EVALUATION OF THE RADIOACTIVE POLLUTION OF THE TERRITORIES AROUND KRASNOYARSK-26

J Smith-Briggs, D Craig, J Watterson,
AEA Technology

Y. Kuznetsov, Y. Panteleev, V. Legin,
VG Khlopin Radium Institute

A. Schischlov
Mining and Chemical Combine

ABSTRACT

The objective of this study, funded by DGXI (now DGEnv (Directorate General Environment) of the European Commission, was to evaluate the impact of radiological contamination in the environment, which has resulted from the operations at the nuclear establishments at Krasnoyarsk-26, and to determine whether any remedial actions are required. Environmental data has been obtained for a wide range of environmental media and for a number of years covering the period before and after the closure of the single pass reactors at Krasnoyarsk-26. These media include air (aerosols and deposition), water, sediments, soil, floodplain deposits, foodstuffs and vegetation. Information was also obtained on demography, meteorology and the general geological, geographical and hydrological characteristics of the regions. A GIS (Geographical Information System) system was established to contain all of this data and to display a number of themes. The sampling and analytical methodologies were evaluated and the data were examined to determine any trends with time or distance from the contamination sources. The data were compared with other global data available in the open literature. A methodology for the radiological assessment of the contamination was established and the calculations performed. The results of the radiological assessments showed that the most significant exposures resulted from the occupancy and use of the contaminated floodplains and from the potential exposure to hot particles. Doses of several millisieverts a year could result from some of the most contaminated sites and of tens to hundreds of millisieverts from hot particles. Criteria for remedial action and a number of remediation options were evaluated.

INTRODUCTION

Much of the emphasis of recent and current projects in the former Soviet Union has been targeted at addressing the problems resulting from the accident at Chernobyl, spent fuel management in North West Russia and environmental contamination at Chelyabinsk-65 (Mayak, e.g. Karachai Lake). Relatively little is known about the problems of the nuclear facilities in Siberia and, particularly, about their impact on the environment.

Concerns about these Siberian combines have been raised recently [1, 2]. These reports suggested that significant levels of environmental contamination are present around these sites. For example, contamination of the riverbanks of the Yenisei River, downstream from Krasnoyarsk-26, is reported with measurements of ^{137}Cs up to 220,000 Bq/kg. About 2 PBq has been reported to be discharged to the Yenisei River and 17 EBq are reported to have been discharged to deep wells near the site.

The European Commission, DGXI, commissioned this project in 1997 to evaluate the consequences of the radiological contamination that may exist around Krasnoyarsk-26.

Krasnoyarsk-26, now known as Zheleznogorsk, is the site of a nuclear reactor plant and reprocessing facility in Siberia in the Russian Federation. These facilities are known as the Mining Chemical Combine (MCC). Operations began there in late 1958. It is located approximately 50 km north from the city of Krasnoyarsk in Siberia. The reactor plant consists of three industrial graphite reactors for the production of weapon-grade plutonium from irradiated natural uranium. The radiochemical plant at the site produces weapons grade plutonium from processing the reactor fuel.

Two of the reactors were designed to abstract cooling water directly from the Yenisei River and then to discharge it back to the river without any purification. The water discharged into the river contained both activation and fission products. These radionuclides have been adsorbed onto sediments that, in turn were distributed by flood action onto the riverbanks and islands. As a result there are areas of contamination for hundreds of kilometres downstream. The pattern of radioactive contamination of river sediments and the floodplains are complex. The maximum contamination density observed was 200 Ci/km^2 (7.42 MBq/m^2) of Cs-137 on the island of Atamanovo where the exposure rate was 150 μR/hr (13 mSv/year). The two single pass reactors were shut down in 1992.

Environmental contamination has also arisen as a result of the aerial dispersion of radionuclides from surface ponds on the industrial site of the MCC old liquid radioactive waste reservoirs.

The project was restricted to evaluating radioactive contamination outside of the sanitary protection zone (SPZ). Information concerning the level of discharges from the sites to the environment were also outside the scope of the project, with the exception of that already in the public domain.

The project was managed by AEA Technology and included contributions from the VG Khlopin Radium Institute, based in St Petersburg and the Mining Chemical Combine at Krasnoyarsk-26. VNIPIPT, Moscow, also provided comments on the reports generated during the project.

A series of technical reports have been prepared by AEA Technology and the VG Khlopin Radium Institute, which cover the technical work programmes described earlier in detail [3]. It is the intention of this paper to summarise the findings of each of these reports, to bring out their salient points and outstanding issues and to describe the main findings and conclusions from the project.

METHODOLOGY

The methodology for achieving the objectives of the project was developed during an initial series of discussions within project team. These discussions established the scope of the project, what information could be made available and the responsibilities for obtaining the information and for the subsequent evaluation and assessments of its implications.

It was agreed that information concerning radionuclide concentrations in environmental media outside of the SPZ for Krasnoyarsk-26 would be made available for a number of years of measurements, including the periods before and after the single pass reactors ceased operation, where possible. However, it was not possible for information concerning the SPZ, and/or discharges from the sites, to be made available.

In order to specify the exact requirements for the radionuclide information a questionnaire was prepared to guide the dissemination of this information and to allow for it to be recorded electronically and facilitate the production of a GIS. The purpose of the GIS was to provide a structured system for the recording of the environmental radioactivity data, and other supporting data, and to allow this information to be represented spatially on maps of the area. The GIS also facilitated interpolation between data points and correlations between different sets of data.

When the datasets were complete for each site they were evaluated against the following criteria:

- sampling methodology
- analytical methodology
- comparison with global data for
    - sites near nuclear establishments
    - non-industrial locations
- comparison with other reported data for the area

With the validity and representativeness of the data established, the potential radiological impact was assessed. The radiological assessment considered the intake of radionuclides from inhalation of air (aerosols and particulate material), ingestion of foodstuffs, water and soils/sediments (inadvertent), and external exposures from γ-emitting radionuclides. The preliminary calculations considered a pessimistic scenario whereby the highest observed levels for each environmental medium were used together with very conservative generic assumptions for dietary and occupancy habits. This simple approach allowed the screening out of unimportant pathways for exposure.

Finally, potential remediation options to reduce or remove the exposure pathways were evaluated and recommendations for further work to improve the understanding of potential future exposures from each site were made.

ENVIRONMENTAL RADIOACTIVITY NEAR KRASNOYARSK-26

In this section the most relevant environmental data obtained for Krasnoyarsk-26 is summarised. All of the original datasets, and their detailed evaluation, are presented in [3].

Information in the following areas was obtained for Krasnoyarsk-26:

- Atmospheric radionuclide concentrations
- Background concentrations in all media
- Demography
- Deposition of radionuclides (bulk)
- Deposition of radionuclides to soil
- Diet
- External dose
- Radionuclide content in fish
- Radionuclide content in food stuffs
- Frequency and radionuclide content in hot particles
- Lifestyle
- Meteorology
- River chemistry & flow
- Radionuclide content in sediment
- Radionuclide content in soil
- Soil type
- Radionuclide content in vegetation
- Radionuclide content in water
- Plutonium data

This data were provided by the MCC and the VG Khlopin Radium Institute and collated by the VG Khlopin Radium Institute. The radionuclide concentrations in environmental media are reported from the late 1980's up to 1997 in most cases. Most of the data are also reported as monthly and/or annual averages.

Atmospheric radionuclide concentrations

Measurements of radionuclide concentrations in air are made at three sites, two of which are downwind and one is upwind of the MCC. A range of radionuclides was measured, including Be-7, Co-60, Sr-90, Ru-106, Cs-137 and levels of gross alpha and beta activity. There is little evidence for a significant influence of plant plume on air concentrations at the measurement sites since concentrations at the three measurement sites all quite similar (upwind and down wind).

Deposition of radionuclides (bulk)

Measurements are of bulk deposition (wet and dry) at several sites around the MCC. Two of these sites are approximately 70 km to the west of the MCC, and represent sites that receive background levels of deposition predominantly from global fallout (plus contributions from locally resuspended soil). Annual mean deposition fluxes of Cs-137 are small, and up to 1995, appear to have been declining.

External dose

Annually averaged external doses were measured at a number of locations within 20 km of the MCC in 1992 and 1994. There are also data for gamma dose rates in air at the locations where soil and sediment samples were taken. The annually averaged external doses, other than those from the contaminated floodplains and islands, range from between 5 and 10 μR/h, which is well within the normal range expected for natural background radiation. The dose rates measured at the locations where contaminated floodplain soils were samples were considerably higher than this average value. Up to 150 μR/h has been reported for some locations.

Radionuclide content in fish

Concentrations of γ-emitting radionuclides are reported for a number of sites upstream of the release point from the MCC and up to 37 km downstream. Extensive data are available in terms of sample frequency, sample locations and species type.

The short lived isotopes P-32, Na-24 and S-35 were responsible for the majority of the dose to the local population from eating fish during the period when the single-pass reactors were operating. Concentrations of Cs-137 in fish downstream of the release point do not exceed about 20 Bq kg^{-1}.

Radionuclide content in foodstuffs

Information on radionuclide levels was reported for a number of foodstuffs sampled between 1990 to 1997. The foodstuffs included milk, beef, potato, cabbage and carrot. The locations ranged from within a few kilometres of the site to up to 250 kms downstream. Average milk concentrations for the region as a whole are reported as being 0.79Bq/kg for Cs-137.

Frequency and radionuclide content in hot particles

A number of hot particles have been detected in several areas downstream of the MCC at Krasnoyarsk-26. The areas where particles have been found have been described together with the analysis of the radionuclide activities of the particles and of the floodplain deposits where they were located. The source of the hot particles is likely to be from spent fuel fragments from the single pass reactors at Krasnoyarsk-26. There has however been little analysis of the particles structures, one measurement suggested a graphite-like matrix. The lack of information on plutonium and uranium isotope ratios makes it difficult to be certain about the particles origins. There is evidence that the

particles found are of different ages implying that the release of the hot particles has taken place over a number of years. There is a wide range in the specific activities of the particles (from 10^4 to 10^7 Bq Cs-137) implying that the particles may come from different sources and originated at different times.

Radionuclide content in sediment

A substantial dataset was provided about radionuclide levels in the sediments of the Yenisei River. Most measurements were made within 300 km of the discharge point. Upstream background concentrations of Cs-137 are three orders of magnitude lower than the peak concentrations in the sediments down stream of the reactor discharge; however, a few down stream concentrations are comparable to background levels. Concentrations of up to 1600 Bq/kg of Cs-137 have been observed up to 650 kms from the discharge point.

Radionuclide content in soil

Essentially, there are two sets of measurement sites: ones which are associated with the floodplains and islands of the Yenisei river, 'down river' from the MCC, and others not adjacent to the river, 'non-river' sites located ten's of kilometres from the MCC. Data are reported from 1992 to 1997. The measurements made in floodland deposits are referred to here as 'soils' but these are in fact hybrid solids, which are neither true soils nor sediments and have quite distinctive properties.

The 'non-river' sites have background concentrations of Cs-137 are about 1 kBq m^{-2} which are smaller than those at many European sites, even making allowances for the small amounts of radioactive decay between the sampling dates.

The data for 'down-river' sites show that there are elevated concentrations of radionuclides at several locations downstream of the MCC (Fig.1 and Fig. 2). The concentrations do not fall predictably with downriver distance from the discharge point; for example the concentrations of Cs-137 in soil are not highest close to the discharge point. The data show that concentrations of Cs-137 in soil are elevated at up to 1000 km down river of the discharge point. The vertical profiles of Cs-137 concentrations indicate that the highest concentrations are not always within the rooting zone and the profiles of concentration with depth are not consistent between different sites.

There is considerable spatial variability in the reported measurements. At one site for Cs-137 (176 km downstream of the discharge point), variability of about 2 orders of magnitude is apparent for samples collected at the same time within a few 10's of metres. Such large variability over a small scale could be explained by inhomogeneous contamination from flooding events or perhaps by the presence of 'hot' particles.

The radionuclides present in floodplain soils, and river sediments, are a potential source of future contamination of other areas in the Yenisei/Kara Sea system through re-mobilisation and dispersion of the radionuclides themselves or re-suspension and deposition of the contaminated soils/sediments.

Plutonium data

During the summer of 1997, soil was sampled at locations very close to the perimeter of MCC and at two locations downwind which might have received deposition, and at a further two locations which are subject to episodic flooding from the Yenisei. Levels of Pu-239+240 were determined in the soil samples, together with some other radionuclides (Cs-137, Sr-90 and Co-60). The concentrations of Cs-137 and Pu at two locations near the complex (storage accumulator of liquid wastes and storage area for non-technological wastes) were elevated over background levels. Measurements a few hundred metres downwind show concentrations in soil close to background. This suggests the transfer mechanism from the liquid waste storage areas is aerosol transfer of material from the liquid surface. Concentrations of Pu do not seem particularly high at the downriver sites, although Cs-137, Sr-90 and Co-60 are elevated over background levels. The Pu concentrations very close to the perimeter of the SPZ of the MCC are approximately an order of magnitude greater than concentrations in rural UK and similar to those at a village near the Sellafield site.

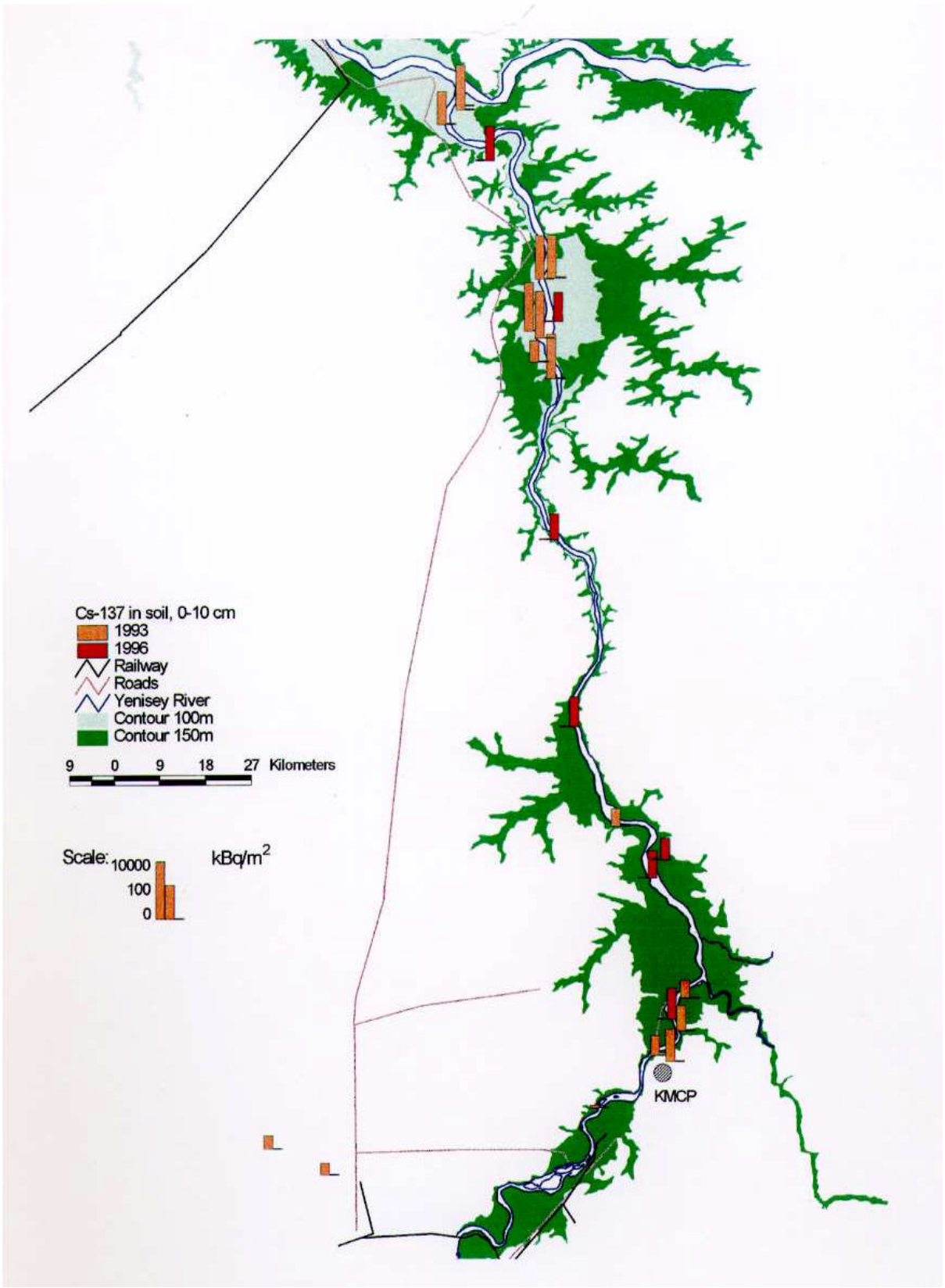

Fig. 1 Cs-137 distribution in floodplain soils near the River Yenisei.

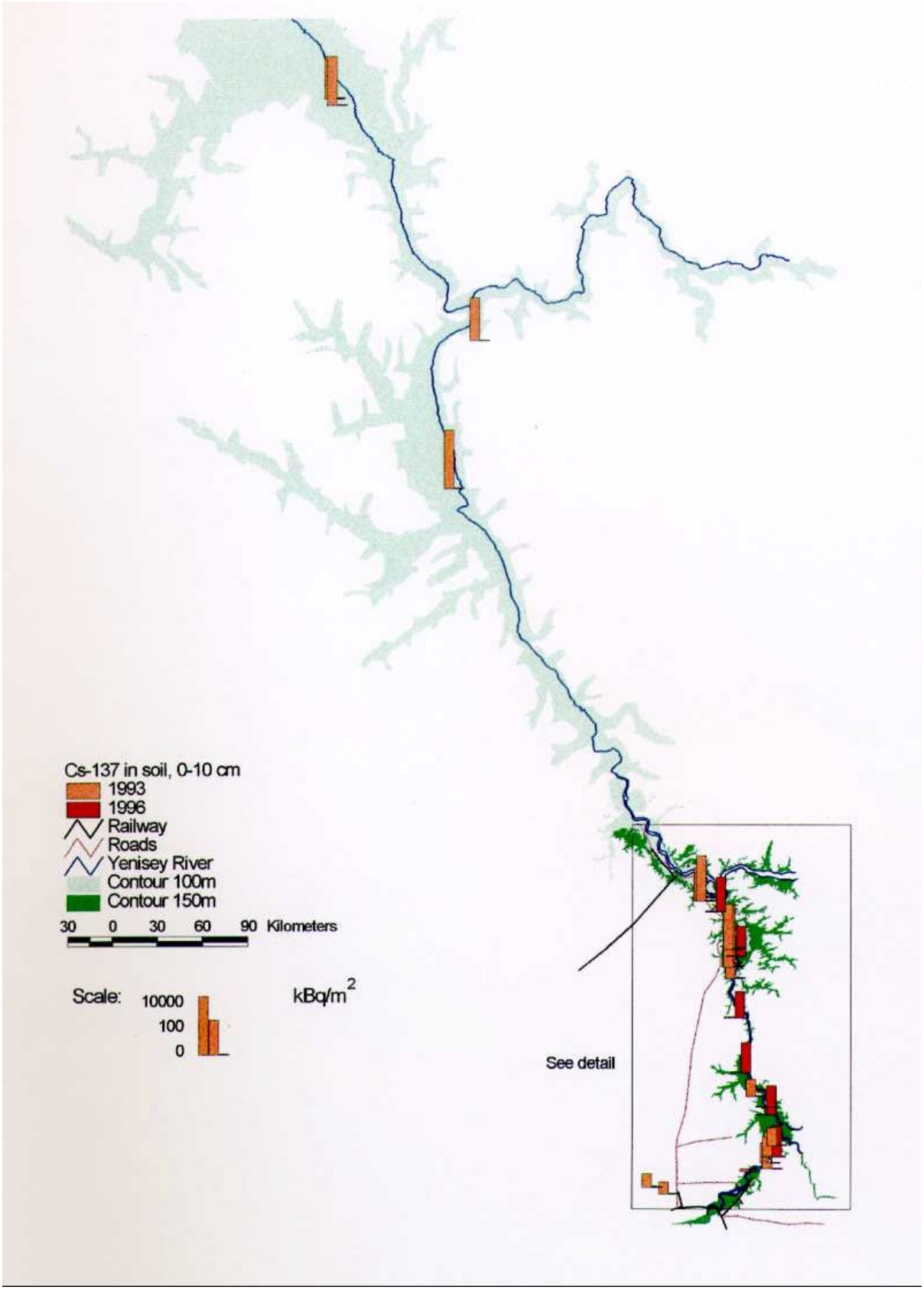

Fig. 2 Cs-137 distribution in floodplain soils near Krasnoyarsk-26

RADIOLOGICAL ASSESSMENT OF ENVIRONMENTAL CONTAMINATION

The radiological assessment of the contaminated territories around Krasnoyarsk-26 has been based on the following pathways:

- the ingestion of radionuclides via contaminated foodstuffs and drinking water
- the ingestion of radionuclides via contaminated soils and sediments
- the inhalation of aerosols
- the inhalation of particulate material
- occupancy of contaminated land (via external exposure)

The radiological parameters used in the dose assessment, that is doses per unit intake (Sv per Bq) are based on internationally accepted criteria [4]. The rates for inhalation and ingestion (mass/volume per unit time) are arbitrarily based on standard reference parameters. External exposure models are based on simple models of exposure to infinite plane sources. Occupancy and habit data were based on locally observed patterns [3]. Calculations were carried out for the highest values observed for each sample type. This allowed the most important pathways to be identified.

Table I: Summary of doses from environmental radioactivity at Krasnoyarsk-26

Pathway	**Maximum Annual Individual dose, Sv/y**		
	Adult	**Child**	**Infant**
Inhalation of air	1.66E-08	1.59E-08	1.34E-08
Inhalation of soil	5.62E-05	1.98E-05	5.96E-06
Inhalation of soil (2)	9.15E-07	7.28E-07	4.69E-07
Inhalation of sediment (1996)	1.17E-06	1.04E-06	6.83E-07
Ingestion of foodstuffs	5.03E-05	5.49E-05	1.20E-04
Ingestion of fish	5.73E-05	1.14E-05	2.85E-06
Ingestion of water	3.12E-06	2.41-06	3.67E-06
Ingestion of soil (1)	1.30E-05	5.88E-06	3.44E-05
Ingestion of soil (2)	4.03E-06	1.03E-06	4.8E-06
Ingestion of hot particles, mean (3) Ingestion of hot particles, max (4)	2.32E-03 3.84E-02 3.77E-01	1.80E-03 2.96E-02 2.90E-01	2.20E-03 3.55E-02 3.48E-01
Ingestion of sediments (1996)	4.17E-06	1.27E-06	6.81E-06
External irradiation	5.34E-03	5.34E-03	5.34E-03

(1) from 'down-river' soils
(2) from 'non-river' soils
(3) mean values from 13 particles

(4) max values are derived from the highest activity observed in a particle
(5) detailed assessments of the potential exposures to hot particles are given in [3]

The floodplains and hot particles therefore represent the most significant exposure pathways for the radiation exposure of the population in the region near, or downstream from, Krasnoyarsk-26. A further potential source of exposures, particularly to infants, could arise from the ingestion of contaminated milk. There is evidence at one site, on the contaminated floodplain that levels of Cs-137 contamination in milk is relatively high. Other sites on the contaminated floodplain did not show elevated levels in locally produced milk. The highest values were used in the above calculations and these are therefore conservative. The population also have a wide variety of food sources and are not dependent solely on locally produced food.

A key feature of the contaminated floodplain is the complex nature of the distribution of the radioactivity. There is no simple correlation with distance downstream and samples taken at apparently adjacent sites, at the same time, can have very different activities.

The maximum doses assessed here, associated with the use and occupancy of the contaminated floodplains and islands of the Yenisei, are higher than the Russian standard for the annual dose limit for member of public (1 mSv/y) by a factor of 2 or 3. The exposures associated with the contaminated floodplain would therefore be classed as requiring further study, and possibly intervention, under the Russian regulatory system.

REMEDIATION OPTIONS

In 1999, Russia adopted new standards of radiation safety, NRB-99, corresponding to international basic safety standards. These standards recommend that the criterion for intervention following radioactive pollution incidents should be where individual doses to the population are more than 0.3mSv per year, based on a collective dose from 70 years occupancy. Based on these criteria the following areas would require remediation.

Areas of contaminated floodplain and riverbank

There is insufficient data to confirm the extent of contamination along the riverbanks. Samples in which contamination has been found have been taken from between 5m and 50m from the edge of the river.

The most recent data from Krasnoyarsk-26, based on a communication received in June 1998, is that for assessment purposes, the following should be assumed:

- only one bank of the river should be assumed to be contaminated ;
- the width of contamination should be assumed to be 20m ;
- the length of this contaminated strip should be assumed to be 1000km ;
- this gives a total area of contamination of 20 km^2 ;

- the gamma dose rate should be assumed to be 30-40 μR/hour (2.6-3.5 mSv/year) ;
- this is equivalent to 50-200 kBq/m^2 for Cs-137.

If the average contamination depth is assumed to be 0.3m, then the volume of contaminated material would amount to 6 million (6 E+06) m^3.

Gorodskoy Island

The island is located in the Yenisei River, approximately 410 km downstream from Krasnoyarsk City at Eniseisk. Average levels of pollution at 1m depth are 1050kBq/m^2 for Cs-137, 3.3kBq/m^2 for Co-60 and 12.5Bq/m^2 for Eu-152. The gamma background dose is 20-60 μR/hour (1.8-5.3 mSv/year). The total area of pollution on the island has been estimated by the Russian partners to be 10000m^2. A vertical profile of contamination down through soil on the island showed contamination present to at least 0.75m depth. This gives an estimated volume of contaminated material to be at least 7500 m^3.

Atamanov Island

Atamanov Island is located about 5km downstream of the MCC nuclear plant. If the nature and depth of contamination are assumed to be similar to that on Gorodskoy Island, and the size of Atamanov Island is estimated to be approximately one third of Gorodskoy Island, then the estimated volume of contaminated material is at least 2500 m^3.

Remediation Options

The overall options for management of the contamination are relatively simple, and include:

- Option 1: excavation of all contaminated material and its removal to approved waste treatment, storage and eventual disposal facilities;
- Option 2: in-situ confinement of the contamination in its present location, through emplacement of engineered barriers, in order to prevent further spread of contamination and to minimise doses to critical groups;
- Option 3: minimisation of dose/risk to the population through "agricultural countermeasures", similar to those employed in areas contaminated from the Chernobyl accident;
- Option 4: removal of the population until risks to them are within acceptable limits
- Option 5: "do nothing".

The conclusions of the project team were that a combination of Options 1 and 5 was likely to be the most pragmatic solution, that is complete removal of contaminated material where demonstrated to be necessary, otherwise, no action needs to be taken.

RECOMMENDATIONS

A number of key recommendations have been made in order to understand the implications of the contaminated floodplains and islands of the Yenisei River more precisely. The recommendations include the following:

1. Define spatial extent of floodplain contamination in a robust and systematic way
   - Aerial gamma survey (Cs-137)
   - Ground gamma survey
   - Examination of hydrological features of the river
   - Evaluation of flooding scenarios and their impact
   - Evaluation of alpha-contamination
2. Identify usage of floodplain site in detail
   - Land use (pasture, crop production, recreation etc)
   - River use (fishing, swimming, etc.)
   - Occupancy times
   - Demographic data
3. Evaluate contamination of food produced on or near contaminated floodplain sites
   - Sampling and analysis programme for foods produced on, or near, contaminated floodplains
   - Modelling of food-chain transfer for representative scenarios
4. Evaluate potential impact from hot particles
   - Small programme currently supported by DGXI
     - Distribution of hot particles
     - Analysis of hot particles
     - Sources of hot particles
     - Critical groups
     - Risk assessment
5. Dose measurements for critical groups
     - Whole body and TLD measurement programmes
     - Dose assessments for representative scenarios, including the impact of potential future flood events
6. Evaluation of key source term data
     - Understanding radwaste disposed of and stored at Krasnoyarsk-26, future operations and the final site closure strategies
     - Understanding (or development) of a strategy for each site for future radwaste management, in order to minimise future releases to the biosphere
7. Detailed evaluation of remediation strategies
     - Cost per averted man-sievert
     - Regulatory issues
     - Pragmatic solutions which can be achieved easily, mainly with existing equipment, and at little cost

8. Evaluate potential for re-contamination of remediated sites
    - Evaluate the potential for re-contamination to occur should areas be remediated
    - Identify patterns of sediment movement and deposition

CONCLUSIONS

The results of the radiological assessments show that the only significant exposures result from the occupancy and use of the contaminated floodplains, particularly if hot particles are present. Doses of several millisieverts a year could result from some of the most contaminated sites. The levels of floodplain contamination are very variable; samples taken at the same approximate location can be very substantially different. There is also no simple correlation with contamination levels and distance from the discharge.

REFERENCES

[1] . D.J. BRADLEY. Behind The Nuclear Curtain: Radioactive Waste Management In The Former Soviet Union. 1997 Battelle Press

[2] N BOHMER, T NILSEN. Reprocessing Plants In Siberia, Bellona Working Paper No. 4, 1995.

[3] J L SMITH-BRIGGS. Evaluation Of The Radioactive Pollution Of The Territories Around Krasnoyarsk-26 And Tomsk-7, Overview Report, Compiled by J L Smith-Briggs, to be published by the European Commission, 2000.

[4] ICRP PUBLICATION 72, *Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 5 - Compilation of Ingestion and Inhalation Dose Coefficients*, Annals of the ICRP, **26** (1), 1996.