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Cs-134 in soils of the Western Canary Islands after the Chernobyl nuclear accident

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ABSTRACT

^{134}Cs was measured in soil samples collected in the Western Canary Islands during a survey carried out in 1990–1991. This was 4 to 5 years after the Chernobyl Nuclear Power plant accident (1986). Models of the radioactive plumes did not show that ^{134}Cs released by the nuclear accident was transported directly from the accident site to these islands. In this work, we provide a possible explanation to how the ^{134}Cs may have been transported and deposited in soils of these islands after the accident. Intermittent inputs of mineral dust from Africa, in the form of intense dust storms, arrive to these islands every year. We believe that the ^{134}Cs from the accident may have been first deposited in Northern Africa, then resuspended and transported to the islands by various dust storms. Atmospheric records of African dust indicate that some strong events (high levels of particulate matter) took place in the 1986–1991 period. This hypothesis is supported by >20 years of aerosol data (2000−2022) collected at this site showing that ^{137}Cs, another isotope of radiocaesium, is resuspended and transported to these islands in connection with these type of African dust storms.

1. Introduction

A radiometric survey carried out in the Western Canary Islands in 1990–1991 reported the presence of caesium-134 (^{134}Cs) in the top layers of soil of three of the 4 investigated islands (Fernández de Aldecoa, 2000). ^{137}Cs, another isotope of caesium, was reported in all 4 islands. A subsequent survey performed in 1994 showed that ^{134}Cs had already decayed below detection limit, approximately 0.5 Bq·kg^{-1}, and confirmed the presence of ^{137}Cs in the soils of these islands (Fernández de Aldecoa, 2000). Because the last atmospheric nuclear weapon test had occurred >20 years prior to the first survey, the possibility of the weapons global fallout being the source of the ^{134}Cs in the soils was discarded. Instead, Fernández de Aldecoa (2000) speculated that the most likely source of this isotope was the Chernobyl Nuclear Power Plant (NPP) accident that took place in 1986 (i.e. 4–5 years earlier). However, given the concentrations measured, this author could not explain how this isotope might have been transported and deposited at the Canary Islands, especially because of the distance to the location of the accident (over 4600 km). Simulations of the dispersion of ^{134}Cs released by the Chernobyl NPP accident and ^{134}Cs deposition maps after the accident did not show any plumes reaching this site or even heading in the direction of the Canary Islands after the accident (De Cort et al., 1998; EEA, 2021; IRSN, 2021; UNSCEAR, 2000). The ^{137}Cs deposition map published by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) did not cover the Canary Islands nor Northern Africa (UNSCEAR, 2000). However, this map showed that Spain mainland and Portugal (approximately 1700 km from the Canary Islands) were barely impacted by the event and received a relatively low ^{137}Cs deposition of <2 kBq·m^{-2}. Baggoura et al. (1998) reported that ^{134}Cs and ^{137}Cs were measured in soils in Algeria (Northern Africa) directly after the Chernobyl NPP accident. A deposition value of 400 Bq·m^{-2} was estimated for ^{137}Cs for this country. The distance between

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Algeria and the Canary Islands is approximately 1000 km.

In this paper, we provide a possible explanation to how Chernobyl-released ^{134}Cs (and ^{137}Cs) may have reached the Western Canary Islands other than by direct atmospheric deposition by a radioactive plume immediately after the accident. This is based on 1) atmospheric records of African dust intrusions collected between 1986 and 1991 and from 2001 to 2021 for this site and 2) the radiometric data (gamma emitting nuclides and a subset of plutonium measurements) that has been collected on aerosol filters over the last 21 years at a sampling station located in the marine boundary layer (MBL) at the island of Tenerife. These data sets show how the Canary Islands regularly receive inputs of ^{137}Cs-loaded particles from Northern and Central Africa. The ^{137}Cs is eroded together with other fine particles from the top layer of soil, resuspended and transported to the islands in dust storms (secondary deposition) (Hernández et al., 2005; Hernandez et al., 2007; Karlsson et al., 2008; López-Pérez et al., 2020; López-Pérez et al., 2021). It is therefore likely that African dust storms that occurred in the months after the Chernobyl accident (as shown by the aerosol records from 1986 to 1991), had been scavenging and had transported both ^{134}Cs and ^{137}Cs to this site. Details follow.

1.1. The study area

The Canary Islands archipelago, Fig. 1, is situated west (approximately 110 km) from the northern coast of Africa, between 27°37′ and 29°25′ N and 13°20′ and 18°10′ W. The location of the Chernobyl NPP is also shown in Fig. 1.

1.2. African dust events

North Africa is Earth's largest and most persistently active mineral dust source accounting for 36 %–79 % of global emissions (Prospero et al., 2021). Dust transport out of Africa, with a pulsing character, undergoes a seasonal cycle that shifts latitudinally with the seasonal changes in the large-scale circulation over the Atlantic. Convection carries dust to altitudes as high as 6–7 km over the Saharan and Sahel regions (Knippertz and Todd, 2012; Prospero et al., 2021). When the dust reaches the west coast of Africa, it is lifted above the cooler trade winds, forming an elevated layer called the Saharan Air Layer (SAL). During transit, dust is transferred from the SAL to the underlying boundary layer, primarily by convective erosion. Some dust might also be injected directly into the boundary layer along the coast of Africa (Prospero et al., 2021; Reid et al., 2003). These dust particles are often deposited over the Canary Islands (Alonso-Pérez et al., 2007; Cuevas et al., 2021; Gelado-Caballero et al., 2012; Goudie and Middleton, 2001). Satellite views of several African dust intrusions over the Canary Islands are shown in Fig. 2.

Díaz et al. (2006) studied the dynamics and the aerosol chemistry of the air masses reaching the free troposphere of the subtropical Northeast Atlantic region during the period 1995–98. They found four different types of air masses: oceanic middle troposphere air masses (OMT), which corresponded to an Atlantic medium troposphere situation and included 50.6 % of the whole set of back trajectories; AfD, which corresponded to African air masses, accounted for the 19.8 % of the data set. Cluster EAM, which was associated with an aerosol mixture of anthropogenic, maritime aerosols and mineral dust, represented 12.7 % of the data; and finally, cluster MaA that was characterised by maritime aerosols represented 16.9 % of the whole data set. The highest quantities of mineral dust were linked with African air masses (the second most frequent air mass type) with a mean value of 86.5 μg·m^{-3}. The lowest dust levels were, on average, associated with OMT and MaA air mass types: 12.7 μg·m^{-3}.

1.3. The Chernobyl NPP accident

The Chernobyl Nuclear Power Plant is in the Kiev region in the north of Ukraine, 7 km south of the Ukrainian-Belarusian border. The NPP started the operation in 1977 (Baggoura et al., 1998). The fourth reactor unit went into operation at the end of 1983. On 26 April 1986, an explosion and fire at reactor number 4 caused the largest uncontrolled

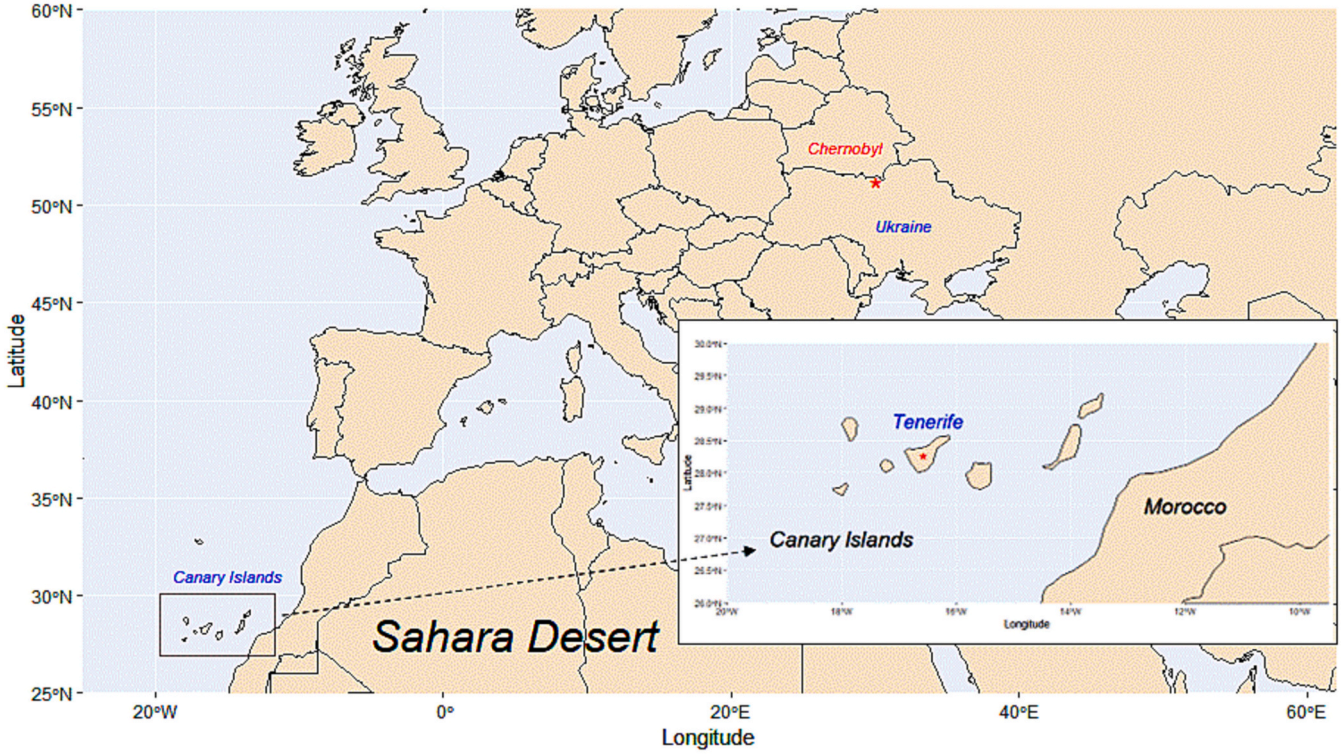

Fig. 1. Geographic location of the Canary Islands. The location of the Chernobyl NPP accident is also indicated for reference.

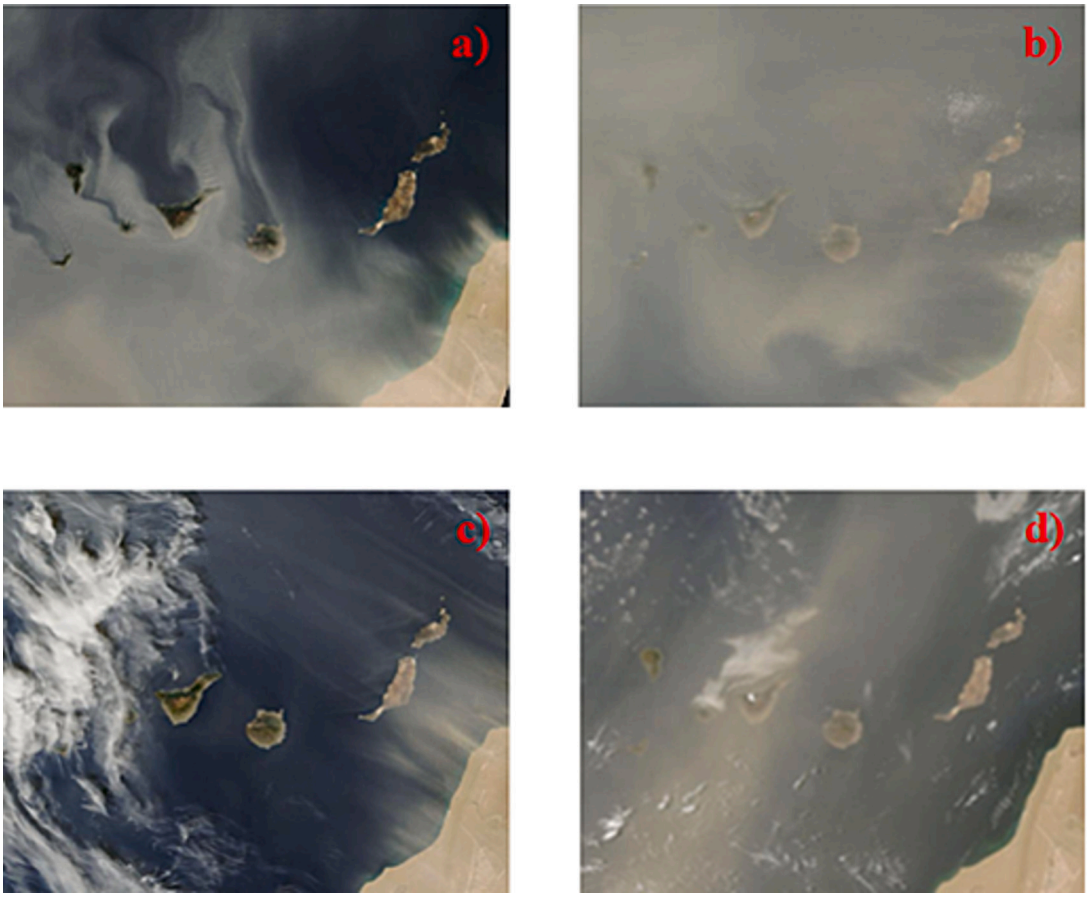

Fig. 2. Moderate Resolution Imaging Spectroradiometers (MODIS) NASA's Terra and Aqua satellites acquired during different dust events crossing the Canary Islands a) 11th February 2001; b) 22nd April 2002; c) 11th November 2006; d) 27th June 2012 (snapshots).

radioactive release in the history of the civil nuclear industry (Evangeliou et al., 2013; Fairlie and Sumner, 2006; Jan, 1996; Kawada and Yamada, 2012; UNSCEAR, 2000; WHO, 2021). Due to the explosion and fire at the Chernobyl NPP, large quantities of both ^{134}Cs and ^{137}Cs isotopes were released into the air. A total of about 44–54 PBq ^{134}Cs and 80–100 PBq ^{137}Cs has been estimated to have been released from Chernobyl according to (Evangeliou et al., 2013; Fairlie and Sumner, 2006; Jan, 1996; Kawada and Yamada, 2012; UNSCEAR, 2008).

The release of radioactive material from the Chernobyl NPP accident lasted approximately 10 days (Kawada and Yamada, 2012). Over that period, due to the changing wind direction and rainfall, the release of the material was widely dispersed and deposited across much of Europe. However, the deposition was very uneven. The largest amounts of material were deposited in Belarus, Ukraine, and Russia but some deposition occurred in most countries within Europe (CRIIRAD, 2017; Fairlie and Sumner, 2006; Kawada and Yamada, 2012). It was even reported that very low levels of radioactive material released from the Chernobyl NPP accident were dispersed throughout the Northern hemisphere and detected as far as Japan and the United States (IAEA, 2006; Kawada and Yamada, 2012). The deposition of ^{137}Cs and ^{134}Cs in Europe one month after Chernobyl appeared to be at least two orders of magnitude, or more, lower than the maximum deposition just after the accident. References to deposition maps and simulations of the dispersion of the radioactive material released during the accident were provided in the introduction section of this paper. None of these suggest a direct deposition path to the Canary Islands immediately after the accident.

1.4. *^{134}Cs and ^{137}Cs isotopes*

^{134}Cs and ^{137}Cs are fission products with half-lives of 2.1 and 30.2 years, respectively. ^{134}Cs is an activation product, while ^{137}Cs is the most common heavy fission product. The chemical properties of the two isotopes are identical so both isotopes behave in the same manner in terms of transport and accumulation. Because of its much shorter half-life, the concentrations of ^{134}Cs decline rapidly in comparison with ^{137}Cs. For example, after 5 years (approximately 2.5 times the half-life of ^{134}Cs) the remaining activity of ^{134}Cs is only 19 % of its initial value. However, for the same period, the remaining activity of the ^{137}Cs is approximately 89 % of its initial value.

^{134}Cs and ^{137}Cs isotopes were released into the environment mainly as the result of the global nuclear weapon testing carried out from the 1950s to the 1970s (Baggoura et al., 1998; Itthipoonthanakorn et al., 2019; McKenzie and Dulai, 2017; UNSCEAR, 1982) and by nuclear power plant accidents, among which, the most relevant were the Chernobyl NPP accident in 1986 (Baggoura et al., 1998; Evangeliou et al., 2013; IAEA, 2006; Jan, 1996; Kawada and Yamada, 2012; Lin et al., 2015; WHO, 2021) and the Fukushima NPP Daiichi accident in 2011 (López-Pérez et al., 2013; Thakur et al., 2013; UNSCEAR, 2000). Because the ratio of ^{134}Cs/^{137}Cs varies on the degree of burn-up of the reactor fuel, a signature ^{134}Cs/^{137}Cs ratio (or vice versa), for these 2 accidents was estimated and reported by various authors (see Table 1). The values for the Fukushima accident are given just for comparison purposes.

^{134}Cs and ^{137}Cs isotopes are of radiological relevance due to the gamma radiation they emit and their bioaccumulation by organisms (Baggoura et al., 1998; Jan, 1996; WHO, 2021). They are chemically analogous to potassium (essential for many organisms) with high mobility in biological systems (Evangeliou et al., 2013). This causes an augmentation of the total radioactivity to which the population is exposed (internal doses mainly caused by ingestion of contaminated

Table 1
$^{134}Cs/^{137}Cs$ signature ratio for the Chernobyl and Fukushima NPP accidents reported by different authors. Values for the Fukushima accident are given for comparison purposes.

Accident	$^{134}Cs/^{137}Cs$ ratio	Reference
Chernobyl	0.4 to 0.7 (average 0.5)	(Thakur et al., 2013)
	0.47–0.55	(Lust and Realo, 2012)
	0.6	(Jan, 1996)
	0.5–0.6	(UNSCEAR, 2000)
Fukushima	1	(UNSCEAR, 2000)
	0.9–1.4	(Baeza et al., 2012)
	0.90 ± 0.11	(Bolsunovsky and Dementyev, 2011; Manolopoulou et al., 2012; Masson et al., 2011)
	0.80–1.35	(López-Pérez et al., 2013)
	0.8–0.9	(Endo et al., 2012; Kinoshita et al., 2011)
	0.90 ± 0.06	(Thakur et al., 2013)

food) (Jan, 1996). In terms of exposure to the population, ^{134}Cs (per unit activity deposited) is about three times greater than ^{137}Cs (Kawada and Yamada, 2012). This is due to the energy of the radiation emitted when ^{134}Cs decays being about three times that for ^{137}Cs. However, the longer half-life of ^{137}Cs makes this isotope a more relevant source from a long-term perspective.

According to Evangeliou et al. (2013), the ecological half-life or residence time of ^{134}Cs and ^{137}Cs in the atmosphere after their release from the Chernobyl accident ranged between 6 and 9 days. This is in good accordance with data collected from the Fukushima NPP accident in Japan, between 5 and 10 days (Evangeliou et al., 2013; Kristiansen et al., 2012), up to 15 days (López-Pérez et al., 2013; Thakur et al., 2013).

For ^{134}Cs, the residence time in soils is limited by its physical half-life (2.1 y). It is therefore expected that this isotope is only present in the top-most layers of the soil where it is deposited. The residence time of ^{137}Cs in soils is longer, and values range widely depending on soil type, season of the year, organic content, and type of vegetation, among other factors (Ager et al., 2019; Burger and Lichtscheidl, 2018). Forest ecosystems contaminated by the Chernobyl NPP accident showed persistent recycling of ^{137}Cs in the topsoil causing vegetation and animals in forests and mountain areas to uptake detectable amounts of this radionuclide (IAEA, 2006). Ager et al. (2019) estimated the residence time of ^{137}Cs in Savannah River floodplain soil and vegetation to be around 17 years. According to Lust and Realo (2012) the residence time for ^{137}Cs in the soil increases from 3.7 years in the top-most compartments (~1 cm) to 36.4 years in deeper compartments (~15–30 cm).

1.5. Resuspension of caesium from soils

There are few studies focused on the resuspension of ^{134}Cs because of its relatively short half-life, e.g. Kinase et al. (2018). However, an analogy can be made based on published ^{137}Cs data. Large-scale resuspension of ^{137}Cs from soils has been reported as the result of either wildfire in areas with contaminated soils or dust storms in arid regions with soils contaminated with this isotope. Details are provided below. The effects of biomass burning have also been reported to contribute to the resuspension of ^{137}Cs to a lesser scale (Strode et al., 2012). Resuspension of Chernobyl-deposited ^{137}Cs has been reported by various authors as the result of forest fires in contaminated regions (Ager et al., 2019; Evangeliou et al., 2014; IAEA, 1991, 2006; Wotawa et al., 2006). This is because ^{137}Cs can be removed from the soils by plants and trees. Pronounced forest fires in 2002, 2010, 2015 and 2020 in Ukraine region and nearby countries remobilised Chernobyl-deposited ^{137}Cs transporting it thousands of kilometres away (Ager et al., 2019; De Meutter et al., 2021; Evangeliou et al., 2014; IAEA, 1991). Resuspension of ^{137}Cs from soils in arid regions has been reported by various authors such as Akata et al. (2007), who studied the atmospheric deposition of ^{137}Cs in Japan and its relationship with Asian dust events. These authors located the arid regions of southern Mongolia and northeast China as the source of the ^{137}Cs. Fukuyama and Fujiwara (2008) found that the deposition of ^{137}Cs from a single Asian dust event was 62.3 $mBq{\cdot}m^{-2}$ and accounted for 67 % of the total ^{137}Cs deposition during their entire monitoring period. Hamadneh et al. (2015) studied the concentrations of radioactive material of seasonal dust storms in the Middle East. Recently, Aba et al. (2018) studied the atmospheric deposition fluxes of ^{137}Cs associated with dust fallout in the north-eastern Arabian Gulf and reported a peak deposition of 50 $Bq{\cdot}m^{-2}$ for ^{137}Cs associated with a single dust storm that hit Kuwait in March 2011. Masson et al. (2011) reported the deposition of ^{137}Cs in France as the result of a large Saharan Dust outbreak that took place in 2004. Pham et al. (2000) also reported the deposition of ^{137}Cs in Monaco in connection with the arrival of sand from North Africa.

Mineral dust from African storms reaching the Canary islands generally contains elements such as iron (Fe) and phosphorous (P), which serve as essential nutrients in marine and terrestrial ecosystems but also radionuclides such as ^{137}Cs (presented herein) and ^{40}K (Hernández et al., 2005; Hernandez et al., 2007; Karlsson et al., 2008; López-Pérez et al., 2020), radon and progeny as well as $^{239+240}Pu$ (presented herein). The environmental radioactivity station FIMERALL (University of La Laguna), located in the MBL at the island of Tenerife, has been monitoring the inputs of ^{137}Cs associated with Saharan dust storms for the last 21 years. This station forms part of the EU's Radioactive Environmental Monitoring (REM) network and its monitoring data can be accessed via (JRC).

2. Materials and methods

2.1. Soil Sampling

A detailed description of the sampling strategy followed in the 1990–1991 and 1994 soil surveys is given in Fernández-Aldecoa et al. (1992) and Fernández de Aldecoa (2000). In summary, 131 locations were sampled from 4 different islands in the first campaign over a period of 9 months and 21 additional locations in the second campaign over a period of 3 months. The size of each sample taken in the two campaigns was 25 cm × 25 cm × 5 cm. Locations that were prompt for soil erosion were avoided during the sampling. All organic material over the soil samples was removed and the soils were dried for 24 h at 110 °C. The material was subsequently sieved to remove stones and then crushed (2 mm mesh). Aliquots of each sample were placed in standard plastic containers used for gamma spectrometry.

2.2. Aerosol sampling

A detailed description of the aerosol sampling carried out at the marine boundary layer at the island of Tenerife between 2001 and 2020 is given in (Hernández et al., 2005; Hernandez et al., 2007; Karlsson et al., 2008; López-Pérez et al., 2020). In summary, aerosols are continuously collected for one-week periods at the monitoring station, located within the Santa Cruz de Tenerife metropolitan area (28° 27′ 18″ N; 16° 17′ 29″ W, 295 m a.s.l.). A Physik Technic Innovation ASS-500 (high-flow station) is used to collect the Total Suspended Particulate (TSP) by making the air flow through polypropylene G3 type square filters (44 cm × 44 cm). The filter used for aerosol collection has no pores, so the particulate matter adheres to the surface without any order (Gordo et al., 2015). The radionuclide aerosol collection efficiency is 93–94 % for the polypropylene filter. TSP matter in the filter was calculated gravimetrically by weighing the filters before and after exposure with a precision balance. The initial sampling rate is approximately 11,500 $l{\cdot}min^{-1}$. At the end of each collection period, a gravimetric analysis is performed to determine the aerosol mass that has accumulated on the filter. Each filter is introduced in a plastic container and kept in a desiccator until measurement with gamma spectrometry 3 days after preparation.

2.3. Gamma spectrometry

All radiometric measurements presented herein were performed by low-level high-resolution gamma spectrometry with coaxial-type germanium detectors (Canberra Industries Inc., USA), with relative efficiency of 20 % and energy resolution (FWHM) of 1.9 keV for the 1.33 MeV ^{60}Co peak. The detectors were shielded with iron blocks to reduce the natural background radiation and regularly calibrated using certified reference gamma-ray cocktails. All measurements were subjected to regular quality control programs including periodical monitoring of the background.

The activity concentrations of ^{134}Cs were measured using the gamma-ray peaks at 605 keV (ca. 98 % intensity) and 796 keV (ca. 85.4 % intensity). The activity concentrations of ^{137}Cs were measured using the gamma-ray peak at 661.65 keV (ca. 85.12 % intensity). The count rates in the full-energy peaks were corrected for the background of the counting system. The average Minimum Detectable Activity (MDA) was 0.5 Bq·kg^{-1} for ^{134}Cs and ^{137}Cs in soil samples as well as 8.1×10^{-4} Bq·m^{-3} for ^{137}Cs in aerosol samples. There are no interfering peaks from natural radionuclides that can cause a false detection of these radionuclides in the samples measured.

The activity concentration measurements of ^{134}Cs and ^{137}Cs in soil samples were performed at the Nuclear Physics Laboratory (Laboratorium voor Kernfysika) of the University of Gent (Belgium) (Fernández-Aldecoa et al., 1992; Fernández de Aldecoa, 2000; Uyttenhove and Poffijn, 1990). The activity concentrations of ^{137}Cs in aerosol samples were measured at the facility of the University of La Laguna, Tenerife, Spain (FIMERALL). The procedure followed at both locations was similar. The FIMERALL Laboratory regularly participates in international and national intercomparison exercises to measure gamma-emitting radionuclides in air filters and soil samples organized by the International Atomic Energy Agency (IAEA), the Joint Research Centre (JRC), and the Spanish Nuclear Safety Council (CSN) (Altzitzoglou et al., 2019; Hult et al., 2019; Máté et al., 2016).

The contour maps of the ^{134}Cs activity concentration for the island of Tenerife presented herein were built by ordinary kriging interpolation. Such probabilistic interpolation methodology is based on the spatial arrangement of the empirical observations and allows to obtain the distribution of the experimental variable in the whole studied area. This method is based on the spatial correlation of the data, semivariogram (Caro et al., 2013). The spatial distribution of ^{134}Cs activity concentrations in soil samples were calculated using the R program (Team_R_Core, 2000). Contour maps of the ^{137}Cs activity concentration for the island of Tenerife, generated also by kriging interpolation, are available in López-Pérez et al. (2021).

2.4. Accelerator Mass Spectrometry

Five aerosol filters, corresponding to weeks with African dust storms, were sent to the Centro Nacional de Aceleradores (CNA) in Seville, Spain (CNA) to determine the concentration of plutonium isotopes by Accelerator Mass Spectrometry (AMS). Two isotopes of Pu were determined separately and together (represented as $^{239+240}$Pu), with half-lives ($T_{1/2}$) of ^{239}Pu and ^{240}Pu being 24,110 y and 6563 y, respectively.

AMS is a technique that allows the determination of long-lived radionuclides that are present in amounts below the detection limits of conventional radiometric and mass spectrometry techniques (Gómez-Camacho et al., 2021). In general, AMS determinations involve the chemical isolation of the element to be studied from the sample and its adjustment to a specific physical-chemical medium. In our case, the plutonium, once chemically isolated, had to be dispersed as Pu_2O_3 in an iron oxide and aluminium matrix for the final preparation of the AMS cathode (Chamizo et al., 2010). Then, the plutonium was extracted from the cathode as PuO in a Cs sputter ion source and stripped to Pu^{3+} in Ar gas at the accelerator terminal.

The 5 aerosol filters were converted to ashes at 600 °C. A ^{242}Pu spike was added on these ashes and homogenized. Finally, leaching was performed with HNO_3 and H_2O_2, and the isolation of the plutonium fraction was achieved with 2 ml prepacked TEVA resins, following the adjustment of the plutonium to Pu(IV) with a two-step redox process (Chamizo et al., 2010). More details about the ^{239,240}Pu measurement technique are reported in Chamizo et al. (2010).

2.5. Calculation of air mass back-trajectories and dust column density

The Modern-Era Retrospective analysis for Research and Applications (MERRA-2) product was used to calculate dust total concentrations. MERRA-2 is a reanalysis database with records since 1980 and is produced by NASA's Global Modeling and Assimilation Office (GMAO). It uses an upgraded version of the Goddard Earth Observing System Model, Version 5 (GEOS-5) data assimilation system (Gelaro et al., 2017). In particular, the variable Dust Column Mass Density (DUCMASS) was used to study the dust intrusions over the Canaries after the Chernobyl NPP accident, specifically in the 1986–1991 period.

To analyse the origin of the air masses that reached the Canary Islands over the last 21 years (2001−2022), 4-day isentropic back-trajectories were computed using the Hysplit-4 dispersion model (Draxler and Rolph, 2003). The back-trajectories were calculated at three heights for the same final point: 300 m (the altitude of the FIMERALL aerosol monitoring station), 1500 m, and 2700 m. The troposphere in this region has a strong stratification and therefore the air masses have different behaviour depending on the height. The height of 1500 m corresponds to the upper limit of the marine boundary layer (MBL) while the height of 2700 m represents the free troposphere (Carrillo et al., 2016; Díaz et al., 2019). To obtain precise results with this software, it was necessary to use accurate meteorological data as input. In this sense, the fifth-generation atmospheric reanalysis of the global climate data (ERA5), computed by the European Centre for Medium-Range Weather Forecasts (ECMWF) was used (Hersbach et al., 2020). Its horizontal and vertical resolutions are $0.25^\circ \times 0.25^\circ$ and 37 pressure levels (1000 hPa to 1 hPa), respectively, with hourly output data from 1970 on a regular grid. The criteria indicated by Díaz et al. (2006) was followed to consider the regions over which the back-trajectories of the air masses moved and loaded with aerosol. Thus, a zone centred on the archipelago was considered and divided into 4 regions: North Atlantic Ocean, MAR; Europe, EUR; Africa, AFR; and Tropical Atlantic Ocean, TRO. A fifth region for the back-trajectories obtained into a circle close to the Canary Islands for the four days was also considered (LOC). The AFR sector included those trajectories that originate or crossed areas of the North African continent. MAR corresponded to maritime air masses with a trajectory over the North Atlantic Ocean. EUR involved air masses that crossed the European continent and the Atlantic Ocean before reaching the FIMERALL station. And finally, TRO, were also marine trajectories but they developed at the southwest of the Canary Islands. Because this area is also frequently affected by Saharan dust intrusions, TRO air masses can occasionally sweep and drag dust to the station.

3. Results and discussion

3.1. ^{134}Cs and ^{137}Cs activity concentrations in soils

The ^{134}Cs and ^{137}Cs activity concentrations, in Bq·kg^{-1} dry weight, measured in the first soil survey carried out in 1990–1991 in the Western Canary Islands are given in Table 2. Only the values for locations where ^{134}Cs was detected above the MDA are given in this table. This data has been extracted from Fernández de Aldecoa (2000) and corrected using the radioactive decay to May 1986 to be able to compare the results (order of magnitude) with other data reported from the Chernobyl NPP accident. There is, however, uncertainty in the applied decay correction, especially for ^{134}Cs activities (short half-life), because we do not know to which date the data from Fernández de Aldecoa (2000) was corrected to.

Table 2
^{134}Cs and ^{137}Cs activity concentrations, in Bq·kg^{-1}, measured in the first soil survey carried out in 1990–1991 in the Western Canary Islands (Fernández de Aldecoa, 2000). Only data for locations where ^{134}Cs was detected above MDA are given. Results are decay-corrected to May 1986. The range of ^{134}Cs/^{137}Cs ratios provided represent the uncorrected data as presented by (Fernández de Aldecoa, 2000) and data corrected to May 1986.

Island	UTM X	UTM Y	^{134}Cs	^{134}Cs Uncertainty	^{137}Cs	^{137}Cs Uncertainty	^{134}Cs/^{137}Cs Range with uncertainty
Tenerife	372,382	3,147,621	11.86	1.53	9.21	0.55	0.36 ± 0.05–1.28 ± 0.18
Tenerife	367,539	3,147,552	24.52	4.53	13.36	1.10	0.53 ± 0.10–1.83 ± 0.37
Tenerife	362,587	3,147,578	14.81	1.56	38.41	0.88	0.10 ± 0.01–0.38 ± 0.04
Tenerife	361,709	3,123,609	19.98	2.61	6.52	0.55	0.77 ± 0.12–3.06 ± 0.47
Tenerife	359,410	3,150,355	7.79	1.56	38.41	1.10	0.05 ± 0.01–0.20 ± 0.04
Tenerife	357,499	3,127,199	17.96	2.24	11.53	0.78	0.38 ± 0.05–1.55 ± 0.22
Tenerife	357,795	3,117,745	33.87	3.04	1.66	0.66	5.2 ± 2.13–20.4 ± 8.37
Tenerife	352,265	3,142,346	9.74	1.95	5.16	0.55	0.53 ± 0.12–1.88 ± 0.42
Tenerife	352,424	3,137,696	11.06	1.98	58.12	0.99	0.05 ± 0.00–0.19 ± 0.03
Tenerife	352,796	3,117,129	25.14	2.69	7.09	0.66	0.87 ± 0.12–3.54 ± 0.50
Tenerife	347,573	3,141,606	9.48	1.98	31.31	1.10	0.08 ± 0.01–0.30 ± 0.06
Tenerife	346,957	3,127,607	15.75	1.75	8.52	0.55	0.46 ± 0.06–1.84 ± 0.23
Tenerife	347,189	3,118,092	30.75	3.56	11.85	1.00	0.64 ± 0.09–2.59 ± 0.37
Tenerife	347,518	3,107,591	20.41	3.40	11.04	1.10	0.48 ± 0.09–1.84 ± 0.35
Tenerife	342,535	3,141,487	14.89	1.21	9.02	0.44	0.45 ± 0.04–1.65 ± 0.15
Tenerife	341,407	3,138,547	10.27	1.19	2.53	0.33	1.13 ± 0.19–4.06 ± 0.70
Tenerife	344,808	3,132,098	26.32	3.76	12.01	0.89	0.51 ± 0.08–2.19 ± 0.35
Tenerife	342,835	3,127,168	28.96	4.01	34.56	1.33	0.20 ± 0.02–0.83 ± 0.12
Tenerife	342,548	3,107,501	25.51	3.40	10.05	0.66	0.65 ± 0.09–2.53 ± 0.37
Tenerife	342,321	3,102,702	13.65	3.52	10.63	0.89	0.32 ± 0.08–1.28 ± 0.34
Tenerife	337,361	3,141,342	16.90	2.82	8.14	0.66	0.56 ± 0.10–2.07 ± 0.38
Tenerife	339,008	3,128,328	21.39	2.73	16.75	0.78	0.31 ± 0.04–1.27 ± 0.17
Tenerife	337,825	3,107,472	15.73	2.13	7.18	0.55	0.56 ± 0.08–2.19 ± 0.34
Tenerife	337,180	3,102,186	16.73	2.20	22.80	1.00	0.18 ± 0.02–0.73 ± 0.10
Tenerife	332,010	3,140,277	14.89	2.01	12.98	0.66	0.31 ± 0.04–1.14 ± 0.16
Tenerife	324,466	3,114,403	13.11	1.59	6.26	0.55	0.57 ± 0.08–2.09 ± 0.31
La Palma	213,444	3,174,360	10.65	1.77	17.17	0.79	0.11 ± 0.02–0.62 ± 0.10
El Hierro	191,261	3,069,274	8.00	1.45	18.33	0.34	0.06 ± 0.01–0.43 ± 0.07

Only records for the sampling dates remain. There are no records of the time of measurement. We have used the known sampling dates to perform the decay correction of the activity concentrations shown in Table 2. For the ^{134}Cs/^{137}Cs ratios, also provided in this table, we have calculated a range of values based on: 1) uncorrected data as published by Fernández de Aldecoa (2000) and 2) data decay- corrected using the sampling date.

The ^{134}Cs activity concentration presented in Table 2 varied between 7.79 and 33.87 Bq·kg^{-1} and ^{137}Cs activity concentration varied between 1.66 and 58.12 Bq·kg^{-1}. The ^{134}Cs activity concentration values presented in Table 2 are higher than the values reported by Llaurado et al. (1994) for mineral soils collected in the NE region of Spain (the area of Spain most impacted by the Chernobyl NPP accident as per (Evangeliou et al., 2013; IRSN, 2021; Yablokov et al., 2006) and higher than the

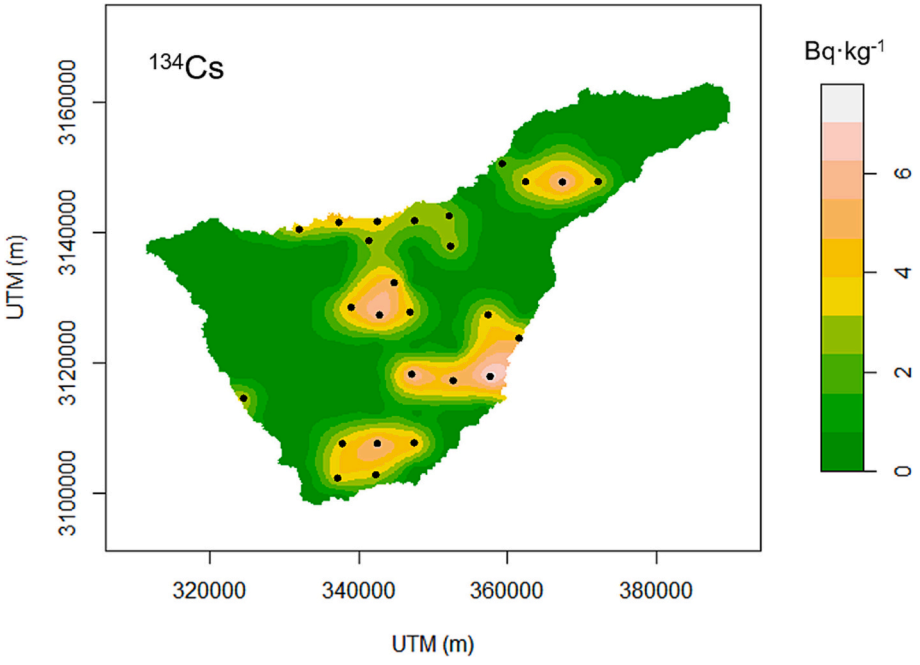

Fig. 3. Spatial distribution of ^{134}Cs activity concentrations in soils collected in the island of Tenerife (Canary Islands, Spain) in 1990–1991. Dots represent sampling sites where the^{134}Cs activity concentrations were above the MDA. All other sampled areas had concentrations below MDA.

values reported for Jordan (CRIIRAD, 2017). No data could be found to compare with other sites in Northern Africa. However, the activity concentrations reported here are much lower than the activity concentrations of other European sites that had direct deposition of ^{134}Cs immediately after the Chernobyl accident. For example, activity concentrations above 1000 Bq·kg^{-1} were recorded in various locations in Eastern France (CRIIRAD, 2017).

A map for the island of Tenerife with the activity concentrations of ^{134}Cs is displayed in Fig. 3, using kriging interpolation. Tenerife is the largest of the studied islands and it had most of the data points. The activity measured in samples from La Gomera were all below MDA (approximately 0.5 Bq·kg^{-1}), whereas only one sample collected in La Palma and one in El Hierro exhibit activity above MDA. A map of the island of Tenerife with ^{137}Cs activity concentrations measured in the first soil survey in 1990–1991 can be found in Fernández de Aldecoa (2000) and a more recent survey, 2013, in López-Pérez et al. (2021). The different spatial distributions of ^{134}Cs and ^{137}Cs in the soils of this island do not seem to indicate that remobilisation of soil by erosion may explain the range of activity values reported in Table 2.

The range of values obtained for the ^{134}Cs/^{137}Cs ratios shown in Table 2 cannot be used to confirm the source of the ^{134}Cs because of the large uncertainty related to the decay correction. The high energy resolution of the Germanium detectors used to identify and measure the ^{134}Cs in the soil samples allowed to resolve the main gamma peaks from this isotope from other interfering peaks and background subtraction were performed. Thus, we can be certain that ^{134}Cs was detected on the soils at this site. Based on the year when the soil samples were collected in the first campaign (1990–1991), the limited number of sources of ^{134}Cs to the global environment at the time and the short half-life of this isotope, the most logical source of the ^{134}Cs activity concentrations recorded in the Western Canary Islands was the Chernobyl NPP accident (1986).

In terms of areal activities, the ^{134}Cs activity concentrations displayed in Table 2 correspond to values ranging between 400 and 1700 Bq·m^{-2}. This is assuming that all the ^{134}Cs was deposited in the top 5 cm of soil. These values are, for example, higher than those reported by De Cort et al. (1998) for Algeria (average of 200 Bq·m^{-2}, estimated from the average ^{137}Cs deposition multiplied by the 0.5 ratio applicable to Chernobyl NPP accident) and Spain mainland (<1 kBq·m^{-2}, estimated also by applying the Chernobyl 0.5 ratios) (UNSCEAR, 2000). This seems to indicate that the presence of ^{134}Cs in the soils of the Western Canary Islands may not be the result of direct atmospheric deposition immediately after the Chernobyl NPP accident.

A comparison of the ^{137}Cs activity concentrations determined in soils of the Western Canary Islands by López-Pérez et al. (2021) showed that the values recorded in samples collected in 2013 had not differed significantly from those in samples collected in 1991. It was argued that, despite the 22-year period between the two sampling surveys, the concentrations had not changed much due to regular transport and deposition of ^{137}Cs to the soils at these islands in association with the arrival of Saharan dust events. The maximum areal activity reported in that paper for this site (decay corrected to 2013), 11 kBq·m^{-2}, was found to be higher than the expected bomb-derived fallout for the northern hemisphere (20°–30° latitude band) (UNSCEAR, 1982), higher than values reported for Spain mainland, 1.5 kBq·m^{-2} by Caro et al. (2013) and comparable to values found in Austria (16 kBq·m^{-2}) (Bossew et al., 2001).

3.2. Activity concentrations in aerosol samples

Fig. 4 displays the weekly activity concentration of ^{137}Cs, in Bq·m^{-3}, determined in aerosol samples, collected at the FIMERALL sampling station over a period of 20 years (2001–2021). Of the measurements collected over the years, approximately 90 % of the data points are correlated with Saharan dust events (Hernández et al., 2005; Hernandez et al., 2007; Karlsson et al., 2008; López-Pérez et al., 2020). Data corresponding to the arrival of ^{137}Cs plume from the Fukushima NPP accident in 2011, with activity levels >40 times higher than those from African events, can be found in López-Pérez et al. (2021) and are not depicted in this Figure. Fig. 4 shows the regular arrival of aerosols loaded with ^{137}Cs to the Canary Islands (at the marine boundary layer).

Saharan dust events impacting the MBL at the Canary Islands are common in the autumn-winter periods, while dust events at higher altitudes are more common over the summer periods (Gelado-Caballero et al., 2012; Karlsson et al., 2008; López-Pérez et al., 2020). Therefore, the data shown in Fig. 4 may underestimate the amount of ^{137}Cs that is deposited in locations in the Western Canary Islands which are above approximately 1000 m a.s.l. (top of the MBL). Because the nature of each dust event is different and ^{137}Cs concentrations differ depending on the main source of aerosols, altitude of the intrusion, duration, wind speed, etc., no estimation of a deposition rate for this isotope is possible from this data.

Saharan dust events reaching the Canary Islands can last from a few hours to several days. Fig. 5, shows 4 clusters of 4-day back-trajectories reaching the islands of Tenerife at noon at an altitude of 300 m a.s.l. These 4 clusters were associated with different Saharan dust events that reached the island during the monitored period. The specific dates for these four dust events are given at the top of each plot and the exact day

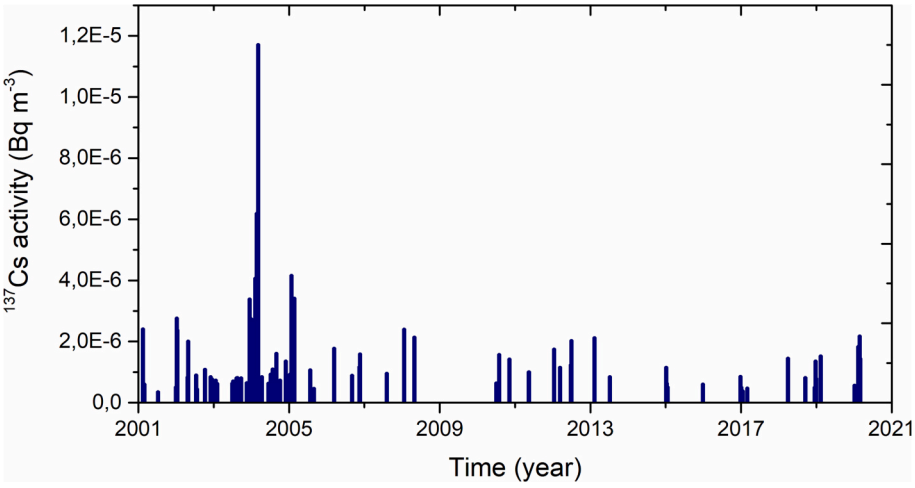

Fig. 4. Weekly activity concentration of ^{137}Cs, in Bq·m^{-3} in filtered air, determined in aerosol samples, collected at the FIMERALL sampling station over a period of 20 years. This sampling station is located in the marine boundary layer at 300 m a.s.l. in the island of Tenerife (Canary Islands, Spain).

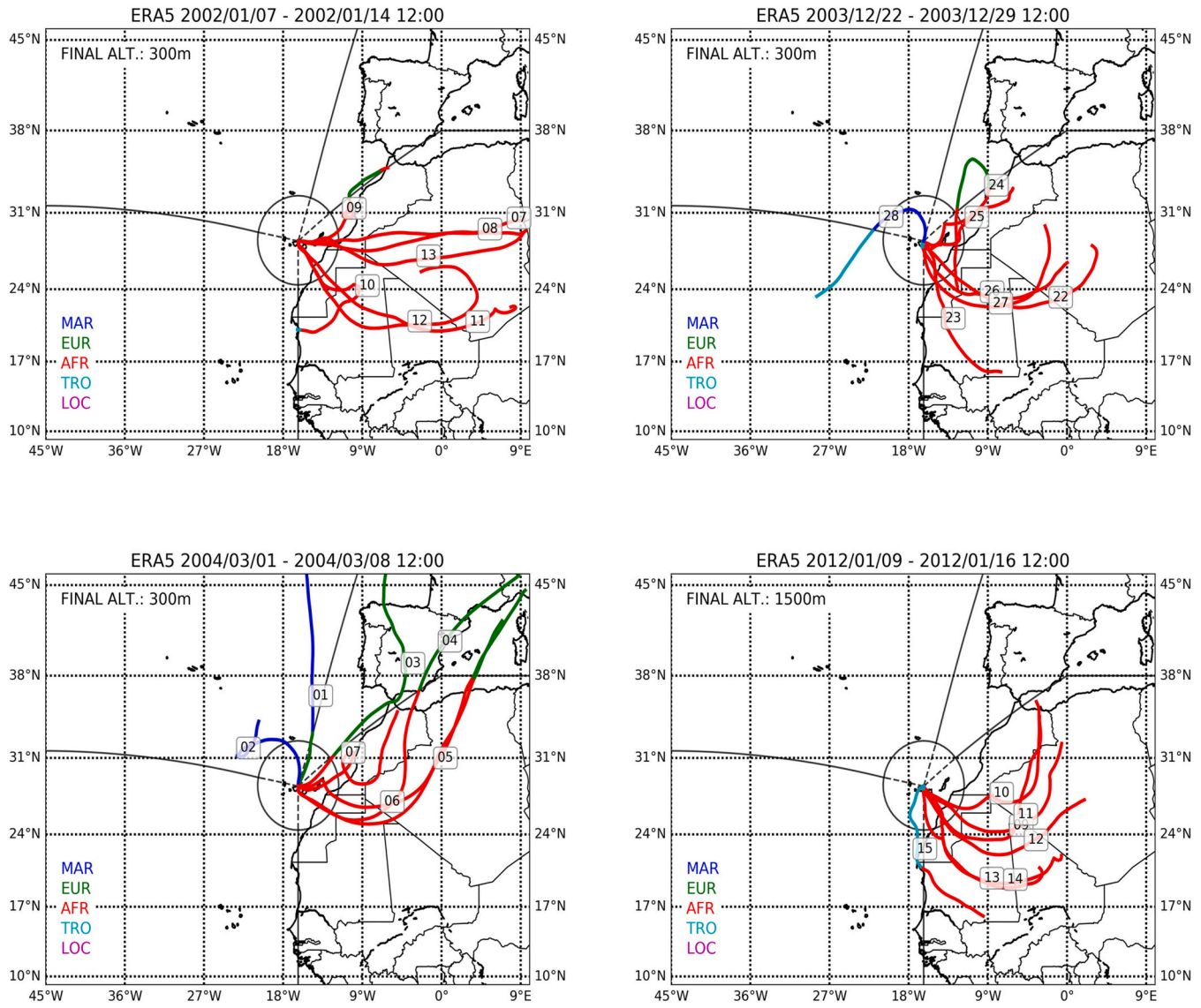

Fig. 5. Examples of 4-day back-trajectories reaching the island of Tenerife at noon at 300 m a.s.l. calculated for periods when Saharan dust events took place: a) 7th–14th January 2002; b) 22nd–29th December 2003; c) 1st–8th March 2004 and d) 9th–16th January 2020.

for each of the back-trajectories is given next to each curve. These are just some examples of what has been observed over the last 20 years. These intrusions of Saharan sand caused the concentrations of particulate matter in the atmosphere above the sampling region (PM10, particle size >10 nm) to increase from background levels of approximately 40 $\mu g \cdot m^{-3}$ to values over 100 $\mu g \cdot m^{-3}$ (GOBCAN, 2021). In the March 2004 event, the PM10 concentrations at this site reached values above 1000 $\mu g \cdot m^{-3}$. The highest PM10 concentration recorded so far at this site was over 3000 $\mu g \cdot m^{-3}$ and corresponded to an African dust event that took place in February 2020 (López-Pérez et al., 2020). All the red colour trajectories shown in Fig. 6 indicate that the air masses crossed the Africa region (blue trajectories denote Atlantic zone, green is for Europe and cyan for the southwestern Atlantic). In all cases, the African air masses crossed Algeria. We know from De Cort et al. (1998) that Chernobyl-related ^{134}Cs and ^{137}Cs was deposited in Algeria. It is, therefore, possible that the resuspension of soils from this region in the following years after the Chernobyl NPP accident may have been transported and caused the deposition of ^{134}Cs and ^{137}Cs over the Canary Islands.

Fig. 6 shows the dust column density data, in $g \cdot m^{-2}$, during the 1986–1991 period (after the Chernobyl accident and prior to the soil survey), obtained from the database MERRA-2. This data supports our hypothesis. In those years, several intense dust events were recorded during winter in the Canary Islands, e.g. February 1989 (Torre Estupiñán and Dáviña Tovar, 1995). Antequera et al. (2015) studied the frequency, seasonality and trends of the Saharan air advections in the Canary Islands (1976–2003). They observed that the total amount of lithogenic material that reached the islands was very high, with average daily values per year that exceeded 50 $\mu g \cdot m^{-3}$, with winter being the time when transport was most relevant. Although the majority of the dust events resulted in dry deposition, on some occasions, precipitation enhanced the deposition of mineral dust "rain of mud" (Criado and Dorta, 2003).

Plutonium isotopes (^{239}Pu, ^{2240}Pu and $^{239+240}Pu$) were determined in 5 aerosol filters collected during 5 different Saharan dust events that took place between 2018 and 2020. The $^{239+240}Pu/^{137}Cs$ and $^{240}Pu/^{239}Pu$ ratios were determined for these samples and the results are shown in Table 3. Because of the long half-lives of plutonium isotopes, no decay correction was necessary. However, the activity concentrations of ^{137}Cs were corrected for radioactive decay to 1998 to be able to compare this data with the results from Baggoura et al. (1998). The $^{239+240}Pu/^{137}Cs$ ratios obtained in the aerosol filters ranged between

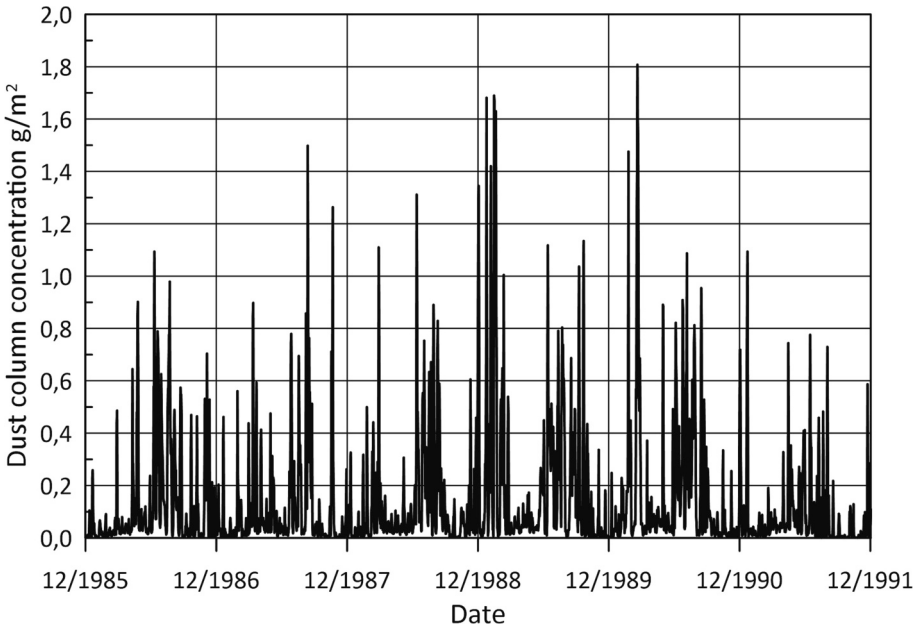

Fig. 6. Dust column mass density (DUCMASS) at the Canary Islands from 1986 to 1991.

Table 3
^{239}Pu, ^{240}Pu, $^{239+240}$Pu and ^{137}Cs activity concentrations, in μBq·g^{-1}, and the corresponding ratio for 5 aerosol filters collected at the monitoring station of FIMERALL during different Saharan dust events. The dates for each of the sampling periods are given in the table. The ^{137}Cs activity concentrations were corrected for decay to 1998. Back-trajectories for these events are given in Fig. 7.

Sampling period	Air volume (m^3)	TSP mass (g)	^{239}Pu	U	^{249}Pu	U†	$^{239+240}$Pu	U	^{137}Cs (corrected to 1998)	U†	^{240}Pu/^{239}Pu ratio	U†	$^{239+240}$Pu/^{137}Cs ratio	U†
2018-03-26/ 2018-04-02	71,552	11.31	206	4	143	5	349	6	14,409.9	1537.0	0.189	0.007	0.024	0.004
2018-09-10/ 2018-09-17	84,563	10.99	170	5	114	4	283	6	9886.1	1716.6	0.182	0.009	0.029	0.004
2019-02-04/ 2019-02-11	56,509	6.83	245	7	169	6	414	9	20,207.7	3817.0	0.187	0.008	0.020	0.002
2020-02-03/ 2020-02-10	50,176	11.89	168	6	102	7	270	9	12,660.4	2541.9	0.165	0.013	0.021	0.004
2020-02-24/ 2020-03-02	57,973	9.57	190	6	120	6	311	8	14,318.8	2610.9	0.172	0.010	0.022	0.003

† Uncertainty (U) is reported with a coverage factor of k = 2, indicating approximately 95 % confidence.

0.021 and 0.029. Baggoura et al. (1998) reported a ratio of 0.02 in the soils collected in Argelia after the Chernobyl NPP accident and 0.04 for the soils collected and measured before the accident. Our results are closer to the post-Chernobyl values than those measured before the accident. This indicates that the source/s of ^{137}Cs captured in the aerosol filters collected at the FIMERALL station, is not solely due to resuspended nuclear bombs fallout. Chernobyl deposited ^{137}Cs and contamination from the French nuclear tests carried out in Algeria in the 60s and 70s are also likely contributors to the transported ^{137}Cs (Menut et al., 2009). This is confirmed also by the recorded ^{240}Pu/^{239}Pu ratios below 0.18 for two of the selected weeks, when the air masses travelled over Algeria where the French nuclear tests were conducted (see Fig. 7; except for the week of September 10–17, 2018, the rest have at least one day where the back-trajectories show that they have passed through Algeria at all three altitudes studied). The other 3 weeks have ratios of 0.18 indicating that the resuspended material is fallout ^{137}Cs.

The results presented in Table 3 indicate that it is possible that Chernobyl-related ^{134}Cs, previously deposited in Algeria and other countries in Northern Africa, could have been resuspended, transported, and deposited in the Western Canary Island during the years that followed the NPP accident. This would explain why ^{134}Cs activity concentrations were measured in the soil samples collected in 1991 at these islands.

4. Conclusion

A radiometric survey carried out in the Western Canary Islands in 1990–1991 reported the presence of ^{134}Cs in some of the soils collected in three of the 4 investigated islands. ^{134}Cs released to the global environment by atmospheric nuclear tests up to the 1970s had already decayed by 1990 due to the relatively short half-life of this isotope. Despite the large distance between the studied site and the location of the nuclear plant at Chernobyl, the most likely source for the ^{134}Cs activity concentrations recorded at the studied site was the Chernobyl NPP accident that occurred in 1986 (4 years earlier). Isotopes released during the Chernobyl accident, in accordance with the available literature, were scattered mostly over Europe. However, small amounts were also dispersed all over the northern hemisphere. Simulations of the radioactive plumes released by the Chernobyl NPP accident did not show a direct deposition path to the Western Canary Islands during the days that followed the accident. We believe that the most likely scenario was that the isotopes were first deposited in the soil surface over Northern Africa after the accident and then resuspended into the atmosphere, transported, and deposited over the islands by dust storms (secondary

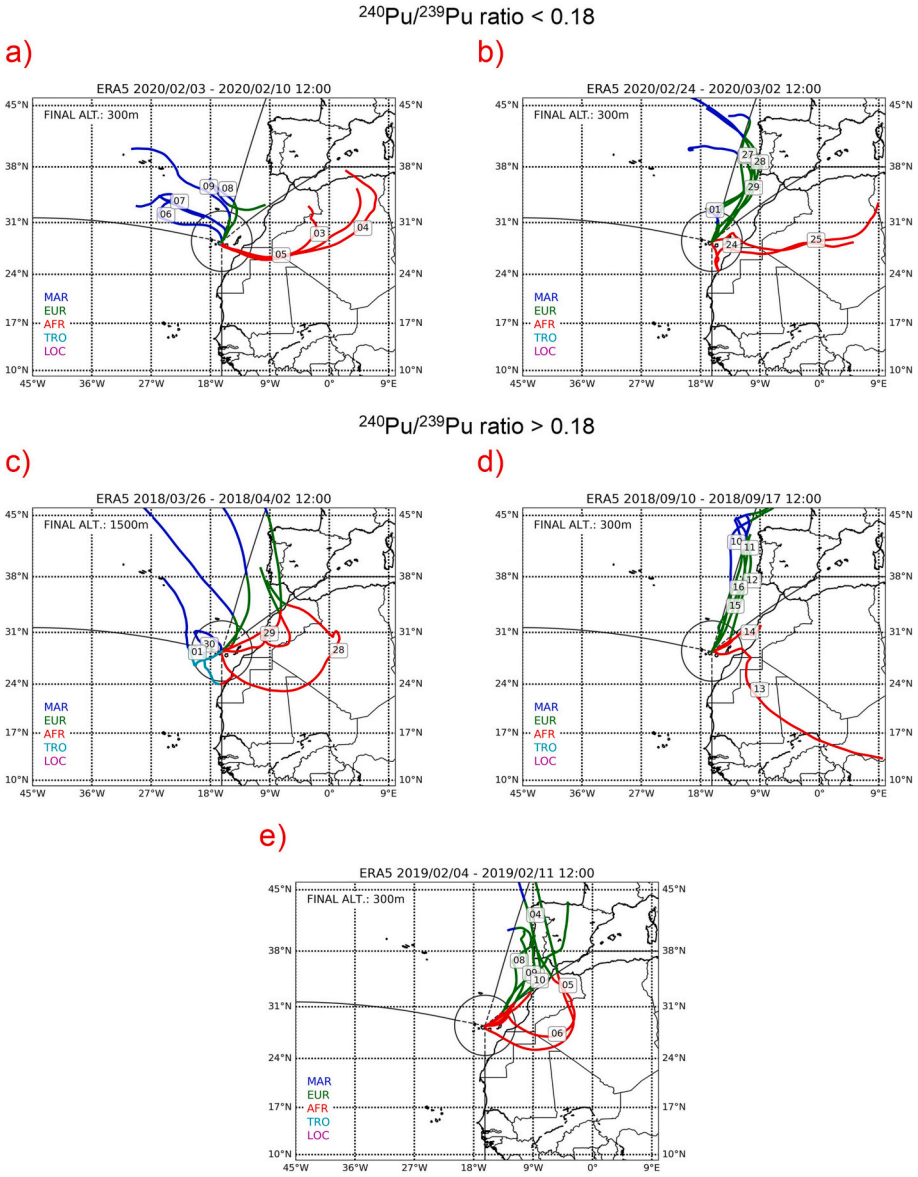

Fig. 7. 4-day return trajectories arriving at Tenerife Island at noon at 300 m a.s.l. calculated for periods in which polonium isotopes were determined: a) 3rd–10th, 2020 February; b) 24th February–2nd March 2020; c) 26th March–2nd April 2018; d) 10th–17th September 2018 and e) 4th–11th February 2019.

deposition). This kind of dust storms are common in northern Africa. Aerosol data collected at the Canary Islands over the last 20 years (2001–2021) and dust column density data for the 1986–1991 period show that there is a continuous input of mineral dust and ^{137}Cs from Northern Africa associated with the dust storms and supports the scenario described above.

CRediT authorship contribution statement

María López-Pérez: Investigation, Methodology, Formal analysis, Writing – original draft, Supervision. **Francisco Hernández:** Investigation, Formal analysis, Writing – original draft. **Esperanza Liger:** Investigation, Formal analysis, Writing – original draft. **Elisa Gordo:** Investigation, Formal analysis, Writing – original draft. **José Carlos Fernández-Aldecoa:** Methodology, Investigation, Formal analysis, Writing – original draft. **Francisco Javier Expósito:** Investigation, Methodology, Formal analysis, Writing – original draft. **Juan Pedro Díaz:** Investigation, Formal analysis, Writing – original draft. **José Hernández-Armas:** Methodology, Investigation, Formal analysis, Writing – original draft. **Pedro A. Salazar-Carballo:** Investigation, Methodology, Formal analysis, Writing – original draft, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

Aba, A., Al-Dousari, A.M., Ismaeel, A., 2018. Atmospheric deposition fluxes of ^{137}Cs associated with dust fallout in the northeastern Arabian Gulf. J. Environ. Radioact. 192, 565–572.

Ager, A.A., Lasko, R., Myroniuk, V., Zibtsev, S., Day, M.A., Usenia, U., Bogomolov, V., Kovalets, I., Evers, C.R., 2019. The wildfire problem in areas contaminated by the Chernobyl disaster. Sci. Total Environ. 696, 133954.

Akata, N., Hasegawa, H., Kawabata, H., Chikuchi, Y., Sato, T., Ohtsuka, Y., Kondo, K., Hisamatsu, S.i., 2007. Deposition of ^{137}Cs in Rokkasho, Japan and its relation to Asian dust. J. Environ. Radioact. 95, 1–9.

Alonso-Pérez, S., Cuevas, E., Querol, X., Viana, M., Guerra, J.C., 2007. Impact of the Saharan dust outbreaks on the ambient levels of total suspended particles (TSP) in the marine boundary layer (MBL) of the Subtropical Eastern North Atlantic Ocean. Atmos. Environ. 41, 9468–9480.

Altzitzoglou, T., Hult, M., De Cort, M., 2019. Evaluation of the 2016 ENV57 MetroERM measurement comparison on 137Cs, 134Cs and 131I in air filters. Appl. Radiat. Isot. 143, 123–131.

Baeza, A., Corbacho, J.A., Rodríguez, A., Galván, J., García-Tenorio, R., Manjón, G., Mantero, J., Vioque, I., Arnold, D., Grossi, C., Serrano, I., Vallés, I., Vargas, A., 2012. Influence of the Fukushima Dai-ichi nuclear accident on spanish environmental radioactivity levels. J. Environ. Radioact. 114, 138–145.

Baggoura, B., Noureddine, A., Benkrid, M., 1998. Level of natural and artificial radioactivity in Algeria. Appl. Radiat. Isot. 49, 867–873.

Bolsunovsky, A., Dementyev, D., 2011. Evidence of the radioactive fallout in the center of Asia (Russia) following the Fukushima Nuclear Accident. J. Environ. Radioact. 102, 1062–1064.

Bossew, P., Ditto, M., Falkner, T., Henrich, E., Kienzl, K., Rappelsberger, U., 2001. Contamination of austrian soil with caesium-137. J. Environ. Radioact. 55, 187–194.

Burger, A., Lichtscheidl, I., 2018. Stable and radioactive cesium: a review about distribution in the environment, uptake and translocation in plants, plant reactions and plants' potential for bioremediation. Sci. Total Environ. 618, 1459–1485.

Caro, A., Legarda, F., Romero, L., Herranz, M., Barrera, M., Valiño, F., Idoeta, R., Olondo, C., 2013. Map on predicted deposition of Cs-137 in spanish soils from geostatistical analyses. J. Environ. Radioact. 115, 53–59.

Carrillo, J., Guerra, J.C., Cuevas, E., Barrancos, J., 2016. Characterization of the Marine Boundary Layer and the Trade-Wind Inversion over the Sub-tropical North Atlantic. Bound.-Layer Meteorol. 158, 311–330.

Criado, C., Dorta, P., 2003. An unusual 'blood rain' over the Canary Islands (Spain). The storm of January 1999. J. Arid Environ. 55, 765–783.

CRIIRAD, 2017. http://www.criirad.org/english/presentation.html.

Cuevas, E., Milford, C., Barreto, A., Bustos, J.J., García, R.D., Marrero, C.L., Prats, N., Bayo, C., Ramos, R., Terradellas, E., Suárez, D., Rodríguez, S., De la Rosa, J., Vilches, J., Basart, S., Werner, E., López-Villarrubia, E., Rodríguez-Mireles, S., Pita Toledo, M.L., González, O., Belmonte, J., Puigdemunt, R., Lorenzo, J.A., Oromí, P., R, D.C.-H., 2021. Desert Dust Outbreak in the Canary Islands (February 2020): Assessment and Impacts.

Chamizo, E., García-León, M., Enamorado, S.M., Jiménez-Ramos, M.C., Wacker, L., 2010. Measurement of plutonium isotopes, 239Pu and 240Pu, in air-filter samples from Seville (2001–2002). Atmos. Environ. 44, 1851–1858.

Torre Estupiñán, C.G., Dáviña Tovar, M.P., 1995. Estudio de las lluvias torrenciales acaecidas en febrero de 1989 en Lanzarote. In: IV Jornadas de Estudios sobre Lanzarote y Fuerteventura. Cabildo Insular de Lanzarote y Cabildo Insular de Fuerteventura, Arrecife.

De Cort, M., Dubois, G., D, F., Germenchuk, M., A, I., Janssens, A., Jones, A., N, K., Knaviskova, E., I, M., M, N., Ym, P., Va, S., Ed, S., Ly, T., Ys, T., 1998. Atlas of Caesium 137 Deposition on Europe After the Chernobyl Accident. EUR 1673 EN/RU.

De Meutter, P., Gueibe, C., Tomas, J., Outer, P.D., Apituley, A., Bruggeman, M., Camps, J., Delcloo, A., Knetsch, G.-J., Roobol, L., Verheyen, L., 2021. The assessment of the April 2020 chernobyl wildfires and their impact on Cs-137 levels in Belgium and The Netherlands. J. Environ. Radioact. 237, 106688.

Díaz, A.M., Díaz, J.P., Expósito, F.J., Hernández-Leal, P.A., Savoie, D., Querol, X., 2006. Air masses and aerosols chemical components in the free troposphere at the subtropical northeast atlantic region. J. Atmos. Chem. 53, 63–90.

Díaz, J.P., Expósito, F.J., Pérez, J.C., González, A., Wang, Y., Haimberger, L., Wang, J., 2019. Long-term trends in marine boundary layer properties over the Atlantic Ocean. J. Clim. 32, 2991–3004.

Draxler, R.R., Rolph, G.D., 2003. Hysplit (Hybrid Single-Particle Lagrangian Integrated Trajectory) Model.

EEA, European Enviroment Agency, 2021. https://www.eea.europa.eu/data-and-maps/figures/deposition-from-chernobyl-in-europe, 09-16.

Endo, S., Kimura, S., Takatsuji, T., Nanasawa, K., Imanaka, T., Shizuma, K., 2012. Measurement of soil contamination by radionuclides due to the Fukushima Dai-ichi Nuclear Power Plant accident and associated estimated cumulative external dose estimation. J. Environ. Radioact. 111, 18–27.

Evangeliou, N., Balkanski, Y., Cozic, A., Hao, W.M., Møller, A.P., 2014. Wildfires in Chernobyl-contaminated forests and risks to the population and the environment: a new nuclear disaster about to happen? Environ. Int. 73, 346–358.

Evangeliou, N., Balkanski, Y., Cozic, A., Møller, A.P., 2013. Simulations of the transport and deposition of 137Cs over Europe after the Chernobyl Nuclear Power Plant accident: influence of varying emission-altitude and model horizontal and vertical resolution. Atmos. Chem. Phys. 13, 7183–7198.

Fairlie, I., Sumner, D., 2006. The Other Report on Chernobyl (TORCH). Kiev, Berlin, Brussels.

Fernández-Aldecoa, J.C., Robayna, B., Allende, A., Poffijn, A., Hernández-Armas, J., 1992. Natural Radiation in Tenerife (Canary Islands). Radiat. Prot. Dosim. 45, 545–548.

Fernández de Aldecoa, J.C., 2000. Radiación natural en aire y suelos de las Islas Canarias Occidentales. Universidad de La Laguna.

Fukuyama, T., Fujiwara, H., 2008. Contribution of Asian dust to atmospheric deposition of radioactive cesium (^{137}Cs). Sci. Total Environ. 405, 389–395.

Gelado-Caballero, M.D., López-García, P., Prieto, S., Patey, M.D., Collado, C., Hérnández-Brito, J.J., 2012. Long-term aerosol measurements in Gran Canaria, Canary Islands: particle concentration, sources and elemental composition. J. Geophys. Res. Atmos. 117.

Gelaro, R., McCarty, W., Suárez, M.J., Todling, R., Molod, A., Takacs, L., Randles, C.A., Darmenov, A., Bosilovich, M.G., Reichle, R., Wargan, K., Coy, L., Cullather, R., Draper, C., Akella, S., Buchard, V., Conaty, A., da Silva, A.M., Gu, W., Kim, G.-K., Koster, R., Lucchesi, R., Merkova, D., Nielsen, J.E., Partyka, G., Pawson, S., Putman, W., Rienecker, M., Schubert, S.D., Sienkiewicz, M., Zhao, B., 2017. The modern-era retrospective analysis for research and applications, version 2 (MERRA-2). J. Clim. 30, 5419–5454.

GOBCAN, 2021. Gobierno de Canarias, Red de Control y Vigilancia de la Calidad del Aire de Canarias. https://www3.gobiernodecanarias.org/medioambiente/calidaddelaire/inicio.do.

Gómez-Camacho, J., García López, J., Guerrero, C., López Gutiérrez, J.M., García-Tenorio, R., Santos-Arévalo, F.J., Chamizo, E., Ferrer, F.J., Jiménez-Ramos, M.C., Balcerzyk, M., Fernández, B., 2021. Research facilities and highlights at the Centro Nacional de Aceleradores (CNA). Eur. Phys. J. Plus 136, 273.

Gordo, E., Dueñas, C., Fernández, M.C., Liger, E., Cañete, S., 2015. Behavior of ambient concentrations of natural radionuclides (7)Be, (210)Pb, (40)K in the Mediterranean coastal city of Málaga (Spain). Environ. Sci. Pollut. Res. Int. 22, 7653–7664.

Goudie, A.S., Middleton, N.J., 2001. Saharan dust storms: nature and consequences. Earth Sci. Rev. 56, 179–204.

Hamadneh, H.S., Ababneh, Z.Q., Hamasha, K.M., Ababneh, A.M., 2015. The radioactivity of seasonal dust storms in the Middle East: the May 2012 case study in Jordan. J. Environ. Radioact. 140, 65–69.

Hernández, F., Hernández-Armas, J., Catalán, A., Fernández-Aldecoa, J.C., Karlsson, L., 2005. Gross alpha, gross beta activities and gamma emitting radionuclides composition of airborne particulate samples in an oceanic island. Atmos. Environ. 39, 4057–4066.

Hernandez, F., Karlsson, L., Hernandez-Armas, J., 2007. Impact of the tropical storm Delta on the gross alpha, gross beta, 90Sr, 210Pb, 7Be, 40K and 137Cs activities measured in atmospheric aerosol and water samples collected in Tenerife (Canary Islands). Atmos. Environ. 41, 4940–4948.

Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., Simmons, A., Soci, C., Abdalla, S., Abellan, X., Balsamo, G., Bechtold, P., Biavati, G., Bidlot, J., Bonavita, M., De Chiara, G., Dahlgren, P., Dee, D., Diamantakis, M., Dragani, R., Flemming, J., Forbes, R., Fuentes, M., Geer, A., Haimberger, L., Healy, S., Hogan, R.J., Hólm, E., Janisková, M., Keeley, S., Laloyaux, P., Lopez, P., Lupu, C., Radnoti, G., de Rosnay, P., Rozum, I., Vamborg, F., Villaume, S., Thépaut, J.-N., 2020. The ERA5 global reanalysis. Q. J. R. Meteorol. Soc. 146, 1999–2049.

Hult, M., Jobbágy, V., Sobiech-Matura, K., Centre, E.C.J.R., 2019. Radioactivity Monitoring: How the JRC Verifies Results From Monitoring Within the European Union: A Quick Guide. Publications Office of the European Union.

IAEA, 1991. International Atomic Energy Agency, Seminar on Comparative Assessment of the Environmental Impact of Radionuclides Released During Three Major Nuclear Accidents. Kyshtym, Windscale, Chernobyl, Luxembourg (Luxembourg).

IAEA, 2006. International Atomic Energy Agency, Environmental Consequences of the Chernobyl Accident and Their Remediation: Twenty Years of Experience, Radiological Assessment Reports Series No. 8 Vienna.

IRSN, Institut de Radioprotection et de Sûreté Nucléaire, 2021. https://www.irsn.fr/en/publications/thematic-safety/chernobyl/pages/the-chernobyl-plume.aspx, 09-16.

Itthipoonthanakorn, T., Dann, S.E., Crout, N.M.J., Shaw, G., 2019. Nuclear weapons fallout 137Cs in temperate and tropical pine forest soils, 50 years post-deposition. Sci. Total Environ. 660, 807–816.

Jan, O.S., 1996. 1996:08 Contamination and Radiation Exposure, Evaluation and Measures in the Nordic Countries after the Chernobyl Accident. SSI.

Karlsson, L., Hernandez, F., Rodríguez, S., López-Pérez, M., Hernandez-Armas, J., Alonso-Pérez, S., Cuevas, E., 2008. Using 137Cs and 40K to identify natural Saharan dust contributions to PM10 concentrations and air quality impairment in the Canary Islands. Atmos. Environ. 42, 7034–7042.

Kawada, Y., Yamada, T., 2012. Radioactivity ratios of 134Cs/137Cs released by the nuclear accidents. Isotope News 16–20.

Kinase, T., Kita, K., Igarashi, Y., Adachi, K., Ninomiya, K., Shinohara, A., Okochi, H., Ogata, H., Ishizuka, M., Toyoda, S., Yamada, K., Yoshida, N., Zaizen, Y., Mikami, M., Demizu, H., Onda, Y., 2018. The seasonal variations of atmospheric 134,137Cs activity and possible host particles for their resuspension in the contaminated areas of Tsushima and Yamakiya, Fukushima, Japan. Prog Earth Planet Sci 5, 12.

Kinoshita, N., Sueki, K., Sasa, K., Kitagawa, J.-I., Ikarashi, S., Nishimura, T., Wong, Y.-S., Satou, Y., Handa, K., Takahashi, T., Sato, M., Yamagata, T., 2011. Assessment of individual radionuclide distributions from the Fukushima nuclear accident covering central-East Japan. Proc. Natl. Acad. Sci. 108, 19526.

Knippertz, P., Todd, M.C., 2012. Mineral dust aerosols over the Sahara: meteorological controls on emission and transport and implications for modeling. Rev. Geophys. 50.

Kristiansen, N.I., Stohl, A., Wotawa, G., 2012. Atmospheric removal times of the aerosol-bound radionuclides 137Cs and 131I measured after the Fukushima Dai-ichi nuclear accident – a constraint for air quality and climate models. Atmos. Chem. Phys. 12, 10759–10769.

Llaurado, M., Vidal, M., Rauret, G., Roca, C., Fons, J., Vallejo, V.R., 1994. Radiocaesium behaviour in Mediterranean conditions. J. Environ. Radioact. 23, 81–100.

Lin, W., Chen, L., Yu, W., Ma, H., Zeng, Z., Lin, J., Zeng, S., 2015. Radioactivity impacts of the Fukushima Nuclear Accident on the atmosphere. Atmos. Environ. 102, 311–322.

López-Pérez, M., Lorenzo-Salazar, J.M., Expósito, F.J., Díaz, J.P., Salazar, P., 2020. Impact of a massive dust storm on the gross alpha, gross beta, 40K, 137Cs, 210Pb, 7Be activities measured in atmospheric aerosols collected in Tenerife, Canary Islands. Atmos. Environ. 239, 117806.

López-Pérez, M., Martín-Luis, C., Hernández, F., Liger, E., Fernández-Aldecoa, J.C., Lorenzo-Salazar, J.M., Hernández-Armas, J., Salazar-Carballo, P.A., 2021. Natural and artificial gamma-emitting radionuclides in volcanic soils of the Western Canary Islands. J. Geochem. Explor. 229, 106840.

López-Pérez, M., Ramos-López, R., Perestelo, N.R., Duarte-Rodriguez, X., Bustos, J.J., Alonso-Pérez, S., Cuevas, E., Hernández-Armas, J., 2013. Arrival of radionuclides released by the Fukushima accident to Tenerife (Canary Islands). J. Environ. Radioact. 116, 180–186.

Lust, M., Realo, E., 2012. Determination of dose rate from Chernobyl-derived radiocaesium in estonian soil. J. Environ. Radioact. 112, 118–124.

Manolopoulou, M., Stoulos, S., Ioannidou, A., Vagena, E., Papastefanou, C., 2012. Radiation measurements and radioecological aspects of fallout from the Fukushima nuclear accident. J. Radioanal. Nucl. Chem. 292, 155–159.

Masson, O., Baeza, A., Bieringer, J., Brudecki, K., Bucci, S., Cappai, M., Carvalho, F.P., Connan, O., Cosma, C., Dalheimer, A., Didier, D., Depuydt, G., De Geer, L.E., De Vismes, A., Gini, L., Groppi, F., Gudnason, K., Gurriaran, R., Hainz, D., Halldórsson, Ó., Hammond, D., Hanley, O., Holeý, K., Homoki, Z., Ioannidou, A., Isajenko, K., Jankovic, M., Katzlberger, C., Kettunen, M., Kierepko, R., Kontro, R., Kwakman, P.J.M., Lecomte, M., Leon Vintro, L., Leppänen, A.P., Lind, B., Lujaniene, G., Mc Ginnity, P., Mahon, C.M., Malá, H., Manenti, S., Manolopoulou, M., Mattila, A., Mauring, A., Mietelski, J.W., Møller, B., Nielsen, S.P., Nikolic, J., Overwater, R.M.W., Pálsson, S.E., Papastefanou, C., Penev, I., Pham, M. K., Povinec, P.P., Ramebäck, H., Reis, M.C., Ringer, W., Rodriguez, A., Rulík, P., Saey, P.R.J., Samsonov, V., Schlosser, C., Sgorbati, G., Silobritiene, B.V., Söderström, C., Sogni, R., Solier, L., Sonck, M., Steinhauser, G., Steinkopff, T., Steinmann, P., Stoulos, S., Sýkora, I., Todorovic, D., Tooloutalaie, N., Tositti, L., Tschiersch, J., Ugron, A., Vagena, E., Vargas, A., Wershofen, H., Zhukova, O., 2011. Tracking of airborne radionuclides from the damaged Fukushima Dai-Ichi nuclear reactors by European Networks. Environ. Sci. Technol. 45, 7670–7677.

Máté, B., Sobiech-Matura, K., Altzitzoglou, T., 2016. Evaluation of the 2014 EC measurement comparison on 137Cs in air filters. Appl. Radiat. Isot. 109, 36–40.

McKenzie, T., Dulai, H., 2017. Fukushima-derived radiocesium fallout in hawaiian soils. J. Environ. Radioact. 180, 106–113.

Menut, L., Masson, O., Bessagnet, B., 2009. Contribution of Saharan dust on radionuclide aerosol activity levels in Europe? The 21–22 February 2004 case study. J. Geophys. Res. Atmos. 114.

Pham, M., Povinec, P., Lee, S.-H., Oregioni, B., 2000. Atmospheric transport of particles from the north Africa to Monaco. Bull. Bureau Natl. Metrologie 123, 143–149.

Prospero, J.M., Delany, A.C., Delany, A.C., Carlson, T.N., 2021. The discovery of african dust transport to the Western Hemisphere and the Saharan air layer: a history. Bull. Am. Meteorol. Soc. 102, E1239–E1260.

Reid, J.S., Kinney, J.E., Westphal, D.L., Holben, B.N., Welton, E.J., Tsay, S.-C., Eleuterio, D.P., Campbell, J.R., Christopher, S.A., Colarco, P.R., Jonsson, H.H., Livingston, J.M., Maring, H.B., Meier, M.L., Pilewskie, P., Prospero, J.M., Reid, E.A., Remer, L.A., Russell, P.B., Savoie, D.L., Smirnov, A., Tanré, D., 2003. Analysis of measurements of Saharan dust by airborne and ground-based remote sensing methods during the Puerto Rico Dust Experiment (PRIDE). J. Geophys. Res. Atmos. 108.

Strode, S.A., Ott, L.E., Pawson, S., Bowyer, T.W., 2012. Emission and transport of cesium-137 from boreal biomass burning in the summer of 2010. J. Geophys. Res. Atmos. 117.

Team_R Core, 2000. https://www.r-project.org/ (2021/09/22).

Thakur, P., Ballard, S., Nelson, R., 2013. An overview of Fukushima radionuclides measured in the northern hemisphere. Sci. Total Environ. 458–460, 577–613.

UNSCEAR, 1982. Ionizing Radiation, Sources and Biological Effects, United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) 1982 Report. United Nations.

UNSCEAR, 2000. Sources and Effects of Ionizing Radiation, United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) 2000 Report, Volume I. United Nations.

UNSCEAR, 2008. Effects of Ionizing Radiation, United Nations Scientific Committee on the Effects of Atomic Radiation. Report, Volume I. United Nations.

Uyttenhove, J., Poffijn, A., 1990. Considerations on the use of high resolution gamma spectrometry in environmental measurements. Radiat. Prot. Dosim. 34, 207–209.

WHO, World Health Organization, 2021. https://pdf4pro.com/view/1986-2016-chernobyl-at-30-an-update-who-1280c9.html, 09-16.

Wotawa, G., De Geer, L.E., Becker, A., D'Amours, R., Jean, M., Servranckx, R., Ungar, K., 2006. Inter- and intra-continental transport of radioactive cesium released by boreal forest fires. Geophys. Res. Lett. 33.

Yablokov, A., Labunska, I., Blokov, I., Santillo, D., Johnston, P., Stringer, R., Sadownichik, T., Antipkin, Y.G., Arabskaya, L.P., Bazyka, D.A., 2006. The Chernobyl Catastrophe Consequences on Human Health, Amsterdam, the Netherlands.