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Inventory and vertical distribution of ^{137}Cs, $^{239+240}Pu$ and ^{238}Pu in soil from Raivavae and Hiva Oa, two French Polynesian islands in the southern hemisphere

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ABSTRACT

Atmospheric nuclear weapons tests carried out by the United States, the former Soviet Union, the United Kingdom, France and China between 1945 and 1980 resulted in radioactive fallout over the earth's surface of long-lived radionuclides, such as ^{137}Cs, $^{239+240}Pu$ and ^{238}Pu that could be detected more than 50 years after their production. In addition, the burnup in the upper atmosphere of a thermoelectric generator fueled by ^{238}Pu, SNAP-9A, contributed to the inventory of ^{238}Pu deposited on the ground. In order to estimate the deposition densities of ^{137}Cs, $^{239+240}Pu$ and ^{238}Pu in French Polynesia, we collected undisturbed soil samples up to 30 cm deep at eight sites in two islands (Hiva Oa, 139°W - 10°S and Raivavae, 148°W - 24°S) in 2015–2016. The top 0–10 cm of the soil cores were sliced into five 2-cm layers and the bottom 10–30 cm into four 5-cm layers for gamma spectrometry and alpha spectrometry measurements. We found that more than 50% of the radioactive inventories are still contained within the first 10 cm and that the average vertical migration velocities of ^{137}Cs and Pu are less than 0.2 cm y^{-1}. The average accumulated depositions, deduced from the profile measurements, are 236 ± 11 Bq.m^{-2} and 313 ± 39 Bq.m^{-2} for ^{137}Cs, 12.1 ± 1.5 Bq.m^{-2} and 22.1 ± 1.7 Bq.m^{-2} for $^{239+240}Pu$, and 1.23 ± 0.46 Bq.m^{-2} and 1.58 ± 0.60 Bq.m^{-2} for ^{238}Pu, in Hiva Oa and Raivavae, respectively. The $^{238}Pu/^{239+240}Pu$ ratios are 0.102 ± 0.050 at Hiva Oa and 0.072 ± 0.033 at Raivavae. Both values are higher than the ratio in nuclear weapons tests fallout estimated to be 0.016 in 2016 (Hardy et al., 1973), because of the contribution of ^{238}Pu fallout from SNAP-9A, which is latitude dependent. The $^{137}Cs/^{239+240}Pu$ ratios, 19.5 ± 3.2 at Hiva Oa and 14.2 ± 2.8 at Raivavae are in the lower part of the range of values observed in other regions of the world.

1. Introduction

The United States, the former Soviet Union, the United Kingdom, France and China conducted 502 atmospheric nuclear weapons tests (NWTs) between 1945 and 1980 for a total yield of 440 Mt (UNSCEAR, 2008). The total fission energy liberated into the atmosphere is estimated to be about 160 Mt including 16.9 Mt originating from tests conducted in the southern hemisphere and from tests conducted in the northern hemisphere at near equatorial latitudes. The atmospheric NWTs in the southern hemisphere were carried out by the United Kingdom between 1952 and 1957 (12 tests in Australia and 3 tests in the Malden Island) and by France between 1966 and 1974 (41 tests in French Polynesia), with a fission energy estimated to be 7 Mt injected in the atmosphere.

The majority of the radioactive debris was dispersed into the stratosphere and fell down gradually on the earth's surface within a few years; this part is called “global fallout”. Ground depositions from global fallout was highly latitude-dependent with a maximum in the 40–50° bands, and lower values by a factor of 2 or more, in higher and lower latitude bands. Based on an empirical compartment model of atmospheric dispersion (Bennett, 1978) UNSCEAR evaluated that about 25% of the fine particles injected in the stratosphere by atmospheric NWTs in one hemisphere was transferred to the other. Thus the global fallout (stratospheric) was distributed for 77% in the northern hemisphere and for 23% in the southern hemisphere. This distribution is in agreement with the total inventory of the $^{239+240}Pu$ estimated from measurements made in 65 sites around the world (Hardy et al., 1973). The results indicated that around 80% (9.47 ± 1.23 PBq) of the

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$^{239+240}$Pu fallout took place in the northern hemisphere and 20% (2.55 ± 0.52 PBq) in the southern hemisphere. The ^{238}Pu/$^{239+240}$Pu activity ratio was 0.237 ± 0.054 as a global average in 1970, i.e. 0.0165 in 2016 (0.0161 from 31 data of the southern hemisphere).

The activity that did not reach the stratosphere was divided into two fractions: "tropospheric" and "local/regional" fallout. The local/regional fallout, consisting mainly of large particles, was deposited during the few days following the nuclear explosion, within a few hundred kilometers from the test site, according to a pattern determined by the wind speed and direction at tropospheric altitudes. The other fraction, tropospheric fallout, consisted mainly of small particles that deposited on the ground within a few weeks following the nuclear weapon test; tropospheric fallout took place in, or close to, the latitude band of the hemisphere where the test had been conducted. The local/regional and tropospheric fallout, with a nonhomogeneous distribution mainly contained in the explosion's latitude band, includes practically all of the relatively short-lived radionuclides, such as ^{140}Ba ($T_{1/2}$ = 12.8 days), ^{131}I ($T_{1/2}$ = 8.0 days), ^{103}Ru ($T_{1/2}$ = 39.3 days) and ^{141}Ce ($T_{1/2}$ = 32.5 days), which account for most of the population's exposure (Drozdovitch et al., 2008). The tropospheric fallout in the southern hemisphere also contributes to about 10% of fallout from radionuclides of longer half-lives, such as ^{90}Sr, ^{137}Cs and ^{238,239,240,241}Pu.

The local/regional and tropospheric partitioning of fission energy in the southern hemisphere is 0.266 Mt and 3.55 Mt respectively. The timing and origin of the local/regional and tropospheric injections are indicated in Fig. 1. One part of the tropospheric injections arose from tests carried out in the southern hemisphere and another part, based on the assumption that the tropospheric debris from near equatorial latitude tests are equally partitioned between the two hemispheres (UNSCEAR, 2008), was due to tests carried out in the Christmas Islands in the northern hemisphere near the equator by the United Kingdom in 1957–1958 and by the United States in 1962. About 1.5%, 0.054 Mt (tests in Emu and Maralinga in Australia), of this tropospheric fallout is centered at 30°S of latitude. Nearly 17%, 0.049 Mt (tests in Monte Bello Islands) and 0.54 Mt (tests in Moruroa and Fangataufa Islands), is centered at latitude of 20°S and 82%, 2.36 Mt (tests in the Christmas Islands) and 0.56 Mt (tests in the Malden Island), was dispersed near the equator in the 0–10°S latitude band. Concentrations in the surface waters of the South Pacific reflect the radioactive fallout from equatorial tests of the United States and the United Kingdom with higher concentrations in the 0–10°S latitude band until 1964. From 1965, the atmospheric tests conducted in French Polynesia in 1966–1974 resulted in slightly higher concentrations in surface waters of the 10–30°S latitude band (Hamilton et al., 1998).

The second source of radioactive deposition on the ground of the southern hemisphere results from the burnup of the thermoelectric generator SNAP-9A, fueled with ^{238}Pu, which occurred on April 21st, 1964 at an altitude of about 50 km above the Indian Ocean in the southern hemisphere. The ground deposition of 600 TBq of ^{238}Pu was distributed for 20% in the northern hemisphere and for 80% in the southern hemisphere, with maximum activity in the 20–50°S latitude bands (Hardy et al., 1972, 1973; Harley, 1980). Measurements in Antarctica ice shelf have shown that ^{238}Pu from the SNAP-9A deposited from 1964 to the 1970s, corresponding to a stratospheric residence time of more than one year, at levels of an order of magnitude greater that the earlier values from NWTs fallout (Koide et al., 1979).

The inventories of long-lived radionuclides in radioactive fallout can be derived from the ^{90}Sr inventory (UNSCEAR, 2000a). The activities of the other fission products were inferred from their relative fission yield. For example, the ^{137}Cs/^{90}Sr fission yield ratio of 1.5 (UNSCEAR, 2008) led to the deposition estimates of ^{137}Cs. $^{239+240}$Pu deposition can also be inferred from ^{90}Sr deposition using environmental measurements of these radionuclides between 1965 and 1976 in mid-latitude bands of both hemispheres (Bennett, 1978). The correlation of ^{137}Cs and Pu concentrations in soils was frequently observed (Bunzl and Kracke, 1988; Mitchell et al., 1990, Hodge et al., 1996; Quang et al., 2004; Le Roux et al., 2010). The ^{137}Cs concentrations in ground level air at Tahiti (17°S, French Polynesia) have been measured with daily or weekly frequency since 1970 (Bouisset et al., 2004). These ^{137}Cs concentrations divided by 1.5 (Fig. 2) are in good agreement with the average annual ^{90}Sr concentrations in ground-level aerosols at mid-latitudes in the southern hemisphere reported by UNSCEAR (2000a). These ^{90}Sr concentrations were measured in several sampling sites, mainly situated in Chile and Peru, of the Health and Safety Laboratory's measurement network (Volchok, 1965). The annual periodic fluctuation of the ^{137}Cs concentration in surface air in Tahiti is shown to follow a seasonal variability in phase with the cosmogenic ^{7}Be concentration produced in the upper atmosphere, indicating that the stratosphere is the common reservoir of both radionuclides (Bouisset et al., 2004).

The activities of long-lived radionuclides deposited on the ground are related to their concentrations in the lower atmosphere but their values vary substantially from one location to another according to the annual rainfall (Bennett, 1978; Mitchell et al., 1990; Bunzl and Kracke, 1998, 1994; Schuller et al., 2002a; Le Roux et al., 2010). Due to their different mobility in soils (Bunzl et al., 2001; Lujaniene et al., 2002), the posterior inventory of ^{137}Cs cannot be accurately derived from the inventory of ^{90}Sr as indicated for example by the high ^{137}Cs/^{90}Sr ratios measured in soils from different regions (Baeza et al., 1994; Kim et al., 1998; Quang et al., 2004; Igarashi et al., 2011).

Whereas inventories in the northern hemisphere are reported in many publications, few studies are related to atmospheric deposition in the southern hemisphere. Inventories in ^{137}Cs in Antarctica, Patagonia, Chile or Brazil show a significant latitude-dependent effect with

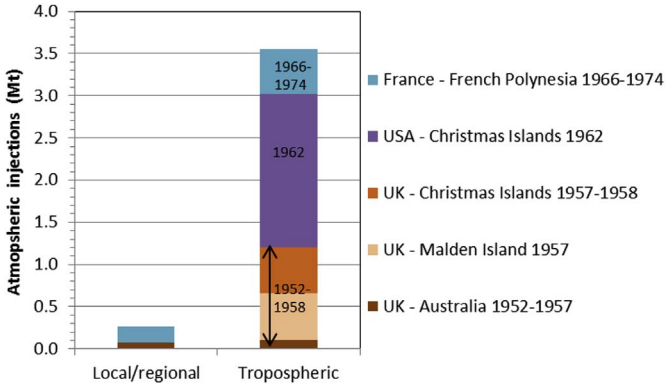

Fig. 1. Local/regional and tropospheric injections in the southern hemisphere, estimated using the assumption that tropospheric injections originating from tests in the Christmas Islands were evenly distributed between both hemispheres (UNSCEAR, 2008).

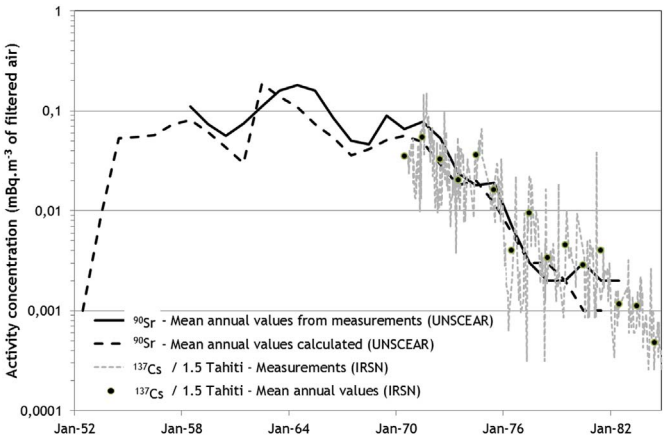

Fig. 2. Concentration of ^{137}Cs in ground-level air in Tahiti (17°S) from 1971 until 1984, normalized to ^{90}Sr using to a ^{137}Cs/^{90}Sr ratio of 1.5. The curves show the annual average concentrations of ^{90}Sr that were either measured or calculated (UNSCEAR, 2000a).