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RESEARCH ARTICLE

Artificial radioactivity in environmental media (air, rainwater, soil, vegetation) in Austria after the Fukushima nuclear accident

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Abstract Several environmental media in Austria were monitored for artificial radionuclides released during the Fukushima nuclear accident. Air (up to 1.2 mBq/m^3 particulate ^{131}I) and rainwater (up to 5.2 Bq/L ^{131}I) proved to be the media best suited for the environmental monitoring, allowing also a temporal resolution of the activity levels. Significant regional differences in the wet deposition of ^{131}I with rain could be observed within the city of Vienna during the arrival of the contaminated air masses. Forward-trajectory analysis supported the hypothesis that the contaminated air masses coming from the northwest changed direction to northeast over Northern Austria, leading to a strong activity concentration gradient over Vienna. In the course of the environmental monitoring of the Fukushima releases, this phenomenon—significant differences of ^{131}I activity concentrations in rainwater on a narrow local scale (8.1 km)—appears to be unique. Vegetation (grass) was contaminated with ^{131}I and/or ^{137}Cs at a low level. Soil (up to 22 Bq/kg ^{137}Cs) was only affected by previous releases (nuclear weapon tests, Chernobyl). Here, also significant local differences can be observed due to different deposition rates during the Chernobyl accident. The effective ecological half-lives of ^{137}Cs in soil were calculated for four locations in Austria. They range from 7 to 30 years. No Austrian sample investigated herein exceeded the detection limit for ^{134}Cs; hence, the Fukushima nuclear accident did not contribute significantly to the total radiocesium inventory in Austrian environmental media. The levels of detected radioactivity were of no concern for public health.

Keywords Fukushima Daiichi nuclear power plant · Nuclear reactor accident · Environmental monitoring · Artificial radionuclides · Radioiodine · Radiocesium · Austria

Introduction

During the Fukushima nuclear accident large amounts of radionuclides were released into the atmosphere (order of magnitude approximately 10^{18}Bq). Radioactive noble gases (e.g. ^{133}Xe and ^{85}Kr) and several radionuclides, in particular those of iodine (e.g. ^{131}I, ^{133}I and ^{135}I), tellurium (e.g. ^{129m}Te) and cesium (e.g. ^{134}Cs, ^{136}Cs and ^{137}Cs) were released from the damaged reactors due to their volatility. Several estimations for the source terms were published (Hamada and Ogino 2012). The Japanese Nuclear and Industrial Safety Agency reported a release of $1.6{\cdot}10^{17}$Bq for ^{131}I and $1.5{\cdot}10^{16}$Bq for ^{137}Cs (NERH 2011). The Nuclear Safety Commission estimated the amount of certain nuclides discharged into the atmosphere from 11 March to 5 April with assistance from the Japan Atomic Energy Agency as $1.5{\cdot}10^{17}$Bq ^{131}I and $1.2{\cdot}10^{16}$Bq ^{137}Cs (Chino et al. 2011; NERH 2011). Stohl et al. (2011) reported higher releases, in particular $6.25{\cdot}10^{16}$Bq ^{137}Cs.

Other radionuclides such as actinoids or radiostrontium are less volatile and hence were released from the molten nuclear fuel of the Fukushima nuclear power plant to a much lower extent. On a global scale, their occurrence in the environment still is largely due to atmospheric nuclear explosions of the twentieth century (with minor contributions from the Chernobyl accident). In fact, in the course of a large European study (Masson et al. 2011), no significantly

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increased ^{90}Sr levels were observed in Europe after the Fukushima accident.

For environmental monitoring and atmospheric dispersion of the γ-emitting radionuclides ^{131}I, ^{134}Cs and ^{137}Cs, however, a couple of studies have been published in the past months from stations on all continents of the northern hemisphere (e.g. Kim et al. 2012; Masson et al. 2011; Norman et al. 2011; Melgunov et al. 2012). These radionuclides are characterised by sufficiently long half-lives and high volatility to be transported over large distances in significant amounts.

The Fukushima accident impressively illustrated the need of published environmental radioactivity data in order to be in a position to distinguish 'fresh' (artificial) activities after a nuclear accident from the background levels of a certain location. This paper represents the Austrian contribution to this constantly growing set of data.

Material and methods

Sampling

Several samples from various environmental media were taken in Northern and Eastern Austria (Vienna and Lower Austria), in particular air (aerosol fraction), rainwater, soil and vegetation.

Aerosol samples were taken in a self-constructed air-filtering station, which was originally dedicated to the emission surveillance of the Atominstitut's research reactor during operation, collecting the aerosol fraction of 65 m^3 of air per day. The station is operated with Whatman® GF 10 glass fibre filters.

Rainwater was sampled in two spots in Vienna with self-constructed samplers. One spot was the Atominstitut in Vienna's southeastern district no. II (N48.196625, E 16.412725), the other one was located in Vienna's northwestern district no. XIX (N48.249528, E16.338292) providing the comparison of two spots in Vienna. The amount of rain collected was comparable in both spots at least in the beginning of the sampling campaign.

Several soil samples (topsoil 0–15 cm; sampled amount approximately 500 g) were taken from the Atominstitut's garden (N48.196625, E16.412725) and measured after removal of grass and roots.

Vegetation primarily included grass samples from the Atominstitut's garden. After we had surmised a positive activity concentration gradient in environmental samples from south to north, one grass sample from Northern Austria (Magersdorf in Lower Austria, north of Vienna; N48.561045, E16.100621) was taken at a time, when the detection limit in vegetation samples was hardly exceeded in Vienna.

Radioactivity measurements

For the determination of the radioactivity, γ-spectrometry was performed on the low-level counting facility of the Atominstitut, consisting of a 226 cm^3 HPGe-detector (Canberra™, detector model GC5020; 2.0-keV resolution at the 1,332 keV ^{60}Co peak; 52.8 % relative efficiency), connected to a PC-based multi-channel analyzer with preloaded filter and located inside an ORTEC™ HBLBS1 shielding. Air filters were measured directly in a calibrated geometry. Samples of rainwater and vegetation were measured in 1-L Marinelli beakers in a calibrated geometry. If necessary, rainwater was diluted with distilled water to obtain the desired volume of 1 L. Soil was measured in polyethylene beakers, for which the geometry and γ-ray self-shielding was calibrated by using quartz sand soaked with a diluted radionuclide calibration solution (see below). The measurement times of the samples were at least 20 h for air filters or, for other sample types, typically 2 or 3 days, or longer, until no significant improvement of the counting error of the most interesting peaks could be yielded by a reasonably longer measurement time of a couple of days. Uncertainties given in the figures and tables are due to counting statistics. The targeted nuclides were ^{131}I ($T_{1/2}$= 8.03 d), ^{134}Cs ($T_{1/2}$=2.07 y), ^{136}Cs ($T_{1/2}$=13.04 d) and ^{137}Cs ($T_{1/2}$=30.08 y). Peak summation effects were considered for the activities of ^{134}Cs. All nuclear data in this paper and for the evaluation of data such as half-lives and γ-ray intensities were taken from the National Nuclear Data Center (NNDC 2012).

Validation

The amount of sampled air was determined with a gauged gas flow counter. This air sampling procedure is also part of the Austrian licencing requirements for the operators of research reactors. For quality control, the activities of natural, cosmogenic ^{7}Be were used as an internal standard, whose typical activity levels are well-known from background measurements. For calibration of the γ-detector's efficiency for the investigated radionuclides, a QCY48 (Amersham® Ltd.) certified standard solution was used. Aliquots of the freshly opened QCY48 vial were weighed quickly into calibration containers that were identical in shape and material with the sample containers and diluted with 4 M hydrochloric acid (p.a., Rotipuran®). For quantification of the detector efficiency for soil samples, the standard solution was diluted and filled with Quartz sand (p.a., Merck™) to the desired sample volume. This completely soaked sand simulated the bulk density of the soil well. For quality control, the activity of a solution of ^{137}Cs with known activity was spiked into a volume of non-radioactive soil and could be recovered with good reproducibility, hence approving the suitability of the calibration of the

detector's efficiency. For the dilution of rainwater, distilled water was used that was produced prior to the Fukushima nuclear accident. Hence, no disturbing radioactivity was introduced in the course of sample preparation.

Results and discussion

Whereas ^{137}Cs can regularly be determined in the environment in Austria, the appearance of ^{131}I and ^{134}Cs can be clearly attributed to the Fukushima nuclear accident. Due to its short half-life, ^{131}I from previous releases has decayed completely; ^{134}Cs, if still present from the Chernobyl accident in minute amounts, can no longer be monitored in typical samples of Central Europe since the 1990s. Thus, both radionuclides are clear indicators for Fukushima-related activities, whereas ^{137}Cs can be present also from previous events. For many decades, soil has been measured by several Austrian institutions more or less regularly. It has been observed that approximately 10 % of the ^{137}Cs activity inventory in Austria is related to the nuclear weapon testing of the twentieth century and 90 % to the Chernobyl accident (Bossew et al. 1995).

Aerosols

Activities determined in aerosol filters from 23 March 2011 to 12 April 2011 are shown in Fig. 1. The ^{7}Be activity concentrations in air were consistent with the levels typically observed in Central Europe, hence errors in the course of sampling (e.g. due to pump failure) can be excluded. No radiocesium could be determined due to the limited capacity of our pump compared with CTBTO stations using high volume samplers. In Europe the levels of airborne radiocesium were approximately one order of magnitude lower than activity concentrations of radioiodine in spring 2011 (Masson et al. 2011). Not all European monitoring stations, as described below, could determine radiocesium from Fukushima.

The results are in very good agreement with previously published European data (Masson et al. 2011). That study suggested that Eastern Austria was a region in Europe with a high ^{131}I activity gradient in March and April 2011. Stations located in countries northeastern of Austria monitored a higher activity concentration of up to 6 mBqm^{-3}; stations located south and southwestern of Austria revealed much lower activity concentrations. On the Iberian Peninsula, significantly higher ^{131}I concentrations (up to 3.69 mBqm^{-3}) were measured, compared to Austria (Lozano et al. 2011); however, the contaminations over Central Europe and the Iberian Peninsula, respectively, entered the European airspace via two different meteorological pathways (Masson et al. 2011); Central Europe was hit by air masses from the polar regions, and the Iberian Peninsula from air masses flowing eastwards over the North American continent and the Atlantic Ocean. Figure 1 illustrates both the temporal evolution of the activity concentrations in air as well as the relationship with the occurrence of natural ^{7}Be. We found this figure also to be useful to illustrate for the public that the levels of radioactivity in Europe were of no concern for human health.

Rainwater and trajectory analysis

Only ^{131}I could be determined in rainwater from both spots in Vienna (districts XIX and II). The results are tabulated in Table 1. The temporal evolution of the activity concentration is illustrated in Fig. 2.

From this figure, it becomes obvious that the decrease in the ^{131}I activity concentration was faster than solely due to radioactive decay with the physical half-life of 8 days. This indicates the efficiency of the wash-out and dilution effects that have already been topic in the study of Pittauerová et al.

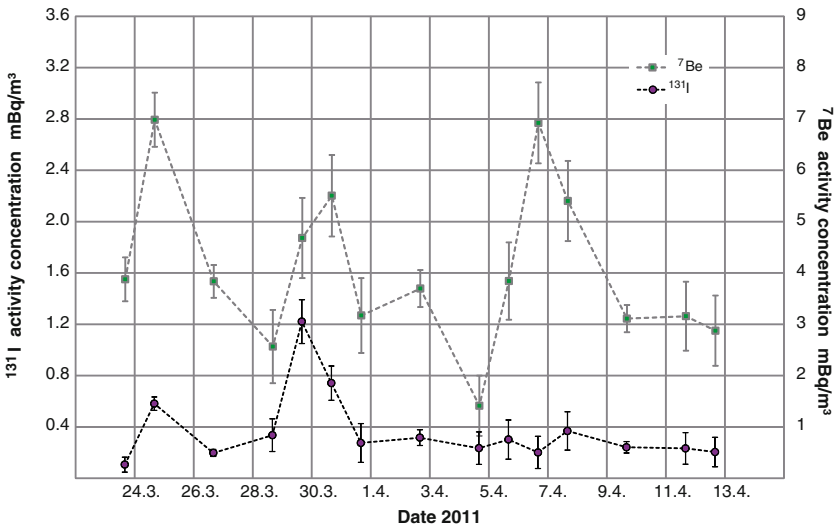

Fig. 1 Temporal evolution of the particulate ^{131}I activity concentrations in air in Vienna compared to natural ^{7}Be

Table 1 Activity concentrations of ^{131}I in rainwater in the northwest of Vienna (district XIX) as well as in the southeast (district II)

Date	Vienna, district XIX		Vienna, district II	
	^{131}I, BqL^{-1}	±	^{131}I, BqL^{-1}	±
26 March 2011	5.2	0.94	1.1	0.19
27 March 2011	<2.4		0.39	0.051
31 March 2011	0.34	0.019	<3.3	
01 April 2011	1.1	0.10	Not collected	
04 April 2011	0.35	0.035	Not collected	
12 April 2011	0.20	0.018	0.37	0.029
13 April 2011	0.16	0.018	0.14	0.020
14 April 2011	0.16	0.013	Not collected	
25 April 2011	0.033	0.0089	<0.055	
29 April 2011	<0.22		<0.043	
30 April 2011	<0.39		Not collected	
02 May 2011	<0.25		Not collected	

Uncertainties are due to counting statistics. Decay correction to the approximate beginning of the rainfall

(2011). Further, it is interesting to note the significant regional differences in wet deposition within the city of Vienna. Despite the relatively short distance of 8.1 km (which represents almost the maximum possible distance within the City of Vienna), the activity concentrations in both spots did not correlate in the beginning of the sampling campaign: Rain from the northwest of Vienna (district XIX) revealed higher ^{131}I activity concentrations in the very beginning than samples from the southeast (district II). Only later, the activity concentrations found in both spots were comparable. Masson et al. (2011) showed that the contaminated air masses entered European airspace along a relatively narrow corridor in a southeastern direction; hence, it is likely that they have passed Vienna's sampling station in district XIX before the one in district II. From the activities measured in rainwater, it appears possible that the air masses changed to another direction over Eastern Austria, thus affecting district XIX to a greater extent than district II. In this respect it is also noteworthy that the radionuclide monitoring station of the Federal Austrian Agency for Health and Food Safety (AGES)—located in the northeast of Vienna—also monitored a slightly higher maximum activity concentration of particulate ^{131}I in air (1.44 mBqm^{-3}) than the station of the Atominstitut (1.22 mBqm^{-3}) on 29 March 2011 (Masson et al. 2011). Unfortunately, no activity data for rainwater have been published by the AGES yet.

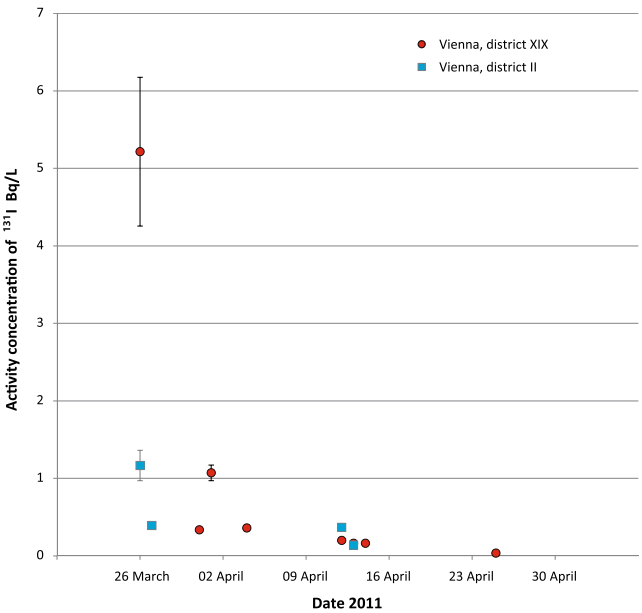

Fig. 2 Temporal evolution of the ^{131}I activity concentrations in rainwater in Vienna, district XIX (northwestern district of Vienna) and Vienna, district II (southeastern district of Vienna)

In order to test the hypothesis of a contribution of sudden wind direction changes to the activity concentration gradient, forward-trajectory analysis was performed using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess 1998; Draxler and Rolph 2012; Rolph 2012; see Fig. 3). The HYSPLIT model is a complete and well-suited system for computing single air parcel trajectories (Draxler and Rolph 2012). It uses available meteorological data and can be operated in forward or backward trajectory mode.

The starting point of the trajectories was chosen to fit the corridor of the contaminated air masses over the North Sea, as suggested by Masson et al. (2011). According to the trajectory calculation, the air masses headed towards Austria in the relevant time frame (26–28 March), where they arrived at an altitude of <2,000 m, which is the typical altitude for rainclouds. In fact, the trajectories support the hypothesis that the centre of the corridor switched to northeastern direction over northeastern Austria heading for the Ukraine and Belarus, hence leaving the sampling station in

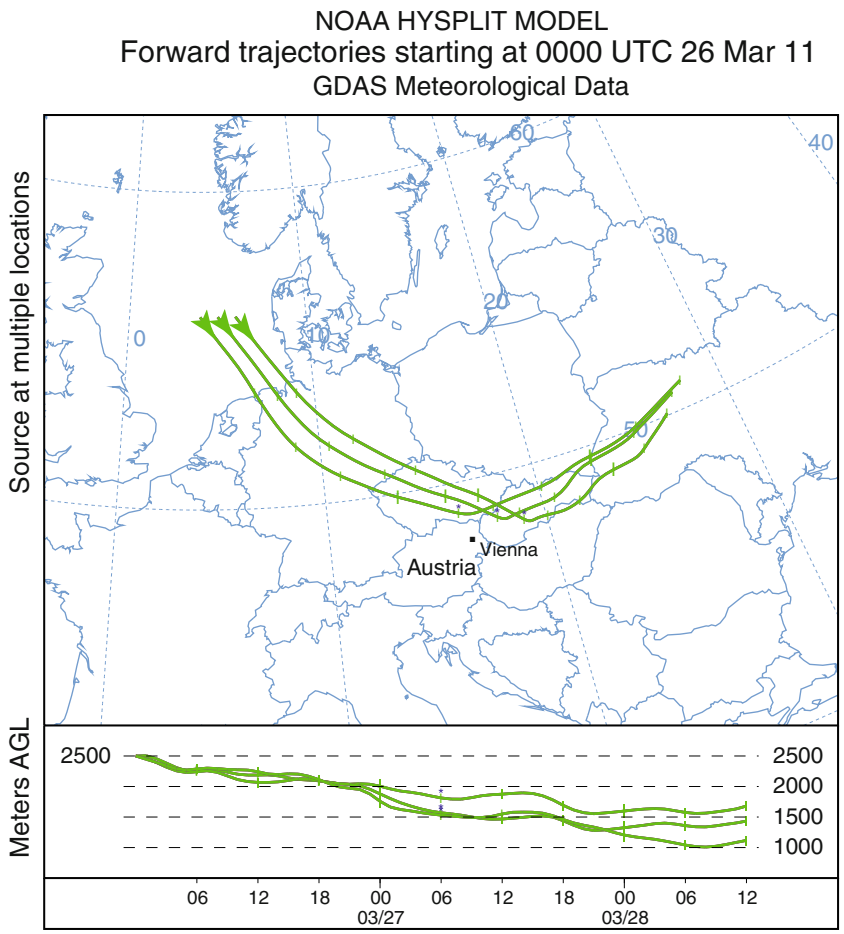

Fig. 3 Forward trajectories of air masses over Central Europe from 26 to 28 March 2011

Vienna's district XIX more in the centre and the one in district II more at the edge of the relatively narrow corridor of contaminated air masses. In fact, the stations located next to this trajectory in Central Belarus (Minsk) and Eastern Belarus (Mstislavl), the latter which is located almost in the centre of the corridor, monitored increased ^{131}I particulate concentrations of 2–3 mBq m^{-3} in the relevant time frame of 29–31 March (Masson et al. 2011). Since it can be assumed that radioiodine is dispersed in the form of localised meteorological corridors, the exemplified model indicates that a strong gradient in activity concentrations (i.e. large activity differences on a narrow local scale) can be resolved, despite the fact that the source is located some 10 000 km away.

In comparison with previous studies, German (Pittauerová et al. 2011) and Korean rainwater (Kim et al. 2012) showed lower ^{131}I activity concentrations than Austrian rainwater; rainwater from San Francisco (Norman et al. 2011) was contaminated with radioiodine up to 16 Bq L^{-1} (three times more than the maximum measured in Vienna, district XIX).

For comparison with Japan, three rain samples were obtained from Tokyo, Japan and measured in the course of this study. The activity concentrations determined in those samples are summarised in Table 2. They showed much higher activities, also of radionuclides that did not exceed the minimum detectable activity in Austrian rainwater (including ^{136}Cs with a half-life of 13 days, as well as ^{134}Cs and ^{137}Cs). As already discussed above, the study by Masson et al. revealed that radiocesium activities in Europe were approximately one order of magnitude below the respective ^{131}I activity concentrations.

Assuming the utilisation of the Austrian rainwater (Table 1) as potable water, in theory, at least 24 L (up to more than 3,000 L) would have to be consumed by an adult every day over the period of a year (with constant activity concentrations), to receive a committed effective dose of 1 mSv. This is regarded as an acceptable dose for members of the public according to European law (The Council of the European Communities 1996). Drinking of the Japanese rainwater (Table 2) from 4 April 2011 would not be advisable, since the activity concentrations are above the Japanese regulatory limits valid at that time (Hamada and Ogino 2012).

Other samples

Since radioiodine is characterised by high volatility, a part of the soil and vegetation samples was measured in fresh state;

Table 2 Activity concentrations of artificial radionuclides in rainwater from Tokyo, Japan

Date and time of sampling	Sample volume (mL)	^{131}I (BqL^{-1})	±	^{134}Cs (BqL^{-1})	±	^{136}Cs (BqL^{-1})	±	^{137}Cs (BqL^{-1})	±
04 April 2011, 13:00 hours	359	284	6	228	4	17	1	236	4
19 April 2011, 20:00 hours	35.5	<11		26	3	<6		19	2
24 April 2011, 07:00 hours	417	1.4	0.2	24.3	0.3	0.56	0.08	27.9	0.4

Uncertainties are due to counting statistics. Decay correction to the time of sampling

another part was dried at room temperature before measurement. In soil, the analyses revealed no other activities than ^{137}Cs (Table 3) that were all in the same range that have been determined prior to the Fukushima accident. Hence, no significant contribution from the Fukushima nuclear accident (i.e. ^{134}Cs) was detectable.

In soil from Athens, ^{134}Cs could be determined in activity concentrations up to 1.4 Bqkg^{-1} (Kritidis et al. 2012). The activity concentrations of ^{137}Cs were significantly lower compared to Austrian soil though, indicating that the contamination with ^{134}Cs was caused by fallout from the Fukushima plume. Small amounts of ^{134}Cs were also detected in soil from Milano, Italy (Ioannidou et al. 2011). Like in Vienna, no ^{134}Cs was detected in soil from Bremen (Germany; Pittauerová et al. 2011).

For validation of the ^{137}Cs activities obtained, a comparison with ^{137}Cs activities from measurements prior to the Fukushima nuclear accident is shown in Fig. SM1 in the "Electronic Supplementary Materials" (ESM). For this comparison, raw data of ^{137}Cs activity concentrations in soil were taken from the Government of Upper Austria (2012). Data from the Atominstitut in Vienna (biannual data since 2006) were compared to ^{137}Cs contaminations from three locations in Upper Austria (annual data since 1992): Linz, Schärding and Vöcklabruck (see ESM). The distances between each of these locations are approximately 50–65 km; they are located 150–220 km west of Vienna. Despite the relatively short distances, the stations in Upper Austria showed highly different activity concentrations of ^{137}Cs in soil. This can be explained with the wet deposition of radiocesium after the Chernobyl accident due to rainfall around Vöcklabruck and other factors (Bossew et al. 1995; Machart et al. 2007). This once again evidences that the degree of contamination after a nuclear accident is not only a function of distance. In any case, in none of the locations a significant increase in the levels of ^{137}Cs contamination could be observed after the Fukushima accident. Although only radiocesium from Chernobyl and nuclear weapon tests was detected in Austria, this figure provides some insight for the understanding of the environmental behaviour of radiocesium, which may be of interest for the assessment of highly contaminated areas after the Fukushima nuclear accident.

Persistence and bioavailability of radiocesium in soil depends on several factors such as the composition of soil, the amount of rain, vegetation and, to a large extent, on the presence of fungal communities (Zhdanova et al. 2005). These factors will result in an effective ecological half-life

Table 3 Radioanalytical results for the measurement of soil and grass

Sample type	Date	^{131}I (Bqkg^{-1})	±	^{134}Cs (Bqkg^{-1})	±	^{137}Cs (Bqkg^{-1})	±
Soil, Vienna, air dried	12 April 2011	<0.4		<0.44		21.4	0.7
	12 April 2011	<0.9		<1.1		13.5	1
	24 October 2011	N/A		<1.4		26	2
	24 October 2011	N/A		<1.2		9.0	1
Soil, Vienna, fresh	30 September 2011	N/A		<0.3		22.0	0.3
	01 November 2011	N/A		<0.1		6.6	0.1
Grass, Vienna, air dried	12 April 2011	4.8	7.4	<5.2		<4.6	
	12 April 2011	19	13	<5.3		<6.2	
	24 October 2011	N/A		<3.6		<3.5	
	24 October 2011	N/A		<4.1		<4.0	
Grass, Vienna, fresh	15 April 2011	<0.64		<0.53		<0.58	
	19 April 2011	<0.63		<0.53		<0.57	
Grass, Magersdorf, fresh	20 April 2011	1.38	0.03	<0.052		0.34	0.02

Uncertainties are due to counting statistics. Decay correction to the date of sampling

N/A not applicable due to the radionuclides's short half-life (for determination of ^{131}I in samples measured later than May 2011)

(T_e), which may differ significantly from the physical half-life of the radionuclide. ESM Fig. SM1 allows the estimation of the T_e of ^{137}Cs in the above mentioned locations, which show large variations despite relatively short distances: Vienna T_e=7.1 years; Vöcklabruck T_e=12.4 years; Linz T_e=18.7 years; and Schärding T_e=29.8 years. Lastly, it must be mentioned that the concept of the ecological half-life can be impaired if the distribution of the ^{137}Cs in soil is nonuniform and if unexpected mixing and transport processes occur within this medium (Pröhl et al. 2006).

The results of the analysis of vegetation are included in Table 3. Grass samples showed low activity values for ^{131}I and ^{137}Cs. Since no ^{134}Cs was determined, it can be concluded that no Fukushima-derived radiocesium was detected. The concentrations of ^{131}I in Austrian grass were higher compared with Italy (0.037–0.066 Bqkg^{-1}) (Ioannidou et al. 2011) but in the same range as in grass from France (up to 9 Bqkg^{-1}) (Parache et al. 2011), Germany (0.12–3.58 Bqkg^{-1}) (Pittauerová et al. 2011), Greece (2.1 Bqkg^{-1}) (Kritidis et al. 2012) or Romania (1.54 Bqkg^{-1}) (Cosma et al. 2012).

Conclusions

Several environmental media were monitored for artificial radionuclides released during the Fukushima nuclear accident, in particular ^{131}I. Air (up to 1.2 mBqm^{-3} particulate ^{131}I) and rainwater (up to 5.2 BqL^{-1} ^{131}I) proved to be the media best suited for the environmental monitoring, allowing also a temporal resolution of the activity levels over Austria.

A trajectory analysis of air masses entering European airspace over the North Sea shows a change of wind directions over the northeastern part of Austria and supports the soundness of a joint European study (Masson et al. 2011) in this respect. Analysis of rainwater revealed significantly higher ^{131}I activity concentrations in samples from northwestern Vienna than from southeastern Vienna in the very beginning of the sampling campaign. This can be explained, at least in part, by the strong activity gradient due to the changing direction of the air masses from southeastern to northeastern direction over northern Austria. To our knowledge, this is the first report of such significant differences of ^{131}I activity concentrations in rainwater on a narrow local scale. Previous reports from short-distant stations reported rather similar activity concentration values in precipitation, obviously lacking the necessary local activity concentration gradient (Bolsunovsky and Dementyev 2011). The temporal evolution of the activity concentrations in rainwater clearly showed wash-out and dilution effects.

Vegetation (grass) was contaminated with ^{131}I and/or ^{137}Cs at a lower level. Soil (up to 22 Bqkg^{-1} ^{137}Cs) was only affected by previous releases (nuclear weapon tests, Chernobyl). Here, also significant local differences can be observed due to different deposition rates. The effective ecological half-lives of ^{137}Cs in soil were calculated for four locations in Austria. They range from 7 to 30 years. No Austrian sample investigated herein exceeded the detection limit for ^{134}Cs; hence, the Fukushima nuclear accident did not contribute significantly to the total radiocesium inventory in Austrian environmental media. The levels of detected radioactivity were of no concern for public health.

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