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# Radioactive inventories from the Kyshtym and Karachay accidents: estimates based on soil samples collected in the South Urals (1990–1995)

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**Abstract**

The implementation of the nuclear programme in the Cheliabinsk region in the Ural, where plutonium for the first Soviet nuclear weapons was produced, involved radioactive contamination of the environment. The end of the cold war in the late 1980s initiated a fruitful co-operation between Russian and Western radioecologists. The present study is a joint Russian–Ukrainian–Danish effort to make an independent estimate of the inventories of $^{90}$Sr, $^{137}$Cs and $^{239,240}$Pu from two major contamination events in the South Urals, namely, the Kyshtym accident in 1957 and the Karachay wind dispersion in 1967. The calculations are based upon deposition measurements of the radionuclides carried out on soil samples assuming that the depositions decreased exponentially with distance from the two sources. The inventory estimates are compared with the available Russian information on the two accidents. © 1997 Elsevier Science B.V.

*Keywords:* MAYAK; Radioactive contaminations; Strontium-90; Caesium-137; Plutonium; Americium

## 1. Introduction

In 1990 a delegation from the International Union of Radioecologists (IUR) was invited to Ekaterinburg (at that time Sverdlovsk) by the Russian Academy of Science Ural Branch. The IUR delegation visited some of the contaminated sites from the so-called Kyshtym accident, which

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occurred in 1957 (Romanov et al., 1990). Together with the Institute of Plant and Animal Ecology, Russia (IPAE) the Institute of Biology of the Southern Seas, Ukraine (IBSS) samples of soil were collected and analysed for radioactive contamination. It appeared that the samples were contaminated not only by radioactivity from the Kyshtym accident, but also by global fallout from nuclear weapons testing in the 1950s and 1960s, by fallout from the Chernobyl accident in 1986 and by (at that time) an unknown source (Aarkrog et al., 1992). The unknown source was airborne debris from Lake Karachay in 1967, which since the early 1950s had been used by the nuclear weapon plutonium production association 'MAYAK' for storage of liquid radioactive waste (Academy of Science, 1991.

Earlier studies of the Kyshtym and Karachay contaminations have been carried out throughout the years (Izrael et al., 1993; Tsaturov and Anisimova, 1993). Western scientists have, however, first in this decade been able to participate in the studies because the two contamination events were kept secret until the cold war ended in the late 1980s.

The aim of the present study, which has been carried out in a co-operation between IPAE, IBSS and Risø National Laboratory was to make an independent estimate of the inventories of $^{90}$Sr, $^{137}$Cs and transuranic elements from the Kyshtym and Karachay accidents with special emphasis on plutonium, for which the information is less comprehensive (Pavlotskaya et al., 1992). Such an estimate is difficult because the events to some extent cover the same areas and the soil samples available may not be sufficient to describe the situation. It is complicated also because radionuclides originating from other, more or less well known, sources may interfere. Finally, exact information on the radionuclide composition from the various sources are only partly known. Any estimate of the contributions from 'Kyshtym' and 'Karachay' has thus to be based on several assumptions and is consequently encumbered with a considerable uncertainty. We can, however, compare our measurements with earlier data obtained for the same locations and see if any systematic differences appear.

## 2. Materials and methods

The soil samples were collected between 1990 and 1995 by IPAE after a method described earlier (Aarkrog et al., 1992), where cross-contamination between the soil layers is avoided. Most samples were collected to a depth of 30 cm and divided in 5-cm layers. Plant material (grass and litter) was usually analysed separately. Fig. 1 shows the sample locations. In the tables with the results the co-ordinates of the sample sites are given together with the distances from MAYAK, which was assigned the co-ordinates of Lake Karachay: 55°42′N 60°48, 5′E. After sampling, the soil was dried and sieved (1 mm mesh size) before analysis. All samples were measured by Ge $\gamma$-spectroscopy. Radiochemical analysis for $^{90}$Sr (Harley, 1972) and $^{239,240}$Pu (Talvitie, 1971) were carried out on 10-g aliquots of the treated soil samples. The results were reported as kBq m$^{-2}$ with $\pm 1$ S.D. due to counting statistics.

## 3. Results and discussion

The results of the radionuclide determinations are given in Tables 1–25. From the vertical distribution of the various radionuclides it can be concluded that for all locations except No. 19 (Tygish NW-bank) essentially all activity was found in the upper 30 cm. In case of $^{90}$Sr, the 25–30 cm layer contained on the average 1.5% of the total inventory of $^{90}$Sr in the 0–30 cm layer, vegetation included; the median value of the 23 locations sampled to 30 cm or more was 1%. In the case of $^{137}$Cs and transuranics (Pu and Am) even less was found in the deeper parts of the soil column. The activity levels in the 25–30 cm layer were for these radionuclides often below the limits of detection, i.e. $\leq 0.1$ kBq $^{137}$Cs m$^{-2}$ and $\leq 1$ Bq $^{239,240}$Pu m$^{-2}$.

Location No. 19 is special because at this site the contaminated top soil layers were displaced to a greater depth after the Kyshtym accident in 1957. Hence we find the highest $^{90}$Sr concentrations, representing the Kyshtym debris, in a depth of 50–100 cm at this location.

In order to estimate the contributions of the

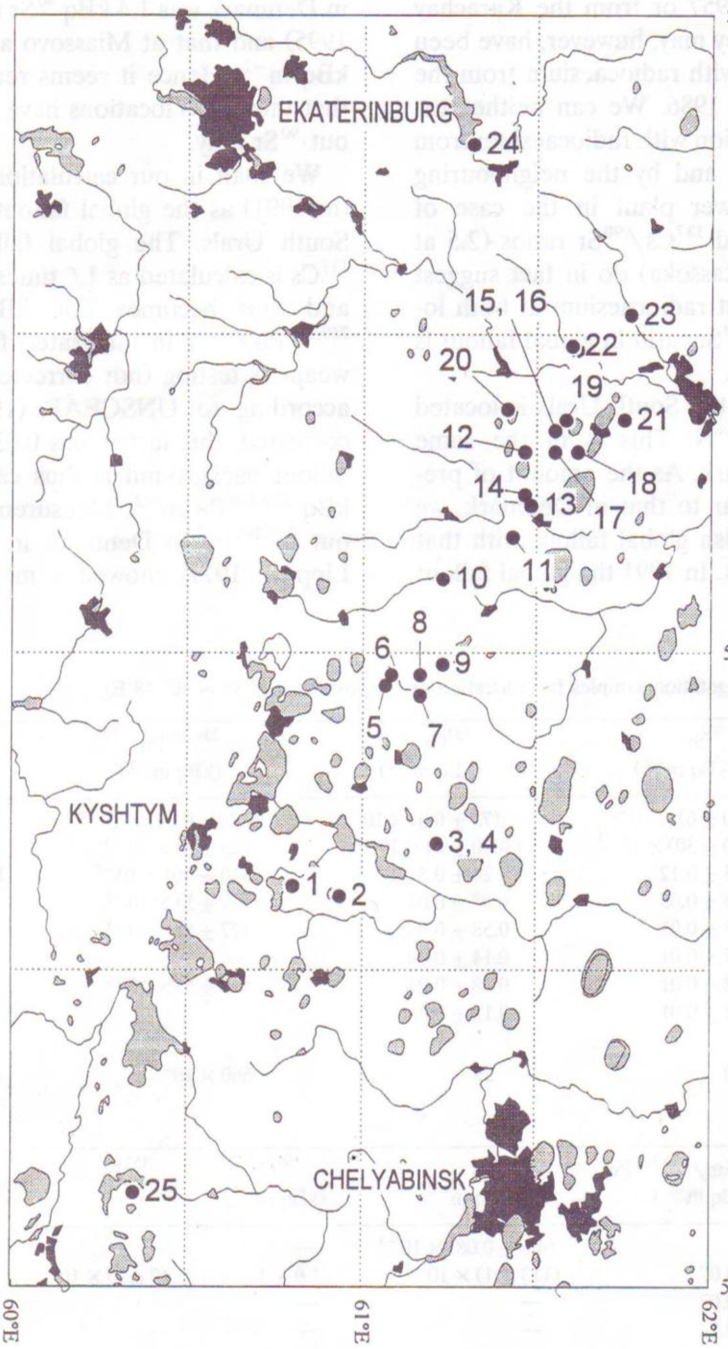


Fig. 1. Sampling locations for soil (1990–1995) in the South Urals.

various radionuclides from the Kysthtym and Karachay accidents a number of assumptions were made. Firstly we had to establish the background of global fallout from nuclear weapons testing. Two locations No. 24 (Rassoka) and No. 25 (Miassovo) were both considered to represent global fallout background with regard to $^{90}$Sr. None of these locations received fallout from the

Kyshtym accident in 1957 or from the Karachay dispersion in 1967. They may, however, have been slightly contaminated with radiocaesium from the Chernobyl accident in 1986. We can neither exclude some contamination with radiocaesium from MAYAK at Miassovo and by the neighbouring Beloyarsk nuclear power plant in the case of Rassoka. The enhanced $^{137}Cs/^{90}Sr$ ratios (2.5 at Miassovo and 3.2 at Rassoka) do in fact suggest some non-global fallout radiocaesium at both locations, as the $^{137}Cs/^{90}Sr$ ratio in global fallout is 1.6 (UNSCEAR, 1993).

The studied area in the South Urals is located between 55°N and 57°N. This is in the same latitude belt as Denmark. As the amount of precipitation also is similar to that in Denmark, we may compare the Danish global fallout with that in this part of the Urals. In 1991 the global fallout in Denmark was 1.4 kBq $^{90}Sr$ $m^{-2}$ (Aarkrog et al., 1995) and that at Miassovo and Rassoka was 1.6 kBq $m^{-2}$. Hence it seems reasonable to assume, that these two locations have received global fallout $^{90}Sr$ only.

We shall in our calculations use 1.6 kBq $^{90}Sr$ (in 1991) as the global fallout background in the South Urals. The global fallout background of $^{137}Cs$ is calculated as 1.6 times the $^{90}Sr$ deposition and thus becomes 2.56 kBq $m^{-2}$. The ratio $^{239,240}Pu/^{90}Sr$ in integrated fallout from nuclear weapons testing (not corrected for decay $^{90}Sr$) is according to UNSCEAR (1993) 0.0182. Decay corrected, this factor was 0.0367 in 1991 and the fallout background is thus calculated to be 0.06 kBq $^{239,240}Pu$ $m^{-2}$. Measurements of global fallout $^{239,240}Pu$ in Denmark in 1976 (Aarkrog and Lippert, 1977) showed a mean of $0.061 \pm 0.011$

Table 1
Radionuclides in soil and vegetation samples from location no. 1 (position: 55° 38′N 60° 48′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{239,240}Pu$ (kBq $m^{-2}$) | $^{238}Pu/^{239,240}Pu$ |
|---|---|---|---|---|
| Grass | $(390 \pm 6) \times 10^{-3}$ | $(73 \pm 0.3) \times 10^{-3}$ | $(0.28 \pm 0.02) \times 10^{-3}$ | $1.61 \pm 0.16$ |
| Litter | $(2030 \pm 30) \times 10^{-3}$ | $(3000 \pm 6) \times 10^{-3}$ | $(25 \pm 3) \times 10^{-3}$ | $0.64 \pm 0.11$ |
| Soil 0–5 cm | $9.3 \pm 0.12$ | $24 \pm 0.3$ | $(540 \pm 40) \times 10^{-3}$ | $0.085 \pm 0.02$ |
| Soil 5–10 cm | $3.3 \pm 0.05$ | $0.98 \pm 0.04$ | $(47 \pm 5) \times 10^{-3}$ | $0.23 \pm 0.07$ |
| Soil 10–15 cm | $0.53 \pm 0.01$ | $0.58 \pm 0.05$ | $(27 \pm 5) \times 10^{-3}$ | — |
| Soil 15–20 cm | $0.137 \pm 0.01$ | $0.14 \pm 0.04$ | — | — |
| Soil 20–25 cm | $0.158 \pm 0.01$ | $0.08 \pm 0.03$ | $(23 \pm 5) \times 10^{-3}$ | — |
| Soil 25–30 cm | $0.142 \pm 0.01$ | $0.12 \pm 0.03$ | — | — |
| Total deposition | 16.0 | 29 | $660 \times 10^{-3}$ | 0.111 |

| Sample | $^{241}Am/^{239,240}Pu$ (kBq $m^{-2}$) | $^{134}Cs$ (kBq $m^{-2}$) | $^{60}Co$ (kBq $m^{-2}$) | $^{152}Eu$ | $^{244}Cm/^{239,240}Pu$ |
|---|---|---|---|---|---|
| Grass | — | $(4.0 \pm 0.08) \times 10^{-3}$ | — | — | — |
| Litter | $0.37 \pm 0.07$ | $(113 \pm 1) \times 10^{-3}$ | $(1.9 \pm 1$ | $(7 \pm 1) \times 10^{-3}$ | $0.16 \pm 0.04$ |
| Soil 0–5 cm | $0.21 \pm 0.02$ | — | — | — | — |
| Soil 5–10 cm | $0.51 \pm 0.12$ | — | — | — | — |
| Soil 10–15 cm | — | — | — | — | — |
| Soil 15–20 cm | — | — | — | — | — |
| Soil 20–25 cm | — | — | — | — | — |
| Soil 25–30 cm | — | — | — | — | — |
| Total deposition | 0.22 | $117 \times 10^{-3}$ | $1.9 \times 10^{-3}$ | $7 \times 10^{-3}$ | — |

Samples were collected in September 1992, 7 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 2
Radionuclides in soil and vegetation samples from location no. 2 (position: 55° 37′N 60° 56′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{239,240}Pu$ (kBq $m^{-2}$) | $^{238}Pu/^{239,240}Pu$ | $^{241}Am/^{239,240}Pu$ | $^{134}Cs$ (kBq $m^{-2}$) | $^{60}Co$ (kBq $m^{-2}$) |
|---|---|---|---|---|---|---|---|
| Grass | $(53 \pm 0.8) \times 10^{-3}$ | $(67 \pm 1) \times 10^{-3}$ | $(0.033 \pm 0.003) \times 10^{-3}$ | $2.4 \pm 0.30$ | $1.36 \pm 0.173$ | $(2.8 \pm 0.1) \times 10^{-3}$ | — |
| Litter | $(804 \pm 10) \times 10^{-3}$ | $(2900 \pm 6) \times 10^{-3}$ | $(67 \pm 4) \times 10^{-3}$ | $0.101 \pm 0.010$ | $0.51 \pm 0.043$ | $(84 \pm 20 \times 10^{-3}$ | $(1.9 \pm 0.3) \times 10^{-3}$ |
| Soil 0–5 cm | $12.0 \pm 0.16$ | $27 \pm 0.3$ | $(1080 \pm 65) \times 10^{-3}$ | $0.148 \pm 0.015$ | $0.185 \pm 0.019$ | $(110 \pm 30 \times 10^{-3}$ | — |
| Soil 5–10 cm | $3.8 \pm 0.05$ | $64 \pm 0.6$ | $(136 \pm 18) \times 10^{-3}$ | $0.37 \pm 0.114$ | $0.096 \pm 0.039$ | $(270 \pm 70) \times 10^{-3}$ | — |
| Soil 10–15 cm | $0.53 \pm 0.01$ | $0.24 \pm 0.08$ | $(11 \pm 2) \times 10^{-3}$ | — | $0.27 \pm 0.104$ | — | — |
| Soil 15–20 cm | $0.34 \pm 0.01$ | $0.14 \pm 0.06$ | $(5 \pm 2) \times 10^{-3}$ | — | — | — | — |
| Soil 20–25 cm | $0.26 \pm 0.01$ | $0.27 \pm 0.09$ | $(5 \pm 2) \times 10^{-3}$ | — | — | — | — |
| Soil 25–30 cm | $0.187 \pm 0.07$ | $0.16 \pm 0.06$ | — | — | — | — | — |
| Total deposition | 18.0 | 94 | 1.30 | 0.166 | 0.192 | $470 \times 10^{-3}$ | $1.9 \times 10^{-3}$ |

Samples were collected in August 1993, 12 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

kBq $m^{-2}$ (±1 S.D., $N=8$). The fallout background of $^{241}Am$ is according to UNSCEAR 0.025 kBq $^{241}Am$ $m^{-2}$.

In an earlier study (Aarkrog et al., 1992) we assumed that the $^{90}Sr/^{137}Cs$ ratio in Kyshtym debris was 71 and that the deposition from the Karachay dispersion showed a $^{90}Sr/^{137}Cs$ ratio of 0.3. These assumptions were based on information given by Nikipelov (1989) and Nikipelov et al. (1990). Romanov et al. (1990) have given a $^{90}Sr/^{137}Cs$ in Kyshtym debris of 75 and if the ratio between $^{90}Sr$ and $^{137}Cs$ disposed in Lake Karachay (Nikipelov et al., 1990) is used for the Karachay dispersion we get 0.2 instead of 0.3. In the calculations shown below we have however, used the first mentioned ratios, i.e. 71 and 0.3 because we don't think that the other ratios differ significantly from those previously used.

A set of four equations were set up for each location and solved in order to calculate the contributions of $^{90}Sr$ and $^{137}Cs$ from the Kyshtym accident and the Karachay dispersion respectively:

$$x+y=a;\ v+p=b;\ -71x+v=0;\ -0.3y+p=0$$

$x$: kBq $^{137}Cs$ $m^{-2}$ from Kyshtym
$y$: kBq $^{137}Cs$ $m^{-2}$ from Karachay
$v$: kBq $^{90}Sr$ $m^{-2}$ from Kyshtym
$p$: kBq $^{90}Sr$ $m^{-2}$ from Karachay
$a$ is the total measured $^{137}Cs$ deposition at the location in kBq $m^{-2}$ minus the contribution from global fallout and minus the contribution from other sources, e.g. Chernobyl determined from the $^{134}Cs$ content.
$b$ is the total measured $^{90}Sr$ deposition at the location in kBq $m^{-2}$ minus the contribution from global fallout.

Table 3
Radionuclides in soil and vegetation samples from location no. 3 (position: 55° 42′N 61° 13′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{239,240}Pu$ (kBq $m^{-2}$) | $^{238}Pu/^{239,240}Pu$ |
|---|---|---|---|---|
| Grass | $(161 \pm 2) \times 10^{-3}$ | $(25 \pm 0.1) \times 10^{-3}$ | $(0.105 \pm 0.008) \times 10^{-3}$ | $0.64 \pm 0.075$ |
| Litter | $(1530 \pm 20) \times 10^{-3}$ | $(1460 \pm 3) \times 10^{-3}$ | $(39 \pm 3.5) \times 10^{-3}$ | $0.41 \pm 0.051$ |
| Soil 0–5 cm | $7.50 \pm 0.10$ | $43 \pm 0.1$ | $(629 \pm 38) \times 10^{-3}$ | $0.045 \pm 0.007$ |
| Soil 5–10 cm | $6.8 \pm 0.09$ | $3.2 \pm 0.06$ | $(67 \pm 7) \times 10^{-3}$ | $0.075 \pm 0.031$ |
| Soil 10–15 cm | $2.3 \pm 0.03$ | $0.45 \pm 0.03$ | $(18 \pm 4) \times 10^{-3}$ | — |
| Soil 15–20 cm | $0.53 \pm 0.01$ | $0.26 \pm 0.03$ | $(7.9 \pm 1.4) \times 10^{-3}$ | — |
| Soil 20–25 cm | $0.115 \pm 0.007$ | $0.18 \pm 0.03$ | $(6.9 \pm 1.6) \times 10^{-3}$ | — |
| Soil 25–30 cm | $0.047 \pm 0.005$ | $0.08 \pm 0.03$ | — | — |
| Total deposition | 19.0 | 49 | $770 \times 10^{-3}$ | 0.064 |

| Sample | $^{241}Am/^{239,240}Pu$ (kBq $m^{-2}$) | $^{134}Cs$ (kBq $m^{-2}$) | $^{60}Co$ (kBq $m^{-2}$) | $^{152}Eu$ | $^{244}Cm/^{239,240}Pu$ |
|---|---|---|---|---|---|
| Grass | $0.47 \pm 0.067$ | $(1.5 \pm 0.04) \times 10^{-3}$ | — | — | $0.21 \pm 0.041$ |
| Litter | $0.49 \pm 0.210$ | $(27 \pm 0.4) \times 10^{-3}$ | $(1.5 \pm 0.2) \times 10^{-3}$ | $(3.4 \pm 0.6) \times 10^{-3}$ | $0.022 \pm 0.005$ |
| Soil 0–5 cm | $0.186 \pm 0.026$ | — | — | — | — |
| Soil 5–10 cm | — | — | — | — | — |
| Soil 15–20 cm | — | — | — | — | — |
| Soil 20–25 cm | — | — | — | — | — |
| Soil 25–30 cm | — | — | — | — | — |
| Total | 0.177 | $29 \times 10^{-3}$ | $1.5 \times 10^{-3}$ | $3.4 \times 10^{-3}$ | — |

Samples collected in September 1992, 25 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 4
Radionuclides in soil and vegetation samples from location no. 4 (position: 55° 42′N 61° 13′E)

| Sample | $^{90}Sr$ (kBq m$^{-2}$) | $^{137}Cs$ (kBq m$^{-2}$k) | $^{239,240}Pu$ (kBq m$^{-2}$) | $^{238}Pu/^{239,240}Pu$ | $^{241}Am/^{239,240}Pu$ | $^{134}Cs$ (kBq m$^{-2}$) | $^{244}Cm/^{239,240}Pu$ |
|---|---|---|---|---|---|---|---|
| Grass | $(21 \pm 0.3) \times 10^{-3}$ | $(13.7 \pm 0.1) \times 10^{-3}$ | $(0.062 \pm 0.006) \times 10^{-3}$ | $1.65 \pm 0.205$ | $0.85 \pm 0.105$ | $(0.5 \pm 0.03) \times 10^{-3}$ | $0.065 \pm 0.017$ |
| Litter | $(1230 \pm 16) \times 10^{-3}$ | $(6200 \pm 19) \times 10^{-3}$ | $(40 \pm 2) \times 10^{-3}$ | $0.20 \pm 0.020$ | $0.31 \pm 0.025$ | $(57 \pm 3) \times 10^{-3}$ | $0.043 \pm 0.0005$ |
| Soil 0–5 cm | $7.1 \pm 0.10$ | $37 \pm 1.1$ | $(560 \pm 39) \times 10^{-3}$ | $0.030 \pm 0.007$ | $0.138 \pm 0.017$ | — | — |
| Soil 5–10 cm | $4.6 \pm 0.06$ | $1.68 \pm 0.1$ | $(68 \pm 9) \times 10^{-3}$ | — | $0.103 \pm 0.032$ | — | — |
| Soil 10–15 cm | $1.32 \pm 0.02$ | $0.78 \pm 0.05$ | $(12 \pm 2) \times 10^{-3}$ | — | — | — | — |
| Soil 15–20 cm | $0.59 \pm 0.01$ | $2.8 \pm 0.1$ | $(22 \pm 3) \times 10^{-3}$ | — | — | — | — |
| Soil 20–25 cm | $0.39 \pm 0.01$ | $1.21 \pm 0.1$ | $(7.3 \pm 1.1) \times 10^{-3}$ | — | — | — | — |
| Soil 25–30 cm | $0.125 \pm 0.004$ | — | $(3.4 \pm 0.7) \times 10^{-3}$ | — | — | — | |
| Total deposition | 15.4 | 50 | $710 \times 10^{-3}$ | 0.035 | 0.135 | $58 \times 10^{-3}$ | |

Samples were collected in August 1993, 25 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 5
Radionuclides in soil and vegetation samples from location no. 5 (position: 55° 57′N 61° 04′E)

| Sample | $^{90}$Sr (kBq m$^{-2}$) | $^{137}$Cs (kBq m$^{-2}$) | $^{239,240}$Pu (kBq m$^{-2}$) | $^{238}$Pu/$^{239,240}$Pu | $^{241}$Am/$^{239,240}$Pu | $^{60}$Co (kBq m$^{-2}$) | $^{152}$Eu (kBq m$^{-2}$) |
|---|---|---|---|---|---|---|---|
| Grass (no sample) | | | | | | | |
| Litter (no sample) | | | | | | | |
| Soil 0–5 cm | $830 \pm 11$ | $73 \pm 1$ | $(3600 \pm 300) \times 10^{-3}$ | $0.008 \pm 0.0018$ | $0.097 \pm 0.011$ | $0.24 \pm 0.01$ | $0.59 \pm 0.04$ |
| Soil 5–10 cm | $980 \pm 13$ | $17.1 \pm 0.2$ | $(1340 \pm 80) \times 10^{-3}$ | $0.010 \pm 0.0016$ | $0.093 \pm 0.013$ | $0.09 \pm 0.02$ | $0.21 \pm 0.06$ |
| Soil 10–15 cm | $250 \pm 3$ | $1.4 \pm 0.03$ | $(129 \pm 13) \times 10^{-3}$ | $0.047 \pm 0.024$ | — | — | — |
| Soil 15–20 cm | $1.49 \pm 0.03$ | $0.1 \pm 0.03$ | $(11 \pm 3) \times 10^{-3}$ | $0.545 \pm 0.31$ | — | — | — |
| Soil 20–25 cm | $2.8 \pm 0.04$ | $0.3 \pm 0.02$ | $(49 \pm 8) \times 10^{-3}$ | $0.429 \pm 0.14$ | — | — | — |
| Soil 25–30 cm | $3.5 \pm 0.05$ | $0.9 \pm 0.04$ | $(48 \pm 9) \times 10^{-3}$ | — | — | — | — |
| Total deposition | 2068 | 93 | 5.2 | 0.014 | 0.092 | 0.33 | 0.80 |

Samples were collected in October 1991, 32 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

At location 2 the contribution from other sources (probably local fallout from the operation of MAJAK) was estimated by assuming the contributions of $^{90}$Sr and $^{137}$Cs from the Kyshtym accident to be zero at this location, because the wind came from the south-west during the accident.

In order to calculate the unknown ratios: $^{239,240}$Pu/$^{137}$Cs in Karachay debris and $^{239,240}$Pu/$^{90}$Sr in Kyshtym debris, seven locations were selected for the first ratio and six for the second. The selection was based on the above calculations of the contributions of $^{90}$Sr and $^{137}$Cs from the two accidents. The first set of locations i.e. numbers 1, 2, 3, 4, 14, 20 and 23 were identified as sites almost free of $^{137}$Cs contamination from the Kyshtym accident. These locations were assumed to have contained $^{239,240}$Pu from global fallout and the Karachay dispersion only. The $^{239,240}$Pu/$^{137}$Cs ratio in Karachay debris were calculated for these locations by dividing the global fallout corrected $^{239,240}$Pu with the above calculated $^{137}$Cs deposition from the Karachay accident. It appeared that this ratio ($r$) varied with

Table 6
Radionuclides in soil and vegetation samples from location no. 6 (position: 55° 58′N 61° 05q′E)

| Sample | $^{90}$Sr (kBq m$^{-2}$) | $^{137}$Cs (kBq m$^{-2}$) | $^{239,240}$Pu (kBq m$^{-2}$) | $^{238}$Pu/$^{239,240}$Pu | $^{241}$Am/$^{239,240}$Pu |
|---|---|---|---|---|---|
| Litter | $0.80 \pm 0.012$ | $0.24 \pm 0.002$ | $(3.2 \pm 0.26) \times 10^{-3}$ | $0.079 \pm 0.023$ | $0.29 \pm 0.037$ |
| Turf | $76 \pm 1.0$ | $10.5 \pm 0.10$ | $(320 \pm 29) \times 10^{-3}$ | $0.0128 \pm 0.0025$ | $0.110 \pm 0.013$ |
| Soil 0–5 cm | $143 \pm 1.8$ | $9.8 \pm 0.98$ | $(147 \pm 9.0) \times 10^{-3}$ | $0.0140 \pm 0.0030$ | $0.114 \pm 0.012$ |
| Soil 5–10 cm | $126 \pm 1.8$ | $1.34 \pm 0.053$ | $(88 \pm 6.1) \times 10^{-3}$ | — | $0.136 \pm 0.022$ |
| Soil 10–15 cm | $31 \pm 0.4$ | $0.134 \pm 0.019$ | $(8.5 \pm 1.02) \times 10^{-3}$ | — | $0.44 \pm 0.119$ |
| Soil 15–20 cm | $11.5 \pm 0.15$ | $0.084 \pm 0.018$ | $(5.9 \pm 0.95) \times 10^{-3}$ | — | — |
| Soil 20–25 cm | $5.9 \pm 0.08$ | — | $(3.1 \pm 0.65) \times 10^{-3}$ | — | — |
| Soil 25–30 cm | $3.0 \pm 0.04$ | — | $(2.7 \pm 0.63) \times 10^{-3}$ | — | — |
| Total deposition | 397 | 22 | $580 \times 10^{-3}$ | 0.0111 | 0.119 |

Samples collected in 1995, 34 km from MAYAK.
The error term is 1 S.D. due to counting statistics.
$^{60}$Co, Turf: $0.028 \pm 0.0071$ kBq m$^{-2}$.

Table 7
Radionuclides in soil and vegetation samples from location no. 7 (position: 55° 56′N 61° 10′E)

| Sample | $^{90}$Sr (kBq m$^{-2}$) | $^{137}$Cs (kBq m$^{-2}$) | $^{239,240}$Pu (kBq m$^{-2}$) | $^{238}$Pu/$^{239,240}$Pu | $^{241}$Am/$^{239,240}$Pu |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter | 0.77 ± 0.011 | 1.57 ± 0.031 | (13.5 ± 1.22) × 10$^{-3}$ | 0.117 ± 0.026 | 0.24 ± 0.033 |
| Soil 0–5 cm | 3.3 ± 0.045 | 5.3 ± 0.105 | (81 ± 9.7) × 10$^{-3}$ | 0.187 ± 0.066 | 0.21 ± 0.039 |
| Soil 5–10 cm | 1.29 ± 0.020 | 0.174 ± 0.028 | (3.8 ± 0.64) × 10$^{-3}$ | — | — |
| Soil 10–15 cm | 0.28 ± 0.008 | 0.119 ± 0.037 | (3.7 ± 0.95) × 10$^{-3}$ | — | — |
| Soil 15–20 cm | 0.163 ± 0.007 | 0.120 ± 0.035 | — | — | — |
| Soil 20–25 cm | 0.124 ± 0.006 | 0.101 ± 0.031 | (4.3 ± 0.77) × 10$^{-3}$ | — | — |
| Soil 25–30 cm | 0.079 ± 0.007 | — | — | — | — |
| Total deposition | 6.0 | 7.3 | 106 × 10$^{-3}$ | 0.157 | 0.193 |

Collected in June 1995, 34 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 8
Radionuclides in soil and vegetation samples from location no. 8 (position: 55° 58′N 61° 10′E)

| Sample | $^{90}$Sr (kBq m$^{-2}$) | $^{137}$Cs (kBq m$^{-2}$) | $^{239,240}$Pu (kBq m$^{-2}$) | $^{238}$Pu/$^{239,240}$Pu | $^{241}$Am/$^{239,240}$Pu |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter | 0.58 ± 0.008 | 0.83 ± 0.017 | (8.7 ± 0.70) × 10$^{-3}$ | 0.115 ± 0.015 | 0.23 ± 0.030 |
| Soil 0–5 cm | 2.0 ± 0.028 | 7.3 ± 1.10 | (88 ± 7.0) × 10$^{-3}$ | 0.040 ± 0.009 | 0.27 ± 0.031 |
| Soil 5–10 cm | 1.11 ± 0.016 | 0.44 ± 0.032 | (8.7 ± 0.87) × 10$^{-3}$ | — | 0.34 ± 0.108 |
| Soil 10–15 cm | 0.64 ± 0.013 | 0.109 ± 0.030 | (3.4 ± 0.82) × 10$^{-3}$ | — | — |
| Soil 15–20 cm | 0.159 ± 0.0071 | 0.058 ± 0.026 | — | — | — |
| Soil 20–25 cm | 0.048 ± 0.0074 | — | — | — | — |
| Soil 25–30 cm | 0.045 ± 0.0066 | — | — | — | — |
| Total deposition | 4.6 | 8.7 | 109 × 10$^{-3}$ | 0.041 | 0.27 |

Samples were collected in June 1995, 37 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 9
Radionuclides in soil and vegetation samples from location no. 9 (position: 55° 59′N 61° 14′E)

| Sample | $^{90}$Sr (kBq m$^{-2}$) | $^{137}$Cs (kBq m$^{-2}$) | $^{239,240}$Pu (kBq m$^{-2}$) | $^{238}$Pu/$^{239,240}$Pu | $^{241}$Am/$^{239,240}$Pu |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter | 2.6 ± 0.04 | 6.9 ± 0.07 | (101 ± 7.1) × 10$^{-3}$ | 0.096 ± 0.012 | 0.22 ± 0.022 |
| Soil 0–5 cm | 3.3 ± 0.04 | 9.9 ± 0.10 | (82 ± 5.7) × 10$^{-3}$ | — | 0.25 ± 0.036 |
| Soil 5–10 cm | 1.74 ± 0.025 | 0.64 ± 0.045 | (13.5 ± 1.22) × 10$^{-3}$ | 0.21 ± 0.041 | 0.098 ± 0.034 |
| Soil 10–15 cm | 0.37 ± 0.008 | 0.159 ± 0.027 | (4.4 ± 1.24) × 10$^{-3}$ | — | — |
| Soil 15–20 cm | 0.092 ± 0.0068 | 0.182 ± 0.036 | — | — | — |
| Soil 20–25 cm | 0.051 ± 0.0053 | — | — | — | — |
| Soil 25–30 cm | — | — | — | — | — |
| Total deposition | 8.2 | 17.8 | 200 × 10$^{-3}$ | 0.063 | 0.22 |

Samples were collected in June 1995, 41 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 10
Radionuclides in soil and vegetation samples from location no. 10 (position: 56° 07′N 61° 14′E)

| Sample | $^{90}$Sr (kBq m$^{-2}$) | $^{137}$Cs (kBq m$^{-2}$) | $^{239,240}$Pu (kBq m$^{-2}$) | $^{238}$Pu/$^{239,240}$Pu | $^{241}$Am/$^{239,240}$Pu |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter | $1.98 \pm 0.027$ | $10.6 \pm 0.11$ | $(40 \pm 2.4) \times 10^{-3}$ | $0.046 \pm 0.006$ | $0.31 \pm 0.031$ |
| Soil 0–5 cm | $10.5 \pm 0.14$ | $4.3 \pm 0.09$ | $(69 \pm 6.9) \times 10^{-3}$ | $0.116 \pm 0.042$ | $0.152 \pm 0.054$ |
| Soil 5–10 cm | $4.2 \pm 0.06$ | $0.79 \pm 0.055$ | $(10.1 \pm 1.11) \times 10^{-3}$ | $0.188 \pm 0.062$ | $0.20 \pm 0.069$ |
| Soil 10–15 cm | $0.70 \pm 0.012$ | $0.36 \pm 0.029$ | $(3.8 \pm 0.69) \times 10^{-3}$ | | |
| Soil 15–20 cm | $0.22 \pm 0.008$ | $0.193 \pm 0.041$ | $(2.9 \pm 0.68) \times 10^{-3}$ | | |
| Soil 20–25 cm | $0.091 \pm 0.0052$ | $0.055 \pm 0.029$ | $(4.7 \pm 0.65) \times 10^{-3}$ | | |
| Soil 25–30 cm | $0.072 \pm 0.0051$ | $0.033 \pm 0.017$ | $(2.8 \pm 0.84) \times 10^{-3}$ | — | |
| Total deposition | 17.8 | 16.4 | $134 \times 10^{-3}$ | 0.088 | 0.188 |

Samples were collected in June 1995, 53 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 11
Radionuclides in soil and vegetation samples from location no. 11 (position: 56° 11′N 61° 26′E)

| Sample | $^{90}$Sr (kBq m$^{-2}$) | $^{137}$Cs (kBq m$^{-2}$) | $^{239,240}$Pu (kBq m$^{-2}$) | $^{238}$Pu/$^{239,240}$Pu | $^{241}$Am/$^{239,240}$Pu |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter | $15.9 \pm 0.21$ | $4.7 \pm 0.09$ | $(75 \pm 6.0) \times 10^{-3}$ | $0.035 \pm 0.0059$ | $0.173 \pm 0.0184$ |
| Soil 0–5 cm | $4.4 \pm 0.06$ | $3.7 \pm 0.15$ | $(67 \pm 5.4) \times 10^{-3}$ | — | $0.24 \pm 0.036$ |
| Soil 5–10 cm | $3.1 \pm 0.05$ | $0.48 \pm 0.030$ | $(9.0 \pm 1.16) \times 10^{-3}$ | — | $0.43 \pm 0.125$ |
| Soil 10–15 cm | $1.25 \pm 0.022$ | $0.64 \pm 0.052$ | $(8.4 \pm 1.26) \times 10^{-3}$ | — | — |
| Soil 15–20 cm | $0.62 \pm 0.014$ | $0.37 \pm 0.040$ | $(9.6 \pm 1.64) \times 10^{-3}$ | — | $0.26 \pm 0.089$ |
| Soil 20–25 cm | $0.36 \pm 0.012$ | $0.098 \pm 0.038$ | — | — | — |
| Soil 25–30 cm | $0.148 \pm 0.010$ | $0.110 \pm 0.046$ | — | — | — |
| Total deposition | 26 | 10.1 | $169 \times 10^{-3}$ | 0.0153 | 0.21 |

Samples were collected in June 1995, 66 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 12
Radionuclides in soil and vegetation samples from location no. 12 (position: 56° 19′N 61° 28′E)

| Sample | $^{90}$Sr (kBq m$^{-2}$) | $^{137}$Cs (kBq m$^{-2}$) | $^{239,240}$Pu (kBq m$^{-2}$) | $^{238}$Pu/$^{239,240}$Pu | $^{241}$Am/239,$^{240}$Pu |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter | $3.8 \pm 0.054$ | $2.3 \pm 0.045$ | $(24 \pm 1.92) \times 10^{-3}$ | $0.030 \pm 0.006$ | $0.23 \pm 0.024$ |
| Soil 0–5 cm | $48 \pm 0.68$ | $3.5 \pm 0.104$ | $(108 \pm 8.6) \times 10^{-3}$ | — | $0.161 \pm 0.021$ |
| Soil 5–10 cm | $34 \pm 0.45$ | $0.29 \pm 0.057$ | $(28 \pm 3.9) \times 10^{-3}$ | — | — |
| Soil 10–15 cm | $16.1 \pm 0.22$ | $0.155 \pm 0.061$ | $(16.3 \pm 2.3) \times 10^{-3}$ | — | — |
| Soil 15–20 cm | $5.8 \pm 0.078$ | $0.098 \pm 0.032$ | — | — | — |
| Soil 20–25 cm | $2.5 \pm 0.036$ | $0.078 \pm 0.030$ | $(7.2 \pm 0.93) \times 10^{-3}$ | $0.50 \pm 0.13$ | $2.2 \pm 0.44$ |
| Soil 25–30 cm | $9.8 \pm 0.128$ | $0.100 \pm 0.040$ | $(8.0 \pm 1.04) \times 10^{-3}$ | — | — |
| Total deposition | 119 | 6.5 | $191 \times 10^{-3}$ | 0.022 | 0.20 |

Samples were collected in June 1995, 80 km from MAYAK.
The error term is 1 S.D. dueto counting statistics.

the distance ($d$ km) from Lake Karachay after the regression: $r = 0.025e^{-0.0186d}$. From this equation and from the calculated $^{137}Cs$ deposition from the Karachay dispersion the Pu deposition from Karachay was calculated for the six other locations.

These sites (numbers 5, 6, 12, 13, 19 and 22) were selected from their high contamination levels with Kyshtym debris. In this way the error from the above calculation of the Karachay-derived Pu was minimized, because this contribution would anyway be minor compared with the contribution of Pu from the Kyshtym accident. The $^{239,240}Pu/^{90}Sr$ ratio in Kyshtym debris was calculated for the six locations and as there was no significant variation with distance the mean ratio was calculated. The mean ratio for the six locations was $0.0018 \pm 0.0007$.

We may, however, use another approach in order to estimate the unknown $^{239,240}Pu/^{90}Sr$ ratio and $^{239,240}Pu/^{137}Cs$ ratios in Kyshtym and Karachay debris, respectively. If the $^{137}Cs/^{90}Sr$ ratios in the various layers at the above mentioned location 19 (see Table 19) were compared with the $^{90}Sr$ concentrations two distinct groups of data appeared: one with relatively high

Table 13
Radionuclides in soil and vegetation samples from location no. 13 (position 56° 14′N 61° 29′E)

| Sample | (kBq m$^{-2}$) | $^{137}Cs$ (kBq m$^{-2}$) | $^{239,240}Pu$ (kBq m$^{-2}$) | $^{238}Pu/^{239,240}Pu$ | $^{241}Am/^{239,240}Pu$ |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter | $0.67 \pm 0.009$ | $(5.2 \pm 0.31) \times 10^{-3}$ | $(0.058 \pm 0.046) \times 10^{-3}$ | | $0.41 \pm 0.077$ |
| Soil 0–5 cm | $25 \pm 0.31$ | $3.7 \pm 0.073$ | $(94 \pm 7.5) \times 10^{-3}$ | $0.018 \pm 0.005$ | $0.160 \pm 0.017$ |
| Soil 5–10 cm | $55 \pm 0.71$ | $5.6 \pm 0.11$ | $(175 \pm 12) \times 10^{-3}$ | — | $0.140 \pm 0.017$ |
| Soil 10–15 cm | $25 \pm 0.31$ | $0.56 \pm 0.084$ | $(27 \pm 2.1) \times 10^{-3}$ | — | — |
| Soil 15–20 cm | $3.5 \pm 0.048$ | $0.025 \pm 0.014$ | $(1.73 \pm 0.43) \times 10^{-3}$ | | |
| Soil 20–25 cm | $2.4 \pm 0.035$ | $0.061 \pm 0.024$ | — | — | — |
| So.02 ± 0.018 | $0.042 \pm 0.026$ | $(3.0 \pm 0.62) \times 10^{-3}$ | — | — | |
| Total deposition | 112 | 9.9 | $300 \times 10^{-3}$ | 0.0057 | 0.132 |

Samples were collected in June 1995, 73 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 14
Radionuclides in soil and vegetation samples from location no. 14 (position: 56° 15′N 61° 28′E)

| Sample | $^{90}Sr$ (kBq m$^{-2}$) | $^{137}Cs$ (kBq m$^{-2}$) | $^{239,240}Pu$ (kBq m$^{-2}$) | $^{238}Pu/^{239,240}Pu$ | $^{241}Am/^{239,240}Pu$ |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter | $0.30 \pm 0.004$ | $1.35 \pm 0.013$ | $(9.9 \pm 0.59) \times 10^{-3}$ | $0.046 \pm 0.005$ | $0.32 \pm 0.034$ |
| Soil 0–5 cm | $1.44 \pm 0.021$ | $4.1 \pm 0.083$ | $(54 \pm 4.9) \times 10^{-3}$ | $0.052 \pm 0.021$ | $0.37 \pm 0.065$ |
| Soil 5–10 cm | $0.67 \pm 0.012$ | $0.41 \pm 0.050$ | $(6.2 \pm 0.98) \times 10^{-3}$ | — | $1.32 \pm 0.40$ |
| Soil 10–15 cm | $0.23 \pm 0.007$ | $0.36 \pm 0.043$ | $(7.0 \pm 1.48) \times 10^{-3}$ | — | — |
| Soil 15–20 cm | $0.22 \pm 0.008$ | — | — | — | — |
| Soil 20–25 cm | $0.075 \pm 0.006$ | $0.041 \pm 0.023$ | — | — | — |
| Soil 25–30 cm | $0.075 \pm 0.006$ | $0.072 \pm 0.023$ | — | — | — |
| Total deposition | 3.0 | 6.4 | $77 \times 10^{-3}$ | 0.042 | 0.40 |

Samples were collected in June 1995, 74 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 15
Radionuclides in soil and vegatation samples from Tygish cemetary (location no. 15; position: 56° 21′N 61° 33′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{239,240}Pu$ (kBq $m^{-2}$) | $^{238}Pu/^{239,240}Pu$ | $^{241}Am/^{239,240}Pu$ |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter (no sample) | | | | | |
| Soil 0–5 cm | $22 \pm 0.2$ | $7.9 \pm 0.08$ | $(100 \pm 6) \times 10^{-3}$ | $0.027 \pm 0.005$ | $0.173 \pm 0.019$ |
| Soil 5–10 cm | $25 \pm 1.3$ | $2.0 \pm 0.02$ | $(38 \pm 3) \times 10^{-3}$ | — | $0.21 \pm 0.041$ |
| Soil 10–15 cm | $4.5 \pm 0.1$ | $0.15 \pm 0.01$ | $(3.5 \pm 0.5) \times 10^{-3}$ | — | $0.60 \pm 0.218$ |
| Soil 15–20 cm | $10.8 \pm 0.5$ | $0.63 \pm 0.01$ | $(1.1 \pm 0.3) \times 10^{-3}$ | — | — |
| Soil 20–25 cm | $0.80 \pm 0.02$ | $0.05 \pm 0.01$ | — | — | — |
| Soil 25–30 cm | $0.130 \pm 0.02$ | — | — | — | — |
| Total deposition | 63 | 10.7 | $143 \times 10^{-3}$ | 0.0189 | 0.193 |

Samples were collected in 1990, 86 km from MAYAK.
The error term is 1 S.D. due to counting statistics, but for $^{90}Sr$ it is 1 SE of double determinations.
$^{134}Cs$, 0–5 cm: $0.20 \pm 0.01$ kBq $m^{-2}$.

$^{137}Cs/^{90}Sr$ ratios and low $^{90}Sr$ concentrations and another with low ratios and high concentrations. The 0–25 cm layer represented the first group of data and the 45–95 cm layer the second one. We assume that the 0–25 cm layer with respect to $^{137}Cs$ and $^{239,240}Pu$ was contaminated by Karachay debris only and that the 45–95 layer with respect to $^{90}Sr$ and $^{239,240}Pu$ contained Kyshtym fallout only. The two unknown ratios were determined from the two data sets. For Karachay debris (0–25 cm) we got $^{239,240}Pu/^{137}Cs = 0.0118 \pm 0.0024$ ($\pm 1$ S.D.); $N = 5$) and for Kyshtym (45–95 cm) we found $^{239,240}Pu/^{90}Sr = 0.0029 \pm 0.0008$ ($\pm 1$ S.D.; $N = 5$). The ratios may be compared with those estimated above. It appears that the new ratios are both somewhat higher than the previous estimates. The difference between the two estimates for the ratio: (Pu/Sr) Kyshtym was probably significant ($P < 0.05$) and for the (Pu/Cs) Karachay the difference between the two determinations was highly significant ($P < 0.001$).

In order to calculate the radionuclide inventories from the Kyshtym and Karachay accidents we assume that the $^{90}Sr$ deposition in kBq $m^{-2}$ from the Kyshtym accident followed an exponential decrease with distance from MAYAK and that

Table 16
Radionuclides in soil and vegetation samples from Tygish cementary (location no. 16; Position: 56° 21′N 61° 33′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{134}Cs$ (kBq $m^{-2}$) |
|---|---|---|---|
| Grass (no sample) | | | |
| Litter (no sample) | | | |
| Soil 0–5 cm | $8.4 \pm 0.1$ | $7.7 \pm 0.3$ | $0.15 \pm 0.01$ |
| Soil 5–10 cm | $11.9 \pm 0.2$ | $0.96 \pm 0.01$ | — |
| Soil 10–15 cm | $2.8 \pm 0.04$ | $0.17 \pm 0.01$ | — |
| Soil 15–20 cm | $0.48 \pm 0.01$ | $0.03 \pm 0.01$ | — |
| Soil 20–25 cm | $0.07 \pm 0.01$ | $0.05 \pm 0.01$ | — |
| Soil 25–30 cm | $0.27 \pm 0.01$ | $0.41 \pm 0.02$ | — |
| Total deposition | 24 | 9.3 | 0.15 |

Samples were collected in May 1991, 86 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 17
Radionuclides in soil and vegetation samples SW of Tygish cementary (location no. 17; position: 56° 19′N 61° 33′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{134}Cs$ (kBq $m^{-2}$) |
|---|---|---|---|
| Grass (no sample) | | | |
| Litter (no sample) | | | |
| Soil 0–5 cm | $1.32 \pm 0.02$ | $3.5 \pm 0.07$ | $0.13 \pm 0.02$ |
| Soil 5–10 cm | $1.21 \pm 0.02$ | $0.11 \pm 0.01$ | — |
| Soil 10–15 cm | $0.63 \pm 0.01$ | $0.05 \pm 0.01$ | — |
| Soil 15–20 cm | $0.28 \pm 0.01$ | $0.06 \pm 0.01$ | — |
| Soil 20–25 cm | $0.12 \pm 0.01$ | $0.02 \pm 0.01$ | — |
| Soil 25–30 cm | $0.06 \pm 0.01$ | $0.06 \pm 0.01$ | — |
| Total deposition | 3.6 | 3.8 | 0.13 |

Samples were collected in May 1991, 83 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 18
Radionuclides in soil and vegetation samples SE of Tygish cementary (location no. 18; position: 56° 19′N 61° 37′E)

| Sample | $^{90}$Sr (kBq m$^{-2}$) | $^{137}$Cs (kBq m$^{-2}$) | $^{134}$Cs (kBq m$^{-2}$) |
|---|---|---|---|
| Grass (no sample) | | | |
| Litter (no sample) | | | |
| Soil 0–5 cm | 46 ± 0.6 | 7.9 ± 0.3 | 0.12 ± 0.00 |
| Soil 5–10 cm | 28 ± 0.4 | 4.5 ± 0.1 | |
| Soil 10–15 cm | 10.5 ± 0.1 | 1.3 ± 0.03 | |
| Soil 15–20 cm | 5.2 ± 0.07 | 0.43 ± 0.03 | |
| Soil 20–25 cm | 2.6 ± 0.04 | 0.14 ± 0.02 | |
| Soil 25–30 cm | 0.38 ± 0.01 | 0.05 ± 0.01 | |
| Total deposition | 93 | 14.3 | 0.12 |

Samples were collected in May 1991, 85 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

the activity is confined within a 15° sector out to a distance of 300 km, which approximately describes the contamination pattern as e.g. shown by Romanov et al. (1990). We have furthermore neglected the effect of remedial measures, which at some locations apparently have removed most of the $^{90}$Sr contamination from the Kyshtym accident. Such sites were consequently not included in the calculations.

In the case of Karachay debris we also assume an exponential decrease of kBq $^{137}$Cs m$^{-2}$ with distance. We further assume that the debris was confined within a 60° sector out to a distance of 150 km from MAYAK. This model was derived from the contamination pattern shown by Izrael et al. (1993) and Tsaturov and Anisimova (1993). In this case all locations were included, which showed $^{137}$Cs from Karachay, as there apparently had been no decontamination efforts carried out after the Karachay accident in 1967.

For the $^{90}$Sr deposition from the Kyshtym accident (see Fig. 2) we have: $y = e^{(a+bx)}$,where

$a = 8.08 \pm 0.35$, $b = -0.048 \pm 0.015$
$y$ is kBq $^{90}$Sr m$^{-2}$ and $x$ = distance from MAYAK in km.

The $^{90}$Sr inventory ($I$) was calculated for the distance interval 30–300 km

$$I = 15/360 \cdot 2\pi \cdot 3240 \cdot 10^{-6} \int_{30}^{300} x \cdot e^{-0.048}\, dx$$

$$= 0.2\ \text{PBq}^{90}\text{Sr}$$

The uncertainty of the integral is about a factor

Table 19
Radionuclides in soil and vegetation samples from Tygish NW-bank (location no. 19; position: 56° 22′N 61° 35′E)

| Sample | $^{90}$Sr (kBq m$^{-2}$) | $^{137}$Cs (kBq m$^{-2}$) | $^{239,240}$Pu (kBq m$^{-2}$) | $^{238}$Pu/$^{239,240}$Pu | $^{241}$Am/$^{239,240}$Pu |
|---|---|---|---|---|---|
| Grass | (2900 ± 42) × 10$^{-3}$ | (1.9 ± 0.2) × 10$^{-3}$ | — | — | — |
| Litter | (400 ± 5) × 10$^{-3}$ | (133 ± 4) × 10$^{-3}$ | 1.5 ± 0.1) × 10$^{-3}$ | 0.067 ± 0.020 | 0.21 ± 0.036 |
| Soil 0–5 cm | 3.4 ± 0.1 | 1.58 ± 0.14 | (17 ± 1) × 10$^{-3}$ | 0.071 ± 0.018 | 0.19 ± 0.032 |
| Soil 5–10 cm | 5.9 ± 0.1 | 2.3 ± 0.16 | (24 ± 2) × 10$^{-3}$ | 0.042 ± 0.021 | 0.19 ± 0.029 |
| Soil 10–15 cm | 8.5 ± 0.1 | 2.8 ± 0.1 | (36 ± 3) × 10$^{-3}$ | 0.064 ± 0.020 | 0.21 ± 0.040 |
| Soil 15–20 cm | 9.5 ± 0.1 | 2.9 ± 0.2 | (45 ± 3) × 10$^{-3}$ | 0.033 ± 0.011 | 0.29 ± 0.069 |
| Soil 20–25 cm | 3.3 ± 0.1 | 0.65 ± 0.09 | (6 ± 0.7) × 10$^{-3}$ | — | 0.23 ± 0.057 |
| Soil 25–35 cm | 6.1 ± 0.1 | 0.97 ± 0.24 | (102 ± 10) × 10$^{-3}$ | — | — |
| Soil 35–45 cm | 13.2 ± 0.2 | 0.96 ± 0.16 | (55 ± 3) × 10$^{-3}$ | — | — |
| Soil 45–55 cm | 26 ± 0.4 | 1.53 ± 0.24 | (86 ± 6) × 10$^{-3}$ | — | 0.16 ± 0.026 |
| Soil 55–65 cm | 29 ± 0.4 | 1.47 ± 0.14 | (76 ± 5) × 10$^{-3}$ | — | 0.18 ± 0.029 |
| Soil 65–75 cm | 25 ± 0.4 | 1.02 ± 0.12 | (59 ± 6) × 10$^{-3}$ | — | 0.20 ± 0.040 |
| Soil 75–85 cm | 41 ± 0.5 | 2.5 ± 0.20 | (170 ± 14) × 10$^{-3}$ | — | 0.08 ± 0.013 |
| Soil 85–95 cm | 36 ± 0.4 | 1.24 ± 0.14 | (79 ± 8) × 10$^{-3}$ | — | 0.16 ± 0.030 |
| Total deposition | 211 | 20.0 | 760 × 10$^{-3}$ | 0.0081 | 0.127 |

Samples were collected in July 1993, 88 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

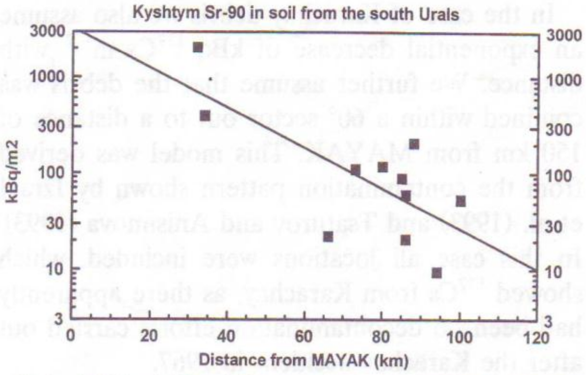


Fig. 2. Sr-90 deposited from the Kyshtym accident related to distance from MAYAK kBq $m^{-2}$ = 3243 $e^{-0.048\ km}$

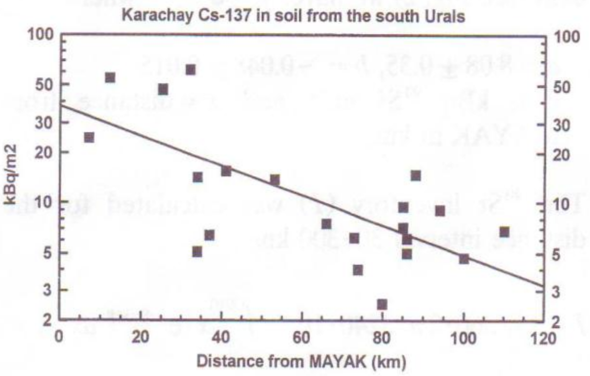


Fig. 3. Cs-137 deposited from the Karachay accident related to distance from MAYAK kBq $m^{-2}$ = 37.8 $e^{-0.0206\ km}$

Fig. 2. $^{90}$Sr deposited from the Kyshtym accident related to distance from MAYAK (kBq $m^{-2} = 3243\ e^{-0.048\ km}$).
Fig. 3. $^{137}$Cs deposited from the Karachay accident related to distance from MAYAK.

of 2.5, i.e. the 1 S.D. range is 0.1–0.5 PBq. If the integration had been carried out from 30 to 100 km (i.e. the distance where samples were collected) the inventory would have been 10% lower. In other words only 10% were according to the model, deposited further away than 100 km.

The deposition from 0 to 30 km would according to the model be 0.18 PBq. This figure is, however, very uncertain because we have no samples from this sector. The theoretical deposition from the Kyshtym accident should be about 1 PBq $^{90}$Sr in 1996 and of this at least half was deposited within 30 km from MAYAK (Romanov et al., 1990). Hence our estimate of 0.2 PBq between 30 and 300 km is not incompatible with the earlier data.

For the $^{137}$Cs deposition from the Karachay incident (see Fig. 3) we have: $y = e^{(a+bx)}$ where

$a = 3.68 \pm 0.144$, $b = -0.021 \pm 0.0048$
$y$ = kBq $^{137}$Cs $m^{-2}$ and $x$ = distance from MAYAK in km.

The $^{137}$Cs inventory ($I$) was calculated for the distance interval of 7–150 km:

$$I = 60/360 \cdot 2\pi \cdot 38 \cdot 10^{-3} \int_{7}^{150} x e^{-0.021x}\, dx$$

Table 20
Radionuclides in soil and vegetation samples from location no. 20 (position: 56° 23′N 61° 25′E)

| Sample | $^{90}$Sr (kBq $m^{-2}$) | $^{137}$Cs (kBq $m^{-2}$) | $^{239,240}$Pu (kBq $m^{-2}$) | $^{238}$Pu/$^{239,240}$Pu | $^{241}$Am/$^{239,240}$Pu |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter (no sample) | | | | | |
| Soil 0–5 cm | $0.90 \pm 0.02$ | $8.4 \pm 0.2$ | $(50 \pm 3) \times 10^{-3}$ | $0.040 \pm 0.006$ | $0.24 \pm 0.025$ |
| Soil 5–10 cm | $1.49 \pm 0.02$ | $2.5 \pm 0.1$ | $(34 \pm 2) \times 10^{-3}$ | — | $0.24 \pm 0.033$ |
| Soil 10–15 cm | $0.75 \pm 0.01$ | $0.73 \pm 0.05$ | $(8 \pm 0.8) \times 10^{-3}$ | — | $0.38 \pm 0.073$ |
| Soil 15–20 cm | $0.45 \pm 0.01$ | $0.34 \pm 0.06$ | $(6 \pm 0.7) \times 10^{-3}$ | — | $0.33 \pm 0.092$ |
| Soil 20–25 cm | $0.18 \pm 0.01$ | $0.33 \pm 0.06$ | $(3 \pm 0.5) \times 10^{-3}$ | — | — |
| Soil 25–30 cm | $0.10 \pm 0.01$ | — | $(2 \pm 0.4) \times 10^{-3}$ | $0.020 \pm 0.006$ | — |
| Soil 30–35 cm | $0.06 \pm 0.01$ | — | — | — | — |
| Soil 35–40 cm | $0.04 \pm 0.01$ | — | $(1 \pm 0.3) \times 10^{-3}$ | — | — |
| Total deposition | 3.97 | 12.3 | $104 \times 10^{-3}$ | 0.020 | 0.24 |

Samples were collected in October 1992, 85 km from MAYAK.
The error term is 1 S.D. due to counting statistics.
$^{134}$Cs. 0–5 cm: $0.22 \pm 0.04$ kBq $m^{-2}$.

Table 21
Radionuclides in soil and vegetation samples from location no. 21 (position: 56° 22′N 61° 45′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{239,240}Pu$ (kBq $m^{-2}$) | $^{238}Pu/^{239,240}Pu$ | $^{241}Am/^{239,240}Pu$ | $^{134}Cs$ (kBq $m^{-2}$) |
|---|---|---|---|---|---|---|
| Grass (no sample) | | | | | | |
| Litter (no sample) | | | | | | |
| Soil 0–2 cm | $1.21 \pm 0.02$ | $1.49 \pm 0.013$ | $(26.3 \pm 1.6) \times 10^{-3}$ | $0.029 \pm 0.005$ | $0.21 \pm 0.021$ | $(62 \pm 2) \times 10^{-3}$ |
| Soil 2–7 cm | $6.6 \pm 0.70$ | $7.7 \pm 0.077$ | $(120 \pm 8) \times 10^{-3}$ | $0.031 \pm 0.004$ | $0.23 \pm 0.032$ | $(75 \pm 4) \times 10^{-3}$ |
| Soil 7–17 cm | $6.0 \pm 0.10$ | $3.5 \pm 0.035$ | $(21.7 \pm 2.2) \times 10^{-3}$ | — | $0.34 \pm 0.103$ | — |
| Total deposition | 13.8 | 12.7 | $168 \times 10^{-3}$ | 0.026 | 0.24 | $137 \times 10^{-3}$ |

Samples were collected in May 1990, 94 km from MAYAK.
The error term is 1 S.D. due to counting statistics, but for $^{90}Sr$ it is 1 SE of double determinations.

$= 72$ TBq $^{137}Cs$

The uncertainty of the integral is about a factor of 1.5 i.e. the 1 S.D. range is 50–110 TBq.

The total released activity from the Karachay accident in 1967 was 600 Ci (Academy of Science, 1991). If 75% of this was $^{137}Cs$, i.e. 450 Ci this would now, 30 years later, have decayed to 225 Ci or 8.3 TBq. Our estimate is nearly an order of magnitude higher. A comparison between our data and Russian aerial survey measurements in 1991 (Izrael et al., 1993; Tsaturov and Anisimova, 1993) show reasonable agreement for stations 1 and 2 (southward of MAYAK), but our stations NE of MAYAK show in general levels twice as high as the Russian 1991 aerial data. We thus believe that the $^{137}Cs$ contamination from the Karachay incident may either have been somewhat higher than earlier assumed or there may have been other contamination events in the South Urals than those reported so far. Anyway our soil data indicates higher $^{137}Cs$ levels than in the Russian aerial data.

We estimated above a mean $^{239,240}Pu/^{90}Sr$ ratio in Kysthym debris of $0.0018 \pm 0.0007$. If the estimated $^{90}Sr$ Kyshtym inventory is multiplied with this ratio we get 0.4 TBq $^{239,240}Pu$. Had we instead used the $^{239,240}Pu/^{90}Sr$ ratio of $0.0029 \pm 0.0008$, estimated from location 19, we would have got 0.6 TBq $^{239,240}Pu$ deposited in the 30–300

Table 22
Radionuclides in soil and vegetation samples from location no. 22 (position: 56° 29′N 61° 36′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{239,240}Pu$ (kBq $m^{-2}$) | $^{238}Pu/^{239,240}Pu$ | $^{241}Am/^{239,240}Pu$ |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter (no sample) | | | | | |
| Soil 0–5 cm | $16.1 \pm 0.20$ | $6.1 \pm 0.1$ | $(130 \pm 9) \times 10^{-3}$ | $0.019 \pm 0.005$ | $0.26 \pm 0.029$ |
| Soil 5–10 cm | $15.7 \pm 0.2$ | $1.2 \pm 0.07$ | $(32 \pm 2) \times 10^{-3}$ | — | $0.22 \pm 0.064$ |
| Soil 10–15 cm | $9.0 \pm 0.1$ | $0.30 \pm 0.07$ | $(6 \pm 0.5) \times 10^{-3}$ | — | — |
| Soil 15–20 cm | $7.9 \pm 0.1$ | $0.19 \pm 0.06$ | $(6 \pm 0.6) \times 10^{-3}$ | — | — |
| Soil 20–25 cm | $2.9 \pm 0.05$ | $0.22 \pm 0.08$ | $(4 \pm 0.5) \times 10^{-3}$ | — | — |
| Soil 25–30 cm | $2.3 \pm 0.03$ | — | $(3 \pm 0.4) \times 10^{-3}$ | — | — |
| Soil 30–35 cm | $1.7 \pm 0.03$ | — | $(2 \pm 0.4) \times 10^{-3}$ | — | — |
| Soil 35–40 cm | $1.0 \pm 0.02$ | — | $(2 \pm 0.3) \times 10^{-3}$ | — | — |
| Total deposition | 57 | 8.0 | $185 \times 10^{-3}$ | 0.0135 | 0.22 |

Samples were collected in October 1992, 100 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 23
Radionuclides in soil and vegetation samples from location no. 23 (position: 56° 32′N 61° 46′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{239,240}Pu$ (kBq $m^{-2}$) | $^{238}Pu/^{239,240}Pu$ | $^{241}Am/^{239,240}Pu$ |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter (no sample) | | | | | |
| Soil 0–5 cm | $1.61 \pm 0.03$ | $7.6 \pm 0.2$ | $(65 \pm 5) \times 10^{-3}$ | — | $0.31 \pm 0.025$ |
| Soil 5–10 cm | $0.79 \pm 0.01$ | $1.13 \pm 0.11$ | $(8 \pm 1) \times 10^{-3}$ | — | $0.38 \pm 0.033$ |
| Soil 10–15 cm | $0.42 \pm 0.01$ | $0.36 \pm 0.08$ | $(3 \pm 0.5) \times 10^{-3}$ | — | — |
| Soil 15–20 cm | $0.24 \pm 0.01$ | — | $(2 \pm 0.4) \times 10^{-3}$ | — | $1.50 \pm 0.092$ |
| Soil 20–25 cm | $0.11 \pm 0.01$ | $0.2 \pm 0.08$ | $(2 \pm 0.7) \times 10^{-3}$ | — | — |
| Soil 25–30 cm | — | — | $(2 \pm 0.4) \times 10^{-3}$ | — | — |
| Soil 30–35 cm | $0.04 \pm 0.01$ | — | $(1 \pm 0.3) \times 10^{-3}$ | — | — |
| Soil 35–40 cm | — | — | — | — | — |
| Total deposition | 3.2 | 9.3 | $83 \times 10^{-3}$ | — | 0.31 |

Samples were collected in October 1992, 110 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

km zone. If the total inventory of $^{90}Sr$ from Kyshtym is 1 PBq the total $^{239,240}Pu$ inventory from Kyshtym becomes $2.4 \pm 0.7$ TBq.

Concerning the $^{239,240}Pu$ inventory from the Karachay accident we calculated this by multiplying the equations for the $^{239,240}Pu/^{137}Cs$ ratio and the equation for the deposition density of $^{137}Cs$ for Karachay debris. We got the equation kBq $^{239,240}Pu$ $m^{-2} = 0.91\ e^{-0.039x}$. From this equation we calculated in analogy $y$ with the $^{137}Cs$ inventories the inventory of $^{239,240}Pu$ from Karachay to 0.6 TBq in the 7–150 km zone from MAYAK. If we instead had used the $^{239,240}Pu/^{137}Cs$ found at location 19, i.e. $0.0118 \pm 0.0024$ and multiplied this with the inventory of $^{137}Cs$ from the Karachay incident i.e. 71 TBq we would have got 0.8 TBq $^{239,240}Pu$. As a mean estimate for the Karachay- derived plutonium we may use $0.7 \pm 0.2$ TBq $^{239,240}Pu$. It thus seems that the inventories of $^{239,240}Pu$ from the Kyshtym and Karachay accidents both are in the order of 1 TBq.

At the three locations, which contained the highest contamination levels with Kyshtym debris i.e. numbers 5, 6 and 19 the mean $^{238}Pu/^{239,240}Pu$ ratio in the total deposit was $0.0115 \pm 0.024$ ($\pm 1$ S.D.; $N = 3$) and the $^{241}Am/^{239,240}Pu$ ratio was $0.117 \pm 0.027$ ($\pm 1$ S.D.; $N = 3$). These ratios are about three times lower than those found in global fallout from nuclear weapons testing (UNSCEAR, 1993). They are also low compared with those found in fuel elements in nuclear power reactors. The low ratios suggest 'low burn up' in the nuclear fuel, which is characteristic for nuclear weapons plutonium.

If we in a similar way looked at the three most contaminated locations with Karachay debris, i.e. numbers 2, 3 and 4 we found $^{238}Pu/^{239,240}Pu = 0.088 \pm 0.069$ and $^{241}Am/^{239,240}Pu = 0.169 \pm 0.026$. These ratios are higher than from those in

Table 24
Radionuclides in soil and vegetation samples from Rassoka (location no. 24; position: 56° 48′N 61° 19′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{134}Cs$ (kBq $m^{-2}$) |
|---|---|---|---|
| Grass (no sample) | | | |
| Litter (no sample) | | | |
| Soil 0–5 cm | $1.12 \pm 0.02$ | $4.5 \pm 0.1$ | $0.19 \pm 0.01$ |
| Soil 5–10 cm | $0.30 \pm 0.01$ | $0.28 \pm 0.01$ | — |
| Soil 10–15 cm | $0.03 \pm 0.00$ | $0.05 \pm 0.01$ | — |
| Soil 15–20 cm | $0.03 \pm 0.02$ | $0.03 \pm 0.01$ | — |
| Soil 20–25 cm | $0.03 \pm 0.01$ | $0.06 \pm 0.02$ | — |
| Soil 25–30 cm | $0.06 \pm 0.01$ | $0.04 \pm 0.02$ | — |
| Total deposition | 1.57 | 5.0 | 0.19 |

Samples were collected in June 1991, 126 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

Table 25
Radionuclides in soil and vegetation samples from Miassovo (location no. 25; position: 55° 09′N 60° 21′E)

| Sample | $^{90}Sr$ (kBq $m^{-2}$) | $^{137}Cs$ (kBq $m^{-2}$) | $^{239,240}Pu$ (kBq $m^{-2}$) | $^{238}Pu/^{239,240}Pu$ | $^{241}Am/^{239,240}Pu$ |
|---|---|---|---|---|---|
| Grass (no sample) | | | | | |
| Litter (no sample) | | | | | |
| Soil 0–5 cm | 0.79 ± 0.01 | 2.7 ± 0.1 | $(57 \pm 4) \times 10^{-3}$ | 0.077 ± 0.017 | 0.27 ± 0.038 |
| Soil 5–10 cm | 0.67 ± 0.02 | 1.08 ± 0.03 | $(32 \pm 4) \times 10^{-3}$ | 0.016 ± 0.007 | 0.33 ± 0.075 |
| Soil 10–15 cm | 0.16 ± 0.01 | 0.26 ± 0.02 | $(7 \pm 2) \times 10^{-3}$ | 0.003 ± 0.002 | — |
| Soil 15–20 cm | — | — | — | — | — |
| Soil 20–25 cm | — | — | — | — | — |
| Soil 25–30 cm | — | — | — | — | — |
| Total deposition | 1.62 | 4.0 | $96 \times 10^{-3}$ | 0.051 | 0.271 |

Samples were collected in July 1991, 68 km from MAYAK.
The error term is 1 S.D. due to counting statistics.

the Kyshtym debris and suggest thus a higher burnup for the Pu deposited in Lake Karachay. The reason for the very high $^{238}Pu/^{239,240}Pu$ ratio at location 2 (= 0.166) may be that this location as mentioned above could have received some airborne deposition from the daily operation of MAYAK, which is only 12 km away from this location. A relatively high deposition of $^{134}Cs$ at location 2 (470 Bq $m^{-2}$) may be indicative for such a contamination. It might therefore be more appropriate to look at the ratios from Bashakul only (Tables 3 and 4). The $^{238}Pu/^{239,240}Pu$ mean ratio then becomes $0.050 \pm 0.020$ and $^{241}Am/^{239,240}Pu = 0.16 \pm 0.03$. These ratios still suggest that the plutonium from Lake Karachay may have another isotopic composition than the plutonium from the Kyshtym accident.

## 4. Conclusion

The contaminations from the 1957 Kyshtym accident and the 1967 Lake Karachay incident was studied by a soil sampling programme carried out between 1990 and 1995 in the South Urals. The samples were analysed for $^{90}Sr$, $^{137}Cs$ and transuranic elements. From models assuming an exponential decay of deposited activity with distance from the source, inventories from the two events were estimated. For the Kyshtym accident the $^{90}Sr$ inventory deposited between 30 and 300 km from the source was estimated at 0.1–0.5 PBq, which is compatible with Russian information. In the case of Karachay the $^{137}Cs$ inventory found outside the MAYAK area (7–150 km) was estimated to be higher than that found from Russian measurements. Whether the discrepancy is due to a higher release from Lake Karachay than earlier assumed or to contamination from unreported sources have not been clarified.

The plutonium contaminations from Kyshtym and Karachay were both in the order of 1 TBq $^{239,240}Pu$ or a total of about 1 kg plutonium was deposited from these two accidents.

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