Evaluation of *In Utero* Doses from Maternal Ingestion of Strontium Radioisotopes at the Techa River

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Abstract

A cohort of persons exposed *in utero* due to radioactive contamination of the Techa River (Southern Urals, Russia) in the early 1950s represents a unique opportunity for studying radiation risks from chronic exposures in early life. Bone-seeking strontium radioisotopes were the main contributors to haemopoietic tissue doses received *in utero* from maternal ingestion of radionuclides before and during pregnancy. Collaborative studies have been conducted between the Urals Research Center for Radiation Medicine, UK Health Protection Agency and University of Florida to provide the best possible evaluations of the doses from strontium radioisotopes incorporated during foetal development to support companion epidemiological studies of the Techa River population. This paper provides insights to the dose reconstruction efforts and presents the first estimates of doses of *in utero* exposure to strontium radionuclides for children born in Muslyumovo located on the Techa River.

Key words: Strontium radioisotopes, *in utero* exposure, Techa River

1. Introduction

A large cohort of individuals living in villages along the Techa River (Southern Urals, Russia) was exposed in the early 1950s due to releases of significant amounts of liquid radioactive wastes (LRW) from the Mayak Production Association (MPA), which was the first site in Russia to produce weapon-graded plutonium (Degteva *et al* 2012). The major intakes of radionuclides by inhabitants of the area occurred in 1950–1951 through drinking contaminated river water. Strontium radioisotopes (^{89}Sr and ^{90}Sr) contained in the releases were the main contributors to the internal doses in the exposed population. Strontium behaves in the human body similarly to calcium; it accumulates in the maternal skeleton, easily transfers through the placenta and incorporates in the skeleton of the growing foetus. Thus, ingestion of strontium radioisotopes by the mother consequently leads to exposure of a child's bone marrow from the earliest stages of life. A study of persons who were exposed *in utero* due to maternal residency in the Techa riverside villages provides a unique opportunity to investigate risks of health effects resulting from protracted exposures during gestation and in early life. For this purpose, a cohort of persons born to mothers of the Techa River population, identified as the Techa River in-utero exposed Cohort (TRC-IU), has been formed for studies of exposure to ionising radiation in early life and the risk of cancer (Ostroumova and Akleyev 2004).

Assessment of doses from ingested ^{90}Sr by members of the Techa River exposed population is largely based on a unique database of ^{90}Sr measurements in humans established at the Urals Research Center for Radiation Medicine (URCRM). The database includes about 33,000 measurements made with a tooth-beta counter, about 10,000 measurements of bones collected *post mortem*, and about 38,000 measurements made with a special whole body counter (WBC) SICH-9.1 that allows *in vivo* measurements of ^{90}Sr in the whole body (Degteva *et al* 2006). Unique measurements of ^{90}Sr in the maternal skeleton and in the skeleton of their newborns allowed, for the first time, the evaluation of *in utero* doses for the Techa population using an empirical approach (Tolstykh *et al* 2001). Many efforts have been made over the past ten years that allow significant improvement in the estimates of doses resulting from maternal ingestion of strontium radionuclides in the Techa River villages.

First of all, maternal ^{90}Sr-dietary intake has been re-evaluated from measurements of the radionuclide in human teeth and body and provides more accurate evaluations of the time dependence of ^{90}Sr-dietary intake in 1950-1954 (Tolstykh *et al* 2011). Secondly, a detailed examination of archival information on the LRW releases in 1949-1951 has allowed a refined evaluation of the time dependence and amounts of strontium radioisotopes in the discharges (Degteva *et al* 2012). This has led to more accurate modelling of the transfer of radionuclides along the Techa River that allows assessment of maternal ^{89}Sr-dietary intake (Shagina *et al* 2012).

Collaborative studies have been initiated between the URCRM and the UK Health Protection Agency (HPA) to provide models to make best estimate evaluations of doses resulting from maternal ingestion of strontium isotopes at the Techa River using approaches presented in Publication 88 of the International Commission on Radiological Protection (ICRP) (ICRP 2001). In these studies the ICRP biokinetic model for strontium transfer from mother to the foetus has been specifically adapted for the Techa River population to quantify ^{89}Sr and ^{90}Sr accumulation in foetal tissues during gestation. Preliminary estimates of doses in the red bone marrow (RBM) resulting from maternal ingestion of ^{90}Sr obtained on the basis of this population-specific biokinetic model have shown some reduction in the doses in comparison with ICRP-88 estimates (Shagina *et al* 2007). However, a dosimetric model provided in ICRP Publication 88 is considered to be conservative for use in epidemiological studies of the Techa River in-utero exposed cohort and requires further improvement to take into account the anatomical description of a developing skeleton and features of haemopoiesis during *in utero* life (Shagina *et al* 2007).

Finally, a series of phantoms has recently been developed at the University of Florida (UF) under the framework of collaborative URCRM-HPA-UF studies that provides a sufficiently realistic description of the foetal skeleton at different gestational ages (Maynard *et al* 2011). Application of these phantoms for radiation transport simulation now allows a more reliable evaluation of *in utero* doses from incorporated strontium isotopes.

This paper summarises the progress made in recent years for more accurate evaluation of the doses of *in utero* exposure as a result of maternal ingestion of strontium radioisotopes at the Techa River and provides the first dose estimates obtained with the biokinetic and dosimetric models improved in the collaborative studies for children born in Muslyumovo on the Techa River.

2. Data and models for assessment of in-utero doses from maternal ingestion of strontium radioisotopes at the Techa River

The first improved estimates of the *in-utero* doses were made for TRC-IU members born from mothers who were residents of Muslyumovo located in the middle-Techa region at a distance of 78 km from the site of releases. Of the largest (population of about 3,500 persons in 1950-1951) settlements on the Techa River, this settlement is located closest to the site of releases. Extensive dosimetric investigations have been performed in this settlement and most of its residents have been investigated for their ^{90}Sr-body burden. The doses were evaluated in this study for infants born in Muslyumovo during the first period after the beginning of LRW releases into the Techa River, i.e. for infants born in 1950-1952.

2.1. Maternal intake of strontium radioisotopes

The details for reconstruction of dietary intake of ^{90}Sr for Muslyumovo residents of different ages are provided by Tolstykh *et al* (2011). The ^{90}Sr-intake function was reconstructed from measurements performed on Muslyumovo residents at later periods after the beginning of contamination, including WBC data on ^{90}Sr-body burdens and data on beta-activity of front teeth obtained with a tooth-beta counter. As a result the reconstructed ^{90}Sr intake was solely based on human data and was independent of the information on the amounts of ^{90}Sr discharges into the Techa River (Tolstykh *et al* 2011).

Reconstruction of ^{89}Sr-dietary intake could not be based on human data due to its relatively short decay half-life ($T_{1/2}$=50.5 days). For this reason, an approach was developed for reconstruction of intakes of short-lived radionuclides by the Techa River residents on the basis of the following assumptions: Because most of the ingestion of radionuclides occurred with the consumption of river water in 1950–1952, the intake of ^{89}Sr was derived from estimates of age-dependent intakes of ^{90}Sr scaled in terms of radionuclide composition of the river water (Degteva *et al* 2006). The Techa River Model developed by Shagina *et al* (2012) was used in this study to evaluate ratios of ^{89}Sr concentrations to ^{90}Sr in the river water as a function of calendar year and distance downstream from the site of release for period of 1950-1951. For 1952 the ratio of ^{89}Sr-to-^{90}Sr concentrations in the Techa River water was taken from Degteva *et al* (2000).

The diet of pregnant women differs from the normal adult diet in a number of ways. Nutritional requirements during pregnancy increase to support foetal growth and development as well as pregnancy-related changes in maternal metabolism and tissue development. Water requirements also increase. An increase by 5% in ^{89}Sr and ^{90}Sr dietary intakes during pregnancy compared to non-pregnant woman was assumed for the Techa River residents exposed in 1950-1952 according to Tolstykh *et al* (2008).

Table 1 shows estimated average ^{90}Sr and ^{89}Sr intakes due to the Techa River contamination for adult women and pregnant women resident in Muslyumovo in 1950-1952. The estimates of ^{90}Sr-dietary intake were evaluated for 6 month intervals since the beginning of the intake in September 1950. The intakes of ^{89}Sr are provided in table 1 for the corresponding time intervals. The contribution of ^{89}Sr to the total ingestion of strontium isotopes from drinking river water amounted to 70-80% in 1950-1951. The peak intakes of the radionuclides observed during the first year since the beginning of massive releases into the Techa River in September 1950 (table 1).

Table 1 Average dietary intakes of ^{90}Sr and ^{89}Sr for pregnant women evaluated in this study on the basis of the average ^{90}Sr intakes for adult Muslyumovo residents (Tolstykh et al 2011) and a model for radionuclide transfer along the Techa River (Shagina et al 2012)

Period of intake	Total intake (kBq)			
	Adults		Pregnant women	
	^{90}Sr	^{89}Sr	^{90}Sr	^{89}Sr
Sep 1950 – Feb 1951	870	1,700	910	1,790
Mar 1951 – Aug 1951	740	3,200	780	3,360
Sep 1951 – Feb 1952	550	1,850	580	1,950
Mar 1952 – Aug 1952	390	1.8	410	1.9
Sep 1952 – Feb 1953	275	0.9	290	0.9

2.2. Population-specific biokinetic model for strontium transfer to the foetus

A general approach to the modelling of strontium transfer to the foetus as a result of intake by the mother adapted for the Techa River population was described by Shagina *et al* (2007). This approach makes use of the foetal model from ICRP Publication 88 (2001) developed by Fell *et al* (2001). The model is based on a model for a woman of reproductive age from the Techa River population (Shagina *et al* 2003a) with adjustment made for the period of pregnancy and inclusion of the foetal model. The maternal and foetal models were adopted specifically for the studied population on the basis of measurements of calcium, strontium and ^{90}Sr-body burden in maternal and foetal skeletons obtained for the Russian and Techa River populations (Shagina *et al* 2007). The model for strontium transfer to the foetus was successfully validated using independent data on the content of ^{90}Sr and stable Sr in foetal and maternal bone samples (Shagina *et al* 2007) and allows reliable quantification of retention of strontium radioisotopes during in-utero development as a result of maternal intakes at the Techa River. Figure 1 shows examples on the total amounts of incorporated ^{89,90}Sr in the foetal skeleton at term for newborns born in December of each year in 1950-1952 evaluated with the population-specific biokinetic models. It should be noted, that the amount of ^{89,90}Sr retained in the foetal soft tissues is two to three orders of magnitude lower than in the skeleton.

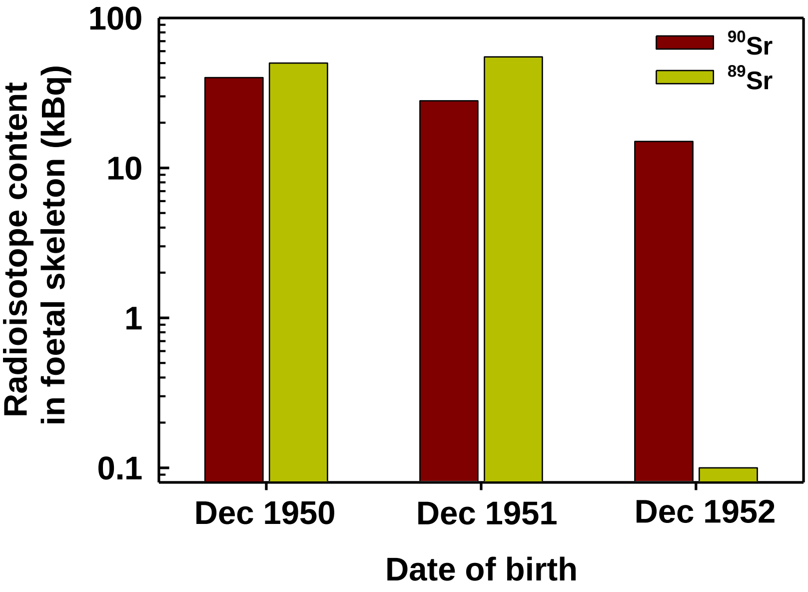

Figure 1. Total amount of ^{89,90}Sr incorporated in the foetal skeleton at term for newborns born in December of each calendar year from 1950 to 1952 in Muslyumovo on the Techa River

It can be seen from figure 1 that due to shorter decay half-life of ^{89}Sr the difference in the amounts of ^{89}Sr and ^{90}Sr in the skeleton of children born in December 1950 and December 1951 is not large despite the more significant ingestion of ^{89}Sr with maternal diet. The levels of ^{90}Sr transferred to the foetus gradually decrease due to a decrease in maternal dietary intakes (table 1).

2.3. Dosimetric model for strontium radioisotopes in the developing foetus

Two foetal hybrid computational phantoms were constructed using high-resolution magnetic resonance imaging and computed tomography image sets obtained for two well-preserved foetal specimens aged 11.5 and 21 weeks post-conception (Maynard *et al* 2011). These phantoms along with a modified version of the 38 week UF hybrid newborn phantom (Pafundi *et al* 2009) were used as a basis to construct a series of hybrid computational phantoms for foetal ages 8, 10, 15, 20, 25, 30, 35, and 38 weeks post-conception. The methodology used to construct the series of phantoms is described in detail by Maynard *et al* (2011). This methodology accounts for age-dependent variations in foetal skeletal size and proportion, levels of bone growth, individual organ and total foetal masses as well as statistical percentile variations of these parameters at each gestation age from 15 weeks post-conception.

The developed phantoms allow detailed description of all foetal organs and tissues; however, in regard to exposure from ^{89,90}Sr the most affected organ is the bone marrow due to their predominant incorporation in the bones of the foetal skeleton. The bone marrow originates as early as 11-12th weeks of gestation (Enzan *et al* 1983, Ogata and Uhthoff 1990) and gradually acquires its haemopoietic function (Tavian and Peault 2005). For this reason, the bone marrow is considered in this study as a target organ during the second and the third trimesters of foetal development.

The series of computational phantoms, starting from gestational age of 11.5 weeks, was used to perform the Monte Carlo radiation transport using radiation transport code MCNPX v2.7. The beta particle spectra of ^{89}Sr, ^{90}Sr and ^{90}Y were directly sampled during transport simulation using the differential probability distributions reported in ICRP Publication 107 (IRCP 2008). All ossified portions of the skeleton were combined into a single source for radiation transport. The ossified bone was treated as a homogenous mass of bone and marrow with a uniform distribution of radionuclides and sensitive cells. Radionuclide S values, defined as the mean absorbed dose to target tissue per nuclear transformation in source tissue, were calculated for each foetal age. Figure 2 provides S values for beta-particle sources within all ossified portions of the foetal skeleton irradiating the sensitive cells. These values are considered as surrogates for absorbed doses in the foetal bone marrow per nuclear transformation.

It can be seen from figure 2 that for all radionuclides, the radiation dose per nuclear transformation decreases with increasing foetal age, a consequence of the increasing mass of the target offsetting any additional increase in energy deposition; and as expected, ^{90}Y (a daughter of ^{90}Sr) consistently contributes the highest target dose per nuclear transformation, followed by ^{89}Sr, then ^{90}Sr, a consequence of the decreasing average beta electron energies (0.93 MeV, 0.58 MeV, and 0.20 MeV, respectively).

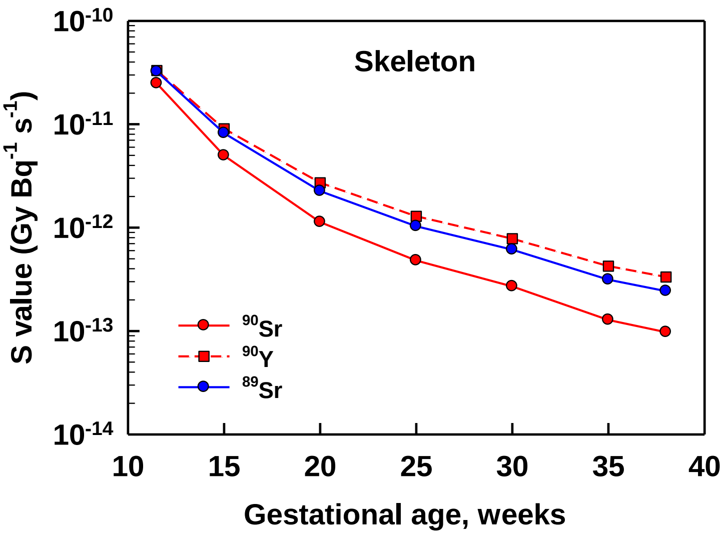

Figure 2 Age-dependent S values of irradiation of ossified portions of the skeleton obtained with the UF series of foetal phantoms considered as a surrogate for bone marrow irradiation from radionuclides incorporated in the bone

3. Results of evaluation of the doses of in-utero exposure due to maternal intake of ^{89,90}Sr in Muslyumovo

Doses were evaluated in this study for infants born during the first period after the beginning of LRW releases into the Techa River, i.e. for infants born in 1950-1952. For dose calculations, infants born at the end of each calendar year, i.e. in Dec 1950, Dec 1951 and Dec 1952, were considered. In all cases the maternal exposure (intake of strontium radioisotopes) began in Sep 1950. For dose calculations, the ^{89,90}Sr dietary intakes and biokinetic models for adult and pregnant women were used in corresponding periods. Doses were calculated to the foetal bone marrow for period of gestation from 12 to 38 weeks.

Figure 3 shows the results of dose calculations with the biokinetic models for strontium adapted for the Techa River population and improved UF dosimetric models for children born in Muslyumovo in the three calendar periods from maternal ingestion of ^{89}Sr and ^{90}Sr.

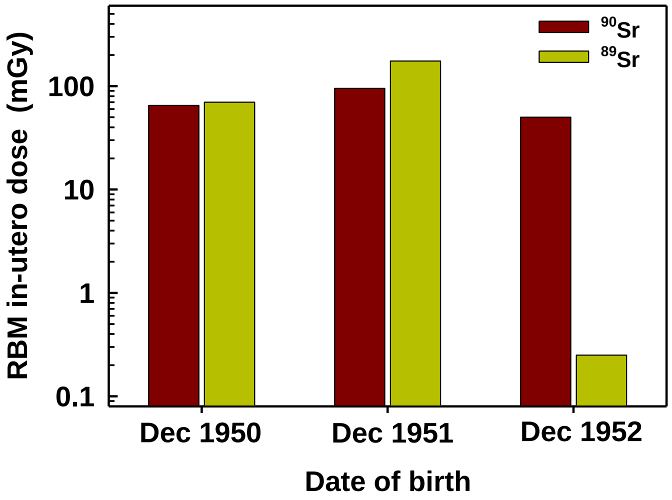

Figure 3 Doses of in-utero RBM exposure of children born in Muslyumovo at different calendar periods. The doses from ^{89}Sr and ^{90}Sr maternal intakes have been calculated with biokinetic and dosimetric models improved for the Techa River studies

It can be seen from figure 3 that doses from ingested ^{89}Sr were predominant in 1951 when the releases of the radionuclide substantially increased. Since 1952 and onwards the internal in-utero doses are determined by ^{90}Sr ingested with maternal diet and incorporated in the maternal skeleton.

Improved estimates of the doses of the foetal bone marrow exposure obtained in this study have been compared with the doses estimated using ICRP-88 methodology (ICRP 2001) recommended for radiation protection purposes. Figure 4 shows the results of these comparisons.

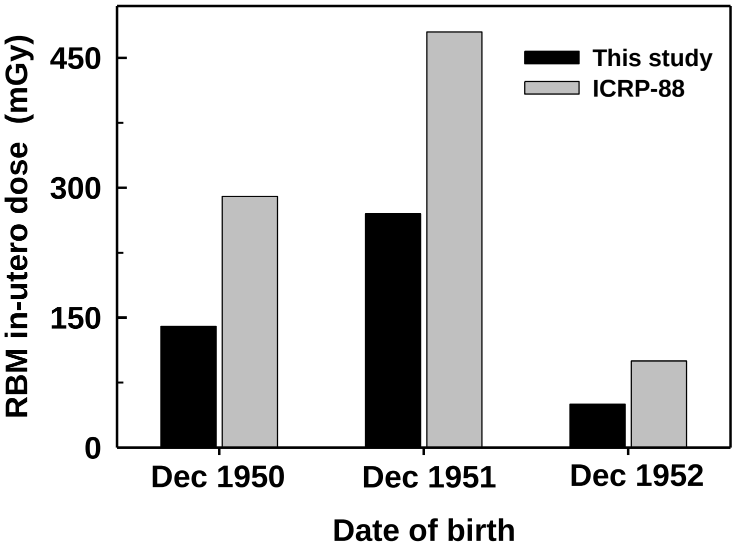

Figure 4 Doses of in-utero RBM exposure of children born in Muslyumovo at different calendar periods due to maternal intakes of strontium isotopes calculated with the models improved in this study and ICRP-88 models (ICRP 2001)

4. Discussion

The levels of maternal ingestion of ^{89}Sr and ^{90}Sr with river water peaked during the first years of LRW discharges into the Techa River (table 1) and consequently resulted in significant levels of foetal exposure. Children, born in Muslyumovo in 1950-1951, during the maximum releases into the river, on the average, accumulated about 25-35 kBq of ^{90}Sr in their skeletons by the time of birth (figure 1). These levels were three to four orders of magnitude higher than ^{90}Sr content in newborns from global fallout after the nuclear weapon tests (Lenihan 1967, Marey 1968). For example, the highest levels of ^{90}Sr concentration in the femur bone reported by Lenihan (1967) for newborns (aged 0-14 days) born in Glasgow in 1964 was 3.5-4.1 pCi/g Ca. The skeletal-averaged modelled ^{90}Sr concentration for children born in Muslyumovo in 1950-1951, expressed in the same units, is about $(2.4\text{-}3.4)\cdot 10^4$ pCi/g Ca.

The maximum levels of *in utero* bone marrow exposure from maternal ingestion of strontium radioisotopes in Muslyumovo were observed in 1951 and amounted to about 300 mGy (figure 4). Short-lived ^{89}Sr present in LRW discharges in significant amounts contributed to these maximum doses (figure 3). In the subsequent period, the in-utero exposure from ^{89}Sr declined substantially due to a decrease in maternal intakes and radioactive decay. The long-lived ^{90}Sr is retained in the maternal skeleton for a long period of time after the intake and becomes a source of ^{90}Sr for the foetus due to the following processes: ^{90}Sr is released from the skeleton mostly due to the process of bone resorption (Shagina *et al* 2003b); the rate of bone resorption doubles during pregnancy (Fell *et al* 2001), leading to ^{90}Sr transfer to maternal blood and, subsequently, to the foetus. As a result, the contribution of ^{90}Sr transferred from maternal skeleton to the foetus increased after 1951 and onwards when the levels of ^{90}Sr transferred from maternal diet decreased. It should be noted that due to long-term ^{90}Sr incorporation in the maternal skeletal tissues, *in utero* exposures occurred even after migration of mothers from the Techa River area.

Comparison of the dose estimates obtained in this study with corresponding estimates evaluated with ICRP-88 models showed that the doses evaluated with the ICRP-88 models are about two times higher (figure 4). These two-fold differences in dose estimates mostly result from differences in dosimetric models. For radiation protection purposes, the ICRP conservatively considers foetal bone of infinite size, therefore, all the initial energy is deposited in the bone. In the current study the dose evaluations are based on skeletal phantoms that provide sufficient anatomical detail to make allowances for finite sizes of bones and deposition of energy in adjacent tissues. The series of phantoms used in this study was constructed taking into account biometrical parameters (such as femur length and bi-parietal diameter) typical for a modern European population (Maynard *et al* 2011). Current studies are now in progress to provide dosimetric models that are based on biometrical and anatomical data that are more typical of the Techa River population. For this purpose, data are used on weights of individual bones and the whole skeleton for foetuses of different gestational ages obtained in the 1960s in Moscow (Borisov 1973). This refined model will also attempt to take account of development of haemopoietic sites in each individual bone with gestational age.

Another area to be considered in the dosimetric model is treatment of the foetal bone microstructure. The approach used in this study considers bones as a homogenous mass due to the absence of data on microstructure of the foetal bone tissues. However, approaches had been developed at the UF for modelling the 3D microstructure of bone tissue. These approaches have been already applied for electron dosimetry in a newborn skeleton (Pafundi *et al* 2010) and can be further extended to the foetal skeleton when data on bone microstructure become available.

The assessment of risks of leukaemia in Techa River populations depends on estimates of doses to haemopoietic tissue. Thus, another important direction of future investigations is more accurate determination of the location of haemopoietic stem cells in the developing foetus. In this study, the doses were calculated for the exposure of the bone marrow on the basis that it is the most affected organ from exposure to ^{89,90}Sr. The bone marrow acquires its haemopoietic function in the beginning of the second trimester and becomes the predominant site of haemopoiesis in the third trimester (Tavian and Peault 2005, ICRP 2001). However, in the developing embryo and foetus, the liver is an active site of haemopoiesis and the stem cells that seed the liver and bone marrow may originate in the yolk sac or in an intra-embryonic site, the aorta-gonad-mesonephros region (Tavian and Peault 2005). While strontium radioisotopes have only a minor contribution to the exposure of these organs, other pathways of exposure need to be taken into account in the Techa River settlements. These pathways included external exposure predominantly in villages located in the upper Techa River region and exposure from ^{137}Cs ingested with maternal diet. These pathways are taken into consideration in evaluation of the total dose to haemopoietic tissue for members of the Techa River in-utero exposed cohort.

5. Conclusion

This paper summarises the progress made in collaborative studies between the Urals Research Center for Radiation Medicine, UK Health Protection Agency and University of Florida to provide the best possible evaluation of the doses of *in utero* exposure from maternal intake of strontium radioisotopes at the Techa River. The dose reconstruction process used in the Techa River study has evolved considerably from the application of empiric approaches to the use of sophisticated state-of-the-art models describing biokinetics and dosimetry of bone seeking radionuclides in foetal tissues. The study continues to adjust the dosimetric models to become more population specific and evaluate correctly the other pathways of *in utero* exposure of members of the Techa River in utero cohort. Epidemiological studies of this cohort (with a follow-up of over 50 years) may provide invaluable information on the risks of health effects from radiation exposure during gestation and early life.

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