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Traces of Fukushima fallout in the environment of Northwest Germany

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Abstract. Traces of short- and long-lived fallout isotopes (^{131}I, ^{134}Cs and ^{137}Cs) were found in environmental samples obtained in northwest Germany (river sediment, rainwater, grass and milk) from March to May 2011, following the radioactivity releases after the nuclear accident in Fukushima, Japan. The found concentration values are consistent with reported concentrations in air, amount of rainfall and expected values applying simple radioecological models. The [^{134}Cs]/[^{137}Cs] ratio reported for air (about 1:1) allows for discrimination between "recent" and "old" ^{137}Cs. Expected ^{136}Cs values fell below the detection limits of the instrumentation, despite large sample masses and long counting times.

1. INTRODUCTION

Following the continuing air releases of radionuclides after the accident in the Fukushima Daichi nuclear power plants, Japan, starting on March 12, 2011, traces of short-lived fission products were recorded in the air by a number of CTBTO radionuclide monitoring stations. First they were detected on March 12 in the Takasaki monitoring station in Japan (220 km away from Fukushima Daichi NPP), followed by eastern Russia on March 14 and US west coast two days later [1]. The dispersion of the radioactivity was detected at Iceland on March 20 [2] and a first German station reported ^{131}I on March 21 [3].

Within an ad hoc monitoring program in the Radioactivity measurements laboratory at the University of Bremen, samples of rain water, sediment, grass and fresh cow milk were taken and analyzed for traces of short-lived isotopes indicating Fukushima fallout.

2. MATERIALS AND METHODS

2.1 Sampling

Samples of rain water were taken in the city of Bremen and in the village of Seefeld (Stadland) in the federal state of Lower Saxony by collecting water during indicated time period. Samples of sediment were collected by sediments traps in small streams situated in the cities of Bremen and Bremerhaven (Kleine Wümme and Markfleth). Grass samples were taken by lawn mowing at the University of Bremen campus and in private gardens in villages of Seefeld and Schiffdorf in Lower Saxony. Cow milk was obtained from a farmer in Seefeld, Lower Saxony. Sample masses varied between 0.5 and 1.5 kg for sediment, 2 to 2.6 kg for rain water, 0.42 to 0.85 kg for grass and 1.8 kg for milk. Locations of sampling points are shown in Figure 1. The timeline of sampling, together with the period of the highest reported Fukushima-related radionuclide air concentration in Germany, is shown in Figure 2.

2.2 Gamma spectroscopy

The samples were placed in Marinelli beakers and measured in fresh state by low-level low-background gamma spectroscopy using standard coaxial HPGe detectors (Canberra Industries, Meriden, CA)

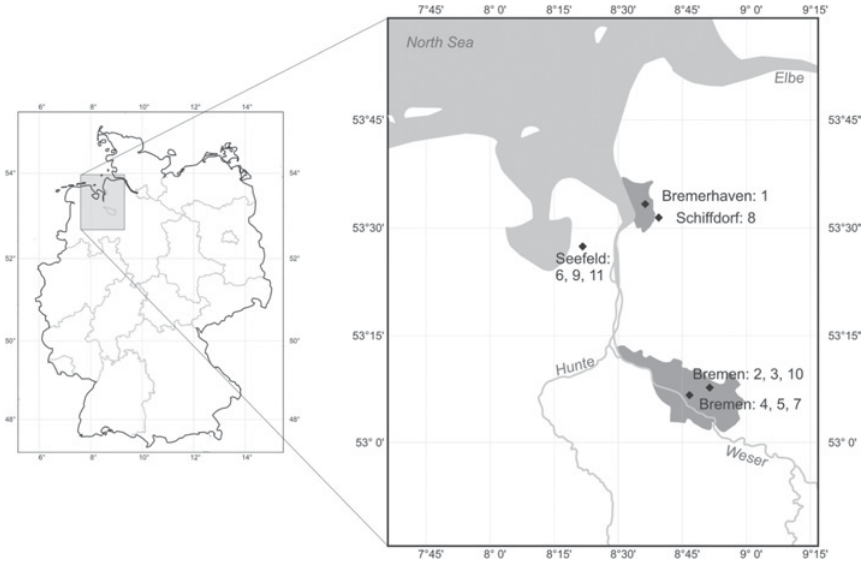

Figure 1. Location of the sampling points. For each location, sample numbers are indicated according to the Table 1.

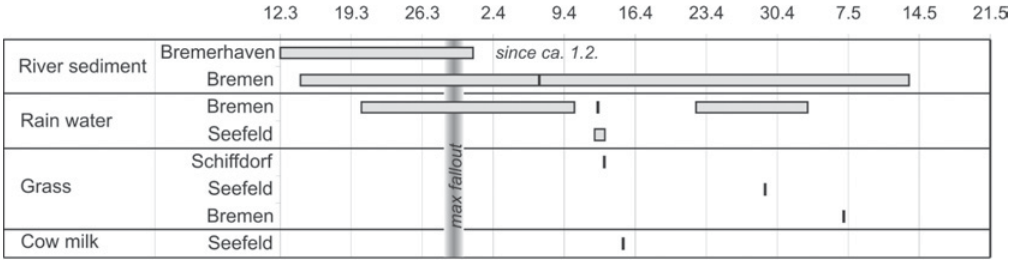

Figure 2. Timeline of the sampling campaign. Maximal concentration in the air, marked as "max fallout" in the figure, was detected (e.g. [5]) between 28.-29.3.2011.

of about 50% relative efficiency housed in 10 cm Pb shieldings with Cu, Cd and plastic lining. Measurement times ranged between about 85,000 and 335,000 seconds. The spectra were analyzed using the Genie 2000 software (Canberra Industries). Efficiency calibration was performed using either data obtained with calibration solutions in suitable geometries or the Monte Carlo based LabSOCS calibration tool (Canberra Industries) [4], which takes into account not only sample to detector geometry, but also sample density and composition, as well as measurement container properties. The precision of the obtained results is verified regularly e.g. by participation in laboratory intercomparison tests.

3. RESULTS AND DISCUSSION

The obtained experimental values are listed in Table 1. The reference date for ^{131}I was either set to the date of sampling (for sediment, grass and milk) or to the date of maximum rainfall (for rainwater).

Table 1. Measured values in Bq/kg (fresh mass).

No.	Type of sample	Date	^{131}I	^{137}Cs	^{134}Cs
1	River sediment (Bremerhaven)	ca. 1.2. – 31.3.	0.113 ± 0.046	2.67 ± 0.07	< 0.084
2	River sediment (Bremen)	14.3. −6.4.	0.45 ± 0.06	1.76 ± 0.04	< 0.06
3	River sediment (Bremen)	6.4. – 13.5.	0.031 ± 0.03	1.73 ± 0.05	< 0.05
4	Rain water (Bremen)	20.3. −10.4.	0.43 ± 0.03	< 0.04	< 0.04
5	Rain water (Bremen)	12.4.	0.10 ± 0.02	< 0.04	<0.04
6	Rain water (Seefeld)	13.4.	0.14 ± 0.02	< 0.03	< 0.03
7*	Rain water (Bremen)	22.4. – 3.5.	0.031 ± 0.008	< 0.02	< 0.02
8	Grass (Schiffdorf)	13.4.	3.58 ± 0.13	1.59 ± 0.07	0.32 ± 0.03
9	Grass (Seefeld)	28.4.	0.31 ± 0.04	0.26 ± 0.04	0.062 ± 0.014
10	Grass (Bremen)	6.5.	0.12 ± 0.03	0.18 ± 0.03	< 0.08
11	Milk (Seefeld)	14.4.	0.08 ± 0.02	< 0.02	< 0.03

*Mixed with old rain water.

3.1 Comparison with concentrations in air

3.1.1 Reported concentrations and isotope ratios in air

After the arrival of the radioactive plume in Germany, several organizations have reported isotope concentrations in air on their internet websites [3, 5–7]. All stations reported values for ^{131}I and ^{137}Cs, and the data are quite consistent among each other, showing a maximum around March 29. Data on other isotopes are more scarce, the information used here was obtained from the Website of the German national metrology institute, PTB [5], which reports data also on ^{134}Cs and ^{136}Cs.

3.1.2 Deposition with rain

Wet deposition by rain is known to be the most effective transfer path for airborne radioisotopes to ground and water bodies. It can be estimated by applying simple environmental models, e.g. those applied for the prediction of environmental contamination after accidental releases of radioisotopes. Here, the German model system documented in [8] has been used. It contains a "washout coefficient" $\Lambda_0 = 7 \cdot 10^{-5}\,\mathrm{s}^{-1}$, indicating the fraction of aerosols washed out from $1\,\mathrm{m}^3$ of air per second, at a standard rain intensity of 1 mm/h. Deposition can then be calculated using

$$D = C_{air}\,\Lambda_0\,t_{rain}\,h \tag{1}$$

with D: deposition in Bq/m^2, C_{air}: isotope concentration in air, Λ_0: standard washout coefficient, t_{rain}: rainfall duration and h: height of atmospheric mixing layer. For the period between March 21 (first detection of the Fukushima plume in Germany) and April 6 (collection of a two-week sediment sample), 8.5 mm of rainfall were reported for Bremen by the German meteorological service, DWD [9]. A rough estimate of the expected deposition can then be obtained assuming e.g. a value for h of 1000 m, a rain intensity of 1 mm/h, a mean C_{air} of 1 mBq/m^3 for ^{131}I and 0.1 mBq/m^3 for ^{137}Cs. These input values would yield a surface deposition of 2.14 Bq/m^2 for ^{131}I and of 0.214 Bq/m^2 for ^{137}Cs and concentrations in rain water of 0.252 and 0.0252 Bq/l, respectively.

3.2 Comparison of measured and derived expected values

3.2.1 Rain water

As can be seen from Table 1, estimated and measured values coincide in a satisfactory manner, given the large uncertainties in the chosen parameters. Logically, values for ^{131}I decrease with time in correspondence with the published air concentration values.

3.2.2 River sediment

The employed sediment traps have a cross section of 0.16 m^2. The mass of sample 2 was 0.520 kg. Assuming 100% transfer of washed out activity into the sediment, these values together result in a ^{131}I deposition of 1.46 Bq/m^2, close to the value calculated in the previous section. Concentration in sample 3, collected at the same site as sample 2, is about a factor of 10 lower. The reported air concentration had decreased by about the same factor between the two sampling periods. Sample 1 has about 25% of the concentration found in sample 2, and it had been collected over a period about 3 times longer, starting long before the emissions from Fukushima. A lower ^{131}I concentration is thus compatible with the other data.

^{137}Cs concentrations found in sediment are much higher than those for ^{131}I, and similar to those found in samples collected in earlier times. Together with the fact that no ^{134}Cs could be detected, it can be concluded that ^{137}Cs must originate from atmospheric bomb test and Chernobyl contributions (which in northern Germany have about the same magnitude).

3.2.3 Grass

Measured concentrations in grass vary considerably. One reason might be the relatively early time of the year: in the investigated region grass had just begun to grow, and thus the collectable mass per area varied considerably. Nevertheless, the change in activity ratio between ^{131}I and ^{137}Cs with time of sampling is clearly visible – it can be attributed to the radioactive decay of ^{131}I. The Cs isotope ratio will be discussed below.

3.2.4 Milk

Although at first sight it was surprising to detect ^{131}I in milk, application of emergency models like SBG §49 StrlSchV [10] with the concentration found in grass would lead to similar values. The concentration in milk is obtained using the following equation:

$$C_{milk} = C_{grass} \dot{M}_{grass} T_{milk} \tag{2}$$

with C_{milk}: activity concentration in milk, C_{grass}: activity concentration in grass, $\dot{M}_{grass}$: daily grass consumption rate of cattle (65 kg/d) and T_{milk}: element-specific transfer factor grass-milk (0.003 for iodine). Using the indicated values, the ^{131}I concentration of sample 9 (0.31 Bq/kg) decay corrected to the time of milk sampling (1.04 Bq/kg) yields a concentration in milk of 0.20 Bq/kg, somewhat higher than measured. Given the small amount of grass available on the meadows, it can be assumed that the cattle was fed only partially from fresh grass, consistent with a lower ^{131}I concentration in milk.

3.2.5 Isotope ratios

Activity ratios can reveal additional information. In northern Germany, a residual deposition of about 2 kBq/m^2 ^{137}Cs remains from bomb test fallout and Chernobyl. It is thus not surprising that the ratio [^{131}I]/[^{137}Cs] can differ from the atmospheric data, depending on the type of sample. At the time of maximal concentration (end of March), this ratio was about 10. For the sediment samples the maximum

value is 0.256, indicating a strong contribution from "old" ^{137}Cs. In fact, ^{137}Cs was detected at these locations earlier and in similar concentrations.

In two grass samples, ^{134}Cs could be detected together with ^{137}Cs and ^{131}I. This offers the possibility to discriminate between "recent" and "old" Cs isotopes, using the [^{134}Cs]/[^{137}Cs] ratio from atmospheric measurements. This value is close to 1 in most published data. Assuming this value, it can be concluded that the main ^{137}Cs contribution in the grass samples is "old".

In order to investigate the presence of ^{136}Cs (found in air at the end of March at concentrations about 1 order of magnitude lower than for ^{134}Cs and ^{137}Cs [5], sample 8 was ashed (to increase detector efficiency) and remeasured. Even so, no ^{136}Cs could be detected due to the late measurement date. If the sampling and measurement had been performed at the beginning of April, probably a ^{136}Cs signal would have been detectable.

4. CONCLUSIONS

Despite the large distance between source and deposition area and the usage of standard equipment, it was possible to detect traces of the emissions from Fukushima in northern Germany in various environmental media. Values were plausible when compared to reported air concentrations and predictions from simple radioecological models. Isotope ratios could be used to discriminate between "recent" and "old" deposition.

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