

Article

Combined Contaminant Levels from Local Harvested Food Items in the Norwegian–Finnish–Russian Border Region

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Abstract: This paper presents the results of a multidisciplinary study with the aim of assessing the potential combined risk from consuming locally harvested food products in the Euro-Arctic region of Norway, Finland, and Russia. The three important contaminant groups—radioactive substances, heavy metals, and persistent organic pollutants (POPs)—were measured in food samples such as berries, mushrooms, fish, birds, reindeer, and moose; they were sampled in 2013–2015. To assess the combined pollution levels and investigate the trends, similarities, and variations between different contaminant groups, subsequent multivariate statistical analysis was performed. The results showed that, in general, the levels of radioactive substances, toxic elements, and POPs were below the permitted EU maximum content in food products. However, statistical analysis revealed some correlations, similarities, and peculiarities between the accumulation of different contaminants in various species, which allowed for a better understanding of the mechanisms of accumulation and interaction between different contaminant groups. It also gave a better insight into the possible added risks and helped pinpoint species that could serve as reference markers for the accumulation of different contaminants in food. Mushrooms, fish, and reindeer were found to be important markers in the combined risk assessments for the contents of metals and radioactive substances. Further research, as well as the development of methodologies for combined assessments, are recommended.

Keywords: Arctic; natural resources; food safety; environmental pollution; radioactive substances; toxic elements; POPs; multivariate analysis; Nikel smelter; Kola Peninsula



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1. Introduction

The issue of environmental contamination and food safety is of special concern to the Arctic region due to numerous existing and potential sources of pollution in the immediate and adjacent areas. Populations living in the Arctic are mainly exposed to long-range pollution from the southern part of the globe, especially those residents whose diets comprise a large proportion of traditional terrestrial, marine, and freshwater food items [1]. Contaminants such as radioactive substances, heavy metals, and persistent organic pollutants (POPs) have long ecological half-lives [1,2]. Arctic residents are exposed to the highest radiation levels due to artificial radionuclides in the Arctic [3]. Radiocaesium (Cs-137) is one of the most important longest-living artificial radionuclides, which has contributed to the radioactive contamination of natural food products, especially reindeer meat and semi-natural flora, via global fallout from the atmospheric nuclear weapon testing that occurred in the 1950s and 1960s and from fallout following the Chernobyl accident in 1986 [3].

An important example of a time-trend study is of the food chain lichen–reindeer/caribou–man, which started in the Nordic countries after launching nuclear weapon testing in the 1950s [2]. In addition, some amounts of radiocaesium were detected in the Arctic after the accident at the Fukushima Daiichi NPP in 2011 [4].

For the people living in the border regions, it is important to be aware of the status of the pollution level and the safety of food items that are regularly harvested or hunted in the area. It is also important to obtain an overview of the possible risks of ingesting locally harvested food.

In some areas across the Euro-Arctic region, local contamination sources have raised concern among residents who harvest food from wild places (e.g., berries, mushrooms, fish, ptarmigans, moose) or who consume semi-domesticated reindeer as an essential constituent of their diet. Such is the case with the border region between Norway, Finland, and Russia, where large smelters in Nikel and Zapoljarny, as well as the mining and pellet industry in Kirkenes [5,6], are reported to be sources of heavy metals [6] and possibly some POPs [5,6]. Both local industrial pollution and the long-range transport of contaminants are sources of overall pollution, but metals and dioxins are more closely related to local and regional sources. In addition to airborne pollution and wastewater discharge from the Russian smelters and the city of Nikel, toxic elements are released into Kuetsjarvi Lake, located in the Pasvik watercourse (forming the border between Norway and Russia), close to the smelter [5]. Such pollution, especially by heavy metals emitted from the Pechenga Nickel complex in the Kola Peninsula, has caused concern about its potential effects on local food in the border regions between Norway, Finland, and Russia during the last few decades.

In 2012–2015, a joint cross-border research project (EU Kolarctic KO467) was implemented with the purpose of assessing the levels of radioactive substances, heavy metals, POPs, and other toxic substances in natural food products from that area of interest (Figure 1). As a result, a combined dataset was compiled, which included the contents of different contaminant types in the same food items.

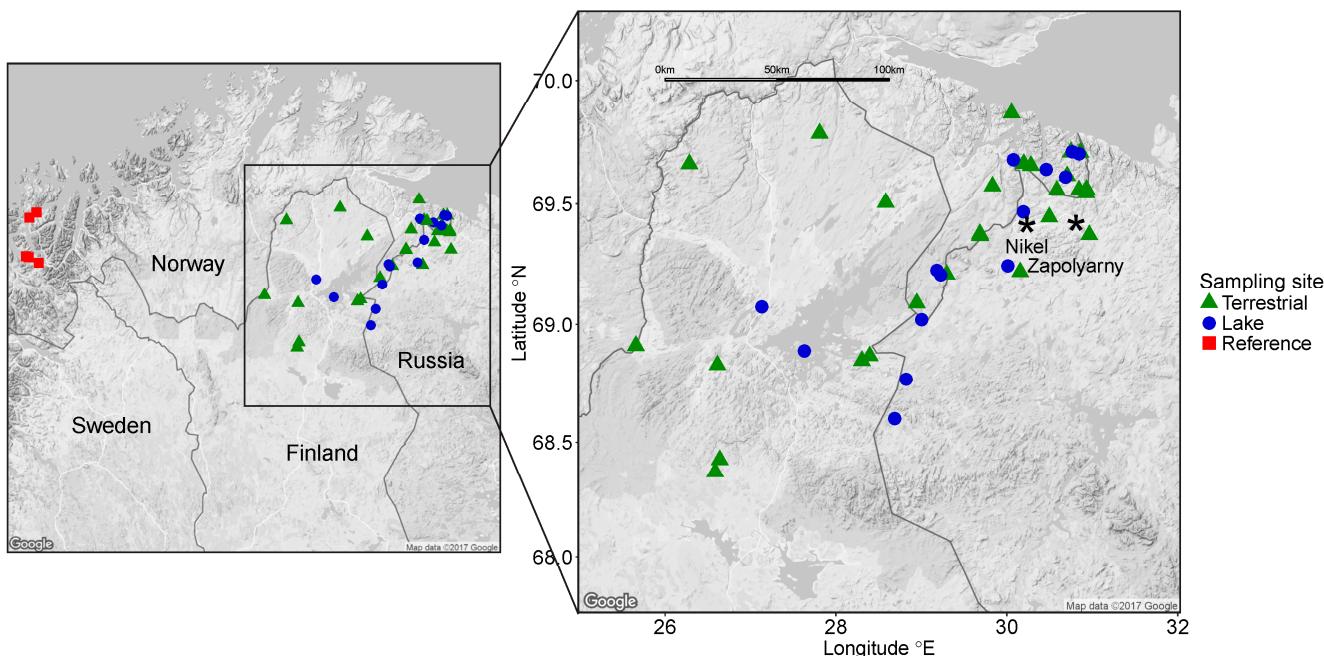


Figure 1. Map of sampling locations. The map to the right is an enlarged view of sampling locations across all three countries, seen on the left: terrestrial sites (green triangles and red squares), lakes (blue dots), reference sites (red squares), and the cities of Nikel and Zapoljarny (black stars). Source: Hansen et al. 2017 [7].

Although substantial research has been carried out with the identification of different types of contaminants in food items, little is known about the interactions between the

different groups of contaminants, possible similarities in accumulation and distribution, their added risks, and possible harmful effects, especially when assessing the total risk of the consumption of these food products.

This study builds on the results of the Kolarctic project to promote the multidisciplinary approach used for assessing toxic compound contents in foraged local food products. This research was designed to assess the potential combined risk from pollutants contained in local food products used by populations living in the northern border regions of Norway, Finland, and Russia by analyzing the combined datasets for three important contaminant groups—radioactive substances, heavy metals, and persistent organic pollutants (POPs)—in the same food items, and comparing their accumulation trends, similarities, and variations through multivariate statistical analysis. We also calculated the relationships between the levels of different types of pollutants in the same food items in order to estimate the potential risks for humans and to identify food items that could be markers of contamination.

2. Materials and Methods

2.1. Sample Collection in Northern Norway, Finland, and Northwest Russia

The studied food items were different types of berries, mushrooms, fish, birds (ptarmigan), reindeer, and moose, sampled in the period 2013–2015 across the border regions of the three countries. Species sampled in the frames of this project were berries (blueberry, cowberry, lingonberry, bog bilberry, bunchberry, and cloudberry), mushrooms (Birch Bolete, Orange Birch Bolete, Brown roll-rim, Velvet bolete, Rufous Milkcap, Red-banded webcap, Gypsy mushroom, Bearded milkcap, Copper Brittlegill, and Gassy webcap), and freshwater fish (perch, pike, and whitefish). Food item sampling sites (Figure 1) were selected across areas used by the local populace for berry picking, freshwater fishing, and hunting. Priority was given to sites northwest of the Ni-Cu smelter because of their position in the path of prevailing winds. In all, across the border regions between Norway, Finland, and Russia, 41 sampling locations were selected (Figure 1), namely 21 sites in Norway (14 terrestrial sites and 7 lakes)—Sør-Varanger municipality, Jarfjord, and Pasvik; 11 in Finland (9 terrestrial sites and 2 lakes)—Inari and Kuusamo regions; and 9 in Russia (4 terrestrial sites, 4 lakes, and 1 water reservoir)—Lakes Virtuovoshjaur, Kuetsjarvi, Shuonijarvi, and Kochejaur, and Rajakovski reservoir. Also, there was a reference site close to Tromsø, Norway [7].

Berry, mushroom, fish, ptarmigan, reindeer, and moose sampling were done using indigenous food harvesting techniques; internal protocols were developed, too. The samples to be analyzed for metals and radioactivity were placed in plastic bags, and those to be analyzed for POPs were placed in glass containers and then kept at -20°C until analysis. All samples were analyzed within 6 months to 1 year.

Table 1 shows the type and number of samples for each food item collected in the border region of the three countries (see more details on species in Appendix A).

Table 1. Overview of analyzed samples. HM = heavy metals, RN = radionuclides, POPs = persistent organic pollutants. Numbers provide the number of samples for that specific contaminant class.

Samples	Norway: 64 Samples	Finland: 35 Samples	Russia: 32 Samples
Berries	24: HM	12: HM, RN, POPs	10: HM, RN
Mushroom	8: HM (incl. background)	3: HM, RN, POPs	8: HM
Fish	19: HM, POPs, RN	8: HM, RN, POPs	13: HM, RN
Ptarmigan	---	3: HM, RN, POPs	---
Moose	3: HM, RN	3: HM, RN, POPs	1: HM
Reindeer	10: HM, RN, POPs	6: HM, RN, POPs	--

All the fish, birds, reindeer, and moose specimens investigated by the project were for commercial use. Reindeer samples were taken from slaughterhouses (only a small piece

was used for the project, the rest for food), moose and birds were taken from the freezers of hunters; fish were caught either by fishermen having fishing permits or in free fishing areas.

2.2. Chemical Analyses and Measurements of Contaminants

2.2.1. Radioactive Substances

All environmental samples collected in Northern Norway, Finland, and Northwest Russia were analyzed for radioactive substances at the DSA's accredited laboratory in Svanhovd, Norway. The samples were dried at 105 °C, homogenized, and placed into respective measuring geometries (sample containers) for subsequent laboratory measurements via gamma spectrometry using HPGe detectors (CANBERRA) [4]. The overview of the measurements of radioactive substances is in Appendix B.

2.2.2. Heavy Metals

Ni, Cu, Co, As, Pb, Hg, and Cd were measured from the collected food items in Norway and Finland. The selected metals are the three main toxic metals monitored in the AMAP monitoring program (Hg, Cd, and Pb) [1], as well as other relevant metals emitted from the Nikel smelter [7]. Norwegian samples were analyzed at the laboratory of NILU, Norway, and the Finnish ones at Jozef Stefan Institute, Department of Environmental Sciences, Ljubljana, Slovenia (see details in [7]). Ni concentrations were measured at NILU, following the procedure used for Norwegian samples [7]. Russian samples (13 metals, Pb, As, Cd, Hg, Cu, Zn, Ni, Cr, Fe, Mn, Co, Sr, and V) were analyzed at Typhoon Laboratory in St Petersburg, Russia (see details in [7–9]). The list of heavy metals analyzed is in Appendix B.

2.2.3. POPs

The selection of organochlorine compounds was based on the knowledge of ubiquitous presence, particularly PCBs and the DDT group, in the environment and humans. Dioxins (PCDD/Fs) and dioxin-like PCBs (DL-PCBs) were analyzed using Norwegian samples due to previous food advice for fish and reindeer in the area near Kirkenes. The analyses of organochlorine compounds in food items (Table 1; i.e., ptarmigan, reindeer, moose, fish, berries, and mushrooms) collected in Finland were performed in the Department of Environmental Chemistry, Institute of Environmental Assessment and Water Research (IDAEA), Barcelona, Catalonia, Spain (see details in [10]). In Norway, the analyses of the organochlorine groups (PCBs, HCB, PeCB, dioxins, and furans (PCDD/Fs)) in fish and reindeer were performed by NILU with the analytical method described in Langdal et al. [11]. The list of the organochlorines analyzed is given in Appendix B.

2.3. Multivariate Statistics

Multivariate statistics were applied to the dataset, specifically principal component analysis (PCA). Some of the advantages of applying PCA are that it copes with collinearity between variables and provides plots of data compressed to fewer dimensions than the original dataset [12]. Loading plots show the influence of pollutants on the location of samples in the scores plot. Pollutants located furthest from the origin (0,0) have the highest influence.

A variety of species were collected and measured from each food item group to screen the levels of contaminants (see Section 2.1). However, not all species were analyzed for all the pollutants. In the multivariate statistical analyses, only those species were included where more than one pollutant group was analyzed.

SIMCAP11 Software was used for the principal component analysis modeling. PCAs of the pollutant concentrations for 131 samples of berries, mushrooms, fish, and game (reindeer, moose, and ptarmigan) were used to evaluate the differences and similarities within the data. Since the pollutant concentrations varied in magnitude and units, the data were logarithmically transformed and subsequently centered and scaled to variance. The number of significant components was determined by cross-validation. Score plots were

used to visualize the similarities/differences in properties, and loading plots were used to visualize which descriptors (properties) have a strong influence on the distribution in the score plot. These are found far from the origin in the loading plot, and positively correlated descriptors are projected close to each other, while negatively correlated descriptors are projected opposite to each other with respect to the origin.

2.4. Risk Assessment

The measured values of different groups of contaminants were compared with the accepted maximum values in the national guidelines and EU recommendations. It should be noted that currently, there are no appropriate methods for assessing a combined risk from different groups of contaminants in foods.

The limits for contaminants in foodstuffs are regulated nationally. Different countries have established different limits for certain foodstuffs.

The limits of radioactive substances, POPs, and metals acceptable in the EU, Norway, Finland, and Russia for natural food products are in Appendix C and the AMAP report 2021, Section 5 [1].

3. Results

3.1. Radioactivity

Measurements of radioactive substances in berries, mushrooms, freshwater fish, reindeer, moose, and ptarmigan sampled in Northern Norway, Finland, and Northwest Russia in 2013 and 2014 have shown that, in general, the Cs-137 levels are low and below the national activity limits for foods set up for commercial retails (Appendices A and C; ref. [4,13]). The results showed large variations in activity concentrations of radiocaesium between different environmental species and sampling areas. Analyses of different types of berries and mushrooms (see details in Section 2.1) proved the presence of Cs-137, with activity concentrations compatible with earlier data from the same region [13–16]. The highest activity of Cs-137 in berries was observed in cloudberry and for mushroom species, in the Gypsy mushroom (up to 1381 Bq/kg dw., which is 125 Bq/kg ww—a factor of 11 difference) [13]. Freshwater fish samples (see details in Section 2.1) from Northwest Russia were compared with those collected from Northern Finland and Norway [4]. Despite the different contents of Cs-137 in the same species from Norway, Finland, and Russia, all levels of this radionuclide were well below the national activity limits for freshwater fish, making, respectively, 3000 Bq/kg, 600 Bq/kg fw., and 130 Bq/kg fw. (Appendix B). All the reindeer samples from Norway contained both Cs-137 and Cs-134, indicating that the small traces of radioactive substances that reached Norway after the Fukushima accident were still observable [4]. Activity concentrations of Cs-137 in the Pasvik area were around 130 Bq/kg ww., and those of Cs-134 were under 1 Bq/kg ww. Radioactivity in all samples was below the activity limits set up for reindeer meat. The ptarmigan samples collected from the Sevettijarvi (2013) and Utsjoki (2013 and 2014) areas showed Cs-137 activity concentration variations of 27.0 to 32.4 Bq/kg dw. in 2013 and 38.9 Bq/kg dw. in 2014 [13].

3.2. Heavy Metals

The concentrations of the study toxic elements were the highest in mushrooms, with the exception of Hg, which was the highest in fish (Appendix B, see [7]). The EU maximum levels of Pb, Cd, and Hg for safe human consumption in commercial food were enhanced in some samples, but most samples showed concentrations below the maximal levels and thus were considered safe [7]. Compared with the border regions of Norway and Russia, those from Finland showed lower Ni and As concentrations in bilberries and lower Ni, As, and Cd concentrations in lingonberries [7]. None of the observed levels of toxic elements in the food items exceeded the accepted safety limits in Finland [10]. In all, 84% of the samples from all border regions overstepped the maximal concentrations detected at a reference site in Tromsø. In fish, Ni, Co, and Cd concentrations were 3.5 to 45 times higher at the

sampling sites closest to the Nikel smelter when compared with other lakes in Norway, Finland, and Russia.

3.3. POPs (or Organochlorines)

PCBs, HCB, PeCB, dioxins, and furans were analyzed in fish and reindeer from Norway. In the three pooled trout and the three pooled char samples, the sum of non-dioxin-like PCBs (ICES-6: PCB-28, -52, -101, -138, -153, and -180, or sumPCB₇: ICES-6 congeners + PCB-118) dominated, followed by the sum of dioxins/furans and dioxin-like PCBs (PCB-77, -81, -126, and -169). PCB-153 was the dominating PCB congener, varying from 0.73 to 4.6 ng/g ww. In the three pooled muscle samples of reindeer, HCB was the dominating compound, followed by PCB ICES-6.

For both fish and reindeer samples, the dioxin-like PCBs dominated the sum of dioxin-like compounds. Dioxin-like compounds in the three pooled reindeer muscle samples exceeded the maximum level of 4 pg TE/g fat—set by the EU for meat from bovine animals—and the maximum level of 7.5 pg TE/g fat for venison; however, none of those fish samples exceeded the maximum limit of 6.5 pg TE/g wet weight for dioxins and dioxin-like PCBs or 125 ng/g wet weight for non-dioxin-like PCBs (ICES-6).

Organochlorine compounds were analyzed in all collected food items (moose, reindeer, ptarmigan, fish, berries, and mushrooms) and had detectable levels in the study in Finnish Lapland [10]. Among them, levels of DDTs and PCBs were found to be the highest in fish (Appendix B). The sumPCB₇ was lower in fish from Finnish Lapland than in the fish samples collected in the border region of Norway and Russia.

None of the analyzed food samples from Russia exceeded the Russian maximum permissible concentrations of POPs in raw foods [9]; data are not included in the present study and PCA analysis. There were no spatial differences in the DDT levels between the lakes closest to the Pechenga nickel plant and the others, whereas PCB levels in perch, whitefish, and pike showed an obvious strong decreasing trend with an increase in distance of sampling locations from the location of the smelter plant, where PCBs' levels were reported to be the highest [9].

3.4. Comparisons of Different Contaminants and Results

3.4.1. Identifying Relevant Markers for Pollutants (Based on Trends in the Dataset)

Pollutant levels were (in large parts of the dataset) below the threshold values for food consumption. However, there are no established threshold values set up by authorities for the contents of multiple contaminants or a mixture of pollutants in foods, and the cumulative effects are unknown. The only exception is the mixture and class of dioxin and dioxin-like PCBs for which the maximum level has been set by the European Commission. Hence, it is necessary to use specialized methods for investigating potential combined patterns and trends of different pollutants in the dataset as well as identifying food items to be employed as markers of potential risk for humans from exposure to pollutants.

3.4.2. Trends for Radioactivity, Metals, and POPs

The first step in the multivariate analysis was to uncover trends in the samples that were analyzed for radioactivity, metals, as well as POPs. Although this only covers part of the dataset, it provides a foundation for further investigations of the trends in the dataset and provides a good insight into potential variations for the different pollutants and samples.

Three PCA models were calculated as follows:

Model 1a: Samples analyzed for radioactivity, metals, HCB, PCB, and DDT (33 samples)

Model 1b: Samples analyzed for radioactivity, metals, HCB, and PCB (46 samples)

Model 1c: Samples analyzed for radioactivity, metals, PCDD/F, and PCB (13 samples)

The PCA models were good, explaining 57–86% of the modeled data, and were stable (Q^2 0.45–0.57). The PCA scores plot showed no clustering in model 1a and no clustering of samples in models 1b and 1c. The reason that there were no clear clustering patterns related

to the samples in model 1a is related to the high weight of DDT/DDE at the location of the samples in the scores plot. The concentrations of DDT/DDE were low and had a similar variance in the samples (apart from one fish sample), hence resulting in a scores plot with no clustering. The score and loading plots of model 1a are included in Appendix B.

The score and loading plots of models 1b and 1c are shown in Figure 2. In model 1b, there is a clear clustering of samples according to whether these are berry, fish, or game samples. The loading plot reveals that PCB congeners and Hg have the highest influence on the first component, t1 (furthest from the origin on the p1 axis).

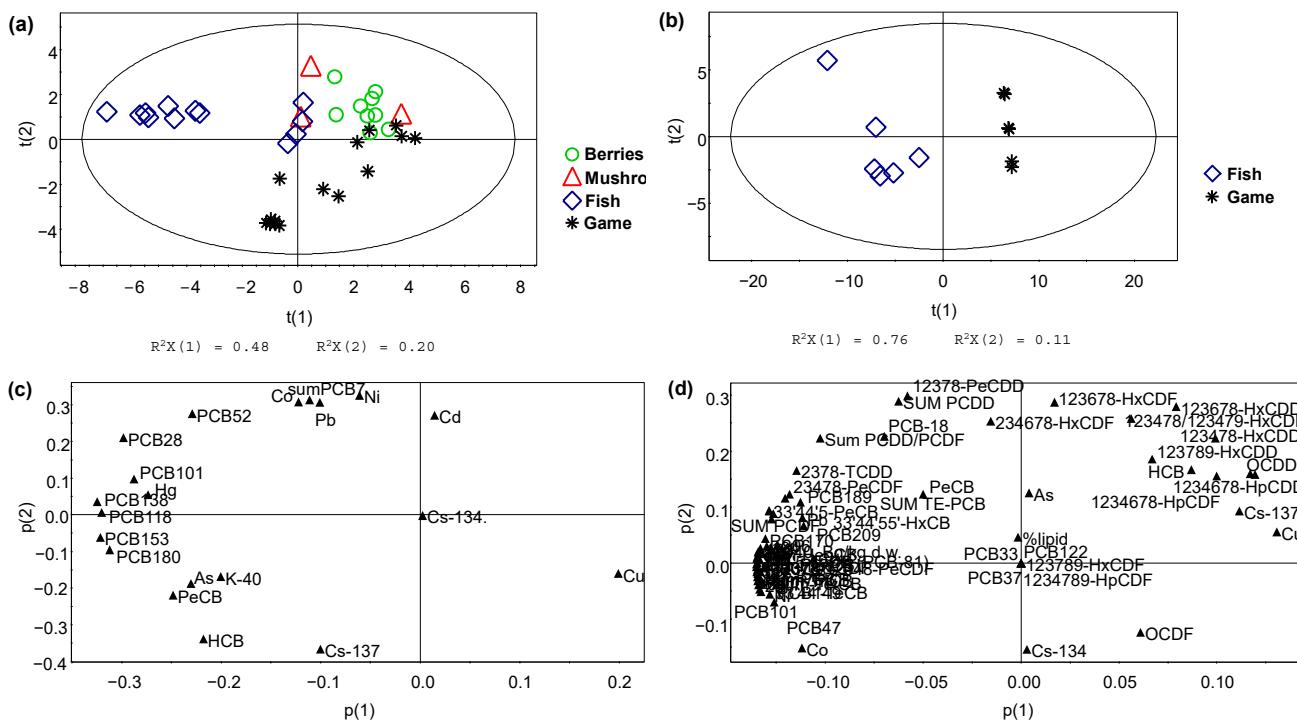


Figure 2. PCA and score plots of (a) model 1b and (b) model 1c. Samples located in the same area of the plot have similar compositions of pollutants. Loading plots of (c) model 1b and (d) model 1c.

The fish samples, hence, have the highest concentration of PCB congeners and Hg, as these are (mostly) located farthest from the origin in the same direction in the scores plot. The loading plot also shows that Cs-137 has the highest influence on the second component, t2, indicating that the game samples located in the third quadrant of the scores plot have the highest level of radioactivity. It is worth noting that the mushroom samples with the highest analyzed radioactivity were not analyzed for POPs (available data only from Finland) and are, hence, not included as data in model 1b. In the loading plot, the PCB congeners are clustered in the same area, meaning that there is a correlation between these in the samples, whereas the radioactive substances and metals are not clustered. Accordingly, a high concentration of one metal in each sample does not entail a generally high level of all metals in that sample. In model 1c, there is a clear clustering of fish and game. The loading plot reveals that the highest concentrations of PCB congeners (furthest from the origin of p1 in the loading plot) were found in the fish samples (furthest from the origin of t1 in the scores plot). The game samples had higher levels of Cs-137 and Cu. The loading plot showed clustering of PCB congeners and some clustering of HxCDD components, indicating that these are correlated. As was the case in model b, no clustering of metals or radioactive substances was observed.

The three first models' implementation showed that the pollutant concentrations depended on the type of sample (berries, mushrooms, fish, or game). Not all the samples were, however, analyzed for all pollutants, and to obtain an overview of the whole dataset, three new models were calculated, focusing on fewer pollutants in each model:

Model 2a: Samples analyzed for radioactivity and metals (128 samples)

Model 2b: Samples analyzed for HCB, PCB, and DDT (45 samples)

Model 2c: Samples analyzed for PCDD/F and PCB (13 samples)

The PCA models were good, explaining 59–89% of the modeled data, and were stable (Q^2 0.3–0.8). A clustering of the samples according to sample type (berries, mushrooms, fish, and game) was observed in the score plots of all the models. The clustering in model 2c was, however, like the trend of model 1c. The score and loading plots (Appendix B) of models 2a and 2b are shown in Figure 3.

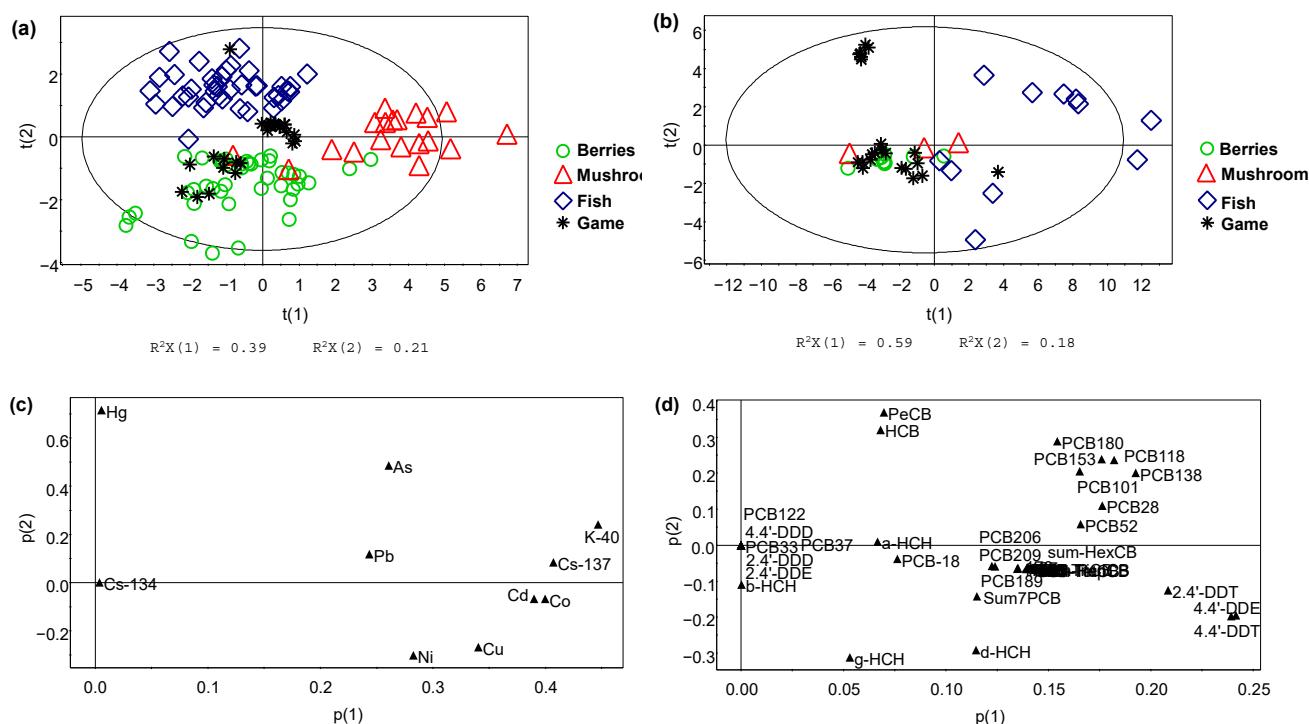


Figure 3. PCA and score plots of (a) model 2a and (b) model 2b. Samples located in the same area of the plot have similar compositions of pollutants. Loading plots of (c) model 2a and (d) model 2b.

In model 2a, there is a clear clustering of berries, mushrooms, and fish, with some overlap of game with berries and fish. The location of mushrooms on the positive side of the t_1 axis is correlated to the radioactivity and concentrations of some metals, especially Cd and Co, as seen in the loading plot (furthest from the origin on the p_1 axis). The location of fish in the fourth quadrant is likely due to low radioactivity and the highest concentrations of Hg (located opposite on the t_1 axis in the scores plot compared to radioactivity on the p_1 axis in the loading plot, and Hg is furthest from the origin on the p_2 axis). The location of most berries in the third quadrant is likely due to low radioactivity, lower concentrations of Hg, and higher concentrations of Ni and Cu. In the loading plot, Cu and Co are clustered, indicating a correlation between the two in the samples. The other metals are not clustered, showing that the metal concentrations vary between the samples.

The fish samples are in the first and second quadrants, which correlates to higher concentrations of POPs in the loading plot. There are, however, some differences in the distribution along the second component (t_2 axis), suggesting a variation in the composition of POPs in the fish samples. The berries and mushroom samples are largely located in the third quadrant close to the t_1 axis, indicating lower concentrations of POPs. The game samples are clustered in several groups, suggesting variations in the composition of POPs in the game samples. In the loading plot, the PCB congeners, apart from PCB-18, are clustered, suggesting a correlation between these in the samples.

Models 1a-1c and 2a-2c showed some clustering for the different groups of samples; for game, there was an additional indication of difference in the composition of pollutants. To further investigate if these are related to the cluster patterns within the type of sample (i.e., type of berry, mushroom, fish, or game), PCA models of each sample type were calculated.

3.4.3. Pollutant Trends for Berries

For the berries, two PCA models were calculated as follows:

Model 3a: Samples analyzed for radioactivity and metals (43 samples)

Model 3b: Samples analyzed for HCB, PCB, and DDT (9 samples)

The PCA models were good, explaining 69–71% of the modeled data. In the model of radioactivity and metals, there was a clustering of cloudberry in quadrants 1 and 4 of the scores plot (Figure 4), while the other berries were not clustered according to berry type and were primarily located in the second and third quadrants. The location of cloudberry, separated from the other berries, is related to the second component (t_2 axis), and the loading plot shows that radioactivity and the concentration of Cd had the highest influence on the second component (furthest from the origin on the p_2 axis). The positive correlation indicates that cloudberry generally have higher radioactivity and Cd concentrations. In the loadings plot, Co and Ni were clustered, meaning they were correlated in the berry samples. All other radionuclides and metals were not correlated, indicating different concentration trends in the berries. In the PCA scores plot of the samples analyzed for POPs, there was no clear clustering according to berry type and, therefore, no clear trends for POP concentrations. In the loading plot, the PCB congeners with five or more chlorides were clustered, indicating a correlation of these in the nine berry samples.

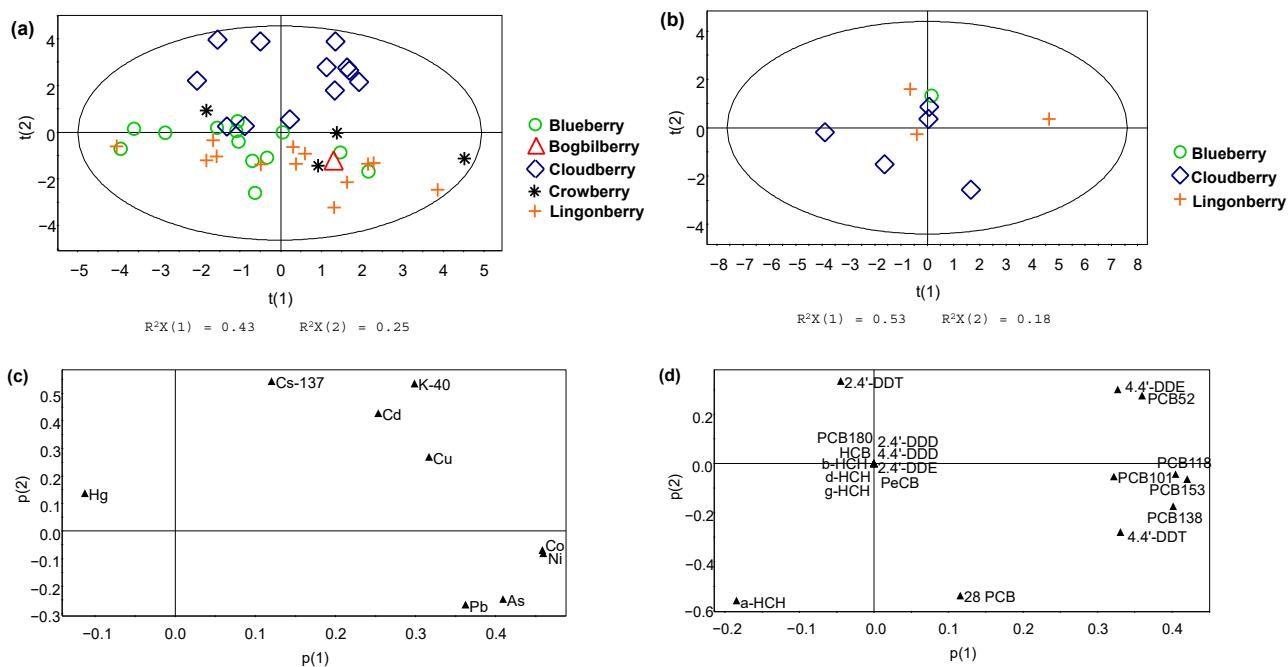


Figure 4. PCA and score plots of (a) model 3a (radioactivity and metals) and (b) model 3b (POPs). Samples located in the same area of the plot have similar compositions of pollutants. Loading plots of (c) model 3a and (d) model 3b.

3.4.4. Pollutant Trends for Mushroom

For mushrooms, one PCA model was calculated that included 19 samples analyzed for radioactivity and metal concentrations (model 3c). Only three samples of mushrooms were analyzed for POPs, and a PCA model was not calculated for such a limited dataset. The model was good, explaining 65% of the variance in the dataset. In the scores plot (Figure 5), there appears to be some clustering of Cortinarius (Gypsy mushroom) and Leccinum

(Orange Birch Bolete) located in the first and second quadrants close to and almost along the t1 axis, meaning that they are primarily affected by the first component and loadings along the p1 axis. In the scores plot, there are also a couple of examples of samples located opposite; for instance, Lactarius, with two samples located opposite on the t1 axis, and the mixed mushroom samples, located opposite on the t2 axis, indicating a difference in metal/radionuclide composition. In the loading plot, As, Cu, Ni, and radionuclides are clustered, indicating that they are correlated in the mushroom samples. Due to their location furthest from the origin on the p1 axis but close to the origin of the p2 axis, they have the highest influence on the distribution of scores on the t1 axis. Samples in the first and second quadrants of the scores plot (mostly Cortinarius and Leccinum), hence, have the highest measured radioactivity and concentration of As, Cu, and Ni.

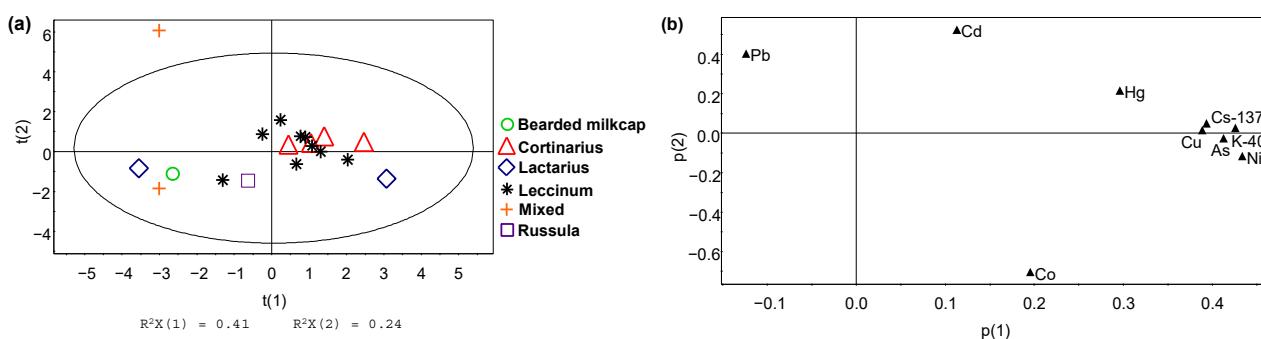


Figure 5. PCA and score plots of (a) model 3c (radioactivity and metals, mushroom). Samples located in the same area of the plot have similar compositions of pollutants. Loading plots of (b) model 3c.

3.4.5. Pollutant Trends for Fish

For the fish, three PCA models were calculated as follows:

Model 3d: Samples analyzed for radioactivity and metals (40 samples)

Model 3e: Samples analyzed for HCB, PCB, and DDT (11 samples)

Model 3f: Samples analyzed for dioxins (6 samples)

All models were good, explaining 57–87% of the datasets. In model 3f, only two types of fish were included (char and trout), and there was no clear trend for the differences in the composition of dioxins in the fish. For this reason, the result of the model is not presented here; please refer to the Appendix B for details and plots. The results of the PCA models 3d and 3e are illustrated in Figure 6. In model 3d, there is some clustering in the scores plot related to the type of fish. Trout and char are located furthest to the right (quadrants 1 and 2), and pike and white fish are located furthest to the left (quadrants 3 and 4), while perch is overlapping between these clusters. The loading plot reveals that Co, Cd, and Ni have the highest influence on the distribution along the t1 axis in the scores plot, meaning that char and trout have higher concentrations of these metals. Radioactivity has the highest influence on the distribution along the t2 axis in the scores plot; this is, however, not related to the type of fish, as all of them are spread along the t2 axis. In the loading plot, there is a clustering of, respectively, radionuclides and Cd and Ni, indicating a correlation in the fish samples. The other metals are not correlated.

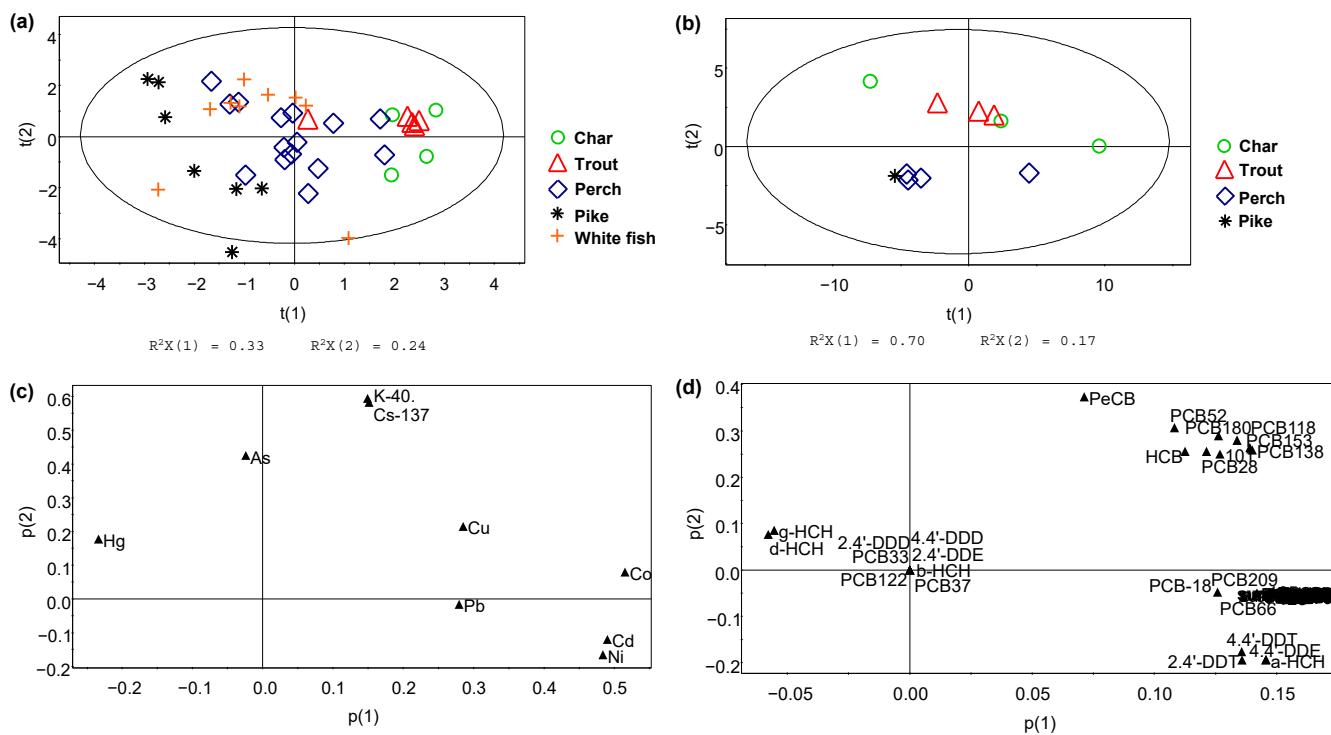


Figure 6. PCA and score plots of (a) model 3d (radioactivity and metals, fish) and (b) model 3e (POPs, fish). Samples located in the same area of the plot have similar compositions of pollutants. Loading plots of (c) model 3d and (d) model 3e.

The scores plot of model 3f shows that the char and trout samples are in the first and fourth quadrants, while perch and pike are in the second and third quadrants. The loading plot shows that the difference between char/trout and perch/pike in the scores plot (related to the second component, t_2) is largely concentrations of PeCB and PCB congeners. Char and trout have higher concentrations of these pollutants. In the loading plot, there is a clustering of PCB congeners, HCH compounds, and DDT compounds, respectively, indicating a correlation in the composition of these pollutants in the fish samples.

3.4.6. Pollutant Trends for Game

For the game, three PCA models were calculated as follows:

Model 3g: Samples analyzed for radioactivity and metals (26 samples)

Model 3h: Samples analyzed for HCB, PCB, and DDT (22 samples)

Model 3i: Samples analyzed for dioxins (7 samples)

All models were good, explaining 67–98% of the datasets. Model 3i only included reindeer, and there was no clear trend for the content of dioxins and PCB in the reindeer. For this reason, the result of the model is presented in Appendix B. The results of the PCA models 3g and 3h are illustrated in Figure 7. In the PCA scores plot of radioactivity and metals, there is clear clustering occurring according to reindeer, moose, and ptarmigan. The distribution of the game samples along the first component, t_1 , is highly influenced by K-40, Cs-137, and Hg (positively correlated), and Co (negatively correlated), meaning that the reindeer have higher levels of radioactivity and Hg, and lower concentrations of Co than the moose and ptarmigan samples. The distribution of moose along the second component, t_2 , is highly influenced by Pb (positive correlation) and Cu (negative correlation). The moose located further up on the t_2 axis has a higher concentration of Pb and the lowest concentration of Cu. K-40 and Cs-137 are clustered in the loading plot and are correlated in the game samples. The metals are not clustered in the loading plot and, hence not correlated in the game samples.

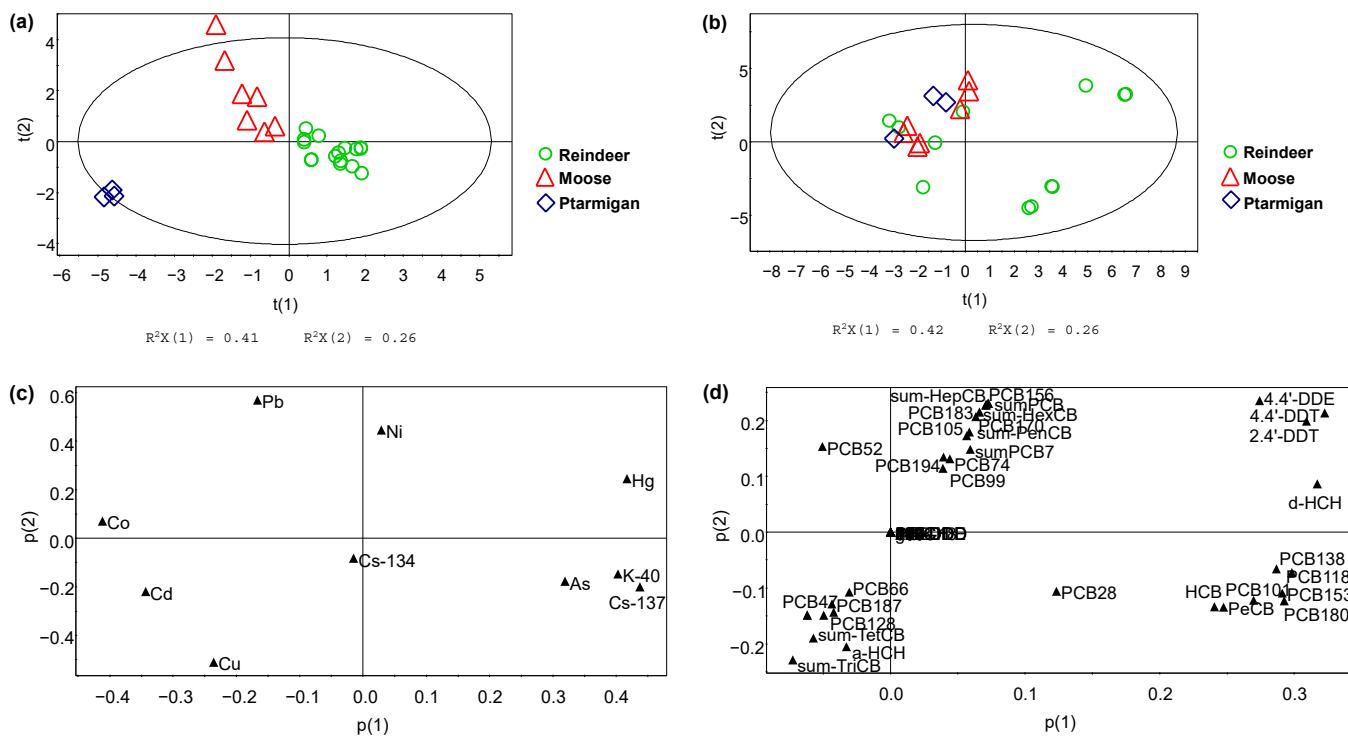


Figure 7. PCA and score plots of (a) model 3g (radioactivity and metals, game) and (b) model 3h (POPs, fish). Samples located in the same area of the plot have similar compositions of pollutants. Loading plots of (c) model 3g and (d) model 3h.

In model 3h, some of the reindeer are in the first and second quadrants; there are, however, overlaps between some reindeer, all moose, and all ptarmigan samples in quadrants 3 and 4. The loading plot shows that the reindeer samples in the first and second quadrants have the highest concentrations of PCB congeners, with a minimum of five chlorides, a-HCH, and DDE/DDT compounds.

4. Discussion

4.1. Contaminant Concentrations and Pollution Sources

The analyzed natural food items from the Arctic regions of Norway, Finland, and Russia showed low levels of radioactive substances, metals, and POPs without any clear indication of significant pollution from a single point source. Levels above the background could hence be a result of several diffuse sources, and it is not possible to clearly distinguish between local and global sources. There are, however, some trends for the different pollutants that may indicate some important sources.

There were large variations in the activity concentrations of Cs-137 in different species of berries, mushrooms, freshwater fish, and game. However, all environmental samples from Northern Norway and Finland, and freshwater fish samples from Northwest Russia proved the presence of Cs-137, and the activity concentrations were compatible with earlier data from the same regions [12–16]. In the reindeer from Northern Norway sampled in 2013, Cs-134 was detected, indicating that the small traces of radioactive substances that reached Norway after the Fukushima accident were still observable. The atmospheric concentrations from the Fukushima accident were very small. This was due to the dilution in the atmosphere because of the long distance between the Euro-Arctic region and Japan. This led to a very small deposition of Cs-134 and Cs-137 at the level of 0.3–0.8 Bq/m². Very small traces of Cs-134 in environmental samples have been observed in Norway, Finland, and Russia after the accident. The influence of the Fukushima accident on the Arctic environment was observable but insignificant, and it had no effects on human health or the state of the environment [4].

The metal concentrations in the food items were generally low to moderate. Hansen et al. [7] found gradients for the metals As, Co, and Ni, with the highest concentrations close to the Nickel smelter area and decreasing concentrations along the dominating wind direction. The study concluded that the Nickel smelter was the main source of the elevated levels of As, Co, and Ni [7]. No trends for the other metals (Cd, Cu, Hg, and Pb) were found; however, it does not mean that there was no pollution with these metals. Properties of the selected food items and local soil influence the pathway to and uptake of the food items. Cu and Pb are, for instance, amongst the metals with the highest affinity for organic matter, which may prevent uptake in plants. The uptake of metals in plants depends on the metal; e.g., it has been found that the uptake of Pb is more affected by pollutant levels in soil, whereas elevated concentrations of Cu in soil do not necessarily cause elevated concentrations in plants. Based on the Cd, Cu, Hg, and Pb concentrations, it is not possible to neither confirm nor disprove that the Nickel smelter is the major source of elevated concentrations. Thus, to identify and assess the source of elevated concentrations, follow-up studies into environmental compartments are required.

DDT is a legacy pollutant that is no longer in use in all three countries across the study area. Since there are no point sources of DDT in the three countries, the source is diffuse; however, it is not possible to assess whether DDT and its metabolites occur as a result of either this pesticide use in the past (before 1990) or from long-range transportation.

Also, HCH is a legacy pollutant that is no longer in use in Finland, Norway, or Russia. Most samples had concentrations below the detection limit. Eight samples above the detection limit for the HCH group indicated a different uptake in the food items, e.g., berries had concentrations of a-HCH above the detection limit, while fish and game showed an uptake of α -, δ - and γ -HCH. This is also apparent in the PCA loading plots, in which α -, δ - and γ -HCH are not clustered; i.e., there is no correlation between the different forms of HCH. The low concentrations and absence of point sources across the study area suggest a diffuse source. It is, however, not possible to assess whether it is a local diffuse source from past practices or a global diffuse source.

PeCB concentrations in berries were below the detection limit in all samples, whereas all fish and game samples analyzed for PeCB had concentrations above the detection limit. There were, however, only nine samples of char, trout, and reindeer. More samples would be needed to assess the source of PeCB.

The PCA plots of all food items revealed that fish had the highest concentrations of PCBs. The PCA loading plots of each food item (berries, fish, and game) showed different trends in the clustering of PCB congeners; i.e., for berries, the clustering pattern was different from that for fish and game. This can be an indication of different pathways for uptake in different food items or different sources of PCB. PCBs are also legacy pollutants; however, since they are connected to construction and building materials, such as paint, insulation, hydraulic oil, electric devices, and cables, they can persist in buildings for many years. That is why PCB levels are usually higher when closer to settled areas. This may be the cause for which a gradient of decreasing PCB concentrations in fish with distance to nickel was observed in an earlier study [5] and the cause for higher concentrations of PCBs close to larger settlements in Norway and Russia (towns like Kirkenes and Nickel) than in rural areas in Finland. PCBs are known to deposit in the high north after long-range transport. Such is the case with the study area, which is most likely affected by diffuse global pollution; however, based on the observed gradients, the local impact on the food items is likely higher.

In the Norwegian part of the study area, concentrations of dioxins and dioxin-like PCBs in reindeer meat were found to be above the maximum levels set by the EU. Analysis of the dioxins and dioxin-like PCBs was not conducted in the Finnish or Russian part of the study area. A local point source of dioxins is known to exist in Kirkenes up to 1996: the iron-ore pellets production of the Sydvaranger A.S. mine, where the process of incineration required for production entailed dioxin emissions. From 1997 through 2009, elevated levels of dioxins were observed in fish from lakes close to Kirkenes. However, according to the

measurement data, over the studied period, fish samples were low in dioxins. The pathway of dioxin emissions to reindeer is unknown and it is impossible to confirm whether the pellets production is the main source of elevated dioxin concentrations.

Overall, current concentrations of most POPs, heavy metals, and radioactive substances are declining in the Arctic regions where the time-trend data exist [1]. However, climate change, permafrost thaw, and new chemicals may change the exposure situation of Arctic populations and wildlife. Climate change may liberate pollution from frozen grounds and glaciers (and increase the risk of re-mobilization of pollutants) as well as cause new chemicals, including micro- and nano-plastics, to spread all over the globe.

4.2. Common Trends and Possible Reference Indicators of Pollution

The composition and concentration of pollutants varied in the different food items. However, some general trends were identified for different pollutants and food items.

The highest radioactivity was observed in mushrooms, game, and berries, in that order. In the PCA plot of mushrooms (Figure 5), it appeared that *Cortinarius* and *Leccinum* had higher radioactivity, but there were not enough samples of other mushroom species ($n = 1$ for most others) to confirm that. However, this corresponds to earlier studies in this field of research [12–16]. The *Cortinarius* or Gypsy mushroom accumulates Cs-137 at a higher rate compared with other mushroom species, and this is a specific feature of this particular mushroom. This species is edible, both by people and animals, so it may be used as a marker for the accumulation of Cs-137 and for monitoring radioactivity transfer via the food chain [4]. The PCA plots for game and berries (Figures 4 and 7) showed that the highest level of radioactivity was found in reindeer and cloudberry, respectively. To monitor the radioactivity levels, it is critical to include mushrooms, reindeer, and cloudberry as markers in food safety assessments. Reindeer, too, is a good indicator in the case of metals (Hg and Cu) and thus may be used in combined risk assessments.

The PCA plot (Figure 6) of fish and metals has indicated that both pike and whitefish (from Russia) have the highest concentrations of mercury; the lowest concentrations are detected in char and trout. High levels of Hg have previously been observed in a study of freshwater fish from the Pasvik watercourse, where the highest levels were measured in the piscivorous fish species, and pike in particular [17]. In berries, mushrooms, and game, the concentrations of mercury were below levels of concern. All the samples except eight samples of berries, mushrooms, and game analyzed in this research were found to be low in other metals. In the PCA plot, there was generally a lack of correlation between the analyzed metals (a limited clustering of metals in the PCA loading plots was demonstrated). This shows that the accumulation of metals in food items varies depending on the metal. Therefore, the same type of food item cannot be a good indicator of all types of metal pollution.

In general, the highest levels of POPs, except dioxins, were measured in fish. This is expected, as the freshwater food chain up to piscivorous fish generally is longer than the terrestrial one and because the fish have higher levels of lipids in their muscle than the game samples. Pike and perch are, therefore, often used in monitoring programs for POPs and mercury.

Game had concentrations of PCB above the detection limits, with the highest concentrations in reindeer sampled in Norway. However, there were not enough data to assess if reindeer are a better marker for POPs in the terrestrial environment than, for e.g., moose or ptarmigan. The highest concentrations were in reindeer from Pasvik, Norway, with no analysis for PCBs in moose or ptarmigan.

PCDD/F were only analyzed in reindeer and a few fish samples (char and trout), and the PCA scores and loading plots in Figure 2 show that the congener patterns differed between reindeer and fish. Generally, the PCDD congeners had higher concentrations in reindeer, while the PCDF congeners had higher concentrations in fish. Due to a limited number of samples ($n = 6$ reindeer; $n = 6$ fish), it is not clear if this trend is general. More analyses of fish and game are required to confirm and build on these initial findings.

4.3. Safety of Locally Harvested Foods

4.3.1. Radioactive Substances, Metals, POPs, and Dioxins

Radioactive substances of natural or artificial origin are present in our food. Some of them are absorbed by plants and animals from the soil or water, entering the food chains that make up our diet. Potassium-40, polonium-210, carbon-14, and lead-210 are among the substances that contribute much to the radiation dose from food in the population. The most problematic in the longterm (after the Chernobyl accident of 1986) has been radioactive cesium-137 (Cs-137), as its half-life is 30 years, and it is readily absorbed into the food chains. Cs-137 may be found in low levels in most foods [18].

After the Chernobyl accident, the authorities introduced national maximum permitted levels for Cs-137 applicable only to food for sale. Different countries could introduce their own national limits. For example, in Finland and Russia, the national limits for Cs-137 in edible products are given for fresh weight. In the case of dried products, the activity concentrations must be converted from dry to fresh weight. If so, all the mushrooms and berries would stay below the limits, with the approximation of 10% dry matter content. In Norway, the same limit is applied for dried and fresh samples. Thus, several mushroom samples can exceed this limit if they are dried, while all berries are clearly below it. If measured mushrooms are fresh, they will also stay below the limit. If we assume that the average dry matter content of these products is 10%, it will mean that the activity concentration is 10 times higher in dried products compared with those fresh. It needs to be mentioned, too, that the consumption of such dried products as berries, mushrooms, or meat is much less than their consumption of fresh foods, and the actual dose caused by eating dried products can be assumed to be about the same as if they were eaten as fresh. It is estimated that in Finland and Norway, only one percent of the annual dose of radioactive exposure is caused by foodstuffs. From positions of radiation safety, the edible natural products in the Euro-Arctic area are safe for use and do not pose health risks [18,19]. In general, eating foodstuffs with activity concentrations over the national limits does not cause any immediate health risks; however, the risk of cancer could increase together with the level of radiation exposure [20]. In emergency cases, different (higher) limits could be applied. The maximum permitted levels for concentrations of radioactive elements in emergency situations are provided in the Euratom Council regulation 2016/52 [21].

The concentrations of metals were generally low in all the food samples in comparison with the EU food safety threshold values. A few samples exceeded the maximum levels [7]. Two berry and five mushroom samples exceeded the threshold value for Cd, one mushroom and one game sample exceeded the threshold value for Pb, and two fish samples exceeded the threshold value of Hg. One mushroom sample from Finland exceeded the threshold value for Cd by 80 times and the threshold value for Pb by 30 times. The EU has not set threshold values for As, Co, Cu, and Ni in food items, and to assess the risk of these metals in food items, tolerable daily intake (TDI) values are often used. The European Food Safety Authority (EFSA) advises TDI values for Co (0.0014 mg/kg bw/d), Cu (0.07 mg/kg bw/d), and Ni (0.013 mg/kg bw/d) based on exposure assessments of all sources [22–24]. For As, the EFSA has established a benchmark dose lower confidence limit (BMDL), that can be used as a reference point to derive the TDI [25]. Based on the 95% confidence level of concentrations of the food items in this study, daily consumption of several kg of berries, mushrooms, fish, or game daily would be necessary to exceed the TDI values. For this reason, the consumption of food items in themselves is not assessed as posing a risk for excessive exposure to Co, Cu, and Ni.

POPs and mercury (i.e., methylmercury) are lipophilic and persistent compounds, have higher levels in more lipid-rich tissues of fish and other animals, and tend to accumulate as the species mature. Most of the examined food samples showed detectable levels of POPs, but these levels were very low in berries, game, and fish from the border regions of the three countries and were well below the threshold values set by the EU for the safe consumption of commercial food, except for the reindeer samples from Norway.

Six additional muscle and kidney fat samples of reindeer from 2015 and 2016 were analyzed for dioxins by NILU, cooperatively with the County Governor of Finnmark and the Norwegian Food Safety Authority. These samples came from the Norwegian part of the border area (the results were included and used in the risk assessment report by Knutsen et al. [26]). When we combine these new results with those from the present study, we see that dioxins and dl-PCBs in 55% of the nine reindeer samples exceeded the maximum levels set by the EU for dioxins and dioxin-like PCBs in meat and meat products of venison (7.5 pg TE/g fat [1]). The levels of non-dioxin-like PCBs (ICES-6) did not exceed the EU's maximum levels.

4.3.2. Combination of Different Contaminant Groups and Risks

Currently, the chemical risk assessment in the European Food Safety Authority (EFSA) and the EU mainly relies on the assessment of individual substances or, in a few cases, groups of substances that are expected to occur together. The EFSA aims that by 2030, the agency and its partners will be ready for the routine implementation of a human health risk assessment of multiple chemicals—also referred to as a mixture risk assessment [27]. The EU maximum threshold and TDI values are appropriate tools for assessing the toxicological risk of individual pollutants. However, even though the effect of pollutants individually is negligible, the combination of several pollutants, even at low concentrations, may pose a risk to human health. The exposure-caused risks must be compared with other risks that may be present—a synergy of risks. Additional supplementing doses/risks should be considered along with the existing exposure/doses. The degree of impact can vary largely, depending on an individual contaminant, its accumulation and distribution peculiarities, level of toxicity, etc. A combination of combined pollution from different contaminants can trigger different types of health effects, depending on the doses/quantity [28].

There is a need for governmental agencies to develop methods and derive threshold values to be used to consider the potential synergic effects of radioactive substances, metals, and POPs in natural food products. It is impossible at this point to assess the potential synergic effects of exposure by only considering the concentrations of different pollutants in this study. One of the potential risk evaluation ways is by using a tolerable daily or weekly intake (TDI or TWI) of various contaminants or a similar threshold, such as an acceptable daily intake (ADI), and so on. This was done by Hansen et al. [7] for a few cases where the metals Cd, Hg, and Ni exceeded the EU maximum levels in food [7]. TWIs are available for Cd (2.5 µg/kg body weight (b.w.) and Hg (4 and 1.3 µg/kg b.w. for Hg and methyl-Hg, respectively). A TDI has also been set for Ni (2.8 µg/kg b.w.).

Based on the samples with the highest concentrations and those exceeding the EU's maximum levels, calculations revealed that over 5 kg of perch may be eaten yearly before the TWI for methyl-Hg was reached. Almost the same amount of mushrooms may also be consumed before thresholds were reached for Cd and Ni [7]. These calculations did not consider food intake from other sources and were also limited by the lack of food intake information from local people. However, this implied that moderate consumption of wild food from the study area, except the surroundings of the smelter, could be considered safe.

In 2018, the TWI of dioxins and dl-PCBs was lowered from 14 to 2 pg toxic equivalents (TEQ) per kilogram of body weight per week [29] due to new knowledge regarding the adverse effects on semen quality and the possible effects on male fertility. Five reindeer neck muscle samples from 2013 and 2015 exceeded the EU's maximum limit of 7.5 pg TE/g Iw for dioxin and dl-PCBs in venison meat. A risk assessment of dioxins and dl-PCBs in reindeer meat and liver in the Norwegian population has been performed by the Norwegian Scientific Committee for Food and Environment [26]. These data from the present study and additional data from 2015 and 2016 analyses were used in the calculations and the assessment; for more details, see the report [26]. Knutsen et al. [26] concluded that one weekly meal of reindeer meat contributes approximately 23% of the TWI to adults, whereas a weekly portion of reindeer fat contributes approximately 34% of the TWI to adults. Reindeer meat is included in a regular Norwegian diet. However, we expect it to

be consumed relatively rarely by most people [26]. Some sub-populations, e.g., reindeer herders, may consume much more than the average person and potentially more fat-rich parts of the animal, and for those who consume the most, the dioxin levels may pose a health risk.

Recently, the European Commission for Chemical Strategy for Sustainability has announced the introduction of a mixture assessment factor (MAF) to address mixtures' assessments under REACH [30,31]. An example is the approach with the use of MAF in a single substance risk assessment [30]. A MAF can be a pragmatic solution that circumvents the need to assess every possible substance combination. With this approach, the possible health risk was calculated with individual chemicals' risk quotients using health-based guidance values and then adding them up to derive the hazard index for the mixture [30].

To the best of our knowledge, a few studies have tried to assess the risk of combined exposure through diet from the occurrence data of radioactive substances, metals, and organic pollutants. A study of a high natural background radiation area in China assessed the dietary exposure of radionuclides and heavy metals through food consumption [32]; the result obtained indicated that the health risk posed by the radioactivity from food intake was probably small, but the obtained heavy metal hazard index (HI) was much higher than 1.0, indicating that heavy metals may have adverse effects on human health, especially lead, cadmium, and arsenic pollution [32].

With an overview of how people can be exposed to different contaminants, it will also be possible to develop a strategy for the optimal management of dose and consequence reduction.

5. Conclusions

To comprehensively assess human health, it is important to have a combined approach for the identification of all contaminant sources and assess the cumulative effects and potential risks from pollution exposure, as in their everyday life, humans are affected not by one single group but a mixture of various groups of contaminants (exposure). This is also true in terms of food safety. For the local population and people who regularly harvest or hunt, it is important to obtain scientific advice and information on food safety, food-caused possible risks, and diet recommendations. This is especially true with respect to areas with existing exposure situations and added industrial pollution.

Due to efficient multidisciplinary cooperation in our project, we had a unique opportunity to measure levels of different contaminant groups in the same food items. The differences in pollutant compositions and levels allowed us to assess how local and global pollution sources might influence food safety in the border regions of Norway, Finland, and Russia in the Arctic. Statistical analyses made it possible to reveal pollutant–food item correlations, identify the indicators or reference markers to be used in future monitoring of pollution and risks for human health, and perform the initial step of analysis of the cumulative effects. However, at present, we lack sufficient health guideline values for the various pollutants in order to estimate the combined exposure from a mixture of contaminants in food items; for instance, with the mixture assessment factor (MAF) recommended by the EU. In this pilot study, we characterized the cumulative effects by using principal component analysis (PCA). Through novel analysis, we identified mushrooms, inland fish, and reindeer as important food items that enhance our understanding of combined exposures to toxic elements and radioactivity. We also could identify and provide recommendations for certain reference species in different contaminant groups. For more detailed investigations, conducting more comprehensive multidisciplinary studies and developing a methodology for combined risk assessments are required. This is also advisable for governmental institutions and ministries that are involved in human health and environmental risk assessments. Moreover, the outcomes are also of interest to the authorities that are providing recommendations on acceptable limits, harmful effects, and doses, as well as issuing regulations.

To ensure that natural food products are safe for consumption, continuous monitoring of foodstuff pollution (metals, POPs, and radioactive substances) is important. Having

in mind the cross-border nature of any potential contamination, one should consider the risks related to pollution by different types of contaminants through monitoring and protection at a regional and international level. In this context, a key role is given to cross-border cooperation.

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Appendix A

Table A1. Overview of samples and species collected from the border regions across Norway, Finland, and Russia.

	Norway		Russia		Finland	
	n Sites	n Samples	n Sites	n Samples	n Sites	n Samples
Berries	16		56	4	10	3
Bilberry (<i>Vaccinium myrtillus</i>)	8	20	4	4	1	3
Lingonberry (<i>Vaccinium vitis-idaea</i>)	6	16	4	4	2	3
Cloudberry (<i>Rubus chamaemorus</i>)	7	11	-	-	1	3
Crowberry (<i>Empetrum nigrum</i>)	3	9	1	1	-	-
Bog bilberry (<i>Vaccinium uliginosum</i>)	-	-	1	1	-	-
Mushrooms	6	24	4	8	2	3
Orange birch bolete (<i>Leccinum versipelle</i>)	4	12	-	-	-	-

Table A1. Cont.

	Norway		Russia			Finland
	n Sites	n Samples	n Sites	n Samples	n Sites	n Samples
Orange oak bolete (<i>Leccinum aurantiacum</i>)	-	-	4	4	-	-
Birch bolete (<i>Leccinum scabrum</i>)	-	-	1	1	-	-
Gypsy mushroom (<i>Cortinarius caperatus</i>)	4	12	-	-	-	-
Rufous milkcap (<i>Lactarius rufus</i>)	-	-	-	-	1	1
Bearded milkcap (<i>Lactarius torquatus</i>)	-	-	1	1	-	-
Rollrim milkcap (<i>Lactarius resimus</i>)	-	-	1	1	-	-
Russula (<i>Russulaceae</i>)	-	-	1	1	-	-
Mixed mushrooms	-	-	-	-	1	2
Fish muscle	7 lakes	19	5 lakes	14	2 lakes	8
Whitefish (<i>Coregonus lavaretus</i>)	2	2	4	4	1	3
Perch (<i>Perca fluviatilis</i>)	3	8	4	4	2	4
Pike (<i>Esox lucius</i>)	2	2	4	4	1	1
Arctic char (<i>Salvelinus alpinus</i>)	3	3	1	1	-	-
Brown trout (<i>Salmo trutta</i>)	4	4	1	1	-	-
Game muscle	2 regions	13	1 region	1	6 regions	9
Reindeer (<i>Rangifer tarandus</i>)	1	10	-	-	1	3
Moose (<i>Alces alces</i>)	1	3	1	1	3	3
Ptarmigan (<i>Lagopus lagopus</i>)	-	-	-	-	2	3

Source: Hansen et al., 2017 [7].

Appendix B

Overview of compounds analyzed for radioactivity, heavy metals, and POPs in all countries.

Table A2. Laboratory measurements of different contaminants in the same food items.

Berries	Units	N	Min	Max	Average	Median
Cs-137	Bq/kg dw	25	3	175	43	35
K-40	Bq/kg dw	24	0.3	23	5.7	4.7
As	mg/kg	43	0.0001	0.042	0.00690	0.00330
Cd	mg/kg	43	0.0001	0.04	0.0050	0.0008

Table A2. *Cont.*

Berries	Units	N	Min	Max	Average	Median
Co	mg/kg	34	0.001	0.18	0.027	0.014
Cu	mg/kg	43	0.32	1.7	0.85	0.83
Hg	mg/kg	35	0.0003	0.009	0.0022	0.0022
Ni	mg/kg	43	0.03	2.3	0.61	0.51
Pb	mg/kg	43	0.0004	0.06	0.008	0.004
PeCB	ng/g ww	9	<0.003	<0.003	-	-
HCB	ng/g ww	9	<0.014	<0.014	-	-
PCB-28	ng/g ww	9	0.0049	0.008	0.005	0.005
PCB-52	ng/g ww	9	0.0041	0.040	0.013	0.013
PCB-101	ng/g ww	9	0.0007	0.098	0.017	0.008
PCB-118	ng/g ww	9	0.0087	0.118	0.048	0.045
PCB-138	ng/g ww	9	0.03	0.137	0.060	0.055
PCB-153	ng/g ww	9	0.0061	0.104	0.034	0.027
PCB-180	ng/g ww	9	<0.0055	<0.0055	-	-
a-HCH	ng/g ww	9	0.0035	0.0319	0.0085	0.0035
b-HCH	ng/g ww	9	<0.0052	<0.0052	-	-
g-HCH	ng/g ww	9	<0.0064	<0.0064	-	-
d-HCH	ng/g ww	9	<0.01	<0.01	-	-
2,4'-DDE	ng/g ww	9	<0.0064	<0.0064	-	-
4,4'-DDE	ng/g ww	9	0.0065	0.0338	0.0135	0.0103
2,4'-DDD	ng/g ww	9	<0.0036	<0.0036	-	-
4,4'-DDD	ng/g ww	9	<0.001	<0.001	-	-
2,4'-DDT	ng/g ww	9	0.0024	0.0038	0.0025	0.0024
4,4'-DDT	ng/g ww	9	0.0026	0.0197	0.0071	0.0071
Mushrooms	Units	N	Min	Max	Average	Median
Cs-137	Bq/kg dw	11	3.6	1479	426	143
K-40	Bq/kg dw	11	86	1475	761	813
As	mg/kg	19	0.004	0.15	0.04	0.03
Cd	mg/kg	19	0.009	42	2.5	0.20
Co	mg/kg	16	0.018	0.91	0.16	0.08
Cu	mg/kg	19	1.2	33	7.3	6.2
Hg	mg/kg	19	0.003	0.15	0.05	0.02
Ni	mg/kg	19	0.037	15	1.7	0.79
Pb	mg/kg	19	0.004	31	1.7	0.03
HCB	ng/g ww	3	<0.014	<0.014	-	-
PCB-28	ng/g ww	1	0.008	0.008	-	-
PCB-52	ng/g ww	3	0.003	0.055	0.027	0.022
PCB-101	ng/g ww	3	0.01	0.121	0.056	0.038
PCB-118	ng/g ww	3	0.026	0.112	0.063	0.052
PCB-138	ng/g ww	3	0.02	0.136	0.074	0.071
PCB-153	ng/g ww	3	0.004	0.114	0.054	0.044
PCB-180	ng/g ww	3	<0.0055	<0.0055	-	-

Table A2. *Cont.*

Berries	Units	N	Min	Max	Average	Median
a-HCH	ng/g ww	3	<0.003	<0.003	-	-
b-HCH	ng/g ww	3	<0.0052	<0.0052	-	-
g-HCH	ng/g ww	3	<0.0064	<0.0064	-	-
d-HCH	ng/g ww	3	<0.01	<0.01	-	-
2,4'-DDE	ng/g ww	3	<0.0064	<0.0064	-	-
4,4'-DDE	ng/g ww	3	0.007	0.042	0.024	0.023
2,4'-DDD	ng/g ww	3	<0.0036	<0.0036	-	-
4,4'-DDD	ng/g ww	3	<0.001	<0.001	-	-
2,4'-DDT	ng/g ww	3	<0.0024	<0.0024	-	-
4,4'-DDT	ng/g ww	3	0.003	0.032	0.017	0.017
Fish	Units	N	Min	Max	Average	Median
Cs-137	Bq/kg dw	26	1.6	96	37	34
K-40	Bq/kg dw	25	74	713	390	474
As	mg/kg	37	0.002	0.14	0.03	0.019
Cd	mg/kg	37	0.0002	0.01	0.002	0.002
Co	mg/kg	32	0.001	0.06	0.01	0.008
Cu	mg/kg	37	0.09	0.67	0.28	0.23
Hg	mg/kg	37	0.012	0.86	0.16	0.11
Ni	mg/kg	38	0.004	0.16	0.04	0.023
Pb	mg/kg	37	0.0004	0.18	0.02	0.007
2378-TCDD	pg/g ww	6	0.02	0.12	0.06	0.05
12378-PeCDD	pg/g ww	6	0.06	0.27	0.12	0.09
123478-HxCDD	pg/g ww	6	0.01	0.04	0.02	0.02
123678-HxCDD	pg/g ww	6	0.02	0.09	0.04	0.03
123789-HxCDD	pg/g ww	5	0.01	0.02	0.02	0.02
1234678-HpCDD	pg/g ww	6	0.02	0.04	0.03	0.03
OCDD	pg/g ww	6	0.07	0.14	0.11	0.11
SUM PCDD	pg/g TE	6	0.09	0.40	0.18	0.15
2378-TCDF	pg/g ww	6	0.48	2.98	1.04	0.68
12378/12348-PeCDF	pg/g ww	6	0.04	0.20	0.09	0.07
23478-PeCDF	pg/g ww	6	0.12	0.56	0.24	0.19
123478/123479-HxCDF	pg/g ww	5	0.01	0.05	0.03	0.04
123678-HxCDF	pg/g ww	6	0.01	0.05	0.03	0.03
234678-HxCDF	pg/g ww	5	0.01	0.04	0.02	0.03
1234678-HpCDF	pg/g ww	3	0.01	0.02	0.01	0.01
SUM PCDF	pg/g TE	6	0.09	0.49	0.19	0.14
33'44'-TeCB (PCB-77)	pg/g ww	6	8.5	33	13.6	10.0
344'5-TeCB (PCB-81)	pg/g ww	6	0.61	2.8	1.4	1.2
33'44'5-PeCB (PCB-126)	pg/g ww	6	3.4	18	7.3	5.7
33'44'55'-HxCB (PCB-169)	pg/g ww	6	1.0	4.8	1.9	1.3
SUM TE-PCB	pg/g TE	6	0.38	1.9	0.79	0.62

Table A2. *Cont.*

Berries	Units	N	Min	Max	Average	Median
Sum PCDD/PCDF (TE(2005))	pgTE/g	6	0.23	0.89	0.37	0.26
SUMPCDD/PCDF + TE-PCB	pgTE/g	6	0.61	2.8	1.2	0.88
SUMPCDD/PCDF + TE-PCB	pgTE/g lw	6	18	126	44	32
PeCB	ng/g ww	11	0.003	0.029	1.3	0.84
PeCB	ng/g lw	6	0.63	0.01	0.86	0.40
HCB	ng/g ww	11	0.01	0.02	0.71	0.49
HCB	ng/g lw	6	12	32	19	17
PCB-18	ng/g ww	1	<0.013	<0.013	-	-
PCB-28	ng/g ww	11	0.005	0.07	0.04	0.04
PCB-31	ng/g ww	6	0.02	0.05	0.03	0.03
sum-TriCB	ng/g ww	6	0.07	0.16	0.11	0.10
PCB-47	ng/g ww	6	0.02	0.05	0.04	0.03
PCB-52	ng/g ww	11	0.003	0.16	0.05	0.04
PCB-66	ng/g ww	6	0.07	0.20	0.14	0.14
PCB-74	ng/g ww	6	0.04	0.16	0.10	0.09
sum-TetCB	ng/g ww	6	0.18	0.56	0.37	0.35
PCB-99	ng/g ww	6	0.15	0.74	0.41	0.37
PCB-101	ng/g ww	11	0.004	0.92	0.33	0.35
PCB-105	ng/g ww	6	0.17	0.81	0.42	0.39
PCB-114	ng/g ww	6	0.01	0.07	0.03	0.03
PCB-118	ng/g ww	11	0.05	1.9	0.65	0.55
PCB-123	ng/g ww	6	0.01	0.05	0.02	0.02
sum-PenCB	ng/g ww	6	0.94	4.5	2.4	2.2
PCB-128	ng/g ww	6	0.13	0.66	0.31	0.26
PCB-138	ng/g ww	11	0.08	3.3	1.0	1.0
PCB-141	ng/g ww	6	0.06	0.40	0.18	0.16
PCB-149	ng/g ww	6	0.15	0.86	0.39	0.35
PCB-153	ng/g ww	11	0.08	4.6	1.4	1.3
PCB-156	ng/g ww	6	0.07	0.39	0.19	0.16
PCB-157	ng/g ww	6	0.02	0.09	0.04	0.04
PCB-167	ng/g ww	6	0.04	0.21	0.09	0.09
sum-HexCB	ng/g ww	6	1.9	10.5	5.0	4.7
PCB-170	ng/g ww	6	0.07	0.37	0.17	0.15
PCB-180	ng/g ww	11	0.01	1.9	0.47	0.33
PCB-183	ng/g ww	6	0.05	0.51	0.19	0.16
PCB-187	ng/g ww	6	0.17	1.55	0.60	0.49
PCB-	ng/g ww	4	0.01	0.03	0.02	0.01
sum-HepCB	ng/g ww	6	0.53	4.4	1.8	1.5

Table A2. *Cont.*

Berries	Units	N	Min	Max	Average	Median
PCB-194	ng/g ww	6	0.04	0.46	0.16	0.13
PCB-206	ng/g ww	5	0.01	0.05	0.02	0.02
PCB-209	ng/g ww	4	0.01	0.03	0.02	0.02
Sum7PCB	ng/g ww	6	2.3	12.9	6.2	6.0
Sum7PCB	ng/g lw	6	67	580	232	184
sumPCB	ng/g ww	6	3.7	21	9.8	9.4
sumPCB	ng/g lw	6	106	929	366	286
a-HCH	ng/g ww	5	0.003	0.023	0.007	0.003
b-HCH	ng/g ww	5	<0.005	<0.005	-	-
g-HCH	ng/g ww	5	0.006	0.023	0.010	0.006
d-HCH	ng/g ww	5	0.010	0.015	0.012	0.010
2,4'-DDE	ng/g ww	5	<0.006	<0.006	-	-
4,4'-DDE	ng/g ww	5	0.08	1.9	0.47	0.10
2,4'-DDD	ng/g ww	5	<0.004	<0.004	-	-
4,4'-DDD	ng/g ww	5	<0.001	<0.001	-	-
2,4'-DDT	ng/g ww	5	0.002	0.14	0.03	0.01
4,4'-DDT	ng/g ww	5	0.02	0.61	0.16	0.05
Game	Units	N	Min	Max	Average	Median
Cs-134	Bq/kg dw	10	0.53	1.5	1.0	1.1
Cs-137	Bq/kg dw	25	3.7	523	248	271
K-40	Bq/kg dw	25	3.5	144	69	75
As	mg/kg	26	0.001	0.05	0.02	0.020
Cd	mg/kg	26	0.0007	0.02	0.003	0.002
Co	mg/kg	17	0.005	0.01	0.01	0.008
Cu	mg/kg	26	1.2	5.2	2.3	2.1
Hg	mg/kg	26	0.0005	0.0047	0.003	0.004
Ni	mg/kg	26	0.003	0.06	0.01	0.008
Pb	mg/kg	26	0.0016	0.34	0.02	0.004
2378-TCDD	pg/g ww	6	0.02	0.03	0.02	0.03
12378-PeCDD	pg/g ww	6	0.05	0.13	0.09	0.10
123478-HxCDD	pg/g ww	6	0.04	0.13	0.09	0.09
123678-HxCDD	pg/g ww	6	0.06	0.16	0.11	0.11
123789-HxCDD	pg/g ww	6	0.02	0.03	0.02	0.02
1234678-HpCDD	pg/g ww	6	0.09	0.19	0.15	0.16
OCDD	pg/g ww	6	0.31	0.68	0.52	0.56
SUM PCDD	pg/g TE	6	0.08	0.20	0.14	0.15
2378-TCDF	pg/g ww	6	0.02	0.03	0.02	0.03
23478-PeCDF	pg/g ww	6	0.06	0.10	0.07	0.06
123478/123479-HxCDF	pg/g ww	6	0.03	0.06	0.04	0.03
123678-HxCDF	pg/g ww	6	0.02	0.04	0.03	0.03
234678-HxCDF	pg/g ww	6	0.01	0.02	0.02	0.01

Table A2. *Cont.*

Berries	Units	N	Min	Max	Average	Median
1234678-HpCDF	pg/g ww	6	0.02	0.03	0.02	0.03
OCDF	pg/g ww	2	<0.03	<0.03	-	-
SUM PCDF	pg/g TE	6	0.03	0.05	0.03	0.03
33°44'-TeCB (PCB-77)	pg/g ww	6	0.41	0.81	0.58	0.52
344°5-TeCB (PCB-81)	pg/g ww	6	0.09	0.14	0.11	0.10
33°44°5-PeCB (PCB-126)	pg/g ww	6	1.1	2.3	1.6	1.2
33°44°55'-HxCB (PCB-169)	pg/g ww	6	0.17	0.48	0.28	0.19
SUM TE-PCB	pg/g TE	6	0.12	0.25	0.16	0.12
Sum PCDD/PCDF (TE(2005))	pgTE/g	6	0.11	0.25	0.18	0.19
SUMPCDD/PCDF + TE-PCB	pgTE/g	6	0.23	0.50	0.35	0.31
SUMPCDD/PCDF + TE-PCB	pgTE/g lw	3	11	12	12	12
PeCB	ng/g ww	19	0.003	0.03	0.01	0.003
PeCB	ng/g lw	3	0.66	0.83	0.75	0.76
HCB	ng/g ww	22	0.01	2.14	0.58	0.11
HCB	ng/g lw	3	49	60	53	50
PCB-28	ng/g ww	6	<0.005	<0.005	-	-
sum-TriCB	ng/g ww	6	0.01	0.02	0.01	0.01
PCB-47	ng/g ww	2	<0.01	<0.01	-	-
PCB-52	ng/g ww	16	0.003	0.02	0.01	0.01
PCB-66	ng/g ww	6	0.02	0.03	0.02	0.02
PCB-74	ng/g ww	6	0.02	0.02	0.02	0.02
sum-TetCB	ng/g ww	6	0.05	0.07	0.06	0.06
PCB-99	ng/g ww	6	0.05	0.06	0.06	0.06
PCB-101	ng/g ww	22	0.001	0.03	0.01	0.001
PCB-105	ng/g ww	6	0.05	0.06	0.05	0.06
PCB-118	ng/g ww	22	0.02	0.15	0.07	0.05
sum-PenCB	ng/g ww	6	0.24	0.29	0.27	0.28
PCB-128	ng/g ww	6	0.015	0.023	0.018	0.017
PCB-138	ng/g ww	22	0.02	0.18	0.08	0.06
PCB-153	ng/g ww	22	0.006	0.26	0.096	0.042
PCB-156	ng/g ww	6	0.015	0.024	0.018	0.015
sum-HexCB	ng/g ww	6	0.42	0.47	0.43	0.42
PCB-170	ng/g ww	6	0.021	0.043	0.030	0.026
PCB-180	ng/g ww	22	0.006	0.10	0.026	0.006
PCB-183	ng/g ww	2	<0.012	<0.012	-	-
PCB-187	ng/g ww	6	0.016	0.019	0.017	0.016
sum-HepCB	ng/g ww	6	0.11	0.17	0.14	0.13
PCB-194	ng/g ww	4	0.012	0.017	0.014	0.014
Sum7PCB	ng/g ww	6	0.58	0.68	0.62	0.60

Table A2. Cont.

Berries	Units	N	Min	Max	Average	Median
Sum7PCB	ng/g lw	3	16	30	24	25
sumPCB	ng/g ww	6	0.89	1.0	0.95	0.92
sumPCB	ng/g lw	3	23	46	36	38
a-HCH	ng/g ww	16	0.0035	0.0054	0.0036	0.0035
b-HCH	ng/g ww	16	<0.0052	<0.0052	-	-
g-HCH	ng/g ww	16	<0.0064	<0.0064	-	-
d-HCH	ng/g ww	16	0.010	0.015	0.010	0.010
2,4'-DDE	ng/g ww	16	<0.0064	<0.0064	-	-
4,4'-DDE	ng/g ww	16	0.007	0.043	0.019	0.017
2,4'-DDD	ng/g ww	16	<0.004	<0.004	-	-
4,4'-DDD	ng/g ww	16	<0.001	<0.001	-	-
2,4'-DDT	ng/g ww	16	0.0024	0.014	0.0045	0.0024
4,4'-DDT	ng/g ww	16	0.0026	0.040	0.011	0.0069

Appendix C

National activity limits for Cs-137 in edible products and drinking water in Norway, Finland, and Russia.

Table A3. National activity limits for Cs-137 in edible products and drinking water.

	Finland	Russia	Norway
Berries (wild)	600 Bq/kg f.w. ¹	160 Bq/kg f.w. ³	600 Bq/kg ⁵
Mushrooms	600 Bq/kg f.w. ¹	500 Bq/kg f.w. ³	600 Bq/kg ⁵
Reindeer, moose, and wild animals	600 Bq/kg f.w. ¹	320 Bq/kg f.w. ³	3000 Bq/kg ⁵
Livestock (pork, beef, mutton)	600 Bq/kg f.w. ¹	160 Bq/kg f.w. ³	600 Bq/kg ⁵
Poultry and wild birds	600 Bq/kg f.w. ¹	180 Bq/kg f.w. ³	600 Bq/kg ⁵
Fish	600 Bq/kg f.w. ¹	130 Bq/kg f.w. ³	600 Bq/kg ⁵
Milk	600 Bq/kg ¹	100 Bq/kg f.w. ³	370 Bq/kg ^{5,6}
Drinking water	50 Bq/kg ²	11 Bq/kg ⁴	No specific limit ⁷
Fruits	600 Bq/kg f.w. ¹	40 Bq/kg f.w. ³	600 Bq/kg ⁵
Vegetables	600 Bq/kg f.w. ¹	120 Bq/kg f.w. ³	600 Bq/kg ⁵

¹ The Finnish limit of 600 Bq/kg fresh weight (f.w.) for edible products is based on Regulations 733/2008 and 1048/2009 of the Council of the European Union and the recommendation 2003/274/Euratom of the Council of the European Union.

² When water does not contain other radionuclides. The total dose from communally produced drinking water cannot exceed 0.5 mSv/year. (STUK's Instruction ST-12.3).

³ Sanitary regulations and standards 2.3.2.1078-01.

⁴ Standards of radiation safety 99/2009.

⁵ Limits are applied both for fresh and dried products.

⁶ Includes also baby food.

⁷ Norway has no specific concentration limit of Cs-137 in drinking water but has a total dose limit of 0.1 mSv/year for all radionuclides, incl. Cs. For Cs-137, 0.1 mSv/year corresponds 10 Bq/L.

Source: [13,14].

Appendix D

Main highlights of statistical analyses by contaminants and food species.

Berries

- Different concentration trends for radioactivity and metals.
- Cloudberry clustered, other berries not. Cloudberry has higher radioactivity (Cs-137) and Cd contents.
- Co and Ni are correlated in berries (clustered).
- Cu and Co are correlated (clustered).
- Low radioactivity and lower Hg, but higher concentrations of Ni and Cu in berries.
- No clear trend for POPs concentration; low concentration.
- PCB congeners with five or more chlorides clustered, thus correlation of PCB in berries.

Mushrooms

- Mushrooms indicated the highest radioactivity and correlation with Cd and Co.
- Clustering of *Cortinarius* and *Leccinum* for metals and radioactivity.
- They have the highest radioactivity and As, Cu, and Ni contents.
- As, Cu, Ni, and radionuclides are in correlation (clustered).
- Low concentration of POPs.

Fish

- Radioactivity correlates with Cd and Ni.
- Dioxins were measured only in char and trout.
- Char and trout clustered together. They have the highest concentrations of PeCB and PCB congeners and also have the highest concentrations of Co, Cd, and Ni.
- Pike and white fish clustered together. They have an opposite correlation and lower concentrations.
- Perch overlapping between clusters.
- Overall, there is a correlation between PCB congeners, HCH compounds, and DDT compounds in fish samples.
- They have the highest concentration of PCB congeners and Hg.
- They have the highest concentration of POPs; however, there is variation in the composition of POPs in fish.
- They have the lowest concentration of radionuclides compared with other contaminants.

Game

- Dioxins are measured only in reindeer; there is no clear trend between dioxins and PCB.
- Variations in the composition of POPS, clustered in several groups and pollutants.
- Overall, they have the highest level of radioactivity and the highest levels of Cu.
- Clear clustering in radioactivity (K-40, Cs-137) and metals in reindeer, moose, and ptarmigan.
- Reindeer have higher levels of radioactivity (Cs-137) and Hg and lower concentrations of Co than moose and ptarmigan.
- Reindeer also have the highest concentration of PCB congeners with min 5 chlorides, a-HCH, and DDE/DDT compounds.
- Moose have a higher concentration of Pb and the lowest of Cu.
- K-40 and Cs-137 are clustered and correlated, whereas metals are not clustered.

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