Artificial radionuclides in the troposphere of Seville (Spain) due to the Fukushima accident, associated fallout and impact on the trophic chain

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Abstract

Radioactive emissions into the atmosphere from the damaged reactors of the Fukushima Daiichi nuclear power complex were transported in part across the Pacific Ocean to North America and then to Europe, where arrived quite diluted due to the dispersion experimented by the masses of contaminated air and their washing by rainfall during their displacement.

This paper shows the magnitude and temporal evolution of the radionuclide concentrations detected in the cited contaminated air masses at their arrival to Seville (Spain), where there is a station of the nation's environmental monitoring network equipped with high sensitivity devices for uninterrupted radiation detection. In the collected aerosol filters and during about two weeks were detected the presence of the following radionuclides: ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{131}I and ^{132}Te (together with its short-lived daughter ^{132}I) at minute levels and with characteristics ^{134}Cs/^{137}Cs and ^{136}Cs/^{137}Cs isotope ratios. The associated ^{131}I fallout due to this episode was also roughly estimated from additional wet and dry deposition measurements, while the presence of ^{131}I in gaseous form was evaluated through its collection with a pumping system equipped with a charcoal filter

It should also be noted that in Seville and surroundings, the presence of ^{131}I with origin in the Fukushima episode was detected in several key links in the human food chain: samples of milk (goat and cow) and derivative dairy products, as well as in various broadleaf plants. The maximum levels measured for this radionuclide were 1.11 Bq/l in samples of milk and 1.42 Bq/kg wet wt in broadleaf plants, with obviously negligible radiological implications. However, no presence of ^{132}Te or ^{134}Cs was detected in the samples of products for human consumption tested, while the possible changes in ^{137}Cs activities were in most cases masked within the global fallout levels.

Key Words: Fukushima accident; radioactive aerosols; wet and dry fallout; radiological impact in Spain

1.- Introduction

An earthquake on 11 March 2011 in the Pacific Ocean at 130 km off the coast of Honchu, in Japan, followed by a large tsunami that, amongst other unfortunate consequences, damaged the refrigeration systems of the Fukushima Dai-ichi nuclear reactor complex, caused a major release of radioactive material to the atmosphere for several days following the accident.

The anthropogenic radionuclides emitted into the atmosphere from the Fukushima Daiichi reactors were partially transported across the Pacific Ocean. The radioactive cloud first reached North America (Diaz-León et al., 2011), moving afterwards towards the Atlantic Ocean and finally reaching Europe. By this time, the radioactive cloud had already been strongly diluted, and the activity had decreased due to the washout and dry deposition during the time of transport, but still detectable. The total activities of radioiodines and radiocaesiums released into the atmosphere in the Fukushima Daiichi accident have been estimated in several works (for instance, IRSN, 2011a; Stohl et al., 2011), being the most reliable values $2\ 10^{17}$ and $3.5\ 10^{16}$ Bq for ^{131}I and ^{137}Cs respectively, which correspond to approximately 6% and 42% of the activity

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of these radionuclides released into the environment after the nuclear reactor accident of Chernobyl.

On 21 March 2011, the Icelandic CTBTO International Monitoring System network station was the first in Europe to detect the presence of ^{131}I traces in aerosols, at volumetric concentrations of 10 μBq/m^3 as reported elsewhere (ZAMG, 2011). On March 24, the Institut de Radioprotection et de Sureté Nucléaire, IRSN (France) informed of the presence of ^{131}I in the air (volumetric concentration of 12 μBq/m^3) measured at one of its radiological surveillance monitoring network stations (IRSN, 2011b). In Seville, the first traces of ^{131}I from the Fukushima accident were detected in an air filter collected in the period 21-28 March.

The present work has three main objectives. First, to make a detailed analysis of the temporal evolution of the activity levels found in the troposphere at the Sevilla station belonging to the Spanish radiological monitoring network. Second, to estimate the associated fallout in the studied area. And third, to evaluate the transfer dynamics of ^{131}I and radiocaesiums from the atmosphere to different environmental compartments which can be considered as characteristic receptors in the trophic chain (rainwater, different types of plants and milk), assessing in all cases the corresponding radiological significance. In this way, the environmental and radiological impact can be properly assessed.

2.- Materials and Methods

2.1. Sampling: Sites and Equipment

The station that provides the data is located on the roof of the Physics Faculty at Seville (South-West of Spain). High-volume aerosol filter samples, wet- and dry-deposition samples and charcoal cartridge samples of gaseous ^{131}I were collected. In addition, and in order to assess the influence on the food chain, samples of pasture, vegetables, milk, and cheese were collected from the vicinity of the station.

The high-volume aerosol collectors pumps in daily air volumes in the range of 10 000-16 000 m^3.The aerosols are retained on polypropylene filters, with dimensions of 440 × 440 mm, and have a retention efficiency greater than 93%. A low-volume suction collectors was also used for the collection in series of: (a) aerosols onto filters with a 19.6 cm^2 cellulose surface (0.8 μm, Millipore); and (b) gaseous ^{131}I in charcoal cartridges impregnated with TEDA (30-50 mesh). The daily volume drawn in by this low-volume samplers is in the range of 30-50 m^3.The collection times used for the aerosol filters and the gaseous iodine samplers varied greatly – from a few hours up to a week – depending on the levels of radioactive contamination which were being detected.

On days without rain, the dry fallout was sampled in the station using a custom-made polyethylene collector with a surface area of 500 cm^2 containing distilled water to trap the deposited aerosol, while the wet deposition samples were obtained by using a rainwater collector with conic shape and a surface area of 1.0 m^2.

Finally, and during the days corresponding to the highest presence of anthropogenic radionuclides in the lower atmosphere over Spain, different types of organic samples were collected: grass samples and broad-leaf vegetables (chard and spinach), as well as cow's and goat's milk and cheese. All of the above were acquired using the following protocol: a) The plants were collected on different dates, between 28th March and middle April. The grass was cut very short (to ground level) over controlled areas of known dimensions. The vegetables were bought from local farmers in areas located within the vicinity of the station, b) The cow's and goat's milk and the goat cheese samples were obtained from farms relatively close to the station on different dates between 28th March and 18th April, but with the animals feeding differently. In the case of cows, in most cases their diet consisted of commercial feed and they

had not grazed, while in the case of goats, their diet consisted only of fresh pasture. The cow cheese samples were purchased at different markets in Seville produced on known dates by a local food company.

2.2. Sample preparation and Measurements

Due to the very short time that had elapsed between sampling and the corresponding analyses, in order to provide data to the Spanish authorities as quickly as possible, preparation of the samples was done as simply and as fast as possible.

The high-volume air filters were folded and pressed to obtain surface areas of 10×10 cm^2, equal to the geometry of the standards used for the gamma spectrometer calibration. The vegetable and grass samples were chopped finely, without washing, and then directly measured without drying. In the case of milk samples, they were measured directly in a Marinelli beaker (1 dm^3), as well as the rainwater samples (without evaporation). The cheese samples were measured directly after adapting them to the calibrated geometry of the corresponding gamma spectrometer.

All the samples were analysed by low-level gamma-ray spectrometry using one of the two following detectors: a) a extended range (XtRa) germanium detector, made by Canberra (model GX4020), with a volume crystal of 160 cm^3 (relative efficiency 37,8%), and b) a REGe detector with relative efficiency of 30%. All the detectors were shielded with 10 cm of o lead, while the Xtra system was additionally equipped with an anti-coincidence device in order to increase their sensitivity.

The reported uncertainties (coverage factor k=2, unless another coverage factor is indicated) for gamma spectrometry were derived from counting statistics and uncertainties in the mixed gamma-ray reference solution. The Sevilla laboratory has participated in several national and international intercomparison exercises involving the measurement of different environmental matrices by gamma-ray spectrometry with very satisfactory results.

3.- Results and Discussion

3.1.- Aerosol filters

The first clear detection of Fukushima released radioisotopes in Seville were made in the high-volume filter collected during the period 21-28 March, At the time when the highest concentrations of artificial radionuclides in the atmosphere were over Seville, it was possible to detect the presence of ^{131}I, ^{137}Cs, ^{134}Cs, ^{136}Cs, and ^{132}Te.

The greatest ^{131}I concentration was detected for the sampling period 28-29 March, and a similar temporal pattern was found for other isotopes released from the Fukushima accident, for instance ^{137}Cs. As can be seen in Table 1, the highest activity concentrations always correspond to ^{131}I due to its high volatility. The highest activity concentrations of ^{132}Te and ^{136}Cs were 240 and 34 $\mu Bq/m^3$, corresponding to the air filter collected in the period 28-29 March. These last nuclides were only detectable in the filters collected until the end of March.

After the maximum reached the 28-29 of March, two plateaus covering the temporal intervals 29 March- 2 April and 3 -9 April were observed (with ^{131}I concentrations ranging between 650-850 $\mu Bq/m^3$ and 200-400 $\mu Bq/m^3$, respectively). These plateaus can be associated to the stable and static atmospheric conditions prevailing during the mentioned dates, with high pressures over the Iberian Peninsula.

In addition to the measurement of independent radionuclides, a study was made of their ratios,

looking for some additional information about the characteristics of the release. The $^{134}Cs/^{137}Cs$ activity ratio remained almost constant during the studied period, with a value close to unity. A ratio of this magnitude leads to an estimate of an average fuel burn-up of the order of 25 000 Mwd/TU, which is consistent with the evaluation provided by TEPCO that considers an average burn-up of Unit 2 of 23000 MWd/tU (Stohl et al., 2011).

Another activity ratio studied was $^{136}Cs/^{137}Cs$. This ratio was less than 0.3, in agreement with the ratios found at other international stations such as those in the Ro5 (ring of five) European network (Masson et al., 2011). This low value would normally imply that the radionuclides come from fuel several days after shut-down (Stohl et al., 2011). This could indicate some release from the Unit 4 spent fuel pool (SFP) with contained a mixture of fresh and old fuel.

Date Start sampling	Date Finish Sampling	Volume (m^3)	^{131}I ($\mu Bq/m^3$)	^{137}Cs ($\mu Bq/m^3$)	^{134}Cs ($\mu Bq/m^3$)
10:00 7 March	9:45 14 March	95503	N.D.	N.D.	N.D.
10:00 14 March	9:45 21 March	101662	2.10 ± 0.25	N.D	N.D.
10:00 21 March	9:45 28 March	90851	620 ± 12	34 ± 3	32 ± 2
10:00 28 March	9:45 29 March	16500	1740 ± 40	300 ± 18	290 ± 12
10:00 29 March	9:45 30 March	16148	766 ± 23	129 ± 5	129 ± 7
10:00 30 March	9:45 31 March	17133	644 ± 20	53 ± 4	49 ± 4
10:00 31 March	9:45 1 April	16222	820 ± 26	70 ± 10	68 ± 5
10:00 1 April	9:45 2 April	16097	857 ± 25	89 ± 5	90 ± 5
10:00 2 April	9:45 3 April	19000	359 ± 17	65 ± 10	68 ± 6
10:00 3 April	9:45 4 April	16433	401 ± 32	75 ± 10	82 ± 6
10:00 4 April	9:45 5 April	16595	254 ± 19	43 ± 4	50 ± 4
10:00 5 April	9:45 6 April	15645	446 ± 19	26 ± 4	27 ± 3
10:00 6 April	9:45 7 April	14147	274 ± 21	32 ± 9	40 ± 4
10:00 7 April	9:45 8 April	15004	239 ± 16	16 ± 5	24 ± 10
10:00 8 April	9:45 9 April	14850	204 ± 18	14 ± 4	15 ± 4
10:00 9 April	9:45 11 April	30426	40 ± 6	6 ± 1	7 ± 1
10:00 11 April	9:45 13 April	35200	61 ± 7	8 ± 2	7 ± 2
10:00 13 April	9:45 15 April	27841	37 ± 3	N.D.	N.D.
10:00 15 April	9:45 18 April	38083	30 ± 2	9 ± 1	8 ± 1

Table 1.- *Activity concentrations of ^{131}I, ^{137}Cs and ^{134}Cs ($\mu Bq/m^3$) determined in filters collected at the Seville station*

3.2 Gaseous ^{131}I

The temporal evolution of the ^{131}I activity concentration in gaseous form can be deduced from the data collected in Table 2. The uncertainty estimates do not consider external physical processes that might increase their values such as the particle to gas transformation in the prior filter of the charcoal cartridge, but comparing the gaseous ^{131}I results with the results compiled in Table 1, it seems evident that a majoritary fraction of the ^{131}I which reaches Seville from the Fukushima accident was in gaseous form.

(It is necessary to mention that due to the small volumes pumped with the gaseous iodine sampler, the limit of detection in the gaseous ^{131}I determinations is quite high, several hundred $\mu Bq/m^3$).

Date Start Sampling	Date Finish Sampling	Volume (m^3)	^{131}I gas ($\mu Bq/m^3$)
10:00 14 March	**9:45 21 March**	**250**	**N.D.**
10:00 21 March	**9:45 28 March**	**245**	**2000 ± 185**
10:00 28 March	**9:45 31 March**	**114**	**8310 ± 660**
10:00 31 March	**9:45 4 April**	**156**	**4590 ± 380**
10:00 4 April	**9:45 8 April**	**148**	**N.D.**
10:00 8 April	**9:45 13 April**	**182**	**N.D.**

Table 2.- *Activity concentrations of gaseous ^{131}I (mBq/m^3) at Seville, determined through charcoal canister measurements.*

3.3 Fallout

Table 3 gives the results for rainfall collected at the Seville station and at rural area 60 km far away from the town. During the period in which the plume containing radioisotopes from the Fukushima accident was detected over Seville, there was limited rainfall. For that reason, the data are quite sparse.

In all the cases, the gamma spectrometric determinations detected only one radionuclide, ^{131}I, in volumetric concentrations in the range 0.27-2.06 Bq/L. Neither ^{137}Cs nor ^{134}Cs were detected (minimum detectable activity in the range 0.1-0.2 Bq/L). The corresponding wet-deposition measurements indicated that the total wet-fallout of ^{131}I at the three stations was limited (clearly less than 10 Bq/m^2). The wet deposition levels found were well below the values which could be the cause for any radiological concern.

Wet-fallout		
Station	**Date**	**^{131}I (Bq/m^2)**
Sevilla	29 March	**3.30 ± 0.27**
Sevilla	3 April	**3.65 ± 9,38**
Rural area 50 km Seville	3 April	**2.97 ± 0.27**
Dry-fallout		
Station	**Date**	**^{131}I (Bq/m^2)**
Sevilla	26-28 March	< 3.0
Sevilla	28-29 March	< 2.6
Sevilla	30- 1 April	< 1.4
Sevilla	1-3 April	<1.8

Table 3.- *Wet- and dry ^{131}I fallout (Bq/m^2) determined in Seville at dates when traces the contaminated plume originated in the Fukushima accident were affecting the South-West of Spain*

The analyses of ^{131}I in the dry deposition at the Seville station (4 independent samples) give no value above the detection limit (see also Table 3). It is then evident from comparison of the wet and dry depositions and their associated limits of detection that, as was to be expected, a major proportion of the fallout was deposited in rainfall rather than as dry fallout. This allows us to state that the total fallout (dry + wet) of ^{131}I over South-West of Spain did not surpass the value of 20 Bq/m^2. However, due to the possibility that the ^{131}I deposited on the filter and that collected in the rainwater could have partially been transformed to gaseous ^{131}I due to its complex chemical dynamics, and moved back into the atmosphere, the latest measurements

need to be interpreted with caution and as minimum concentrations.

For ^{137}Cs, one can estimate a total deposition value in South-West of Spain due to the Fukushima accident of at least a factor of ten lower than for ^{131}I. This is not significant compared with the global accumulated ^{137}Cs fallout from the nuclear weapons tests performed mostly during the sixties, which, decay corrected to current dates, is of the order of 10^3 Bq/m^2 (Sanmiguel et al., 2003).

The order of magnitude of the total ^{131}I fallout over Seville estimated through direct measurements of the wet and dry depositions was confirmed by means of an indirect set of measurements: undisturbed grass growing in a part of the gardens belonging to the Physics Faculty were collected. On 30 March, just after the observation of the main peak of particulate ^{131}I airborne concentrations in Spain, grass from the lawn that had been growing for more than one week was cut in a defined area, and a representative sample was analyzed by gamma spectrometry. The value determined for the ^{131}I mass activity concentration was 9.5 ± 0.8 Bq/kg wet weight, corresponding to an ^{131}I accumulation of 2.1 ± 0.2 Bq/m^2. Taking into consideration the less than 100% effectiveness of the lawn in retaining the deposited material, this accumulation is coherent with the wet deposition in the area up to the date of collection of the sample.

Also detected in the undisturbed lawn grass were ^{134}Cs and ^{137}Cs. A total accumulation of 0.05 Bq/m^2 of ^{134}Cs and 0.09 Bq/m^2 of ^{137}Cs was found. The presence of detectable amounts of ^{134}Cs is indicative of the impact of the Fukushima accident on the compartment analyzed, while the ^{137}Cs/^{134}Cs ratio clearly greater than unity indicates that the ^{137}Cs in the lawn has another source in addition to the Fukushima accident, i.e., the deposition of some re-suspended material including ^{137}Cs originating from nuclear weapon tests, because the Chernobyl accident had a negligible impact in the south-west of Spain, the area in which the lawn grass analyzed was collected. Lawn samples collected at the sample place the 4th and the 12th of April give ^{131}I activity concentrations of 4.8 and 2.8 Bq/kg wet weight in good concordance with the observed decrease of the ^{131}I activity concentrations in the troposphere and the quite short half-life of this radionuclide,

3.4. Impact on the trophic chain

Table 4 lists the range of activity concentrations detected for ^{131}I in the different types of vegetables collected in Seville at different dates. ^{131}I was the only radionuclide with origin in the Fukushima accident detected in these samples.

Vegetables		
Type	**Collection date**	**^{131}I (Bq/kg w,.w)**
Chards	**28 March**	**1.42 ± 0.22**
Chards	**3 April**	**0.36 ± 0.06**
Chards	**4 april**	**1.05 ± 0.15**
Chards	**9 april**	**0.32 ± 0.05**
Spinachs	**7 April**	**0.78 ± 0.09**
Spinachs	**11 April**	**0.31 ± 0.03**
Spinachs	**12 April**	**0.10 ± 0.02**

Table 4.- *^{131}I activity concentrations (Bq/kg w.w.) determined in different types of vegetabklles collected in the vicinity of Sevilla station.*

These obtained levels should be compared with the maximum permitted levels of radioactive

contamination of foodstuffs and feedstuffs following a nuclear accident in Europe (EURATOM, 1989), that for vegetables and ^{131}I are 2000 Bq/kg w.w. In any case, the maximum levels detected in the analyzed samples were some orders of magnitude below the aforementioned EURATOM limits.

A conclusion, in addition to the very weak levels detected, can be drawn from the results obtained in the vegetables: the maximum levels of ^{131}I were detected in both locations around 29 March for all the plant types, with a clear and monotonic decrease after this date, in clear correlation with the evolution of the ^{131}I activity concentrations in the troposphere of Sevilla. The ^{131}I contamination of the vegetables is due to foliar deposition and attachment of aerosols (aerial route) with concentration factors in the order of 10^3.

Milk		
Type	**Collection date**	**^{131}I (Bq/L)**
Goat milk	**1 April**	**1.04 ± 0.10**
Goat milk	**4 April**	**1.11 ± 0.11**
Goat milk	**7 April**	**0.73 ± 0.07**
Goat milk	**9 april**	**0.40 ± 0.05**
Goat milk	**14 April**	**0.38 ± 0.04**
Cow milk	**28 March**	**< 0.08**
Cow milk	**29 March**	**< 0.18**
Cow milk	**1 April**	**< 0.20**
Cow milk	**4 April**	**< 0.14**
Cow milk	**6 April**	**< 0.15**
Cow milk	**8 April**	**<0.10**
Cow milk	**15 April**	**< 0.10**

Table 5.- *^{131}I activity concentrations (Bq/L) in cow and goat milk collected in the vicinity of the Sevilla station.*

Table 5 presents the range of ^{131}I activity levels found in cow's and goat's milk samples collected in the location of Seville at different dates. These results show that the ^{131}I levels detected in cow's milk in no case surpassed the procedure's detection limit, LID. This may be at least partially explained by diet type. The cows under study were feeded in most cases with commercial preserved feed and they had not grazed fresh pasture.

On the contrary, detectable ^{131}I levels were observed in all the goat's milk samples analyzed. This is in agreement with the type of diet of the goats in comparison with that of the cows (see Section 2.1), and also with the known fact that the daily proportion of ^{131}I intake which is secreted in milk is one order of magnitude greater for sheep and goats than for cows (Howard et al., 1993). Because of this high transfer factor, goat's milks are regarded as key indicators with which to monitor or evaluate contamination after nuclear accidents. The levels detected in all the milk samples analyzed have no radiological significance because the maximum level found for ^{131}I is at least two orders of magnitude below the limit established in Europe for the maximum permitted levels of radioactive contamination in milk following a nuclear accident in Europe (EURATOM, 1989). Only in the goat´s milk sample collected the 4th of April it was possible to detect ^{134}Cs with an activity concentration as low as 0.19 ± 0.05 Bq/L.

If we analyze the temporal evolution of the ^{131}I levels detected in the goat's milk samples collected in Seville, it can be additionally deduced that the timing of the maximum levels does not coincide with the maximum levels found in aerosols. There is a slight lag in the maximum corresponding to the goat's milk samples, due to the path followed by the anthropogenic radionuclides from the troposphere until their incorporation into the milk.

Finally, we have also analyzed the radioactive content in different cheese samples purchased in the vicinity of Seville station and produced during the days when the Fukushima plume passed over Spain. The only radionuclide detected was ^{131}I, the levels measured being 0.85 ± 0.07 and 0.17 ± 0.04 Bq/kg (wet weight) in goat's and cow's cheese, respectively. The conclusion to be derived from these results is evident: the higher concentrations were found in the goat's cheese as corresponded to the higher levels that had been found in this milk type.

4. Conclusions

Detectable levels of some of the anthropogenic gamma-emitters (^{131}I, ^{137}Cs, ^{134}Cs, ^{136}Cs and ^{132}Te) originating from the Fukushima accident have been measured in the troposphere over Spain. The characteristic activity ratios encountered for the contaminated plume which reached Spain are a $^{134}Cs/^{137}Cs$ close to unity, and a ^{131}I gaseous/^{131}I particulate ratio greater than unity.

The highest volumetric activity concentrations in aerosols were observed for ^{131}I (maximum concentrations of the order of mBq/m^3), with the ^{137}Cs and ^{134}Cs concentrations being at least one order of magnitude lower. Consequently, the associated fallout has had negligible radiological consequences in Spain. Indeed, the only radionuclide detected in key components of the trophic chain (vegetables, milk) was ^{131}I at levels of no radiological significance because they were several orders of magnitude lower than the maximum permitted levels of radioactive contamination of foodstuffs and feedstuffs following a nuclear accident in Europe (EURATOM, 1989).

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References

Diaz Leon, J., Jaffe, D.A., Kaspar, J., Knecht, A., Miller, M.L., Robertson, R.G.H., Schubert A.G., 2011. Arrival time and magnitude of airborne fission products from the Fukushima, Japan, reactor incident as measured in Seattle, WA, USA. J. Environ. Radioactiv. 102, 1032-1038

EURATOM, 1989. Reglamento EURATOM Nº 2218/89 del Consejo, de 18 de Julio de 1989, por el que se modifica el Reglamento (Euratom) nº 3954/87. (in Spanish).

Howard, B.J., Beresfold, N.A., Mayes, R.W., Lamb, C.S., 1993. Transfer of ^{131}I to sheep milk from vegetation contaminated by the Chernobyl accident. J. Environ. Radioactiv. 19, 155-161..

IRSN, 2011a. Évaluation de la radioactivité rejeteé par la centrale de Fukushima Daiichi (Fukushima I). Report 22 march 2011.

IRSN, 2011b. Synthèse des resultats des mesures de la radioactivité dans le cadre de la surveillance de l´impact à très longue distance des rejects de l`accident de Fukushima. Report 26 march 2011.

Masson, O., Baeza, A., Bieringer, J., Brudecki, K., Bucci, S., Cappai, M., Carvalho, F. P., Connan, O., Cosma, C., Dalheimer, A., Didier, D., Depuydt, G., De Geer, L.E., De Vismes, A., Gini, L., Groppi, F., Gudnason, K., Gurriaran, R., Hainz, D., Halldórsson, Ó., Hammond, D., Hanley, O., Holeý, K., Homoki, Zs., Ioannidou, A., Isajenko, K., Jankovic, M., Katzlberger, C., Kettunen, M., Kierepko, R., Kontro, R., Kwakman, P.J.M., Lecomte, M., Leon Vintro, L., Leppänen, A.-P., Lind, B., Lujaniene, G., Mc Ginnity, P., Mc Mahon, C., Malá, H., Manenti, S.,

Manolopoulou, M., Mattila, A., Mauring, A., Mietelski, J.W., Møller, B., Nielsen, S.P., Nikolic, J., Overwater, R.M.W., Pálsson, S.E., Papastefanou, C., Penev, I., Pham, M.K., Povinec, P.P., Ramebäck, H., Reis, M.C., Ringer, W., Rodriguez, A., Rulík, P., Saey, P.R.J., Samsonov, V., Schlosser, C., Sgorbati, G., Silobritiene, B. V., Söderström, C., Sogni, R., Solier, L., Sonck, M., Steinhauser, G., Steinkopff, T., Steinmann, P., Stoulos, S., Sýkora, I., Todorovic, D., Tooloutalaie, N., Tositti, L., Tschiersch, J., Ugron, A., Vagena, E., Vargas, A., Wershofen, H., Zhukova, O., 2011. Tracking of Airborne Radionuclides from the damaged Fukushima Dai-ichi nuclear reactors by European networks. Environ. Sci. Technol. 45, 7670-7677

Sanmiguel, E.G., Bolivar, J.P., García-Tenorio, R., 2003. Mixing, sediment accumulation and focusing using Pb-210 and Cs-137. J. Paleolimnol. 29, 1-11.

Stohl, A., Seibert, P., Wotawa, G., Arnold, D., Burkhart, J. F., Eckhardt S., Tapia, C., Vargas, A., Yasunari, T.J., 2011. Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition, Atmos. Chem. Phys. 11, 28319-28394 (http://www.atmos-chem-phys-discuss.net/11/28319/2011/acpd-11-28319-2011-discussion.html).

ZAMG (Zentralanstalt für Metereologie und Geodynamik), 2011. Accident in the Japanese NPP Fukushima: Large emissions of Cesium-137 and Iodine-131 / Spread Radioactiviy / weather currently favourable / low level radioactivity observed in Iceland. Report 24 March 2011.