

Impact of Saharan dust events on radionuclide levels in Monaco air and in the water column of the northwest Mediterranean Sea

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ABSTRACT

Characterization of atmospheric aerosols collected in Monaco (2004–2008) and in sediment traps at 200 m and 1000 m water depths at the DYFAMED (Dynamics of Atmospheric Fluxes in the Mediterranean Sea) station (2004) was carried out to improve our understanding of the impact of Saharan dust on ground-level air and on the water column. Activity concentrations of natural (²¹⁰Pb, ²¹⁰Po, uranium and radium isotopes) and anthropogenic (¹³⁷Cs, ²³⁹Pu, ²⁴⁰Pu, and ²³⁹⁺²⁴⁰Pu) radionuclides and their isotopic ratios confirmed a Saharan impact on the investigated samples. In association with a large particulate matter deposition event in Monaco on 20 February 2004, the ¹³⁷Cs (~40 Bq kg⁻¹) and ²³⁹⁺²⁴⁰Pu (~1 Bq kg⁻¹) activities were almost a factor of two higher than other Saharan deposition dust events. This single-day particle flux represented 72% of the annual atmospheric deposition in Monaco. The annual deposition of Saharan dust on the sea was 232–407 mBq m⁻² for ¹³⁷Cs and 6.8–9.8 mBq m⁻² for ²³⁹⁺²⁴⁰Pu and contributed significantly (28–37% for ¹³⁷Cs and 34–45% for ²³⁹⁺²⁴⁰Pu) to the total annual atmospheric input to the northwest Mediterranean Sea. The ¹³⁷Cs/²³⁹⁺²⁴⁰Pu activity ratios in dust samples collected during different Saharan dust events confirmed their global fallout origin or mixing with local re-suspended soil particles. In the sediment trap samples the ¹³⁷Cs activity varied by a factor of two, while the ²³⁹⁺²⁴⁰Pu activity was constant, confirming the different behaviors of Cs (dissolved) and Pu (particle reactive) in the water column. The ¹³⁷Cs and ²³⁹⁺²⁴⁰Pu activities of sinking particles during the period of the highest mass flux collected in 20 February 2004 at the 200 m and 1000 m water depths represented about 10% and 15%, respectively, of annual deposition from Saharan dust events.

1. Introduction

Radionuclides found in ground-level air are of natural or anthropogenic origin. Sources of natural radioactivity include cosmogenic radionuclides such as ⁷Be and ²²Na and radiogenic radionuclides such as ²¹⁰Pb that emanate (via ²²²Rn) from the earth's crust to the atmosphere. Anthropogenic radionuclides including ¹³⁷Cs, isotopes of plutonium have been introduced to the atmosphere via nuclear weapons tests and releases from nuclear reprocessing facilities and accidents such as Chernobyl and

Fukushima (e.g. Livingston and Povinec, 2000, 2002). Radionuclides in the atmosphere rapidly attach on submicron-sized aerosols, and their variability in ground-level air is driven by the behavior of aerosols (Masson et al., 2009; Sýkora et al., 2012; Povinec et al., 2012; Hirose and Povinec, 2015).

Atmospheric radionuclides are deposited from the air onto the land and sea surface by wet and dry deposition. In this way, the terrestrial and marine environments are labelled by natural and anthropogenic radionuclides that can be used as tracers of environmental processes. Radionuclide activities and ratios in the environment can vary due to changes in radioisotope production, such as releases from nuclear installation or radon emanating from soil. Variability in radionuclides can also arise from processes such as soil resuspension or biomass burning (Amiro et al., 1996;

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Wotawa et al., 2006; Povinec et al., 2012; Hirose and Povinec, 2015).

Saharan dusts events, which can be observed in the atmosphere of southern Europe, are associated with the re-suspension source term. This transport of dust particles from northern Africa to Europe can have a substantial impact on the atmospheric processes in southern Europe. It has also been identified as an important pathway for particle delivery into surface seawater of the north-west (NW) Mediterranean (Moulin et al., 1997; Lee et al., 2002, 2003; Pham et al., 2003, 2005; TERNON et al., 2010). Similar dust events originating in Mongolian and Chinese deserts have also been reported in Japan (e.g. Igarashi et al., 2005).

Out of the nine significant Saharan dust events registered between 1998 and 2013 at the Monaco air monitoring station of the International Atomic Energy Agency, two major deposition events were detected on 23–24 November 2002 and 20–21 February 2004, when significant amounts of red-colored particles were collected (Pham et al., 2013). The impact of the exceptional Saharan dust event in February 2004 was also studied by French colleagues using aerosol samples collected in southern France (Masson et al., 2010; Menut et al., 2009).

Atmospheric aerosol radioactivity has been continuously monitored and studied in the Principality of Monaco by International Atomic Energy Agency - Environment Laboratories (IAEA-EL) since the 1980s to understand sources and variations of natural (^7Be , ^{40}K , ^{210}Po , ^{210}Pb , radium and uranium isotopes) and anthropogenic (^{137}Cs , ^{239}Pu , ^{240}Pu , and $^{239+240}\text{Pu}$) radionuclides in the atmosphere, including impact studies of the Chernobyl and Fukushima accidents on the atmospheric and marine environments (Then et al., 1980; Ballestra et al., 1987; Lee et al., 2002; Pham et al., 2011, 2012, 2013). In our previous studies we focused on variations of radionuclides in the atmosphere, on their dry and wet depositions, and on their inputs to the Mediterranean Sea (Lee et al., 2001, 2003; Pham et al., 2011, 2012, 2013). Other studies were devoted to investigations of Saharan dust particles in ground-level air (Lee et al., 2002; Pham et al., 2003, 2005, 2013).

The aim of the present work was to study the impact of Saharan dust events between 2004 and 2008 on atmospheric radioactivity and the transport of dust particles in seawater. The main focus is on the Saharan dust event observed in Monaco on 20 February 2004, and its impact on natural and anthropogenic radionuclides in ground-level air, and the water column of the NW Mediterranean Sea using sediment trap samples at the DYFAMED (Dynamics of Atmospheric Fluxes in the Mediterranean Sea) station off the Monaco coast (Fig. 1). The characterization of collected particles was done using X-ray fluorescence, gamma-spectrometry, alpha-spectrometry, ICP-MS (Inductively Coupled Plasma - Mass Spectrometry), and AMS (Accelerator Mass Spectrometry) techniques, which represent the state of the art technologies applied in environmental radioactivity research. Data on concentrations and isotope ratios of natural (^{40}K , ^{210}Pb , ^{210}Po , ^{226}Ra , ^{238}Ra , ^{234}U , ^{235}U and ^{238}U) and anthropogenic (^{137}Cs , ^{239}Pu , ^{240}Pu and $^{239+240}\text{Pu}$) radionuclides as well as major and trace elements are presented and discussed. This is the first time that such an extensive set of natural and anthropogenic radionuclides emanating from a Saharan dust event has been studied simultaneously in the atmosphere and marine environment using sediment traps.

2. Materials and methods

2.1. Collection of Saharan dust samples in Monaco

The collection of Saharan dust was carried out on the roof of the IAEA-EL premises in Monaco (43°45' N, 07°25' E) (Fig. 2) using a $2 \times 2 \text{ m}^2$ stainless steel funnel-type collector located about 15 m above the ground (Fig. 2). At the bottom of the collector a

rectangular container made of high density polypropylene was placed to collect the dry and wet deposition (Pham et al., 2013). The dust samples were collected by rinsing the surface of the collector with acidified distilled water. Finally, the collected liquid with the dust particles was slowly evaporated to dryness and placed in polyethylene containers for analysis by gamma spectroscopy.

Several Saharan dust deposition events were recorded in Monaco between 2002 and 2008. The most two important events were detected during 23–24 November 2002 and 20–21 February 2004 (Pham et al., 2005, 2013). In this study we will mainly focus, however, on the latter event which happened in the night of 20 February 2004 (coded as SD1, Table 1a), when a significant amount of red-colored particles (mass flux of $19,800 \text{ mg m}^{-2}$) was collected. Smaller dust events, which were observed on 1 May 2004, 29 July 2005, and 26 May 2008 (named as SD2, SD3 and SD4, respectively) are also investigated in the present work (Table 1a).

2.2. Sediment trap samples collected in the NW Mediterranean

The input of the 20 February 2004 Saharan dust event to the water column of the Mediterranean Sea was studied using sediment trap samples collected at 200 m and 1000 m at the DYFAMED station located at 43°25' N and 07°52' E in the Ligurian Sea (NW Mediterranean), about 52 km off the Monaco coast (Fig. 1). Details of the deployment of the DYFAMED sediment traps has been described in previous work (Martín et al., 2009; Miquel et al., 2011). The collection of the sediment trap samples started on 21 December 2003 and ended on 23 May 2004, with a 14 day collection periods (Table 1b). A total of eight sediment trap samples were collected at a water depth of 200 m, and 11 samples were collected at a depth of 1000 m. The sample coded as ST 9–12 was a combination of four samples (i.e. ST9, ST10, ST11 and ST12) due to the scarcity of material over the sampling periods. The Saharan dust event of 20 February 2004 manifested as red particles in the samples collected at the 200 m, mainly during the deployment time interval of 15–29 February (ST6, mass flux of $1228 \text{ mg m}^{-2} \text{ d}^{-1}$), with a smaller contribution ($853 \text{ mg m}^{-2} \text{ d}^{-1}$) up to 14 March (ST7). In the 1000 m trap samples the peak-particle flux ($893 \text{ mg m}^{-2} \text{ d}^{-1}$) was found between 29 February and 14 March (ST7), with smaller fluxes in the preceding interval, and the two subsequent intervals.

2.3. Analytical methods

Activity concentrations of gamma-emitters (^{40}K , ^{137}Cs , ^{210}Pb , ^{226}Ra and ^{228}Ra) were determined by gamma-ray spectrometry. Well-type HPGe detectors of 150% and 200% relative efficiency (compared to 7.6 cm in diameter and 7.6 cm high NaI(Tl) detector), operating in the IAEA-EL's underground laboratory with very low background, were used for the analysis of collected samples. The samples were counted for one to two weeks. Further details of the low-background gamma-ray detection system and calibration procedures can be found in Povinec et al., 2004, 2005. The uncertainties (below 10% at 1 sigma) include the counting statistics, detector efficiency calibration, and a background correction.

^{210}Pb was measured non-destructively (4 g in the well-type HPGe detector) in the whole sample (or a subsample for a big event such as SD1) by gamma-spectrometry. ^{210}Po was determined by alpha-spectrometry on a sub-sample (around 300 mg each) after radiochemical separation using an Ortec counting system. The calculated activities were corrected for the radioactive decay to the mid-collection period. Typical propagated uncertainties at 1 sigma were around 10%.

Uranium isotopes (^{234}U , ^{235}U , and ^{238}U) were determined by ICP-MS at the CITIUS laboratory (Departamento de Física Aplicada I, Universidad de Sevilla, Spain) after a total decomposition of the

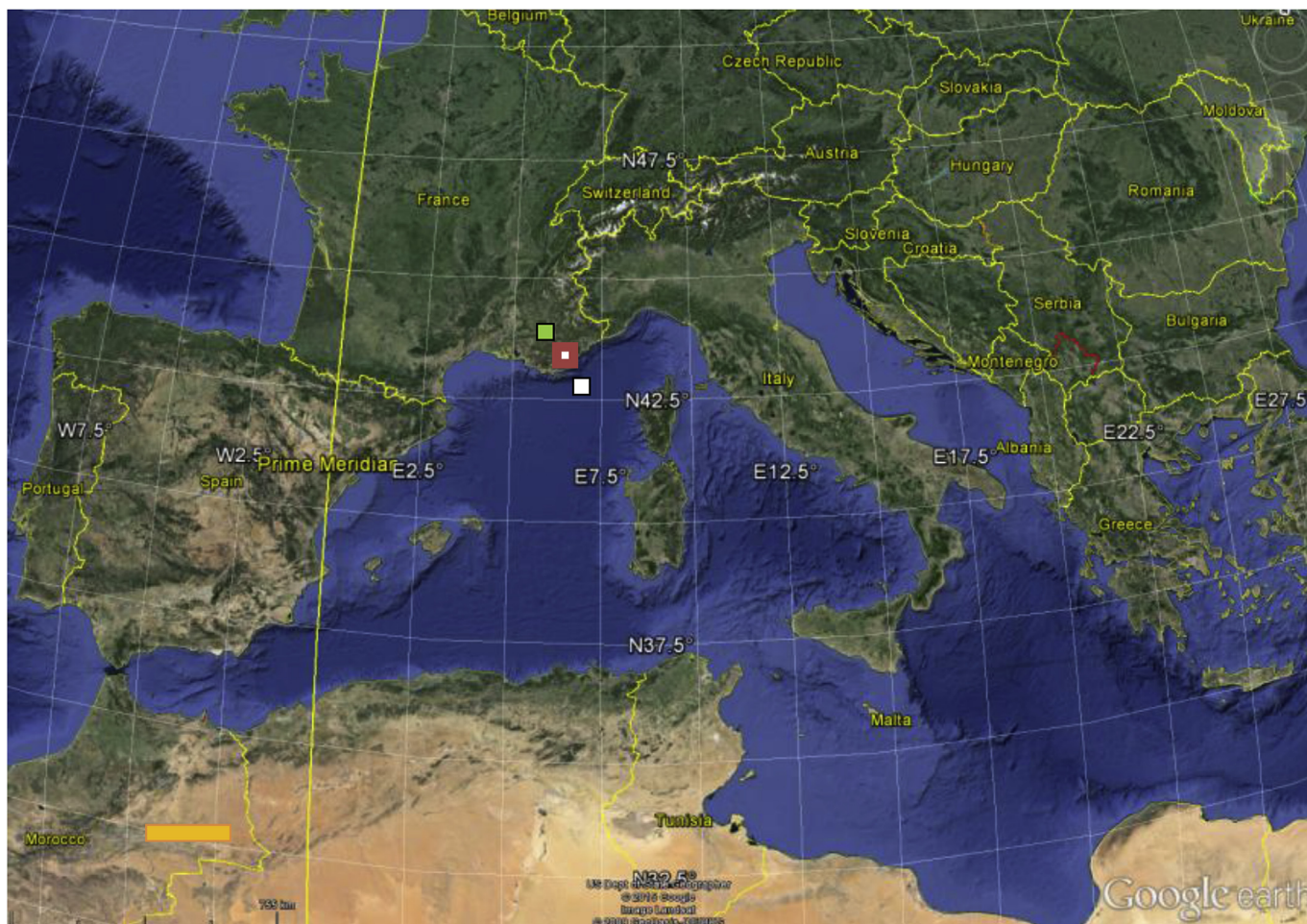


Fig. 1. Map of the Northwestern Mediterranean basin with two studied stations: (■) Monaco principality ($43^{\circ}45' \text{ N}$, $07^{\circ}25' \text{ E}$) and (□) DYFAMED site ($43^{\circ}25' \text{ N}$, $07^{\circ}52' \text{ E}$). The comparison with Manosque station (■) (southern France: $43^{\circ}50' \text{ N}$, $5^{\circ}47' \text{ E}$, [Masson et al., 2010](#)) is also shown. Note (■) is eastern Magreb region where 80% of Saharan dust event, 21–22 February 2004, arrived to southern France ([Menut et al., 2009](#)).

samples. Plutonium isotopes (^{239}Pu and ^{240}Pu) were determined by AMS at the CAN laboratory (Seville, Spain) after applying radiochemical procedures described in [Chamizo et al. \(2015\)](#).

The elemental composition of particles was determined using X-ray fluorescence spectrometry (Spectro X-Lab 2000). Details of the X-ray fluorescence technique can be found in [Martín et al. \(2009\)](#) and [Pham et al. \(2005\)](#).

2.4. Quality assurance of the analytical methods

In order to achieve high quality in the reported radionuclide results, regular analyses of reference materials were carried out during the sample measurements. A certified reference material provided by the IAEA (IAEA-385, Irish Sea sediment) ([Pham et al., 2008](#)), and a standard reference material (SRM 4353, soil) developed by NIST (National Institute of Standards and Technology, Gaithersburg, USA) were analyzed by gamma- and alpha-spectrometry, ICP-MS and AMS. To check for possible contamination problems, blank samples were also analyzed during ICP-MS and AMS measurements.

A Saharan soil sample (SS) ($26^{\circ}02'34.2'' \text{ N}$ and $14^{\circ}21'17.6'' \text{ E}$) from nearby the Gerboise site, where French nuclear tests were carried out in the 1960s ([Menut et al., 2009](#); [Masson et al., 2010](#)) was also analyzed for radionuclides and trace elements. The source of the Saharan dust collected at the Monaco station may not come

from the exact place where the Saharan soil (SS) was collected. The SS sample was sieved, and the $<63 \mu\text{m}$ fraction was used for comparison with the deposit samples collected in Monaco.

3. Results and discussion

3.1. Elemental composition

The major element composition of the Saharan dust deposits collected in Monaco during the four studied events, in the Saharan soil (SS) and in the sediment trap samples collected at the DYFAMED station, were similar consisting mainly of Al (5–7%), Si (up to 20%), Ca (up to 12%) and Fe (4%). The concentrations of Al, Si, Ca and Fe were comparable with those observed previously in Saharan dust samples collected in Monaco ([Pham et al., 2005](#)), and with those observed in marine particles collected in the Southeast Mediterranean ([Herut et al., 2001](#)). However, their concentrations were higher than in the dust originating from the European continent ([Guieu et al., 1997](#); [Pham et al., 2003](#)).

Trace metal concentrations were similar between samples, except for Cu, Zn, Cr, Sn and Ni, whose concentrations were higher in the Saharan dust deposits compared to in the sediment trap samples and the SS. It has been previously demonstrated that Zn decreases notably from the atmosphere through the water column to the sea floor ([Martín et al., 2009](#), and references therein). A



Fig. 2. Stainless steel collector on the roof of IAEA-EL premises in Monaco for collection of dry and wet deposits (Note in the lower part shown the important residue of dust deposition SD1 collected on the 20 February 2004).

Table 1a

Saharan dust deposits collected at Monaco station (43°45' N, 07°25' E).

Sample code	Sampling date start	Sampling date end	Mass flux (mg m ⁻² d ⁻¹)
SD1	2004-2-20	2004-02-21	19,800
SD2	2004-05-01	2004-05-02	2675
SD3	2005-07-29	2005-07-31	1275
SD4	2008-05-26	2008-05-27	1375

sediment core collected from the sea floor at the DYFAMED site revealed that Pb, Zn and Cu were enriched over background levels in the upper centimeters of the sediment column (Martín et al., 2009).

The concentration of Zr was exceptionally high in the SS sample (7900 µg/g) compared to the dust and sediment trap samples (maxima at 200 µg/g). Conversely, S, Br, Sr and I were higher in the sediment trap samples; the highest S concentration (up to 3770 µg/g) was found in particles collected between 28 March 2004 and 23 May 2004 at 200 m depth, and between 9 May 2004 and 23 May 2004 at 1000 m depth. It is well known that particles collected at the coastal sites in the NW Mediterranean are mixtures of urban-dominated (European) and crust-dominated (Saharan) components (Chester et al., 1997), and that concentrations of some trace

elements (Fe, Mn, Cr, Ni, Cu, Zn, Cd and Pb) in particles coming from Sahara Desert are lower than the ones of European origin (Gueiu et al., 1997; Pham et al., 2003).

3.2. Radionuclide compositions

3.2.1. Natural radionuclides

Activities of ²¹⁰Pb, ²¹⁰Po, ²²⁶Ra, ²²⁸Ra, ²³⁴U, ²³⁵U, ²³⁸U and ⁴⁰K are listed in Tables 2a and b. Activity ratios for some of the radionuclides are given in Table 4.

3.2.1.1. Saharan soil and Saharan dust deposition. The activity of uranium isotopes (²³⁴U, ²³⁵U and ²³⁸U) was similar in SD1, SD2, and SD3 samples and slightly higher in SD4 (Table 2b). The reported activities for SD1 are comparable with results obtained at the Manosque station (southern France: 43°50' N, 5°47' E; Fig. 1) one or two days later (Masson et al., 2010). They are also comparable with previous results from the Monaco station (Pham et al., 2005). The activities of ²²⁶Ra, ²²⁸Ra (Table 2a) in the Saharan dust samples were very similar, but much lower than the soil sample (SS).

The ²¹⁰Pb and ²¹⁰Po levels determined by gamma and alpha spectrometry, respectively for SD and SS samples were in good agreement (Table 2a). Over the sampling period, their values in the Saharan dust deposit samples varied four-fold (Table 2a). ²¹⁰Pb is produced by radioactive decay from its progenitor, ²²²Rn, a noble gas in the ²³⁸U decay chain, and emanates primarily from land surface. The ²¹⁰Po/²¹⁰Pb activity ratio for SD1, SD2 and SD3 as well as for SS was 1 (Table 4), almost two times higher than ratios found in a previous study of Saharan dust deposits at the Monaco station (Pham et al., 2005). The ²¹⁰Po activity at the time of radiochemical separation from ²¹⁰Pb was the same as of ²¹⁰Pb, thus confirming an equilibrium between ²¹⁰Pb and ²¹⁰Po at the sampling date. Lower concentrations of ²¹⁰Po and ²¹⁰Pb in the SD1 sample compared to the other SD samples (Table 2a) could be explained by a direct (unmixed) transfer from the Saharan Desert or in the case of SD2 and SD3 by partial mixing with local re-suspended dust particles. The concentrations of ²¹⁰Pb, ²¹⁰Po and uranium isotopes in the SS sample were lower than in the SD samples, while the ⁴⁰K activity was similar.

The activity ratios of ²³⁵U/²³⁸U were ~0.046, confirming that the

collected particles were of natural origin (crust-dominated from Saharan desert), as demonstrated in previous studies (Pham et al., 2003, 2005). The activity ratios of ²³⁴U/²³⁸U were ~1, indicated a radioactive equilibrium between ²³⁸U and its daughter ²³⁴U. However, a further look at other ²³⁸U daughter product, ²²⁶Ra and ²¹⁰Pb calls into question this equilibrium as ²²⁶Ra and ²¹⁰Pb were enriched relative to the ²³⁸U in all the Saharan dust deposition samples.

In the Saharan soil sample (which may not be representative of the Saharan dust deposits collected in Monaco), the ²³⁵U/²³⁸U and ²³⁴U/²³⁸U activity ratios were respectively about 0.046 and 1, confirming that they were of natural origin (terrestrial crust). Similar to the Monaco dust samples, ²²⁶Ra and ²¹⁰Pb were enriched relative to ²³⁸U. Uranium and radium have different behaviors in the

Table 1b

Depth, date and mass flux of the mooring deployments during the time-series DYFAMED 39 (DYF 39) survey (43°25' N, 07°52' E). Note the sediment trap samples in this series were principally influenced from the SD1 event.

Depth (m)	Sample code	Sampling date start	Sampling date end	Mass flux (mg m ⁻² d ⁻¹)
200	ST2	2003-12-21	2004-01-04	584.8
200	ST3	2004-01-04	2004-01-18	175.8
200	ST4	2004-01-18	2004-02-01	130.9
200	ST5	2004-02-01	2004-02-15	41.7
200	ST6	2004-02-15	2004-02-29	1228
200	ST7	2004-02-29	2004-03-14	852.7
200	ST8	2004-03-14	2004-03-28	234.1
200	ST9-12	2004-03-28	2004-05-23	209.1
1000	ST2	2003-12-21	2004-01-04	156.3
1000	ST3	2004-01-04	2004-01-18	140.6
1000	ST4	2004-01-18	2004-02-01	85.3
1000	ST5	2004-02-01	2004-02-15	114.9
1000	ST6	2004-02-15	2004-02-29	723.7
1000	ST7	2004-02-29	2004-03-14	893.4
1000	ST8	2004-03-14	2004-03-28	558.7
1000	ST9	2004-03-28	2004-04-11	564.9
1000	ST10	2004-04-11	2004-04-25	173.0
1000	ST11	2004-04-25	2004-05-09	243.8
1000	ST12	2004-05-09	2004-05-23	248.1

Table 2a

Radionuclide activities (in Bq kg⁻¹) determined by gamma-spectrometry and alpha-spectrometry (for ²¹⁰Po).

Sample	¹³⁷ Cs	Unc.	²²⁶ Ra	Unc.	²²⁸ Ra	Unc.	⁴⁰ K	Unc.	²¹⁰ Pb	Unc.	²¹⁰ Po	Unc.
SD1	40.7	2.1	38.7	2.4	42.2	1.6	633	15	980	10	1050	20
SD2	37.9	1.9	31.1	1.6	34.6	0.5	574	12	2450	6	2370	40
SD3	38.1	2.2	26.4	2.4	42.4	2.3	680	19	2940	30	2930	50
SD4	23.2	1.2	32.2	1.7	44.5	0.5	531	11	4032	6	n.d.	
SS	23.0	1.3	117.7	6.1	171.4	1.9	497	11	188	5	215	4
200 m												
ST2	12.2	0.9	39.0	3.3	38.8	2.6	581	18	323	20	1030	30
ST3	10.2	1.9	82.7	4.8	34.4	1.7	489	13	1510	60	1040	30
ST4	10.6	1.0	95.1	5.2	41.7	1.8	465	12	534	16	1660	50
ST6	22.9	1.3	59.5	3.9	32.0	2.3	483	14	298	12	740	30
ST7	25.4	1.4	51.0	3.6	33.4	2.2	545	15	302	12	780	30
ST8	23.3	1.3	50.9	3.3	34.0	1.8	494	13	287	10	820	30
ST9-12	14.2	0.9	83.2	4.6	24.0	1.7	383	11	347	11	830	30
1000 m												
ST2	12.7	0.9	61.3	3.5	34.6	1.8	553	14	372	11		
ST3	10.4	4.8	86.0	4.6	35.3	1.4	498	12	1760	200		
ST4	10.4	0.9	95.4	5.4	37.8	2.2	508	13	509	16		
ST5	8.5	2.0	108.6	5.8	32.3	1.4	477	12	1420	70		
ST6	22.2	1.6	61.6	3.7	31.0	1.9	538	14	714	20		
ST7	24.1	1.7	60.4	4.1	34.9	2.4	559	17	781	22		
ST8	19.4	1.2	62.4	3.6	31.4	1.8	527	14	405	12		
ST9	16.3	1.5	96.5	5.4	27.8	1.7	480	13	928	26		
ST10	14.5	1.5	82.4	4.7	28.3	1.9	467	13	1140	30		
ST11	12.1	3.2	85.6	4.5	27.3	1.2	471	11	1130	80		
ST12	9.7	0.8	102.0	5.3	23.6	1.2	340	8	366	11		

n.d. not determined.

environment: uranium is soluble (U⁴⁺ form being oxidized to uranyl anion UO₂²⁺), while radium is insoluble (existing as RaO) (Cochran, 1992). This may explain the depletion of uranium (compared to radium) in the soil and dust samples. The ²¹⁰Po/²¹⁰Pb activity ratio of Saharan soil was about 1.14 (Table 4), confirming that ²¹⁰Pb and ²¹⁰Pb were in equilibrium at the sampling date.

3.2.1.2. *Sediment trap samples.* The ²²⁶Ra levels in sediment trap samples were higher during the three Saharan dust events (samples # ST6, ST7 and ST8) at both depths compared to the atmospheric level (Table 2a). The concentration of uranium isotopes (²³⁴U, ²³⁵U and ²³⁸U) varied little between depth and sampling date (Table 2b).

The ²²⁶Ra/²³⁸U activity ratios of sediment trap samples were

similar at both depths and higher than the dust deposition samples (Table 4). This is likely due to solubility of uranium and insolubility of radium in seawater.

The levels of ²¹⁰Pb and ²¹⁰Po (Table 2a) varied in time at both depths, with lower activities in samples ST6, ST7 and ST8 compared to samples collected before the SD1 event (for instance ST4 sampled at 200 m, and ST5 sampled at 1000 m). The ²¹⁰Po/²¹⁰Pb activity ratios (Table 4) in samples ST3 to ST9-12 at 200 m varied from 0.69 to 2.9. The ²¹⁰Po activity at the time of radiochemical separation was higher than that for ²¹⁰Pb for all samples at 200 m (except for the sample ST3), suggesting that there was already some ²¹⁰Po in the Saharan dust deposit before it was transported into the water column. The ²²⁶Ra and ²¹⁰Pb showed enrichment compared to ²³⁸U and ²³⁴U. Radon progeny likely adsorbed onto atmospheric

Table 2b
Activity concentration of uranium isotopes (in Bq kg⁻¹) determined by ICP-MS.

Sample	²³⁴ U	Unc.	²³⁵ U	Unc.	²³⁸ U	Unc.
SD1	28.0	0.6	1.17	0.01	25.2	0.2
SD2	28.0	0.5	1.17	0.01	25.4	0.2
SD3	28.6	2.1	1.20	0.01	25.9	0.2
SD4	36.8	0.7	1.51	0.01	32.6	0.3
SS	16.3	0.6	0.71	0.01	15.4	0.1
200 m						
ST3	19.1	1.1	1.02	0.01	22.0	0.2
ST6	21.4	0.5	0.95	0.01	20.5	0.1
ST7	22.7	0.4	0.98	0.01	21.4	0.2
ST8	25.7	0.5	1.02	0.01	22.2	0.2
ST9-12	20.5	0.3	0.84	0.01	18.4	0.1
1000 m						
ST3	19.4	0.8	1.03	0.01	22.3	0.2
ST6	23.3	0.5	0.98	0.01	21.2	0.1
ST7	18.5	0.9	0.91	0.01	19.9	0.2
ST8	22.7	0.5	1.00	0.01	21.5	0.2
ST9	23.2	0.4	1.01	0.01	21.8	0.2

aerosol particles, leading to ²¹⁰Pb enrichment relative to the soil and the degree of enrichment increased with travel time through the atmosphere. Both polonium and lead have affinity for particles; however, lead preferentially adsorbs to inorganic particles, whereas polonium has a high affinity for biological particles via biological uptake. Therefore particles traveling in the water column incorporated more Po than Pb from the seawater. Similarly, the other elements with a high affinity for organic matter (e.g. Ca) were also enriched in sediment trap samples when compared to Saharan dust and Saharan soil.

⁴⁰K levels varied little between samples, as potassium dissolves readily in the water column. The ²²⁸Ra (the decay product in the ²³²Th decay chain) concentrations were comparable with ²²⁶Ra levels.

3.2.2. Anthropogenic radionuclides

The concentrations of ¹³⁷Cs, ²³⁹Pu, ²⁴⁰Pu and ²³⁹⁺²⁴⁰Pu in Saharan dust deposit samples (SD), Saharan soil (SS) sample as well as in sediment trap samples (ST) are shown in Tables 2a and 3. The atom (²⁴⁰Pu/²³⁹Pu) ratio and the activity (²³⁹⁺²⁴⁰Pu/¹³⁷Cs, ¹³⁷Cs/²³⁹⁺²⁴⁰Pu) ratios are shown in Table 4.

Table 3
Activity concentrations of plutonium isotopes (in Bq kg⁻¹) determined by AMS.

Sample	²³⁹ Pu	Unc.	²⁴⁰ Pu	Unc.	²³⁹⁺²⁴⁰ Pu	Unc.
SD1	0.57	0.01	0.41	0.01	0.98	0.02
SD2	0.54	0.01	0.39	0.01	0.93	0.02
SD3	0.44	0.01	0.32	0.01	0.76	0.02
SD4	0.37	0.01	0.31	0.01	0.68	0.01
SS	0.059	0.003	0.037	0.004	0.096	0.01
200 m						
ST3	0.61	0.01	0.41	0.01	1.02	0.02
ST6	0.64	0.01	0.46	0.01	1.09	0.02
ST7	0.66	0.01	0.47	0.01	1.13	0.02
ST8	0.68	0.02	0.47	0.01	1.16	0.02
ST9-12	1.16	0.02	0.86	0.02	2.02	0.03
1000 m						
ST3	0.68	0.01	0.49	0.01	1.17	0.02
ST6	0.34	0.01	0.24	0.01	0.58	0.01
ST7	0.64	0.01	0.46	0.01	1.10	0.02
ST8	0.66	0.01	0.49	0.01	1.15	0.02
ST9	0.65	0.01	0.48	0.01	1.14	0.02

3.2.2.1. *Saharan dust deposits and Saharan soil.* The activity concentration of ¹³⁷Cs in Saharan dust deposits was rather stable from one event to another with higher concentrations (from about 38 to 40 Bq kg⁻¹) in the first three events SD1, SD2 and SD3 compared to the last event SD4 (23 Bq kg⁻¹) (Table 2a), which was closest to Saharan soil (Table 2a). The ¹³⁷Cs deposition flux from the Saharan dust event SD1 was 0.79 Bq m⁻². This deposition event, which lasted just one day, corresponds to 72% of the cumulative ¹³⁷Cs deposition during 2004 at the Monaco station (1.1 Bq m⁻² y⁻¹; Pham et al., 2013). It is comparable to the ¹³⁷Cs activity (38.7 Bq kg⁻¹) and 75% cumulative ¹³⁷Cs deposition flux found by Masson et al. (2010) from the same event collected one or two days later at the Manosque station. The ¹³⁷Cs activity in the Saharan dust deposits was of the same order of magnitude as the activity of local re-suspended particles collected during dry seasons (Pham et al., 2013). The main sources of ¹³⁷Cs variations in the atmosphere may be summarized as follows:

- (i) Local re-suspension of ¹³⁷Cs from soil (Pham et al., 2011; Sýkora et al., 2012);
- (ii) ¹³⁷Cs input from Saharan dust events (Masson et al., 2010; Pham et al., 2005, 2011);
- (iii) Contributions from field/forest fires burning biota containing ¹³⁷Cs of the Chernobyl and global fallout origin (Wotawa et al., 2006; Masson et al., 2009; Hirose and Povinec, 2015).

Menut et al. (2009) showed that in the case of the 21–22 February 2004 Saharan dust event, the main part (80%) of the dust found in southern France was from the eastern Maghreb region. Only 0.7% of the African emissions were from the Gerboise site, which could explain 1–5% to the peak-day value observed at southern France station.

The ²³⁹⁺²⁴⁰Pu levels in Saharan dust deposits ranged from 0.98 Bq kg⁻¹ to 0.68 Bq kg⁻¹ significantly higher than in the Saharan soil (0.096 Bq kg⁻¹) (Table 3). The plutonium value obtained for SD1 event was also similar to the activity measured in the samples collected at Manosque station (Masson et al., 2010).

The ²⁴⁰Pu/²³⁹Pu atom ratio was similar in all dust samples (~0.20) (Table 4) and within uncertainty the same as the Saharan soil (0.17 ± 0.02). These values are similar to the global fallout ratio of 0.186 ± 0.009 reported by Kelley et al. (1999). The ²³⁹⁺²⁴⁰Pu/¹³⁷Cs activity ratios in dust samples also varied little (Table 4) and were similar to the global fallout value (0.024) reported by Ikeuchi et al. (1999). These results are in agreement with results obtained from other Saharan dust events previously observed at the Monaco station (Pham et al., 2005), and also those measured at the Manosque station (Masson et al., 2010). We can therefore conclude that the origin of anthropogenic radionuclides in Saharan dust particles is primarily global fallout. The ¹³⁷Cs/²³⁹⁺²⁴⁰Pu activity ratios in SD1, SD2 and SD4 also support this hypothesis (Menut et al., 2009, and references therein), however the dust collected on 29 July 2005 (SD3) may have been influenced by another source than global fallout. The higher ¹³⁷Cs/²³⁹⁺²⁴⁰Pu ratio of SD3 may be due to the contribution of locally re-suspended ¹³⁷Cs from soil. This hypothesis is supported by the presence of grey/reddish colored particles, compared to the red or reddish-colored particles found in the SD1, SD2 and SD4 samples. The ¹³⁷Cs/²³⁹⁺²⁴⁰Pu activity ratio in the Saharan soil was much higher (239 ± 19), reflecting the ¹³⁷Cs dominance in the Saharan region.

3.2.2.2. *Sediment trap samples.* In sediment trap time-series, cesium and plutonium isotopes exhibited different behaviors. The activity of ¹³⁷Cs was double in samples ST6-ST8 relative to samples collected before and after the deposition event. These activities were however only half the activity of ¹³⁷Cs in the dust sample from

Table 4
Activity and atom ratios for natural and anthropogenic radionuclides.

Sample	$^{240}\text{Pu}/^{239}\text{Pu}$	Unc.	$^{239+240}\text{Pu}/^{137}\text{Cs}$	Unc.	$^{137}\text{Cs}/^{239+240}\text{Pu}$	Unc.	$^{226}\text{Ra}/^{238}\text{U}$	Unc.	$^{210}\text{Po}/^{210}\text{Pb}$	Unc.
SD1	0.20	0.01	0.024	0.001	41.4	2.3	1.54	0.10	1.08	0.02
SD2	0.20	0.01	0.024	0.001	40.9	2.2	1.22	0.06	0.97	0.02
SD3	0.20	0.01	0.020	0.001	50.3	3.0	1.02	0.09	1.00	0.02
SD4	0.23	0.01	0.029	0.002	34.1	1.9	0.99	0.05		
SS	0.17	0.02	0.004	0.001	239	19	7.64	0.40	1.14	0.04
200 m										
ST3	0.18	0.01	0.100	0.019	10.0	1.9	3.76	0.22	0.69	0.02
ST6	0.20	0.01	0.048	0.003	21.0	1.3	2.90	0.19	2.48	0.14
ST7	0.20	0.01	0.044	0.003	22.6	1.3	2.38	0.17	2.58	0.14
ST8	0.19	0.01	0.050	0.003	20.1	1.2	2.29	0.15	2.86	0.14
ST9-12	0.20	0.01	0.142	0.010	7.0	0.5	4.52	0.25	2.39	0.11
1000 m										
ST3	0.20	0.01	0.113	0.052	8.8	4.1	3.86	0.21		
ST6	0.20	0.01	0.026	0.002	38.1	2.9	2.91	0.18		
ST7	0.20	0.01	0.046	0.003	21.9	1.6	3.04	0.21		
ST8	0.20	0.01	0.059	0.004	16.9	1.1	2.90	0.17		
ST9	0.20	0.01	0.070	0.006	14.3	1.3	4.43	0.25		

SD1. It is well known that cesium is soluble in water (Pham, 1997, and references therein); therefore its levels in particulate matter should be low.

The activity of ^{239}Pu , ^{240}Pu , $^{239+240}\text{Pu}$ were similar for sediment trap samples ST6, ST7 and ST8 which were collected during the Saharan dust event and sample ST3 collected before the event (Table 3). However, in sample ST9-12 collected after the event (between 28 March 2004 and 23 May 2004 at 200 m), the plutonium concentration was two times higher. This sample is unique in that it is a combination of four very low-flux sediment trap samples. In the 1000 m sediment trap sample, ST6 (sampled between 15 February 2004 and 29 February 2004), which was the first sample of sinking particles from the Saharan dust event at this depth, the plutonium activity was two times lower than other samples. There was little variation in the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios at either depth across the entire sampling period (Table 4). The $^{137}\text{Cs}/^{239+240}\text{Pu}$ activity ratio of ST6, ST7 and ST8 samples were two times higher than for other ST samples. Still, these ratios were half those measured in the atmospheric particles (SD1). With the exception of sample ST9-12 at 200 m and ST6 at 1000 m, the $^{239+240}\text{Pu}$ activity was around 1.1 Bq kg⁻¹. This behavior of plutonium isotopes reflects its particle-reactivity compared to the high solubility of cesium in seawater.

Taking into account the ^{137}Cs and $^{239+240}\text{Pu}$ activities in dust particles and the reported fluxes of marine particles (around 1 mg cm⁻² yr⁻¹ in the western Mediterranean (Guieu et al., 1997), the annual deposition from Saharan dust was estimated to 232–407 mBq m⁻² for ^{137}Cs and to 6.8–9.8 mBq m⁻² for $^{239+240}\text{Pu}$. These represent significant contributions (28–37% for ^{137}Cs and 34–45% for $^{239+240}\text{Pu}$) to the total annual atmospheric input of radionuclides to the NW Mediterranean Sea. The ^{137}Cs and $^{239+240}\text{Pu}$ activities during ST6 (period of the highest mass flux at 200 m) and ST7 (period of the highest mass flux at 1000 m) represented about 10% of ^{137}Cs and about 15% of $^{239+240}\text{Pu}$ annual Saharan dust fluxes at each depth. It was worth noting that the total flux of ^{137}Cs in sediment traps at both depths in 2004 was 3–5 times greater than the annual deposition reported in the previous studies (Lee et al., 2003; Pham et al., 2005). Although cesium has been diluted through the water column, its downward transport appears to be very efficient.

4. Conclusions

To understand the impact of Saharan dust on the air and sea of the northwest Mediterranean region, radioisotopes in aerosol and marine particles were characterized from 2004 to 2008. Four specific dust events were captured including a particularly large event in February 2004. The main observations were:

- (i) The activity concentrations of natural radionuclides (^{210}Pb , ^{210}Po , uranium and radium isotopes) and their activity ratios, as well as the elemental composition of collected particles, confirmed a Saharan-dust source fingerprint. In a huge particulate matter deposition event captured at the Monaco station on 20 February 2004, the ^{137}Cs (about 40 Bq kg⁻¹) and $^{239+240}\text{Pu}$ (about 1 Bq kg⁻¹) activities were almost a factor of two higher compared to the other Saharan deposition events. This single-day particle flux represented 72% of annual atmospheric deposition in Monaco.
- (ii) The annual deposition of the Saharan dust on the sea was estimated to be 232–407 mBq m⁻² for ^{137}Cs and 6.8–9.8 mBq m⁻² for $^{239+240}\text{Pu}$, accounting for a significant proportion of the total annual atmospheric input to the NW Mediterranean Sea (28–37% for ^{137}Cs and 34–45% for $^{239+240}\text{Pu}$).
- (iii) The $^{137}\text{Cs}/^{239+240}\text{Pu}$ activity ratios of different Saharan dust events ranged from 31 to 40 with one exceptional value of 50, showing, a global fallout origin for the former cases, or their mixing with local re-suspended soil particles for the latter case.
- (iv) While the ^{137}Cs activity varied by a factor of two in marine particles, the $^{239+240}\text{Pu}$ activity remained unchanged at the time scale studied at both depths, thus reflecting the different behaviors of Cs (soluble in seawater) and Pu (particle reactive) in the water column. The ^{137}Cs and $^{239+240}\text{Pu}$ activities of the sinking particles at the peak mass fluxes at 200 m and 1000 m represented about 10% of ^{137}Cs , and about 15% of $^{239+240}\text{Pu}$ total annual deposition fluxes from Saharan dust deposits at each depth.

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