffin
Lin-PFOS	Linear perfluorooctanesulfonate
MCCP	Medium chainchlorinatedparaffin
MeP	Methylparaben
MPFOA	Perfluoro-n-(1, 2, 3, 4-13C4) octanoicacid
MPFOS	Perfluoro-1-(1, 2, 3, 4-13C4) octanesulfonicacid
NBFRs	Novel brominated flame retardants
NFRs	Novel flame retardants
NP	4-nonylphenol
OC	2-ethylhexyl 2-cyano-3,3-diphenyl acrylate
ODPABA	2-ethylhexyl 4-(dimethylamino) benzoate
4-OH-BzP	4-Hydroxybenzophenone
OH-EtP	Ethylprotocatechuate
OH-MeP	Methylprotocatechuate
OP	4-tert-octylphenol
OOPFRs	Organophosphorus flame retardants
PBEB	Pentabromoethylbenzene
PeBT	Pentabromotoluene
PCNs	Polychlorinated naphthalenes

PeP	Pentylparaben
PFASs	Per- and Polyfluoroalkyl substances
PFBA	Perfluorobutanoicacid
PFBS	Perfluorobutanesulfonate
PFCAs	Perfluoroalkyl carboxylic acids
PFDcA	Perfluorodecanoanoicacid
PFDcS	Perfluorodecanesulfonate
PFDoA	Perfluorododecanoicacid
PFHpA	Perfluoroheptanoicacid
PFHpS	Perfluoroheptanesulfonate
PFHxA	Perfluorohexanoicacid
PFHxS	Perfluorohexanesulfonate
PFNA	Perfluorononanoicacid
PFNS	Perfluorononanesulfonate
PFOA	Perfluorooctanoicacid
PFOS	Perfluorooctanesulfonate
PFOSA	Perfluorooctanesulfonamide
PFPA	Perfluoropentanoicacid
PFPeA	Pefluoropentadecanoicacid
PFPS	Perfluoropentanesulfonate
PFTeA	Perfluorotetradecanoicacid
PFTriA	Perfluorotridecanoicacid
PFUnA	Perfluoroundecanoicacid
PFSAs	Perfluoroalkyl sulfonates
PhP	Phenylparaben
PrP	Propylparaben
SCCP	Short chainchlorinatedparaffin
2-4-6-TBA	2-4-6-Tribromoanisole
TBB	2-Ethylhexyl-2,3,4,5-tetrabromobenzoate
TBOEP	Tri-(2-butoxyethyl)-phosphate
2-4-6 TBP	2,4,6-Tribromophenol
TBPH	3,4,5,6-Tetrabromophthalate
TCEP	Tris(chloroethyl)phosphate
TCiPP	tris (1-chloro-2-propyl) phosphate
TCPP	tris(2-chloroisopropyl) phosphate
TDCiPP	Tris-(2,3-dichloropropyl)-phosphate
THC	Thiacloprid
TPhP	Tris(phenyl)phosphate

Table 2. Raptor species and number of analyzed individuals (sample population), countries where the studies were performed, matrices and concentrations of CECs in European raptors (2002-2020). The studies are ordered by year of publication, starting with the most recent ones.

Species (sample size)	**Country/ sampling year**	**Contaminants**	**Matrix**	**Concentration range ($ng\ g^{-1}$ or $ng\ mL^{-1}$)**	**Concentration median or mean ($ng\ g^{-1}$ or $ng\ mL^{-1}$)**	**Reference**
White-tailed eagle *(Haliaeetus albicilla)* (n=16) Eurasian sparrowhawk *(Accipiter nisus)* (n=2) Long-eared owl *(Asiootus)* (n=2)	Greenland and France/ 1997-2011	BPs: BPA, BPAF, BPAP, BPF, BPB, BPM, BPP, BPS. BzPs: BzP-1, BzP-2, BzP-3, BzP-8, 4-OH-BzP.	Preen gland	BPs: 0.2-149 (w.w.) BzPs: 0.3-7 (w.w.)	BPs median range: 0.05-70.9 (w.w.) BzPs median range: 0.3-47 (w.w.)	González-Rubio et al. (2020)
			Liver	BPs: 0.05-5 (w.w.) BzPs:0.9-44 (w.w.)		
			Kidney	BPs: 0.02-7 (w.w.) BzPs: 0.7-8 (w.w.)		
			Muscle	BPs: 0.1-3.5 (w.w.) BzPs: 0.3-47 (w.w.)		
Northern goshawk (*Accipiter gentilis*) (n=61)	Norway/ 2015-2016	PFASs: -PFCAs: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA, PFHxDcA -PFSAs: PFBS, PFPS, PFHxS, PFHpS, PFOS, PFDcS -PFOSA NFRs: -DP -NBFRs: BTBPE, TBB, TBPH -OPFRs: TCEP, TBOEP, TPHP, EHDPHP, TCIPP, TDCIPP	Feathers	∑PFASs: 1.9-12.5 (Trøndelag, 2016); 0.3-9.6 (Troms, 2016); (w.w.) ∑NFRs: 17-203(Trøndelag, 2015); 2.6-41.1 (Trøndelag, 2016); 135-314 (Troms, 2015); 8-72 (Troms, 2016); (w.w.)	∑PFASs: mean (median): 5.7 (6.2) (Trøndelag, 2016); 1.8 (1.8) (Troms, 2016); (w.w.) ∑NFRs mean (median): 46.1 (47.7) (Trøndelag, 2015); 19.4 (22.2) (Trøndelag, 2016); 204 (206) (Troms, 2015); 27.5 (25.5) (Troms, 2016); (w.w.)	Briels et al. (2019)

			Plasma	∑PFASs: 2.-36(Trøndelag, 2015); 1.4-23.8 (Trøndelag, 2016); 1.4-28.4 (Troms, 2015); 2-16 (Troms, 2016) ∑NFRs: n.d.	∑PFASs: 14.6 (17.5) (Trøndelag, 2015); 6.3 (6.6) (Trøndelag, 2016); 8.8 (9.2) (Troms, 2015); 6.3 (5.0) (Troms, 2016) ∑NFRs: n.d.	
Western marsh harrier (*Circus aeruginosus*) (n=1)	Spain/ 2010	NFRs: Dec, DP	Eggs	NFRs:-	∑NFRs: 10 (l.w.)	Eljarrat et al. (2019)
White-tailed eagle (*Haliaeetus albicilla*) (n=70)	Norway/ 2015-2016	PFASs: -PFCAs: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDcA PFUnA, PFDoA, PFTrA, PFTeA, -PFSAs: PFBS, PFPS, PFHxS, PFHpS, Lin-PFOS, Br-PFOS, PFNS -PFOSA NFRs: -NBFRs: TBB, TBPH, BTBPE -OPFRs: TCEP, TBOEP, EHDPHP, TPHP, TCPP, TCIPP, TDCIPP -DPs	Plasma	PFUndA: 2.4-4.4 (Smøla, 2015); 0.7-2.1 (Smøla, 2016); 2.3-5.1 (Steigen, 2015); 0.9-2.2 (Steigen, 2016) ; (w.w.) NFRs: 0.0002-0.3 (Smøla, 2015); 0.002-0.1 (Smøla, 2016); 0.01-1.4 (Steigen, 2015); 0.01-0.08 (Steigen, 2016); (w.w.)	PFUndA (median): 3.59 (Smøla, 2015); 1.15 (Smøla, 2016); 3.36 (Steigen, 2015); 1.40 (Steigen, 2016); (w.w.) NFRs median range: 0.01-0.2 (Smøla, 2015); 0.002-0.1 (Smøla, 2016); 0.01-0.55 (Steigen, 2015); 0.01-0.08 (Steigen, 2016); (w.w.)	Løseth et al. (2019a)
			Feathers	PFUndA: 0.05-0.6 (Smøla, 2015); 0.07-1 (Smøla, 2016); 0.1-0.8 (Steigen, 2015); 0.3-1.1 (Steigen, 2016); (w.w.) NFRs: 0.1-1229 (Smøla, 2015); 0.1-168 (Smøla, 2016); 0.0001-63 (Steigen, 2015); 0.1-68.4 (Steigen, 2016); (w.w.)	PFUndA (median): 0.15 (Smøla, 2015); 0.58 (Smøla, 2016); 0.38 (Steigen, 2015); 0.28 (Steigen, 2016); (w.w.) NFRs median range: 0.1-21.6 (Smøla, 2015); 0.1-27 (Smøla, 2016); 0.1-14 (Steigen, 2015); 0.2-21 (Steigen, 2016); (w.w.)	

White-tailed eagle (*Haliaeetus albicilla*) (n=70)	Norway/ 2015-2016	PFASs: -PFCAs: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTrA, PFTeA PFSAs: PFBS, PFPS, PFHxS, PFHpS, Lin-PFOS, Br-PFOS, PFNS - PFOSA	Plasma	∑PFASs: 10-47 (Smøla, 2015); 5-13 (Smøla, 2016); 18-53 (Steigen, 2015); 7-33 (Steigen, 2016)	∑PFASs median: 26 (Smøla, 2015); 9 (Smøla, 2016); 32 (Steigen, 2015); 13 (Steigen, 2016)	Løseth et al. (2019b)
Cinereousvulture (*Aegypius monachus*) (n=16)	Spain/ 2016	NFRs: - OPFRs: TCEP, TCIPP, TPHP, TDCIPP	Feathers	∑NFRs: 1-65; (d.w.)	∑NFRs: 18 (13); (d.w.)	Monclús et al. (2019)
Egyptian vulture (*Neophron percnocterus*) (n=77)	Spain/ 2012-2016	PFASs: -PFCAs: PFBA, PFOA, PFNA, PFDA, PFUnA, PFDoA ,PFTrA, PFTeA -PFSAs: PFHxS, PFOS -MPFOA, MPFOS	Plasma	-	PFASs: 0.03-2.3; (w.w.)	Ortiz-Santaliestra (2019)
White tailed-eagle (*Haliaeetus albicilla*) (n=147)	Norway, Sweden and Greenland/ 1968-2015	PFASs: -PFCAs: PFHxA, PFHpA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTrA,,PFTeA -PFSAs: PFBS, PFHxS, PFHpS, PFOS, PFDS -PFOSA	Feathers	-	∑PFCAs mean (median) range: 1.8-6.5 (2-6.1) (Norway); 0.2-8.7 (0.1-9) (Sweden); 3.1-9 (3.1-8.2) (Greenland) PFOS: 2.5-11.6 (1.6-9.5) (Norway); 3-22.3 (3-20.1) (Sweden);2.7-4.1 (2.6-4) (Greenland)	Sun et al. (2019)

					FOSA: 0.7-3.6 (0.7-3.5) (Norway); 0.4-1.6 (0.3-1.5) (Sweden); 1.3-8.3 (1.1-8.3) (Greenland)	
Peregrine falcon (*Falco peregrinus*) (n=26)	Greenland/ 1986-2014	PFASs: -PFCAs: PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA ,PFTrA, PFTeA -PFSAs: PFBS, PFHxS, PFHpS, PFOS, PFDS -PFOSA PCNs: CN 42-52/60, 53, 54, 63, 66/67, 68/64, 69	Eggs	∑PFSAs: 113-3900; (d.w.) ∑PFCAs: 29-410; (d.w.) ∑PCNs: 7-493; (d.w.)	∑PFSAs mean (median): 474 (303); (d.w.) ∑PFCAs: 138 (100); (d.w.) ∑PCNs: 40 (21); (d.w.)	Vorkamp et al. (2019)
White-tailed eagle (*Haliaaetus Albicilla)* (n=4) Eagle owl (*Bubo bubo*) (n=10) Golden eagle (*Aquila chrysaetos*)	Sweden, Denmark and the Baltic Sea (Poland)/ 2012-2017	Chlorinated paraffins: SCCPs, MCCPs, LCCPs.	Muscle	-	SCCPs: median range: 540-730; (l.w.) MCCPs: 360-720; (l.w.) LCCPs: 210-1200; (l.w.)	Yuan et al. (2019)

(n=10) Peregrine falcon (*Falco peregrinus)* (n=10) Common kestrel (*Falco tinnunculus*) (n=3) Tawny owl (*Strix aluco)* (n=4)			Eggs	-	SCCP: median range: 85-220; (l.w.) MCCPs: 85-250; (l.w.) LCCPs: 34-51; (l.w.)	
Black kite (*Milvus migrans*) (Madrid, n=28; Doñana, n=23)	Spain/ 2007-2016	NFRs: -Dec:Dec 602, 603, 604 -DP:*syn*-DP, *anti*-DP	Eggs	NFRs: n.d.-33 (Doñana); n.d.-2452 (Madrid); (l.w.)	NFRs: n.d.-11 (Doñana); n.d.-353 (Madrid); (l.w.)	Blanco et al. (2018)
European honey buzzard (*Pernis apivorus*) (n=10)	Finland/ 2013	Neonicotinoids: ACE, IMC, THC	Blood	ACE: <LOD-0.008 IMC: <LOD-0.04 THC: <LOD-0.03	-	Byholm et al. (2018)
Cinereousvulture (*Aegypius monachus*) (n=16)	Spain/ 2016	NFRs: - OPFRs: TCEP, TCiPP, TPhP, TDCiPP	Contourf eathers	∑NFRs: 0.4-24.6; (d.w.)	∑NFRs: 0.4-10.5 (d.w.)	Monclús et al. (2018)
			Down feathers	∑NFRs: 0.4-49 ; (d.w.)	∑NFRs: 0.4-13.6 ; (d.w.)	

Northern goshawk (*Accipiter gentilis)* (n=11) White-tailed Eagle (*Haliaeetus albicilla)* (n=14)	Norway/ 2014	PFASs: -PFCAs: PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA, -PFSAs: PFBS, PFHxS, lin-PFOS, br-PFOS, PFDcS - PFOSA	Plasma	∑PFASs: 5-42 (*Accipiter gentilis*); 28-79 (*Haliaeetus albicilla*)	∑PFASs mean (median): 19 (17) (*Accipiter gentilis*); 45 (46) (*Haliaeetus albicilla*)	Gómez-Ramírez et al. (2017)
			Feathers	∑PFASs: 5-26 (*Haliaeetus albicilla*); (w.w.)	∑PFASs mean (median): 13 (12) (*Haliaeetus albicilla*); (w.w.)	
Common kestrel (*Falco tinnunculus*) (n=10)	Spain/ 2010-2012	UV-filters: BP1, BP3, 4HB, 4DHB, OC, ODPABA, AllylBZT	Eggs	BP1: 28-54; (d.w.) BP3: 19-35; (d.w.) 4HB: 20-1200; (d.w.) 4DHB: <MLOQ-132; (d.w.) AllylBZT: 0.4-3; (d.w.)	BP1 mean: 39 (d.w.) BP3: 25 (d.w.) 4HB: 210 (d.w.) 4DHB: 132 (d.w.) AllylBZT: 1.5 (d.w.)	Molins-Delgado et al. (2017)
European herring gull (*Larus argentatus*) (non raptor species) (n=26)	Poland/ 2010-2012	BPA OP NP	Feathers	BPA: 29-512; (d.w.) OP: 28-564; (d.w.) NP: 5-151; (d.w.)	BPAmean: 145 (d.w.) OP: 162 (d.w.) NP: 38 (d.w.)	Nehring et al. (2017)
Eurasian eagle-owl (*Bubo bubo*) (n=30)	Spain/ 2004-2009	Neonicotinoids: ACE, IMC, THC, Clothianidin, Dinotefuran, Nitenpyram, Thiamethoxam	Blood	IMC: 3.3 Other neonicotinoids: n.d.	-	Taliansky-Chamudis et al. (2017)

Peregrine falcon (*Falco peregrinus)* (n=11) Eurasian eagle-owl (*Bubo bubo*) (n=3) White-tailed Eagle (*Haliaeetus albicilla)* (n=1) Osprey (*Pandion haliaetus*) (n=2)	Germany/ 2014	NFRs: - NBFRs: DPTE, BATE, ATE, HBCD, HBB, 2-4-6-TBA, 2-4-6 TBP, PBEB, PBT	Eggs	NFRs: 0.03-66 (*Falco peregrinus*); 0.6-81 (*Bubo bubo)*; 2-11 (*Haliaeetus albicilla*); 1-9 (*Pandion haliaetus*); (l.w.)	NFRs mean range: 1-33 (*Falco peregrinus*); 2-24 (*Bubo bubo)*; 5 (*Haliaeetus albicilla*); 3-5 (*Pandion haliaetus*); (l.w.)	Vetter et al. (2017)
Peregrine falcon (*Falco peregrinus)* (n=10)	Greenland/ 1986-2014	NFRs: TBB, BTBPE, DPTE, DBDPE, DP	Eggs	∑NFRs: 0.2-54; (l.w.)	NFRs: 0.5-4.7 (l.w.)	Vorkamp et al. (2017)
Osprey (*Pandion haliaetus*) (n=30) Tawny owl (*Strix aluco)* (n=10) Common kestrel (*Falco tinnunculus*) (n=40)	Sweden/ 1997-2014	PFASs: -PFCAs: PFBA, PFHxA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, -PFSAs: PFHxS, PFOS, -PFOSA	Eggs	PFASs: 0.08-22 (*Falco tinnunculus*, 2014); 0.08-35 (*Strixaluco*, 2014); 0.08-224 (*Pandion haliaetus*, 2013); 0.3-337 (*Pandion haliaetus*, 2008-2009); 0.08-667 (*Pandion haliaetus*, 1997-2001); (w.w.)	PFASs median range: 0.3-4 (*Falco tinnunculus*, 2014); 0.3-8 (*Strix aluco*, 2014); 1-70 (*Pandion haliaetus*, 2013); 0.5-64 (*Pandion haliaetus*, 2008-2009); 0.15-103 (*Pandion haliaetus*, 1997-2001); (w.w.)	Eriksson et al. (2016)

White tailed eagle (*Haliaeetus albicilla*) (n= -)	Sweden/ 1966-2010	PFAS: -PFCAs:PFHxA, PFOA, PFNA, PFDcA, PFUnA, PFDoA,PFTrA, PFTeA, and PFPeA -PFSAs: PFBS, PFHxS, PFOS, PFDS -PFOSA	Eggs	-	PFASs geometric mean range: 0.005-1513 (Location 1 from 1969 to 2010); 0.009-837 (Location 2 from 1966 to 2010); 0.007-370 (Location 3 from 1986-2010); 0.006-174 (Location 4 from 1966-2009); (w.w.)	Faxneld et al. (2016)
White-tailed eagle (*Haliaeetus albicilla*) (n=35)	Norway/ 2011-2012	PFASs: -PFCAs: PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA, -PFSAs: PFBS, PFHxS, Lin-PFOS, Br-PFOS	Plasma	∑PFASs: 1-14	∑PFASs mean (median): 50 (42)	Sletten et al. (2016)
White-tailed eagle (*Haliaeetus albicilla*) (n=20)	Germany and Polish coastal areas of the Baltic Sea/ 1979-1999	Parabens: MeP, EtP, PrP, BuP, BnP, HepP, OH-MeP, OH-EtP, 3,4-DHB, 4-HB	Liver	MeP:<8.-657; (w.w.) 4-HB: 545-68600 Others: n.d.	MeP: mean: 112 4-HB: 11500 Others: n.d	Xue&Kannan (2016)
Black kite (*Milvus migrans*)	Spain/ 1999-2013	NFRs: -Dec:Dec 602, 603, 604	Eggs	∑NFRs: 1-76 (1999); 5-74 (2011); (l.w.)	∑NFRs median: 17 (1999); 19 (2011); (l.w.)	Barón et al. (2015)

(n=10)		-DP				
Black kite (*Milvus migrans*) (n=22) Red kite (*Milvus milvus*) (n=2) Western marsh harrier (*Circus aeruginosus*) (n=1) Booted eagle (*Aquila pennata*) (n=6) Common kestrel (*Falco tinnunculus*) (n=13) Black-winged kite (*Elanus caeruleus*) (n=1) Barn owl (*Tyto alba*) (n=1)	Spain/ 2010-2012	NFRs: Dec:Dec 602, 603, 604 DP:*anti*-DP, *syn*-DP	Eggs	ΣNFRsorNFRs(l.w.): 7.5-74.4 (*Milvus migrans*);43.2-48.7 (*Milvus milvus*); 0.69-88.9 (*Circus aeruginosus*); 3.6-83.8 (*Aquila pennata*); 1-19.6 (*Falco tinnunculus*); 0.6-20.9 (*Elanus caeruleus*); 0.4-5.4 (*Tyto alba*) .	ΣNFRs mean (l.w.): 30.9 (*Milvus migrans*);46 (*Milvus milvus*); 161 (*Circus aeruginosus*); 29.9 (*Aquila pennata*); 8.8 (*Falco tinnunculus*); 22.6 (*Elanus caeruleus*); 7.3 (*Tyto alba*) .	Barón et al. (2014)
Tawny owl (*Strix aluco*) (n=107)	Norway/ 1986-2009	PFASs: -PFCAs: PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA-PFSAs: PFHxS, PFHpS, PFOS, PFDcS -PFOSA	Eggs	∑PFASs: 0.06-6 (1986-2009); 0.06-6 (1986-1995); 0.06-5 (2001-2009); (w.w.)	∑PFASs mean (median): 2 (1) (1986-2009); 1 (0.9) (1986-1995); 2 (1.8) (2001-2009); (w.w.)	Bustnes et al. (2014)

Northern goshawk (*Accipiter gentilis*) (n=16) White-tailed eagle (*Haliaeetus albicilla*) (n=24)	Norway/ 2008-2010	PFASs: PFOS	Plasma	PFOS: (*Accipiter gentilis)*: 0.3-36 (w.w.); (*Haliaeetus albicilla)*: 0.2-34 (w.w.)	PFOS: mean (median): (*Accipiter gentilis)*: 7.5 (6.2) (w.w.); (*Haliaeetus albicilla)*: 16 (13) (w.w.)	Bustnes et al. (2013)
White-tailed eagle (*Haliaeetus albicilla*) (n=21)	Norway/ 2011	NFRs: OPFRs: TPHP, TCIPP, TDCPP, TCEP, TBOEP, EHDPP	Plasma	NFRs: 0.1-0.7	NFRs:<LOQ-0.22	Eulaers et al. (2014)
			Feathers	NFRs: 1-3000; (w.w.)	∑NFRs: 4-110; (w.w.)	
Barn owl (*Tyto alba*) (n=15)	Belgium/ 2008-2009	PFASs: -PFCAs: PFBA, PFPA, PFHxA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA -PFSAs: PFHxS, PFOS, PFDcS -PFOSA	Muscle	PFASs: 11-477; (w.w.)	PFASs median range: n.d.-135; (w.w.)	Jaspers et al. (2013)
			Liver	PFASs: 6-116; (w.w.) PFOS: 42-992; (w.w.)	PFASs: 16-21; (w.w.) PFOS: 304 (w.w.)	
			Preen oil	PFASs: 2-1208; (w.w.)	PFASs: 22-431; (w.w.)	
			Fat	PFASs: 0.6-609; (w.w.)	PFASs: 0.6-203; (w.w.)	
			Feathers	PFASs: 2-670; (w.w.)	PFASs: 2-37; (w.w.)	
Northern goshawk (*Accipiter gentilis*) (n=56) White-tailed eagle (*Haliaeetus albicilla*)	Norway/ 2008-2010	PFASs: -PFCAs: PFPA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA -PFSAs: PFHpS, PFOS, PFDcS	Plasma	∑PFASs (*Accipiter gentilis)*: 9-99 (2008); 6-608 (2009) ∑PFASs (*Haliaeetus albicilla*): 33-71 (2008); 16-197 (2009); 10-60 (2010) ∑PFASs (*Aquila chrysaetos)*: 0.9-1	∑PFASs: (*Accipiter gentilis)*: 21 (2008); 151 (2009) ∑PFASs: (*Haliaeetus albicilla*): 55 (2008); 57 (2009); 32 (2010) ∑PFASs: (*Aquila chrysaetos)*: - (2008);	Sonne et al. (2012)

(n=36) Golden eagle (*Aquila chrysaetos*) (n=12)				(2008); 3-30 (2010)	12 (2010)	
White-tailed eagle (*Haliaeetus albicilla)* (n=5) Golden eagle (*Aquila chrysaetos*) (n=2) Northern goshawk (*Accipiter gentilis*) (n=16)	Norway/ 2008-2009	PFASs: -PFCAs: PFPA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA -PFSAs: PFHpS, PFOS, PFDcS	Plasma	∑PFASs: 0.9-1 (*Aquila chrysaetos*); 9-99 (*Accipiter gentilis*); 33-70 (*Haliaeetus albicilla*)	∑PFASs: - (*Aquila chrysaetos*); 21 (*Accipiter gentilis*); 55 (*Haliaeetus albicilla*)	Sonne et al. (2010)
Tawny owl (*Strix aluco*) (n=107)	Norway/ 1986-2009	PFASs: -PFCAs: PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA -PFSAs: PFHxS, PFHpS, PFOS, PFDcS -PFOSA	Eggs	PFASs (1986-2009): 0.04-12; (w.w.)	PFASs geometric mean range (1986-2009): 0.05-10; (w.w.)	Ahrens et al. (2011)
Peregrine falcon (*Falco peregrinus*) (n=13)	Spain/ 2003-2006	NFRs: -Dec: 602, 603, 604 -DP	Eggs	Dec: n.d.-25; (l.w.) ∑DP: 0.3-17; (l.w.)	Dec geometric mean range: 0.2-8; (l.w.) ∑DP: 2; (l.w.)	Guerra et al. (2011)
Peregrine falcon (*Falco peregrinus*) (n=10)	Sweden/ 1974-2007	PFASs: -PFCAs: PFHxA, PFHpA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFTriA, PFTeA, PFPeA -PFSAs: PFBS, PFHxS, PFOS, PFDcS -PFOSA	Eggs	PFASs: 0.3-220; (w.w.)	PFASs arithmetic mean range: 0.6-83; (w.w.)	Holmström et al. (2010)
Eurasian sparrowhawk (*Accipiter nisus*) (n=10)	Belgium/ -	PFASs: -PFCAs: PFOA, PFNA -PFSAs: PFHxS, PFOS	Liver	PFOSs: 47.6-775; (w.w.) Other PFASs: 3.2-40.6; (w.w.)	PFOS mean: 236; (w.w.)	Meyer et al. (2009)

			Feathers	-	PFOS mean: 102; (d.w.)	
White-tailed eagle (*Haliaeetus albicilla*) (n=-)	Germany and Poland/ 1979-1999	PFASs: -PFCAs: PFOA -PFSAs: PFOS -PFOSA	Liver	PFOS: 3.9-127;(w.w.) Other: n.d.	-	Kannan et al. (2002)

n.d.: non detected;**w.w.:** wet weight; **d.w.:** dry weight;**l.w.:** lipid weight

1383

1384

Table 3. Sample treatment strategies and analytical techniques and quantification limits for the determination of CECs in European raptors (2002-2020). The studies are ordered by year of publication, starting with the most recent ones.

Sample treatment technique(s) (extraction and clean-up)	Analytical technique(s)/column/mobile phase	LOQs (ng mL^{-1} ; ng g^{-1})	References
BPs and BzP UV-filters: (0.1-0.2 g tissue) Solvent extraction with ethyl acetate.	LC-MS-MS / C_{18} (2.1× 50 mm, 1.3µm)/(A) 0.1% v/v ammonium hydroxide in water and (B) methanol	0.01-6.5	González-Rubio et al. (2020)
NFRs: Solvent extraction with n-hexane:DCM (4:1, v/v) (~160 mg feathers and 1mL plasma) or DCM (preen oil) and clean-up with SPE (Supelclean ENVI Florisil; only for preen oil). PFASs: Solvent extraction with MeOH (0.2 mL plasma) or NaOH (200 Mm) in MeOH(~160 mg feathers) and clean-up with d-SPE (Supelclean ENVI-Carb and acetic acid)	NFRs: GC-MS/ DB-5 (30 m x 0.25 mm, 0.25 µm) for PBDEs and NBFRs or a HT-8 (25 x 0.22 mm, 0.25 µm) for OPFRs. PFASs: LC-MS-MS/ HSS-3T (2.1 x 100 mm, 1.8 µm); HSS-T3 guard column (2.1 x 5 mm, 1.8 µm)/)/ 2 mM NH_4OAc in 90:10 MeOH:water and 2 mM NH4OAc in MeOH	0.1-0.4	Briels et al. (2019)
NFRs: (1 g egg) PLE with hexane:DCM (1:1) and clean-up with SPE (alumina cartridge).	GC-(NCI)-MS-MS/ DB-5 (15 m x 0.25 mm, 0.1 µm)	0.007-32.2	Eljarrat et al. (2019)
NFRs: Solvent extraction with n-hexane (0.013-0.04 g preen oil) orn-hexane:DCM (4:1 v/v) (1 mL plasma and 0.2 g feathers) PFASs: Solvent extraction with MeOH (200-300 µL plasma) or 2M HCl in MeOH (0.2 g feathers) and clean-up with d-SPE (Supelclean ENVI-Carb)	NFRs: GC-MS/ DB-5 (30 m x 0.25 mm, 0.25 µm)/- PFASs: LC-MS-MS/ C_{18} (2.1× 50 mm, 5 µm)/ 2 mM NH_4OAc in 90:10 MeOH:water and 2 mM NH4OAc in MeOH	NFRs: 0.002-4 PFASs: 0.05-0.4	Løseth et al. (2019a)

PFASs (200-300 µL plasma): Solvent extraction with MeOH and clean-up with d-SPE (Supelclean ENVI-Carb, 50 µL of glacial acetic acid)	LC-MS-MS/ C_{18} (2.1× 50 mm, 5 µm)/ 2 mM NH4OAc in 90:10 MeOH:water and 2 mM NH4OAc in MeOH	0.05-0.1	Løseth et al. (2019b)
NFRs (0.14-0.22 g feathers): Solvent extraction with n-hexane-DCM (4:1, v:v) and clean-up with SPE (Supelclean ENVI Florisil topped with anhydrous Na_2SO_4)	GC-MS/HT-8 (25 m x 0.22 mm x 0.25 µm)/-	1	Monclús et al. (2019)
PFASs (0.2 mL plasma): Solvent extraction with ACN and clean-up with d-SPE with activated carbon and glacial acetic acid.	LC-MS-MS/ C_{18}(4.6 x 50 mm, 3.5 µm)/ water with 10 mM ammonium acetate/MeOH (80:20, v/v) and CAN with 10 mM ammonium acetate	0.01-0.1 (LOD)	Ortiz-Santaliestra (2019)
PFASs (~0.18 g feathers): Solvent extraction with MeOH and clean-up with SPE (ENVI-Carb with acetic acid column)	HPLC-QTRAP-MS-MS/-	0.006-0.15 (MDL)	Sun et al. (2019)
PFASs (1 g eggs): Solvent extraction with ACN and clean-up with d-SPE (Supelclean ENVI-Carb) PCNs (1 g eggs): SE withn-hexane:acetone (4:1 v/v) and clean-up with a multi-layer column of aluminium oxide, silica and acid-treated silica	PFASs: LC-QTRAP-MS-MS/C_{18} (2.1 x 150 mm)/ 2mM ammonium acetate:MeOH (90:10, v/v) and 2 mM ammonium acetate:MeOH (10:90, v/v) PCNs: GC-MS/DB-5 (0.25mm, 0.25 µm)/-	PFASs: 0.004-0.2 PCNs: 0.003-0.02	Vorkamp et al. (2019)
Paraffins (2-3 g muscle): Accelerated solvent extraction with DCM:n-hexane (1:1) and clean-up with SPE (multilayered column of Florisil, silica gel, acid silica gel and anhydrous sodium sulfate). Paraffins: (10 g eggs): Solvent extraction with isopropanol and n-hexane:diethyl ether (3:1, v:v) and clean-up with a multilayer SPE column.	APCI-QTOF-MS	0.4-41	Yuan et al. (2019)

NFRs (1.5 g eggs): Pressurized liquid extraction (PLE) with n-hexane-DCM (1:1 v/v) and clean-up SPE (neutral alumina)	GC-(NCI)-MS-MS/ DB-5 (15 m × 0.1 mm, 0.1 μm)/-	0.007-0.07	Blanco et al. (2018)
Neonicotinoids (100 μL blood): Modified QuEChERS method with ACN extraction and clean-up with d-SPE (Supel QuE PSA-C18-GCB)	LC-MS-MS/C_{18} (1.7 μm, 2.1 mm × 100 mm); C_{18} pre-column (130 Å, 1.7 μm, 2.1 mm × 5 mm)/95 % water, 5 % ACN, 5 mM ammonium formate, 0.1 % formic acid and 95 % ACN, 5 % water, 5 mM ammonium formate, 0.1 % formic acid	0.0006-0.03	Byholm et al. (2018)
NFRs (0.14-0.22 g feathers): Solvent extraction with n-hexane-DCM (4:1, v:v) and clean-up with SPE (Supelclean ENVI Florisil topped with anhydrous Na_2SO_4)	GC-MS/HT-8 (25 m x 0.22 mm x 0.25 μm)/-	1	Monclús et al. (2018)
PFASs (200 μL plasma and <0.1 g feathers): Solvent extraction with MeOH and clean-up with d-SPE (graphitized carbon, 50 μL of glacial acetic acid)	LC-MS-MS/ C_{18} (2.1 x 50 mm, 5 μm)/ 2 mM NH_4OAc in 90:10 MeOH: water and 2 mM methanolic NH_4OAc	-	Gómez-Ramírez et al. (2017)
UV-filters (0.1 g eggs): PLE with EthylAcetate:DCM (1:1 v/v) and clean-up SPE(C_{18})	LC-QTRAP-MS-MS/ C_{18} (50mm × 2.0 mm, 5 μm); Guard column of the same packaging material/ Water and ACN, both with 0.15% formic acid	0.0005-0.02	Molins-Delgado et al. (2017)
BPA, OP, NP (0.1 g feathers): Solvent extraction with methanol, 0.01 M ammonium acetate and chloric acid (VII) and clean-up with SPE (Oasis HLB)	HPLC-Fluorescence detector/C_{18} PAH (250 x 4.6 mm; 5 μm)/ACN and water	0.5-2	Nehring et al. (2017)
Neonicotinoids (500 μL blood): modified QuEChERS extraction with ACN (acetic acid 1% v/v) and clean-up with d-SPE (magnesium sulphate, PSA and polymerically bonded trifunctional C_{18} silica sorbent)	LC-TOF-MS/ C_{18} (250 mm × 4.6, 25 μm)/ 0.1% formic acid in water/ammonium formiate 5 mM and 0.1% formic acid in acetonitrile	2-10	Taliansky-Chamudis et al. (2017)

NFRs(0.1 g eggs): Solvent extraction with n-hexane and iso-octane and clean-up with deactivated silica gel (30% water, w/w)	GC-MS/HP-5 (30 m x 0.25 mm, 0.25 mm)/-	1.6-17	Vetter et al. (2017)
NFRs (- g eggs): SE with hexane:acetone (4:1) and clean-up with SPE (column with alumimium oxide, silica (with and without H_2SO_4) and Na_2SO_4.	GC-MS/DB-1701 (60 m) and DB-1 (15 m) for DBDPE and BEH-TEBP		Vorkamp et al. (2017)
PFASs (0.25 g eggs): Solvent extraction with ACN and clean-up with d-SPE (ENVI-carb, 100 µL glacial acetic acid)	LC-MS-MS/C_{18} (2.1 mm, 1.7 µm)/2 mM ammonium acetate	-	Eriksson et al. (2016)
PFASs (0.5 g eggs): Solvent extraction with ACN and clean up with d-SPE(graphitized carbon and acetic acid)	LC-MS-MS/C_{18} (50 x 2.1 mm, 1.7 µm)/2 mM ammonium acetate in MeOH and water.	0.01-0.3	Faxneld et al. (2016)
PFASs (200 µL plasma): Solvent extraction with MeOH and clean-up with d-SPE (ENVI-carb,100 µL glacial acetic acid)	LC-MS-MS/C_{18} (2.1 × 50 mm,5 µm)/2 mM NH_4OAc in 90:10 MeOH:water and 2 mM NH_4OAc in MeOH	-	Sletten et al. (2016)
Parabens (0.2-0.3 g liver): Solvent extraction with MeOH:ACN (1:1, v/v).	HPLC-MS-MS/ Zorbax SB-Aq (2.1 x 150mm, 3.5 µm); Guard column C_{18} (2.1 x 20mm, 5 µm)/ MeOH and water with 0.4% (v/v) acetic acid.	0.1-0.5	Xue&Kannan (2016)

NFRs (1 g eggs): PLE with n-hexane:DCM (1:1 v/v) and clean-up with SPE (alumina)	GC-MS-MS/DB-5 (15 m × 0.1 mm, 0.1 μm)/-	0.05-0.8	Barón et al. (2015)
NFRs (1.5 g eggs): PLE with hexane: DCM (1:1 v/v) and clean-up with SPE (Alumina)	GC-MS-MS/(15 m × 0.1 mm, 0.1 μm)/-	0.03-5	Barón et al. (2014)
PFASs (1 g eggs): Solvent extraction with ACN and clean-up with d-SPE (ENVI-Carb, glacial acetic acid)	LC-QTOF-MS/C_{18} column (150 x 2.1mm, 3 μm)/ MeOH and water (both with 2 mM NH_4OAc)	LODs: 0.06	Bustnes et al. (2014)
PFASs (0.2 g plasma): Solvent extraction with ACN and clean up with d-SPE (ENVI-Carb)	LC-QTOF-MS/ C_{18} (150 x 2.1mm, 3 μm)/ MeOH:water (both with 2 mM NH4OAc)	-	Bustnes et al. (2013)
NFRs (1 mL plasma and 0.2 g feathers): Solvent extraction with hexane:DCM (4/3:1, v:v) (NFRs) or hexane (PBDEs) and clean-up with SPE (Supelclean ENVI-Florisil topped with anhydrous Na_2SO_4)	GC-MS/ DB-5 (30 m x 0.25 mm, 0.25 μm) for PBDEs; HT-8 (25 m x 0.22 mm, 0.25 μm) for NFRs/-	0.02-5	Eulaers et al. (2014)
PFASs (1 g tissues and 0.3 g preen oil): Solvent extraction with MeOH and clean-up with d-SPE (ENVI-Carb, glacial acetic acid)	LC-TOF-MS/C_{18} (150 × 2.1 mm, 3 μm); Guard column (5 × 2.1 mm, 3 μm)/MeOH and water (both containing 2 mM NH_4OAc)	LODs: 0.5-60	Jaspers et al. (2013)

PFASs(0.2 g plasma): Solvent extraction with ACN and clean-up with d-SPE (ENVI-Carb).	LC-QTOF-MS/ C_{18} (150 x 2.1mm, 3 μm)/ MeOH:water (both with 2 mM NH4OAc)	-	Sonne et al. (2012)
PFASs (0.2 g plasma): Solvent extraction with ACN and clean up with d-SPE (ENVI-Carb)	LC-TOF-MS/ C_{18} (150 x 2.1mm, 3 μm)/ MeOH:water (both with 2 mM NH4OAc)	-	Sonne et al. (2010)
PFASs (1 g eggs): Solvent extraction with ACN and clean up with d-SPE (ENVI-Carb)	LC-QTOF-MS/ C_{18} (150 x 2.1mm, 3 μm)/ MeOH:water (both with 2 mM NH4OAc)	LODs: 0.01-0.05	Ahrens et al. (2011)
NFRs (3-5 g eggs): SE with DCM and clean-up with GPC (Bio Beads SX-3 column) and a silica gel column	GC-MS/ DB-5HT (0.25 mm, 0.10 μm)/-	0.01-1	Guerra et al. (2011)
PFASs (0.2-0.3 eggs): Solvent extraction with ACN and clean-up with d-SPE (Supelclean ENVI-Carb)	LC-MS-MS/ C_{18} (150 × 2.1 mm, 3 μm)/ (2 mM ammonium acetate) MeOH and Milli-Q water	LODs: 0.04-2	Holmström et al. (2010)
PFASs Solvent extraction with ACN and clean-up with d-SPE (activated carbon, glacial acetic acid) (1 g liver) or SPE (Oasis Plus) (1 g feathers)	LC-MS-MS/Fluophase PFP column (50mm×1 mm); C_{18} pre-column (10 mm×1 mm)/ 2 mM NH_4OAc and ACN	LOD: 3.2	Meyer et al. (2009)

PFASs (1 g liver): Solvent extraction with methyl tert-butyl ether	LC-MS-MS/C_{18} (50 x 2mm, 5 µm)/2 mM NH_4OAc and MeOH	1-72	Kannan et al. (2002)

PSA: Primary secondary amine; **GCB:**Gratiphized carbon black; **PLE:**Pressurized liquid extraction; **SE:** Soxhlet extraction; **Na_2SO_4**: sodium sulfate; **SPE:** solid phase extraction; **GPC:** gel permeation chromatography; **d-SPE:**dispersive solid phase extraction; **GC-MS:** Gas chromatography coupled to mass spectrometry; **LC-MS-MS:**Liquid chromatography coupled to a triple quadrupole spectrometer; **LC-QTOF-MS**: Liquid chromatography coupled to a quadrupole-time of flight mass spectrometer; **LOD:** Limit of detection; **MDL:** Method detection limit; **PFP:** Pentafluorophenyl

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