A tool for the survey of low level radiological and natural events in the atmosphere

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Abstract:
Expertise has to deal with the evaluation of current atmospheric background levels in order to characterize all situations before they could represent the least radioecological impact for the population or the environment. IRSN strategy to face all situations corresponds to a range of four tools organised according their sensitivity *vs* reactivity ratio and allowing a continuum in the management of artifical radioactivity in the atmosphere. In the context of the lowest artificial activity levels bound to aerosols since several decades, natural events, mainly linked to or induced by meteorological conditions, can explain variations and magnitude of current activity levels in the air. This knowledge based on very low activity level determination highlights some natural processes that contribute to the persistence of radionuclides in the atmosphere during long term post accidental stage. If such processes occurred simultaneously during a release, they can also modify initial conditions of distribution in the environment. This sensitive expertise suits with the capability to check if routine nuclear plant releases are able to increase levels beyond the current background.
IRSN strategy combines several response tools based on varying sampling devices, treatment and metrology approaches according to the desired position of the cursor sensitivity / reactivity. This paper focuses on a high sensitive network operating at IRSN and able to highlight natural events and determines the reference to be taken to evaluate the impact to the atmosphere. Several references of ^{137}Cs activity levels can be chosen according to the space and/or time scales considered.

1 FROM MONITORING TO TRACE SURVEY

During the five past decades, we have inherited sampling and measurments technics to follow the overall decrease of radioactivity bounded to aerosols. This was mainly accomplished considering ^{137}Cs in the air at ground level after the nuclear weapon tests and after the Chernobyl acident. Beside the developpement of a network devoted to alert and rapid response system, specific technics were regularly upgraded to follow the average background level that now reaches about 0,2 $\mu Bq.m^{-3}$ in France.
IRSN operates four networks with different sensitivity / reactivity ratios.

- Teleray network with 180 gamma dose rate sensors is devoted to rapid detection and alert during an accident. Those sensors are localised near nuclear sites, meteorological stations, prefecture buildings and at the top of some mountains,
- SARA network (13 stations) for in situ determination of artificial radionuclides in some few hours and above limits concerning
- AS network of 70 aerosol stations with daily sampling at 10 m^3/h devoted to the monitoring of accidental impact.

- The highest sensitivity network is represented by the 9 stations (aerosol + bulk deposition) from the Permanent Observatory of Radioactivity (OPERA network), a tool based on high volume sampling of aerosol (up to 700 m^3/h) and high deposition surface (up to 5 m^2) suitable for precise but delayed caracterisation of trace activity levels over five to ten days periods. Research and survey studies concerns the transfers of radionuclides between aerosol and liquid phase and between atmosphere and soil and vice versa. Some of those studies are devoted to natural and meteorological events able to modify the background level.

Those networks are being reshaping and will be converted soon on the one hand into a wide trace survey network of about 70 stations (upgraded AS and OPERA) devoted to the characterisation of low level activities and including gazeous radionuclides (iodine, carbon-14, krypton-85, tritium) (Cf. figure 1). On the other hand, a real time monitoring network of about one hundred stations (SARA + TELERAY) with early detection and in situ gamma spectrometry designed for significant or major radiological event.

Another major improvment for the next two years concerns a mobile network split up into 2 operational bases (1 in the north, 1 in the south of France) with sampling devices (AS or OPERA) on trail that could be send in some few hours to the concerned location and that wil stay as long as the ambiant level will fall down to the level prior to the release.

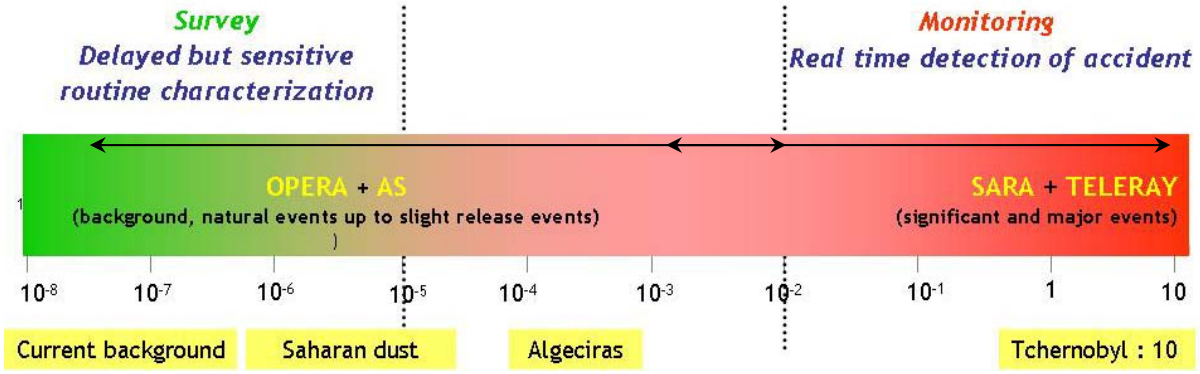


Figure 1: IRSN Networks devoted to monitoring and expertise upon radioactivity in the atmosphere

2 SURVEY AND STUDIES

Until fifteen years ago the explicit consideration of natural feeder mechanisms of the atmosphere from former artificial radionuclide deposits was impossible because their role was hiden by the prominent importance of global fallout due to nuclear weapon tests or Chernobyl accident. This impregnation gradually diseappeared and made room for those natural and timeless mechanisms to be highlighted. This also means that ^{137}Cs which had a pollutant status can now be considered as a tracer of natural events or meteorological processes.
In the frame of the OPERA network, the routine detection capablity for ^{137}Cs at about some tenth of nanoBecquerels per cubic meter is still possible thanks to high volume sampling of filtered air at about 300 to 800 m^3/h on a five days period and counting with low background facilities (HP Ge detectors, antiCompton, anticosmic devices,…). According to this low level detection capability it is possible to refine the reference to be considered for the evaluation of the whole radioecological situations whatever they will have an impact for the atmospheric compartment or not.

The sampling period of five days used in OPERA allows to a better link between changes of activity levels with meteorological conditions than weekly period. Meteorological changes are often of the same duration such as : dust events, temperature inversion events, rain events, origin of air masses. The perfect sampling frequency would be to match the closest possible the duration of those events but this is not often compatible with the obtaining of the required amount of materials for analysis purposes.

Thus, the least input of particles coming from substantially marked areas due to former deposits is sufficient to lead to a slight increase of the airborne activity level.
The main processes involved are resuspension of soil particles through wind erosion or re-emission from biomass burnings. The least resuspension of dust from soils having activity level of radionuclide like ^{137}Cs at several hundreds or more $Bq.m^{-2}$ is able to enhanced activity level in the air. Apparently this process remains insignificant but in case of accidental release it could delay the time to return to previous activity levels. Among natural phenomenon, saharan dust and wildfires are the most noticeable events responsible for such temporary rising activities up to several tenth times the prior values.
Except volcanic eruptions, those phenomena correspond to the highest flux events of particle to the atmosphere. Combined with trace levels of ^{137}Cs in those particles yields to high fluxes of radionuclide formerly deposited.

2.1 Meteorological conditions

2.1.1 Scavenging and dry deposition

Rain is the most effective phenomenon to remove particles from the atmosphere. This is visible in the figure below where lowest activity levels are obtained during rain events.

^{137}Cs ($\mu Bq/m^3$)

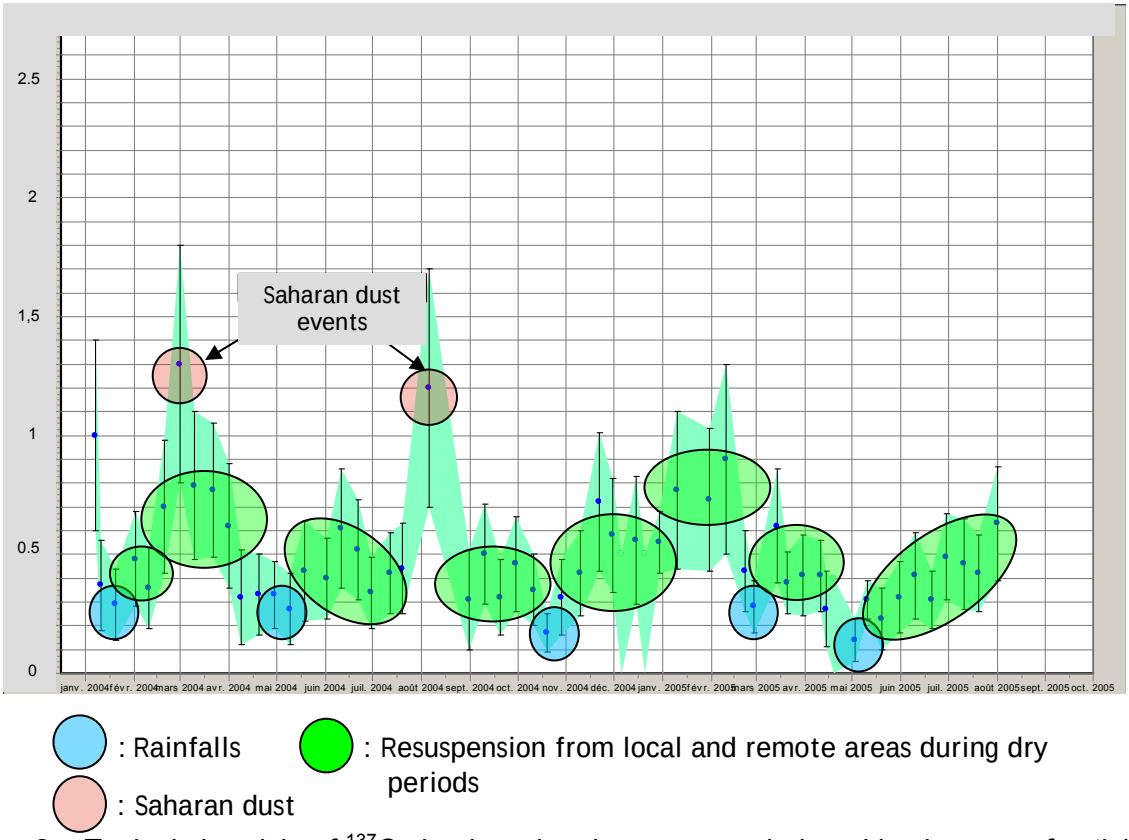


Figure 2 : Typical chronicle of ^{137}Cs in air and main processes induced in changes of activity levels

Some scavenging experiments were recently performed at the Puy de Dôme moutain by comparing ^{137}Cs activity level in cloud water (in droplets) and in rain water samples. Thanks to high volume of cloudwater sampled and to a very low background metrology, ^{137}Cs was mesured in cloud droplets at level about 5 times higher than mean value for rain water (1 mBq/l). This difference can be explained by a dilution effect during the groth of the drop when falling. Occult deposition by fog and cloud can thus explains excess of ^{137}Cs soil inventory observed in altitude of some cloudy locations [Le Roux et al. (2007)].
Since the end of 1990's, the atmosphere is no more considered as the major tank for artificial radionuclides. Thus wet deposition of radionclides only apply on some residue that are mainly supplied through resuspension. Under the present circumstances, it happens that deposition operating in wet conditions to be of the same order of size than deposition in dry conditions. This is especially the case during the dry season.

2.1.2 Temperature inversion

About one third of the peaks of activity levels is encountered in winter season. This holds for a lot of atmospheric pollutants wether gazeous or particular and is due to the inversion of the vertical temperature gradient that traps pollutants in the lower layers above the ground (see also the § 2.1.4 Seasonal aspects)

2.1.3 Origin of air masses

We show that a relevant information of the current reference level to be considered for expertise could be refined according to the origin of air masses. Using back trajectory

analysis, we show that the values lower than the 10th percentile of the activity levels (≤ 0,1 µBq.m^{-3}) encountered over a six year period correspond to oceanic air masses while continental air masses yield to values higher than the 90th percentile (≥ 1 µBq.m^{-3}). ^{137}Cs activity levels encountered on France during oceanic air mass untrusions are on average locally up to 9 times lower (mean value of 4 for the whole France) those registered for continental air masses.

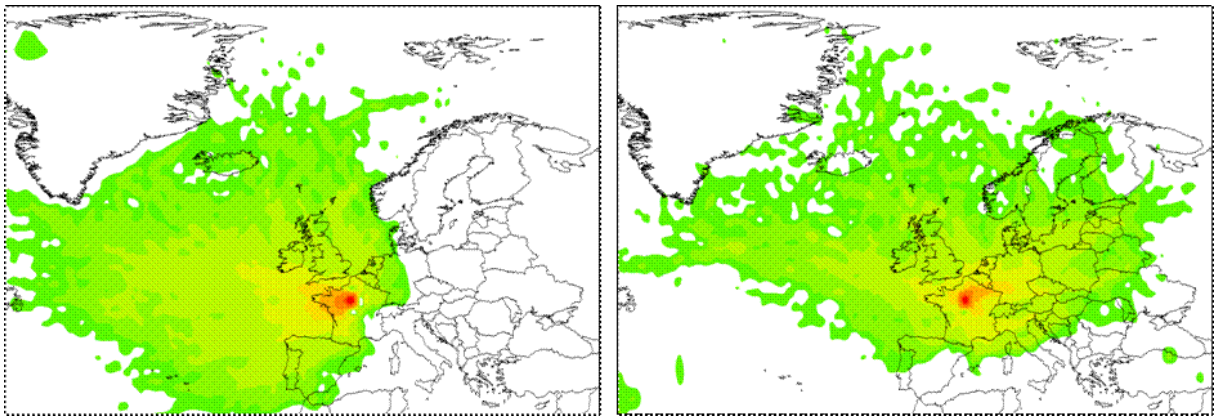


Figure 2 : Backward trajectory analysis computed with Hysplit model and corresponding to the ^{137}Cs lowest activity levels in the air

^{137}Cs activity levels in the range 0,03 to 0,2 µBq.m^{-3} obtained during oceanic air mass intrusions represent the absolute low level reference for France.

2.1.4 Seasonal aspects

Instead of considering a mean (time and space) value, it may be usefull to consider seasonal reference to characterize current situation. Seasonal approach leads to the same variability than the air mass origin (about one order of magnitude). Dry season is propicious to resuspension or re-emission from widfires and dry deposition. Winter season is not suitable for resuspension from soil due to an enhanced cohesion of particles from the moisty soils or in relation to a snow cover. Otherwise winter is often subject to temperature inversion and in some particular case to an additional small ^{137}Cs source term from the use of wood in fireplaces. As the current background level is very low, this small source term could be significant enough to be detected.

2.1.5 Role of the altitude

In case of a major accident with release to the atmosphere, it can be supposed, as it was the case for the Chernobyl releases, that a thermal and kinetic effect could spread radionuclides in altitude. Long range transport of the plume was first observed in altitude above Finland. Otherwise, recent studies conducted over 3 years at the altitude research station of the Puy de Dôme (1465 m a.s.l., France) by IRSN have demonstrated that mean activity levels of ^{137}Cs are about twice those registered at a second station located at about 800 m below. The reason is linked to long range transport of saharan dust that travel between 1 and 3 km high or elevated plumes from wildfires occuring in foreign countries, especially when fires happen in countries with high amount of ^{137}Cs in soil and vegetation. This points out the interest for such sentinel locations in the frame both of monitoring and survey networks. Futur networks operated by IRSN will include about half a dozen of such locations

2.2 Natural events

2.2.1 Biomass burnings

About one third of the peaks of ^{137}Cs in the air observed in France is due to biomass burning including local burnings mainly at the wet season or remote contribution from wildfires and long range transport of particles especially at the dry season. Those last events yield to some of the highest particle fluxes. In conjunction with some trace of artificial radioactivity in the biomass and forest litter especially when fires took place in contaminated areas, this yields to high flux of radionuclides.
We showed that emissions from wildfires in Eastern European countries at the beginning of September 2002, introduce significant amounts of ^{137}Cs in the atmospheric compartment over Europe (Cf. figures 3 and 4).

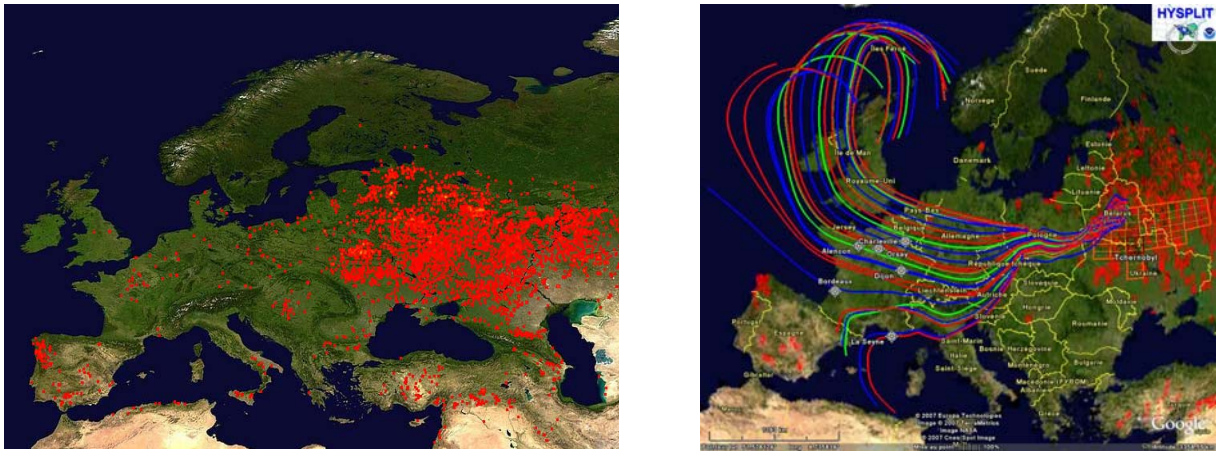


Figure 3 : Spot fires detected by Modis spectrometer onboard Terra and Aqua satellites and forward trajectories from the fire area corresponding to the contaminated area (up to 40 kBq/m^2)

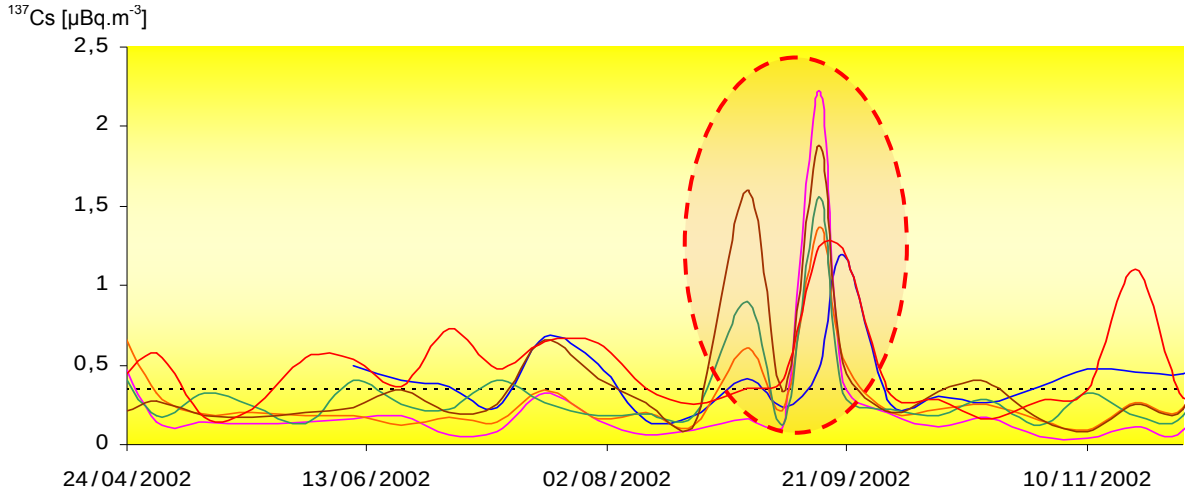


Figure 4 : Rising activity levels in France due to the transport of smoke plume from fires in eastern Europe

During the plume travel, airborne activity levels in France increase simultaneously and up to one order of magnitude compared to the 2002 mean level (Cf. figure 5). Thanks to several atmospheric sampling stations through Europe, we determine the impact of this event on

airborne activity levels at the continental scale and study long range transport of ^{137}Cs emitted (figure 1).

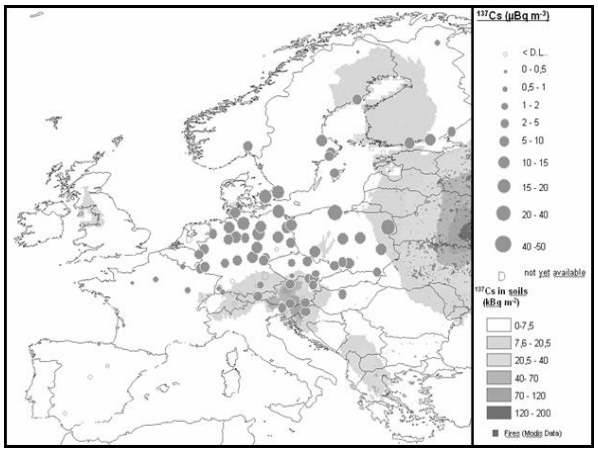


Figure 5 : Map of airborne activity levels in ^{137}Cs over Europe during and after the fire event in eastern Europe in summer 2002.

2.2.2 Saharan dust

From time to time, long range transport of particles from Sahara affects European countries causing a non negligible impact on the suspended particulate matter < 10 µm (PM_{10}) surface concentrations (Antoine et Nobileau [2006], Escudero et al. [2007], Gobbi et al. [2008], among others). Their contribution is added to the local production (Cf. § 2.2.3). The long range transport of dust coming from the Saharan region is more sporadic and intense. Both on a quality air aspect regarding PM_{10} content or radioecological aspect, those events may contribute significantly to the local PM budget (Colette et al. [2008]) and local ^{137}Cs deposition fluxes or airborne activity levels (temporarily up to 10 to 30 times the ambiant level), even far from the sources (Masson et al. [2005]). Arrival of saharan air masses correspond to radiological events in the present context of ultra low background ^{137}Cs activity level in France that can explain up to 30 % of peaks depending on the location.

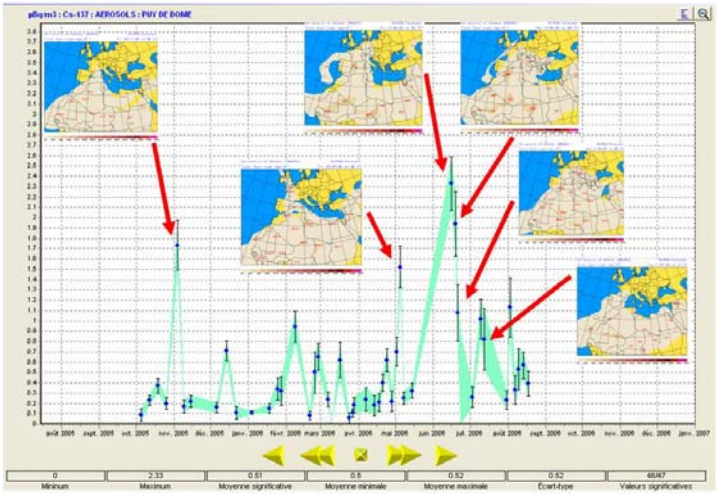
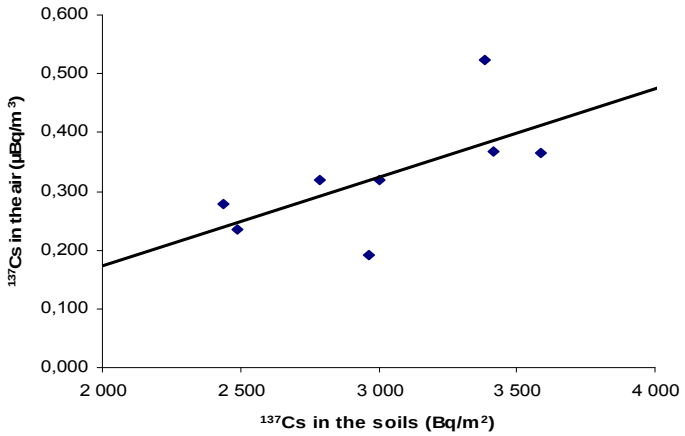


Figure 6 : ^{137}Cs rising activity levels at the top of the Puy de Dôme moutain in relation with long range transport of saharan air masses

2.2.3 Local resuspension

This local production is viewed as a 'background' contribution, regular in time and mainly depending on the season contrary to sporadic events of long range transport of wind eroded dust or re-emissions of particles by biomass burnings. A noticeable signature of this background contribution can be appreciated when comparing the relation between ^{137}Cs in soil and in aerosol (average values over the last 6 years), collected at the OPERA sampling locations.



3CONCUSION

Management of the airborne radioactivity in France is based on 4 complementary networks with different sensitivity *vs* reactivity ratios. Current activity levels of ^{137}Cs in France is performed routinely in the frame of the OPERA network and is about 0,25 µBq/m^3. Thanks to high sampling and very low level metrology capabilities, this network allows natural events to be distinguished through changes of airborne ^{137}Cs activity levels while it was not possible in the past due to the prominent contribution of nuclear weapon test fallouts. Resuspension and re-emission from biomass burnings are the main processes involved in rising activity levels. Those peaks are linked to long range transport of suspended particles originating from eastern european countries or Saharan area. Those processes generate high fluxes of particle and correspond to the extrem expression of some more common and local events. This local production is viewed as a 'background' contribution, regular in time and mainly depending on the season. Knowledge about those processes can help to understand the persistence of radionuclide in the air after their initial deposition in post-accidental survey or research studies both shortly or at long time scale.