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# Radioactivity from Fukushima nuclear accident detected in Lisbon, Portugal

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### ABSTRACT

The radioactivity released from the Fukushima Dai-ichi nuclear accident was transported around the globe by atmospheric processes. Several artificial radionuclides were detected and measured in aerosols and atmospheric surface depositions in the Lisbon area during late March and early April 2011. The highest concentrations measured in aerosols were those of particulate <sup>131</sup>I, 1.39  $\pm$  0.08 mBq m<sup>-3</sup>. Cesium-134, <sup>137</sup>Cs and <sup>132</sup>Te were also determined but at lower concentrations. The total atmospheric depositions on the ground were higher on the first week of April with values for <sup>131</sup>I, <sup>134</sup>Cs, and <sup>137</sup>Cs of 0.92  $\pm$  0.11, 0.59  $\pm$  0.06, and 0.62  $\pm$  0.12 Bq m<sup>-2</sup>, respectively. The four artificial radionuclides measurable, <sup>131</sup>I, <sup>132</sup>Te <sup>134</sup>Cs, and <sup>137</sup>Cs, caused little radiation exposure to the members of the public, that was five orders of magnitude lower than the ionizing radiation effective dose limits for members of the public for one year (1 mSv y<sup>-1</sup>).

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## 1. Introduction

Following the earthquake that occurred on March 11, 2011 and the subsequently generated tsunami, the east coast of Japan suffered one of the major natural catastrophes of the last one hundred years. Amongst the impacts of this natural catastrophe, serious damage was caused to the Fukushima Dai-ichi nuclear power plant, triggering hydrogen explosions in reactor buildings and releases of radioactivity into the environment on the following days. As reported by the Japanese Government to the IAEA on June 2011, the total amount of radioactivity released into the atmosphere might have been around  $1.5 \times 10^{17}$  Bq of  $^{131}$ I and  $1.2 \times 10^{16}$  Bq of  $^{137}$ Cs, and the total radioactivity discharged into the sea might have been around  $4.7 \times 10^{15}$  Bq (Report of Japanese Government, 2011).

Artificial radionuclides such as <sup>133</sup>Xe, <sup>131</sup>I, <sup>134</sup>Cs, and <sup>137</sup>Cs were detected in the atmosphere, in Japan as well as on the west coast of the United States and transported by the westerly winds over the North Atlantic ocean, reaching Europe (Hosoda et al., 2011; IRSN, 2011; Leon et al., 2011; Manolopoulou et al., 2011; Masson et al., 2011; Pittauerová et al., 2011).

The airborne radioactivity is continuously monitored in Lisbon, Portugal, at the Atlantic edge of the European continental landmass by an aerosol monitoring station. Following the Fukushima nuclear accident additional atmospheric sampling and radioactivity monitoring was performed. This paper gives an account of the results of these radionuclide measurements in the Lisbon area, and an assessment of the radiation dose exposure to members of the public.

#### 2. Materials and methods

Surface air was sampled with a high volume sampler (F&J Specialty Products Inc., USA), placed 1.5 m above the ground in an open field at Sacavém (38° 48′ 50″ N, 09° 05′ 38″ W), near Lisbon. Filters used were of borosilicate glass microfiber FP47M, 10 cm in diameter, and replaced every two days. The air pump digital controller maintained a continuous volume measurement at normal pressure and temperature (NPT), and an average flow of around 60 m<sup>3</sup> h<sup>-1</sup> over the sampling period. This aerosol sampling procedure allowed highly efficient sampling, >98% for sub-micron aerosols, of airborne particulate radionuclides, (HI-Q Environmental Products Company). However, radioactive iodine from nuclear releases into the atmosphere is known to be present in particulate and gaseous forms, and glass microfiber filters do not retain gaseous radioiodine (Masson et al., 2011).

Sample filters were removed from the pump holder and placed in Petri dish plastic polyethylene containers, with constant geometry, and immediately analyzed with a broad energy range HPGe detector (Canberra BEGe model BE5030) with an active volume of 150 cm<sup>3</sup> and a carbon window. The software Genie 2k (Canberra) was used in spectral analysis. The detector was calibrated in efficiency by using a radioactive standard customized in epoxy resin

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Filter n.	Beginning of sa	Beginning of sampling		End of sampling		Average	Activity concentration Bq m <sup>-3</sup>				
	Date (d-m-y)	Hour (h:min)	Date (d-m-y)	Hour (h:min)	filtered (m <sup>3</sup> )	flux (m <sup>3</sup> h <sup>-1</sup> )	Be-7	I-131	Te-132	Cs-134	Cs-137
F8	17-03-2011	14:05	18-03-2011	15:50	1.64E+03	63.78	4.00E-03 ± 5.40E-04	n.d.	n.d.	n.d.	n.d.
F10	18-03-2011	15:55	21-03-2011	09:20	3.02E+03	46.14	$4.43\text{E-03} \pm 5.30\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F11	21-03-2011	09:25	23-03-2011	10:20	2.74E+03	55.86	$4.02\text{E-03} \pm 5.00\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F12	23-03-2011	10:26	25-03-2011	16:15	2.99E+03	55.56	$2.43\text{E-03} \pm 2.30\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F13	25-03-2011	16:17	28-03-2011	09:53	4.14E+03	64.56	$3.19E-03 \pm 2.20E-04$	$1.05\text{E-03} \pm 6.40\text{E-05}$	$6.60\text{E-}05 \pm 2.20\text{E-}05$	$6.50\text{E-}05 \pm 1.10\text{E-}05$	$6.50\text{E-}05 \pm 1.10\text{E-}05$
F14	28-03-2011	09:54	30-03-2011	10:00	3.18E+03	65.82	$2.84\text{E-03} \pm 2.40\text{E-04}$	$1.39\text{E-03} \pm 8.40\text{E-05}$	$6.20\text{E-}05 \pm 2.50\text{E-}05$	$1.53E-04 \pm 1.90E-05$	$1.39\text{E-04} \pm 1.70\text{E-05}$
F15	30-03-2011	10:01	31-03-2011	09:10	1.57E+03	68.58	$2.32\text{E-03} \pm 2.40\text{E-04}$	$8.35\text{E-04} \pm 7.30\text{E-05}$	n.d.	$7.90\text{E-}05 \pm 1.40\text{E-}05$	$9.60\text{E-}05 \pm 1.60\text{E-}05$
F17	31-03-2011	09:11	02-04-2011	16:55	3.39E+03	60.90	$3.15E-03 \pm 2.30E-04$	$3.88\text{E-04} \pm 3.10\text{E-05}$	n.d.	$4.90\text{E-}05 \pm 1.20\text{E-}05$	$4.10\text{E-}05 \pm 9.70\text{E-}06$
F18	02-04-2011	16:56	04-04-2011	09:51	2.80E+03	68.46	$2.34\text{E-03} \pm 2.30\text{E-04}$	$3.30\text{E-}04 \pm 3.10\text{E-}05$	n.d.	$2.90\text{E-}05 \pm 1.10\text{E-}05$	$2.30\text{E-}05 \pm 1.20\text{E-}05$
F19	04-04-2011	09:52	06-04-2011	09:30	2.74E+03	57.42	$3.97E-03 \pm 3.20E-04$	$2.42E-04 \pm 3.20E-05$	n.d.	$3.57E-05 \pm 7.30E-06$	$3.92E-05 \pm 8.10E-06$
F20	06-04-2011	09:31	08-04-2011	16:40	2.50E+03	45.48	$4.90\text{E-03} \pm 3.40\text{E-04}$	$2.08\text{E-}04 \pm 2.90\text{E-}05$	n.d.	$2.79E-05 \pm 7.10E-06$	$2.13E-05 \pm 7.30E-06$
F21	08-04-2011	16:42	11-04-2011	09:40	3.92E+03	60.36	$3.34E-03 \pm 2.10E-04$	$4.60\text{E-}05 \pm 1.20\text{E-}05$	n.d.	n.d.	n.d.
F22	11-04-2011	09:45	13-04-2011	09:00	3.18E+03	48.96	$2.93\text{E-03} \pm 1.80\text{E-04}$	$3.10\text{E-}05 \pm 1.10\text{E-}05$	n.d.	n.d.	n.d.
F23	13-04-2011	09:00	15-04-2011	16:00	3.04E+03	46.80	$4.51E-03 \pm 3.20E-04$	$2.50\text{E-}05 \pm 1.50\text{E-}05$	n.d.	n.d.	n.d.
F24	15-04-2011	16:00	18-04-2011	09:10	3.77E+03	57.48	$4.54\text{E-03} \pm 2.80\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F25	18-04-2011	09:05	20-04-2011	09:05	3.29E+03	50.70	$2.76E-03 \pm 2.20E-04$	n.d.	n.d.	n.d.	n.d.
F26	20-04-2011	09:10	26-04-2011	09:45	6.31E+03	_	$2.20\text{E-03} \pm 1.54\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F27	Sampling inter	ruption (bre	eakdown of sam	pler)			n.a.	n.a.	n.a.	n.a.	n.a.
F28	27-04-2011	09:45	29-04-2011	15:15	1.92E+03	36.00	$6.35\text{E-03} \pm 5.12\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F29	29-04-2011	15:30	02-05-2011	11:50	2.46E+03	36.00	$5.71\text{E-03} \pm 4.53\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F30	02-05-2011	12:00	04-05-2011	10:30	1.68E+03	36.00	$4.54\text{E-03} \pm 4.06\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F31	04-05-2011	10:35	06-05-2011	15:33	1.90E+03	36.00	$6.40\text{E-03} \pm 5.42\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F32	06-05-2011	15:40	09-05-2011	09:30	2.36E+03	36.00	$2.56\text{E-03} \pm 2.36\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F33	09-05-2011	09:30	12-05-2011	10:00	1.74E+03	36.00	$7.53\text{E-}03 \pm 5.88\text{E-}04$	n.d.	n.d.	n.d.	n.d.
F34	12-05-2011	10:00	14-05-2011	15:00	1.91E+03	36.00	$8.48\text{E-03} \pm 7.07\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F35	14-05-2011	15:00	16-05-2011	12:00	2.60E+03	37.98	$7.44\text{E-03} \pm 5.60\text{E-04}$	n.d.	n.d.	n.d.	n.d.
F36	16-05-2011	12:00	18-05-2011	10:20	1.76E+03	37.98	$5.92E-03 \pm 4.89E-04$	n.d.	n.d.	n.d.	n.d.
Mean + 1SD							(4.34 + 1.79)E-03				

Table 1	
Radionuclide activity concentrations in aerosol particulates measured at Sacavém, Lisbon.	

n.a., not analyzed; n.d., not detected; Minimum Detectable Activity (MDA) values: <sup>7</sup>Be 2.6E-04 Bq m<sup>-3</sup>, <sup>131</sup>I 3.0E-05 Bq m<sup>-3</sup>, <sup>132</sup>Te 4.4E-05 Bq m<sup>-3</sup>, <sup>134</sup>Cs 2.6E-05 Bq m<sup>-3</sup>, <sup>137</sup>Cs 3.0E-05 Bq m<sup>-3</sup>.

matrix in the Petri dish container, customized by Eckert & Ziegler Isotope Products, Inc. This radioactive standard ranged in energy from 46.5 keV to 1836 keV and is traceable to the National Institute of Standards and Technology (NIST, USA). The software GESPECOR (Sima et al., 2001) was used for correcting coincidence summing effects (<sup>131</sup>I, <sup>134</sup>Cs) and for calculating the efficiency transfer from the calibration filter geometry to the measurement filter geometry. This procedure was validated through good results in proficiency tests organized by the International Atomic Energy Agency (IAEA, 2009). Minimum detectable activity (MDA) was computed for each radionuclide of interest.

Atmospheric total (wet plus dry) depositions were collected with a 1 m<sup>2</sup> cross-section area surface collector, with the shape of an inverted pyramid trunk permanently open and pouring through the vertices into a polyethylene drum placed underneath the collector. Atmospheric depositions were collected each 8 days, and at the end of every sampling period the surface of the collector was washed with HCl acidified distilled water (pH = 1) in order to ensure complete recovery of dry depositions from the collector surface. The total deposition collected in the drum was transferred to 1.0 dm<sup>3</sup> capacity polyethylene Marinelli beakers and counted on an HpGe detector (Canberra Model GC4019). Efficiency calibration for this geometry was performed with a radioactive standard prepared in the same Marinelli beakers used for samples, and ordered from Eckert & Ziegler Isotope Products, Inc.

## 3. Results and discussion

The gamma emitting radionuclides of artificial origin detected on filters were  $^{131}I(T_{1/2}=8.0\,d)\,^{134}Cs\,(T_{1/2}=2.06\,y),\,^{137}Cs\,(T_{1/2}=30.0\,y),$  and  $^{132}Te\,(T_{1/2}=3.26\,d)\,(Table\,1).$  Be-7  $(T_{1/2}=53.3\,d)$  of cosmogenic natural origin is also reported. Those radionuclides were firstly detected on March 25, 2011 and attained maximum activity concentration values on March 29. The highest concentrations

recorded were those of particulate  $^{131}I$  at 1.39  $\pm$  0.08 mBq m $^{-3}$ . Concentrations of  $^{134}Cs$  and  $^{137}Cs$  were always lower, with maximal concentrations around 150  $\mu$ Bq m $^{-3}$ , and those of  $^{132}Te$  even lower and at up to 66  $\mu$ Bq m $^{-3}$ . Other gamma emitting fission products were specifically looked for, such as  $^{129m}Te$ , I,  $^{135}Cs$  and  $^{136}Cs$  but could not be detected. As an indication of detection limits, the MDAs for  $^{129m}Te$  and  $^{136}Cs$  were about 2.64  $\times$  10 $^{-04}$  and 3.05  $\times$  10 $^{-05}$ Bq m $^{-3}$ , respectively. In surface air  $^{131}I$  was measurable during about 3 weeks from

In surface air <sup>131</sup>I was measurable during about 3 weeks from March 25 to April 15, and lasted longer than detection of cesium isotopes. Cs-134 and <sup>137</sup>Cs were detected for about 2 weeks with <sup>134</sup>Cs/<sup>137</sup>Cs activity concentration ratios averaging nearly 1 during that time (Fig. 1).

Particulate <sup>131</sup>I collected on filters was only part of the total <sup>131</sup>I present in the air, such as observed during the Chernobyl and Fukushima nuclear accidents (Masson et al., 2011). In the radioactivity from Fukushima, the ratio of gaseous to total over Europe averaged 0.772  $\pm$  0.136 based on pooled measurements of both <sup>131</sup>I forms, with a noticeable constant value both geographically as well as at altitude (Masson et al., 2011). Therefore, the total <sup>131</sup>I concentration in the air mass over Lisbon would have been nearly four times the particulate <sup>131</sup>I concentration measured on filters, i.e., at the peak it would have reached about 5.6 mBq m<sup>-3</sup>. Cesium has a different chemical behavior and most, or even all the radioactivity associated with cesium isotopes was attached to aerosol particulates.

The concentrations of these artificial radionuclides measured in surface air were consistently low and, for example, were generally lower than the concentrations of <sup>7</sup>Be, a gamma emitting radionuclide of natural origin averaging  $4.34 \pm 1.79$  mBq m<sup>-3</sup> in surface air during the same period (Table 1).

The integrated exposure from March 25 to April 15 of members of the public to artificial radionuclides present in surface air of the Lisbon area was computed for the inhalation pathway using the



Fig. 1. Radionuclides determined in air filters at Sacavém, Lisboa, following the Fukushima Dai-ichi nuclear accident. Arrows indicate the collection date of atmospheric deposition samples.

dose conversion factors recommended by the International Atomic Energy Agency (IAEA, 1996) and applying an inhalation rate of 22 m<sup>3</sup> per day for an adult person. Through inhalation, the four artificial particulate radionuclides determined on filters accounted for a committed effective dose of 2.4 nSv ( $2.4 \times 10^{-09}$  Sv). Assuming, based on measurements made in Europe on both iodine forms in the atmosphere that the activity concentration of gaseous <sup>131</sup>I was four times the particulate <sup>131</sup>I, the integrated effective dose from total <sup>131</sup>I would increase the committed effective dose from 2.4 to about 6.8 nSv (Table 2). Both <sup>131</sup>I forms (gaseous + particulate) contributed with 87%, and <sup>137</sup>Cs plus <sup>134</sup>Cs with 12% to the estimated total radiation dose exposure caused by artificial radionuclides inhaled with surface air.

The radiation dose due to inhalation of artificial radionuclides released by the Fukushima nuclear accident added to the background radiation dose from inhaled naturally-occurring radionuclides. For comparison, the committed effective radiation dose to members of the public through inhalation of the average <sup>7</sup>Be activity concentration measured on the same filters was 1.9 nSv  $y^{-1}$ . Other naturally-occurring radionuclides, such as the long lived radon daughters  $^{210}\text{Pb}\,(T_{1/2}=22.3\,\text{y})$  and  $^{210}\text{Po}\,(T_{1/2}=138.4\,\text{d})$  are present in the aerosol particulate fraction in the Lisbon area in annual average concentrations of 181  $\pm$  111  $\mu$ Bq m<sup>-3</sup>, and 94  $\pm$  73  $\mu$ Bq m<sup>-3</sup>, respectively (Carvalho, 1995a,b). The inhalation of these naturallyoccurring radionuclides contributed to a committed effective dose of about 8.1  $\times$  10<sup>+03</sup> nSv y<sup>-1</sup> and 3.2  $\times$  10<sup>+03</sup> nSv y<sup>-1</sup> to an adult member of the public. Radon gas ( $^{222}$ Rn, T<sub>1/2</sub> = 3.8 d) in surface air in the Lisbon area displayed average concentrations of 1-10 Bg m<sup>-3</sup> in outdoor air and gives rise to even higher radiation doses (Carvalho, 1995a). The whole body committed effective dose by inhalation of artificial and several naturally-occurring radionuclides are compared in Table 2.

During the air monitoring period in Sacavém, the atmospheric total depositions of  $^{131}$ l,  $^{134}$ Cs, and  $^{137}$ Cs on the ground were 0.92  $\pm$  0.11, 0.59  $\pm$  0.06, and 0.62  $\pm$  0.12 Bq m $^{-2}$ , respectively

(Table 3). These radioactive depositions took place during the first week after the arrival of the airborne radioactive materials from Fukushima nuclear accident. In the following weeks no further radioactive depositions were detectable, both in periods of dry deposition only and in periods with rainfall (Fig. 1). Other gamma emitting fission products such as <sup>129m</sup>Te, <sup>132</sup>I, <sup>135</sup>Cs and <sup>136</sup>Cs could not be detected in the atmospheric depositions by the applied methods.

The deposition of <sup>131</sup>I on grasslands and pastures,  $0.92 \pm 0.11$  Bq m<sup>-2</sup> in the Lisbon area determined with the surface collector, was also lower than recorded in other locations in Europe (Beresford et al., 2012). An estimate of <sup>131</sup>I that could be transferred to milk of outdoor grazing cattle led to insignificant activities, much lower than 1 Bq dm<sup>-3</sup> (IAEA, 2010). Similar results were generally achieved in other European regions (Beresford et al., 2012). Consumption of leafy vegetables grown in the Lisbon area, assuming 100% interception of radioactive fallout by plant leaves followed by full transfer of deposited radionuclides to consumers with vegetable intake, could lead to a worst case scenario (although unlikely) dose of near 39 nSv, using IAEA recommended dose conversion factors for the ingestion pathway (IAEA, 1996). A more realistic assessment would place this dose at around 10% (4 nSv) of that value. For comparison, the ingestion of <sup>210</sup>Pb and <sup>210</sup>Po naturally-occurring radionuclides in the average diet of the Portuguese population originates a dose rate of about  $3.2 \times 10^{+02}$  nSv and  $5.2 \times 10^{+05}$  nSv, respectively (Table 2).

The total radiation dose exposure caused by radionuclides from Fukushima, through combined inhalation and ingestion pathways for adult members of the public, is estimated at about 10 nSv, and at up to 46 nSv in the worst case scenario. This radiation dose exposure can be compared with the 1 mSv per year of dose limit for radiation doses from radiation practices to members of the public (EURATOM, 1996).

Due to their short half-lives, deposited <sup>131</sup>I, <sup>132</sup>Te, and even <sup>134</sup>Cs did not persist long time in the environment. The longer lived

Table 2

Committed effective radiation doses through inhalation pathway computed for adult members of the public in the Lisbon area, assuming 22 m<sup>3</sup> air inhaled per day, and radiation dose though ingestion pathway assuming full transfer of deposited radionuclides to the consumer.

Radionuclide	Average concentration in surface air Bq m <sup>-3</sup>	Inhalation in the period	Inhalation dose conversion factor (Sv $Bq^{-1}$ )	Effective dose nSv
Artificial radionuclides				
I-131 particulate		25-3-2011 to 15-4-2011	7.4E-09	1.5 <sup>a</sup>
I-131 particulate + gaseous				5.9 <sup>b</sup>
Te-132 particulate		25-3-2011 to 15-4-2011	2.00E-09	0.01
Cs-134 particulate		25-3-2011 to 15-4-2011	2.00E-08	0.4
Cs-137 particulate		25-3-2011 to 15-4-2011	3.90E-08	0.5
All particulate only				2.4
All particulate + gaseous				6.8
Natural radionuclides				
Be-7 particulate	$(4.34 \pm 1.79)$ E-03	One year	5.5E-11	1.9
Pb-210 particulate	$(181 \pm 111)$ E-06	One year	5.6E-06	8.1E+03
Po-210 particulate	$(94\pm73)$ E-06	One year	4.3E-06	3.2E+03
Radionuclide		Ingestion in the period	Ingestion dose conversion factor (Sv $Bq^{-1}$ )	Effective dose nSv
Artificial radionuclides	Deposition Bq $m^{-2}$			
I-131	9.20E-01	25-3-2011 to 13-4-2011	2.20E-08	20
Te-132	n.d.	25-3-2011 to 13-5-2011		
Cs-134	5.91E-01	25-3-2011 to 13-5-2011	1.90E-08	11
Cs-137	6.20E-01	25-3-2011 to 13-5-2011	1.30E-08	8
All above				39
Natural radionuclides	Intake with diet Bq d $^{-1}$			
Be-7		One year	2.80E-11	
Pb-210	0.47 <sup>c</sup>	One year	6.90E-07	3.2E+02
Po-210	1.2 <sup>c</sup>	One year	1.20E-06	5.2E+05

<sup>a</sup> Particulate I-131 (measured).

<sup>b</sup> Estimated total I-131 (particulate + gaseous)

c Carvalho, 1995b

Table 3				
Radioactivity in total atm	ospheric depositions at Sac	avém, Lisbon area, in the	e weeks following the	nuclear accident of Fukushima.

Sampling period		Type of deposition during the period	Deposited activity (Bq m <sup>-2</sup> )				
Beginning (d-m-y)	End (d-m-y)		Be-7	I-131	Cs-134	Cs-137	
11-03-2011	04-04-2011	Dry	$5.99E{+}01 \pm 4.00E{+}00$	9.20E-01 ± 1.10E-01	$5.91\text{E-}01 \pm 6.30\text{E-}02$	6.20E-01 ± 1.20E-01	
04-04-2011	11-04-2011	Dry	$8.00E{+}00\pm1.10E{+}00$	n.d.	n.d.	n.d.	
11-04-2011	19-04-2011	Dry + Wet ( $R = 30 \text{ Lm}^{-2}$ )	$5.31E{+}01 \pm 1.77E{+}01$	n.d.	n.d.	n.d.	
19-04-2011	26-04-2011	Dry + Wet ( $R = 30 \text{ Lm}^{-2}$ )	< 3.75E + 01	n.d.	n.d.	n.d.	
26-04-2011	02-05-2011	Dry + Wet ( $R = 16 \text{ Lm}^{-2}$ )	$2.46E{+}01 \pm 8.48E{+}00$	n.d.	n.d.	n.d.	
02-05-2011	09-05-2011	Dry + Wet ( $R = 1.6 \text{ Lm}^{-2}$ )	$4.13E{+}00 \pm 9.92E{-}01$	n.d.	n.d.	n.d.	
09-05-2011	16-05-2011	Dry	$3.00E{+}00 \pm 1.00E{+}00$	n.d.	n.d.	n.d.	
16-05-2011	23-05-2011	Dry + Wet ( $R = 14.6 \text{ Lm}^{-2}$ )	$1.49E{+}01 \pm 7.76E{+}00$	n.d.	n.d.	n.d.	
23-05-2011	30-05-2011	Dry + Wet ( $R = 30 \text{ Lm}^{-2}$ )	<3.39 E+01	n.d.	n.d.	n.d.	

n.d., not detected; R, rainfall collected in the sampling period.

radionuclide <sup>137</sup>Cs has the potential to stay in soils for many years. Before the Fukushima nuclear accident, <sup>137</sup>Cs was present already in the surface layer of soils throughout the country as a result of radioactive fallout deposition from nuclear tests in the sixties and from the Chernobyl nuclear accident in April 1986, and its activity concentrations ranged from 50 to 250 Bq kg<sup>-1</sup> (unpublished results). The <sup>137</sup>Cs activity with origin in Fukushima and deposited on the ground in the Lisbon area, 0.62  $\pm$  0.12 Bq m<sup>-2</sup>, is about 1.6  $\times$  10<sup>-2</sup> % of the Cs inventory existing already in soils of this area. Therefore, deposition of <sup>137</sup>Cs from Fukushima nuclear accident is unlike to add a detectable enhancement of <sup>137</sup>Cs in soils of Lisbon area as well as of other regions in the country.

Following the Fukushima nuclear accident, the activity values of artificial radionuclide depositions determined at Sacavém were much lower than recorded after the Chernobyl accident in 1986 in the same area. Following the Chernobyl accident, the highest depositions measured in the grass at Sacavém were  $3.8 \pm 0.2$  Bq m<sup>-2</sup> and  $6.8 \pm 1.6$  Bq m<sup>-2</sup> of <sup>131</sup>I and <sup>137</sup>Cs, respectively, and in cow milk the maximum concentrations determined in continental Portugal were 1.5 Bq dm<sup>-3</sup> and 2 Bq dm<sup>-3</sup> for <sup>131</sup>I and <sup>137</sup>Cs, respectively (LNETI, 1991).

## 4. Conclusions

The radioactivity from the Fukushima Dai-ichi nuclear accident, triggered by the earthquake and tsunami of March 11, 2011, was detected in the surface air near Lisbon 14 days later, and indeed 11 days only after the steam venting and explosions that took place at Dai-ichi Unit 3 on March 14. Airborne artificial radioactivity in the Lisbon area could be measured for a period of about 3 weeks, although activity concentrations remained low. The four artificial radionuclides measurable, <sup>131</sup>I, <sup>132</sup>Te <sup>134</sup>Cs, and <sup>137</sup>Cs, originated little radiation exposures to the members of the public, estimated at about 10 nSv, and remained five orders of magnitude lower than the ionizing radiation effective dose limits for members of the public for one year (1 mSv y<sup>-1</sup>). Radionuclide activity concentrations recorded in the surface air in Lisbon area were of the same order of magnitude as recorded in other cities of central and southern Europe during the same period and slightly lower than measured in monitoring stations at the northern European countries (Masson et al., 2011).

The deposition of the short lived isotope <sup>131</sup>I, usually of great concern due to known impact on public health (Bard et al., 1997), was low and unlikely to originate meaningful contamination of grazing cattle feeding in the open air, including milk and beef production. The longer lived isotopes <sup>134</sup>Cs and <sup>137</sup>Cs were deposited also onto the ground carried by dry and wet atmospheric depositions. However, the total activity deposited was not sufficient to generate a recognizable Fukushima <sup>137</sup>Cs signal in soils, as the activity deposited was much lower than <sup>137</sup>Cs from the Chernobyl accident and radioactive fallout from previous nuclear tests.

From the radiological point of view the exposure of members of the public to the radioactivity originating from the nuclear accident of Fukushima, in Japan, and transported to the Lisbon area by atmospheric processes was very low, and with no measurable impact on public health.

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