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Deposition of artificial radionuclides from atmospheric Nuclear Weapon Tests estimated by soil inventories in French areas low-impacted by Chernobyl

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ABSTRACT

Soil inventories of anthropogenic radionuclides were investigated in altitudinal transects in 2 French regions, Savoie and Montagne Noire. Rain was negligible in these 2 areas the days after the Chernobyl accident. Thus anthropogenic radionuclides are coming hypothetically only from Global Fallout following Atmospheric Nuclear Weapon Tests. This is confirmed by the isotopic signatures ($^{238}Pu/^{239+240}Pu$; $^{137}Cs/^{239+240}Pu$; and $^{241}Am/^{239+240}Pu$) close to Global Fallout value. In Savoie, a peat core age-dated by $^{210}Pb_{ex}$ confirmed that the main part of deposition of anthropogenic radionuclides occurred during the late sixties and the early seventies. In agreement with previous studies, the anthropogenic radionuclide inventories are well correlated with the annual precipitations. However, this is the first time that a study investigates such a large panel of annual precipitation and therefore of anthropogenic radionuclide deposition. It seems that at high-altitude sites, deposition of artificial radionuclides was higher possibly due to orographic precipitations.

1. Introduction

Artificial radionuclides in soils of Western Europe are mainly due to global fallout from Nuclear Weapon Tests (NWTs) in the late fifties and sixties, to the American SNAP 9A satellite explosion in 1964, and to deposition from Chernobyl accident in May 1986. To discriminate and evaluate separately these three causes of soil contamination, different strategies were considered in the past:

- measurements of soils before and after 1986 (e.g. Aoyama et al., 2006),
- measurements of radionuclide or radionuclide ratio specific of one event (e.g. Mitchell et al., 1990),
- use of rain-deposition map considering that annual precipitations have driven the deposition of artificial radionuclides due to global fallout and that precipitations at the beginning of May 1986 have driven deposition of artificial radionuclides after the Chernobyl accident (e.g. Almgren et al., 2006).

In France, no reliable maps for artificial radionuclides in soils before 1986 were available and therefore Renaud et al. (2003) used annual precipitation map and map of precipitation of 1–5 May 1986 to estimate respectively global fallout deposition and Chernobyl deposition. To estimate global fallout, Renaud et al. used the relationship established in Ireland (Mitchell et al., 1990), which is based on $^{134}Cs/^{137}Cs$ ratio considering that there was no ^{134}Cs in western Europe coming from Nuclear Weapon Test (NWT). It appeared that some French areas were theoretically unaffected by wet deposition of artificial radionuclides from Chernobyl accident.

We investigate soil inventories of artificial radionuclides in two of these areas: Savoie and Montagne Noire. In addition, these 2 areas are mountain ranges and give us information on how the deposition of artificial radionuclides was in altitude sites. Finally, the contamination of the Montagne Noire was already investigated by a non-profit organisation (CRIIRAD, 2003) and it was shown that the contamination was quite high in the soil up to 1800 Bq kg^{-1} for ^{137}Cs.

The aims of this research study were 1) to validate rain-deposition estimation of global fallout and check if there were also

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artificial radionuclides from Chernobyl accident; 2) to investigate into details global fallout in mountain areas where deposition of atmospheric pollutants (POP, heavy metals) is known to be higher due to occult and orographic deposition (Graustein and Turekian, 1989; Roe, 2005; Daly and Wania, 2005; Le Roux et al., 2008).

2. Methods

2.1. Site description

Soils were sampled between 2004 and 2006 along transects of increasing altitude and subsequently increasing annual precipitation rate. Annual precipitation rates were given at each sampling site by data from Météo-France following AURELHY method (Bénichou and Le Breton, 1986, 1987).

Based on the map established by Renaud et al. (2003, 2004, 2005) and Roussel-Debet et al. (2007), areas where no precipitation occurred in the first week of May 1986 were chosen: Savoie and Montagne Noire (Fig. 1; Table 1). These mountains have different landscape and climate features. The Montagne Noire is a small mountain massif (Pic de Nore – $z_{max} = 1210$ m) in the Southern Part of Massif Central influenced both by Atlantic and Mediterranean air masses under a Mediterranean climate. Savoie (Tarentaise and Beaufortin) is a region part of the Western Alps and is characterised by a high mountain climate bordered by le Mont Blanc Massif ($z_{max} = 4810$ m) in the North and Vanoise Massif in the South.

2.2. Soil sampling and preparation

Sampling sites were located in undisturbed areas, i.e. non-ploughed grassland or woodland, that were assumed flat enough (slope < 5%) to prevent soil migration and heavy run-off erosion. In each site, undisturbed soil samples were collected using an 8 cm diameter stainless steel corer. Three cores (where possible) were collected together at each sampling site in order to minimize heterogeneity. The depth of each core was at least 30 cm. Some cores were sub-sampled (5 or 10 cm) in order to investigate the vertical distribution of the radionuclides. Sub-sections for the same depth interval of the three cores were then bulked and each composite interval was prepared separately. Soil profiles exhibiting very low anomalous inventory were discarded from data set.

Soil samples were homogenised, dried at 80 °C and sieved to remove coarse particles (greater than 2 mm) prior to spectrometry analyses. Soil samples were discarded when the amount of coarse material exceeds 30% of total material.

In addition, a peat core was taken in a minerotrophic peatland at the little Saint-Bernard Pass with a plexiglas tube of 1 m long and 9.4 cm diameter. The core was frozen, transported to the Institute of Environmental Geochemistry, Heidelberg, Germany and was prepared according to the protocol of Givelet et al. (2004). The first 55 cm powder samples were analysed by Gamma spectrometry like soil samples.

2.3. Gamma spectrometry

Direct gamma spectrometry analyses were performed on closed volumes of 60 ml of soil using γ-spectrometers with low background level HPGe-detectors with a 0.5 mm thickness beryllium window at the "Laboratoire de Mesure de la Radioactivité dans l'Environnement" (IRSN Orsay, France) (Bouisset and Calmet, 1997). Samples were measured for 24–48 h. Detectors were located underground, under a 3 m slab of concrete, in a room shielded with 10 cm low activity lead and 5 mm electrolytic copper. Efficiency calibrations were obtained using different densities of pitchblende sources prepared in the same geometry as the samples. The calibration energy range was 46 keV to 2.7 MeV. ^{137}Cs activity (half-life: 30.2 years) was determined based on its peak at 661.7 keV. For peat samples, ^{241}Am, ^{214}Pb and ^{210}Pb were measured respectively based on their peaks at 59.4; 351.9 and 46.5 keV. ^{214}Pb was used to estimate supported ^{210}Pb (Appleby, 2001). Self-adsorption corrections were done according to sample type, density and gamma energy using in-house standards.

2.4. $^{239+240}$Pu and ^{241}Am determination

Pu isotopes and ^{241}Am were measured at the "Laboratoire de Mesure de la Radioactivité dans l'Environnement" (IRSN Orsay, France), by alpha-counting

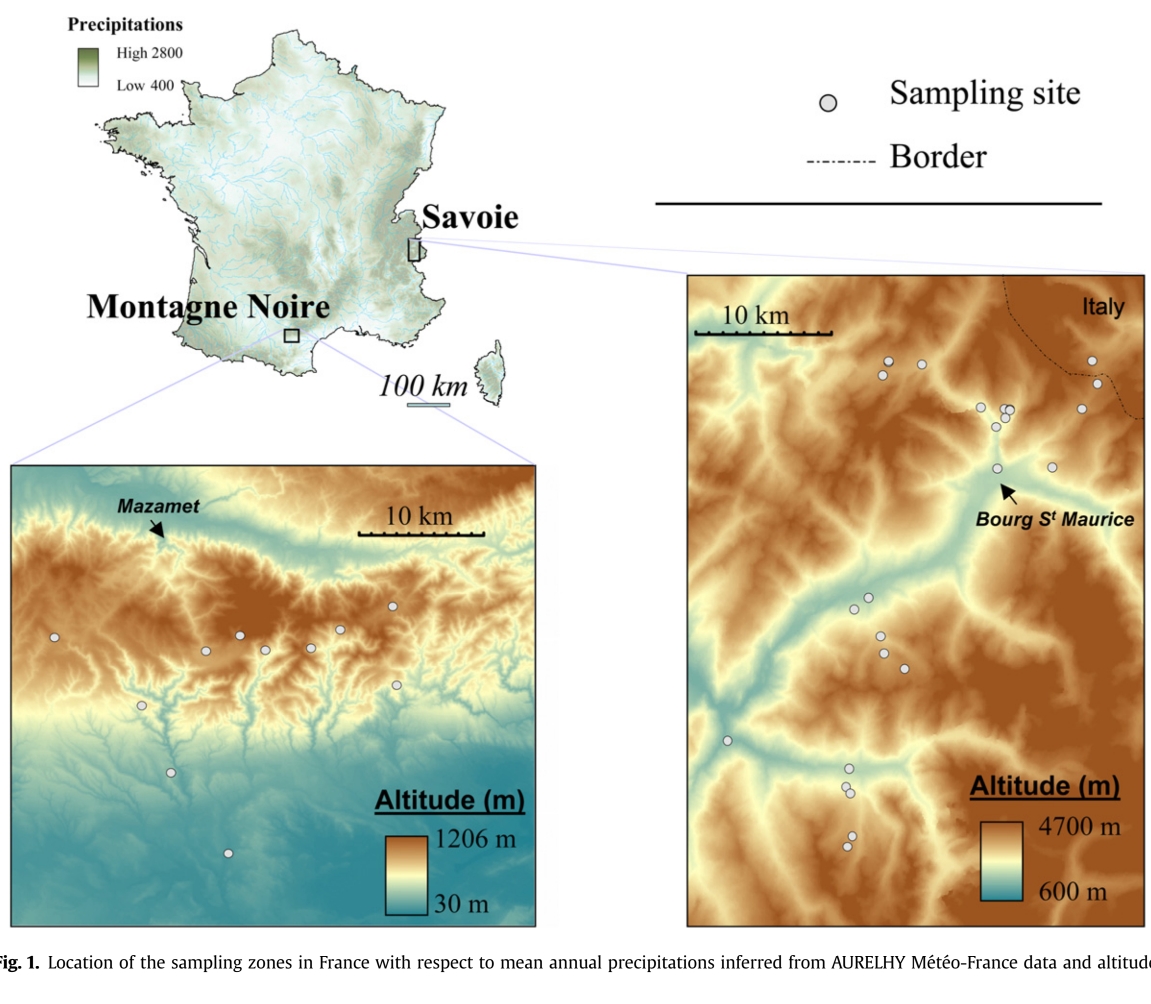

Fig. 1. Location of the sampling zones in France with respect to mean annual precipitations inferred from AURELHY Météo-France data and altitude.