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Environ. Sci. Technol., **Just Accepted Manuscript** • DOI: 10.1021/acs.est.5b04663 • Publication Date (Web): 22 Dec 2015

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Abstract

The northern fulmar (*Fulmarus glacialis*) is defined as an indicator species of plastic pollution by the Oslo-Paris Convention (OSPAR) for the North-East Atlantic, but few data exist for fulmars from Norway. Moreover, the relationship between uptake of plastic and pollutants in seabirds is poorly understood. We analysed samples of fulmars from Norwegian waters and compared the POP concentrations in their liver and muscle tissue with the corresponding concentrations in the loads of ingested plastic in their stomachs, grouped as 'no', 'medium' (0.01 – 0.21 g; 1 – 14 pieces of plastic) or 'high' (0.11 – 0.59 g; 15 – 106 pieces of plastic). POP concentrations in the plastic did not differ significantly between the high and medium plastic ingestion group for sumPCBs, sumDDTs and sumPBDEs. By combining correlations among POP concentrations, differences in tissue concentrations of POPs between plastic ingestion subgroups, fugacity calculations and bioaccumulation modeling, we showed that plastic is more likely to act as a passive sampler than as a vector of POPs, thus reflecting the POP profiles of simultaneously ingested prey.

Introduction

Marine litter and especially plastic debris has emerged as a major environmental concern world-wide and has been recognized as a threat to marine ecosystems due to its large abundancy.[1] The yearly production rates of plastics have increased more than a hundredfold from the onset of plastic mass production (1950: 1.7 million tons) until today (2013: 299 million tons).[2] According to recent estimations, 5-13 million tons have ended up in the oceans by 2010.[1] However, present estimates are still under debate, including the major uncertainty associated with estimating emissions. Plastics are known to slowly weather by UV light and physical abrasion into smaller particles down to the micrometer and nano-scale but total

degradation is slow.[3-5] In terms of particle count, most of the plastic floating around in the world's oceans is microplastic debris, i.e. < 5mm.[6-8] Plastics are released into the environment from industrial activities (e.g. fishing, plastic abrasives, spills of plastic pellets) but also from domestic applications (e.g. washing of plastic microfiber clothes, usage of personal care products containing microplastics). Wear and tear of everyday items and products and use of domestic applications containing microplastics (e.g. car tires, fiber shredding from textiles, household waste, personal care products), have shown to contribute to environmental micro plastic pollution.[9] Climate change and increased ice melt may be an additional source by releasing currently ice-bound plastic particles into the water column.[10] As could be expected from the extensive presence of plastics in the marine environment, plastic fragments have been found in the gut of a wide range of marine species, from plankton to top predators.[4, 11-13] Seabirds are long-lived top predators with average lifespan of adult individuals varying between 5 to more than 30 years depending on species, increasingly recognized as sensitive indicators of the health and condition of the marine ecosystem.[14, 15] Among the most long-lived seabirds in boreal and arctic waters is the northern fulmar (*Fulmarus glacialis*), hereafter fulmar, a surface-feeding petrel with an extensive offshore foraging range during its entire life cycle. This makes it an ideal monitoring sentinel for marine plastic litter.[16–20] Van Franeker et al. (1985) were among the first toreport ingested plastic in fulmars.[21] Since then, reports on ingested plastic in seabirds have been steadily increasing.[12, 22, 23] Within Europe, fulmars are defined as an indicator species of plastic pollution by the Oslo-Paris Convention (OSPAR) for the North-East Atlantic.[24] OSPAR recommendations state that for an acceptable ecological quality objective (EcoQO), less than 10 % of the monitored population of fulmars should have more than 0.1g of plastic in the stomach.[24] Few data exist for fulmars from Norwegian waters, but the load of ingested plastic particles in dead fulmars beached in southwestern Norway is monitored annually as a contribution to the EcoQO monitoring

implemented by OSPAR. For the period 2005-2009 52% of the monitored population had more than 0.1 g plastic ingested.[20] Recently, Trevails and allies reported that 22.5 % of fulmars in the arctic archipelago of Svalbard, Norway, also were found with > 0.1 g of plastic in their stomach.[25] Besides these studies, no further data on ingested plastic in seabirds from Norwegian waters are available from the scientific literature, limiting our current understanding of the sources of contamination and hampering actions for the reduction of emission and subsequently the exposure of marine wildlife to plastic particles.

Marine litter that remains in surface waters can act as a floating artificial compartment accumulating persistent organic pollutants (POPs) that are within reach of marine life.[26-28] Considering that macro- and microplastics cannot be effectively removed from the ocean, research efforts are needed to understand how biological sentinels as seabirds are affected by ingestion, accumulation, possible leakage of chemicals and further breakdown of microplastics. We are aware of only one earlier study providing data on the bioaccumulation of POPs by fulmars from the Norwegian Arctic and Iceland.[25] This study found no significant difference in the tissue concentrations of PCBs, PBDEs, DDTs, HCB, Chlordanes and Mirex between fulmars with a high plastic load in their stomach (on average 0.63 ± 0.12 g) and fulmars that had no plastic in their stomach.[25] Recently, Tanaka and allies described the accumulation of PBDE in seabird tissues, indicating the potential of PBDE 209 to be transferred from ingested plastic to tissues.[26] To decrease the knowledge gaps, we aim at mechanistically explaining the role of plastic on the bioaccumulation of POPs by the fulmar and to increase the knowledge of ingested plastic and related POP concentrations in fulmars from coastal Norway.

The objective of this study was to investigate i) the occurrence of ingested plastic in fulmars collected in coastal Norway, ii) the relationship between ingested plastic particles and tissue concentrations of POPs and iii) the qualitative and quantitative relationship of POPs in

ingested plastic and the tissue concentrations in such individuals, with the final aim iv) to assess the contribution of POPs leaching from ingested plastic to the overall POP burden in fulmars by applying a mechanistic model. We are not aware of earlier studies that have combined statistical analysis of POP and plastic concentration data in fulmars with a mechanistic, plastic-inclusive bioaccumulation model analysis.

Materials and Methods

Sampling and study design. In 2012 and 2013, 72 fulmars were unintentionally caught as by-catch on long-lines off the coast of northern Norway (Figure 1, panel A) and delivered by fishermen to the Norwegian Institute of Nature Research (NINA) in Trondheim. In addition, NINA received 3 birds found dead on beaches in Rogaland county (Figure 1, panel B). During necropsy at NINA, the whole stomach and samples of liver and muscle tissue were collected from each individual. Tissue samples were put in aluminum foil, enveloped and frozen to -18 °C. Plastic particles were extracted from the stomach samples following an internationally standardized procedure[30] by rinsing the proventriculus and gizzard over a 1-mm sieve, drying their content in a Petri dish at 40 °C, and sorting it into different categories (i.e. plastic, non-plastic waste and natural food items), which were later weighed and stored separately in vials until further processing and chemical analyses at the Norwegian Institute for Air Research in Tromsø.

As an indication of body condition, the thickness (mm) of subcutaneous fat deposits was measured over the lower end of the breast bone. In addition, body condition was assessed as the sum of scores from evaluating both the subcutaneous and internal fat deposits and the breast muscle size on a 0-3 scale as described by van Franeker.[27]

Since plastic particles can reside in fulmar stomachs for several months muscle tissue was considered more suitable for assessing exposure than blood or liver tissue as it can be regarded to integrate a longer period of exposure. [20, 28] Only 14 (19%) of the 75 collected birds, had no visible plastic in their stomachs. The weight of ingested plastic in all birds varied between 0 and 0.59 g, with an average of 0.101 g. On the basis of number of plastic pieces found in the stomachs, tissue samples from 30 fulmars with either 'no', 'medium' (0.01 – 0.21 g; 1 – 14 pieces of plastic) and 'high' (0.11 – 0.59 g; 15 – 106 pieces of plastic) plastic ingestion were selected by randomized procedure for chemical analyses of POPs (n = 10 for all groups). Because of the applied method for extraction of plastic from the stomachs, particles <1mm were not included in the analysis. The high and median groups included 1 and 2 birds from Rogaland, respectively, all other birds were from North Norway. Muscle tissue was analysed for all three groups, while liver samples only were analysed for the high plastic ingestion group. In addition, the plastic particles found in the stomachs of the medium and high plastic ingestion groups were analysed for POPs.

Chemical analysis. All samples were analysed for a suite of POPs: PCB 18, 28/31, 52, 99, 101, 105, 118, 138, 153, 170, 180, 183, 194 (Ultra Scientific, Kingstown, USA) and BDE 28, 47, 99, 100, 119, 138, 153, 154, 183, 209 (Wellington laboratories, Ontario, Canada and CIL, Andover, USA) and DDTs (Ultra Scientific, Kingstown, USA). Of the muscle and liver tissue, 2 g were processed for analyses whilst all plastic found in each bird (ranging from 0.01 g and 0.59 g) was subjected to trace analyses. See Supporting Information for details.

Instrumental analysis. A Quattro micro TM mass spectrometer (Micromass MS technologies; Manchester, UK) was used for analyses of PCBs and PBDEs. For more

information regarding the method the reader is referred to Carlsson[29] and Supporting Information (SI).

Quality control. One quantifier and one qualifier ion were acquired for each target substance regardless of the POP group. A laboratory blank and a standard reference material (SRM) were analysed for every 10th sample. The NIST 1945 (whale blubber) was used as reference material. The relative standard deviations in SRMs were 18% for BDE-47 and between 6-14% for the analysed PCB congeners and the measured levels varied within an acceptable range ($\pm$20%) compared to the reference levels. The limit of detection (LOD) was calculated as three times the signal-to-noise ratio for each compound and the limit of quantification (LOQ) was calculated as 10 times the laboratory blank for all target analytes. The LOD for the PCBs ranged between 1-129 pg/g wet weight (ww), and 13-426 pg/g ww for the PBDE congeners, depending on congener and matrix. The median recoveries were 65-70% for the PCB internal standards and 45-54% for the PBDE internal standards. No additional recovery correction was carried out due to the application of the internal standard method.

Data treatment and statistical methods. Summed concentrations for POP groups were calculated from median concentrations of 14 PCBs (PCB 28, 52, 99, 101, 105, 118, 138, 153, 170, 180, 183, 187, 189, 194, of 3 DDTs (*p,p'* -DDT, *o,p* -DDT and *p,p'*- DDE) and 9 PBDEs (PBDE 47, 99, 100, 119, 153, 154, 183, 196, 209). Statistical analyses were executed using R, ver.3.1.1 and IBM SPSS Statistics, ver. 22.0.0.1, and statistical significance defined as $p < 0.05$.

Modeling bioaccumulation. The contribution from plastic to the total bioaccumulation of selected POPs by fulmars was assessed using an established kinetic mass balance approach [30-

[32] in which plastic is included as a component of the diet.[33, 34] The POP concentration in biota over time ($dC_{B,t}/dt$) is quantified using:

$$\frac{dC_{B,t}}{dt} = IR_{FOOD}a_{FOOD}C_{FOOD} + IR_{PL}C_{PLR,t} - k_{loss}C_{B,t} \quad (1)$$

The first term quantifies the uptake of POPs from the natural diet. The second term quantifies exchange of POPs between plastic and biota lipids during transfer of plastic in the birds gut. The third term is a loss term quantifying elimination and egestion. IR_{FOOD} and IR_{PL} are the ingestion rates i.e. the masses of food and plastic particles respectively, ingested per unit of time and organism dry weight, a_{FOOD} is the absorption efficiency from the diet, and C_{FOOD} is the POP concentration in the food. The product $a_{FOOD}\times C_{FOOD}$ quantifies the contaminant concentration that is transferred from food, i.e. prey, to the organism during gut passage.. $C_{PLR,t}$ is the POP concentration transferred from or to plastic during gut passage,[33, 34] and k_{loss} is the first order loss rate constant. Further details on the calculations are provided in the Supporting Information.

Results and Discussions

General condition of the birds. Although the majority of the birds could be considered healthy, the body conditions ranged from high amount of subcutaneous fat and large pectoral muscles to birds that clearly were in poorer condition. The lipid content averaged 4%, 2.5% and 2.5% in muscle tissue of the no, medium and high plastic ingestion group, respectively, and 5.2% in liver of the high ingestion group. The thickness of subcutaneous fat was however not significantly correlated with plastic mass in the stomachs (ANOVA on regression, p = 0.311), and did not differ between the three plastic ingestion groups (ANOVA, p = 0.338) nor between birds with and without ingested plastic (p = 0.573) or below and above the EcoQO of 0.1 g plastic (p = 0.122). Although the median condition index differed between the two latter groups (independent samples median test, p= 0.026), it did not differ significantly between

194 birds with or without plastic (p = 0.268) or between the three study groups of plastic load (p =
195 0.095).

196 **Ingested plastic.** Of the total of 75 birds, 14 individuals fell into the category of "no", 48 in
197 the category of "medium" and 13 in the category of "high" plastic ingestion. In the sub-group
198 selected for chemical analysis, the number of plastic particles per stomach averaged 6 in the
199 group with medium plastic ingestion and 41 in the high ingestion group. The weight of the
200 plastic found in the medium ingestion group averaged 0.08 g (median 0.04 g), which is less
201 than the OSPAR EcoQO maximum of 0.1 g, whereas the corresponding value for the high
202 ingestion group was 0.29 g (median 0.21 g), almost three times higher than the EcoQO limit.
203 For the total sample of fulmars delivered to NINA, 36% exceeded the EcoQO threshold (N =
204 75). The particle size varied between 1.8 mm and 9.1 mm (mean 5.0 mm) in addition to some
205 longer threads, excluding particles < 1 mm by the applied sieve.

206 **Persistent organic pollutants in ingested plastic.** Of the analysed PCBs, all PCBs besides
207 PCB 28, 52, 101 and 189 were detected in more than 70% of all samples. The sumPCB
208 concentrations ranged between 0.08 and 64.4 ng/g with a median of 2.49 ng/g demonstrating
209 large variation among individuals. When comparing the medium and high groups of ingested
210 plastic, a median sumPCB concentration of 2.49 ng/g was found in the high group compared
211 to 4.03 ng/g in the medium group. In both the medium and the high ingestion group, PCB 153
212 was the major PCB found, followed by PCB 118 and 138 (see Table 1 for concentrations).
213 For the DDTs, *p,p'*-DDE was the major DDT compound found with a median of 16.05 ng/g in
214 the high plastic ingestion group and 53.4 ng/g in the medium group. The highest
215 concentrations of sumDDTs were found in one sample from the medium ingestion group with
216 823 ng/g. DDE was dominating over DDT with at least a factor of 10 in all plastic samples,
217 pointing to general old sources and/or previous biological degradation.

When assessing the PBDE data, there is more variation in concentrations among individuals as compared to the PCBs. SumPBDE concentrations ranged between < LOD to 16.7 ng/g with a median concentration of 1.68 and 2.33 ng/g for the high and medium ingestion samples respectively. Furthermore, the detected congeners differed considerably as for example in one sample from the high ingestion batch the high brominated PBDEs as PBDE 183 and 209 were detected whereas PBDE 47, 100 and 154 were detected in most of the other samples. The concentrations found in the ingested plastic per bird were higher in the high ingestion group compared to the medium ingestion group (median of sumPCBs: 1.12 ng/ bird and 0.3 ng/ bird; median of sum PBDE: 0.29 ng/ bird and 0.18 ng/ bird; sumDDTs: 7.32 ng/ bird and 5.43 ng/ bird for high and medium ingestion groups, respectively).

The differences in POP concentrations between the high and medium plastic ingestion groups were however not significant for sumPCBs and sumPBDEs ($p > 0.05$, Wilcoxon Rank Sum Tests) and the somewhat lower sumDDTs in the high ingestion group were only close to significance ($p = 0.07$). The tests were also performed without the extreme values (data not shown), which however did not yield differences in the detected significances (Figure 3).

Persistent organic pollutants in tissue samples. All targeted PCBs could be detected in the analysed muscle and liver samples. The PCB pattern observed in muscle and liver samples was similar to that in the ingested plastic, with PCB 153 as the dominating congener followed by 180, 183 and 118. SumPCB levels in muscle tissues ranged between 69.7 to 2067 ng/g ww with median sumPCB concentrations of 665, 1005 and 607 ng/g ww for the high, medium and no ingestion group, respectively. In liver samples, sumPCB concentrations varied between 183 and 3830 ng/g ww in the high ingestion group with a median sumPCB of 782 ng/g ww (See Table 2 for concentrations).

243 *P,p'*-DDE was the major DDT observed in muscle and liver tissue, ranging between 22.8 and
244 1251 ng/g ww in muscle samples (median of 228, 396 and 209 ng/g ww in high, medium and
245 no ingestion group respectively). In liver, *p,p'*-DDE ranged between 74 and 1634 ng/g ww in
246 the high ingestion group, with a median of 164 ng/g ww. Of the analysed pesticides, *oxy*-
247 chlordane, HCB, Mirex, *t*-nonachlor and *t*-chlordane were detected in decreasing order. The
248 concentrations of *oxy*-chlordane ranged between 112 and 154 ng/g ww in liver and between
249 31 and 690 ng/g ww in muscle.
250 PBDE 153, 47 and 154 dominated the PBDE pattern in muscle tissues. PBDE 209 was only
251 detected in two muscle samples with 259 and 8 ng/g ww. The one elevated PBDE209 muscle
252 sample also demonstrated high levels of PBDE 209 in its ingested plastic, suggesting a
253 plastic-tissue transfer in this one incident. Muscle sumPBDE concentrations varied between
254 0.24 and 9.91 ng/g (not considering the one elevated PBDE 209 sample) with a median of
255 1.26, 1.51 and 0.74 ng/g ww for no, median and high ingestion samples, respectively. Liver
256 tissue had a comparable PBDE pattern, with additional PBDE 183 and 184 detected in the
257 majority of the samples, but no PBDE 209. Liver sumPBDE concentrations ranged between
258 0.28 and 3.15 ng/g ww, with a median of 0.98 ng/g ww. The differences in concentrations in
259 muscle tissues between the plastic ingestion groups were significant for sumPBDEs (based on
260 lipid weight normalized concentrations, Kruskal Wallis test, $p=0.01$), whereas the differences
261 were not significant for sumDDTs ($p=0.07$) and sumPCBs ($p>0.05$) (Figure 4). For all three
262 compound groups, the highest median concentration was found in the medium ingestion
263 group, while the high ingestion group showed the lowest median compared to the two other
264 groups.
265 **Effect of plastic on bioaccumulation: statistical evaluation of concentration data.**
266 Correlation of liver and muscle concentrations in the high ingestion group resulted in Pearson
267 correlation coefficients *r* ranging between 0.93 and 0.99 for the individual PCBs, suggesting

equilibrium of PCB in liver and muscle tissues. The correlation between the POP groups in ingested plastic and muscle tissue on a lipid weight basis also was statistically significant with r^2 of 0.49 ($p = 0.03$) for sumPCBs, and 0.72 ($p < 0.001$) for sumDDT but not significant for sumPBDEs ($r^2 = 0.24$; $p = 0.35$).In summary, PCBs and DDTs in ingested plastic are relatively strong correlated with the concentrations found in muscle tissue, a tissue reflecting a long-term POP exposure. Since plastic particles reside in the stomach of the birds for weeks and up to months[20], they are constantly exposed to the continuously ingested fish diet, also containing POPs. The order of sumPCB, sumDDT and sumPBDE concentrations found in ingested plastic as well as in muscle tissue was: medium > high > no plastic ingestion. Bioaccumulation of POPs thus was not proportional with quantity of plastic ingested, an observation that contradicts the hypothesis that plastic acted as a carrier of POPs. Together with the close correlation of POPs found on ingested plastic with muscle tissues, this suggests that the plastic particles rather reflect the POP levels found in the food of fulmars, i.e. acting as a kind of "passive sampler" due to their lipophilic character and long residence time in the stomach of seabirds, rather than being a direct source of POPs to the birds. With the exception of one individual (showing high PBDE 209 concentrations in both plastic and muscle tissue), the POPs absorbed to the plastic prior to ingestion might be desorbed very soon after ingestion, yet may be of little influence if in fact the influx of POPs by the fulmars' prey would be larger, or if the fugacities of POPs in the fulmar lipids would be higher than in the plastic. The latter two conditions are mechanistically evaluated below, (a) by calculating fugacities, and (b) by a model-assisted quantitative analysis of the bioaccumulation fluxes due to ingestion of plastic and of food items (see section below).

Fugacities of POPs in fulmar lipids versus ingested plastic. To further analyze the likely direction of POP transfer, that is, from plastic to biota lipids or vice versa, we calculated lipid-plastic fugacity ratios. Lipid-plastic fugacity ratios ranged from 2.6×10^3 (PCB28) to 2.3×10^6

293 (PCB194). It appears that the fugacities of POPs in lipids are much higher than in plastic and
294 increase with hydrophobicity (Figure S3 and SI pp. 2), which implies biomagnification from
295 either prey or from plastic or both. A prerequisite for biomagnification is volume reduction of
296 the ingested medium, which for ingested prey is rapid digestion of prey lipids.[35, 36] Plastic
297 inside a fulmars' stomach however, is known to degrade very slowly due to mechanical wear,
298 with half-lives of months.[28] Mechanical wear partly leads to increased numbers of smaller
299 particles, which in turn can be egested, but it does not lead to a proportionally lower total
300 volume of plastic in the intestine. Per unit of time, the volume reduction due to digestion of
301 persistent microplastics would be much smaller than that for more digestible prey items. This
302 implies that the observed fugacity ratio for the main part must be caused by biomagnification
303 of POPs from prey. At the same time, gut residence times of microplastics are long whereas
304 POP exchange kinetics are fast and therefore sufficient to cause chemical equilibrium with
305 ingested microplastics.[37] Given the higher fugacity of POPs in biota lipids compared to
306 microplastics, transfer from the biota lipids to the plastic will occur, which is consistent with
307 our hypothesis of microplastics acting as a passive sampler for POPs in the gut.
308
309 **Modeling the contribution of ingested plastic to the total bioaccumulation of PCBs.**
310 The uptake of PCBs by fulmars was modeled using Eq. 1, with a few key assumptions. The
311 first assumption is that we modeled an 'average' fulmar. This implies that average POP
312 concentrations are used for the fulmars with and without plastic, and that the selected
313 parameters relate to the behavior of the 'mean' fulmar in the sampled population. A second
314 assumption is that the measured and modeled bioaccumulation of plastics and POPs relate to
315 steady state and reflects the time-averaged net result of uptake and loss processes that on
316 shorter time scales may show some seasonal and spatial fluctuations. Parameters were
317 obtained as follows. First, the ingestion rate *IR* of regular prey (i.e. IR_{FOOD}, Eq 1) needs to be

known. Barrett et al. (2002) estimated 365 500 fulmars inhabiting Norwegian waters with an average body mass of 810 g each, which consumed 31 624 metric tonnes of prey per year. This translates into an average 'normal prey' ingestion rate 'IR_{FOOD}' of 0.3 g prey per gram of body mass (g bm) fulmar per day.[38]

The ingestion rate for plastic (IR_{PL}, g/g bm d^{-1}) can be calculated as follows. We assume that the accumulation of plastic in the fulmars' stomach is a balance of accumulation and loss processes:

$$\frac{dC_{PL}}{dt} = IR_{PL} - k_R C_{PL} \quad (2)$$

where C_{PL} is plastic concentration in the bird (g/g), and k_R (d^{-1}) is the first order removal rate constant from the stomach. At steady state it follows from Eq.2 that $IR_{PL} = k_R C_{PL}$. Therefore, IR_{PL} can be calculated from the measured average concentration of plastic in the fulmars stomach (C_{PL} =0.3 g of plastic per 973 g of fulmar weight = 3.083×10^{-4} g/g) and k_R. Van Franeker et al. (2011) provided an estimate of the loss rate of 75% of ingested plastic in one month, which translates into a first order removal rate constant of k_R= 0.0462 d^{-1}.[20] The product of $k_R C_{PL}$ equates to IR_{PL} and is calculated as $3.083\times10^{-4} \times 0.0462$ d^{-1} = 1.43×10^{-5} g plastic per gram fulmar per day. The fraction of plastic in the ingested food equates to $IR_{PL}/IR_{PREY} = S_{PL}$ and is calculated as 1.43×10^{-5} / 0.3 = 4.75×10^{-5}. Recently, it has been argued that the aforementioned loss rate of 75% per month may be overestimated by an order of magnitude.[28] This would imply that S_{PL}would be even an order of magnitude lower than 4.75×10^{-5}. Obviously, such estimations carry uncertainties, yet due to the extremely low value of S_{PL}we can safely conclude that ingestion of plastic mass is negligible compared to the mass of ingested prey per unit of time. To calculate C_{FOOD} (Eq. 1), PCB congener concentration data for in the fulmars' diet shorthorn sculpin *Myoxocephalus scorpius*, Arctic staghorn sculpin *Gymnocanthus tricuspis*, Atlantic cod *Gadus morhua*, polar cod *Boreogadus saida*, capelin *Mallotus villosus*, and haddock *Melanogrammus aeglefinus*, all sampled in

343 Kongsfjorden (78°55'N, 11°56'E), Svalbard, Norway in 2007, were averaged.[39] The average
344 PCB concentration varied among these diet components with a relative standard deviation of
345 ~50%. The loss rate parameter k_{loss} was individually calibrated for each of the PCB congeners,
346 using the known PCB concentrations measured in these diet components, and in the fulmars
347 *without plastic*, the plastic ingestion term in Eq.1 (i.e. IR_{PL}) was set to zero and the a_{FOOD} to
348 0.8.[30] The optimized k_{loss} values decreased linearly with LogK_{OW}, (Figure S2).
349
350 Finally, bioaccumulation of PCBs by the fulmars with plastic was modeled by using all
351 aforementioned parameters including the plastic ingestion term, with S_{PL}= 4.75×10^{-5} and a
352 value for the k_{1G} POP exchange rate constant parameter of 10 d^{-1}. This value is at the higher
353 end of the range calculated for microplastics from first principles [33], as well as of the range of
354 values measured for artificial gut fluids.[37]
355 The modeled lipid normalized PCB concentrations agreed very well to the measured C_{lipid}
356 (µg/g) values, with no significant difference from the 1:1 line (Figure 2). This implies that the
357 k_{loss} values from the fulmars without plastic provided an excellent agreement to the
358 bioaccumulation data for birds with plastic. In the model, the concentration in the plastic at
359 ingestion was equated to the value measured for plastic in stomach, which however is not the
360 same as the concentration in the freshly ingested plastic, which may have been different.
361 Therefore, we explored a scenario where the model was allowed to fit an optimal
362 concentration in the plastic. This optimal PCB concentration appeared to be 'zero', which
363 implies that 'no influence of PCB uptake by plastic' best explains the bioaccumulation in the
364 birds in which a median of 0.3 g of plastic was found. This is consistent with the
365 aforementioned inferences on ingestion rates, which showed that plastic ingestion was
366 negligible, compared to that of regular prey. Results from this second scenario were
367 indistinguishable from those in Figure 2 and therefore not plotted separately. To explore the

368 sensitivity of the model to the concentration in ingested plastic, we also explored a third
369 scenario in which the concentrations in ingested plastic were taken 1000 times higher than the
370 values measured for plastic in the stomach. The intercept of the resulting regression between
371 modeled and measured values now moved away from the 1:1 line (Figure S1). This poorer fit
372 however, still was not dramatic due to the unimportance of plastic ingestion compared to that
373 of regular prey.
374
375 **General discussion and Implications**
376 For the first time, POP concentrations in tissues and ingested plastic from the same individual
377 were analysed for fulmars in Norway. Earlier studies on the diving behavior of chick-rearing
378 fulmars in Shetland, U.K., showed that fulmars forage on their prey through shallow dives (*N*
379 = 97 per day); 85% of these dives less than 1 m deep, potentially exposing them to floating
380 plastic debris.[40] POP concentrations have been reported in fulmars from Norway before,
381 indicating lower PCB and DDT concentrations but higher PBDE concentrations compared to
382 our study.[41-46]
383 In our study, we have provided several lines of evidence suggesting that ingested
384 microplastics can act as 'negligible depletion' passive samplers for POPs originating from
385 ingested food. First, we found that POP concentrations in fulmars were not linked to the
386 magnitude of their stomach plastic concentrations, which would have been the case if plastic
387 acted as a substantial carrier of the POPs to the fulmars. Lack of unidirectional relationships
388 between these variables has also been demonstrated in one other study,[25] supporting our
389 findings are not incidental. Second, we found that POP concentrations in plastic correlated
390 strongly with POP concentrations in fulmars, which implies that chemical transfer still does
391 occur. Thirdly, we found that chemical fugacities in plastic were lower than in the birds lipids,
392 which would suggest transfer of POPs to the plastic i.e. as passive samplers, rather than the

393 other way around. This would explain the aforementioned correlation, and might also explain
394 such correlations reported in earlier studies (eg.,[26]). Fourth, we quantified the fluxes of POPs
395 entering fulmars using a dynamic bioaccumulation model. We calculated that the flux of
396 POPs by ingestion of natural prey would be at least 21 000 times higher than the flux of POPs
397 by ingestion of plastic. The uptake from plastic thus is calculated to be overwhelmed by
398 ingestion via natural pathways aka by ingestion via feed, which also has been recognized by
399 recent modeling studies [33, 37, 47, 48] and in 2015 by the GESAMP Working Group 40 on Marine
400 Litter.[49]The suggested dominance of plastic-mediated internal exposure to PBDE 209 in
401 particular as stated by Tanaka et al., could not be observed when applying average data and in
402 comparison with individuals with no ingested plastic in their guts as a control.[26]
403 In summary, we conclude that bioaccumulation of POPs by fulmars is mainly governed by the
404 ingestion of natural prey. POPs taken up via ingested plastics may equilibrate readily in the
405 intestines of the birds, making a negligible contribution to accumulation, yet absorbing POPs
406 from the ingested food simultaneously such that POP profiles in plastic reflect the profiles
407 observed in tissues. Since the here applied sampling methodology excluded particles smaller
408 than 1mm, follow up studies are recommended to include such smaller sized particles.
409 It has been generally recognized that it is difficult to infer causal relationships from
410 correlative evidence. Here we showed that correlations among POP concentrations in plastic
411 and tissues do not necessarily imply that plastic acts as a substantial carrier for POPs. By
412 combining correlations among POP concentrations, differences between plastic ingestion
413 subgroups, fugacity calculations and bioaccumulation modeling, we showed that ingested
414 plastic is due to its relatively long residence time more likely to act as a passive sampler,
415 reflecting the POP profiles as they occur in the gastro-intestinal tract. Although this study was
416 specific for birds, it is likely that microplastics may act as passive samplers (rather than as
417 vectors for bioaccumulation) also in other species, like invertebrates or fish.

Acknowledgements

The project was partly funded by the EU project CLEANSEA and the FRAM Centre project Microplastics in arctic marine food webs. The collection of fulmars was funded by the Norwegian Environment Agency as part of a study of unintentional by-catch of seabirds in Norwegian fisheries[50] and the EcoQO monitoring of fulmars beached in Rogaland. We also thank Jan van Franeker at IMARES, Texel, for guidance with necropsy and sampling of the fulmars, and Line Christoffersen for assistance in the sample preparation for chemical analyses.

Supporting Information Available

Text, figures and tables addressing i) the model parameters, least squares used in the modelling approach, ii) illustrating the further validation of the model, iii) giving loss rate constants (k_{loss}) estimated for PCBs, based on bioaccumulation data without plastic ingested and iv) presenting the Muscle - Plastic Fugacity ratios for selected individual birds. This information is available free of charge via the Internet at http://pubs.acs.org/.

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Figure captions:

Figure 1: Main sampling regions in coastal Norway for the fulmars examined in this study between 2012 and 2013, Panel A: North Norway, Panel B: South Norway (n=75; $n_{south} = 3$; $n_{north} = 72$).

Figure 2: Log modeled vs log measured lipid based concentration C_{lipid} (µg/g). Fully plastic-inclusive model implemented.

Figure 3: Summed concentrations of A) PCBs, B) DDTs and C) PBDEs (concentrations displayed as ng/g on a log10-scale) for the ingested plastic content in the medium and high plastic ingestion groups in ng/g plastic. (Triangles: individual concentrations; dots: outliers)

Figure 4: Summed wet weight concentrations of A) PCBs, B) DDTs and C) PBDEs in muscle tissue in the no, medium and high plastic ingestion groups in ng/g ww. One extreme value for sumPBDEs (267 ng/g) is excluded. (Triangles: individual concentrations; dots: outliers)

TOC/Abstract art

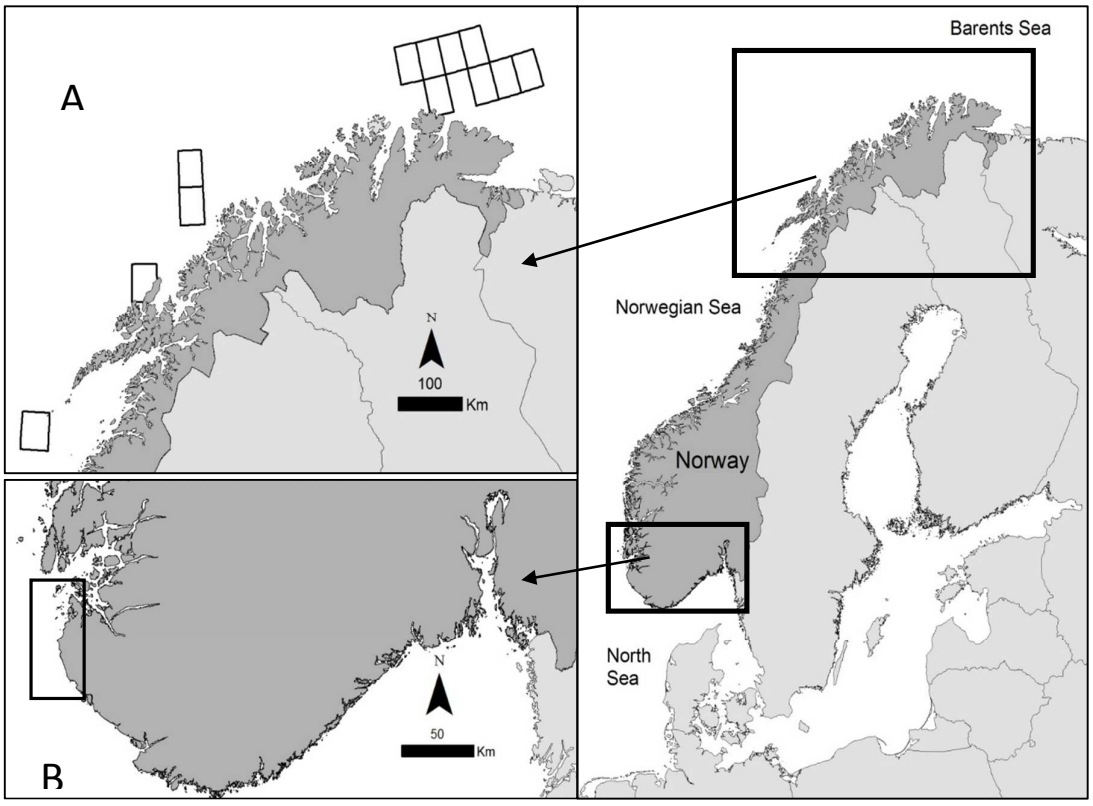

Figure 1

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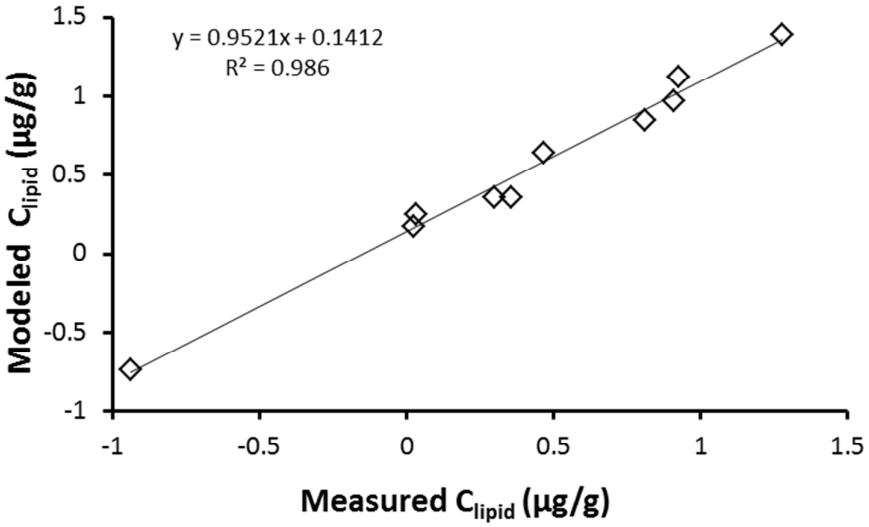

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649 **Figure 2**
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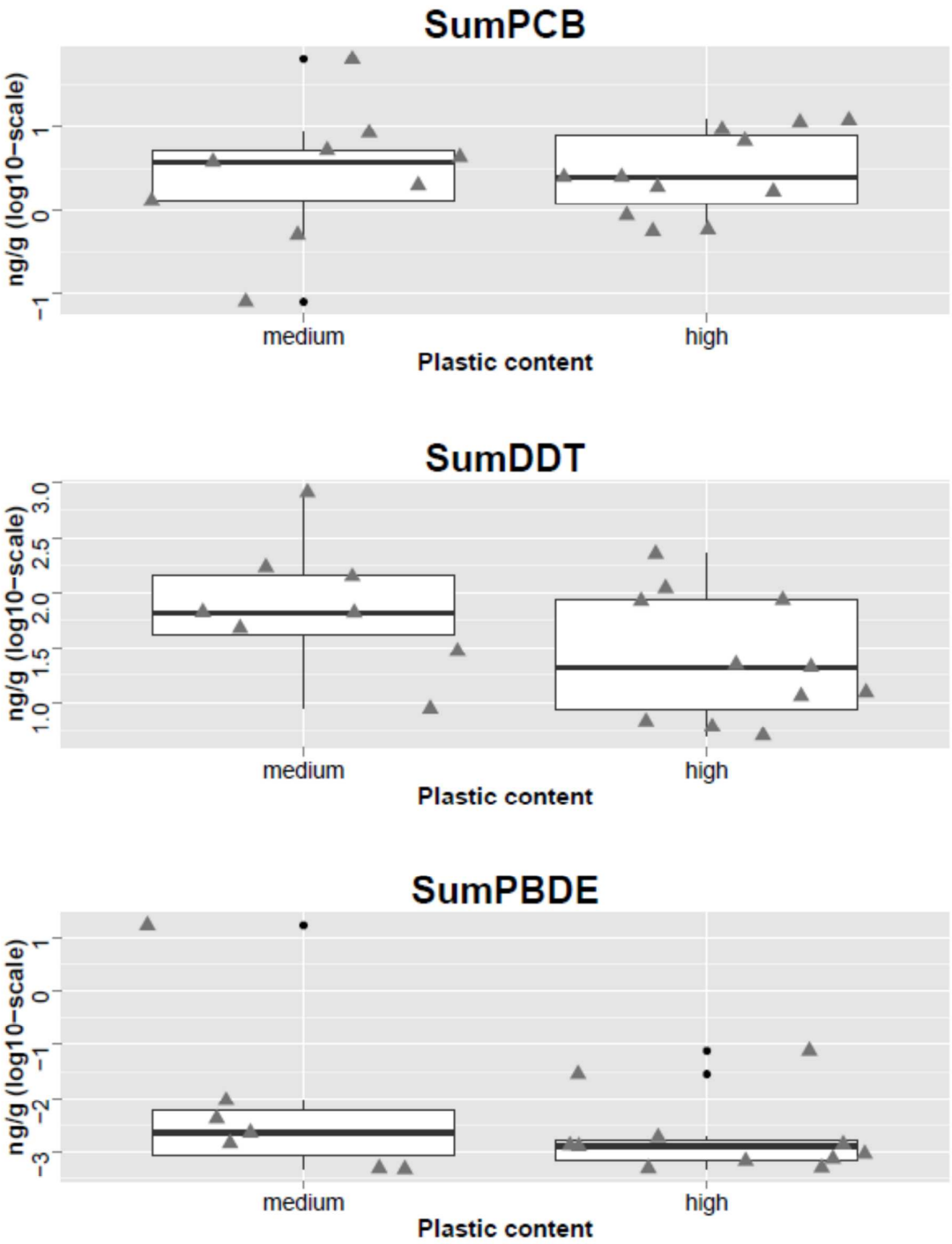
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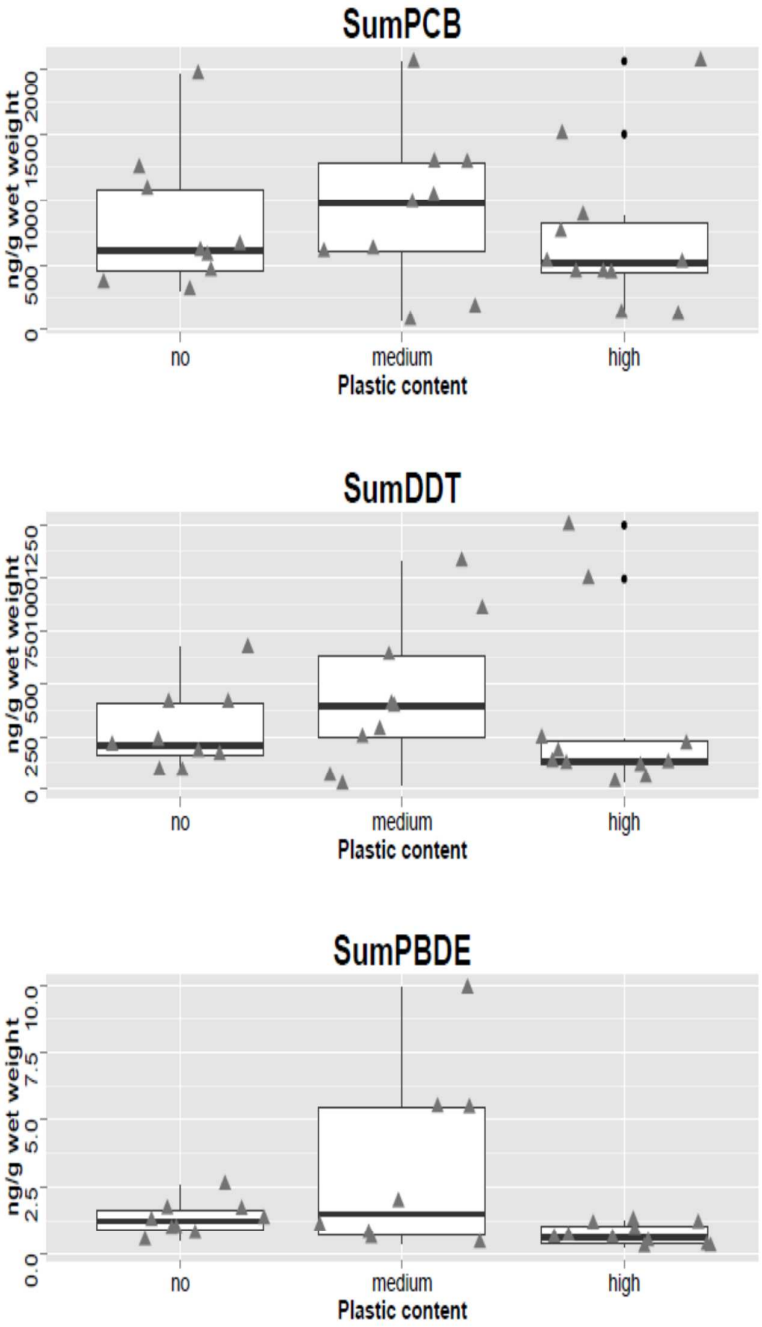
Figure 3



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663 **Figure 4**

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666 **Table 1: Concentrations of POPs in ingested plastic samples of Northern Fulmars in**
667 **ng/g for the different ingestion groups (nd: not detected)**

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	***Medium* plastic ingestion**			***High* plastic ingestion**		
	Median	Mean	± SD	Median	Mean	± SD
PCB 28/31	0.01	0.02	0.03	0.01	0.02	0.02
PCB 52	nd	0.03	0.09	nd	0.01	0.04
PCB 99	0.18	0.43	0.73	0.08	0.21	0.24
PCB 101	0.03	0.16	0.36	nd	0.04	0.09
PCB 105	0.16	0.37	0.60	0.12	0.21	0.22
PCB 118	0.57	1.59	2.69	0.40	0.85	0.89
PCB 138	0.75	2.14	3.77	0.37	0.75	0.73
PCB 153	1.31	3.32	6.24	0.81	1.50	1.49
PCB 170	0.21	0.58	1.16	0.09	0.21	0.21
PCB 180	0.57	1.66	3.29	0.23	0.53	0.53
PCB 183	0.06	0.21	0.43	0.04	0.07	0.08
PCB 187	0.02	0.20	0.41	0.02	0.05	0.08
PCB 189	nd	0.02	0.06	0.002	0.004	0.005
PCB 194	0.07	0.22	0.42	0.03	0.06	0.05
$\sum_{14}$PCB	3.92	10.94		2.21	4.51	
p,p'-DDT	0.23	1.12	2.11	0.53	1.32	1.39
o,p-DDT/ *p,p'*-DDD	1.96	6.67	9.48	0.61	1.19	1.13
p,p'-DDE	53.4	130	239	16.0	50.7	66.3
o,p-DDE	nd	0.04	0.13	nd	0.13	0.33
o,p-DDD	0.20	1.47	2.77	0.06	0.12	0.17
∑DDT	55.8	139		17.2	53.5	

PBDE 28	0.04	0.11	0.16	0.08	0.07	0.06
PBDE 47	0.71	1.82	2.61	0.38	0.44	0.27
PBDE 99	nd	nd	nd	nd	nd	nd
PBDE 100	0.04	0.29	0.55	0.13	0.10	0.10
PBDE 119	nd	nd	nd	nd	nd	nd
PBDE 138	nd	nd	nd	nd	nd	nd
PBDE 153	nd	0.13	0.28	nd	0.02	0.04
PBDE 154	0.09	0.22	0.29	0.04	0.07	0.08
PBDE 183	nd	0.23	0.72	nd	0.10	0.25
PBDE 209	nd	1596	5047	nd	9.05	22.8
$\sum_{10}$PBDE	0.88	1669		0.62	10.3	

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674 **Table 2: Concentrations of POPs in tissue samples of Northern Fulmars in pg/g wet weight for all ingestion groups (nd: not detected)**

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	Muscle	***No***	**Ingestion**	**Muscle**	***Medium***	**Ingestion**	**Muscle**	***High***	**Ingestion**	**Liver**	***High***	**Ingestion**
	Median	Mean	± SD	Median	Mean	± SD	Median	Mean	± SD	Median	Mean	± SD
PCB 28/31	0.98	1.06	0.44	0.89	1.06	0.61	0.96	1.18	0.70	1.20	1.61	1.55
PCB 52	0.10	0.33	0.73	1.20	2.98	4.14	0.05	0.37	0.65	1.44	2.41	3.21
PCB 99	20.7	27.4	17.1	36.5	39.9	28.9	16.6	31.4	34.7	26.8	42.5	58.4
PCB 101	0.25	0.61	0.89	0.13	0.77	1.56	0.07	0.06	0.05	0.12	0.11	0.08
PCB 105	19.2	27.4	17.0	28.2	31.1	20.8	15.9	27.9	28.9	29.26	42.2	57.0
PCB 118	63.2	83.7	52.1	89.9	98.6	65.4	54.8	89.6	89.2	108	150	194
PCB 138	79.7	112	77.2	142	153	108	68.6	112	115	104	162	213
PCB 153	215.	296	188	355	316	218	195	260	205	289	351	397
PCB 170	37.0	52.8	40.3	58.0	53.1	39.4	29.2	40.2	28.8	43.4	52.8	55.8
PCB 180	114	160	121	165	158	118.8	89.5	115	76.3	123	151	153.
PCB 183	12.6	17.7	12.7	18.8	20.9	14.3	10.2	14.5	11.4	14.9	20.3	23.0

PCB 187	0.39	1.03	1.47	0.25	1.62	2.70	0.18	0.26	0.16	0.49	0.56	0.49
PCB 189	1.67	2.20	1.54	1.88	2.07	1.65	1.33	1.59	0.93	2.06	2.18	2.18
PCB 194	18.7	21.4	14.6	15.4	20.1	15.3	12.1	14.7	8.76	16.8	19.3	17.92
$\sum_{14}$PCB	585	805		914	900		495	709		763	999	
p,p'-DDT	0.9	1.5	1.5	0.6	1.6	1.8	0.9	0.8	0.5	0.2	0.5	0.6
o,p-DDT/												
p,p'-DDD	8.6	10.3	8.6	17.6	14.8	12.8	3.5	8.6	13.0	2.0	4.9	7.4
p,p'-DDE	206	260	181	352	424	345	122	305	396	164	381	562
o,p-DDE	nd	0.0	0.1	nd	0.0	0.1	nd	0.0	0.0	nd	0.0	0.0
o,p-DDD	nd	0.1	0.2	nd	0.1	0.3	nd	0.0	0.0	0.0	0.0	0.0
∑DDT	216	272		370	441		127	315		167	386	
PBDE 28	0.04	0.05	0.02	0.04	0.04	0.03	0.02	0.03	0.03	0.04	0.05	0.05
PBDE 47	0.34	0.42	0.31	0.17	0.49	0.74	0.10	0.12	0.05	0.17	0.17	0.13
PBDE 99	0.11	0.16	0.15	0.11	0.45	0.77	0.06	0.07	0.04	0.13	0.13	0.09
PBDE 100	0.09	0.10	0.06	0.04	0.12	0.17	0.02	0.03	0.02	0.05	0.05	0.05
PBDE 119	0.03	0.03	0.01	0.02	0.03	0.03	0.02	0.02	0.02	nd	nd	nd

PBDE 138	nd	0.00	0.00	nd	0.00	0.00	nd	0.00	0.01	0.00	0.01	0.02
PBDE 153	0.30	0.31	0.15	0.56	0.50	0.36	0.24	0.27	0.21	0.32	0.51	0.63
PBDE 154	0.17	0.19	0.08	0.11	0.25	0.27	0.12	0.12	0.07	0.15	0.18	0.16
PBDE 183	nd	0.01	0.01	0.02	0.02	0.01	nd	0.01	0.01	0.03	0.03	0.03
PBDE 209	nd	nd	nd	nd	29.70	86.12	nd	nd	nd	nd	nd	nd
$\sum_{10}$PBDE	1.08	1.30		1.10	32.54		0.59	0.70		0.93	1.17	
Lipid %	4.3	3.95	1.35	3.2	2.3	1.57	2.6	2.7	0.73	4.8	5.2	1.61

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