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Short communication

Fukushima fallout in Northwest German environmental media☆

Daniela Pittauerová*, Bernd Hettwig, Helmut W. Fischer

University of Bremen, Institute of Environmental Physics, Otto-Hahn-Allee 1, D-28359 Bremen, 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 collected in Northwest Germany (rain water, river sediment, soil, grass and cow milk) from March to May 2011, following the radioactivity releases after the nuclear accident in Fukushima, Japan. The measured concentrations 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) 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 (NPP) in Japan, starting on March 12, 2011, traces of short-lived fission products were recorded in the air by a number of the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) Preparatory Commission radionuclide monitoring stations. First they were detected on March 12 in the Takasaki monitoring station in Japan (220 km away from the Fukushima Daichi NPP), followed by eastern Russia on March 14 and US west coast two days later (CTBTO, 2011). The dispersion of the radioactivity was detected at Iceland on March 19–20 (IRSA, 2011) and a first German station reported ^{131}I in an air sample collected from March 21–23 (DWD, 2011a).

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

2. Experimental

2.1. Sampling

Rain water, sediment, soil, grass and milk were collected as standard environmental media for radioactive emissions monitoring. These are also media in which radionuclide concentrations are likely to be observed soon after the deposition and where the radionuclides enter the food chain. Locations of sampling points are indicated in Fig. 1. The timeline of sampling is shown in Fig. 2. Samples 1, 2 and 4 were taken as a part of routine environmental radioactivity monitoring scheme in the federal state of Bremen (comprising the cities of Bremen and Bremerhaven). The rest of the samples were collected by matter of general interest at suitable locations easily accessible to the authors in states Bremen and Lower Saxony after it became obvious from the first sediment data that radioactive substances with presumable Fukushima origin might appear in other environmental media in detectable amounts.

Samples of rain water were taken in the city of Bremen and in the village Seefeld (Stadland, Lower Saxony) by collecting water during the indicated time period (Table 1). River sediment samples were collected by sediment traps in minor streams situated in the cities of Bremen and Bremerhaven (Kleine Wümme and Markfleth). The soil sample was taken from the upper approx. 3 cm of an agricultural field in Bremen – Arsten (Riederdamm street). The field was cropped with young corn plants at the time of the sampling. The soil was sieved and only the fraction <2 mm was

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* Corresponding author. Tel.: +49 421 21862763.
E-mail address: pittauerova@iup.physik.uni-bremen.de (D. Pittauerová).

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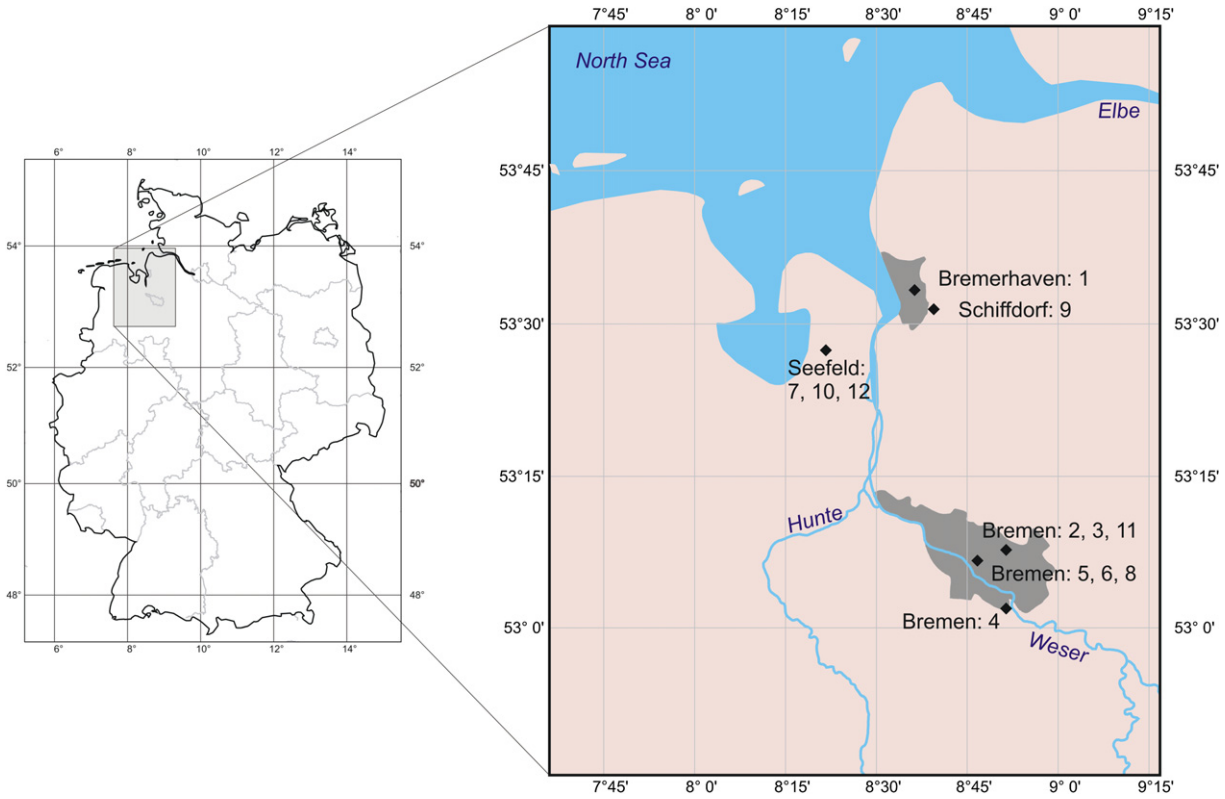

Fig. 1. Location of the sampling points. For each location, sample numbers are indicated according to Table 1.

used for the analysis. Grass samples were taken by lawn mowing at the University of Bremen campus and in private gardens in the villages Seefeld and Schiffdorf, Lower Saxony. Cow milk was obtained from a farmer in Seefeld. Sample masses were between 0.5 and 1.5 kg for sediment, 2.5 kg for soil, 2 and 2.6 kg for rain water, 0.42 and 0.85 kg for grass and 1.8 kg for milk.

2.2. Radiometric analysis

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) of about 50% relative efficiency housed in 10 cm Pb shielding with, depending on each of 4 detectors used, Cu or Cu, Cd and plastic linings. Measurement times ranged between 85,000 and 335,000 s. 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) (Bronson, 2003), 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

The obtained experimental values are listed in Table 1. The reference date for ^{131}I decay correction was either set to the date of sampling (for sediment, soil, grass and milk) or to the date of maximum rainfall (for 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 (DWD, 2011a; PTB, 2011; BfS, 2011; Uni Oldenburg, 2011). All stations reported values for ^{131}I and ^{137}Cs, and the data are quite consistent among each other, showing maxima around March 29 and another short-term increase of activity around April 5. Data on other isotopes are scarcer; the information used here was obtained from the website of the German national metrology institute, PTB (2011), which reports data also on ^{134}Cs and ^{136}Cs. A time series representing the arithmetic means of ^{131}I air concentrations measured at 4 stations in Germany (Braunschweig, Potsdam, Offenbach and Schauinsland) is included in Fig. 2 (DWD, 2011a; PTB, 2011; BfS, 2011).

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 using 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 SSK (2004a) has been used. It contains a "washout coefficient" $\Lambda_0 = 7 \cdot 10^{-5}$ s^{-1}, indicating the fraction of aerosols washed out from 1 m^3 of air per second, at a standard rain intensity of 1 mm·h^{-1}. Deposition can then be calculated using

$$D = C_{air} \Lambda_0 t_{rain} h \quad (1)$$

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. Here, h replaces an expression for the 3-dimensionaldistribution of a radioisotope in the atmosphere,

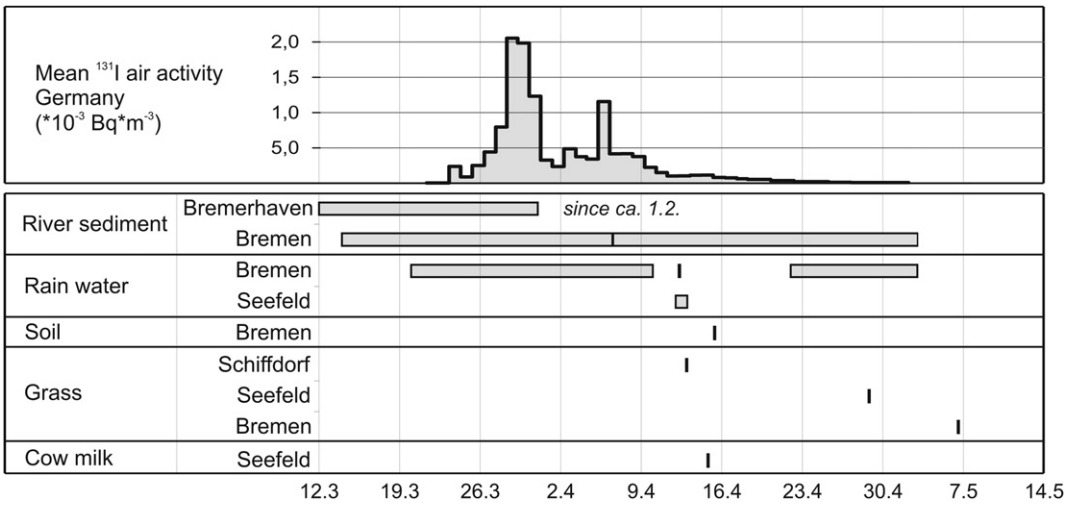

Fig. 2. Timeline of the sampling campaign. In the upper panel, averaged ^{131}I air concentrations reported by 4 German air activity monitoring stations in Braunschweig, Potsdam, Offenbach and Schauinsland (data taken from: PTB, 2011; DWD, 2011a; BfS, 2011) are plotted.

assuming constant concentration within the boundary layer. Given the long travel distance, this simplifying assumption seems justified. For the period between March 21 (first detection of the Fukushima plume in Germany) and April 6 (collection of a two-week sediment sample 2), 8.5 mm of rainfall were reported for Bremen by the German meteorological service, DWD (2011b). 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 0.025 Bq·l^{-1}, respectively.

3.2. Comparison of measured and derived expected values

3.2.1. Rain water

Measured concentrations of ^{131}I (Table 1) agree with the values estimated in the section 3.1.2 in a satisfactory manner, given the large uncertainties in the chosen parameters. Logically, later values for ^{131}I decrease with time in correspondence with the published air concentration values.

3.2.2. River sediment and soil

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 section 3.1.2. Concentration in sample 3, collected at the same site as sample 2, is already below the instrumental detection limit. The reported air concentration had decreased by about a factor of 10 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.

The soil sample 4 was taken from an area of 0.16 m^2 with a mass of 2.47 kg. The calculated areal ^{131}I deposition of 1.05 Bq·m^{-2} decay corrected to the maximum rainfall date (April 4) results in the value of 2.72 Bq·m^{-2}, similar to the deposition predicted in section 3.1.2.

The ^{137}Cs concentrations found in sediment and soil are much higher than those for ^{131}I, and similar to those found in samples collected at the same locations in previous years. Together with the fact that no ^{134}Cs could be detected, it can be concluded that the major portion of ^{137}Cs must originate from atmospheric bomb test and Chernobyl contributions (which in northern Germany have about the same magnitude). This is reflected in the soil sample directly, and most probably ^{137}Cs has reached the sediment by surface erosion and redistribution processes.

3.2.3. Grass

The range of the measured concentrations in grass was found to be very wide (Table 1). One reason might be the relatively early

Table 1
Measured values in fresh mass. Data are decay corrected to the last sampling date, unless noted otherwise in footnote.

No	Type of sample	Moisture content (%)	Date (2011)	^{131}I (Bq·kg^{-1})	^{137}Cs (Bq·kg^{-1})	^{134}Cs (Bq·kg^{-1})
1	River sediment (Bremerhaven)	80.8	~1.2–31.3	0.113 ± 0.046	2.67 ± 0.07	< 0.084
2[a]	River sediment (Bremen)	91.5	14.3–6.4	0.45 ± 0.06	1.76 ± 0.04	< 0.06
3	River sediment (Bremen)	89.9	6.4–3.5	< 0.12	1.73 ± 0.05	< 0.05
4	Soil (Bremen)	10.3	15.4	0.068 ± 0.025	3.00 ± 0.07	< 0.069
5[a]	Rain water (Bremen)	n.a.	20.3–10.4	0.43 ± 0.03	< 0.04	< 0.04
6	Rain water (Bremen)	n.a.	12.4	0.10 ± 0.02	< 0.04	<0.04
7	Rain water (Seefeld)	n.a.	11.4–13.4	0.14 ± 0.02	< 0.03	< 0.03
8[b]	Rain water (Bremen)	n.a.	22.4–3.5	0.031 ± 0.008	< 0.02	< 0.02
9	Grass (Schiffdorf)	53.5	13.4	3.58 ± 0.13	1.59 ± 0.07	0.32 ± 0.03
10	Grass (Seefeld)	73.2	28.4	0.31 ± 0.04	0.26 ± 0.04	0.062 ± 0.014
11	Grass (Bremen)	n.d.	6.5	0.12 ± 0.03	0.18 ± 0.03	< 0.08
12	Milk (Seefeld)	n.a.	14.4	0.08 ± 0.02	< 0.02	< 0.03

Abbreviations: n.a. = not applicable, n.d. = not determined.
[a] Decay corrected to the date of the maximum rainfall (3.4.2011).
[b] Mixed with old rain water.

time of the year: in the investigated region grass had just begun to grow, and thus the collectable mass per area presumably varied considerably. Nevertheless, the decrease in activity ratio between ^{131}I and ^{137}Cs with time of sampling is clearly visible – it is a consequence of the radioactive decay of ^{131}I.

For comparison, the maximal post-Chernobyl concentrations in grass (fresh mass) from a meadow in Bremen were reported to be 2450 Bq·kg^{-1} for ^{131}I and 230 Bq·kg^{-1} for ^{134}Cs on May 5, 1986 (Fischer et al., 1986). This is approximately 700 times higher than activities found in sample 9 (3.58 ± 0.13 Bq·kg^{-1} for ^{131}I and 0.32 ± 0.03 Bq·kg^{-1} for ^{134}C). The maximal ^{137}Cs activity of post-Chernobyl grass reported was 555 Bq·kg^{-1} (Fischer et al., 1986), compared to 1.59 ± 0.07 Bq·kg^{-1} in this study. The Cs isotope ratio will be discussed in section 3.2.5.

3.2.4. Milk

The transfer of the radionuclides from grass to milk can be predicted by applying emergency models like SSK (2004b). There, the concentration in milk is obtained using the following equation:

$$C_{milk} = C_{grass}\dot{M}_{grass}T_{milk} \quad (2)$$

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^{-1}) and T_{milk}: element-specific transfer factor grass-milk (0.003 for iodine). Using the indicated values, the ^{131}I concentration of sample 10 (0.31 Bq·kg^{-1}), decay corrected to the time of milk sampling (1.04 Bq·kg^{-1}), yields a concentration in milk of 0.20 Bq·kg^{-1}, somewhat higher than measured (0.08 ± 0.02 Bq·kg^{-1}). Due to the small amount of grass available on the meadows in early April, only approximately 50% of the cattle's daily diet originated from fresh grass, according to the farmer's estimation. This is consistent with a lower ^{131}I concentration actually present in milk.

Maximum ^{131}I concentrations in the study region in milk after the Chernobyl accident reached up to 40 Bq·kg^{-1} (Fischer et al., 1986).

3.2.5. Isotope ratios

Activity ratios can reveal additional information on radioisotope origin. 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 (PTB, 2011)), sample 9 was ashed (to increase detector efficiency) and measured again. 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 Northwest 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. Maximum registered concentrations of ^{131}I and ^{134}Cs assigned to Fukushima fallout in grass from the study area are about 3 orders of magnitude lower than those reported in the first weeks after the Chernobyl accident in the same region.

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