The Deposition of Chernobyl Fallout in North-east Italy*

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Since April 29th 1986 air radioactivity measurements at ground level in relation to the Chernobyl accident, were performed in our Institute by high resolution gamma-ray spectrometry. Airborne dust samples were collected on 0.45 μm filters, with controlled flow samplers, on a 4 h time basis until May 1st. Afterwards, samples were collected on a daily basis. The maximum air radioactivity was observed on May 1st when the following average values (mBq m^{-3}) were detected: $^{131}I = 15\,577$; ^{132}I (^{132}Te) = 10 550; $^{103}Ru = 4070$; $^{137}Cs = 1783$; $^{134}Cs = 929$; $^{106}Ru = 1100$; $^{99m}Tc = 969$; $^{140}Ba = 944$; $^{140}La = 548$.

In Fig. 1 the histogram of ^{131}I activity in air, clearly identifies the time of the radioactive cloud arrival. The reported values were adjusted taking into account the retention ability of the filtering system (36.5% in respect to ^{131}I).

In the days following there was a steady decrease in radioactivity until May 4th when the long-life ^{137}Cs ($t_{1/2} = 30$ y) was reduced to about 1/2, and ^{131}I ($t_{1/2} = 8$ d) to about 1/4. On May 5th a new increase in radioactivity was observed: ^{137}Cs and other relatively long-life radionuclides increased again to almost the same concentrations as the first

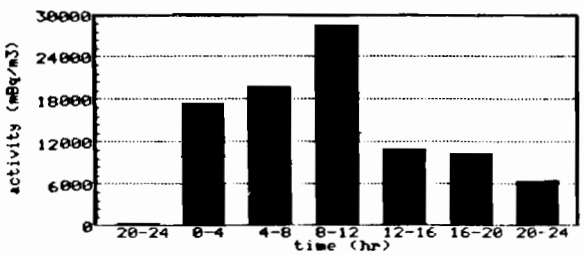

Fig. 1. ^{131}I activity from April 30th to May 1st, 1986.

day whereas the other radionuclides reached lower concentrations in good relation with their shorter half-lives. The same phenomenon has been reported by other authors [1–7]. On May 9th concentrations lower than 1/100 of the initial values were observed in air.

As far as the ground deposition of the cloud is concerned, Fig. 2 shows our first gamma spectrum of a surface soil collected in the garden of our Institute at 9.00 am on May 1st 1986; detector characteristics are also reported. A longer counting time (60 000 s) of the same sample allowed the determination of some other radioisotopes listed in the insert of Fig. 2.

The activity of surface soil samples collected daily showed that the deposition was quite remarkable on the first day corresponding to about the 75% of the total. This is not in agreement with the air radioactivity observed at ground level, from which a clear increase in ground deposition would have been expected after the return of the radioactivity on May 5th. On the other hand, daily deposits collected on wet surfaces are in agreement with ground deposits, hence indicating that the physical characteristics of the airborne radioactive dust changed with time, suggesting that less 'settleable' forms remained in suspension.

Weather conditions, and in particular rainfall, were very important in determining the final degree of soil contamination. As a consequence, very active spots were found in which radionuclide concentrations were even ten times higher than nearby areas. In order to monitor the radioactive fallout in N.E. Italy, a series of surface soil samples (Fig. 3) were collected and analysed during the first days of June. Each sampling area was accurately chosen and for each sampling point only the 0–5 cm top layer was analysed. It is important to point out some observations:

(i) All sampling sites were chosen in open areas far from trees, buildings and other obstacles.

(ii) Passing from the plain to the moutain area it became more and more difficult to find large homogeneous areas suitable for sampling.

(iii) The map reported does not include the whole northern area since deposited radionuclide concentrations were found to be strongly dependent on altitude.

The ^{137}Cs activity values were elaborated by means of a graphic contouring program (GPCP) and the distribution map is reported in Fig. 3. The ^{137}Cs distribution appeared to be related to the rainfall of the first days of May. Rainfall occurred in many places in the areas with $^{137}Cs > 5$ kBq m^{-2} but not in the large areas close to the sea characterized by a lower deposition. In addition, heavy rain in the three

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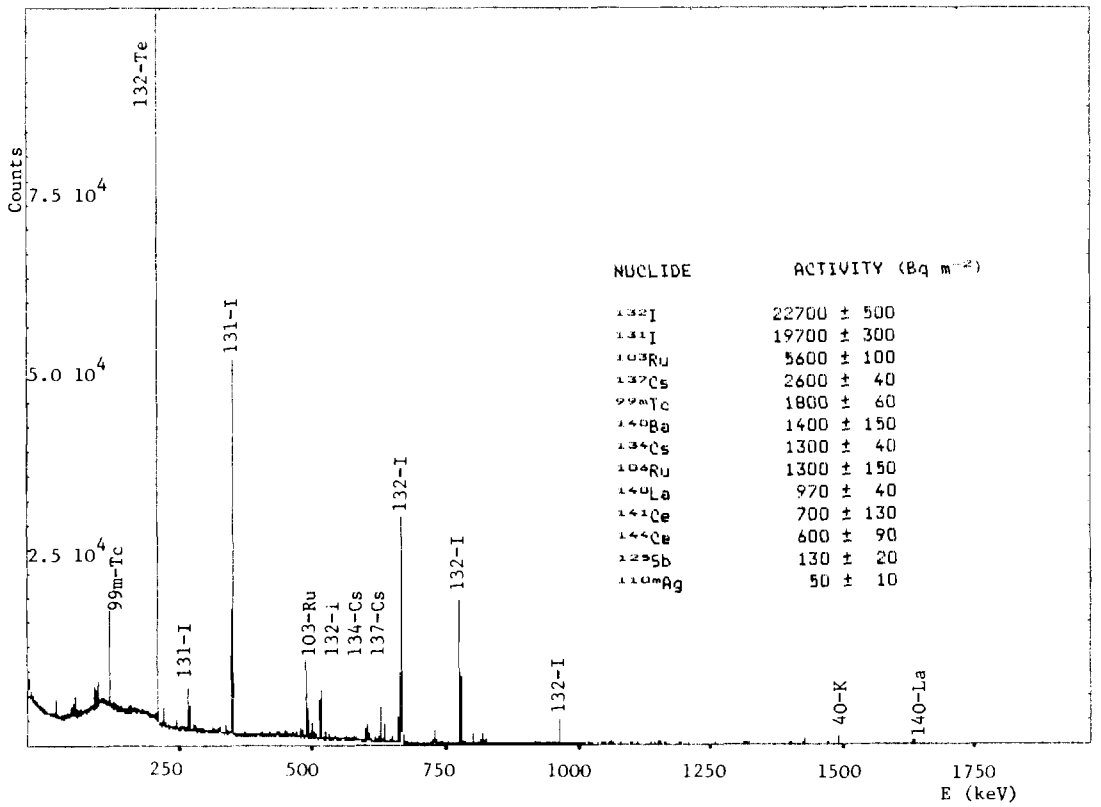

Fig. 2. Gamma-ray spectrum of a surface soil sample collected in the garden of our Institute on May 1st 1986 at 9 am. Sample dimensions: 157 cm^2 × 5 cm. Detector: Ge(Li). Couting time: 16 167 s. *FWHM*: 1.82 keV at 1.33 MeV. Efficiency: 13.2%. *P/C* = 40/1.

areas with about 60 kBq m^{-2} of ^{137}Cs were observed. As far as the activity ratios of the deposited contaminants is concerned, only minor spatial variations were observed in respect to the ratios deducible from the radionuclide list of Fig. 2.

In some areas the ratio $^{137}Cs/^{134}Cs$, which was quite constant in air filters and grass (ranging from 1.84 to 1.90 in about 50 samples from various localities) reached even higher values (3.05 and 3.99). These anomalous values are certainly due to previously deposited ^{137}Cs (from weapons tests fallout), since anomalous ^{137}Cs activities were also observed in deeper soil sections where no other radionuclides from Chernobyl had yet been found.

The average ratios of the main radioisotopes detected in soil in N.E. Italy, calculated to May 1st 1986, are as follows: $^{137}Cs/^{134}Cs = 1.87$; $^{134}Cs/^{103}Ru = 0.25$; $^{103}Ru/^{106}Ru = 4.50$; $^{141}Ce/^{144}Ce = 1.25$; $^{134}Cs/^{144}Ce = 5.50$; $^{134}Cs/^{125}Sb = 8.58$; $^{134}Cs/^{110m}Ag = 32.40$; $^{134}Cs/^{140}Ba = 0.62$.

As it can be noticed, some of the ratios reported refer to ^{134}Cs instead of ^{137}Cs as a consequence of the above considerations. These values are in agreement with analogue ratios reported in the literature [1, 3, 5, 8, 9]. It should be noted that at the time of collection and measurement of soil samples, ^{140}La had already reached radioactive equilibrium with its parent, while this was not observed on the very first days of the event. The only other occurring lanthanides, namely ^{141}Ce and ^{144}Ce, showed a slightly higher variability possibly due to the greater uncertainty in the measurement of such low level activities.

As far as the assessment of actinides transported by the cloud is concerned, the detection of uranium and plutonium was attempted in a small rain water sample, collected on May 3rd 1986, at about 20 km from Padua during the recording of the maximum air radioactivity. In spite of a relevant ^{137}Cs activity (547 Bq m^{-2}, expressed as areal deposition) $^{239+240}Pu$ shows an activity lower than the detection limit (0.078 Bq m^{-2}); therefore, the amount of sprayed $^{239+240}Pu$, if any, can be estimated to be less than 1/100 in respect to that actually present, derived from nuclear weapons tests. Total uranium measured in the same sample corresponds to less than 0.06 $\mu g\ m^{-2}$, and, since natural uranium in soil is orders of magnitude higher than that possibly deposited by the radioactive cloud in Italy, its contribution is negligible [10].

The dry deposition of Chernobyl radionuclides on grass was quite different with respect to wet deposi-

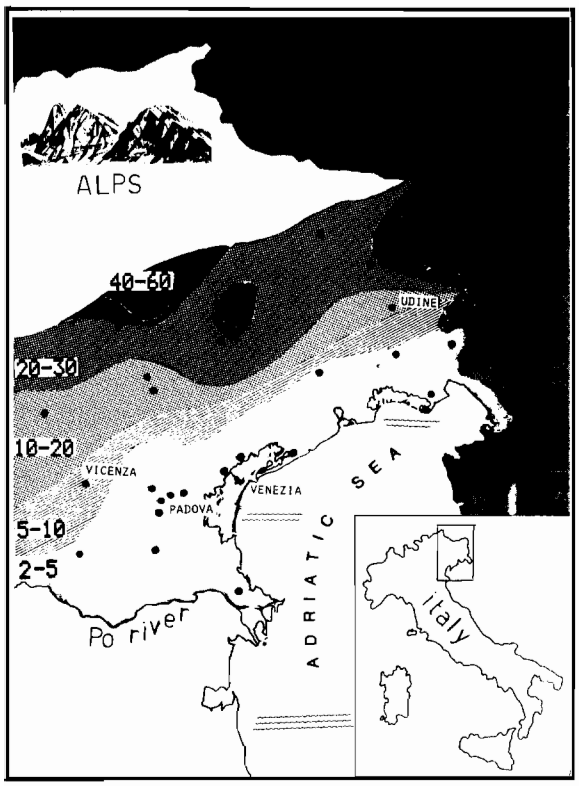

Fig. 3. Map of the area examined, sampling sites (•) and contouring distribution of ^{137}Cs (kBq m^{-2}).

tional areas. In fact out of about 4000 Bq m^{-2} of ^{137}Cs total deposit in Padua (only dry deposition), as much as 1/2 was present in grass whereas, where rainfall had occurred, only a small percentage of the total deposited ^{137}Cs was present in grass. Obviously even in dry depositional areas the radionuclides were subsequently removed from the grass as a consequence of weathering. As an example, in the grass of the garden of our Institute, the amount of ^{137}Cs initially present was reduced to about 10% at the end of June when a total of 7.5 g cm^{-2} of rain had occurred.

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