Environmental Science & Technology

Article

pubs.acs.org/est

# Negligible Impact of Ingested Microplastics on Tissue Concentrations of Persistent Organic Pollutants in Northern Fulmars off Coastal Norway

Dorte Herzke,*[,†] Tycho Anker-Nilssen,[‡] Therese Haugdahl Nøst,[†] Arntraut Götsch,[†] Signe Christensen-Dalsgaard,[‡] Magdalene Langset,[‡] Kirstin Fangel,[⊥] and Albert A. Koelmans[§,∥]

[†]Norwegian Institute for Air Research, FRAM—High North Research Centre on Climate and the Environment, 9296 Tromsø, Norway

[‡]Norwegian Institute for Nature Research, P.O. Box 5685 Sluppen, 7485 Trondheim, Norway

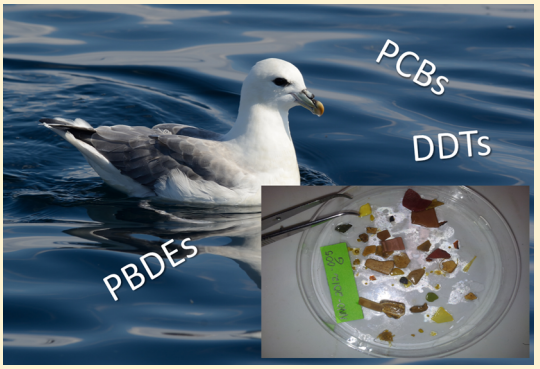
[⊥]Norwegian Institute for Nature Research, Fakkelgården, 2624 Lillehammer, Norway

[§]Aquatic Ecology and Water Quality Management Group, Department of Environmental Sciences, Wageningen University, P.O. Box 47, 6700 AA Wageningen, The Netherlands

[∥]IMARES—Institute for Marine Resources & Ecosystem Studies, Wageningen UR, P.O. Box 68, 1970 AB IJmuiden, The Netherlands

Supporting Information

**ABSTRACT:** The northern fulmar (*Fulmarus glacialis*) is defined as an indicator species of plastic pollution by the Oslo-Paris Convention 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 analyzed 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 worldwide 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 nanoscale 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., <5 mm.[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 the average lifespan of adult individuals varying between 5 to more than 30 years depending on species, increasingly recognized as sensitive

Received: September 23, 2015
Revised: December 9, 2015
Accepted: December 22, 2015
Published: December 22, 2015






DOI: 10.1021/acs.est.5b04663
Environ. Sci. Technol. 2016, 50, 1924−1933

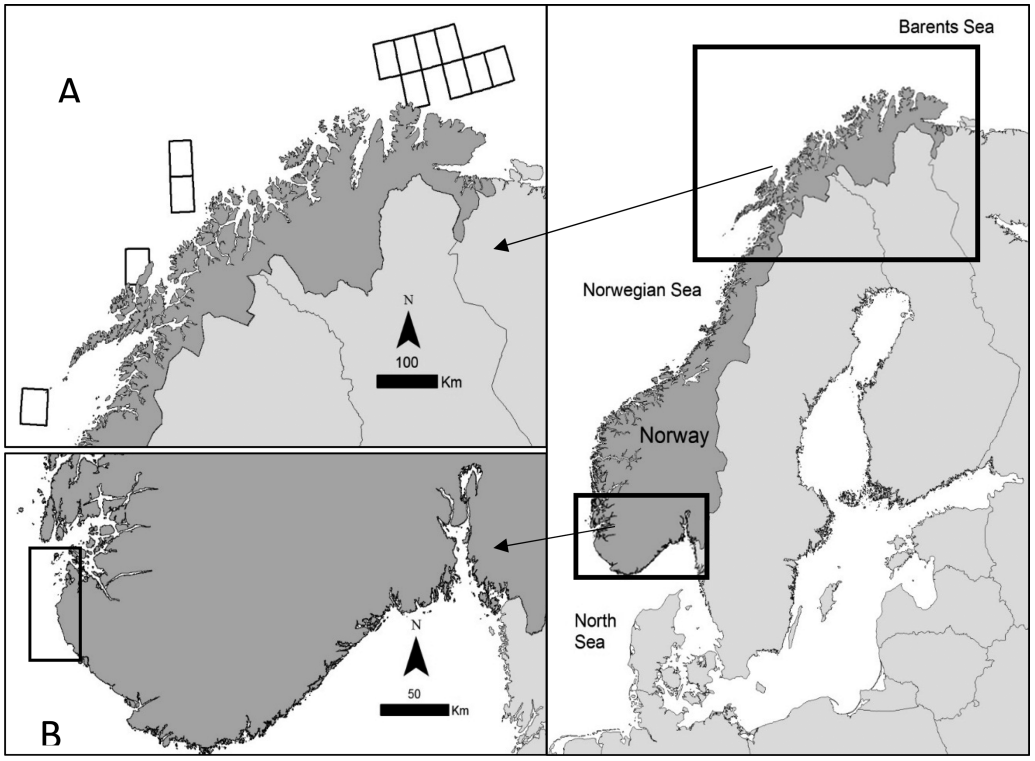



**Figure 1.** Main sampling regions in coastal Norway for the fulmars examined in this study between 2012 and 2013, Panel A: North Norway, Panel B: South Norway ($n$ = 75; $n_{south}$ = 3; $n_{north}$ = 72).

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 to report 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), <10% of the monitored population of fulmars should have >0.1 g 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 >0.1 g plastic ingested.[20] Recently, Trevail 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


DOI: 10.1021/acs.est.5b04663
*Environ. Sci. Technol.* 2016, 50, 1924−1933

off the coast of northern Norway (Figure 1, panel A) and delivered by fishermen to the Norwegian Institute for 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[20] by rinsing the proventriculus and gizzard over a 1 mm sieve. Their content was dried in a Petri dish at 35−40 °C (although the standard is room temperature) and sorted into different categories (i.a. plastic, nonplastic 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, most particles <1 mm were probably lost 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 analyzed for all three groups, while liver samples only were analyzed for the high plastic ingestion group. In addition, the plastic particles found in the stomachs of the medium and high plastic ingestion groups were analyzed for POPs.

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

**Instrumental Analysis.** A Quattro micro TM mass spectrometer (Micromass MS technologies; Manchester, U.K.) was used for analyses of PCBs and PBDEs. For more information regarding the method the reader is referred to Carlsson[29] and 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 analyzed 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 and 14% for the analyzed PCB congeners and the measured levels varied within an acceptable range (±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 and 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 the following:

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

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 SI.

## RESULTS AND DISCUSSIONS

**General Condition of the Birds.** Although the majority of the birds could be considered healthy, the body conditions ranged from high amounts 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 birds with or without plastic ($p$ = 0.268) or between the three study groups of plastic load ($p$ = 0.095).



DOI: 10.1021/acs.est.5b04663
Environ. Sci. Technol. 2016, 50, 1924−1933

**Table 1. Concentrations of POPs in Ingested Plastic Samples of Northern Fulmars in ng/g for the Different Ingestion Groups (nd: Not Detected)**

| | *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 | |

**Ingested Plastic.** Of the total of 75 birds, 14 individuals fell into the category of "no", 48 in the category of "medium" and 13 in the category of "high" plastic ingestion. In the subgroup selected for chemical analysis, the number of plastic particles per stomach averaged 6 in the group with medium plastic ingestion and 41 in the high ingestion group. The weight of the plastic found in the medium ingestion group averaged 0.08 g (median 0.04 g), which is less than the OSPAR EcoQO maximum of 0.1 g, whereas the corresponding value for the high ingestion group was 0.29 g (median 0.21 g), almost three times higher than the EcoQO limit. For the total sample of fulmars from North Norway delivered to NINA, 35% exceeded the EcoQO threshold ($N$ = 72). The particle size varied between 1.8 and 9.1 mm (mean 5.0 mm) in addition to some longer threads, excluding particles <1 mm by the applied sieve.

**Persistent Organic Pollutants in Ingested Plastic.** Of the analyzed PCBs, all PCBs besides PCB 28, 52, 101, and 189 were detected in >70% of all samples. The sumPCB concentrations ranged between 0.08 and 64.4 ng/g with a median of 2.49 ng/g demonstrating large variation among individuals. When comparing the medium and high groups of ingested plastic, a median sumPCB concentration of 2.49 ng/g was found in the high group compared to 4.03 ng/g in the medium group. In both the medium and the high ingestion group, PCB 153 was the major PCB found, followed by PCB 118 and 138 (see Table 1 for concentrations). For the DDTs, *p,p′*-DDE was the major DDT compound found with a median of 16.1 ng/g in the high plastic ingestion group and 53.4 ng/g in the medium group. The highest concentrations of sumDDTs were found in one sample from the medium ingestion group with 823 ng/g. DDE was dominating over DDT with at least a factor of 10 in all plastic samples, pointing to generally 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 and 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



DOI: 10.1021/acs.est.5b04663
*Environ. Sci. Technol.* 2016, 50, 1924−1933

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 (Figures 2 and 3).

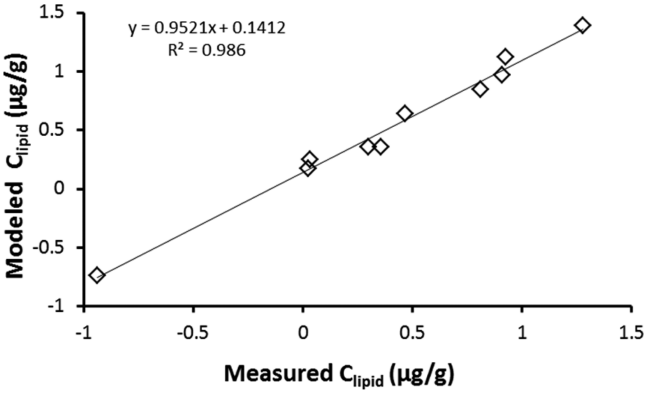



**Figure 2.** Log modeled vs log measured lipid-based concentration $C_{lipid}$ ($\mu$g/g). Fully plastic-inclusive model implemented.

**Persistent Organic Pollutants in Tissue Samples.** All targeted PCBs could be detected in the analyzed 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 and 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).

*p,p′*-DDE was the major DDT observed in muscle and liver tissue, ranging between 22.8 and 1251 ng/g ww in muscle samples (median of 228, 396, and 209 ng/g ww in high, medium and no ingestion group respectively). In liver, *p,p′*-DDE ranged between 74 and 1634 ng/g ww in the high ingestion group, with a median of 164 ng/g ww. Of the analyzed pesticides, *oxy*-chlordane, HCB, Mirex, *t*-nonachlor, and *t*-chlordane were detected in decreasing order. The concentrations of *oxy*-chlordane ranged between 112 and 154 ng/g ww in liver and between 31 and 690 ng/g ww in muscle.

PBDE 153, 47, and 154 dominated the PBDE pattern in muscle tissues. PBDE 209 was only detected in two muscle samples with 259 and 8 ng/g ww. The one elevated PBDE 209 muscle sample also demonstrated high levels of PBDE 209 in its ingested plastic, suggesting a plastic-tissue transfer in this one incident. Muscle sumPBDE concentrations varied between

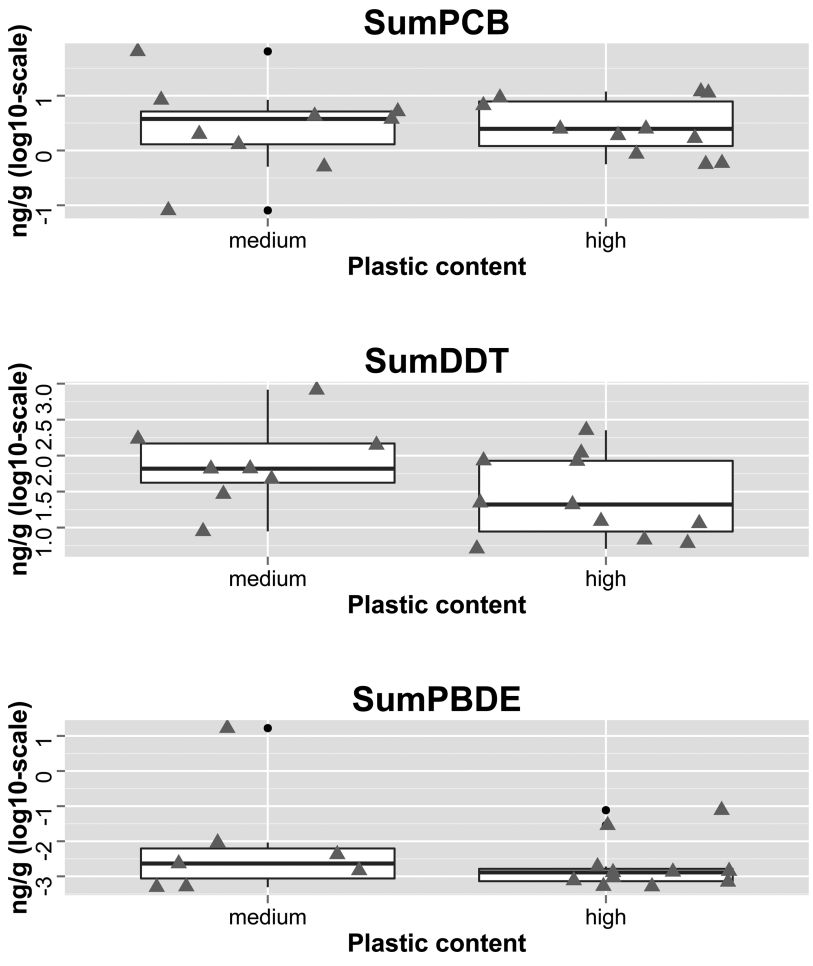



**Figure 3.** Summed concentrations of (A) PCBs, (B) DDTs, and (C) PBDEs (concentrations displayed as ng/g on a log10-scale) for the ingested plastic content in the medium and high plastic ingestion groups in ng/g plastic. (Triangles: individual concentrations; dots: outliers).



DOI: 10.1021/acs.est.5b04663
*Environ. Sci. Technol.* 2016, 50, 1924−1933

Table 2. Concentrations of POPs in Tissue Samples of Northern Fulmars in pg/g Wet Weight for All Ingestion Groups (nd: Not Detected)

| | 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 |

0.24 and 9.91 ng/g (not considering the one elevated PBDE 209 sample) with a median of 1.26, 1.51, and 0.74 ng/g ww for no, median and high ingestion samples, respectively. Liver tissue had a comparable PBDE pattern, with additional PBDE 183 and 184 detected in the majority of the samples, but no PBDE 209. Liver sumPBDE concentrations ranged between 0.28 and 3.15 ng/g ww, with a median of 0.98 ng/g ww. The differences in concentrations in muscle tissues between the plastic ingestion groups were significant for sumPBDEs (based on lipid weight normalized concentrations, Kruskal–Wallis test, $p = 0.01$), whereas the differences were not significant for sumDDTs ($p = 0.07$) and sumPCBs ($p > 0.05$) (Figure 4). For all three compound groups, the highest median concentration was found in the medium ingestion group, while the high ingestion group showed the lowest median compared to the two other groups.

**Effect of Plastic on Bioaccumulation: Statistical Evaluation of Concentration Data.** Correlation of liver and muscle concentrations in the high ingestion group resulted in Pearson 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 strongly 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


DOI: 10.1021/acs.est.5b04663
Environ. Sci. Technol. 2016, 50, 1924–1933

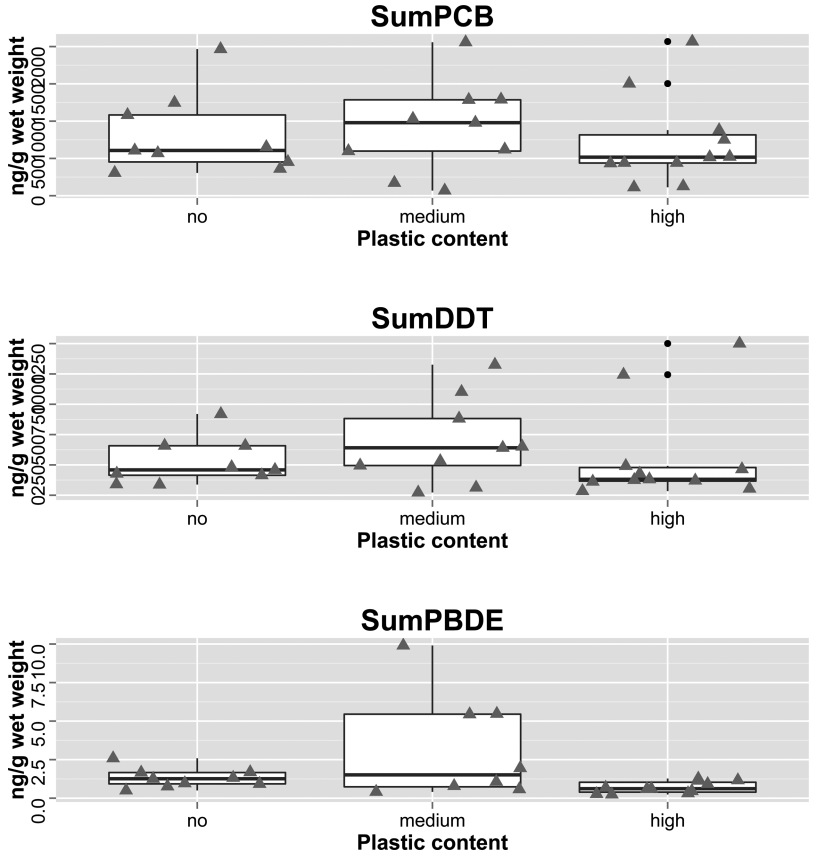



**Figure 4.** Summed wet weight concentrations of (A) PCBs, (B) DDTs, and (C) PBDEs in muscle tissue in the no, medium and high plastic ingestion groups in ng/g ww. One extreme value for sumPBDEs (267 ng/g) is excluded. (Triangles: individual concentrations; dots: outliers).

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 $2.6 \times 10^3$ (PCB 28) to $2.3 \times 10^6$ (PCB 194). It appears that the fugacities of POPs in lipids are much higher than in plastic and increase with hydrophobicity (Figure S3 and SI pp 2), which implies biomagnification from either prey or from plastic or both. A prerequisite for biomagnification is volume reduction of the ingested medium, which for ingested prey is rapid digestion of prey lipids.[35,36] Plastic inside a fulmars' stomach, however, is known to degrade very slowly due to mechanical wear, with half-lives of months.[28] Mechanical wear partly leads to increased numbers of smaller particles, which in turn can be egested, but it does not lead to a proportionally lower total volume of plastic in the intestine. Per unit of time, the volume reduction due to digestion of persistent microplastics would be much smaller than that for more digestible prey items. This implies that the observed fugacity ratio for the main part must be caused by biomagnification of POPs from prey. At the same time, gut residence times of microplastics are long, whereas POP exchange kinetics are fast and therefore sufficient to cause chemical equilibrium with ingested microplastics.[37] Given the higher fugacity of POPs in biota lipids compared to microplastics, transfer from the biota lipids to the plastic will occur, which is consistent with our hypothesis of microplastics acting as a passive sampler for POPs in the gut.

**Modeling the Contribution of Ingested Plastic to the Total Bioaccumulation of PCBs.** The uptake of PCBs by fulmars was modeled using eq 1, with a few key assumptions. The first assumption is that we modeled an "average" fulmar. This implies that average POP concentrations are used for the fulmars with and without plastic and that the selected parameters relate to the behavior of the "mean" fulmar in the sampled population. A second assumption is that the measured and modeled bioaccumulation of plastics and POPs relate to steady state and reflects the time-averaged net result of uptake and loss processes that on shorter time scales may show some seasonal and spatial fluctuations. Parameters were 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:

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



DOI: 10.1021/acs.est.5b04663
Environ. Sci. Technol. 2016, 50, 1924−1933

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 = $3.083 \times 10^{-4}$ 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 $3.083 \times 10^{-4}$ g/g × 0.0462 $d^{-1}$ = $1.43 \times 10^{-5}$ 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 $1.43 \times 10^{-5}/0.3 = 4.75 \times 10^{-5}$. 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 $4.75 \times 10^{-5}$. 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 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 Kongsfjorden (78°55′N, 11°56′E), Svalbard, Norway in 2007, were averaged.[39] The average PCB concentration varied among these diet components with a relative standard deviation of ∼50%. The loss rate parameter $k_{loss}$ was individually calibrated for each of the PCB congeners, using the known PCB concentrations measured in these diet components, and in the fulmars *without plastic*, the plastic ingestion term in eq 1 (i.e., $IR_{PL}$) was set to zero and the $a_{FOOD}$ to 0.8.[30] The optimized $k_{loss}$ values decreased linearly with $Log K_{OW}$, (Figure S2).

Finally, bioaccumulation of PCBs by the fulmars with plastic was modeled by using all aforementioned parameters including the plastic ingestion term, with $S_{PL}$= $4.75 \times 10^{-5}$ and a value for the $k_{1G}$ POP exchange rate constant parameter of 10 $d^{-1}$. This value is at the higher end of the range calculated for microplastics from first-principles,[33] as well as of the range of values measured for artificial gut fluids.[37]

The modeled lipid normalized PCB concentrations agreed very well to the measured $C_{lipid}$ ($\mu$g/g) values, with no significant difference from the 1:1 line (Figure 2). This implies that the $k_{loss}$ values from the fulmars without plastic provided an excellent agreement to the bioaccumulation data for birds with plastic. In the model, the concentration in the plastic at ingestion was equated to the value measured for plastic in stomach, which however is not the same as the concentration in the freshly ingested plastic, which may have been different. Therefore, we explored a scenario where the model was allowed to fit an optimal concentration in the plastic. This optimal PCB concentration appeared to be "zero", which implies that "no influence of PCB uptake by plastic" best explains the bioaccumulation in the birds in which a median of 0.3 g of plastic was found. This is consistent with the aforementioned inferences on ingestion rates, which showed that plastic ingestion was negligible, compared to that of regular prey. Results from this second scenario were indistinguishable from those in Figure 2 and therefore not plotted separately. To explore the sensitivity of the model to the concentration in ingested plastic, we also explored a third scenario in which the concentrations in ingested plastic were taken 1000 times higher than the values measured for plastic in the stomach. The intercept of the resulting regression between modeled and measured values now moved away from the 1:1 line (Figure S1). This poorer fit, however, was still not dramatic due to the unimportance of plastic ingestion compared to that of regular prey.

**General Discussion and Implications.** For the first time, POP concentrations in tissues and ingested plastic from the same individual were analyzed for fulmars in Norway. Earlier studies on the diving behavior of chick-rearing fulmars in Shetland, U.K., showed that fulmars forage on their prey through shallow dives ($N$ = 97 per day); 85% of these dives less than 1 m deep, potentially exposing them to floating plastic debris,[40] and they may also pick floating plastic particles when laying on the surface between dives. POP concentrations have been reported in fulmars from Norway before, indicating lower PCB and DDT concentrations but higher PBDE concentrations compared to our study.[41−46]

In our study, we have provided several lines of evidence suggesting that ingested microplastics can act as "negligible depletion" passive samplers for POPs originating from ingested food. First, we found that POP concentrations in fulmars were not linked to the magnitude of their stomach plastic concentrations, which would have been the case if plastic acted as a substantial carrier of the POPs to the fulmars. Lack of unidirectional relationships between these variables has also been demonstrated in one other study,[25] supporting our findings are not incidental. Second, we found that POP concentrations in plastic correlated strongly with POP concentrations in fulmars, which implies that chemical transfer still does occur. Third, we found that chemical fugacities in plastic were lower than that in the bird's lipids, which would suggest transfer of POPs to the plastic i.e., as passive samplers, rather than the other way around. This would explain the aforementioned correlation, and might also explain such correlations reported in earlier studies (e.g., ref 26). Fourth, we quantified the fluxes of POPs entering fulmars using a dynamic bioaccumulation model. We calculated that the flux of POPs by ingestion of natural prey would be at least 21 000 times higher than the flux of POPs by ingestion of plastic. The uptake from plastic thus is calculated to be overwhelmed by ingestion via natural pathways i.e. by ingestion via feed, which also has been recognized by recent modeling studies[33,34,48] and in 2015 by the GESAMP Working Group 40 on Marine Litter.[49] The suggested dominance of plastic-mediated internal exposure to PBDE 209 in particular as stated by Tanaka et al., could not be observed when applying average data and in comparison with individuals with no ingested plastic in their guts as a control.[26]

In summary, we conclude that bioaccumulation of POPs by fulmars is mainly governed by the ingestion of natural prey. POPs taken up via ingested plastics may equilibrate readily in the intestines of the birds, making a negligible contribution to accumulation, yet absorbing POPs from the ingested food simultaneously such that POP profiles in plastic reflect the profiles observed in tissues. Since the here applied sampling methodology excluded particles smaller than 1 mm, follow-up studies are recommended to include such smaller-sized particles.

It has been generally recognized that it is difficult to infer causal relationships from correlative evidence. Here we showed that correlations among POP concentrations in plastic and



DOI: 10.1021/acs.est.5b04663
Environ. Sci. Technol. 2016, 50, 1924−1933

tissues do not necessarily imply that plastic acts as a substantial carrier for POPs. By combining correlations among POP concentrations, differences between plastic ingestion subgroups, fugacity calculations and bioaccumulation modeling, we showed that ingested plastic is due to its relatively long residence time more likely to act as a passive sampler, reflecting the POP profiles as they occur in the gastro-intestinal tract. Although this study was specific for birds, it is likely that microplastics may act as passive samplers (rather than as vectors for bioaccumulation) also in other species, like invertebrates or fish. However, potential harm caused by ingested plastic due to physical damage or other plastic related chemicals cannot be excluded.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b04663.

Text, figures, and tables addressing (i) the model parameters, least-squares used in the modeling 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. (PDF)

## ■ AUTHOR INFORMATION


### Corresponding Author

*Phone: +47 777 50 39; fax: +47 777 50375; e-mail: dorte.herzke@nilu.no (D.H.).


### Notes

The authors declare no competing financial interest.

## ■ ACKNOWLEDGMENTS

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. Cover photo courtesy of Tycho Anker-Nilssen, Norwegian Institute for Nature Research - NINA.

## ■ REFERENCES


(1) Jambeck, J. R.; Geyer, R.; Wilcox, C.; Siegler, T. R.; Perryman, M.; Andrady, A.; Narayan, R.; Law, K. L. Plastic waste inputs from land into the ocean. *Science* **2015**, *347*, 768−771.

(2) PlasticEurope. *Plastics—the Facts 2014. An analysis of European plastics production, demand and waste data*. 2014, Brussels, Belgium.

(3) Hammer, J.; Kraak, M. H. S.; Parsons, J. R. Plastics in the Marine Environment: The Dark Side of a Modern Gift. *Rev. Environ. Contam. Toxicol.* **2012**, *220*, 1−44.

(4) Law, K. L.; Moret-Ferguson, S.; Maximenko, N. A.; Proskurowski, G.; Peacock, E. E.; Hafner, J.; Reddy, C. M. Plastic Accumulation in the North Atlantic Subtropical Gyre. *Science* **2010**, *329*, 1185−1188.

(5) Koelmans, A. A.; Besseling, E.; Shim, W. J. Nanoplastics in the aquatic environment. Critical Review. In Bergmann, M., Gutow, L., Klages, M., Eds.; *Marine Anthropogenic Litter*; Springer: Berlin, 2015; pp 325−340.

(6) Cozar, A.; Echevarria, F.; Gonzalez-Gordillo, J. I.; Irigoien, X.; Ubeda, B.; Hernandez-Leon, S.; Palma, A. T.; Navarro, S.; Garcia-de-Lomas, J.; Ruiz, A.; Fernandez-de-Puelles, M. L.; Duarte, C. M. Plastic debris in the open ocean. *Proc. Natl. Acad. Sci. U. S. A.* **2014**, *111*, 10239−10244.

(7) Eriksen, M.; Lebreton, L. C. M.; Carson, H. S.; Thiel, M.; Moore, C. J.; Borerro, J. C.; Galgani, F.; Ryan, P. G.; Reisser, J. Plastic Pollution in the World's Oceans: More than 5 Trillion Plastic Pieces Weighing over 250,000 Tons Afloat at Sea. *PLoS One* **2014**, *9* (12), e111913.

(8) Law, K. L.; Moret-Ferguson, S. E.; Goodwin, D. S.; Zettler, E. R.; De Force, E.; Kukulka, T.; Proskurowski, G. Distribution of Surface Plastic Debris in the Eastern Pacific Ocean from an 11-Year Data Set. *Environ. Sci. Technol.* **2014**, *48*, 4732−4738.

(9) NIVA. Microplastics in Marine Environments: Occurrence, Distribution and Effects. 2014, *Report M-319*, Norwegian Institute for Water Research, Oslo, Norway.

(10) Obbard, R. W.; Sadri, S.; Wong, Y. Q.; Khitun, A. A.; Baker, I.; Thompson, R. C. Global warming releases microplastic legacy frozen in Arctic Sea ice. *Earth's Future* **2014**, *2*, 315−320.

(11) Besseling, E. E. M.; Foekema, J. A.; van Franeker, M.; Leopold, E. L.; Bravo Rebolledo, S.; Kuehn, L.; Mielke, E.; Heße, J.; IJzer, P.; Kamminga, A. A.; Koelmans. Microplastic in a macro filter feeder: humpback whale Megaptera novaeangliae. *Mar. Pollut. Bull.* **2015**, *95*, 248−252.

(12) Colabuono, F. I.; Barquete, V.; Domingues, B. S.; Montone, R. C. Plastic ingestion by Procellariiformes in Southern Brazil. *Mar. Pollut. Bull.* **2009**, *58*, 93−96.

(13) Collignon, A.; Hecq, J. H.; Galgani, F.; Voisin, P.; Collard, F.; Goffart, A. Neustonic microplastic and zooplankton in the North Western Mediterranean Sea. *Mar. Pollut. Bull.* **2012**, *64*, 861−864.

(14) Fort, J.; Robertson, G. J.; Gremillet, D.; Traisnel, G.; Bustamante, P. Spatial Ecotoxicology: Migratory Arctic Seabirds Are Exposed to Mercury Contamination While Overwintering in the Northwest Atlantic. *Environ. Sci. Technol.* **2014**, *48*, 11560−11567.

(15) Piatt, J. F.; Harding, A. M. A.; Shultz, M.; Speckman, S. G.; Van Pelt, T. I.; Drew, G. S.; Kettle, A. B. Seabirds as indicators of marine food supplies: Cairns revisited. *Mar. Ecol.: Prog. Ser.* **2007**, *352*, 221−234.

(16) Bond, A. L.; Lavers, J. L. Effectiveness of emetics to study plastic ingestion by Leach's Storm-petrels (Oceanodroma leucorhoa). *Mar. Pollut. Bull.* **2013**, *70*, 171−175.

(17) Rebolledo, E. L. B.; van Franeker, J. A.; Jansen, O. E.; Brasseur, S. M. J. M. Plastic ingestion by harbour seals (Phoca vitulina) in The Netherlands. *Mar. Pollut. Bull.* **2013**, *67*, 200−202.

(18) Avery-Gomm, S.; O'Hara, P. D.; Kleine, L.; Bowes, V.; Wilson, L. K.; Barry, K. L. Northern fulmars as biological monitors of trends of plastic pollution in the eastern North Pacific. *Mar. Pollut. Bull.* **2012**, *64*, 1776−1781.

(19) Kuhn, S.; van Franeker, J. A. Plastic ingestion by the northern fulmar (Fulmarus glacialis) in Iceland. *Mar. Pollut. Bull.* **2012**, *64*, 1252−1254.

(20) van Franeker, J. A.; Blaize, C.; Danielsen, J.; Fairclough, K.; Gollan, J.; Guse, N.; Hansen, P. L.; Heubeck, M.; Jensen, J. K.; Le Guillou, G.; Olsen, B.; Olsen, K. O.; Pedersen, J.; Stienen, E. W. M.; Turner, D. M. Monitoring plastic ingestion by the northern fulmar Fulmarus glacialis in the North Sea. *Environ. Pollut.* **2011**, *159*, 2609−2615.

(21) van Franeker, J. A. Plastic Ingestion in the North-Atlantic Fulmar. *Mar. Pollut. Bull.* **1985**, *16*, 367−369.

(22) Blight, L. K.; Burger, A. E. Occurrence of plastic particles in seabirds from the eastern North Pacific. *Mar. Pollut. Bull.* **1997**, *34*, 323−325.

(23) Provencher, J. F.; Gaston, A. J.; Mallory, M. L.; O'Hara, P. D.; Gilchrist, H. G. Ingested plastic in a diving seabird, the thick-billed murre (Uria lomvia), in the eastern Canadian Arctic. *Mar. Pollut. Bull.* **2010**, *60*, 1406−1411.

(24) OSPAR Commission. Background document for the EcoQO on plastic particles in stomachs of seabirds. 2008, London, UK: 355/2007.

(25) Trevail, A. M.; Gabrielsen, G. W.; Kühn, S.; Bock, A.; van Franeker, J. A. Plastic Ingestion by Northern Fulmars, Fulmarus




DOI: 10.1021/acs.est.5b04663
*Environ. Sci. Technol.* 2016, 50, 1924−1933

glacialis, in Svalbard and Iceland, and Relationships between Plastic Ingestion and Contaminant Uptake. 2014 *Brief Report Series*; Norwegian Polar Institute.
(26) Tanaka, K.; Takada, H.; Yamashita, R.; Mizukawa, K.; Fukuwaka, M.; Watanuki, Y. Accumulation of plastic-derived chemicals in tissues of seabirds ingesting marine plastics. *Mar. Pollut. Bull.* **2013**, *69*, 219−22.
(27) van Franeker, J. A. *Save the North Sea Fulmar-Litter-EcoQO Manual part 1: Collection and dissection procedures*. Alterra Report 2004, *672*: Wageningen.
(28) Ryan, P. G. How quickly do albatrosses and petrels digest plastic particles? *Environ. Pollut.* **2015**, *207*, 438−440.
(29) Carlsson, P. Polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and perfluorinated alkylated substances (PFASs) in traditional seafood items from western Greenland. *Environ. Sci. Pollut. Res.* **2014**, *21*, 4741−4750.
(30) Hendriks, A. J.; van der Linde, A.; Cornelissen, G.; Sijm, D. T. H. M. The power of size. 1. Rate constants and equilibrium ratios for accumulation of organic substances related to octanol-water partition ratio and species weight. *Environ. Toxicol. Chem.* **2001**, *20*, 1399−1420.
(31) Selck, H. K.; Drouillard, K.; Eisenreich, K.; Koelmans, A. A.; Palmqvist, A.; Ruus, A.; Salvito, D.; Schultz, I.; Stewart, R.; Weisbrod, A.; van den Brink, N. W.; van den Heuvel-Greve, M. Explaining differences between bioaccumulation measurements in laboratory and field data using a probabilistic modeling approach. *Integr. Environ. Assess. Manage.* **2012**, *8*, 42−63.
(32) Thomann, R. V.; Connolly, J. P.; Parkerton, T. F. An Equilibrium-Model of Organic-Chemical Accumulation in Aquatic Food Webs with Sediment Interaction. *Environ. Toxicol. Chem.* **1992**, *11*, 615−629.
(33) Koelmans, A. A. Modeling the role of microplastics in Bioaccumulation of organic chemicals to marine aquatic organisms. A Critical Review. In: Bergmann, M., Gutow, L., Klages, M., Eds.; *Marine Anthropogenic Litter*; Springer: Berlin, 2015, ISBN 978-3-319-16510-3, doi 10.1007/978-3-319-16510-3.
(34) Koelmans, A. A.; Besseling, E.; Wegner, A.; Foekema, E. M. Plastic as a Carrier of POPs to Aquatic Organisms: A Model Analysis. *Environ. Sci. Technol.* **2013**, *47*, 7812−7820.
(35) Macdonald, R.; Mackay, D.; Hickie, B. Contaminant amplification in the environment. *Environ. Sci. Technol.* **2002**, *36*, 462A.
(36) Foster, K. L.; Wang, S. W.; Mackay, D.; Mallory, M. L.; Blais, J. M. Preliminary Assessment of Avian Stomach Oils: A Vector of Contaminants to Chicks and Potential for Diet Analysis and Biomonitoring. *Environ. Sci. Technol.* **2010**, *44*, 6869−6878.
(37) Bakir, A.; Rowland, S. J.; Thompson, R. C. Enhanced desorption of persistent organic pollutants from microplastics under simulated physiological conditions. *Environ. Pollut.* **2014**, *185*, 16−23.
(38) Barrett, R. T.; Anker-Nilssen, T.; Gabrielsen, G. W.; Chapdelaine, G. Food consumption by seabirds in Norwegian waters. *ICES J. Mar. Sci.* **2002**, *59*, 43−57.
(39) Evenset, A.; Hallanger, I. G.; Tessmann, M.; Warner, N.; Ruus, A.; Borgå, K.; Gabrielsen, G. W.; Christensen, G.; Renaud, P. E. Seasonal variation in contaminant accumulation in Arctic marine benthic food webs. *Sci. Total Environ.* **2016**, *542*, 108−120.
(40) Garthe, S.; Furness, R. W. Frequent shallow diving by a Northern Fulmar feeding at Shetland. *Waterbirds* **2001**, *24*, 287−289.
(41) Fangstrom, B.; Athanasiadou, M.; Athanassiadis, I.; Bignert, A.; Grandjean, P.; Weihe, P.; Bergman, A. Polybrominated diphenyl ethers and traditional organochlorine pollutants in fulmars (Fulmarus glacialis) from the Faroe Islands. *Chemosphere* **2005**, *60*, 836−843.
(42) Foster, K. L.; Mallory, M. L.; Hill, L.; Blais, J. M. Pcb and Organochlorine Pesticides in Northern Fulmars (Fulmarus Glacialis) from A High Arctic Colony: Chemical Exposure, Fate, and Transfer to Predators. *Environ. Toxicol. Chem.* **2011**, *30*, 2055−2064.
(43) Helgason, L. B.; Arukwe, A.; Gabrielsen, G. W.; Harju, M.; Hegseth, M. N.; Heimstad, E. S.; Jorgensen, E. H.; Mortensen, A. S.; Wolkers, J. Biotransformation of PCBs in Arctic seabirds: Characterization of phase I and II pathways at transcriptional, translational and activity levels. *Comp. Biochem. Physiol., Part C: Toxicol. Pharmacol.* **2010**, *152*, 34−41.
(44) Jorundsdottir, H.; Lofstrand, K.; Svavarsson, J.; Bignert, A.; Bergman, A. Organochlorine Compounds and Their Metabolites in Seven Icelandic Seabird Species - a Comparative Study. *Environ. Sci. Technol.* **2010**, *44*, 3252−3259.
(45) Knudsen, L. B. Halogenated organic contaminants and mercury in northern fulmars (Fulmarus glacialis): levels, relationships to dietary descriptors and blood to liver comparison. *Environ. Pollut.* **2007**, *146*, 25−33.
(46) Nøst, T. H.; Helgason, L. B.; Harju, M.; Heimstad, E. S.; Gabrielsen, G. W.; Jenssen, B. M. Halogenated organic contaminants and their correlations with circulating thyroid hormones in developing Arctic seabirds. *Sci. Total Environ.* **2012**, *414*, 248−256.
(47) Guzzo, M. M.; Eckbo, N. H.; Gabrielsen, G. W.; Fisk, A. T.; Hylland, K.; Borga, K. Persistent organic pollutant concentrations in fledglings of two arctic seabird species. *Environ. Pollut.* **2014**, *184*, 414−418.
(48) Bakir, A.; O'Connor, I. A.; Rowland, S. J.; Hendriks, A. J.; Thompson, R. C. Relative importance of microplastics as a pathway for the transfer of persistent organic pollutants to marine life. Chapter 3. In: O'Connor I. A. *Modelling the oral uptake of chemicals: the role of plastic, passive diffusion and transport proteins*. PhD thesis; Radboud University Nijmegen: The Netherlands, 2014. ISBN: 978-94-6259-386-2.
(49) GESAMP. Sources, fate and effects of microplastics in the marine environment: a global assessment. Kershaw, P. J., Ed.; (IMO/FAO/UNESCO-IOC/UNIDO/WMO/IAEA/UN/UNEP/UNDP Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection). Reports and Studies. GESAMP 2015, No. *90*, 96 p.
(50) Fangel, K.; Aas, Ø.; Vølstad, J. H.; Bærum, K. M.; Christensen-Dalsgaard, S.; Nedreaas, K.; Overvik, M.; Wold, L. C.; Anker-Nilssen, T. Assessing incidental bycatch of seabirds in Norwegian coastal commercial fisheries: Empirical and methodological lessons. *Glob. Ecol. Conserv.* **2015**, *4*, 127−136.

## NOTE ADDED AFTER ASAP PUBLICATION

The values in the Ingested Plastic section were changed in the version of this article published January 14, 2016. The corrected version published January 22, 2016.


DOI: 10.1021/acs.est.5b04663
*Environ. Sci. Technol.* 2016, 50, 1924−1933