Contamination characteristics of podzols affected by the Chernobyl Accident.

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Abstract

In the framework of ECP1 project, the soils from 6 experimental sites contaminated after the Chernobyl NPP accident have been studied in order to characterize source terms for resuspension effects in rural or agricultural areas. Except for one sand deposit located within a few km from the nuclear plant, the selected sites were podzols which had undergone important contamination during cloud transfer above districts of Ukraine, Belarus and Russia. The soils have been sampled during 5 field campaigns carried out in 1992 to 1994. Radionuclides of major importance for dose delivery, i.e. Cs-137, Sr-90, isotopes of Pu and Am-241 were measured by organisations involved. The specific activity distributions in the first 30 cm of top soil and the surface contamination densities representative of each site were determined for the radionuclides above. Complementary experiments and studies such as size specific activity distribution in soil fractions, fuel hot particles numbering and selective extraction, were carried out in order to identify contamination mechanisms and try to predict their evolution. Finally, nuclide ratios were estimated for each site and compared to those representative of fuel composition at the time of accident. Interpretations of the results obtained are given in present paper. It appears that despite the fact that weak retention properties are expected from investigated podzols, the migration of studied nuclides has been rather slow during the past 9 years, allowing 70 to 90% of initially deposited activity to remain within the first 5 cm of soil in almost all cases. Nevertheless, there are some evidence of differences in the nature of deposited radionuclides (condensed forms or fuel particles), increasing with the remoteness of studied sites from accident location. Some attempts have also been made to simulate the evolution of the distribution trends. Results from these attempts are given in present paper.

1 Introduction

The present work was performed in the framework of ECP1 project, aiming at assessing the processes of contamination by resuspension of material from rural areas strongly affected by the Chernobyl Nuclear Power Plant accident. Soil characterization is a necessary step for the determination of sources of resuspended material. A compilation of all results concerning the characterization of 6 contaminated sites and acquired during 5 experimental campaigns from 1992 to 1994, has been achieved. Systematic measurements have been carried out in order to determine surface contamination densities, activity distribution according to depth and particle size, and the distribution of fuel particles in soils. Though a rather wide spectrum of radionuclides has been measured, the present paper focuses on results from Cs-137, Sr-90, Pu-239 and Am-241 measurements, for reasons of their key importance for radiological impact and for the understanding of soil contamination mechanisms. Finally, a few attempts to predict the contamination evolution of studied sites were made. More details can be found in EUR report n° 16527 [1].

2 Material and methods

For soil sampling, top soil cutters were used on 20, 25 and 30 cm depth, 4 cm inside diameter, made from 0,3 cm thick cold-rolled steel. Generally about 20-25 cores were taken. The separation of granulometric classes from soil samples was obtained by wet sieving using polyester meshes disposed on a shaker table. For fractions of lower diameter than 20 µm, a centrifugation and sedimentation of the remaining was necessary. The grain sizes were measured by Laser Beam Diffraction (LBD) using a CILAS HR-850 granulometer. The size distribution of samples are determined by means of a computerized system for particles ranging from 0.1 to 600 µm.

Activity measurements were performed by -γ-spectrometry on hyperpure Germanium detector for Cs-137 and Am-241. Isotopes of Pu were extracted by dissolution in a mixture of acids (HNO3and HF) and separated from Am by standard methods. They were measured by α-spectrometry. Contents of Sr-90 were determined on daughter radionuclide Y-90. Measurements for Y-90 and Pu-241 were carried out with windowless gas-flow counter "TESLA".

Finally, selective extraction was performed by leaching samples with NH_4^+ cations (exchangeable fractions), with a reducing solution (dithionite for determination of oxyde bound fractions), and with oxydizing solutions (H_2O_2 + HNO_3 for determination of organic and sulphide bound fractions). The strongly fixed fractions have been determined by acid leaching of soil samples (1N and 6 N HCl or 7N HNO_3).

3 Results and Discussion

The six selected sites are located over three independent states on areas strongly affected by the Chernobyl NPP accident (3 sites in Ukraine : the "Kopachi", "Zapolye" and "Beach" sites, which can be considered as virgin soils situated within 10 km from the Chernobyl NPP, two sites in Belarus: the "Mikulichi" and "Kovali" sites located at 60 km from the NPP in the Bragin district, which were disturbed by agricultural activities, and one site in Russia : the "Novozybkov" site located at about 150 km from the NPP, for which data related to both virgin and disturbed soils were available. All sites are of similar podzolic nature, except for the Beach site which is an artificial sand deposit. According to direct measurement of grain size by LBD method, the studied podzols textures are mainly constituted of silts and fine sands for about 70% of their volume. The fine volume fraction of soil (<2µm) is about 10%. The main mineral component of former soils is quartz, whatever fine or coarse fraction is considered. Mineralogy studies carried out on the fine fraction showed, nevertheless, some content in clay minerals. For the Beach site, the frequency distribution was found to be very narrow, with more than 75% of coarse sands (>200 µm) and only 1% of particles under 50 µm.

The specific activities of Cs-137, Sr-90 and Pu-239+240 have been measured on samples taken out from the first 30 and 50 cm of soil. For virgin soils, despite a rather high variability of results coming from the same locations, due probably to local perturbations induced by bioactivity or/and heterogeneity of radionuclide initial deposition, the mean vertical distributions show similar trends from one site to another. In nearly all cases, 70 to 90% of Cs, Pu and Sr activity are still present in the first 5 cm of soil 9 years after the accident. Only Sr90 distribution in Novozybkov was found to be significantly different, with a deeper penetration of Sr in soil. Some typical distributions are given in Fig. 1.

On cultivated fields, a good homogenisation of activity was observed as a result of the implementation of standard technogenic activities such as ploughing or harrowing. The amount of total activity remaining in the first 5 cm of soil drops down to 20% in such cases, whatever radionuclide is considered.

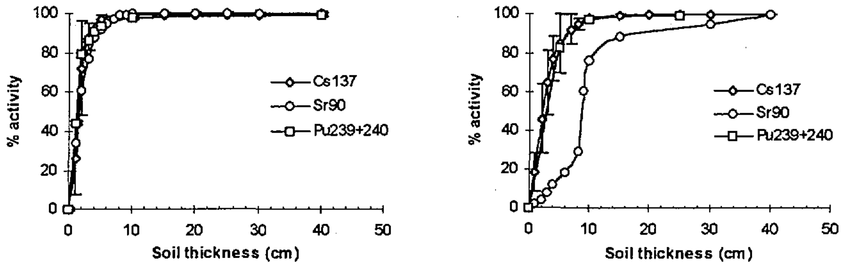

Fig.1 :Activity distributions (as cumulated percentages) in podzols from Kopachi (left) located within 10 km from NPP, and Novozybkov (right) located at 150 km from NPP

Am-241 activity build-up from Pu-241 radioactive decay has been estimated from field data and by use of results on initial fuel composition at the time of accident, taken out from various theoretical and experimental studies [2], [3]. It appears that Am-241 activity will rise until 73 years after the accident and is likely to reach at this date 43 times its initial value, which would lead to activity levels of about 9×10^4 to 4×10^5 Bq/m² (350 to 1500 Bq/kg) in soils near NPP, and 4×10^3 to 5×10^3 Bq/m² (15 to 20 Bq/kg) in soils from the Bragin district. Am-241 may therefore become of prime importance for future dose delivery.

Surface contamination densities (σa) and nuclide activity ratios were reconstructed from activity measurements and are given in table1.

Tab.1: Orders of magnitudes of surface contamination densities (σa in Bq/m²), specific activity values (As in Bq/g) in 0-1 cm layers, and radionuclide ratios related to field or fuel values. Site location is given as the average distance from NPP.

	Cs-137		Sr-90		Pu-239-240		Pu/Sr		Pu/Cs	
Site location	σa	As	σa	As	σa	As	field	fuel	field	fuel
10 km	1E+06	1E+01	1E+06	1E+01	1E+04	1	1E-02		1E-02	
60 km	5E+05	1E+01	2E+04	1E-01	5E+02	5E-03	1E-02	1E-02	1E-03	1E-02
150 km	5E+05	1E+01	2E+04	1E-02	5E+01	5E-04	5E-03		1E-05	

It appears that the levels of Cs-137 activity stay roughly the same in soils located at distances ranging from 4 to 150 km of nuclear plant, but σa for Pu-239+240 and Sr show a steep decrease of 1 to 3 orders of magnitude depending on site location. Comparative studies of Pu/Cs activity ratios measured in the various sites and proposed values of ratios representative of fuel particles [4], [5], show that Cs is probably mainly contained in fuel particles in sites located within a 10 km zone around the Chernobyl NPP. But with increasing distance, the part of Cs activity in soils issued from initial deposition of volatile forms becomes progressively dominant (90% at 60 km and around 100% at 150 km). The distribution of Sr activity observed in Novozybkov, along with the information given by Pu/Sr ratio values as well as some results on exchangeable fractions obtained by selective extraction, which showed increasing values with the distance [6], [7], would indicate a similar trend for Sr90.

Finally, Cs, Ce and Eu distributions according to soil particle sizes have been realized on samples from Zapolye site, located within the 10 km zone. For Ce and Eu, identified as good tracers of fuel particles, the size distribution showed a maximum of activity for particles of 2μm mean diameter, which is consistent with size distributions typical of fuel particle emission during the burning phase [8]. Size distribution of Cs activity realized on the same sample showed a different pattern with a sharp increase of specific activity for the lowest particle sizes, which may be due to the sorption of a small part of « condensed » Cs on clay minerals, though of weak proportion in investigated soils. Such hypothesis needs however to be confirmed.

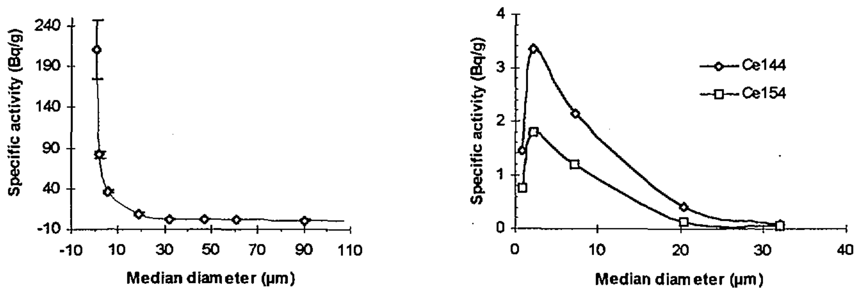

Fig.2 :Size specific activity distributions for Cs-137 (left) and Ce-144, Eu-155 (right) in Zapolye soil

Attempts were made to simulate the basic migration trends observed by means of models based on diffusive and convective transfer of radionuclide in soils or transfer in layers of soils described as discrete compartments. Simulations were performed in order to make a prognosis of the Cs-137 distribution evolution from 1995 to 2005. Results would indicate a decrease of about 20% of Cs-137 activity in the 0-5 cm layer of soil, within 10 years from present date. A decrease of about 40% is expected for Sr-90 under the same conditions. No major differences in migration speeds representative of close and remote sites can be derived from simulations. These results may be consistent with already observed evolution since the time of accident accounting for the rather large uncertainties showed by the activity distributions determined experimentally in the first 10 cm of soil. Additional verification in future would nevertheless be worth to carry out.

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