Ann. Nucl. Energy, Vol. 20, No. 6, pp. 415–420, 1993
Printed in Great Britain.

0306-4549/93 $6.00+0.00

PARTICLE-ASSOCIATED CHERNOBYL FALL-OUT IN THE LOCAL AND INTERMEDIATE ZONES

V. D. KURINY, YU. A. IVANOV, V. A. KASHPAROV, N. A. LOSHCHILOV, V. P. PROTSAK, E. B. YUDIN, M. A. ZHURBA and A. E. PARSHAKOV

Ukrainian Institute of Agricultural Radiology, p.g.t. Chabany, ul. Mashinostroitelei, d.7, 255205 Kiev, Ukraine

(Received 27 April 1992)

Abstract—Fuel and condensed particle components of the Chernobyl fall-out are differentiated and the constitution of the fuel component is displayed on a map of the area to about 60 km from the reactor. The nuclear physics related properties of the fuel "hot particles" are discussed. The possibility of a long-term increase in the bioavailability of Sr and Pu in soils due to dissolution of these hot particles is commented upon.

INTRODUCTION

As a result of the explosion at Unit 4 of the Chernobyl nuclear power plant (NPP) about 3.5% of its fuel was released into the atmosphere (U.S.S.R. State Committee, 1986). Rapid energy release in the fuel rods, the shock wave from the explosion and thermal gradients led to fragmentation of the fuel. The high temperature of the core, due to residual heat release and graphite burning, led to further fragmentation of the fuel, to the release of fuel hot particles (FHPs) from the core and to the leakage of some highly mobile, volatile radionuclides (U.S.S.R. State Committee, 1986; Devell *et al.*, 1986; Van der Veen *et al.*, 1986; Hohenemser, 1986). Fuel rod destruction and heating of the fuel led to release into the atmosphere of a considerable amount of volatile fission products—I, Te, Cs, Sr etc.—some of which condensed on inert carriers—particles of soot, dust, construction materials etc.; thus forming condensation hot particles (HPs) with properties similar to those of HPs formed during the last stage of a nuclear bomb explosion (Bykhovsky and Zaraev, 1974; Lavrenchik, 1965), e.g. they have surface contamination and low specific activity. Thus, the condensed component of fall-out, formed as a result of the Chernobyl NPP accident, has similar properties to the global fall-out of radionuclides after nuclear weapon tests. Consequently, its behaviour in soil can be predicted quite reliably. However, the behaviour of FHPs, most of which are found in the local (up to 60 km) zone, which contain the major fraction of biologically important radionuclides, such as Sr and Pu, still remains unclear.

SAMPLING PROCEDURE

In 1987, the State Committee of Hydrometeorology of the U.S.S.R. and the Scientific Centre of the Defence Ministry of the U.S.S.R. created a regular sampling network in the 60-km zone. 15 points were chosen on each of 36 rays defined at 10° intervals and originating from the site of the Chernobyl NPP. Soil samples were collected at each point, except for those located in woods, rivers and lakes. The samples were 15 cm in diameter and 5 cm deep. The samples were measured in Marinelli beakers using semiconductor γ-spectrometers. The Cs-137 deposition data obtained by this method were shown to agree with the official IAEA data (IAC, 1991). HPs were obtained from the subset of these soil samples collected from within the 10-km zone.

THE FUEL HOT PARTICLES (FHPs)

As early as 1986, the existence of FHPs had been discovered (Devell *et al.*, 1986; Van der Veen *et al.*, 1986; Hohenemser, 1986) and there has been considerable effort devoted to studying HPs from the accident (Baltensperger *et al.*, 1987; Broda, 1988; Broda *et al.*, 1989; Kulmala *et al.*, 1988; Luokkanen *et al.*, 1988; Osuch *et al.*, 1989; Raunemaa *et al.*, 1987; Saari *et al.*, 1989; Schubert and Behrend, 1987; Toivonen *et al.*, 1988; Van der Wijk *et al.*, 1988). However, there have been few efforts to analyse the radionuclide composition of FHPs from the nuclear physics point of view (Broda, 1987; Jaracz *et al.*, 1990; Piasecki *et al.*, 1990). Also, the statistics of the

data presented by Broda (1987), Jaracz *et al.* (1990) and Piasecki *et al.* (1990) are insufficient and, moreover, inventory calculations for a hypothetical light water reactor were used, so the data analysis presented is of doubtful validity.

In contrast, we have used the results of inventory calculations (Begichev *et al.*, 1990; Belyaev *et al.*, 1990) carried out at the Kurchatov Institute of Atomic Energy in Moscow, which take account of the detailed history of the Unit 4 reactor operation. Furthermore, we have studied the γ-spectrometry data on 1200 HPs with characteristic sizes of 20–400 μm and activities (in 1987) of more than 10^{-9} Ci (37 Bq), which were collected from soil at sites located at different distances (up to 10 km) and in different directions from the Chernobyl NPP in 1986–1987.

For statistical processing of the information a "Chernobyl Hot Particles" data base was created. The γ-spectrometry of these particles was performed using a high-purity Ge detector (GEM-30185) of about 30% efficiency and 1.85 keV energy resolution (for Co-60, 1332 keV γ-rays) connected to a multichannel analyser (ADCAM™ MCA 350, EG&G ORTEC system). γ-Spectrometry and neutron activation analyses indicate that 97% of these HPs are FHPs: their radionuclide composition and specific activities, determined for refractory, non-volatile elements, being close to those of Unit 4 reactor fuel. The rest are Ru HPs and Cs-condensed HPs, which are of minor importance in the local zone (up to 60 km).

We estimated the fuel burn-up for FHPs from the activity ratios of Sb-125/Ce-144, Zr-95/Ce-144 and Cs-134/Cs-137, the most probable values being 9, 8 and 11 MWtd/kg, respectively (Fig. 1). These values are approximately two-thirds of the most probable burn-up for the reactor core—14 MWtd/kg (Begichev *et al.*, 1990; Belyaev *et al.*, 1990). So we can conclude that the FHPs were released from the less irradiated (younger) part of the reactor core (about 20% of FHPs have burn-up values <0.9 MWtd/kg)—probably the epicentre of the explosion was situated in one of the fuel rods installed in the reactor not long before the accident.

It should be noted that the fission product fractionation in the fuel took place at the time of the accident as well as during normal reactor operation. Thus, on the one hand, the FHPs were depleted in highly mobile volatile fission products as a result of high-temperature annealing, and, on the other hand, these fission products had condensed and been absorbed on the surface of various carriers, including the FHPs. This leads to the variable content of CS, Sr and Pu in FHPs. The FHPs are most depleted in Cs-134 and Cs-137 (fractionation factor $k_F = 0.7$) and less depleted

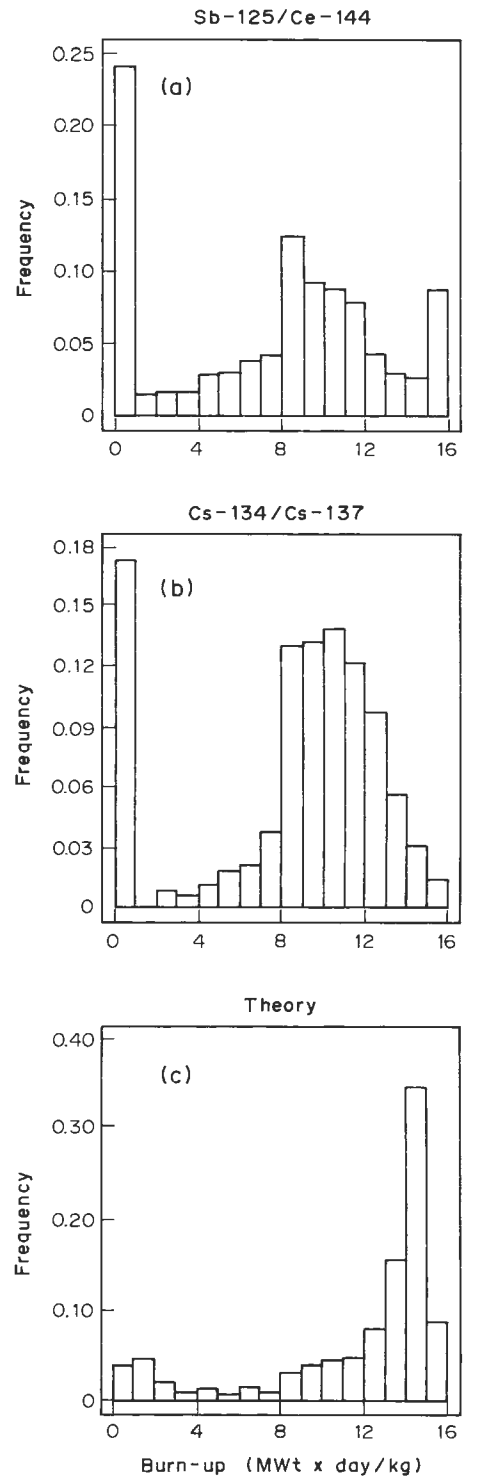

Fig. 1. Frequency histograms presenting the distribution of the burn-up determined by the activity ratios of (a) Sb-125/Ce-144 and (b) Cs-134/Cs-137 and (c) theory (Begichev *et al.*, 1990).

in Sr-90 and Ru-106, while fractionation of Zr, Sb and Eu relative to Ce-144, which is supposed to be refractory, is practically nil—these conclusions are readily derived from analyses of the histograms presented in Fig. 2: if there is a quasi-Gaussian peak in the histogram (Zr-95, Sb-125, Eu-155), then fractionation did not occur. In contrast, for Sr-90, Ru-106 and especially for Cs-137, there is a large group of FHPs depleted in the radionuclide (these FHPs are represented by a column to the very left of the histograms). Large particles (>50 μm in diameter) were found to be less depleted in radio-Cs than small ones. Also, FHPs released westwards contained more Cs-134 and Cs-137 than those released northwards, because of the differences in the temperature and time for annealing subsequent to the explosion. The sample-averaged values of activity ratios for other radionuclides relative to Ce-144 are presented in Table 1. We can see good agreement between the theoretical (Begichev *et al.*, 1990; Belyaev *et al.*, 1990) and experimental results for Sr-90, Ru-106, Sb-125, Cs-134, Cs-137 and Pu. There are discrepancies in the values for Zr-95, Eu-154 and Eu-155. Discrepancies are explained by the systematic under- and over-estimations which occurred in the theoretical calculations: Begichev *et al.* (1990) and Belyaev *et al.*(1990) seem to have used a wrong value for the neutron absorption cross-section for Eu-154, while in case of Zr-95, a more detailed account of the neutron flux dependence on time was necessary because of the rather short half-life of Zr-95.

Table 1. Theoretical and experimental relationship between the ith radionuclide's activity (A_i) and Ce-144 activity ($A_{Ce\text{-}144}$) in FHPs for 26 April 1986, N being the number of particles: $N = 1167$ for Zr-95, Ru-106, Sb-125, Cs-134, Cs-137, Eu-154, Eu-155; $N = 179$ for Sr-90

Radionuclide	Theory $A_i/A_{Ce\text{-}144}$	Experiment $\sum_{i=1}^{N} (A_i/A_{Ce})/N$
Sr-90	0.051	0.050
Zr-95	1.42	2.30
Ru-106	0.29	0.26
Sb-125	0.0078	0.0061
Cs-134	0.036	0.020
Cs-137	0.062	0.041
Eu-154	0.0030	0.0015
Eu-155	0.0043	0.0017
Pu-239+240[a]	0.00046	0.00037
Pu-239+240+238[b]	0.00069	0.001

[a] High resolution α-spectrometry ($N = 40$).
[b] α-Radiometry ($N = 1167$).
The experimental error (standard deviation, SD) for Zr-95, Ru-106, Sb-125, Cs-134, Cs-137, Eu-154, Eu-155 is <10%; for Sr-90, SD < 20%; for Pu, SD ≈ 30%.

THE FUEL AND CONDENSED PARTICULATE COMPONENTS OF THE CHERNOBYL FALL-OUT

Good tracers of the presence of FHPs in the fall-out deposited in a particular area are the refractory radionuclides—Zr-95, Nb-95, Ce-144 etc. There is a good correlation between the contents of Ce-144, Zr-95, Nb-95, Pu-238, 239, 240, Sr-90, Sb-125 and Ru-106 in FHPs (Table 1). On this basis, the contribution of the fuel component to fall-out decreases as the distance from the reactor increases. Supposing our sample of FHPs to be representative, we can estimate the contribution of the condensed component of radio-Cs in the total deposition of Cs-137 by substracting the inferred amount contained in the FHPs (see Table 1) from the total activity deposited; the values obtained have been confirmed by direct isotopic exchange methods in which a solution of stable Cs is used (Bondar *et al.*, 1991). The same approach can be used for differentiating between fuel and condensed components of Sr-90, Ru-106 etc.: there are some areas with almost complete condensed Sr contamination of the soil westwards from Novozybkovo (Russia), and near Polesskoye (Ukraine) a condensed component of Ru-106 contamination has also been observed.

Currently, there are no unequivocal data on the physical–chemical stability and migration of FHPs in realistic environmental conditions, i.e. on the dynamics and rate of radionuclide leaching as influenced by soil type, hydrological regime, composition of the FHPs etc. This is due to the great variety in both the physical–chemical properties of the FHPs (from UO_2 to UO_3), and in the environment in which they are located. The data obtained (Fig. 3) show that the contribution of the fuel component to deposition is most significant in the 30-km zone. Beyond the relocation zone northwards and westwards from the Chernobyl NPP, excluding the narrow plume directed westward, FHPs contain <25% of Cs-137 and, consequently, even total destruction of the FHPs will not lead to a significant increase in the radio-Cs contamination of agricultural products. Southwards and south-eastwards from the reactor, >50% of radio-Cs is contained in the fuel component. However, in the region beyond the relocation zone, the deposition of Cs-137, as a rule, is not in excess of 5 Ci/km^2 (185 kBq/m^2). In contrast, most of the Sr-90 and Pu is contained in FHPs and is, therefore, initially biologically unavailable. Thus, leaching of the FHPs could lead to a significant increase in the contamination of vegetation in the 30-km zone and beyond the zone southwards and westwards from the Chernobyl NPP. It is relevant to note that the HP destruction rate is highest in carbonate soils with

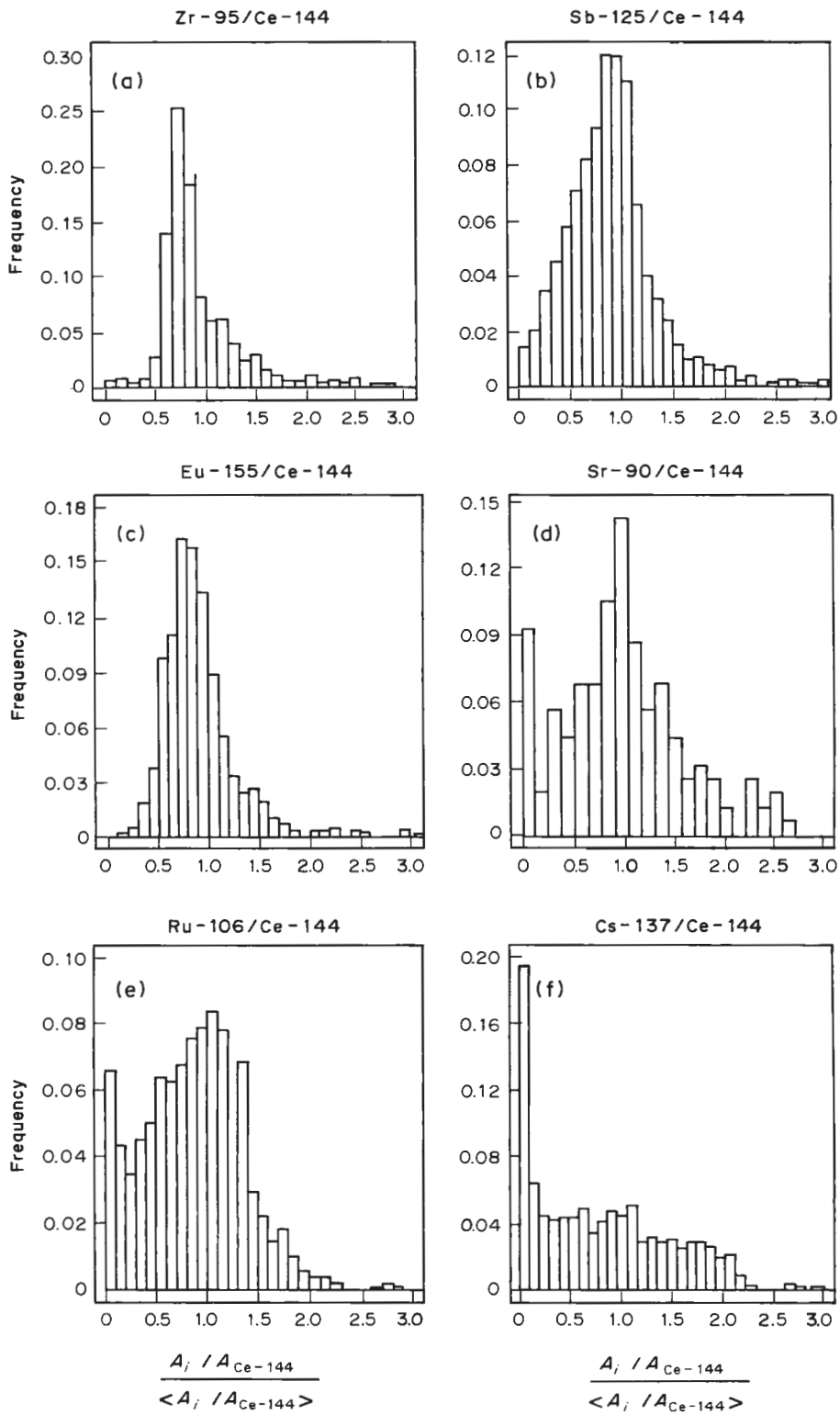

Fig. 2—*caption opposite.*

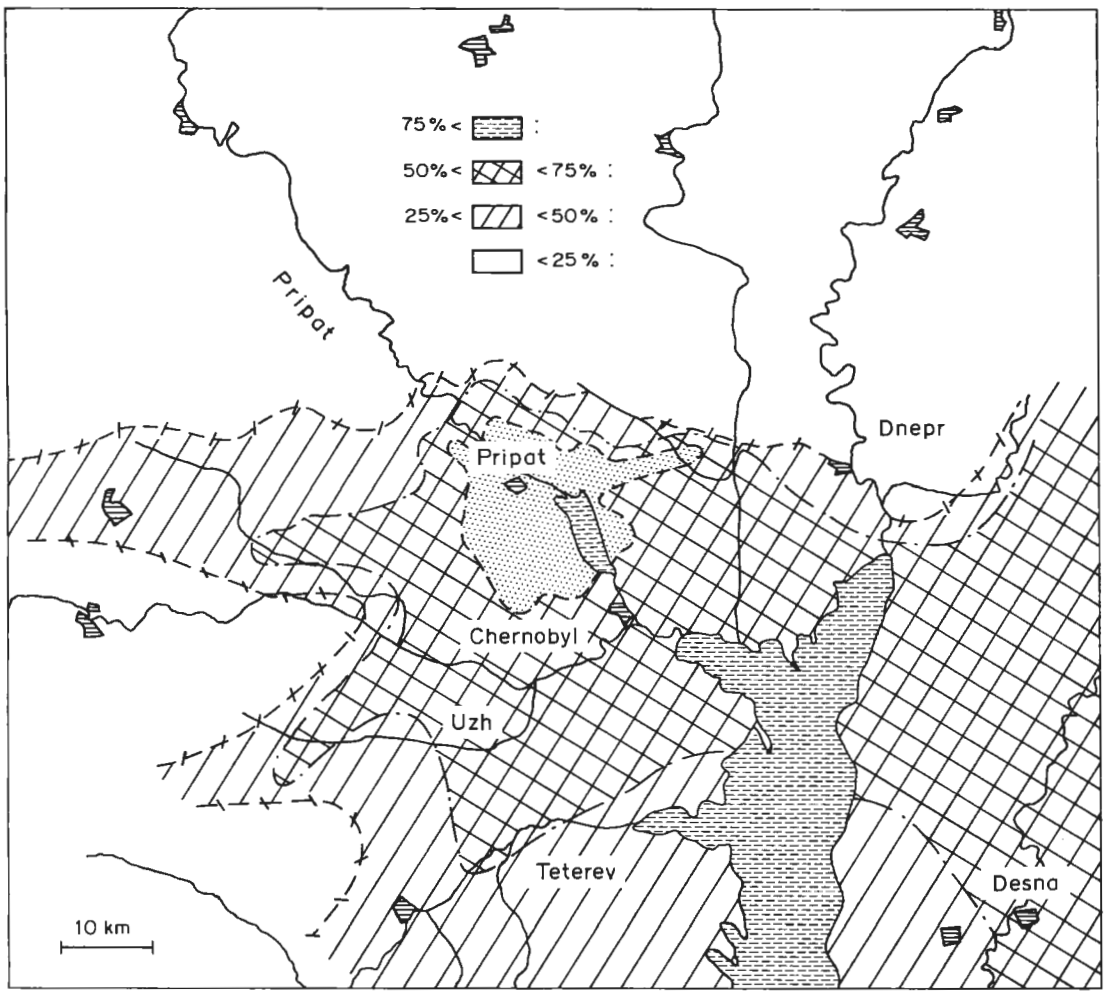

Fig. 3. The 60-km zone map depicting the part of Cs-137 contained in the FHPs relative to the total activity.

pH > 8 and redox potential $E_h > 0.45$ V, so areas with such soils are critical with respect to future enhancement of Sr-90 and, maybe, Pu uptake in vegetation, although the availability of Pu is generally poor for plant root uptake.

CONCLUSION

The Chernobyl accident has presented us with the challenge of understanding the behaviour of FHPs, released in considerable numbers into the environment. The major fraction of some potentially biologically important radionuclides, such as Sr and Pu, is contained in FHPs in biologically unavailable form. Destruction of these FHPs may lead to substantial changes in the radioecological situation. Also, inhalation of α- and β-emitting FHPs with high specific activities may turn out to be particularly harmful (Hohenemser, 1986; Richmond *et al.*, 1970). Using a representative sample of FHPs collected in 1986–1987

(Fig. 2 Opposite)

Fig. 2. Frequency histograms presenting the distribution of the ratios of different radionuclide activities (A_i) to Ce-144 activity ($A_{Ce\text{-}144}$) calculated for the moment of the accident (26 April 1986): (a) Zr-95/Ce-144; (b) Sb-125/Ce-144; (c) Eu-155/Ce-144; (d) Sr-90/Ce-144; (e) Ru-106/Ce-144; (f) Cs-137/Ce-144. Data on Sr-90 are obtained by radiochemistry; data on the rest of the radionuclides are obtained by γ-spectrometry. $\langle A_i/A_{Ce\text{-}144}\rangle$ is the sample-averaged value of the activity ratio.

we have obtained reliable data on their radionuclide composition, burn-up distribution and radionuclide fractionation. (These data can be used to constrain reconstruction of the Chernobyl accident.) Based on these FHP data, a 60-km zone map depicting the relative contributions of fuel and condensed forms of radio-Cs has been compiled.

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APPENDIX
(added in proof by A. E. Parshakov)

Anomalous Hot Particles

There are several scenarios of the accident at Chernobyl. Some of them (Hohenemser, 1986) assume that the accident was brought about by a sharp increase in the fission neutron flux. Still, the Soviet officials (the Resolution of the Central Committee of the Communist Party of the Soviet Union and the Council of Ministers of the U.S.S.R., *Pravda,* 9 June 1986) stated that the accident had a "non-nuclear" character.

Careful analysis of the radionuclide content of the HPs released due to the accident should allow us to choose between the two options: Cs-137 and Eu-155 are fission products whereas Cs-134 and Eu-154 are activation products, so the production of Cs-134 and Eu-154, in the case of a nuclear explosion, is several orders of magnitude less than the corresponding production of Cs-137 and Eu-155. In the case of normal reactor operation there is no great difference in the production of Cs-134 and Cs-137 and of Eu-154 and Eu-155.

I have analysed the information presented in the data base "Chernobyl Hot Particles", which contains information on the radionuclide composition of more than 1200 Chernobyl HPs. I could actually find about 10 anomalous HPs with the activity ratios of Eu-155/Eu-154 and of Cs-137/Cs-134 being in excess of the typical ratios by 1, 2 or even more orders of magnitude. In some of them the content of Eu-154 is below the detection limit. There are well-defined peaks corresponding to Cs-137 and Eu-155 in the γ-spectra of these anomalous HPs, whereas there are no Eu-154 peaks in the spectra or they are just a bit (not more than twice) higher than the noise level. Most of the anomalous HPs are strongly depleted in Cs-137 and Ru-106—this is evidence that they have experienced very high temperatures (a sharp increase in fission neutron flux evidently leads to a great energy release in the same area). However, the Cs-137/Ce-144 activity ratio for some of the anomalous HPs is quite normal, but the Cs-134/Ce-144 activity ratio is extremely small—the extra Cs-137 could condense on the surface of the HPs from the stream of volatile Cs-137, but the stream should have an anomalously small content of Cs-134. The anomalous activity ratios for isotopes of the same element can be explained by nuclear-physics reasons only: no chemical or diffusion processes can lead to a deviation of these ratios from the usual values by several orders of magnitude. The activity ratios for Sb-125, Eu-155 and Ce-144 are within the usual range.

To date, there are operating NPPs with the Chernobyl type reactors (RBMK-1000) in the Ukraine and other republics of the former Soviet Union, so it is extremely important to know what actually happened at Chernobyl. The very existence of the anomalous HPs strongly supports the idea of a sharp fission neutron flux increase, but still it is not clear whether such an increase was the reason for the explosion or if the desctruction of the reactor led to the fission neutron flux increase. To learn more about the parameters of the explosion it is desirable to use optical, solid-state and chemical methods for investigation of the anomalous HPs: the concentration of point defects can give information about temperatures experienced by the HPs, loop dislocation concentration should provide information on the pressures during the explosion etc.