Traces of Fukushima fallout in the environment of Northwest Germany

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Introduction

Following the Fukushima NPP, Japan, radionuclide air releases, samples of environmental media – **rain water, river sediment, soil, grass and cow milk** - from federal states Bremen and Lower Saxony were taken and analyzed by standard **low level low background gamma spectrometry** for traces of isotopes indicating Fukushima fallout in the Radioactivity measurements laboratory at the University of Bremen, partially as a part of routine environmental radioactivity monitoring scheme.

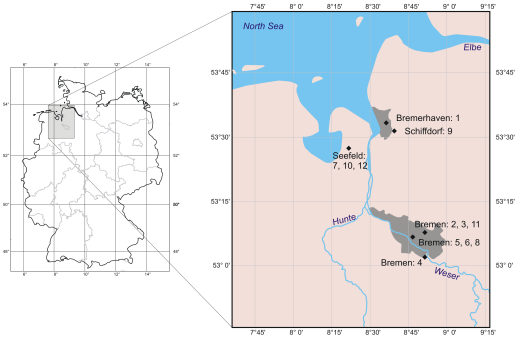

Figure 1: *Location of the sampling points. For each location, sample numbers are indicated according to Table 1.*

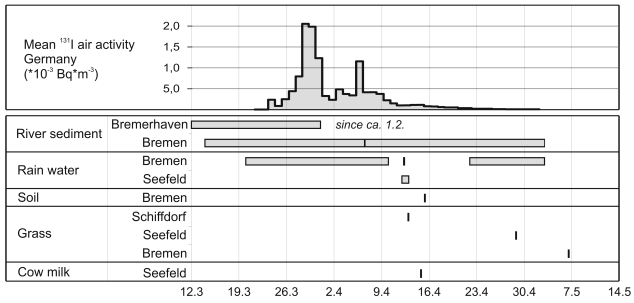

Figure 2: *Timeline of the sampling campaign. In the upper panel, averaged ^{131}I air concentrations reported by 4 German air activity monitoring stations in Braunschweig, Potsdam, Offenbach and Schauinsland (data taken from: PTB, 2011; DWD, 2011; BfS, 2011) are plotted.*

Measured values

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	ca. 1.2. – 31.3.	0.113 ± 0.046	2.67 ± 0.07	< 0.084
2[1]	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[1]	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[2]	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

[1]Decay corrected to the date of the maximum rainfall (3.4.2011). [2]Mixed with old rain water.
Abbreviations: n.a.=not applicable, n.d.=not determined.

Isotope ratios

Activity ratios can reveal additional information on radioisotope origin.

[^{131}I]/[^{137}Cs]

In NW Germany, a residual deposition of about 2 kBq·m^{2} ^{137}Cs remains from bomb test fallout and Chernobyl. Therefore 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.

[^{134}Cs]/[^{137}Cs]

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 air concentration data. Assuming this value, it can be concluded that the main ^{137}Cs contribution in the grass samples is "old".

[^{136}Cs]

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.

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Models

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 (after SSK, 2004a) as:

$$D = C_{air}\,\Lambda_0\,t_{rain}\,h \quad [1]$$

$$C_{rain} = D/P \quad [2]$$

D … areal deposition
C_{air} … isotope concentration in air
Λ_0 … standard washout coefficient (=7·10^{-5} s^{-1})
t_{rain} … rainfall duration
h … contaminated air column height (=1000 m) (replaces an expression for the 3-D distribution of a radioisotope in the atmosphere, assuming constant concentration within the boundary layer)
C_{rain} … activity concentration in rain water
P … precipitation (l·m^{-2})

Transfer of radioiodine from grass to milk

can be predicted by applying emergency models like SSK (2004b).

$$C_{milk} = C_{grass}\,\dot{M}_{grass}\,T_{milk} \quad [3]$$

C_{milk} … activity concentration in milk
C_{grass} … activity concentration in grass
$\dot{M}_{grass}$ … daily grass consumption rate of cattle
T_{milk} … element-specific transfer factor grass-milk (0.003 for iodine)

Table 2: *Comparison of modelled and measured values of Fukushima fallout in NW Germany. Historical data on post-Chernobyl concentrations in the studied area.*

	Modelled concentrations	Measured concentrations	Post-Chernobyl conc.[1]
Areal deposition	2.14 Bq·m^{-2} ^{131}I 0.21 Bq·m^{-2} ^{137}Cs (Eq. 1)	1.46 Bq·m^{-2} ^{131}I (sample 2 – sediment) 2.72 Bq·m^{-2} ^{131}I (sample 4 – soil) Most of ^{137}Cs "old"	ca. 12000 Bq·m^{-2} ^{131}I ca. 3000 Bq·m^{-2} ^{137}Cs
Rain water	0.252 Bq·l^{-1} ^{131}I 0.025 Bq·l^{-1} ^{137}Cs (Eq. 2)	max. 0.43 Bq·l^{-1} ^{131}I < 0.04 Bq·l^{-1} ^{137}Cs	
Grass		max. 3.58 Bq·kg^{-1} (w.m.) ^{131}I max. 1.59 Bq·kg^{-1} (w.m.) ^{137}Cs (Most of ^{137}Cs "old") max. 0.32 Bq·kg^{-1} (w.m.) ^{134}Cs	2450 Bq·kg^{-1} (w.m.) ^{131}I 450 Bq·kg^{-1} (w.m.) ^{137}Cs 230 Bq·kg^{-1} (w.m.) ^{134}Cs
Milk	0.20 Bq·kg^{-1} ^{131}I (Eq. 3)	0.08 Bq·kg^{-1} ^{131}I	40 Bq·kg^{-1} ^{131}I

[1] Fischer et al. (1986)

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 NW 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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