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Recent trends and explanation for airborne ^{137}Cs activity level increases in France

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Abstract. Airborne ^{137}Cs level in France is not decreasing significantly anymore (mean yearly value around 0,25 μBq.m^{-3}) contrarily to what was noticed in the past decades. This observation points out the role of processes that delay the atmospheric cleaning and participate to the persistence of radionuclide in the air at ground-level after a deposit, in the frame of medium as well as long-dated post-accidental contexts. The current background level also yields to consider ^{137}Cs in the atmosphere as a tracer of atmospheric processes like resuspension and re-emission from biomass burnings. This allows us to explain 2/3 of the peaks observed over the last six years. The remaining 1/3 is mainly noticed in winter when spreading of pollutants in the atmosphere is often weak due to temperature gradient inversion. On average, continental air masses are responsible for increases by a factor of 3 while oceanic air masses are characterised by levels 3 times lower, compared to the mean value. Feeding of ^{137}Cs in air at ground-level is the result of both local resuspension that signs ^{137}Cs activity levels in soils to which is added a remote contribution from time to time (resuspension of Saharan dust or re-emission from fires occurring in eastern territories with high ^{137}Cs deposition level). Finally, ^{137}Cs activity levels in air masses crossing over France can be described on average by a longitudinal gradient.

1. INTRODUCTION

Worldwide ground-level deposition of artificial radionuclides released in the atmosphere is no longer coming from the upper layers whereas it was the case during the global fallout period (1945–1980) following nuclear bomb tests or after the Chernobyl accident. Currently, significant amount of radionuclides is still localized in terrestrial ecosystems especially in the topsoil layers and on a lesser extent, in biomass. For example, ^{137}Cs soil activity levels in France are ranging from some hundreds to several tens of thousands Bq.kg^{-1}. Thus, specific events affecting landscape like resuspension or re-emission from biomass burnings can temporarily enhance airborne levels through the raising of particles fluxes with significant specific activities. It is all the more easily to distinguish such impact that the mean ^{137}Cs activity level in the air has now reached its lowest level over the past 50 years. (around 0,25 μBq.m^{-3}). This mean level is not decreasing significantly anymore in France (excepted through the physical decay period of ^{137}Cs). This trend was also noticed in Lithuania [1]. Nevertheless, some variations with peak activities are registered regularly. Since 2000, variation of ^{137}Cs activity levels in aerosols sampled in 8 sites in France over ten-day periods can be explained by processes previously mentioned and enhanced by meteorological parameters.

2. MATERIALS AND METHODS

Aerosol sampling is performed at 1.5 m above ground-level, on a 5 days basis with high volume samplers (300 m^3 h^{-1}). This allows filtration of about 35000 m^3 every five days. Filters are made of 4 layers polypropylene fibres with a minimum collection efficiency of 95% for 30 nm particles.

^{137}Cs is measured by gamma spectrometry through his gamma emitter son ^{137} mBa. Measurements are performed on high purity Germanium low level detectors with relative efficiency higher than 50%. Those detectors are settled underground in a lead + copper chamber and under ventilation conditions and filtered air flow, to avoid radon daughters and lower detection limits. The typical counting time is 240 000 seconds

3. RESULTS

3.1 Meteorological conditions

As a meteorological parameter, we demonstrate from 14 000 daily back-trajectories (2000 to 2006 for six locations) performed with Hysplit model that origin of air masses is a key parameter. On a global time scale, extreme ^{137}Cs activity levels in France (i.e. higher than the 90th percentile or lower than the 10th percentile) depend on the air mass origin. Most of time, eastern air masses correspond to highest values while lowest values are associated with oceanic air masses (see Fig. 1). Southern origin of air masses over France is quite rare but still sufficient to explain the second reason for highest values in southern and central parts of France according to the presence of large amount of dust from Sahara [2].

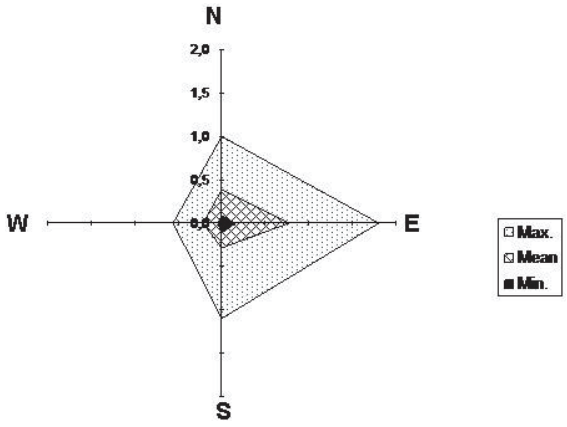

Figure 1. Airborne ^{137}Cs mean activity levels (μBq.m^{-3}) according to the air mass origin, 2000–2006 in France.

In addition we found that during their travel from coastal areas towards inlands, oceanic air masses are subject to a small enrichment of their specific activity levels. As this is not the case with the dust load (expressed through average PM_{10} levels), we concluded that the enrichment of activity level from one place to another is mainly the signature of local resuspension and depends on the soil activity level (higher on the eastern part of France due to the incidence of the Chernobyl fallout).When cruising over France from east to west, continental air masses tend to lay down their overall activity level. Moreover, the east to west gradient is more pronounced (see Fig. 2). Thus we conclude that excess of ^{137}Cs activity compared to the one belonging from local resuspension can be related to the westerly transport of particles from countries with higher ground activity levels. Concomitant increases of airborne ^{137}Cs activity levels in the different sampling locations emphasize the spatial scale of the event and consideration for long range transport.

Seasonal aspect must also be considered in variations of ^{137}Cs airborne activity level. Winter is not propitious for wind dust erosion in relation to the snow cover or soil moisture. Simultaneously, the ventilation level responsible for the dilution of the activity level per cubic meter (μBq.m^{-3}) is considered as to be poor in winter [3]. The average mixing height variation between summer and winter derived

from radio-soundings in several places in Europe leads to an average increasing factor ranging from 2 to 4 depending upon the urban/rural location or distance to the coast [4].

Inversions of the temperature profile especially in the lower layers of the atmosphere are frequent in winter and yield to reduction of the dispersion of pollutants. Even if ^{137}Cs level is quite low in French wood (<10 to 50 Bq.kg^{-1}), its widespread use in fireplaces can thus participate temporarily to the ^{137}Cs level in the lower layers of the atmosphere (see below §*Biomass burnings*). Over the period 2000–2006, we observed that the winter season (DJF) concentrates between 30 to 45% of all the peaks, according to the location.

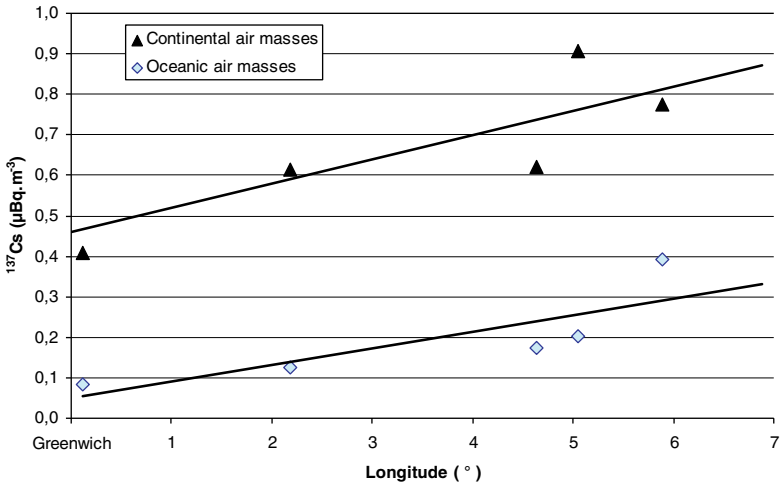

Figure 2. Longitudinal gradient of ^{137}Cs mean activity levels (μBq.m^{-3}) according to the air mass origin, 2000–2006 in France.

4. MAIN FEEDING PROCESSES INVOLVED

4.1 Biomass burnings

Wildfires represent one of the main processes able to redistribute ^{137}Cs from former deposits both at local and remote scales [5–10]. According to the fire intensity, smoke plumes from vegetation fires are able to reach atmospheric layers up to several km from ground level and to travel over distances as long as several thousand kilometres or be maintained in the atmosphere up to 20 days [11] giving thus evidence for radionuclide redistribution and trans-boundary pollution conditions. Another consequence of the spreading and redistribution of radionuclides from biomass burnings is the relative depletion of the radionuclide amounts in the emission area [5]. Among recent studies aimed at the ^{137}Cs distribution in various parts of the forest ecosystem, more than 70% and up to 99% of the ^{137}Cs is concentrated in the forest litter and upper mineral and/or organic layers of the soil [12, 13]. The remainder part, only few per-cents of the ^{137}Cs inventory, is stored in the living biomass [8–13]. Nevertheless it was demonstrated that wood burning for domestic or collective purposes and release of smokes through fireplaces can also explain increases of airborne ^{137}Cs levels [14].

As an example, we present here the specific case of extensive forest fires that took place in Belarus, Federation of Russia and neighbouring countries at the end of August/beginning of September 2002 and that give abnormal activity levels along its course in Lithuania, Scandinavia, Poland, Austria, Czech Republic, Germany and as far as western part of France (see Fig. 3).

In France, those smokes were responsible for an average rising of 6 times the yearly (2002) ^{137}Cs value and up to 10 times according to the location. The end of the fire period was due to rains starting at the end of September.

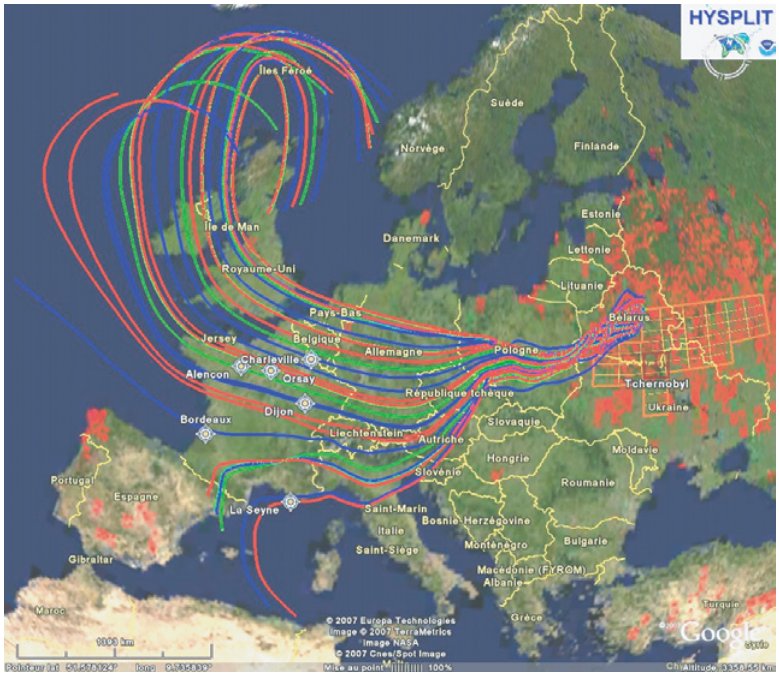

Figure 3. Western trajectories from the fire period at the beginning of September 2002.

4.2 Dust events

Acting also as a major term of resuspension, long range transport and deposition, Saharan particles can explain sporadic but intense fluxes and temporary rising of airborne ^{137}Cs. Deposition fluxes observed during a single event can therefore bring as much as radionuclide than during one year of standard deposition. This was the case for the 21 February 2004 event in France when about 2 millions tons were deposited over the two southern thirds of France. Daily airborne activity levels were 10 times higher than the mean level [2].

5. CONCLUSIONS

In the frame of post-accidental survey, we have to deal with persistence of radionuclides at ground-level air, a long time after their deposition on soil or incorporation in biomass. Mean annual ^{137}Cs activity levels in air at ground level in France remains steady at 0.25 μBq.m^{-3} since 2000. This corresponds to the current airborne ^{137}Cs background level. Thanks to high volume sampling capability and a very low level metrology, such study of the airborne variation of ^{137}Cs is now possible regarding ^{137}Cs as a tracer of atmospheric processes such as resuspension, re-emission from biomass burnings, air mass origin. It was not so evident in the past considering the prevailing atmospheric inputs of radionuclide originating from atmospheric nuclear weapon tests or accidental release that completely hide land to air transfers. On average, ^{137}Cs deposition (dry + wet) processes are counterbalanced by feeding processes

like resuspension and re-emission from biomass burnings that act as secondary and delayed sources of ^{137}Cs for the atmospheric compartment. Both processes are responsible for large amount of particles among them some are ^{137}Cs labelled. Continental air masses (including Saharan air masses) bring more ^{137}Cs labelled particles than other air mass origins. Along the trip, the amount of particles as well as ^{137}Cs content was found to diminish westerly. In France, this remote contribution is added to local resuspension that signs local soil contamination. About 1/3 of ^{137}Cs peaks observed over 10-day periods belong to dust events, according to the localization of the sampling station. Biomass burnings are responsible of about 1/3 of peaks; the rest corresponds to a mixed contribution. About one third to half of peaks are encountered in winter when meteorological conditions are not propitious to wind resuspension (due to soil moisture and/or snow cover) or spreading of atmospheric pollutants (due to the so called "temperature inversion").

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