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Aerosol deposition and origin in French mountains estimated with soil inventories of ^{210}Pb and artificial radionuclides

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Abstract

Radionuclide inventories were measured in soils from different French mountainous areas: Chaîne des Puys (Massif Central), Eastern Corsica, Jura, Montagne Noire, Savoie, Vosges and Rhine Valley. ^{210}Pb soil inventories were used to estimate long-term (> 75 yr) deposition of submicron aerosols. Whereas ^{210}Pb total deposition is explained partly by wet deposition, as demonstrated by increase of ^{210}Pb inventory with annual rainfall; a part of ^{210}Pb in the soils of higher altitude is caused by orographic depositions. Using measurements of radionuclides coming from nuclear aerial weapon tests (^{137}Cs and Pu isotopes), we were able to estimate the origin of aerosols deposited in high-altitude sites and to confirm the importance of occult deposition and feeder–seeder mechanism. Using a simple mass balance model, we estimate that occult deposition and feeder–seeder mechanisms account to more than 50% of total deposition of ^{210}Pb and associated submicron aerosols in French altitude sites.

Keywords: Atmospheric deposition; ^{210}Pb ; ^{137}Cs ; Pu isotopes; Mountain environment; Orographic deposition

1. Introduction

Atmospheric deposition is one of the major pathways of pollutants and nutrients. Deposition of atmospheric contaminants in mountainous areas is controlled by wet and dry depositions. Compared to neighbouring valleys, higher precipitations are occurring on hills and mountains due to several orographic effects (Roe, 2005) like the feeder–seeder effect. In addition, the so-called

“occult” depositions, which are not measurable by classic precipitation gauges (cloud water deposition, haze, canopy interception of fog water, etc.) are also higher in mountainous areas due to the high frequency of fog, haze and low-altitude clouds. The deposition is thus very variable depending on the controlling parameters including topography, dominating winds and type of vegetation.

Pollutants including N and S oxides, metals and artificial radionuclides are known to be concentrated in mountain soils (Branford et al., 1998; Huh and Su, 2004; Pourcelot et al., 2003, 2007; Weathers et al., 2000). In addition, high-elevation ecosystems are also highly sensitive to human impact because

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environmental conditions are extreme and growing seasons are short.

Direct measurement of the rate of deposition of pollutants to mountain terrain is complicated by meteorological conditions typical of high elevations. For example, it is difficult to quantify deposition by direct impact of cloud droplet on exposed surface. As quoted by Weathers et al. (2000): “measurements of all these controlling factors would be nearly impossible with a spatial resolution fine enough to reveal the important variations of deposition”. They therefore use stable Pb inventories in soils to investigate variations of deposition rates in mountain soils controlled by landscape features. The approach based on the use of naturally occurring and/or artificial radionuclides in USA and UK soils to investigate the variations with vegetation type and topography was employed in the same manner (Branford et al., 1998; Graustein and Turekian, 1989). In addition, Graustein and Turekian (1989) suggest that difference in the altitudinal origin between artificial and natural radionuclides can be used to trace sub-micron aerosol sources in the air column. Here, we

examine exhaustively the magnitude and extend of variations of atmospheric deposition in French mountainous areas using soil inventories of unsupported ^{210}Pb . In addition, we combine artificial radionuclides with ^{210}Pb to trace the origin of aerosols deposited in mountains soils.

2. Methods

2.1. Site description

Soils were sampled between 2000 and 2006 along transects of increasing altitude and subsequently increasing annual precipitation rate (Table 2 *electronic supplementary information: esi*). Annual precipitation rates were estimated at each sampling site using the model of Météo-France: AURELHY (Benichou and Le Breton, 1986).

Seven main French mountainous areas, namely; Chaîne des Puys (Massif Central), Eastern Corsica, Jura, Montagne Noire, Savoie, Vosges and Rhine Valley were investigated (Fig. 1, Table 1). These mountains have different landscape and climate features. In addition, they were more or less affected

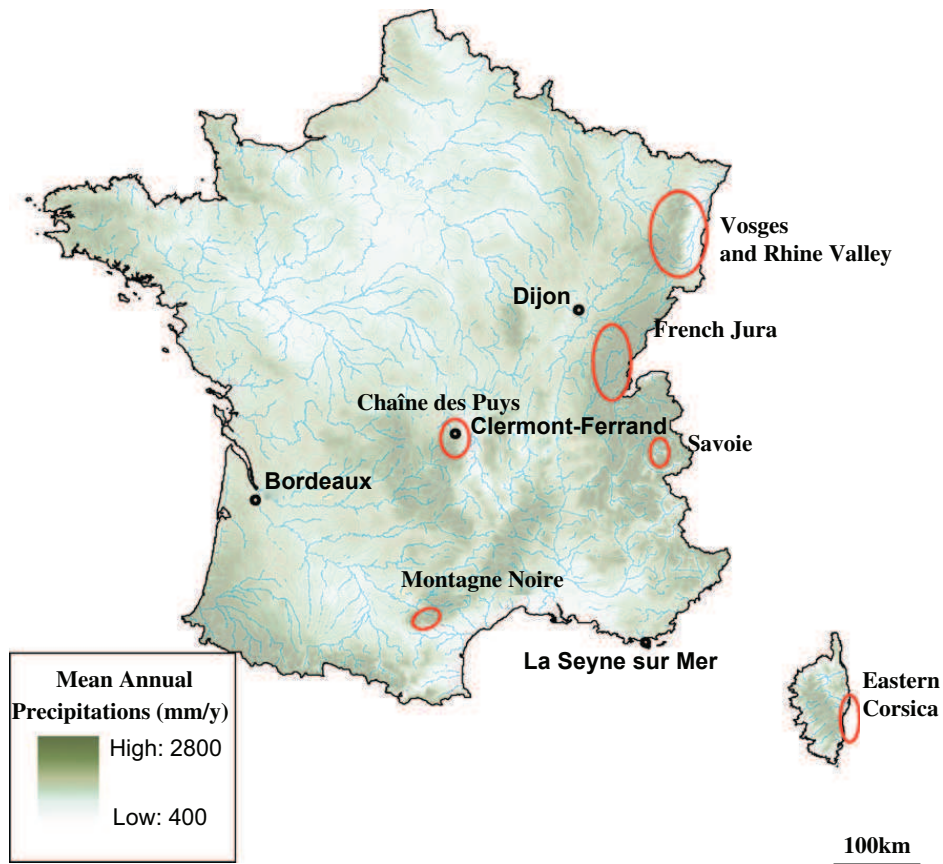


Fig. 1. Location of the sampling zones with respect to mean annual precipitations inferred from Météo-France AURELHY Model.

Table 1
Number of soil samples, range of altitude and precipitation in each studied mountainous area

Mountainous area	Number of soil samples	Sampling period	Altitude range (m)	Mean annual precipitation rate (mm yr ⁻¹) ^a	Precipitation rate 1–5 May 1986 (mm) ^b
Eastern Corsica	22	2000	5–680	690–1520	30–120
Jura	16	2005	280–1200	1250–2030	9–51
Vosges and Rhine Valley	8	2004–2006	130–1050	810–1350	10–30
Savoie	18	2006	725–2175	780–1920	0
Chaîne des Puys (Auvergne)	5	2003	680–1450	790–1900	1–6
Montagne Noire	20	2004–2005	240–1180	738–1500	0

^aAurehly model.

^bMétéo-France data (rain gauges and meteorological model).

by Chernobyl-derived artificial radionuclides. Indeed the deposition of Chernobyl-derived ¹³⁷Cs in soils depends largely on the plume trajectory and on the rainfall amount from the 1 to 5 May 1986 (Almgren et al., 2006; Renaud et al., 2003). Whereas large precipitations occurred at that time over Eastern Corsica, Jura and Vosges (up to 140 mm) and induced local deposition, very low depositions of Chernobyl-derived ¹³⁷Cs are assumed over Montagne Noire and Savoie mountains because no precipitation was recorded in May by Météo-France. In the Chaîne des Puys, low precipitations during the week following Chernobyl explosion did not exceed 6 mm.

2.2. Soil sampling and preparation

Sampling sites were located in undisturbed areas, i.e. non-ploughed grassland or forest, 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 minimise 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 bulked and prepared separately. Although not reported in this paper, results show that up to 80% of the radionuclides (²¹⁰Pb, ¹³⁷Cs and Pu) remains in the first 10 cm of the soil profiles, ascertaining no major loss by vertical migration. Soil profiles exhibiting major vertical

migration of ²¹⁰Pb or low anomalous inventory were discarded from data set.

Soil samples were homogenised, dried at 80 °C and sieved to remove coarse particles (> 2 mm) prior to spectrometry analyses. Soil samples were discarded when the amount of coarse material exceeds 20% of total material, to keep the uncertainty of soil density as low as possible.

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–2.7 MeV. ²¹⁰Pb (half-life = 22.3 yr), ²³⁴Th (24.1 d) and ²¹⁴Pb (26.9 min) activities as well as ¹³⁷Cs activity (30.2 yr) were determined based on their respective peaks at 46.5, 63.3, 351.9 and 661.7 keV. Self-adsorption corrections were done according sample type, density and gamma energy using in-house standards. Measurements were done after at least 4 weeks of equilibrium and therefore it was possible to estimate supported ²¹⁰Pb using ²¹⁴Pb activities (Appleby, 2001). Unsupported ²¹⁰Pb, which is the fraction of ²¹⁰Pb coming from

the atmosphere through the decay of atmospheric ^{222}Rn and sorbed on the surface of aerosols, was obtained by subtraction of the part of supported ^{210}Pb . From direct measurements using cascade impactors in our monitoring network OPERA, we know that ^{210}Pb is mainly bound to aerosols with a diameter between 0.7 and 1 μm .

2.4. $^{239+240}\text{Pu}$ determination

Pu isotopes were measured at the “Laboratoire de Mesure de la Radioactivité dans l’Environnement” (IRSN Orsay, France), by alpha-counting after radiochemical treatment, using a protocol of Goutelard et al. (1998). Briefly, samples are first ashed at 480 °C in order to eliminate organic matter and then spiked with ^{242}Pu , to control the recovery efficiency of the treatment. A double-step leaching using concentrated HNO_3 and H_2O_2 makes the radionuclides soluble. Stable elements and natural alpha-emitting radioisotopes are then separated from Pu by co-precipitation exchange chromatography and extraction chromatography. Pu is then electro-deposited and alpha-counted for 7 days.

3. Results and discussion

3.1. ^{210}Pb inventories in soils vs. altitudes and annual precipitations

Fig. 2a shows that ^{210}Pb soil inventories are increasing with altitude for each mountain range. The ^{210}Pb inventory at the bottom of each mountain (ca. in the valley) is nearly the same for all mountains: around 2000 Bq m^{-2} . Increase of ^{210}Pb inventories with altitude is less pronounced for the Savoie than for other mountains.

Similarly, ^{210}Pb soil inventories are increasing with annual precipitation rates based on the estimation by the model AURELHY. However, Fig. 2b shows that high-altitude soil ^{210}Pb inventories are higher than expected by a simple linear relationship with rainfall. In addition, Eastern Corsica soils have higher ^{210}Pb inventories than other soils for the same annual precipitation rates.

Our data suggest that total deposition of ^{210}Pb largely depends on wet deposition as already demonstrated by other studies (i.e. McNeary and Baskaran, 2003). Even considering a first-order linear relationship between ^{210}Pb inventories and annual precipitations and that the y-intercept is

negative (Fig. 2b), we can conclude that ^{210}Pb dry deposition on a 75 yr scale is negligible.

The atmospheric deposition rate of ^{210}Pb F ($\text{Bq m}^{-2}\text{yr}^{-1}$), can be deduced from soil ^{210}Pb inventories I (Bq m^{-2}), using

$$F = \lambda_{210\text{Pb}} \times I, \quad (1)$$

where $\lambda_{210\text{Pb}}$ the decay constant (0.0311 yr^{-1}) and under the assumptions that ^{210}Pb atmospheric rate is on average constant and that the totality of ^{210}Pb deposited is preserved in the soil column.

If we omit the highest-altitude samples, which will be discussed in the next part, we can assume a linear relationship between ^{210}Pb inventory or flux and precipitations. It is noteworthy that the slope for each sample sets of mountain is more or less identical: the relationship between ^{210}Pb soil inventory and annual precipitation rate (R in mm yr^{-1}) for all the sites except Corsica and the two highest sites of each mountain range is

$$I = (6.4 \pm 1.4) \times R - (2600 \pm 2000) \quad (r^2 = 0.6, n = 57) \quad (2)$$

or in terms of ^{210}Pb flux

$$F = 0.2 \times R - 80. \quad (3)$$

^{210}Pb activity in rainwater is indeed represented by the slope of the relationship. Using the different relationships for each mountain except Eastern Corsica, we can calculate the average ^{210}Pb activity in rainwater: $200 \pm 95\% \ 50 \text{ Bq m}^{-3}$. This is in good agreement with the 6 yr average of ^{210}Pb activities measured in rainwater from precipitation collectors of the OPERA network operated by the IRSN at Clermont–Ferrand (150 Bq m^{-3}), Dijon (200 Bq m^{-3}), Bordeaux (120 Bq m^{-3}) and la Seyne Sur Mer (400 Bq m^{-3}) (data available at <http://opera.irsn.org/opera/>).

^{210}Pb inventories and mean annual precipitation rates in United Kingdom, Inland Europe (Appleby, 2001) and in Spain (Sanchez-Cabeza et al., 2007) also exhibit correlations suggesting that the amount of deposited radionuclide is proportional to the annual intensity of rain (Fig. 3). Most of our data are consistent with those relationships. However, a pronounced higher ^{210}Pb inventories are observed for each massif for the highest rainfall amount—that is to say—for the most elevated sampling points.

More pronounced ^{210}Pb inventories for the highest altitudes can be explained by two different causes. First, estimation of the AURELHY model

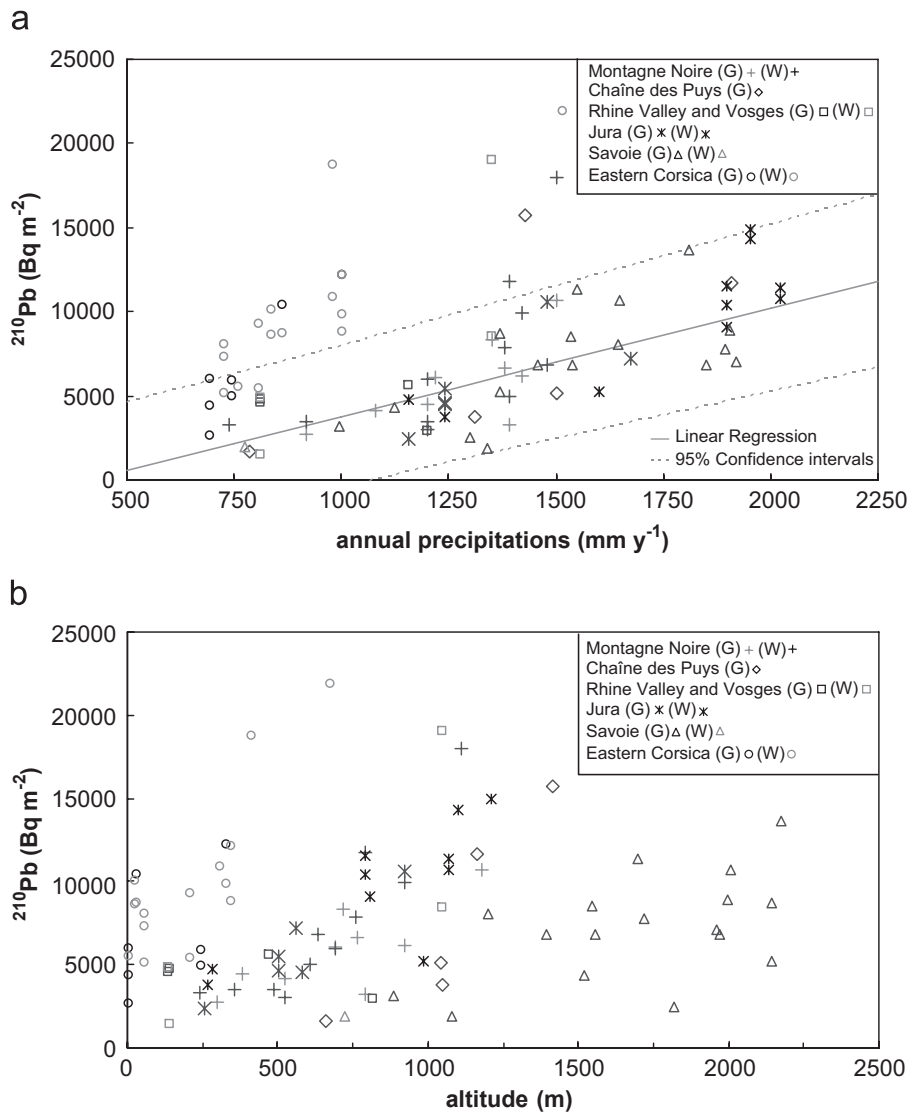


Fig. 2. (a) ^{210}Pb soil inventory vs. altitude and linear regression for soil inventories except Corsica and highest altitude sites. (b) ^{210}Pb soil inventory vs. mean annual precipitation rate.

is perhaps not satisfactory for high-altitude sites, where precipitation data are rather scarce compared to lowland areas. Thus even if AURELHY model takes into account the topography (Benichou and Le Breton, 1986), the meteorological database of Meteo-France is mainly based on measurements made at low-altitude or valley sites. Precipitations for highland sites are mainly extrapolated data. The model does not well describe orographic deposition induced for example by the feeder-seeder effect. Secondly, the model does not describe occult deposition, which is higher in areas with high topographic variations and which can significantly account for pronounced deposition of trace elements (Herckes et al., 2002).

The slope of the relationship between ^{210}Pb inventories and precipitations for Corsica is larger than for other mountains. This can be explained by a difference of ^{210}Pb activity in rain above Corsica than above continental France, as cited before. Higher activity of rain can be explained by higher ^{210}Pb activity in air (i.e. different air mass than above the continent); however ^{222}Rn levels and therefore ^{210}Pb are normally lower in oceanic regions. We suggest rather that Mediterranean rains, short but intense, have a more efficient washout ratio than in oceanic rains (Masson and Le Roux, submitted). Additionally, higher ^{210}Pb flux can also be explained by the influence of Sahara dust (Garcia-Orellana et al., 2006).

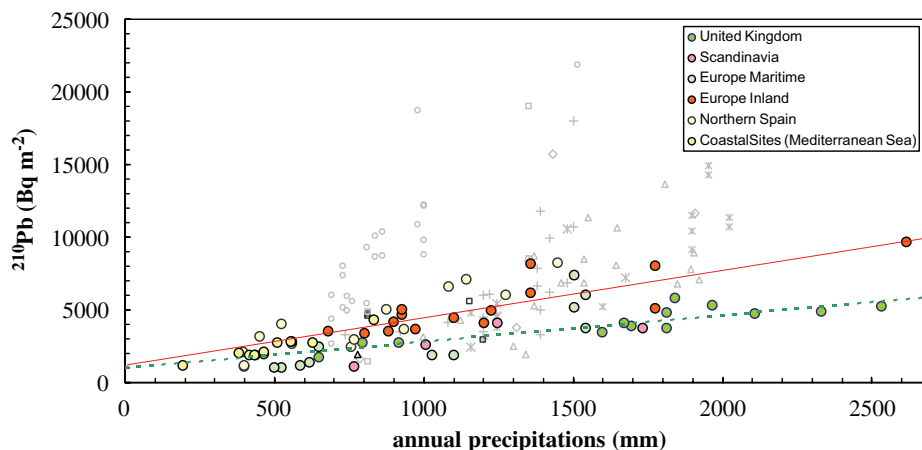


Fig. 3. ^{210}Pb inventory vs. precipitation rate observed in Europe. ^{210}Pb soil inventories in different places in Europe (in grey: this paper): the red continuous line is the correlation line for Inland European sites and the green dashed line is for UK locations (Appleby, 2001). Also plotted samples from the Mediterranean Area (Garcia-Orellana et al., 2006; Preiss et al., 1996; Sanchez-Cabeza et al., 2007) and Scandinavia (Peter Appleby, personal communication).

3.2. Patterns of ^{210}Pb inventories in forest vs. in grasslands soils: example of Montagne-Noire and Jura mountains

Data from Montagne Noire and Jura mountains allow further comparison of ^{210}Pb inventories depending on vegetation cover (Fig. 2). Brandford et al. (2004) suggested that ^{210}Pb can be used as a tracer of higher deposition rate in forested areas, especially at the forest edge. Despite that it was not our original aim; our data suggest that the ^{210}Pb soil inventories are higher in forest soils with respect to grassland soils, especially for the highest sites. For example, the highest woodland soil at the Montagne Noire ($z = 1112\text{ m}$) has a ^{210}Pb inventory of $18,000\text{ Bq m}^{-2}$ ($F = 560\text{ Bq m}^{-2}\text{ yr}^{-1}$) compared to the highest grassland point ($z = 1180\text{ m}$), which has an inventory of $10,700\text{ Bq m}^{-2}$ ($330\text{ Bq m}^{-2}\text{ yr}^{-1}$). Our data and our protocol seem to be insufficient to study fully effects of forest on atmospheric deposition.

3.3. Contributions of surface air to aerosol deposited in mountainous soils deduced from artificial radionuclide- ^{210}Pb ratios

It is now well established that ^{137}Cs from Chernobyl accident was deposited on soils mainly during raining events at the beginning of May 1986 (Almgren et al., 2006; Mitchell et al., 1990; Renaud et al., 2003). In Montagne Noire and Savoie, no precipitation occurred between 1 and 5 May 1986 and our hypothesis was that the artificial radionuclides were mainly coming from nuclear weapon

tests (NWT) global fallout. This is confirmed by the mean ratio $^{137}\text{Cs}_{\text{NWT}}/^{239-240}\text{Pu}$ of 35 (Table 3 *esi*) compared to the ratios measured by Mitchell et al. (1990), Hodge et al. (1996) and Bunzl and Kracke (1988) between 28 and 33. Excess ^{137}Cs with respect to Pu isotopes observed in other mountainous area, like for example Jura ($^{137}\text{Cs}/^{239-240}\text{Pu} \sim 44-116$), is accounted for Chernobyl radiocesium deposition.

Although inventories of ^{210}Pb and artificial radionuclides from atmospheric NWTs (R_{NWT}) roughly increase with mean annual rainfall (Table 3 *esi*); a significant increase of $^{210}\text{Pb}/^{239+240}\text{Pu}$ ratios from lowland to highland sites is observed in measured samples from Savoie, Montagne Noire and la Chaîne des Puys (Fig. 4 and Table 3 *esi*). For example, the $^{210}\text{Pb}/^{239+240}\text{Pu}$ ratio increases from 40 to ~ 95 when altitude increases from ~ 300 to 1100 m in Montagne Noire. Similarly, $^{210}\text{Pb}/^{137}\text{Cs}_{\text{NWT}}$ ratio increases for the same altitude range from 0.9–1.5 to 4.4 (Table 3 *esi*). Following Graustein and Turekian (1986, 1989), we use $^{210}\text{Pb}/R_{\text{NWT}}$ ratio to determine the proportion of ^{210}Pb deposition from the boundary layer. Indeed the $^{210}\text{Pb}/R_{\text{NWT}}$ ratio has been shown in the past—before 1981—to decrease with altitude because R_{NWT} were principally injected in the upper troposphere and stratosphere whereas ^{210}Pb follows the distribution of ^{222}Rn with highest concentrations at the Earth's surface. Assuming that there was no local source of R_{NWT} in the continental boundary layer (Monaghan, 1989) and that any increase in $^{210}\text{Pb}/R_{\text{NWT}}$ is due to the deposition of ^{210}Pb derived recently from the decay of ^{222}Rn in the continental boundary layer (Model scheme *esi*); we calculate that

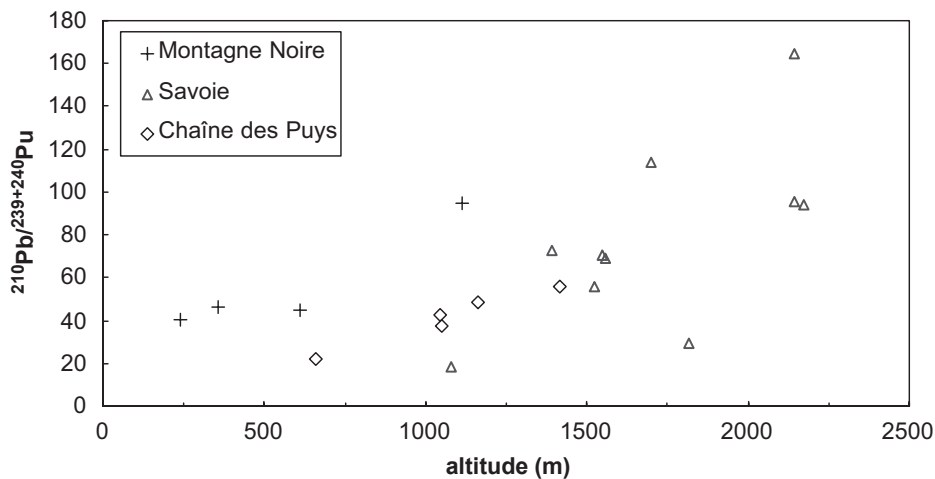


Fig. 4. $^{210}\text{Pb}/^{239+240}\text{Pu}$ ratio vs. altitude for Montagne Noire, Savoie and Chaîne des Puys.

more than 50% of ^{210}Pb in high-altitude sites in Chaîne des Puys, Savoie and Montagne Noire is derived from the boundary layer, that is to say from increased deposition due to cloud interception or seeder–feeder mechanism. For the Chaîne des Puys, a single North–South volcanoes range, the model of Graustein and Turekian seems to work well with an increase of atmospheric deposition mainly due to cloud interception. For Montagne Noire and Savoie, the relationship between $^{210}\text{Pb}/R_{\text{NWT}}$ and altitude is more scattered. In Savoie, soils were sampled in different neighbouring valleys (dashed lines in Table 3 *esi*) and effects of occult deposition are surely different depending on the direction of the slope. In Montagne Noire, samples were collected in different sub-valleys along the southern slope side influenced by Mediterranean incoming wind (called “Autan”). More samples from Savoie and from the northern slope of Montagne Noire are needed to investigate the heterogeneity of occult depositions in complex topographic areas.

Based on the three studied mountain ranges, we can deduce that excess of ^{210}Pb deposition in French mountain soils compared to European lowland sites is due to occult deposition and feeder–seeder mechanisms. Different mountain ranges have different increase in aerosol deposition: low-altitude mountain sites are less likely affected by the process of occult deposition. Branford et al. (1998) showed that the feeder–seeder mechanism in mountainous regions of Scotland decreases in effectiveness as a function of inland distance along the direction of the prevailing wind. In France, W–NW oceanic air masses are the prevailing cause of wet deposition; however it seems that distance to Atlantic Ocean

cannot explain differences in occult deposition in the studied mountains. In France, even if oceanic air masses are dominant, continental air mass and Mediterranean Saharan dust can also considerably influence the ^{210}Pb deposition.

4. Conclusions

Soil inventories of radionuclides in French mountainous areas provide indirect measurements of long-term (>75 yr) total deposition of ^{210}Pb bound to submicron aerosols. For the lowest altitude sites, ^{210}Pb rain activities were estimated to range between 150 and 250 Bq m^{-3} . Deposition of ^{210}Pb in high-altitude sites cannot only be explained by wet deposition as estimated by the meteorological model AURELHY. Soils from Corsica exhibit a specific situation with very high increase of deposition with altitude: this can be explained by Mediterranean rains short in time but very efficient in scavenging and/or Saharan dust inputs. For other French mountains, highest deposition of ^{210}Pb and carrying submicron aerosols in high-altitude sites, compared to European lowland sites with the same annual precipitation rate, is explained by occult deposition and feeder–seeder mechanism as demonstrated by $^{210}\text{Pb}/R_{\text{NWT}}$ ratios. $^{210}\text{Pb}/R_{\text{NWT}}$ ratios in soils are increasing with altitude showing that within the boundary layer, deposition of aerosols is more intense in altitude sites. A simple mass balance model using the respective vertical distributions of ^{210}Pb and R_{NWT} shows that more than 50% of ^{210}Pb deposited in altitude sites is derived from occult deposition and feeder–seeder mechanism. Our conclusion

emphasises the need of data to better understand the spatial distribution of orographic depositions over European mountainous areas.

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Appendix A. Supplementary materials

The online version of this article contains additional supplementary data. Please visit [doi:10.1016/j.atmosenv.2007.10.083](https://doi.org/10.1016/j.atmosenv.2007.10.083).

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