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^7Be , ^{210}Pb and ^{137}Cs concentrations in cloud water

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ABSTRACT

Cloud water was sampled during 8 months, in 2008 and 2009, at the puy de Dome high-altitude atmospheric research station (France). The concentrations of ^7Be and ^{210}Pb , both naturally occurring radionuclides, and ^{137}Cs of anthropogenic origin, were determined. Those values are useful for a better knowledge of the aftermath cloud deposition and more generally for wet deposition assessment of radionuclides. This is of primary interest in case of a nuclear accident, especially considering ^{137}Cs deposition, both for high-altitude locations that are regularly embedded by clouds and also for lowlands where fog can occur.

The ^7Be and ^{210}Pb average activity concentrations in cloud water found were $1.9 \pm 0.11 \text{ mBq m}^{-3}$ air and $140 \pm 10 \text{ } \mu\text{Bq m}^{-3}$ air, respectively. For ^{137}Cs , the average concentration was $0.14 \pm 0.02 \text{ } \mu\text{Bq m}^{-3}$ air. This very low-level is representative of the long term post-accidental background level. Indeed, for the studied period, the last accidental ^{137}Cs release was that of Chernobyl accident, in April 1986. To our knowledge this is the first data about ^{137}Cs reference level determination in cloud water. The comparison between cloud water and rain water concentrations showed a ratio cloud/rain ranging between 3.4 and 8.1, in agreement with previous studies performed on inorganic compounds. Scavenging efficiencies of aerosols by cloud droplets were also calculated with the additional aerosol concentrations routinely measured at the station and were quite low (0.13–0.40) compared to what has previously been observed for inorganic soluble ions.

1. Introduction

Pollutants emitted into the atmosphere or formed there by chemical reactions, as well as radionuclides of natural or anthropogenic origin, can be deposited to the ground by wet and dry removal processes; wet removal being the most effective one. The wet removal includes two mechanisms (Junge, 1963; Engelmann, 1968; Codina and Lorente, 1993). The first one, termed ‘rainout’ or ‘in-cloud scavenging’, corresponds to the mechanism that occurs

within the cloud (i.e. water vapor condensation on aerosol particles, activation of aerosol particles into cloud droplet, gas absorption). The second one refers to the ‘washout’ or ‘below-cloud scavenging’ mechanism that occurs below the cloud base during precipitation events (Seinfeld and Pandis, 2006). Experimental and modeling studies show that the rainout is believed to be the most important mechanism for the wet removal of particulate pollutants from the atmosphere (Perkins et al., 1970; Davis, 1972; Murakami et al., 1983; Schumann et al., 1988; Flossmann, 1994; Flossmann and Wobrock, 2010).

Only very few determination of ^7Be and ^{210}Pb concentrations in the cloud water are available in the literature up to now (Su and Huh, 2006; in Taiwan), while the ^{137}Cs activity level in cloud water was, to our knowledge, never registered before. At the puy de Dôme mountain (central part of France), the chemical composition of clouds has already been studied (Marinoni et al., 2004; Deguillaume et al., 2013) as well as its microbial population (Amato et al., 2005). Aerosol scavenging efficiencies were also calculated for inorganic and organic species (Sellegri et al. 2003a)

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and the contribution from the aerosol and gas phases to the droplet composition were measured (Sellegri et al., 2003b) and simulated (Leriche et al., 2007).

The transfer function between radioactive-labeled aerosols and cloud droplets is a key parameter for modeling studies of scavenging and wet deposition. Up to now, this process is generally not explicitly considered in operational dispersion models used in case of a nuclear accident. Indeed, airborne radionuclides spread as particles in the atmosphere or attached onto ambient aerosols can be activated into cloud droplets that in turn may deposit on mountainous areas or precipitate. In the framework of the OPERA-Air network (Permanent Observatory of the airborne Radioactivity) operated by the IRSN (French Nuclear Safety and Radioprotection Institute), radionuclides (^{137}Cs , ^7Be and ^{210}Pb) are quantified at trace levels on a routine basis in the aerosol phase, using very high-volume samplers and low-level detectors.

2. Material and method

2.1. Sampling site

The high-altitude (1465 m a.s.l) measurement site is the puy de Dôme (pdD) atmospheric research station. This station is located within a topographic North-South barrier in the center of France. The facing dominant westerly winds coming from the Atlantic Ocean led to a frequent cloud formation at the summit (45% of time, on average). The sampling site is located 10 km west of the city of Clermont-Ferrand. The site is generally in the planetary boundary layer (PBL) air during the day and in a mixed free tropospheric/residual air layer at night (Venzac et al., 2009). A stronger influence from PBL air is observed during summer compared to winter (Venzac et al., 2009).

At the summit ($45^\circ 46' 20'' \text{ N}$; $2^\circ 57' 57'' \text{ E}$), the main meteorological parameters (pressure, temperature and relative humidity) and aerosol particle characteristics (number and mass concentration, size distribution and chemical composition) are measured continuously on a weekly basis. The temperature varies between -15 and 10°C during winter and between 5 and 20°C during summer. A particle volume monitor (PVM100; Gerber et al., 1994) measures the liquid water content (LWC) of the cloud as well as the particle surface area and the effective droplet radius.

2.2. Sampling procedures

Measurements were performed from August to November 2008 and in January, September, November and December 2009.

The cloud sampling was performed using a Caltech Active Strand Cloudwater Collector as depicted by Demoz et al. (1996). For the present application, strands were made out of stainless steel having a diameter of 0.5 mm . The flow rate was $640 \text{ m}^3 \text{ h}^{-1}$ and the cut-off diameter of droplets was $3.5 \mu\text{m}$. Samples were collected on a weekly basis, and later gathered on a monthly basis in order to reach the activity levels necessary for the analysis.

Aerosol particles were collected with a high-volume sampler (TGD 152) having a flow rate of $700 \text{ m}^3 \text{ h}^{-1}$ specifically designed for low environmental concentrations. This sampler has a collection efficiency of 91.1% for a particle size of $5 \mu\text{m}$ (Cheng 2004). The sampler was located in the vicinity of the station, with an inlet at about 2 m a.g.l. The filters (polypropylene fibers, Jonell JPME 13 160) were collected on a weekly basis. The atmospheric concentrations sampled at the pressure at pdD (around 850 hPa) were normalized to the standard pressure conditions (1013 hPa).

Rain water was sampled on a monthly basis with a 3 m^2 rain collector. This sampling was performed at Opme, a ground based station located about 800 m lower from the summit of pdD and 10 km far in the horizontal plane (more details in Bourcier et al., 2011).

2.3. Analysis

The liquid samples were evaporated at IRSN (Cadarache, France) until dry extract. The analysis was performed by γ -spectrometry in the underground measurement laboratory LSM (Modane, France) where IRSN owns low-level, well-type high-purity Germanium detectors (Gurriaran et al., 2004).

The γ -spectrometry analysis of the aerosol filters was performed at the LMRE (Laboratory of Measurement of Radioactivity in the Environment, IRSN). For the analysis, the filters were pressed into a

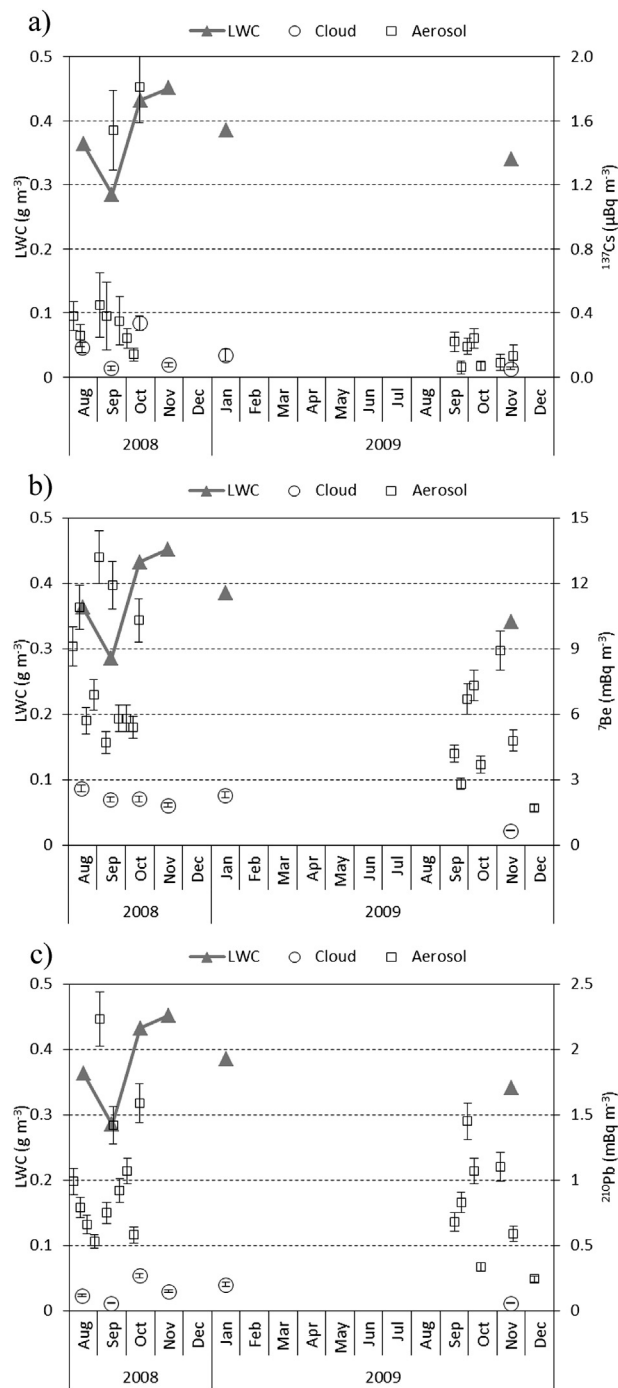


Fig. 1. Monthly cloud water concentration and weekly aerosol particle concentration on the right axis and monthly liquid water content (LWC) on the left axis for a) ^{137}Cs , b) ^7Be and c) ^{210}Pb . The error bars on the graph represent the expanded uncertainties.

tablet 7 cm in diameter and 10 mm high by means of a thermal press. Gamma counting was performed using a high-purity coaxial germanium detector (type N) for the determination of ^{210}Pb , ^7Be and ^{137}Cs using a method described by Kerlau et al. (1999). The counting time was about 2 to 4 d. Long counting times were required for the detection of ^{137}Cs traces. Data reported here were decay corrected to the end-point of the collection period. The decision thresholds and detection limits were calculated with both alpha risk and beta risk equal to 5%, the ^{137}Cs detection limit was 3.5 mBq per sample, corresponding to $0.05 \mu\text{Bq m}^{-3}$; only samples with higher concentrations were considered in the analysis. The result can be found in annex and was expressed as $A \pm UA$ where UA was the expanded uncertainty calculated by $UA = k \cdot uA$ with $k = 2$ and where uA was the absolute uncertainty.

3. Results and discussion

3.1. ^{137}Cs

Cesium-137 is a rather long-lived radionuclide ($T_{1/2} = 30.2$ y.) of anthropogenic origin. It is still persistent in the atmosphere through re-emission during biomass burning events (Bourcier et al., 2010) or resuspension of soil particles, that have received former deposition of radionuclides from past nuclear bomb test fallout and Chernobyl fallout (Masson et al., 2010). The mean ^{137}Cs concentration in cloud water was $320 \pm 60 \mu\text{Bq L}^{-1}$ (corresponding to $0.14 \pm 0.02 \mu\text{Bq m}^{-3}$ air by using a monthly averaged LWC ranging from 0.28 and 0.43 g m^{-3}) and its concentration in the aerosol phase was $0.4 \pm 0.1 \mu\text{Bq m}^{-3}$ for the corresponding period (Fig. 1a). A higher variability in the aerosol concentration can be observed compared to that of the cloud water concentration. This could be due to the fact that some air masses with high activity levels reached the pdD while there was no cloud (case of September 17th 2008 and October 15th 2008 when continental air masses or desert dust plume arrived, respectively). Consequently, by excluding those two outliers, the mean ^{137}Cs concentration in the aerosol phase was $0.23 \pm 0.09 \mu\text{Bq m}^{-3}$.

The comparison between cloud water and rain water showed a mean (\pm standard deviation, SD) cloud/rain ratio of 4.5 ± 2.9 (Fig. 2a). This is likely due to dilution effect while cloud droplets are growing to falling raindrops. According to Lin and Peng (1999), this dilution factor (cloud/rain water) is comprised between 2 and 23.

The scavenging efficiency of the aerosol in cloud droplet is calculated with the following equation (Junge, 1963):

$$F = \frac{[C]_{\text{cloud}} \times \text{LWC}}{[C]_{\text{aerosol}}}$$

where $[C]_{\text{cloud}}$ is the radionuclide concentration in cloud droplets ($\mu\text{Bq L}^{-1}$), $[C]_{\text{aerosol}}$ is the radionuclide concentration ($\mu\text{Bq m}^{-3}$) in aerosol particles for the corresponding period and LWC is the liquid water content (g m^{-3}). For the studied period, the mean LWC was 0.36 g m^{-3} (SD = 0.06). Considering that cloud water has a density of 1000 kg m^{-3} , F is equal to $(\mu\text{Bq kg}^{-1}) / (\text{g m}^{-3}) / (\mu\text{Bq m}^{-3})$ and is unit less.

The scavenging efficiency was calculated on a monthly basis. Its averaged value on the overall period was 0.40 (SD = 0.22) and the monthly means ranged from 0.08 to 0.57. It should be pointed out that this is a very rough estimate of what the scavenging efficiencies might be since the aerosol concentration derives from both cloudy and non-cloudy periods. The theoretical F value ranges between 0 (no aerosol in the cloud droplet) and 1 (all the aerosol particles are in the cloud droplets). Our values were lower than those previously observed for sulfate and black carbon at the pdD (Sellegri et al. 2003a). Since the ^{137}Cs is water soluble (Hernández

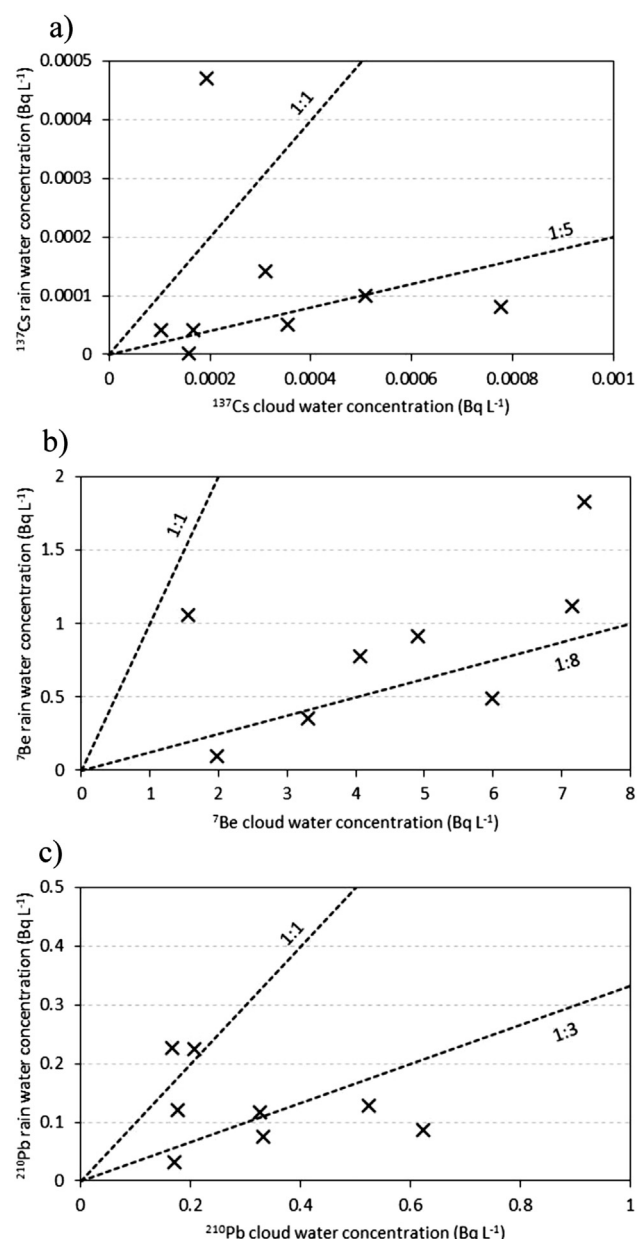


Fig. 2. Monthly rainwater concentration versus monthly cloud water concentration. The dashed lines represent the linear line with 1:1 slope and the linear line with 1:5; 1:8 and 1:3 slope for a) ^{137}Cs , b) ^7Be and c) ^{210}Pb , respectively.

et al., 2005), a higher F was expected. This gap might be attributed to differences in the experimental set-ups and especially different sampling time. Nevertheless, the mean scavenging efficiency found is similar to what was observed for sulfates at Great Dun Fell (847 m a.s.l.; UK) with values ranging between 0.41 and 0.86 (Gieray et al., 1997), or at Kleiner Feldberg (825 m a.s.l.; Germany) with a mean value of 0.53 ± 0.13 (Hallberg et al. 1994). The high variability can be attributed to the fact that the aerosol data were obtained on a monthly basis while the clouds were present only part of the sampling time.

3.2. ^7Be and ^{210}Pb

Beryllium-7 is a cosmogenic radionuclide with a relatively short half-life of 53.2 d. It is mainly (2/3) produced in the lower stratosphere and (1/3) in the upper troposphere. Its concentration in air

depends mostly on altitude but also on latitude. Due to downwind movements of stratospheric air masses in summer there is a strong seasonal dependence at ground level (Kulan et al., 2006; Steinmann et al., 2013). Finally ^7Be production rates also vary with the 11 y solar cycle (Leppänen and Paatero, 2013). Lead-210 is a product of ^{238}U series and comes from the decay of its grandparent ^{222}Rn , a gaseous compound with a short half-life of only 3.8 d that escapes from the soil. Its variation in the ground level air depends mainly on the air mass origin (i.e. continental) and on climatic conditions such as “temperature inversion” or precipitations. Both radionuclides are attached to particles and are thus very sensitive to scavenging processes.

The mean ^7Be concentration was $4.5 \pm 0.3 \text{ Bq L}^{-1}$ in the cloud water (corresponding to $1.9 \pm 0.11 \text{ mBq m}^{-3}$ air) and $6.8 \pm 0.67 \text{ mBq m}^{-3}$ in the aerosol phase (Fig. 1b). For ^{210}Pb , the mean concentration was $0.36 \pm 0.025 \text{ Bq L}^{-1}$ in the cloud water (corresponding to $140 \pm 10 \mu\text{Bq m}^{-3}$ air) and $940 \pm 90 \mu\text{Bq m}^{-3}$ in the aerosol phase (Fig. 1c). In the North of Taiwan, Su and Huh (2006) measured ^7Be cloud water concentrations up to 83 Bq L^{-1} with most of the values lower than 8.3 Bq L^{-1} . For ^{210}Pb , they observed concentrations up to 7.5 Bq L^{-1} with most of the values lower than 1.6 Bq L^{-1} . Our concentrations for ^7Be and ^{210}Pb match in those ranges. The variability of ^7Be cloud water concentration is lower than the one observed for ^{210}Pb . This is likely due to the fact that ^7Be is less influenced by the air mass origin (i.e. in the horizontal plan; Bourcier et al. 2011).

The mean cloud/rain ratio was 8.1 (SD = 5.7) for ^7Be and 3.4 (SD = 2.2) for ^{210}Pb (Fig. 2b and c); consistent with the estimate of Lin and Peng (1999).

Still being a rough estimate, the monthly averaged F value was equal to 0.24 (SD = 0.10; ranging from 0.09 to 0.31) for ^7Be and 0.13 (SD = 0.09; ranging from 0.04 and 0.24) for ^{210}Pb . These values were lower than the one observed for ^{137}Cs . This suggests that ^7Be and ^{210}Pb are less prone to be incorporated into the cloud droplets compared to ^{137}Cs . This may be attributed to higher solubility of the cesium. The solubility product constants are higher for the molecules containing Cs compared to Be and Pb.

Since these measurements and the resulting F computations have not been performed on a cloud event basis, we cannot conclude about their variability according to the cloud type. However, to our knowledge, there is no literature reporting rainout of ^7Be and ^{210}Pb . Thus, these are the first measurements about the in-cloud scavenging of these radionuclides and especially for ^{137}Cs that was determined for the first time.

4. Conclusion

During 8 months in 2008 and 2009, the activity levels of ^7Be , ^{210}Pb and ^{137}Cs have been measured in cloud droplets sampled at the top of the puy de Dôme mountain. The mean ^{137}Cs concentration was $320 \pm 57 \mu\text{Bq L}^{-1}$. To our knowledge, this is the first determination of the ^{137}Cs background level in cloud. In the case of ^7Be and ^{210}Pb , data in cloud water are very scarce. The mean concentration of ^7Be was $4.5 \pm 0.3 \text{ Bq L}^{-1}$ and the mean concentration of ^{210}Pb was $320 \pm 20 \text{ mBq L}^{-1}$. These observations allowed us to increase knowledge about the radionuclides scavenging efficiencies that were calculated for the first time for ^7Be , ^{210}Pb and ^{137}Cs . It has been observed that ^{137}Cs was more efficiently incorporated than ^7Be and ^{210}Pb . This could be attributed to a higher cesium solubility. Considering the small number of samples and the difficulty in analyzing activity levels on a cloud event basis, those values are only estimates of the in-cloud scavenging and remains to be confirmed. Nevertheless, it gives some indications about the mechanism engaged to scavenge the radionuclides in case of nuclear accident within clouds. Those values might also be useful to

study the wet deposition of the radionuclides in lowland areas with foggy conditions; this should be done carefully since cloud droplets and fog droplets present different number size distributions.

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Appendix A. Supplementary data

Supplementary data related to this article can be found in the online version at <http://dx.doi.org/10.1016/j.jenvrad.2013.10.020>.

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