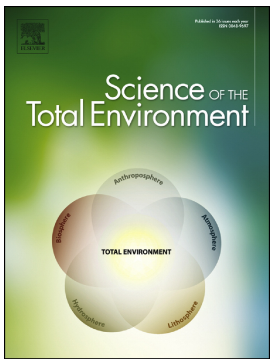

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Snow buntings (*Plectrophenax nivealis*) as bio-indicators for exposure differences to legacy and emerging persistent organic pollutants from the Arctic terrestrial environment on Svalbard.

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Abstract

Eggs of snow buntings (Plectrophenax nivealis) were applied as a bio-indicator to examine differences in exposure to legacy persistent organic pollutants (POPs) and perflouroalkyl subtances (PFAS) from the terrestrial environment surrounding the settlements of Longyearbyen, Barentsburg and Pyramiden on Svalbard, Norway. Significantly higher concentrations of summed polychlorinated biphenyls (sumPCB7) in eggs collected from Barentsburg (2980 ng/g lipid weight (lw)) and Pyramiden (3860 ng/g lw) compared to Longyearbyen (96 ng/g lw) are attributed to local sources of PCBs within these settlements. Similar findings were observed for p,p'- dichlorodiphenyldichloroethylene (p,p'-DDE) where higher median concentrations observed in Pyramiden (173 ng/g lw) and Barentsburg (75 ng/g lw) compared to Longyearbyen (48 ng/g lw) may be influenced by guano inputs from breeding seabird populations, although other point sources cannot be ruled out. Concentrations of perfluorooctane sulphonate (PFOS) and several perfluorinated carboxylic acids (PFCAs) in snow bunting eggs were found to be statistically higher in the populated settlements of Longyearbyen and Barentsburg compared to the abandoned Pyramiden. Narrow foraging ranges of snow buntings during breeding season was useful in assessing point sources of exposure for PCBs and PFAS at particular sites with extreme differences observed between nest locations. SumPCB7 concentrations ranged from 2 μg/g ww to below detection limits between nest sites located less than a kilometer from each other in Pyramiden. Similar findings were observed in Longyearbyen, where several PFCAs ranged from 2 to 55 times higher between nest sites with similar spatial distances. These findings indicate that snow buntings can be a useful bio-indicator offering high spatial resolution for contaminant source apportionment in terrestrial environments on Svalbard

Keywords: persistent organic pollutants, birds, terrestrial exposure, Arctic, contaminated soils

1. INTRODUCTION

Long-range transport and accumulation of persistent organic pollutants (POPs, i.e., polychlorinated biphenyls (PCBs), organochlorine (OC) pesticides) and newer generation pollutants such as perflouroalkylated substances (PFAS) in Arctic biota has been well established (AMAP, 2004; Giesy and Kannan, 2001; Martin et al., 2004; Verreault et al., 2010). A majority of studies have placed focus on the marine environment in Arctic regions as it is the dominant deposition/storage compartment of atmospherically transported pollutants and is an essential source of prey for the diet of Arctic top predators'. High levels of POPs and PFAS have been reported in top marine predators on Svalbard, such as polar bears (*Ursus maritimus*), glaucous gulls (*Larus hyperboreus*) and ivory gulls (*Pagophila eburnea*) (Bytingsvik et al., 2012a; Bytingsvik et al., 2012b; Letcher et al., 2010; Miljeteig et al., 2009; Verreault et al., 2010). However, only a handful of studies have investigated exposure and bioaccumulation of POPs and PFAS within a terrestrial Arctic food web (Bossi et al., 2015; Kelly and Gobas, 2001; Müller et al., 2011), with no information regarding bioaccumulation in terrestrial feeding animals on Svalbard.

Transport and deposition of POPs and PFAS to the Arctic terrestrial environments can occur via atmospheric currents (wet and/or dry deposition) (Kelly and Gobas, 2001; Young et al., 2007) and/or indirectly from oceanic currents via sea spray (Webster and Ellis, 2010). However, local sources of contamination may also be present and act as significant sources to terrestrial biota. Several human settlements exist on Svalbard, which include Longyearbyen, Barentsburg and Pyramiden. High concentrations of PCBs have been reported in the soils surrounding Barentsburg and Pyramiden with concentrations reaching 29 mg/kg and 14 mg/kg, respectively

(Jartun et al., 2009a, b). This high contamination is attributed to inputs from local sources (building paint, small capacitors, electrical waste, building refuse, and scrap metals) within these settlements resulting in a significant exposure risk to local terrestrial wildlife. In addition, old dumping grounds and landfills have also been reported as local sources to OC pesticides (Granberg et al., 2017). Several reports have documented increasing levels of poly- and perfluorinated substances (PFASs) in Arctic biota, but trends vary among the various PFAS due to changing regulations and subsequent emissions and substitution scenarios with increasing, decreasing and stabilizing trends being observed in different environmental media and species (Land et al., 2018). Declining trends have been observed for some PFAS compounds (i.e., perfluorooctane sulphonate (PFOS), perfluorooctanoic acid (PFOA), while several other PFAS compounds continue to be released into the environment where exposure levels continue to increase (Muir et al., 2016; Routti et al., 2017; Wong et al., 2018). As PFAS are used in several commercial and/or household products and applications (e.g., lubricants, textiles, leathers and fire-fighting foams (Hekster and Voogt, 2002)), emissions of PFAS may also occur from settlements present on Svalbard, and thus, potentially biomagnify in Arctic terrestrial food chains (Müller et al., 2011). Exposure and accumulation of PFAS within marine based organisms has been well documented from Svalbard, but little information exists regarding exposure from the terrestrial environment. Recent findings have reported PFAS accumulation in Arctic foxes (*Vulpes lagopus*) on Svalbard (Aas et al., 2014). However, Arctic foxes are opportunistic feeders with varying feeding behavior and feed from both the terrestrial and marine environments depending on prey availability (Eide et al., 2005; Roth, 2003). Thus, contribution of the terrestrial environment to overall accumulated body burden cannot be ascertained.

The snow bunting (*Plectrophenax nivalis*), which is a passerine bird, is one of two terrestrial bird species that breed on Svalbard. Due to their long migratory route across the North-Atlantic from Siberia via Novaya Zemlya, Kola or Northern Norway to Svalbard (Snell et al., 2018), snow buntings are considered income breeders, where energy allocated for breeding originates from recently ingested resources (Drent and Daan, 1980), e.g., seeds, insects and spiders (Skjøstad, 2008). As snow buntings replenish their energy storages after the spring migration for reproduction, their eggs are a useful indicator of local contaminant exposure on Svalbard. In addition, small home ranges, territories and foraging areas of passerine birds (Dauwe et al., 2003) offers high spatial resolution in assessing differences in local pollutant exposure from terrestrial environments. This makes snow buntings an ideal bio-indicator for source apportionment of POPs and PFAS terrestrial-based exposure on Svalbard.

2. MATERIAL AND METHODS

2.1. Study sites

Longyearbyen (78°22'N, 15°65'E) was a coal mining town until its status gradually changed in the 1990's and became an ordinary civil community in 2002, although some coal mining activity remains. It now serves as the main entry point into Svalbard, hosting the largest airport and represents the largest community on Svalbard (approximately 2100 inhabitants). Barentsburg (78°04'N, 14°13'E) is a coal mining town and is the second largest settlement on Svalbard with about 500 inhabitants, but contained as many as 1500 during the Soviet era. Pyramiden

(78°41'N, 16°24'E) also served as a coal mining town until it was abandoned in 1998, where only a few inhabitants remain for tourism purposes and maintaining the current infrastructure.

2.2. Sampling

Eggs of snow buntings were collected in Longyearbyen (n=8), Barentsburg (n=7) and Pyramiden (n=9), in June 2010 and 2011 (Table A1). One egg was randomly collected from each nest, and wrapped in aluminium foil and zip lock bags. The clutch size was noted, the eggs were measured (length and width), and the volume of the eggs were calculated according to Hoyt (1979) (Table 1). The eggs were frozen at -20 °C within a few hours after collection until analysis. Permission to sampling of the eggs was provided by The Governor of Svalbard, 2011/00488-14.

2.3. Sample extraction and analysis

Sample extraction for PCBs, hexachlorobenzene (HCB), organochlorine pesticides (chlordanes, nonachlors, hexachlorocyclohexanes (HCHs), dichlorodiphenyltrichloroethane (DDT) and associated breakdown/metabolite products dichlorodiphenyldichloroethane (DDD) and Dichlorodiphenylcichloroethylene (DDE)) and PFAS; perfluorooctanesulfonamide (PFOSA), perfluorobutanesulfonate (PFBS), perfluorohexanesulfonate (PFHxS), perfluorooctanesulfonate (PFOS), perfluorodecanesulfonate (PFDcS), perfluorohexanoate (PFHxA), perfluoroheptanoate (PFHpA), perfluorooctanoate (PFOA), perfluorononanoate (PFNA), perfluorodecanoate (PFDcA), perfluoroundecanoate (PFUnA), perfluorododecanoate (PFDoA), perfluorotridecanoate (PFTrA), and perfluorotetradecanoate (PFTeA). perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA),

perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTrDA) using previously published techniques (Ahrens et al., 2011; Huber et al., 2015). Prior to extraction and analysis, the eggshell was removed and the egg content was homogenized using a stainless steel Ultra-turrax homogenizer. Lipid content in the eggs was determined gravimetrically and is presented in Table 1. Contaminant analyses were conducted at the Norwegian Institute of Air Research (NILU), Fram Centre, Tromsø, Norway. Further information on the sample extraction, analysis and quality assurance are described in the Supporting Information (A1).

2.4. Statistical analysis

The descriptive statistics of median, mean and standard error of mean (SE) were calculated only for each compound when more than 60 % of the samples were found above the limit of detection (LOD), defined as the 3 times the background instrumental noise/or blank response. Only these compounds were considered in the statistical analyses and the values below LOD were given new value of half the corresponding LOD to avoid missing values in the statistical analyses (Bernhoft et al., 1997). Assessment of variable residuals for constant variance and normal distribution was carried out by entering residuals in quantile-quantile (q-q) plots, a graphical technique used to visualize whether the variable approached normal distribution. As the sample size within each settlement was small, the approach toward the normal distribution was not achieved. The Kruskal-Wallis test was thus used to test if significant differences in contaminant concentration existed between the three settlements followed by rank Tukey HSD to distinguish settlements. A small, but significant difference in egg lipid content among the locations (chi-squared = 9.2, $p = 0.03$) confirms the use of lipid weight (lw) concentrations for the lipid soluble contaminants. The significance level was set at < 0.05 and all statistics were performed with the

statistical software R, version 3.3.2 (R_Core_Team, 2016). No adjustments were made for multiple comparison, since such comparisons decrease the chances of rejecting the null hypothesis that are not null (type II error) (Rothman, 1990).

3. RESULTS AND DISCUSSION

3.1. Spatial distribution of legacy and emerging contaminants

3.1.1. PCBs

Seven PCB-congeners (ΣPCB_7: PCB-28/31, -52, -101, -118, -138, -153, -180) were analysed for and detected in greater than 60 % of samples collected. Concentration of PCBs in snow bunting eggs collected in Pyramiden and Barentsburg were significantly higher compared to eggs collected in Longyearbyen both on $\sum PCB_7$ and individual congener concentration basis (chi-squared = 8.9, p = 0.01) (Table 1, Table A2). This is attributed to differences in soil exposure between the settlements, where soils from Barentsburg and Pyramiden are highly contaminated compared to Longyearbyen (Jartun et al. 2009a, b). The mean concentration of $\sum PCB_7$ in the snow bunting eggs from Barentsburg (4930 ± 1620 ng/g lipid wt (350 ± 322 ng/g ww)) and Pyramiden (12800 ± 6310 ng/g lipid wt (604 ± 821 ng/g ww)) are 40 and 120 times higher, respectively, compared to concentrations found in the Longyearbyen (106 ± 21 ng/g lipid wt (6.93 ± 2.88 ng/g ww), surpassing international health protection guidelines for wildlife on a wet weight basis (> 100 ng/g ww) (Braune et al., 1999). Comparison of PCB congener profiles found within eggs show clear differences between the Norwegian (i.e., Longyearbyen) and Russian

(i.e., Barrentsburg and Pyramiden) settlements (Figure 1A). The relative contribution to the ΣPCB_7 for PCB 52, 101, 118 and 138 in eggs from the Russian settlements was higher compared to those from the Norwegian settlements, whereas the opposite was observed for PCB 153 and 180 (Figure 1A). This is consistent with PCB-congener profiles observed in soil within the Russian settlements and Longyearbyen (Jartun et al. 2009a, b), and indicates contaminant burden reflects recent consumption from the local environment. Difference in congener patterns in eggs between settlements can be attributed to usage of different PCB technical mixtures. The technical mixture Sovol was produced and used within the former Soviet Union. Its chemical composition contained higher relative percentage of the tetra (i.e., PCB 52) and penta-chlorinated congeners (PCB 101 and 118) compared to technical mixtures used within Europe (i.e., Aroclor 1260, 1262) in which higher chlorinated congeners (i.e., PCB 153, 180) were used in greater proportions (Takasuga et al., 2006). Comparison of congener profiles observed in eggs to congener profiles in PCB technical mixtures (Aroclor 1260, 1262 and Sovol) shows that eggs from the Russian settlements closely resemble the Sovol technical mixture (Figure 1B). This pattern is also observed in macro-benthos in Grønfjorden and Billefjorden outside Barentsburg and Pyramiden, respectively (Hop et al., 2001), as well as in liver samples from glaucous gulls (*Larus hyperboreus*) in Barentsburg (Sagerup et al., 2009). Eggs collected from Longyearbyen display PCB congener profiles similar to Aroclor mixtures, with exception to PCB 180. Lower PCB 180 concentrations found within eggs compared to the technical mixture can be attributed to slower diffusion into lipid stores due to its high molecular weight (Fisk et al., 1998) and thus subsequent uptake into eggs through maternal transfer.

Concentrations within eggs collected from the Russian settlements were similar or higher than other findings reported in snow buntings in Arctic regions. The mean whole body $\sum PCB_{37}$ concentrations in 18 snow buntings from Cape Vera, Devon Island, Nunavut, Canada was 5770 ± 1480 ng/g lw (Choy et al., 2010). These concentrations were attributed to contaminant inputs from seabird guano to the local terrestrial environment and are not typically observed in Arctic samples. Within the same study, concentrations of PCBs were below detection limits in snow buntings collected outside seabird colony areas. It should be noted that kittiwakes (*Rissa tridacatyla*) breed within the local buildings at Barentsburg and Pyramiden (Miljeteig and Gabrielsen, 2009). A greater population exists in Pyramiden as most of the infrastructure here is abandoned and birds are able to breed undisturbed, possibly explaining the higher levels observed in Pyramiden compared to Barentsburg. Thus, in addition to contaminated soil from local releases of PCBs, guano from kittiwakes may constitute an additional source of PCBs to the local terrestrial environment.

3.1.2. Organochlorine pesticides

Of the pesticides selected for analysis, only HCB, *p,p*'-DDE and *trans*-nonachlor were detected in the samples (Table 1, Table A3). Concentrations of HCB and *trans*-nonachlor were similar among all the settlements with no statistical differences observed ($\rho > 0.6$). However, significant differences were observed between the settlements for *p,p'*-DDE with the highest concentrations found in snow bunting eggs originating from Pyramiden ($\rho < 0.01$, Table 1). Nevertheless, the concentration of *p,p'*-DDE in eggs from Pyramiden (268 ng/g lipid wt) was far lower than whole body concentrations in snow buntings from Cape Vera, Canada in 2006-07 (3380 ng/g lw) and from West-Greenland in 1973 (1900 ng/g lw) (Burnham and Mattox, 1984; Choy et al., 2010).

This difference can be attributed to the egg vs. tissue concentration comparison, although similar spatial trends have been observed for DDT (and related metabolites) in ringed seals (*Pusa hispida*) and polar bear (*Urus maritimus*) adipose tissue with higher levels observed in the Canadian Arctic compared to the European Arctic (Muir et al., 2000; Norstrom et al., 1998). Concentration of *p,p*'-DDE was found to be statistically different between Pyramiden and Longyearbyen with higher concentrations occurring in Pyramiden (Tukey HSD $\rho < 0.001$). Eggs collected from Barentsburg also contained higher concentration of *p,p*'-DDE compared to Longyearbyen, but lower compared to Pyramiden on an average and median basis. However, no statistical differences was observed between settlements ($\rho = 0.08$ and 0.10, respectively), which is likely attributed to the small sample size of eggs collected. The difference observed in *p,p*'-DDE contamination between Longyearbyen and the two Russian settlements may be attributed to guano inputs. As mentioned previously, kittiwake populations breed within the buildings of Barentsburg and Pyramiden (Miljeteig and Gabrielsen, 2009), whereas this is not observed in Longyearbyen. Higher concentrations of *p,p*'-DDE in Pyramiden may be attributed to the larger kittiwake population within this settlement and supports findings observed for PCBs. This is also supported by findings within the Canadian Arctic where snow buntings feeding outside seabird breeding colonies had lower concentration of *p,p*'-DDE compared to birds feeding inside breeding colonies (Choy et al., 2010; Johnstone et al., 1996).

3.1.3. PFAS

Analysis of snow bunting eggs detected only PFOS and several PFCAs (i.e., PFNA, PFDA, PFUnDA, PFDoDA and PFTrDA) above detection limits (PFOS: 0.5 ng/g ww, PFCAs: 0.02 – 0.06 ng/g ww), with significant differences observed among all settlements (all chi-squared >

7.1, $p < 0.03$), while the PFTrDA and the $\sum_6$PFAS did not differ among the settlements (all chi-squared < 5.8, $p > 0.05$). PFOS and PFCA concentrations were higher in the two largest settlements, Longyearbyen and Barentsburg, with lower concentrations occurring in Pyramiden, albeit with exception of PFTrDA ($p = 0.13$, Table 1). On an average basis, PFOS was the dominating PFAS in the samples from Longyearbyen followed by PFNA, PFUnDA, PFDA, PFTrDA and PFDoDA. Similar to Longyearbyen, PFOS dominated the PFAS profile in Barentsburg. However, PFAS profiles in Barentsburg showed the average concentration to decrease in the order of PFUnDA > PFTrDA > PFDoDA = PFNA > PFDA. Higher variation was observed for PFNA, PFUnDA and PFDA in eggs from Longyearbyen compared to Barentsburg and Pyramiden (Table 1). This can be attributed to several individual eggs from Longyearbyen containing high concentrations of these particular PFAS (Table A4). PFAS profiles between Longyearbyen and Barentsburg were identical on a median concentration basis as the influence of extreme values were minimized (i.e., PFOS > PFUnDA > PFTrDA > PFNA > PFDoDA > PFDA). Lower $\Sigma PFAS_6$ concentrations were observed in Pyramiden, although one individual egg (egg 7, Table A4) contained high PFOS levels (11 ng/g ww, respectively) compared to the other eggs collected within this settlement. The remaining PFAS within this egg were below detection limits, which is strange considering the level of PFOS detected and indicates this sample may have been contaminated during the sample analysis. If we disregard this sample, PFUnDA was found to have the highest mean concentration followed closely by PFTrDA > PFOS > PFNA = PFDoDA > PFDA. This profile is likely attributed to the fact Pyramiden is abandoned and PFAS exposure via local sources is limited. Higher concentrations observed for PFUnDA (C11) and PFTrDA (C13) can be attributed atmospheric transport and subsequent degradation of FTOHs to PFCAs (Ellis et al., 2004), which is the dominant source of longer

chain PFCAs (C > 10) in remote regions (Armitage et al., 2009). Comparison of PFAS patterns between these settlements indicates that PFAS exposure in Longyearbyen and Barentsburg is influenced by local sources, whereas in Pyramiden exposure appears to be driven by long range transport.

The findings in the present study are in agreement with findings of PFAS in Greenland where PFNA (0.1 - 3.6 ng/g ww) was detected in all terrestrial wildlife investigated (i.e., ptarmigan (*Lagopus muta*), reindeer (*Rangifer tarandus*), musk ox (*Ovibos moschatus*) along with PFDA and PFUnDA in reindeer (0.28 and 0.45 ng/g ww, respectively) and musk ox (3.2 and 2.4 ng/g ww, respectively) (Bossi et al., 2015). Sources of PFCAs to terrestrial environments in remote environments can be attributed to atmospheric transport of flourotelomer alcohols and subsequent degradation to PFCAs (Ellis et al., 2004). Partitioning of PFCAs to snow within Arctic regions has been previously demonstrated, where PFNA, PFDA and PFUnDA have been detected in snow from the Canadian and European Arctic (Kwok et al., 2013; Young et al., 2007). Temporal analysis of ice cores from the Devon Ice Cap (Nunavut, Canada) have shown that fluxes of PFCAs to snow has remained constant from 1985 to 2015. The PFCA (C9-C13) profile was dominated by PFNA, followed by PFUnDA and PFDA, while PFDoDA and PFTrDA were rarely detected (Pickard, 2018). Kwok et al. (2013) also observed the dominance of PFNA ice cores taken from the Longyearbreen glacier (Svalbard, Norway). However, unlike findings from the Canadian Arctic, PFDoDA and perflourotetradecanoic acid (PFTeDA) were detected in surface snow samples, with higher proportions occurring near the community of Lonyearbyen. Ski wax from recreational skiing is a suggested source to PFDoDA and PFTeDA to surface snow as these compounds have been found to dominate in air during application of ski

wax as well as in serum from professional skiers (Freberg et al., 2010). However, usage/emission from local communities will also contribute to this observation. This is supported by recent findings of elevated levels of PFAS in soils and run off water from fire-fighter training facilities in Longyearbyen (Skaar et al., 2018), although snow bunting nest sites where not located near these facilities. High amounts of PFTrDA were also found compared to other PFCAs in snow bunting eggs in Longyearbyen. This can be attributed to its higher bioaccumulation potential and exposure from local sources, despite its low or non-existent detection in snow/ice core samples (Kwok et al., 2013; Pickard, 2018). All PFAS (except PFTrDA) were statistically higher in communities with sustained populations (Longyearbyen, Barentsburg) compared to the ones that do not (Pyramiden).

3.2. Spatial resolution in snow bunting egg exposure

Comparison of median and mean concentrations for individual PCB congeners and $\sum PCB_7$ showed considerable variation in PCB concentrations in eggs collected from the Russian settlements, particularly in Pyramiden (Table 1. Figure 2). This is attributed to that two of the 7 eggs collected contained PCB levels below detection limits (0.03 – 1.16 ng/g ww, congener dependent), which may be attributed to small home ranges exhibited by snow buntings during the breeding season. Studies within the literature have suggested snow buntings can forage up to a kilometer from their nest sites while breeding (Hayhow et al., 2018). Feeding rate in breeding snow buntings decreases with the distance birds must travel to acquire food (Falconer et al., 2008). It can therefore be assumed that these birds tend to gather resources for egg production in close proximity to their nesting site. Comparing nest locations to soil concentrations reported in Pyramiden ((Jartun et al. (2009a, b), Table A5), concentrations of $\sum PCB_7$ was highest in eggs

from nests located in close proximity to regions of high soil contamination (Figure 1). Eggs which contained PCB concentrations below detection limits (Pyramiden egg 6 and 7, Table A2) were from nests in areas where PCB soil concentrations were low or below detection limits. Surprisingly, nest sites for eggs containing high levels of PCBs compared to eggs having concentrations below detection limits were within less than 800 meters from each other. The nest site for egg 7, which was below detection limits, was within 150 meters from several sites with soil concentrations of $\sum PCB_7$ ranging between 5 -15 mg/kg dried weight (dw). It should be noted that the laying order of eggs can affect the overall exposure concentration dependent upon bird species. Although no information exists regarding egg exposure concentration and laying order in snow buntings, PCBs and DDT concentrations in eggs have been observed to decrease significantly with egg laying order in other passerine species (i.e., the great tit (*Parus major*) (Van den Steen et al., 2009). However, studies in several species have shown that variation in concentrations within clutches is smaller than among clutches (Custer et al., 2000; Van den Steen et al., 2006; Verreault et al., 2006). For example, variation in PCB and DDT concentrations between nest sites for great tits accounted for 93% and 78% of the total variation (Van den Steen et al., 2006). Due to the larger variation among clutches compared to the variation within clutches, one random egg from a clutch can therefore be regarded as a useful biomonitoring tool for organic pollutants (Van den Steen et al., 2009). Thus, our results indicate that the small home range and foraging behaviour for snow buntings during the breeding season can greatly affect their exposure, providing high spatial resolution within a given area for source apportionment.

Lower variation was observed in egg concentrations of individual PCB congeners and $\sum PCB_7$ concentrations in Barentsburg compared to Pyramiden (Table 1, Table A2). In Barentsburg,

concentrations of PCBs in individual snow bunting eggs ranged from 1.2 – 5.2 mg/kg lw, with the exception of one egg (Barentsburg egg 3, Table A2, Figure 3), which displayed significantly higher individual and $\sum PCB_7$ concentration. Unfortunately, no data is available on PCB soil concentrations around the nest location from which this particular egg was collected to assess nearby exposure. No clear spatial pattern could be observed between individual egg concentrations and soil concentrations reported by Jartun et al. (2009a, b) in Barentsburg, despite several locations showing high contamination (Figure 3). However, over 79 % of soil samples collected had $\sum PCB_7$ concentrations below 0.5 mg/kg dw (Table A6) with fairly uniform distribution across Barentsburg, which is supported by our findings in snow bunting eggs.

Concentration of PCBs in soil from Longyearbyen were significantly lower compared to Barentsburg and Pyramiden (Jartun et al. 2009a, b). Only 6 % of soil samples were above detection limits in Longyearbyen with most PCB concentrations below 0.05 mg/kg dw (Table A7) and the comparison between egg concentrations and the soil concentration adjacent to the nests showed no clear trends (Figure A1). This indicates terrestrial exposure to PCBs is low and uniformly distributed with no indication of local sources in Longyearbyen.

In contrast to the findings for PCBs in Longyearbyen, evidence of spatial gradients for certain PFAS substances was observed in snow bunting eggs. No clear spatial pattern for PFOS concentration in eggs was observed between the different nest sites (Figure 4). PFOS dominated the PFAS profile in a majority of the eggs collected except for eggs 1 and 3 where the longer chained PFCAs (i.e., PFNA, PFUnDA) were 2 to 8 times higher compared to the PFOS concentration (Figure 4, Table A4). Comparison of PFNA, PFUnDA and PFTrDA concentration

in eggs among all sites showed eggs 1 and 3 were 2 to 55 times higher. Although low sample numbers (i.e., 1 egg per nest site) hinders statistical assessment of differences, our findings are in agreement to earlier reports of elevated levels of various PFAS (including PFNA and PFUnDA) in surface snow sampled near these locations (Kwok et al. 2013, Figure A2). Various activities occur at these locations, mainly related to tourist activities where snow mobiles and equipment are stored. Various workshops are also located nearby along with a garbage sorting centre. Landfills have been identified as sources of PFAS exposure on Svalbard (Skaar et al., 2018), thus garbage sorting facilities of PFAS containing materials may act as sources of exposure in this region. Another potential exposure pathway could be attributed to snow buntings feeding from the tidal zone in this region. Choy et al. (2010) showed feeding on chironomids from local ponds to be an important exposure source for snow buntings in Cape Vera. Transport of PFAS bound within snow/glaciers via melt water to river and marine environments has been shown to occur in Longyearbyen (Kwok et al., 2013). PFOS was found in much higher amounts compared to the longer chain PFCAs (i.e., PFNA, PFDA, PFUnDA, PFDoDA) in river water and seawater downstream from melted glaciers in Longyearbyen. We would expect to see higher PFOS contribution in eggs 1 and 3 compared to eggs from other nest sites further inland if snow buntings from this region were feeding from the marine environment. However, PFOS concentration was fairly uniform in eggs across all nest sites, supporting that exposure to PFAS is being driven through terrestrial feeding. Similar to findings for PCBs within Pyramiden, eggs 1 and 3 in Longyearbyen were within 700 m from other nest locations in which eggs contained much lower PFAS concentrations (eggs 2 and 6). Although this could be attributed to small home ranges for feeding by snow buntings, no data is available on soil concentrations to support this. In addition, it is unclear how PFAS concentration will vary within snow bunting clutches

and this needs further investigation. However, high concentrations found in egg 1 and 3 support earlier findings of elevated PFAS concentrations in snow near these nest locations (Kwok et al. 2013, Figure A2) indicating a point source to PFAS exposure in this area.

In Barentsburg, PFOS was the dominating PFAS in eggs sampled at all locations with highest concentrations occurring in eggs 1 and 4, which were located within the settlement (Figure 5). No clear pattern could be observed for PFOS as eggs from nest locations within or near the settlement limits (eggs 1, 2, 4, 5, 7, and 9) showed a wide range in concentration (1.2 – 12 ng/g ww, Table A4). However, nest sites located farthest from the settlement (eggs 3, 6, 8) contained eggs with low PFOS concentrations (0.6 – 1.5 ng/g ww, Table A4), suggesting sources to PFOS exposure in Barentsburg. For PFCAs, no clear pattern could be observed in regards to nest location and concentration. However, concentration of ΣPFCAs in eggs 4 and 9 (15.1 and 13.8 ng/g ww, respectively (Table A4)) were 2 to 3 times higher compared to all other eggs collected. Egg 4 is located within the settlement and would be expected to show higher concentrations of PFAS due to is close proximity potential exposure sources. The nest location for egg 9 lies further from the settlement center, but is located near the power plant and other anthropogenic activity. In is unclear what could be contributing to higher levels but further investigation is needed to clarify if sources of PFAS exposure exist in this area.

Concentrations of PFAS were significantly lower in Pyramiden compared to Barentsburg and Longyearbyen, with exception to PFTrDA ($\rho < 0.03$) (Table 1, Table S3). No clear spatial trends could be observed for PFAS within this settlement. One sample showed high levels of PFOS (egg 7: 11.9 ng/g ww, Table S3, Figure A3) which was comparable to concentrations found in

Barentsburg and Longyearbyen (Table S3). However, PFCAs within the same sample were below detection limits, which may indicate this sample is an artifact and may have been compromised/contaminated during analysis.

4. Conclusion

The results reported in this study to the best of our knowledge are the first to investigate exposure to POPs and PFAS in a bird species feeding solely from the terrestrial environment on Svalbard. Despite lower contamination of both POPs and PFAS in terrestrial biota, local communities may act as local sources of exposure with significant bioaccumulation and toxicological risks to terrestrial feeders for certain contaminants (i.e., PCBs) at particular locations (i.e., Pyramiden, Barentsburg). Although limited sample number restricted the ability to conduct a more thorough statistical analysis, our results indicate narrow home range behavior by snow buntings may provide an advantage for exposure source apportionment. Further investigation is needed to evaluate concentration variability within snow bunting clutches for source apportionment on fine spatial scales. However, snow bunting eggs show great promise as a bio-indicator for contaminant source apportionment in Arctic terrestrial environments.

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construction and Ann Kristin Balto of the Norwegian Polar Institute for Pyramiden photograph usage in the graphical abstract.

Conflict of interests

The authors declare no competing financial interests

TABLE AND FIGURES CAPTIONS

Table 1**.** Concentrations of PCBs, HCB, organochlorine pesticides (ng/g lw) and PFASs (ng/g ww) in eggs of snow buntings (*Plectrophenax nivalis*) from Longyearbyen, Barentsburg and Pyramiden. Footnote*:*Indicates statistical differences in concentrations between settlements.*

Figure 1: Relative percentage of PCB congeners of ΣPCB_7 present in (A) snow bunting eggs collected from Longyearbyen, Barrentsburg and Pyramiden and (B) technical mixtures of Aroclor 1260, Aroclor 1262 and Sovol. Footnote: *Data for determining relative PCB congener percentage in technical mixtures taken from Takasuga et al., 2006.*

Figure 2. Spatial distribution of ΣPCB_7 concentrations in soil (mg/kg dry weight) and snow bunting egg (mg/kg lipid weight) concentrations in Pyramiden. Foot note: *Soil concentrations are extracted from Jartun et al. 2009a, b.*represents ΣPCB_7 concentrations below detection limits in snow bunting eggs.*

Figure 3. Spatial distribution of ΣPCB_7 concentrations in soil (mg/kg dry weight) and snow bunting eggs (mg/kg lipid weight) in Barentsburg. Foot note: *Soil concentrations are extracted from Jartun et al. 2009a, b.*

Figure 4. Spatial distribution of PFOS, PFNA, PFUnDA, and PFTrDA in snow bunting eggs (mg/kg wet weight) in Longyearbyen.

Figure 5. Spatial distribution of PFOS, PFNA, PFUnDA, and PFTrDA concentrations (mg/kg, wet weight (wet weight) in snow bunting eggs in Barentsburg

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Siv Kristoffersen: Investigation

Dr. Dorte Herzke: Writing – Reviewing & Editing, Supervision, Resources, Formal Analysis

Dr. Geir Wing Gabrielsen: Supervision, Writing – Review & Editing

Dr. Bjørn Munro Jenssen: Conceptualization, Writing – Review & Editing, Supervision, Project Administration, Funding Acquisition

Table 1

		Longyearbyen		**Barentsburg**		**Pyramiden**
	n	Median, mean ± SE	n	Median, mean ± SE	n	Median, mean ± SE
Volume egg (cm3)	8	3.06, 3.11 ± 0.18	9	2.79, 2.79 ± 0.08	7	2.99, 3.02 ± 0.08
Lipids (%)*	8	6.40, 7.08 ± 0.70	9	7.52, 7.53 ± 0.81	7	4.59, 4.79 ± 0.27
Clutch size (#)	6	5.50, 5.67 ± 0.49	5	5.00, 5.40 ± 0.24	7	6.00, 5.57 ± 0.20
HCB*	8	44.6, 66.2 ± 19.5	9	49.3, 52.0 ± 7.15	7	37.7, 53.9 ± 15.4
p,p'-DDE*	8	47.8, 62.3 ± 14.5	9	74.7, 92.2 ± 15.5	7	173, 268 ± 98.4
trans-nonachlor*	8	0.94, 1.02 ± 0.19	8	1.02, 1.17 ± 0.28	7	0.68, 0.87 ± 0.15
PCB-28/31*	7	0.12, 0.50 ± 0.22	9	4.61, 8.55 ± 2.73	7	12.0, 28.8 ± 18.5
PCB-52*	7	1.02, 0.94 ± 0.14	9	110, 197 ± 95.6	7	49.9, 404 ± 235
PCB-101*	8	3.89, 5.42 ± 1.53	9	387, 718 ± 317	7	301.5, 2 020 ± 1 080
PCB-118*	8	13.0, 20.4 ± 6.64	9	828, 1 307.1 ± 377	7	988.7, 3 490 ± 1 730
PCB-138*	8	18.2, 23.9 ± 5.74	9	818, 1 316.6 ± 416	7	1 010, 3 390 ± 1 670

PCB-153*	8	34.0, 38.6 ± 6.53	9	716, 1 130 ± 328	7	1 310, 3 020 ± 1 420
PCB-180*	8	14.5, 16.1 ± 4.69	9	144, 250 ± 106	7	230, 394 ± 175
∑7PCB*	8	95.7, 106 ± 21.2	9	2 980, 4 930 ± 1 620	7	3 860, 12 800 ± 6 310
PFOS*	8	4.14, 3.75 ± 0.68	9	2.20, 3.53 ± 1.22	7	0.55, 2.13 ± 1.63
PFNA*	8	0.98, 4.78 ± 2.28	9	0.75, 0.92 ± 0.25	7	0.17, 0.23 ± 0.07
PFDA*	8	0.65, 1.76 ± 0.99	9	0.55, 0.65 ± 0.16	7	0.03, 0.14 ± 0.08
PFUnDA*	8	1.55, 3.93 ± 1.77	9	1.64, 2.08 ± 0.54	7	0.39, 0.56 ± 0.19
PFDoDA*	8	0.66, 0.93 ± 0.31	9	0.65, 0.94 ± 0.23	7	0.24, 0.21 ± 0.07
PFTrDA	8	0.99, 1.28 ± 0.36	9	1.30, 1.76 ± 0.52	7	0.71, 0.54 ± 0.14
∑6PFAS	8	9.45, 16.4 ± 5.61	9	6.69, 9.89 ± 2.44	7	2.48, 3.82 ± 1.46

Highlights

- POPs and PFAS accumulate in Arctic terrestrial birds
- Snow bunting eggs are useful for determining exposure sources for contaminants
- Settlements on Svalbard are sources for POPs and PFAS exposure
- Snow buntings provide high spatial resolution for contaminant source apportionment

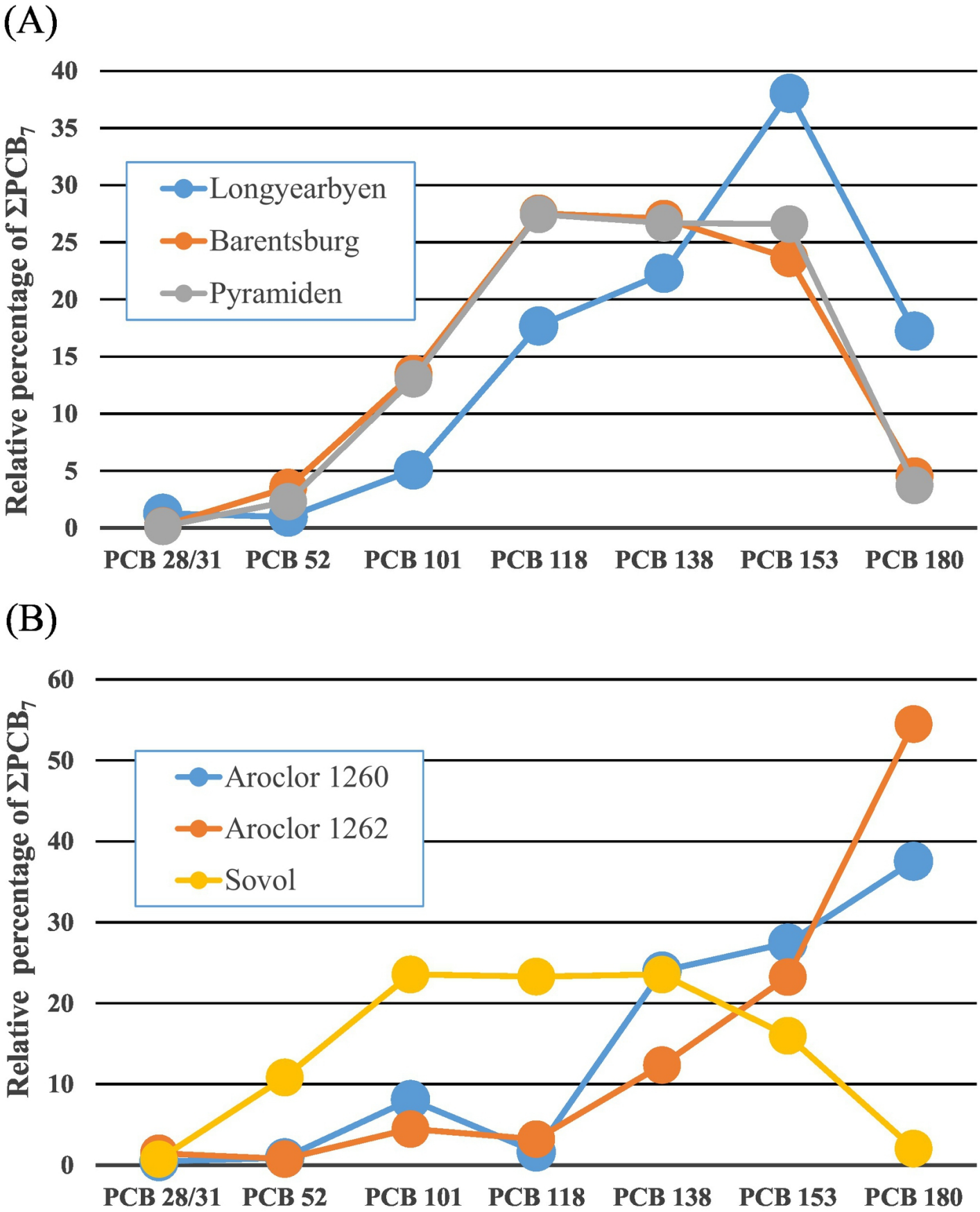
(A)
Relative percentage of ΣPCB7
40
35
30
25
20
15
10
5
0
Longyearbyen
Barentsburg
Pyramiden
PCB 28/31
PCB 52
PCB 101
PCB 118
PCB 138
PCB 153
PCB 180
(B)
Relative percentage of ΣPCB7
60
50
40
30
20
10
0
Aroclor 1260
Aroclor 1262
Sovol
PCB 28/31
PCB 52
PCB 101
PCB 118
PCB 138
PCB 153
PCB 180

Figure 1

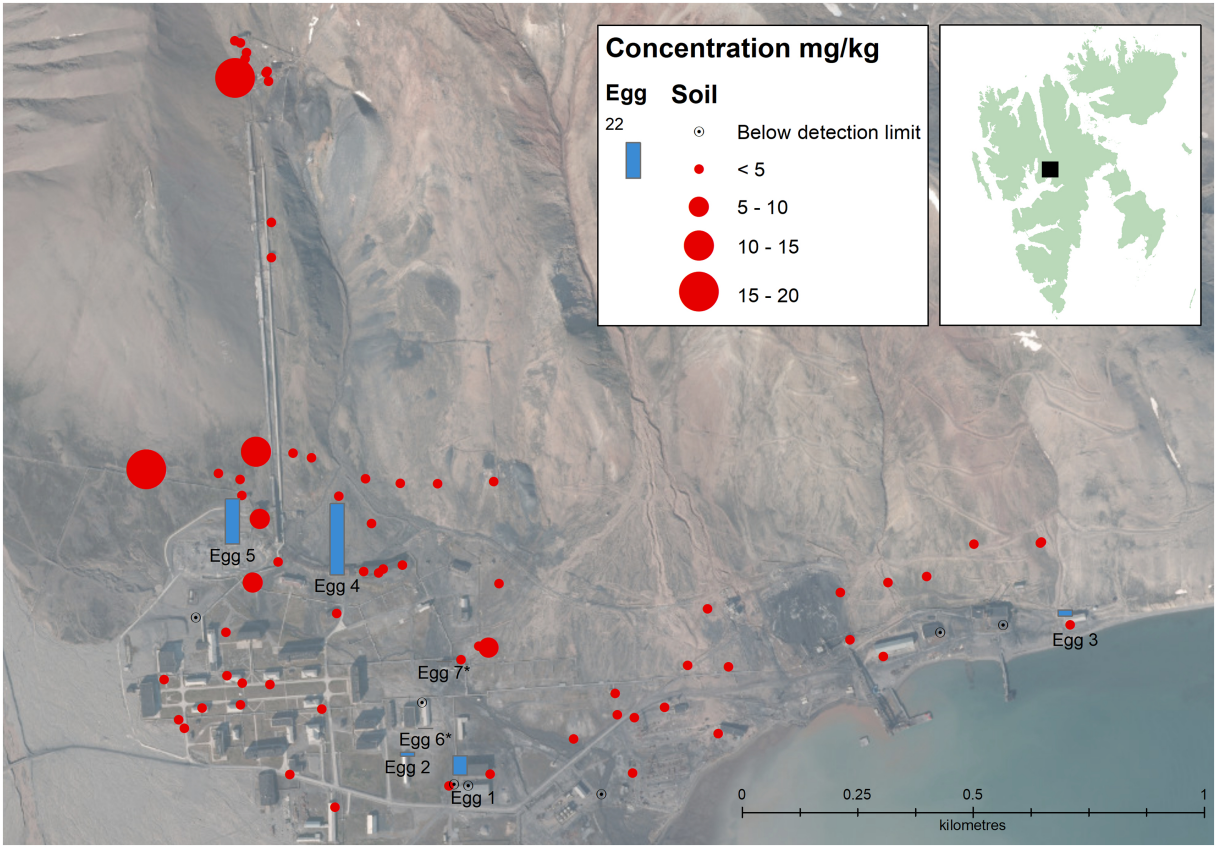
Concentration mg/kg
Egg
Soil
22
Below detection limit
< 5
5 - 10
10 - 15
15 - 20
Egg 5
Egg 4
Egg 7*
Egg 6*
Egg 2
Egg 1
Egg 3
0
0.25
0.5
1
kilometres

Figure 2

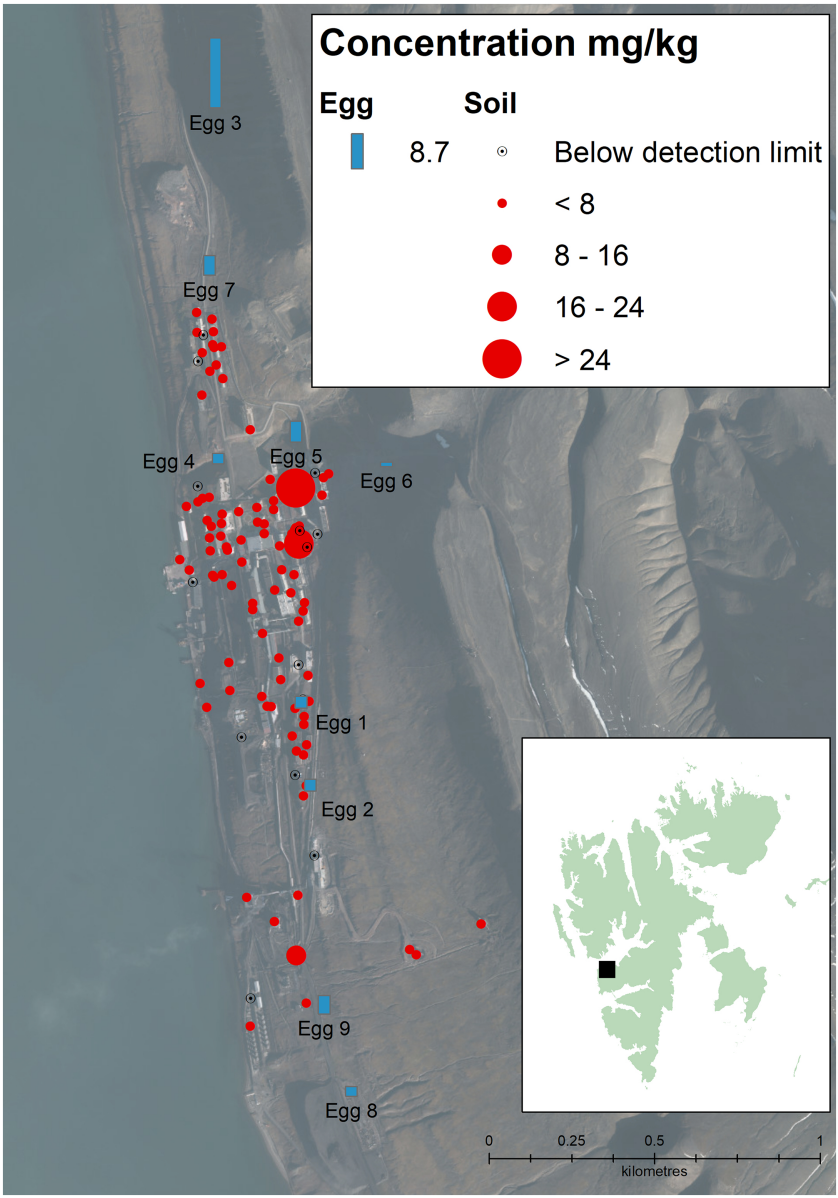
Concentration mg/kg
Egg
Soil
8.7
Below detection limit
< 8
8 - 16
16 - 24
> 24
Egg 3
Egg 7
Egg 4
Egg 5
Egg 6
Egg 1
Egg 2
Egg 9
Egg 8
0
0.25
0.5
1
kilometres

Figure 3

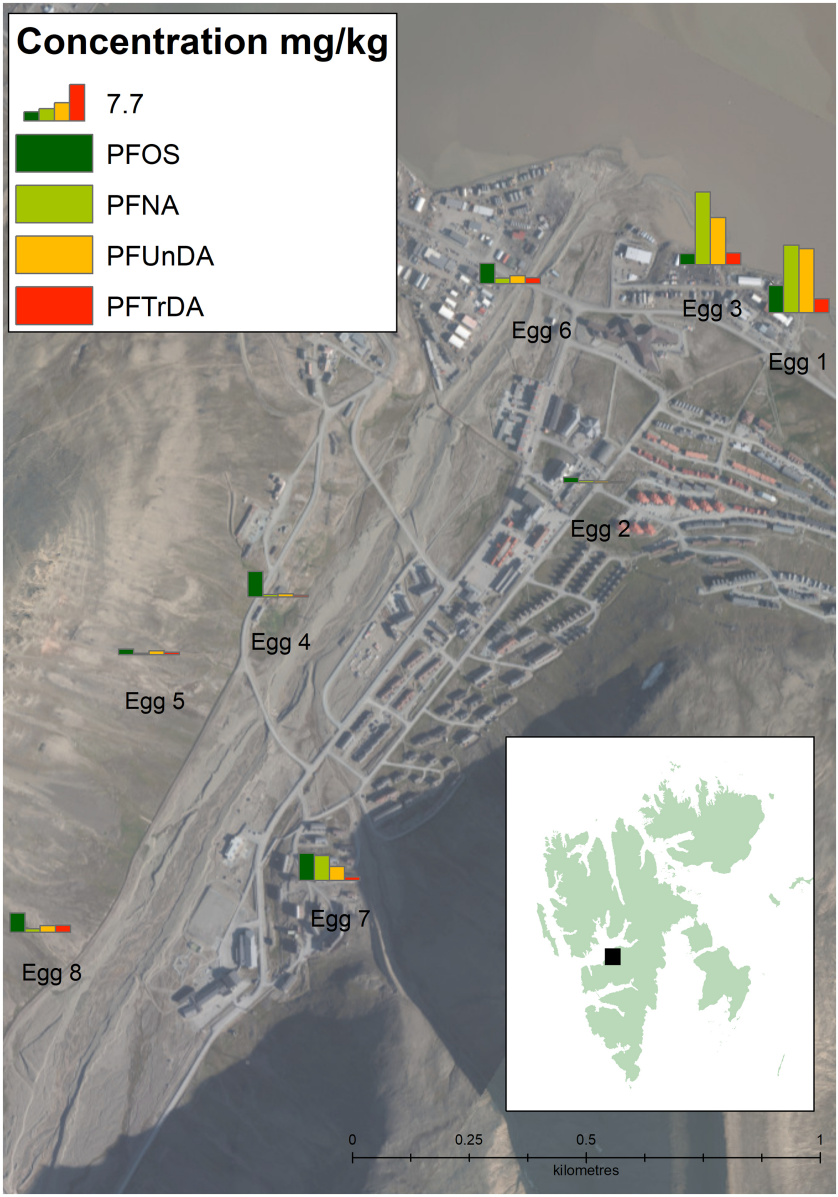
Concentration mg/kg
7.7
PFOS
PFNA
PFUnDA
PFTrDA
Egg 6
Egg 3
Egg 1
Egg 2
Egg 4
Egg 5
Egg 7
Egg 8
0
0.25
0.5
1
kilometres

Figure 4

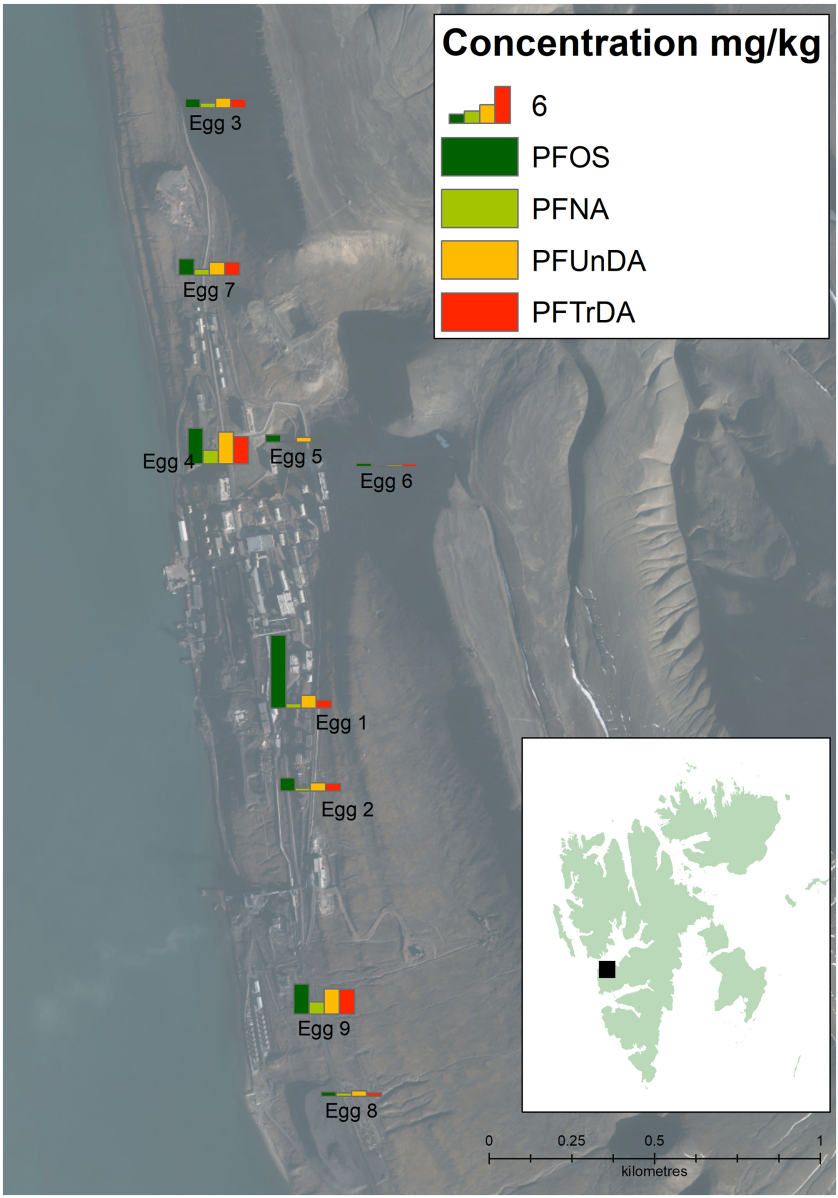
Concentration mg/kg
6
PFOS
PFNA
PFUnDA
PFTrDA
Egg 3
Egg 7
Egg 4
Egg 5
Egg 6
Egg 1
Egg 2
Egg 9
Egg 8
0
0.25
0.5
1
kilometres

Figure 5