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Radioactive impact of Fukushima accident on the Iberian Peninsula: Evolution and plume previous pathway

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ABSTRACT

High activity concentrations of several man-made radionuclides (such as ^{131}I, ^{132}I, ^{132}Te, ^{134}Cs and ^{137}Cs) have been detected along the Iberian Peninsula from March 28th to April 7th 2011. The analysis of back-trajectories of air masses allowed us to demonstrate that the levels of manmade radionuclide activity concentrations in the southwest of the Iberian Peninsula come from the accident produced in the nuclear power plant of Fukushima. The pathway followed by the radioactive plume from Fukushima into Huelva (southwest of the Iberian Peninsula) was deduced through back-trajectories analysis, and this fact was also verified by the activity concentrations measured of those radionuclides reported in places crossed by this radioactive cloud. In fact, activity concentrations reported by E.P.A., and by IAEA, in several places of Japan, Pacific Ocean and United States of America are according to the expected ones from the air mass trajectory arriving at Huelva province.

1. Introduction

On March 11th, 2011, an earthquake happened in the Pacific Ocean (9 degrees on the Richter Scale), east coast of Japan (epicenter 38.32°N, 142.37°E), and 130 km from Sendai (capital city of Miyagi Prefecture, Japan), which caused a tsunami that hit the east coast of Japan.

There are 55 operating nuclear power reactors in Japan, the majority of them being located close to the coast. Several of these plants were affected by this earthquake and the tsunami, which caused an extensive damage in the nuclear plant of Fukushima–Daiichi, whose reactors were seriously affected. Posterior explosions inside the reactors injected fission products (radionuclides) into the atmosphere. These radionuclides are quickly bound to the atmospheric aerosols (Gaffney et al., 2004), and later they can travel big distances around the World with the air masses thanks to the large-scale circulation of the atmosphere, going up to be detected in places far away from their sources (Dueñas et al., 2011; Masson et al., 2010).

A similar episode of man-made radionuclides was produced by the nuclear accident occurred in Chernobyl (Ukraine) in 1986, which caused serious ecological problems and also contaminated a large part of the higher latitudes of the northern hemisphere (Bell and Shaw, 2005). This fact reveals the relationship between the meteorological conditions and the evolution of radionuclide concentrations measured (Erlandsson and Isaksson, 1988; Mattsson and Vesanen, 1988). In this sense, the wind dynamics plays a key role in transport and dispersion (Salazar et al., 1994).

The analysis of back-trajectories has been found to be a valuable tool to indentify the origin of air masses arriving at the aerosol sampling sites. This tool has been often used in several fields as air pollution, attempting to identify the source regions of atmospheric pollutants by estimating the paths of air parcels arriving into the region of interest (Borge et al., 2007; Brankov et al., 1998; Petzold et al., 2009; Sánchez-Ccoyllo et al., 2006). This analysis enables us to know the pathway followed by an air mass over a previous time period, giving a better understanding about its origin, horizontal displacement and circulation altitude (Hondula et al., 2009; Jorba et al., 2004).

Taking into account the facts previously commented, the main objective of this work has been to justify the reasons of the activity concentrations for several man-made radionuclides (such as ^{131}I, ^{132}I, ^{132}Te, ^{134}Cs and ^{137}Cs) detected in the Iberian Peninsula from March 28th to April 7th 2011, through the special synoptic-scale air mass circulation pattern appended during this time period.

2. Materials and methods

2.1. Study area

The monitoring sampling network used in this study comprises three aerosol sampling stations located in the Southwest of the Iberian

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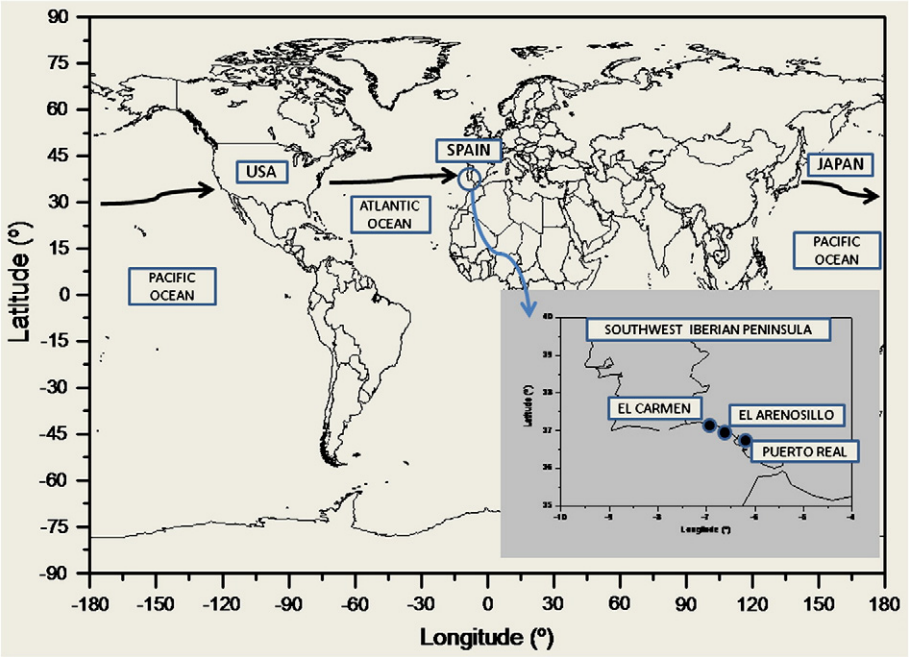


Fig. 1. Location of Iberian Peninsula in the world, and monitoring stations defined as El Arenosillo, Puerto Real, and El Carmen.

Peninsula (Fig. 1): 1) "El Carmen" (CRM), in Huelva city, 2) "Puerto Real" (PRE), close to Cadiz city, and 3) "El Arenosillo" (ARE), close to the National Park of Doñana.

"El Carmen" sampling station is located at the Campus of the University of Huelva (37.28° N, −6.91° W), in the city of Huelva (population ~150,000 inhabitants). The characteristics of the surrounding area allow us to classify this site as an urban sampling station affected by a very close chemical industrial complex (3 km and more). "Puerto Real" sampling station is located at the Campus of the University of Cádiz in Puerto Real village (36.53° N, −6°19° W), and sited very close to the Metropolitan Area of the Bay of Cádiz city (population ~650,000 inhabitants), and for that this sampling station is also considered as industrial site. "El Arenosillo" station (37.10° N, −6.73° W) is sited in a fairly flat area far from important population nucleus, 1 km from the coastline, and for that it is considered as a reference station (natural background or baseline). "El Arenosillo" is located 35 km Southeast of the city of Huelva, and 5 km from the National Park of Doñana (the main natural protected area in the Western Mediterranean region), and being this one considered as the reference station in the Spanish Air Quality Network (Adame et al., 2010). In all sampling stations the air samples were collected at about 12 m above ground.

In addition, activity concentrations measured by E.P.A. and by IAEA have been reported, in sampling sites located in Japan, the Pacific Ocean and the United States of America, according to the expected ones from the air mass trajectory arriving at Huelva province.

2.2. Samplings

In each sampling station, the aerosol samples (PM10, grain size <10 μm) were collected onto quartz fiber filters (QF20 Schleicher & Schuell, 25.4 cm × 20.3 cm) with high volume samplers working at a flow rate of 40 cfm (68 $m^3\ h^{-1}$). The sampling period studied in this work ranges from March 15th 2011 to April 17th 2011, with a sampling time of 24 h during the episode and 48 h for the remaining samplings. Prior to the aerosol sampling, the filters are weighed several times until the mass is constant. The filter weighing was carried out with a precision of 0.1 mg inside a chamber with controlled humidity. After sampling, the filter is newly weighted at the same atmospheric conditions to avoid humidity effects.

2.3. Radionuclide determinations

In order to calculate the activity concentrations, aerosol filters are cut into two halves, and both weighed accurately. One half was wrapped, pressed in a plastic bag and measured by gamma spectrometry. The other half was totally dissolved with a mixture of strong acids (65% HNO_3, 37% HCl, 40% HF). The final solution was used for alpha emitters determination as ^{210}Po, U-isotopes, and Th-isotopes. The results on alpha emitters are not discussed in this paper. A detailed description of pretreatment for air filters can be found elsewhere (Lozano et al., 2011a).

Table 1
Activity concentrations (mBq/m3) in Huelva City station (CRM).

Date	^{131}I	^{132}I	^{132}Te	^{134}Cs	^{137}Cs	^{134}Cs/^{137}Cs
15/17 –03–11	<0.030	<0.027	<0.017	<0.026	<0.032	–
21/23 –03–11	<0.030	<0.027	<0.017	<0.026	<0.032	–
28/29 –03–11	3.69 ± 0.19	0.162 ± 0.013	0.175 ± 0.018	0.88 ± 0.05	0.95 ± 0.05	0.93 ± 0.07
29/30 –03–11	0.78 ± 0.04	<0.027	<0.017	0.078 ± 0.016	0.093 ± 0.014	0.84 ± 0.21
30/31 –03– 11	0.82 ± 0.07	<0.027	<0.017	0.096 ± 0.021	0.114 ± 0.013	0.84 ± 0.21
31–03–11/1–04–11	1.31 ± 0.11	<0.027	<0.017	0.153 ± 0.022	0.175 ± 0.024	0.87 ± 0.17
01/04 –04–11	1.19 ± 0.10	<0.027	<0.017	0.112 ± 0.016	0.131 ± 0.017	0.85 ± 0.17
05/07 –04– 11	<0.030	<0.027	<0.017	0.044 ± 0.007	0.051 ± 0.008	0.86 ± 0.19
08/11 –04–11	<0.030	<0.027	<0.017	<0.026	<0.032	–
15/17 –04–11	<0.030	<0.027	<0.017	<0.026	<0.032	–

Table 2
Radionuclide activity concentrations (mBq m^{-3}) in several places around the world after the Fukushima accident (a: data taken from IAEA (2011); b: data taken from US E.P.A. (2011)).

Place (latitude, longitude)	Sampling dates	^{131}I	^{132}I	^{132}Te	^{134}Cs	^{137}Cs	Reference
Tokyo (Japan) (35.70°N, 139.72E)	March 15–March 19	5300	5200	–	1200	1200	a
Takasaki (Japan) (36.50°N, 138.97°E)	March 15–March 19	3000	2200	5400	1400	3000	a
Saipan Northern Mariana Islands (USA) (15.18°N, 145.76°E)	March 21–March 24	0.33 to 29	ND	0.3 to 3.7	0.21 to 0.25	0.15 to 1.5	b
Anaheim California (USA) (33.83°N, −117.88°W)	March 18–March 22	2.9 to 6.3	0.67 to 0.81	0.70 to 0.81	0.28 to 0.63	0.055 to 0.30	b
Montgomery Alabama (USA) (32.22°N, −86.27°W)	March 24–March 31	2.0 to 5.5	ND	ND–3.5	ND	ND–0.067	b
Orlando Florida (USA) (28.55°N, −81.37°W)	March 24	7.4	0.444	0.888	0.555	0.703	b
El Arenosillo Huelva (Spain) (37.10°N, −6.73°W)	March 29−April 7	0.07 to 0.80	ND	ND	ND–0.084	ND–0.127	This work
Puerto Real Cádiz (Spain) (36.53 N, −6°19°W)	March 29–April 7	0.07 to 0.63	ND	ND	0.045 to 0.052	0.079 to 0.120	This work

Gamma measurements of the atmospheric filters were performed with an XtRa coaxial Ge detector (Canberra), with 38% relative efficiency, FWHM of 0.95 keV at the 122 keV (^{57}Co), and 1.9 keV at the 1333 keV (^{60}Co). The detector is coupled with a set of standard electronic components, including a multichannel analyzer, and it is surrounded with 15 cm thick Fe. Details on the efficiency calibration for air filters are given elsewhere (Martínez-Ruiz et al, 2007). ^{131}I, ^{132}I, ^{132}Te, ^{134}Cs, ^{137}Cs activities were determined through their following photon emissions: 364.48 keV (Pγ 81.2%), 667.69 keV (Pγ 98.7%), 228.16 keV (Pγ 88.0%), 604.7 keV (Pγ 97.6%), and 661.66 keV (Pγ 85.2%), respectively (Kocher, 1981). Background levels for ^{131}I, ^{132}I, ^{132}Te, ^{134}Cs and ^{137}Cs activity concentrations measured from 2004 to 2011 in Huelva were always below the MDA values (Table 1).

2.4. Air masses back-trajectory analysis

In order to calculate the set of back-trajectories during the radioactive episode period, the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model, Version 4, has been used. This model was developed by the NOAA's Air Resources Laboratory (ARL) (Draxler and Hess, 1998; Draxler et al., 2009). The GDAS meteorological files generated from the Global Data Analysis System model, and maintained by the ARL have been used as data input. GDAS files have a temporal resolution of three hours, with a data gridded of $1° \times 1°$ in latitude and longitude, including upper level information in 23 pressure levels.

Kinematic 3D-back-trajectories have been calculated using the vertical wind component information given by GDAS meteorological files. The capability of these files to determine the air mass arrival over this area has been validated through meteorological soundings (Hernández-Ceballos et al., 2011). Trajectory duration of eight days has been considered since this time interval is enough for the influence of synoptic air flows to be adequately represented. In order to understand the behavior of the air masses circulating in lower levels, hourly back-trajectories at three different heights have been calculated at 500 m, 1000 m and 1500 m over Huelva city (37.28° N, −6.91° W) from 26th March to 6th April.

In addition, 8-days back-trajectories at Washington DC (38.89° N, −77.02° W) were calculated at the same heights from 18th to 29th March to check the connection between the air flows from the Pacific Ocean and air masses over the east coast of North America, where activity concentrations were previously measured by E.P.A. This city has been taken as reference station of the east coast of North America, although the E.P.A monitoring data only show values above background in the southeast coast (Montgomery, Jacksonville, Orlando). However, due to the atmospheric dispersion phenomenon, there may be found values above background over Washington.

Once calculated the set of back-trajectories for Washington DC and Huelva city, the daily average path (centroid) was calculated for each day of the period over each city. Each centroid was calculated using 24 hour back-trajectories per day, so, each one represents the 8-day backward dynamic pattern over both sites. This technique has been used to comprise and understand the information inside each set of back-trajectories calculated for both sites.

3. Results and discussion

3.1. Activity concentrations

Table 1 shows the activity concentrations at El Carmen sampling site (Huelva City). As it can be observed from this table, the highest activity concentrations of fission products were obtained on 28th March. After this date, the activity concentrations decreased and remained approximately constant during the following days until reaching values below detection limits. As it can also be observed from this table, the activity concentrations are generally below the Minimum Detectable Activity from 4 April, which have been calculated according to Currie (1968, 1995). These results are in agreement with the change in the wind direction occurring during days 4–5 April. Additionally, the obtained $^{134}Cs/^{137}Cs$ activity ratios in Huelva province (Table 1) are similar to those ones obtained in the area of Japan several days after the Fukushima accident (Table 2).

The temporal evolution of both ^{131}I and ^{137}Cs concentrations in CRM station (Huelva city) are shown in Fig. 2, in order to have a better understanding of this radioactive episode. From this figure, it can be observed that ^{131}I and ^{137}Cs activity concentration levels in Huelva city remained below the detection limits during the previous days of the Fukushima accident. Once the radioactive plume arrived in Huelva, the maximum activity concentrations were obtained during days 28 and 29 of March. After 5 April, once the wind direction changed and another air mass arrived here, the activity concentrations decreased to background levels.

In order to study the routes followed by the air masses from Fukusima (Japan) to reach Spain, the analysis of the spatial and temporal evolution of the radioactive cloud was carried out.

Table 2 shows the radionuclide activity concentrations of interest in different locations crossed by the radioactive plume originated in Fukushima. As it can be expected, the highest values for radionuclide concentrations are obtained in areas close to Fukushima (March 12th–19th). Days after the accident, the cloud traveled through the Pacific Ocean, being detected in Saipan (Commonwealth of the Northern Mariana Islands, USA). The data reported from this station correspond to March 21st to 24th. In the following days, the radioactive cloud reached the west coast of the United States (USA) (Anaheim, California, USA). Later, the cloud continued up to reach

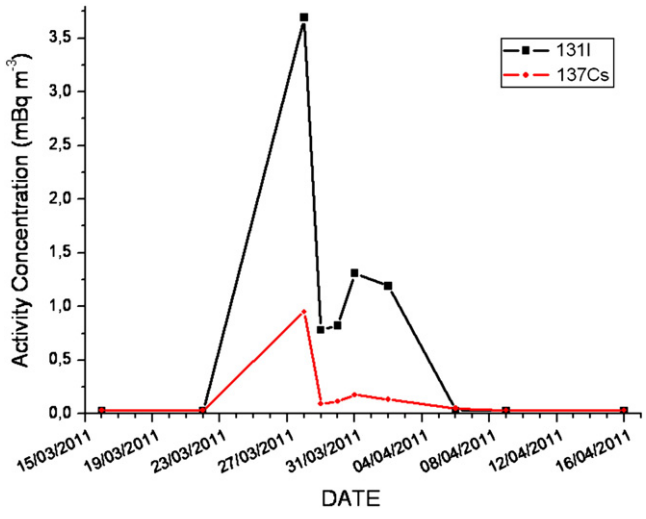


Fig. 2. Temporal evolution of ^{131}I and ^{137}Cs in Huelva City (CRM).

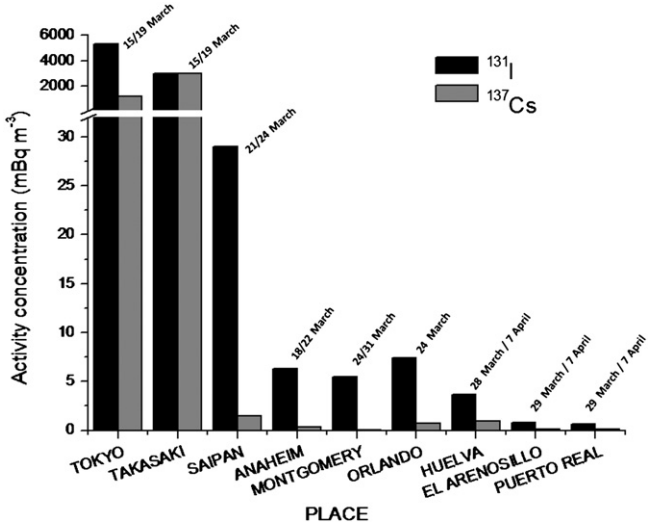


Fig. 3. Temporal evolution of the maximum ^{131}I and ^{137}Cs activity concentration registered in the different monitoring sites around the world after Fukushima nuclear accident.

the east coast of the USA, being detected on March 24th in Montgomery (Alabama, USA) and Orlando (Florida, USA). Thus, the cloud continued into the east crossing the Atlantic Ocean and reaching the Iberian Peninsula approximately on March 28th when the highest values of radionuclide levels were obtained, as it can be seen in Table 1 ("El Carmen" station in Huelva), and in "The Arenosillo" and "Puerto Real" (Table 2). It is worth noting that radionuclide determination in aerosol samples in Huelva province have been carried out for more than 4 years (Lozano et al, 2011b) and only natural occurring radionuclides (^{7}Be, ^{210}Pb, ^{210}Bi, ^{210}Po and ^{40}K) have been detected.

As it can be expected, the highest activity concentrations of the radionuclides determined in this study correspond to Japanese locations, them being three orders of magnitude higher than the ones obtained in North America and Europe. As time passes, the cloud expands and the activity concentrations decreased to values below the detection limits (0.03 mBq m^{-3}). Fig. 3 shows the maximum concentration of ^{131}I and ^{137}Cs recorded in the locations crossed by the radioactive cloud after the Fukushima accident. The dates of measurements are also indicated in this figure in order to infer the time travel between the different sites considered.

3.2. Back-trajectories analysis

To understand the temporal evolution of the activity concentrations from 26th March to 6th April measured in the study area over Huelva city and to justify the transport of radionuclides from Fukushima into the south western of the Iberian Peninsula, which has been indicated in the previous section, the dynamic of air masses has been studied over this area during this period. HSYPLIT model has been used to calculate back-trajectories at three different vertical heights, 500, 1000 and 1500 m over this sampling point.

Trying to minimize the uncertainty calculation associated to each trajectory (Stohl, 1998), due to the accumulating errors caused by the computation of trajectories with a large temporal coverage, the air mass analysis has been carried out in two steps. In a first step, hourly back-trajectories with a temporal coverage of eight days have been calculated over Huelva city (as representative site in the Southwest of Iberian Peninsula) from 26th March to 6th April. And secondly, taking into account that the corresponding radioactive episode comes from North East of the USA (see Table 2 and Fig. 4), the city of Washington DC was selected as a reference point of the east coast of North America to compute the hourly backward trajectories from 18th to 29th March with a temporal coverage of 8 days at 500, 1000 and 1500 m. Thus, joining both studies it was possible to analyze the dynamic of air mass over Huelva city with a temporal coverage of 16 days.

Once calculated the sets of back-trajectories over Washington DC and Huelva at 500, 1000 and 1500 m, the analysis of air mass pathway at each height revealed a similar backward circulation during the previous 8 days over each site respectively. This fact indicates a large coupling degree among these atmospheric layers from 18th to 29th March over Washington DC and from 26th March to 6th April over Huelva. Taking into account this vertical behavior, the set of back-trajectories obtained at 1000 m have been taken as reference to represent the circulation over both sites.

Once the vertical height was set, the corresponding daily centroids at 1000 m (average path of 24 hour trajectories) were calculated. Fig. 4 shows the centroids of the previous 8 days at 1000 m from 18th to 29th March over Washington DC (Fig. 4a), and from 26th March to 6th April over Huelva (Fig. 4b).

As it is shown in both figures, each daily centroid for Washington and Huelva has been consecutively referred with a letter from "a" to "l" to facilitate the connection between both. The circulation of 28th March over Huelva is taken as example to analyze this figure. The centroid of this day was referred by "c". This air mass developed a movement during the previous 8 days over the Atlantic Ocean and an inland origin at the east coast of North America on 20th March. To follow the journey of this air mass, the daily centroid of 20th March at Washington DC, "c", has to be selected. In this case, this centroid showed a displacement along North America with its origin over the Bering Sea, on 12th March. Joining both, it is observed that from 12th to 28th March, the circulation of air masses were predominant from the northeast of the Pacific Ocean to the Iberian Peninsula.

Analyzing the results obtained in Huelva city (Fig. 4b), the 8-day centroids showed a homogeneous westerly circulation from 26th March to 5th April. On 26th March the origin of air masses was located in the surroundings of the Iberian Peninsula, whereas from 27th March to 4th April the air masses over Huelva city had a large westerly displacement, with its origin located in the northeast inland areas of North America. On the other hand, during the last days of the analyzed period, the air mass circulations decreased their displacements, their origins being located inside the Atlantic Ocean (5th April) and the east of the Iberian Peninsula (6th April).

Therefore, and taking into account this air mass behavior over Huelva, it is possible to affirm that from 27th March to 4th April there was a connection between southwest of the Iberian Peninsula and the east coast of North America, due to the origin of these air masses being located inland of North America.

Fig. 4a shows the 8-day centroid obtained for Washington DC back-trajectories from 18th to 29th March at 1000 m. As it is observed, during this period there were two different wind patterns over the east coast of North America. From 18th to 24th March a westerly air mass circulation was detected over this area. During this period, the daily centroids of 19th and 20th March showed a westerly air mass circulation with its origin in the Bering Sea, over the NE Pacific Ocean. In this sense, considering the time covered of these air masses (8 days), the origin of these circulations coincided with the Japanese earthquake and, so, with damages in the nuclear plant of Fukushima. For this reason, we can suggest that this air mass behavior could be the responsibility of the radionuclide concentrations measured in the east coast of North America during the following days, as it is shown in Table 2. On the other hand, from 24th to 29th March northerly flows were predominated over this area during the previous 8 days.

By connecting the results obtained for the back-trajectories over both Huelva and Washington DC during the previous 16 days, a wind corridor from the Bering Sea to the Iberian Peninsula was observed in 27–29 March, coinciding with the maximum of concentrations measured in the southwestern Iberian Peninsula. After these days, the progressive change in the dynamics of air masses over the east coast of North America tended to break this wind channeling, despite the continuous arrival of westerly flow over the southwest of the Iberian Peninsula. This change happened in the same dates that a progressive decreasing in the radionuclides concentrations was observed.

As it is shown, the air mass analysis by centroids has allowed the confirmation of the wind corridor establishment between the northeast of the Pacific Ocean and the Iberian Peninsula during the period of maximum concentration measured in Huelva. However, in this case, the use of centroids limits a clear identification of the air mass location in the surroundings of Japan. In this sense, the centroid of 19th March is the closest to the nuclear plant, but it is originated in the Bering Sea. This fact is associated to the meaning of centroids, due to each one summarizing the daily variability of hourly air masses, and, so, not representing the full range of possible transport pathway.

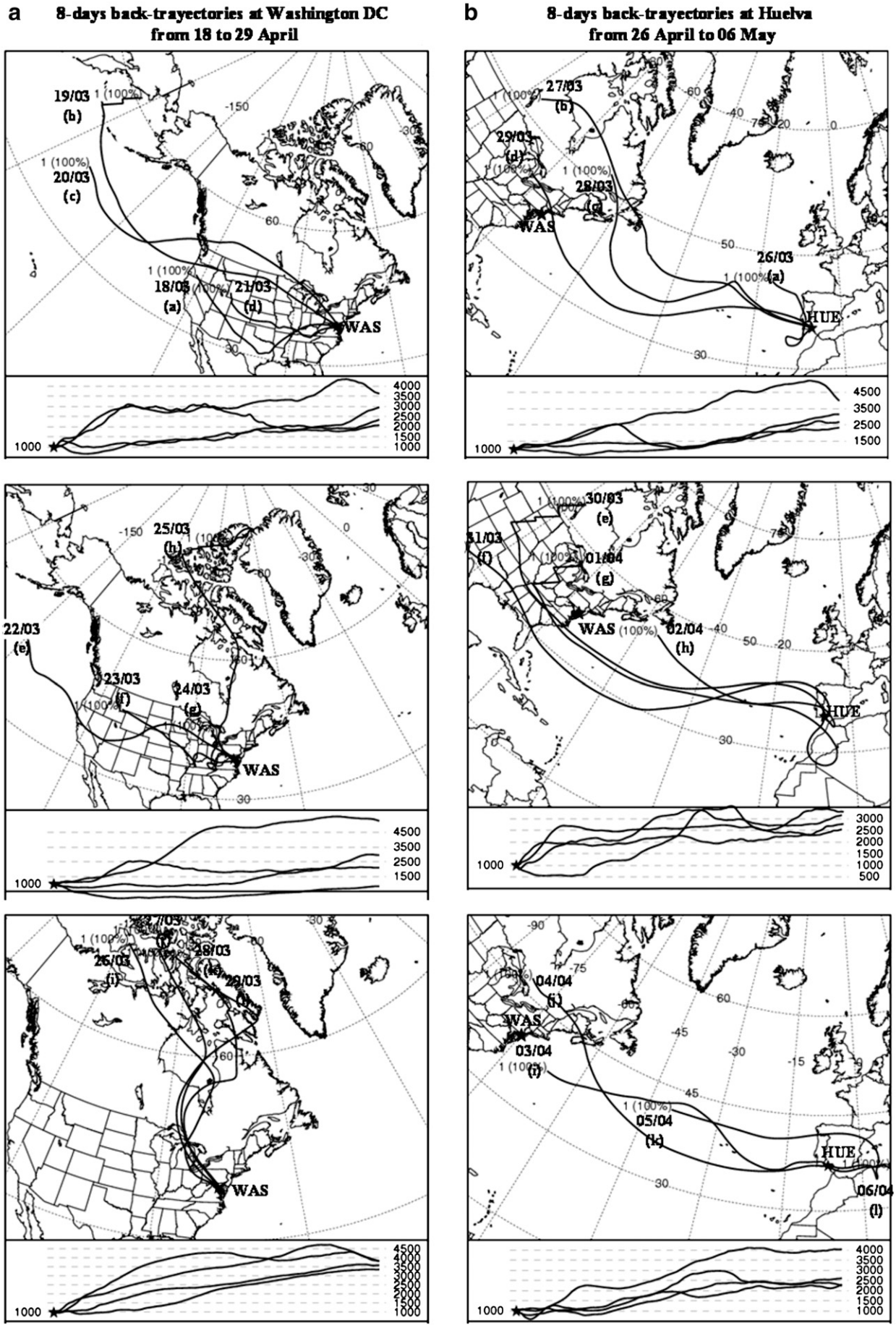
For this reason, to confirm the relationship between the Fukushima accident and the maximum concentrations measured in Huelva from 27th to 29th March, the hourly 3-D forward trajectories from Fukushima have been calculated from 11th to 13th March with a temporal coverage of 16 days.

Fig. 5 shows the set of air masses that connect the Fukushima area and the surroundings of the Iberian Peninsula during 11–13 March. This set of trajectories represents the 19.5% of the hourly forward trajectories computed between 11 and 13 March from Fukushima. The displacement of air masses over the Pacific Ocean, the United States and the Atlantic Ocean, and its arrival in the surrounding of the Iberian Peninsula has been observed. These air mass circulations during this period make possible radionuclides from Fukushima accident reach the Iberian Peninsula, and as a consequence, to confirm the origin of the activity concentrations in Huelva city.

4. Conclusions

In this paper the way the radioactive plume coming from the Fukushima accident has reached the South of Europe (Iberian Peninsula) has been studied and explained. In a first step the increase in air activity concentrations of radionuclides from nuclear fission,

Fig. 4. Horizontal and vertical daily centroid (average path of 24 hourly trajectories) during the previous 8 days at 1000 m at a) Washington from 18 to 29 of March and b) Huelva from 26 March to 6 April. The numbers of the centroids are the percentage of hourly trajectories used, and the identification number. Each daily centroid is consecutively referred with a letter from "a" to "l" in both cases.


a
8-days back-trayectories at Washington DC
from 18 to 29 April
b
8-days back-trayectories at Huelva
from 26 April to 06 May
19/03
(b)
20/03
(c)
18/03
(a)
21/03
(d)
WAS
HUE
27/03
(b)
29/03
(d)
28/03
(c)
26/03
(a)
25/03
(h)
22/03
(e)
23/03
(f)
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30/03
(e)
31/03
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01/04
(g)
02/04
(h)
26/03
(i)
28/03
(k)
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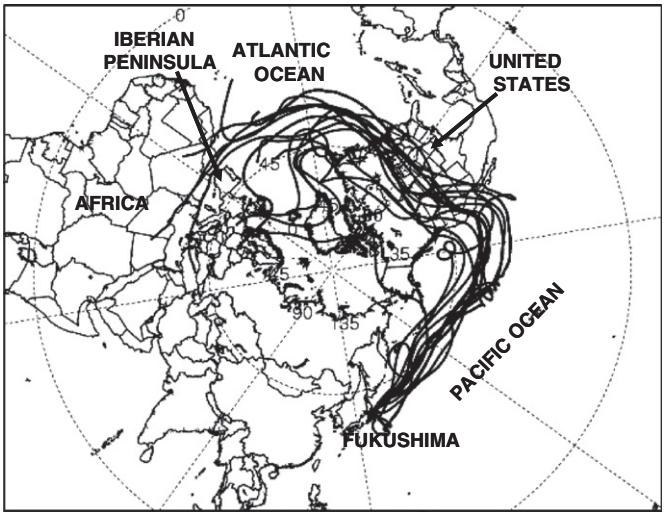

Fig. 5. 16-day hourly forward trajectories from Fukushima from 11 to 13 of March, representing the air mass connection between this area and the surroundings of Iberian Peninsula.

such as ^{131}I, ^{132}I, ^{132}Te, ^{134}Cs and ^{137}Cs in Huelva province has been shown. In a second step, and through back-trajectory analysis, it has been demonstrated that the radioactive plume producing the increase in those radionuclides is clearly coming from the geographical region of Japan (NE Pacific Ocean).

The analysis of back-trajectories has shown that the air mass over the South of the Iberian Peninsula during the radioactive episode came from the east of the USA (8 days before). Furthermore, this air mass was over the NE Pacific Ocean 8 days before. The activity concentrations of different radionuclides determined from aerosol samples collected in Huelva province show that the air mass of this Iberian radioactive episode passed throughout the Pacific Ocean–North America–the Atlantic Ocean until finally reaching the Southwestern Iberian Peninsula. As a final conclusion, it seems that the atmospheric radioactive plume from the Fukushima nuclear accident has extended first by the North Pacific Ocean, crossed the American continent, and reached Europe through the Iberian Peninsula.

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References

Adame JA, De la Morena BA, Bolívar JP. Surface ozone measurements in the southwest of the Iberian Peninsula (Huelva, Spain). Environ Sci Pollut Res 2010;17:355–68.
Bell JNB, Shaw G. Ecological lessons from the Chernobyl accident. Environ Int 2005;31:771–7.
Borge R, Lumbreras J, Vardoulakis S, Kassomeno P, Rodríguez E. Analysis of long-range transport influences on urban PM10 using two-stage atmospheric trajectory clusters. Atmos Environ 2007;41:4434–50. doi:10.1016/j.atmosenv.2007.01.053.
Brankov E, Trivikrama S, Steven P. A trajectory-clustering-correlation methodology of examining the long-range transport or air pollutants. Atmos Environ 1998;32:1525–34. DOI: 1325-2310/98.
Currie LLA. Limits for qualitative detection and quantitative determination: application to radiochemistry. Anal Chem 1968;40:586–93.
Currie LLA. Nomenclature in evaluation of analytical methods including detection and quantification capabilities. Pure Appl Chem 1995;67:1699–723.
Draxler RR, Hess GD. An overview of the HYSPLIT_4 modelling system for trajectories, dispersion, and deposition. Aust Meteorol Mag 1998;47:295–308.
Draxler RR, Stunder B, Rolph G, Taylor A. HYSPLIT_4 User's Guide, via NOAA.ARL. Silver Spring, MD: NOAA Air Resources Laboratory; 2009. Dec., 1997, revised Jan., 2009. Website http://www.arl.noaa.gov/documents/reports/HYSPLIT_user_guide.pdf.
Dueñas C, Orza JAG, Cabello M, Fernández MC, Cañete S, Pérez M, et al. Air mass origin and its influence on radionuclide activities (^{7}Be and ^{210}Pb) in aerosol particles at a coastal site in the western Mediterranean. Atmos Res 2011. doi:10.1016/j.atmosres.2011.02.011.
Erlandsson B, Isaksson M. Relation between the air activity and the deposition of Chernobyl debris. Environ Int 1988;14:165–75.
Gaffney JS, Marley N, Cunningham MM. Natural radionuclides in fine aerosols in the Pittsburgh area. Atmos Environ 2004;38:3191–200.
Hernández-Ceballos MA, Adame JA, Bolívar JP, De la Morena BA. Dynamic of air masses in southwestern the Iberian Peninsula from 1997 to 2007. Dyn Atmos Ocean 2011. (Under revision).
Hondula DM, Sitka L, Davis RE, Knight DB, Gawtry SD, Deaton ML, et al. A back-trajectory and air mass climatology for the Northern Shenandoah Valley, USA. Int J Climatol 2009. Published online www.interscience.wiley.com.
International Atomic Energy Agency (IAEA). http://documents.ccifj.or.jp/lm/documents_radiation/4_-_iaea_march_20th_briefing_short.pdf 2011.
Jorba O, Pérez C, Rocandenbosch F, Baldasano JM. Cluster analysis of 4-day back-trajectories arriving in the Barcelona Area (Spain) from 1997 to 2002. J Appl Meteorol 2004;43:887–901.
Kocher CK. Radioactive decay data tables. National Technical Information Service, U. S. Deparment of Commerce0-87079-124-9; 1981. DOE/TIC-11026.
Lozano RL, Bolívar JP, San Miguel EG, García-Tenorio R, Gázquez MJ. An accurate method to measure alpha-emitting natural radionuclides in atmospheric filters: application in two *norm* industries. Nucl Instrum Methods Phys Res A 2011a. (Under revision).
Lozano RL, San Miguel EG, Bolívar JP, Baskaran M. Depositional fluxes and concentrations of 7Be and 210Pb in bulk precipitation and aerosols at the interface of Atlantic and Mediterranean coast in Spain. J Geophys Res 2011b. (Under revision).
Martínez-Ruiz F, Borrego E, San Miguel EG, Bolívar JP. An efficiency calibration for ^{210}Pb and ^{7}Be measurements by gamma-ray spectrometry in atmospheric filters. Nucl Instrum Methods Phys Res A 2007;580:663–6.
Masson O, Piga D, Gurriaran R, D'Amico D. Impact of an exceptional Saharan dust outbreak in France: PM_{10} and artificial radionuclides concentrations in air and in dust deposit. Atmos Environ 2010;44(20):2478–86.
Mattsson S, Vesanen R. Patterns of Chernobyl fallout in relation to local weather conditions. Environ Int 1988;14:177–80.
Petzold A, Rasp K, Weinzierl B, Esselborn M, Hamburger T, Dornbrack A, et al. Saharan dust absorption and refractive index from aircraft-based observation during SAMUM 2006. Tellus 2009;618:118–30. doi:10.1111/j.1600-0889.2008.00383.x.
Salazar C, Alvarez C, Silva HA, Dorantes C. Radioactivity in air around nuclear facilities in Mexico. Environ Int 1994;20:747–56.
Sánchez-Ccoyllo OR, Silva Dias PL, Fátima Andrade M, Freitas SR. Determination of O3-, CO- and PM10- transport in the metropolitan area of Sao Paulo, Brazil though synoptic-scale analysis of back-trajectories. Meteorol Atmos Phys 2006;92:83–93. doi:10.1007/s00703-005-0139-6.
Stohl A. Computation, accuracy and applications of trajectories — a review and bibliography. Atmos Environ 1998;32:947–66. DOI: S1352-2310(97)00457-3.
US E.P.A. (United States Environmental Protection Agency). Website http://www.epa.gov/japan2011/rert/radnet-sampling-data.html 2011.