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RADIOLOGICAL ASPECTS OF URANIUM CONTAMINATION BY FERTILIZING

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Introduction

Accepted low dose effects of ionising radiation are genetic diseases, tumors, and developmental disorders after in utero exposure. They are initially induced in tissues by mutation of cells or by cell killing. According to dose-effect relationships derived by international expert committees the natural radiation environment contributes to less than 1 % of the occurring malignant and genetic diseases. Natural uranium is mainly composed of the isotope 238 emitting alpha-rays with 12 Bq per mg which is the lowest specific activity of all uranium isotopes. It is therefore usually assumed that the radiation effects of uranium in low concentrations are negligible compared to the heavy metal toxicity. Depleted uranium (DU) used in weapons contains even lower specific activity than natural uranium in its isotopic composition, and there can be no doubt that DU has caused severe health effects in populations of the attacked regions and in army personnel (Bertell 2006). Since these effects became known new discussions arose about the radiological impact or synergistic contribution of the material.

In vitro experiments have shown that chromosome aberrations induced by DU are not explainable by the heavy metal property of the material but should be considered as radiation effect (Miller *et al.* 2002). The exposure conditions in case of natural uranium enhanced by fertilizing are, of course, different compared to DU intake. Chromosome aberrations measured in the blood of Gulf and Balkans War veterans (Schröder *et al.* 2003) show, however, that the conventional methodology to derive doses and dose-effect dependencies is questionable for uranium. Dicentric chromosomes in lymphocytes are a rather radiation-specific mutation and their distribution in cells is a reliable indicator for the effect of alpha-irradiation. The height of the effect found in veterans is in contradiction to the prediction by the physically derived exposure dose.

Dose estimation for incorporated radioactivity

The isotope uranium 238 is the mother of a natural decay chain (Table 4). Natural uranium – and therefore also uranium in fertilizers – contains also the uranium isotope 234 in radioactive equilibrium i.e. their decay rate in Bq is equal. Both isotopes emit alpha-radiation which are double charged particles and are identical with nuclei of helium. Their energy is about 4.5 MeV (only few eV are needed to ionise a molecule or destroy a

molecular binding). Their kinetic energy is rapidly stopped in matter. The range of these alphas in water (tissue) is about 40 μm.

The dose to a tissue which is used to evaluate the biological effect is physically an absorbed energy per tissue mass in Joule/kg (energy dose). Because alpha-radiation of similar energy absorption causes higher biological effects than reference radiation (200 keV X-rays) the energy dose is multiplied with a weighting factor of 20. The weighted energy dose is called "equivalent dose" to a tissue and given in Sv (Sievert).

This kind of dose definition is evidently a very simplified base for evaluating the amount of mutations and other effects, because it averages the energy of the alpha particles over the entire organ in which the radioactive emitter lodges, whereas in reality, the alpha energy is absorbed in a small sphere of surrounding tissue, or in the blood vessels along which the radionuclide travels. It can cause benign tumors in the blood vessels, around which cholesterol plaques form, causing heart attacks and strokes. It also causes depletion of the SOD in cells, which is the enzyme which repairs free radicals (Viglino *et al.* 1986)

In order to calculate the absorbed energy for the case of radioactivity incorporation it must be integrated from the amount of the specific radionuclide entering the regarded organ or tissue and the variation of the nuclide concentration with time. The International Commission on Radiological Protection ICRP has derived dose coefficients for each nuclide in **Sv per Bq** regarding a single intake (Bq) by inhalation or ingestion. In order to do this the human body is simulated by age specific phantoms where the relevant organs and tissues are represented by calculable geometrical structures.

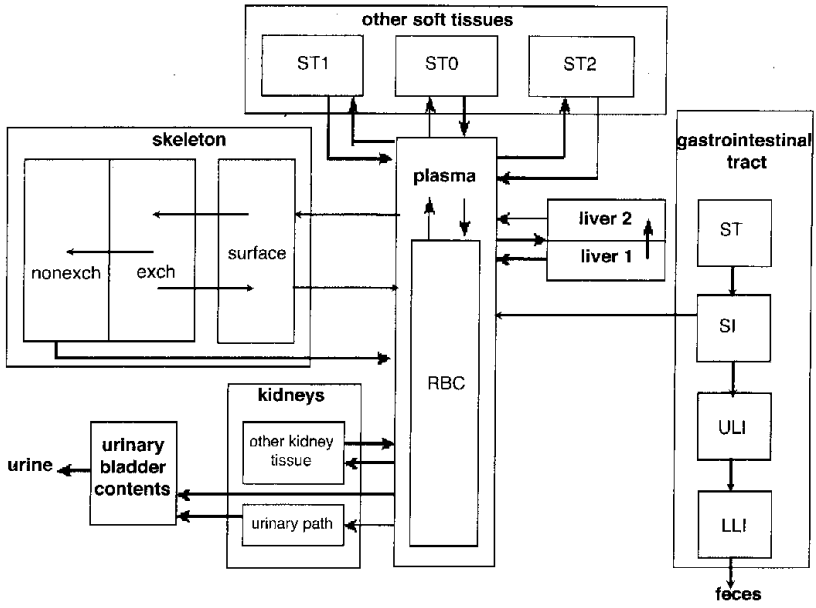

Fig.1. Biokinetic model for ingestion of uranium. STO, tenacious retention; ST1, intermediate retention; ST2, rapid retention; RBC, red blood cellc; ST, stomach contents; SI, small intestine contents; ULI, upper large intestine contents; LLI, lower large intestine contents; EXCH, exchangeable uranium in bone volume; NON-EXCH, non-exchangeable uranium in bone.

In case of ingestion the radioactivity passes the stomach and is partly transfered to the blood from the small intestine. The blood activity is transported to the organs and tissues and deposited and excreted from there. The absorption fraction or "gut factor" f_1 is a most important parameter in dosimetry because it is proportional to the organ doses except for the colon and the other excretion organs. It is clear that f_1 and the tranfer to the organs depend on the solubility of the radioactive material, i.e. the chemical compound and the physical size, as well as of the physiology of the affected person. The metabolism of uranium which has arrived in the blood plasma is simulated in a modification of the "alkaline earth model" (ICRP 1995), the compartments are shown in Fig.1.

The dose coefficients for ^{238}U (Table 1) are considerably lower compared to other natural and man-made actinides emitting alphas (e.g. ^{228}Th, ^{226}Ra, ^{239}Pu) and the ^{238}U-descendant ^{210}Pb. This is partly determined by the low absorption fraction f_1 which is set 2% except for children aged ≤ 1y where it is 4%. The dose coefficients for the natural isotopes ^{234}U and ^{235}U are very similar to the values in Table 1.

Table 1. Committed effective*) dose and dose of the bone surfaces per unit intake of Uranium-238 via ingestion – here in **µSv/Bq** - for members of the public (ICRP 1995)

Age	3 months	1 y	5 y	10 y	15 y	› 17 y
effective	0.33	0.12	0.08	0.068	0.067	0.045
bone surfaces	6.9	1.6	1.2	1.4	2.1	0.71

*) the "effective" dose is a weighted mean body dose

Although the dose is a function of a variety of material and metabolic parameters which show great variations the ICRP dose coefficients are generally used as a kind of natural constant. But they are given without confidence ranges and are, in reality, highly uncertain (Fairlie 2005; Leggett 2001; Harrison *et al.* 2001). It is often stated that the ICRP dose factors are "conservative" which would mean that nearly all of possible constellations would be included. The modelling is, however, based on the most frequently found values of the parameters and therefore simulating a kind of mean result with unknown confidence limits.

Another problem is the accumulation of doses for chronic exposure as in the case of U in the environment (Arruda-Neto *et al.* 2004; Fisenne *et al.* 1988; Paquet *et al.* 2006). While the ICRP dose coefficients are derived for acute, i.e. short-time intake it has been shown in rats that protracted exposure by U may induce changes in biokinetic parameters when compared to acute intake. An accumulation of U was found e.g. in brain and in seminal fluid, which are not described as a target organ (Fig. 1).

Dosimetry for the offspring

The early stages of life are known to be most sensitive for radiation. It is a basic problem to simulate these stages by physical modelling because they are small in comparison to the range of the ionising particles and they are rapidly changing by growing and differentiation. Additionally, there is a general lack of knowledge about the metabolism of radionuclides in the mother-child system and the fetal tissues (NCRP 1998).

The dose coefficients for the offspring (embryo, fetus, and newborn) are given in dependency of the incorporated activity of the mother (ICRP 2001). Besides the effective dose the brain dose is derived because of the experience in Hiroshima and Nagasaki

where the central nervous system was found to be most radiation-sensitive between the 8th and 15th week of development. They are derived for single uptakes at several stages of incorporation and also for chronic uptakes as follows:

1. The dose to the embryo from conception until the end of the 8th week is taken to be the same as to the maternal uterus.

2. For the fetus, from the 9th week after conception until birth, the dose is estimated by similar biometric and biokinetic modelling as in children and adults. The influx is taken from the placenta concentration which is derived from that in maternal tissues for intakes before or during pregnancy. The relation between the fetal concentration and that in the placenta is mainly gained by animal data.

The ICRP states that their approach is judged to give conservative dose values. This must, however, be doubted merely because of the fact that at first the maternal concentrations must be estimated with the discussed uncertainties. Furthermore it must be considered that microdosimetric effects with the incorporation of radionuclides may lead to comparatively enormous tissue exposures in the early stages.

In experiments in mice where pregnant and newborn animals were injected with the same Pu concentrations the fetal concentration was much lower (up to 500 fold) than that in the postnatal contaminated offspring. But the fetuses showed much higher damages of the haemopoietic tissues which are related by the authors to the leukaemia risk (Lord *et al.* 1992). This high fetal sensitivity - found also at very low incorporation doses (Mason 1989) - was specific for alpha-irradiation. Chronic gamma exposure which was applied for comparison showed much lower effect. A Relative Biological Effectiveness (which corresponds to the above mentioned weighting factor for alphas) between 250 and 360 was estimated in this system for alpha-rays (Jiang *et al.* 1994) which is more than tenfold of the value which is thought to be a conservative approach by the ICRP (20).

In NCRP No.128, a problem is also seen in the extra-embryonal deposition of radionuclides in the uterus - especially in the yolk sac - which was measured in experimental studies for the actinides Pu, Am, Np, and Cm. These actinides are chemically and radiologically similar to uranium. Because the first stages of the hemopoetic system are generated in the yolk sac and also germ cells, these stem cells which later migrate into the embryo are affected (Morgan *et al.* 1992; Sikov 1992; Stather *et al.* 1992). The exposure of such stem cells was discussed in connexion with the observed leukemia clusters near nuclear establishments.

Also in experimental studies, extreme effects by uranium-235 on the development of the central nervous system were shown (Gu *et al.* 2001).

Dose-effect relationships in case of incorporated radioactivity

The human reference collective which is used by the ICRP and other committees to evalutae the effects of ionizing radiations are the survivors of the Atomic bomb attacks in Hiroshima and Nagasaki. They had been exposed predominantly by a short-time flash of very high energetic and therefore highly penetrating gamma-rays. The dose-effect estimations are transmitted to all other exposure conditions underlying the concept of the "equivalent" dose in Sv which is thought to achieve similarity for all kind of radiation.

At least, the experience in populations affected by the Chernobyl accident has shown that this assumption is erroneous. Not only are there higher effects than predicted by the estimated doses but also a broad spectrum of health defects has appeared which was not seen in the A-bomb survivors (ECRR 2006). Common dose-effect assumptions are

therefore not sufficiently reliable to deny possible detriments by incorporated radioactivity.

Uranium effects in humans investigated in epidemiological studies
It is well-known that uranium miners have a high risk to die from lung cancer which is caused by radon, the gaseous effluent of the uranium-238 decay chain. Malign and other radiation-linked diseases in other sites of the body are predominantly attributed to uranium deposits because the exposure is accompagnied by the inhalation of uranium dust. Table 2 shows epidemiological findings about such diseases in miners and other persons exposed by uranium.

Table 2. Uranium effects in humans other than diseases of the respiratory tract.

Diseases	Collective	Reference
All solid cancers	Uranium workers	Ritz 1999
Benign & unspecified tumors	Uranium miners	Roscoe 1997
Blood diseases	Uranium miners	Roscoe 1997
Leukemia	Uranium miners	Mohner et al. 2006;Rericha et al. 2006
	Underground miners	Darby et al. 1995
Lymphoma	Uranium workers	McGeoghegan et al. 2000
Multiple Myeloma	Uranium miners	Tomásek et al. 1993
Gastric cancer	Underground miners	Darby et al. 1995; BEIR IV 1988
	Population in U contaminated region	Wilkinson 1985
Liver cancer	Uranium miners	Tomásek et al. 1993
	Underground miners	Darby et al. 1995
Cancer of the gallbladder & extrahepatic bileducts	Uranium miners	Tomásek et al. 1993
Kidney cancer	Uranium workers	Dupree-Ellis et al. 2006
Mental disorders	Uranium miners	Tomásek et al. 1994
Birth defects	Uranium miners	Müller et al. 1992
	Population in U contaminated region	Shields et al. 1992

Natural uranium from the environment and dose enhancement by fertilizing
The dose per year by natural sources – without regarding radon in the lungs – is about 1 mSv. The mean annual contribution of incorporated ^{238}U and ^{234}U is given by the United Nations Scientific Committee on the Effects of Atomic Radiation to 5 µSv (UNSCEAR 1988), i.e. 0.5 % of the background exposure. It is derived from an assumed annual intake of 5 Bq for each isotope by ingestion in normal areas.

Table 3. Specific activity of natural uranium

0.99 mg U-238	0.05 µg U-234	7 µg U-235	1 mg U_{nat}
12.3 Bq	12.3 Bq	0.6 Bq	25 Bq

1 mg natural uranium corresponds to an activity of 25 Bq (Table 3). If 20g uranium is transfered per year by fertilizing to 1 ha of acre (Kratz *et al.* 2007) there will be a deposi-

tion of 50 Bq/m^2. Assuming a distribution in soil of 0.3 m plugging depth and a soil density of 1.5 kg/l the concentration in soil will be enhanced by 0.1 Bq/kg. This is 0.2 % related to the mean basic uranium concentration of soil which is estimated to be about 50 Bq/kg (UNSCEAR 1982). If it is leached completely by annual raining of 600 mm the mean concentration in surface water would be 83 mBq/l. If such a concentration would arrive in drinking water by accumulation the annually added effective dose after ICRP (Table 1) would be 7 µSv for infants (250 l consumption) and 3 µSv for adults (800 l). The corresponding bone surface dose would be 143 µSv and 47 µSv respectively.

Discussing the possible radiological impact of fertilizing it is, however, compelling to consider also other possible contributions of natural radioactivity. Thorium and its derivates are not relevant in the phospate mineral, uranium-238 will be present in radioactive equilibrium with its daughters at least until radium-226 (Table 4). Among these the long-lived alpha-emitters are of dosimetric relevance and are thought to generate in general much higher exposures than uranium (Table 5).

Table 4. Natural decay chain of uranium-238

Nuclide	Half-life	Radiation	Relative activity
U 238	4.5 10^9y	α γ	100
Th 234	24 d	ß γ	″
Pa 234m	1.2 m	ß γ	100
U 234	2.5 10^5y	α γ	100
Th 230	8.0 10^4y	α γ	″
Ra 226	1622 y	α γ	100
Rn 222	3.8 d	α	100
Po 218	3.05 m	α	″
Pb 214	26.8 m	ß γ	″
Bi 214	19.7 m	ß	″
Po 214	1.6 10^{-4}s	α γ	″
Tl 210	1.3 m	ß γ	″
Pb 210	22 y	ß γ	″
Bi 210	5.0 d	ß	″
Po 210	138 d	α	″
Tl 206	4.2 m	ß	″
Pb 206	stable		

Table 5. Ingestion dose coefficients in µSv/Bq derived by the ICRP

		U-238	Th-230	Ra-226	Po-210	Pb-210
Age 3 months	Effective dose	0.33	4.1	4.7	26	8.4
“	Bone Surfaces	6.9	120	160	0.62	67
Adults	Effective dose	0.045	0.21	0.28	1.2	0.69
“	Bone Surfaces	0.71	12	13	0.082	22

The concentration of the radionuclides besides uranium in the fertilizer depends also on the kind of processing. If we follow the UNSCEAR 88 report, where ^{230}Th is assumed to be in equilibrium with ^{238}U, ^{226}Ra to be ¼ of ^{238}U activity, and ^{210}Pb and ^{210}Po to be in

equilibrium with ^{226}Ra, we derive the annual dose contributions of Table 6. This example shows, that the activity of the fertilizer has, indeed, to be controlled.

Table 6. Annual dose enhancement by 83 mBq/l U-nat of drinking water

	U-nat		U-nat+^{230}Th+^{226}Ra+^{210}Po+^{210}Pb	
	Age 3 months	Adults	Age 3 months	Adults
Effective dose	7 µSv	3 µSv	151 µSv	28 µSv
Bone Surfaces	143 µSv	47 µSv	2142 µSv	739 µSv

Conclusions

The Depleted Uranium case is not the only experience where radiation effects are not predicted by standard assumptions. The uranium burden to the environment by phosphate fertilization and that of the other radionuclides of the uranium chain seem to be negligible compared to the natural background. Because the material is, however, physically and chemically processed the biological metabolism may be quite different. This has to be studied predominantly for the early stages of life.

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