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

In this work is shown the magnitude and temporal evolution of the radionuclide concentrations detected in the contaminated air masses with origin in the Fukushioma accident at their arrival to Seville (Spain). 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.

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 weight in broadleaf plants, with obviously negligible radiological implications.

Sampling Sites and Equipments

High-volume aerosol collector

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.

Rainwater collector. Scarce data because limited rain occurs during the arrival of the contaminated air-masses

High-volume air filter before and after aerosol collection

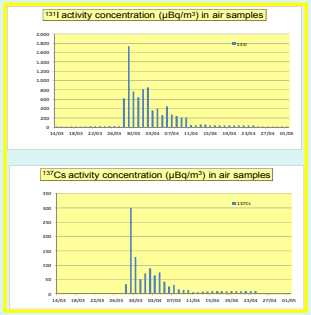
The high-volume aerosol collectors pumps in daily air volumes of 10000-16000 m^3 on polypropylene filters. A low-volume suction collectors was also used for the collection of gaseous ^{131}I in charcoal filters (daily volume of 30-50 m^3). On days without rain, the dry fallout was sampled using a custom-made polyethylene collector 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 a surface area of 1.0 m^2

Flock of goats grazing fresh pasture . Milk and cheese samples with origin in this flock were analyzed

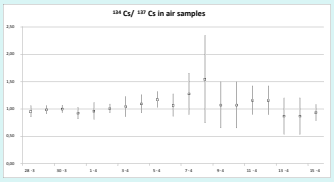
All the samples were analysed by low-level gamma-ray spectrometry using one of the two following detectors: a) a extended range (XtRa) germanium detector, made by Canberra (model GX4020), with a volume crystal of 160 cm^3 (relative efficiency 37,8%), and b) a REGe detector with relative efficiency of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

Results and Discussion

Aerosol Filters



The first detections 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, as ^{137}Cs. The highest activity concentrations of ^{132}Te and ^{136}Cs were 240 and 34 mBq/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.



The ^{134}Cs/^{137}Cs activity ratio was constant during the studied period, with a value close to unity. This ratio leads to an estimate of an average fuel burn-up of 25 000 Mwd/TU, which is consistent with the evaluation provided by TEPCO considering an average burn-up of Unit 2 of 23000.

The ^{136}Cs/^{137}Cs activity ratio in the aerosol filters 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. This low value would normally imply that the radionuclides come from fuel several days after shut-down

Gaseous ^{131}I

Date Start Sampling	Date Finish Sampling	Volume (m^3)	^{131}I gas (μBq/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.

A majoritary fraction of the ^{131}I which reaches Seville was in gaseous form. The ratio ^{131}I-gaseous/^{131}I-total rose over time, increasing from around 0.75 in the last days of March to 0.85 during the first days of April indicating that ^{131}I particulate deposition is not balanced by gas-to-particle conversion, and therefore particulate ^{131}I decreases faster than gaseous ^{131}I.

Fallout

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

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.

Impact on the trophic chain

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

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

^{131}I was the only radionuclide with origin in the Fukushima accident detected in the vegetable samples, with negligible radiological impact.. 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.

The levels detected in all the milk samples have no radiological significance with higher values in the goat milk due to the feeding habits (fresh pasture) and the high transfer food- milk for these animals. The temporal evolution of the ^{131}I levels in the milk show a well defined lag in comparison with the observed in the aerosols

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