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Radiological Hazards in Finnish Cereals: Comparison of Man-made and Natural Sources

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A survey on man-made (^{137}Cs and ^{90}Sr) and natural radionuclides (^{238}U, ^{232}Th, ^{228}Ra, ^{226}Ra) in cereal crops was conducted by collecting 66 cereal samples at 36 flour mills. This was the first time that natural radionuclides were included in such a survey in Finland. Based on the results new domestic reference concentrations for cereals were suggested: 2 $\mathrm{mBq \cdot kg^{-1}}$ for ^{238}U and ^{232}Th, 100 $\mathrm{mBq \cdot kg^{-1}}$ for ^{228}Ra and 200 $\mathrm{mBq \cdot kg^{-1}}$ for ^{226}Ra. The mean committed effective dose from ingestion of all radionuclides in cereal products was assessed as 30 μSv per year. Currently, the man-made radionuclides contribute only one percent to the total dose whereas in 1963 their proportion was about half and in 1987 about 20%, one year after the Chernobyl accident. Even so, the doses are very small and pose insignificant health risk to the consumers.

Keywords: cereals, radioactivity, dose, risk assessment

Introduction

Food production chains are susceptible to both microbiological and chemical hazards which can be attributable to anthropogenic or natural sources. Radiological hazards in food can originate from both sources. For example, anthropogenic contamination can take place, when radionuclides are discharged in a nuclear accident the most infamous being Harrisburg (1979), Chernobyl (1986) and Fukushima (2011). In a large-scale nuclear accident, several radioactive fission products are released and the radioactive plume may travel hundreds of kilometres from the point of origin, and deposit on agricultural soils. This happened in Europe in 1986 when several radionuclides were released during the Chernobyl nuclear accident. Most of these radionuclides had short half-lives (such as ^{131}I) and hence they disappeared in a few weeks and caused little exposure via ingestion of food. The fallout, however, contained radioactive strontium (^{90}Sr) and caesium (^{137}Cs) which have long half-lives, 29 and 30 years, respectively, and these radioactive isotopes are still present in food chains.

There are also traces in the environment from global fallout after nuclear weapons testing carried out in the 1950s and 1960s. Most of the ^{90}Sr in the Finnish environment origi-

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nates from such weapons tests while ^{137}Cs deposition after the Chernobyl accident was notably higher in many parts of the country than the deposition in the 1960s (Fig. S1*) (STUK 1987; Arvela et al. 1990; Paatero et al. 2010).

In addition to anthropogenic radioactivity, there is also naturally occurring radioactivity that has been present in terrestrial matter since the formation of the Earth 4.6 billion years ago. Uranium and thorium are widely distributed in the Earth's crust and can be found in all rocks, soils and natural water bodies. As these elements decay they form chains of radioactive elements known as the uranium, actinium and thorium decay series totalling more than 30 different radionuclides (Fig. S2). As the first radionuclides in the series have extremely long half-lives their concentrations can be assumed constant and hence they are able to continuously produce succeeding radionuclides in the environment. Many of these so called daughter nuclides, however, are short-lived and are of little radiological interest. Considering the radiotoxicity, the most important natural radionuclides in foods are generally isotopes of radium (^{228}Ra and ^{226}Ra), lead (^{210}Pb) and polonium (^{210}Po) (UNSCEAR 2000).

The Radioactivity Environmental Monitoring (REM) project was initiated in Europe in 1986 in order to help improve exchange of information between Member States. In 1987, maximum permitted levels (750 Bq·kg^{-1} for ^{90}Sr and 1250·Bq kg^{-1} for ^{137}Cs+^{134}Cs) were laid down in the event of a future radiological accident (3954/87/EURATOM). In 1990, Council Regulation EEC 737/90 set the maximum level for ^{137}Cs+^{134}Cs in most imported foodstuffs at 600 Bq·kg^{-1}. Natural radioactivity, however, has been omitted from environmental monitoring programs and from legislation concerning the maximum permitted levels. Therefore, data on natural radioactivity in foodstuffs are generally sparse compared to data on man-made radionuclides.

Cereal products form a substantial part of the Finnish diet. Domestic cereal products have therefore been studied as a part of surveying radioactivity in the environment ever since the 1960s (Rajama and Rantavaara 1982; Rantavaara and Haukka 1987; Kostiainen and Rantavaara 2002). In light of recent events in Japan, radioactivity in food products has raised a lot of public interest. Also, aspirations toward commissioning uranium mines in Finland have been actively debated especially among the public living in areas where prospecting reservation permits have been filed. Elevated levels of natural radioactivity in food crops have been observed near certain mining areas (e.g. Louw et al. 2009). This has raised long-neglected questions about baseline concentrations of natural radionuclides in foodstuffs produced in Finland. Thus far natural radioactivity has been investigated only in wild gathered food and in game but not in agricultural produce (Kauranen and Miettinen 1968; Vaaramaa et al. 2009). Dose assessments concerning natural radioactivity in food have therefore been based upon reference values reported by the United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR (2000). These values are calculated on the basis of studies available from four European, three Asian and one North-American countries where different geological and climatic conditions prevail,

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*Further details about the Electronic Supplementary Material (ESM) can be found at the end of the article.

and may hence be misleading in our case. Furthermore, the reported concentrations have ranged greatly.

The main aim of this study was to embark on screening naturally occurring radioactivity in domestically produced foodstuffs. These data will be of use for radiation exposure assessments domestically, for environmental impact assessments near industrial establishments exploiting materials containing natural radionuclides, and for assessing radioactivity levels in European foodstuffs in general. Another aim of the study was to complement data on man-made radionuclides in cereals and to compare the associated risks to those arising from naturally occurring radioactivity.

Materials and Methods

Sampling

Sample collection was organized by the Finnish Food Safety Authority (Evira) and carried out by municipal health officers between September and December, 2007. Sampling was carried out jointly with a project investigating the adult intake of *Fusarium* toxins from cereals and cereal-based products (Rautala et al. 2008). Samples collected at 36 flour mills in 34 municipalities consisted of both flour and unprocessed samples of wheat (15), rye (33), oats (11) and barley (7). Sampling covered the most important cereal production areas in Finland and different soil types of fields under cultivation (Fig. S3). Each sample was a pooled sample (5 kg) created by combining 50–100 gram samples taken by a bore or a scoop from the load. All cereals selected had been grown in Finland. The crops had been harvested between 2004 and 2007. The ages of the cereals were 185–1350 days at the time of the analyses.

Analytical methods

[Read in the online Supplement]

Quality control

[Read in the online Supplement]

Risk assessment

Assessment of dietary intake in this study is based on the National FINDIET 2007 Survey (Korkalo et al. 2008). The major ingredients in foods consumed by adult Finns are vegetables (140 $g \cdot d^{-1}$), fruit and berries (220 $g \cdot d^{-1}$), potato (90 $g \cdot d^{-1}$), cereals (160 $g \cdot d^{-1}$), meat (130 $g \cdot d^{-1}$) and milk (460 $g \cdot d^{-1}$). Of all cereals, wheat, rye, oats and barley cover 90% of consumption. Consumption of rice, maize and other bread grain is low, about 10% (Table S1).

Health detriment caused by ionizing radiation is based on energy imparted to a tissue. This energy can induce chemical reactions in the tissues, such as DNA breakage or formation of free radicals that can further damage DNA. In our assessment and comparison of anthropogenic and natural radioactivity in cereals, we express the radiation exposure as the committed effective dose, the unit of which is the sievert (Sv). The committed effective dose has been calculated from the radionuclide concentrations in cereals, the consumption rates of cereals and the dose coefficients ($Sv \cdot Bq^{-1}$) for internal exposure recommended by International Commission on Radiological Protection (ICRP 1996).

Results

Effect of milling on activity concentrations

There were two sample pairs of wheat and four pairs of rye, of which both the flour and grains originated from the same farms. The mean (and range) ratio of ^{137}Cs concentration in flour samples to that in grain samples was 0.59 (0.46–0.72) for wheat, and 0.54 (0.25–0.85) for rye. These are quite consistent with the corresponding values reported earlier (Bunzl and Kracke 1987; Rantavaara and Haukka 1987). Variation in the ratio is partially due to the amount of bran in the flour, the composition of the flours analysed varied from white flour to whole grain flour. The ^{90}Sr concentrations were determined in two sample pairs of grains and flour, one for wheat and one for rye. The ratios of ^{90}Sr concentrations in flour samples to those in grain samples were 0.49 ± 0.03 and 0.90 ± 0.04 for wheat and rye, respectively.

There was a sample pair of each of wheat and rye in which uranium and thorium concentrations were determined in both flour and unprocessed grain. The ratio of ^{238}U concentration in wheat flour to that in grain was 0.17 ± 0.01. For rye the ratio was 0.53 ± 0.02. The flour-to-grain ^{232}Th concentration ratio was 0.24 ± 0.01 in wheat and 0.57 ± 0.02 in rye.

^{228}Ra concentrations in two sample pairs of both unprocessed wheat grain and flour indicated a mean (and range) flour-to-grain concentration ratio of 0.45 (0.42–0.49). The mean (and range) flour-to-grain concentration ratios from four sample pairs of rye was 0.58 (0.51–0.79). We assume that the two isotopes of radium have similar translocation rates in plants, and the same flour-to-grain concentration factors were applied to isotope ^{226}Ra.

Activity concentrations

^{137}Cs

The arithmetic mean concentrations (and range) of ^{137}Cs in barley, oats, rye and wheat samples (representing both grain and flour) were 0.21 (0.027–0.72), 0.56 (0.27–1.0), 0.42 (0.027–1.4) and 0.14 (0.024–0.37) $Bq \cdot kg^{-1}$, respectively.

The highest ^{137}Cs concentrations in cereals were found in areas where ^{137}Cs deposition was the highest after the Chernobyl accident. In the sampling areas, the mean ^{137}Cs depositions were 16, 18, 20 and 25 $kBq \cdot m^{-2}$ for oats, wheat, rye, and barley samples, respectively, ranging from 1.7 to 62 $kBq \cdot m^{-2}$ (1 Oct. 1987). The correlation of ^{137}Cs concentration with the deposition level in the sampling area was most evident in wheat samples, which reflects the similarity of growing conditions of wheat. The correlation was not so clear with other cereals which are often grown on fields with more variable soil types (Fig. S4).

The ^{137}Cs concentrations decreased in the order: oats > rye > barley >wheat. The higher concentrations and wider range of ^{137}Cs in oats and rye samples reflect variation in soil conditions. Oats and rye are not as demanding in regard to soil type and soil conditions (fertility, moisture and pH), whereas wheat and barley are cultivated on fields with the best conditions. The areas under wheat cultivation in Finland are soils with higher clay content, which is known to reduce uptake of ^{137}Cs.

^{90}Sr

The mean ^{90}Sr concentrations (and range) in wheat, rye, oats and barley samples were 0.12 (0.081–0.17), 0.20 (0.11–0.32), 0.33 (0.22–0.60) and 0.27 (0.18–0.36) Bq·kg^{-1}, respectively. The ^{90}Sr concentrations were found to be slightly higher in the cereal samples from areas with the highest ^{90}Sr deposition in 1963–1985 and in 1986 even though the areal variation is small (Paatero et al. 2010).

^{238}U

The mean activity concentration of ^{238}U in all 30 samples was very low, only 2.2 mBq·kg^{-1}. By using the results of 11 flour samples and 2 unprocessed samples corrected to represent flour (by a factor 0.17) the mean activity concentration (and range) in wheat flour was 1.2 (0.17–3.5) mBq·kg^{-1}. In the 8 samples of rye flour, the mean activity concentration (and range) was 3.5 (0.48–8.8) mBq·kg^{-1}.

Oats is mostly consumed as whole-grain products such as porridge and thus the unprocessed samples represent the general concentrations in consumer products. The mean activity concentration (and range) in the three unprocessed oats samples was 2.9 (2.0–4.7) mBq·kg^{-1}. In the case of barley, bran is normally removed in processing of flour. The hulled grain and flour sample both contained 0.31 ± 0.02 mBq·kg^{-1} of ^{238}U whereas two separate unprocessed samples contained 2.9 ± 0.1 and 3.3 ± 0.1 mBq·kg^{-1} of ^{238}U.

^{232}Th

The mean activity concentration of ^{232}Th in all 30 samples was only 2.1 mBq·kg^{-1}, which is similar to that of ^{238}U. By using the flour-to-grain concentration ratios, the mean activity concentrations (and range) in wheat and rye flour were assessed as 0.78 (0.14–1.5) and 4.2 (0.46–13) mBq·kg^{-1}, respectively. The mean activity concentration (and range) in the three unprocessed oats samples was 2.3 (1.3–3.9) mBq·kg^{-1}. The hulled grain and the flour samples of barley contained 0.20 ± 0.01 and 0.24 ± 0.01 mBq·kg^{-1}, respectively. The separate unprocessed barley samples contained 1.5 ± 0.0 and 1.9 ± 0.1 mBq·kg^{-1} of ^{232}Th.

^{228}Ra

By using the results of 11 wheat flour samples and 2 unprocessed wheat samples corrected to represent flour the mean activity concentration (and range) in wheat flour was assessed as 110 (49–250) mBq·kg^{-1}. Activity concentrations in all rye samples (N=33) were 63–580 mBq·kg^{-1}. By using the results of 16 flour samples and 13 unprocessed samples corrected to represent flour the mean activity concentration (and range) in rye flour was assessed as 170 (63–360) mBq·kg^{-1}.

The mean activity concentration (and range) in 10 unprocessed oats samples was 170 (67–280) mBq·kg^{-1}. The mean concentration (and range) in four unprocessed barley samples was 310 (240–410) mBq·kg^{-1}. Hulled barley grain, barley malt and barley flour samples contained 88 ± 43, 360 ± 60 and 330 ± 30 mBq·kg^{-1} of ^{228}Ra, respectively.

^{226}Ra

By using the results of 11 flour samples and 2 unprocessed samples corrected to represent flour the mean activity concentration (and range) in wheat flour was 180 (68–400) mBq·kg^{-1}. The mean activity concentration (and range) in 8 rye flour samples was 270 (170–410) mBq·kg^{-1}. The highest concentration was in an unprocessed barley sample which was 710 ± 100 mBq·kg^{-1}. The mean activity concentrations of 3 unprocessed oats and 2 unprocessed barley samples were 210 and 530 mBq·kg^{-1}, respectively. The hulled barley grain and barley flour samples contained 300 ± 100 and 200 ± 50 mBq·kg^{-1} of ^{226}Ra, respectively.

^{210}Pb and ^{210}Po

Since most of the samples were older than 185 days, ^{210}Pb and ^{210}Po concentrations were observed to be in secular equilibrium due to radioactive decay, i.e. their activity concentrations were the same. The mean ^{210}Pb concentration (and range) in wheat flour was 120 (58–300) mBq·kg^{-1}. In rye flour, the mean concentration (and range) was 290 (230–380) mBq·kg^{-1}. In oats and barley samples the mean concentrations (and range) were 360 (110–520) and 360 (80–560) mBq·kg^{-1}, respectively (Turtiainen et al. 2011).

A summary of the data is presented in Table S2.

Quality control

[Read in the online supplement]

Intake assessment

The ^{137}Cs concentration remaining in pasta after boiling is 10–40% of the original but no data has been reported for natural radionuclides (IAEA 2010). Consumption of pasta, however, covers only 5–6% of the total consumption of cereal among Finns while most consumption is bakery products and porridges (Korkalo et al. 2008). Considering the preparation technique of bakery products and porridge where little loss of radionuclides occurs, the influence of food preparation on radionuclide concentration was not taken into consideration in the present work (Table S3).

Natural uranium is composed of three isotopes (^{238}U, ^{234}U and ^{238}U). The ^{238}U/^{235}U activity ratio is 21.4 in natural uranium. ^{238}U and ^{234}U belong to the same decay series and hence their activity concentrations in soils are generally similar. According to Pietrzak-

Flis et al. (1997), the ^{234}U/^{238}U activity concentration ratio in flour was 1.12 ± 0.12. In our work, an activity ratio of 1 was used. The values for intake of ^{210}Pb and ^{210}Pb were taken from Turtiainen et al. (2011).

Risk assessment

The mean annual effective dose from all sources (e.g. radon in indoor air, external radiation from soils and building materials, cosmic radiation and X-ray diagnostics) among Finns has been assessed as 3.7 mSv·a^{-1} (Muikku et al. 2005). Compared to this value the committed effective dose from ingestion of radionuclides in cereals was very small, 32 µSv·a^{-1} among men and 24 µSv·a^{-1} among women, or 28 µSv·a^{-1} averaged among the adult population (years 25–75). The highest contribution to the dose was from ^{210}Po and ^{210}Pb which accounted for 70% of the dose received from all radionuclides (Fig. 1). The isotopes of radium (^{228}Ra and ^{226}Ra) contributed about 29% to the dose whereas the actinoid elements (uranium and thorium) made insignificant contributions (only 0.1%).

The committed effective doses from ingestion of ^{90}Sr and ^{137}Cs in cereals were 0.27 and 0.19 µSv·a^{-1} among men and 0.21 and 0.15 µSv·a^{-1} among women, respectively. In other words, man-made radionuclides accounted for only one percent of the dose in 2007. The highest annual dose from ^{137}Cs in cereals was received in 1987 and amounted to 4.4 µSv·a^{-1}. In the same year, the dose from the short-lived radionuclide ^{134}Cs gave an addition of 2.3 µSv·a^{-1} to the dose (Rantavaara 1991). After the rapid decrease in the end of 1980s, the dose from ^{137}Cs has declined slowly. The dose due to ^{90}Sr was highest in 1963, 26 µSv·a^{-1}. In 1987–88, the dose from ^{90}Sr in cereals was 0.2 µSv·a^{-1} and according to the present work it has remained at the same level (Rajama and Rantavaara 1982; Rantavaara 1991).

It is obvious that the committed effective doses from ingestion of both man-made and natural radionuclides in cereals are very low. The ICRP proposes a nominal probability

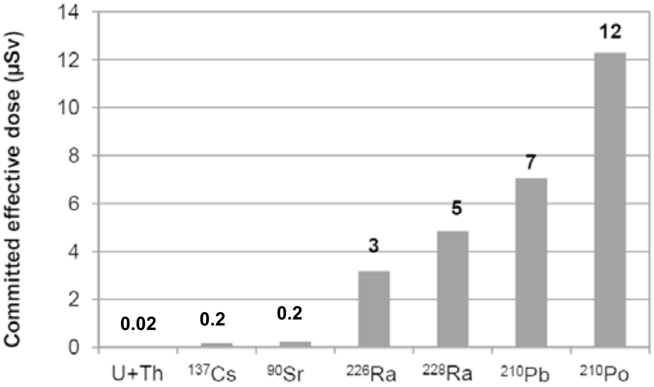

Figure 1. The contribution of different radionuclides to the committed effective doses caused by ingestion of cereal products among Finns (ages 24–75)

coefficient for detriment-adjusted cancer risk of 0.055 $\mathrm{Sv{\cdot}Bq^{-1}}$. This coefficient, however, should not be used for assessing projected cancer cases when the doses are very low or to a large number of people. We can only conclude that the radiation induced health detriment caused by radionuclides in cereals is most probably negligible.

Discussion

The number of cereal samples collected for this survey was 66. This covered only a fraction of Finnish farms which grow cereals (28,000 in 2007) even though one third of the samples were mixed samples representing cereal crops from several farms. Due to analytical costs, the number of samples in which ^{238}U, ^{232}Th, ^{226}Ra and ^{90}Sr were determined was even lower. The areal representativeness of the samples, on the other hand, was good. Furthermore, the measured concentrations of natural radionuclides ranged only by one order of magnitude, at maximum, and no anomalously high or small concentrations were observed which implies that the nominal concentration range could be established.

Activity concentrations of long-lived natural actinoids, ^{230}Th, ^{228}Th and ^{227}Ac were not determined in the present work. The low concentrations determined for actinoids ^{238}U and ^{232}Th suggest that these radionuclides are also found only in very low concentrations and that their contribution to the total effective dose is small. Children were excluded from the risk assessment since there was no data available on the consumption of cereals among Finnish children.

This was the first survey on radionuclides, including natural radionuclides, in cereal crops carried out in Finland and also the first in which natural radionuclides were surveyed in agricultural produce instead of wild gathered food or game. Until now, assessment of exposure to natural radionuclides in food products has mainly been based upon the UNSCEAR reference values. In grain products these are 20, 3, 60, 80, 50 and 60 $\mathrm{mBq \cdot kg^{-1}}$ for ^{238}U, ^{232}Th, ^{228}Ra, ^{226}Ra, ^{210}Pb and ^{210}Po, respectively (UNSCEAR 2000). In our survey, consistently lower ^{238}U concentrations were found, and hence the reference value based on consumption of different cereal species in Finland should be 2 $\mathrm{mBq{\cdot}kg^{-1}}$ for both ^{238}U and ^{232}Th. Furthermore, we suggest 100 $\mathrm{mBq{\cdot}kg^{-1}}$ for ^{228}Ra and 200 $\mathrm{mBq{\cdot}kg^{-1}}$ for ^{226}Ra, ^{210}Pb and ^{210}Po as reference values in Finland. The latter, by contrast, are about double those suggested by UNSCEAR.

We can assume that the concentrations of natural radionuclides remain rather constant in cultivated fields (Bolca et al. 2007). In 1963, the dose from man-made radionuclides was the same magnitude as that from natural radionuclides. The contribution from man-made radionuclides to the total dose from ingestion of cereals has declined, and even after Chernobyl fall-out, was only about 20% of the total. Based on this survey, the proportion today is only one percent.

Normally, radiological surveying of food is carried out by total diet studies and concentrate only on man-made radioactivity. More attention should be paid to natural radioactivity in agricultural produce since currently natural radionuclides account for much of the effective dose that is received from ingestion of cereal products. By surveying different food groups instead of the total diet, more accurate dose assessments at the population level can be made even if the consumption rates of certain products change over the years. Furthermore, industrial activities, such as extracting or processing of minerals, may lead to release of natural radionuclides into agricultural soils and thereby contamine the food chains. Hence, it is important that baseline concentrations of these radionuclides are established.

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Electronic Supplementary Material (ESM)

Electronic Supplementary Material (ESM) associated with this article can be found at the website of CRC at http://www.akademiai.com/content/120427/

Electronic Supplementary *Figure S1.* Caesium (^{137}Cs) deposition in Finland after the Chernobyl accident in 1986

Electronic Supplementary *Figure S2*. Natural decay series. Branching (in %) and decay mode (α, β or IT) are given next to the arrows and the respective half-lives are given in parenthesis

Electronic Supplementary *Figure S3*. Locations of flour mills (numbers) and prevailing soil types of cultivated fields in Finland

Electronic Supplementary *Analytical methods*

Electronic Supplementary *Quality control*

Electronic Supplementary *Risk assessment*

Electronic Supplementary *Table S1*. Consumption of cereals among Finns (aged 24–75 years) presented as kg per year

Electronic Supplementary *Figure S4*. ^{137}Cs concentration in rye and wheat (Bq·kg^{-1} d.w.) samples versus deposition (kBq·m^{-2})

Electronic Supplementary *Table S2*. Summary of the activity concentrations (mBq·kg^{-1} in dry weight) determined in selected cereal samples

Electronic Supplementary *Quality control*

Electronic Supplementary *Figure S5.* Arithmetic mean concentrations of ^{228}Ra in cereal products representing selected samples (n = 30) and all samples (n = 66). The numbers in parentheses indicate the numbers of individual samples in the selected group and in all samples

Electronic Supplementary *Figure S6*. Correlation of potassium concentration in cereal products obtained by ICP-SFMS and gamma spectrometry

Electronic Supplementary *Table S3*. Mean intake of man-made and natural radionuclides (in Bq·a^{-1}) via ingestion of cereal products by the Finnish adult population (ages 24–75)