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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 (^{210}Pb, ^{210}Po, uranium and radium isotopes) and anthropogenic (^{137}Cs, ^{239}Pu, ^{240}Pu, and $^{239+240}$Pu) 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 ^{137}Cs (~40 Bq kg^{-1}) and $^{239+240}$Pu (~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 ^{137}Cs and 6.8–9.8 mBq m^{-2} for $^{239+240}$Pu and contributed significantly (28–37% for ^{137}Cs and 34–45% for $^{239+240}$Pu) to the total annual atmospheric input to the northwest Mediterranean Sea. The ^{137}Cs/$^{239+240}$Pu 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 ^{137}Cs activity varied by a factor of two, while the $^{239+240}$Pu activity was constant, confirming the different behaviors of Cs (dissolved) and Pu (particle reactive) in the water column. The ^{137}Cs and $^{239+240}$Pu 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 ^{7}Be and ^{22}Na and radiogenic radionuclides such as ^{210}Pb that emanate (via ^{222}Rn) from the earth's crust to the atmosphere. Anthropogenic radionuclides including ^{137}Cs, 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 northwest (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 (^{7}Be, ^{40}K, ^{210}Po, ^{210}Pb, radium and uranium isotopes) and anthropogenic (^{137}Cs, ^{239}Pu, ^{240}Pu, and $^{239+240}$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 $^{239+240}$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 × 2 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 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 mg m^{-2} d^{-1}), with a smaller contribution (853 mg m^{-2} d^{-1}) up to 14 March (ST7). In the 1000 m trap samples the peak-particle flux (893 mg m^{-2} 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

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