

## Dose Estimation for Incorporated Radioactivity

The ICRP dose coefficients for the inhalation and ingestion of radionuclides in special view of depleted uranium

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### Introduction

The effective dose and the organ doses in persons who have incorporated radioactivity are calculated by dose coefficients which were derived by the ICRP (International Commission on Radiological Protection). They are set down for each nuclide in **Sv per Bq** for the case of inhalation or ingestion. Although the dose to a tissue is a function of a variety of parameters which depend on the physical and chemical shape of the nuclide and show great differences among the individuals the ICRP dose coefficients are generally used as a kind of natural constant.

The dose coefficients are given without a range of confidence. It can be shown that they are not conservative and that the uncertainties may partly explain the observed discrepancies between effects after incorporation of radioactivity and expected dose-effect relationships which were derived after external exposure.

### Problems in modeling the transfer and retention of nuclides in organs and tissues

The human body is simulated by age specific phantoms where the relevant organs and tissues are represented by calculable geometrical structures (fig.1).

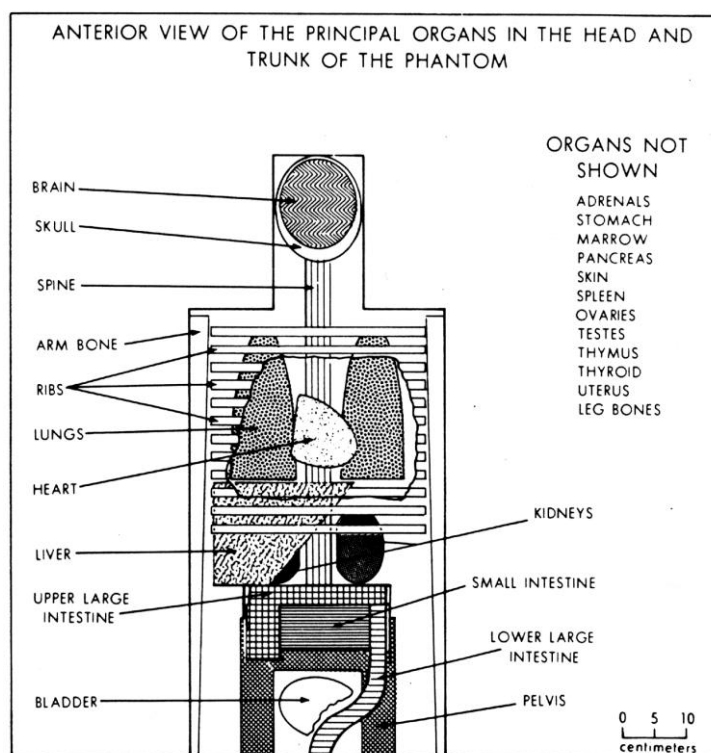


Fig.1  
The ICRP phantom of the reference man

The red bone marrow as origin for the development of leukaemia is assumed to be homogeneously distributed in the whole skeleton.

The dose rate originated by the deposited radioactivity in the organs and tissues is accumulated to the lifetime dose assuming a mean life expectancy of 70 years. The exposures by the daughter products of the decay are included.

In order to simulate the distribution of the radioactive substance in the body and its retention in the different tissues the ICRP has developed a biokinetic model (ICRP No.30), see fig.2. The radionuclide may enter the organism via the respiratory tract or the gastrointestinal tract from where it is absorbed by the body fluids and then translocated to the organs and other tissues. Each organ or tissue of deposition is assumed to consist of one or more compartments, and from each of these compartments the radionuclide is translocated to the excretion pathways. For simplicity, it is usually assumed that there is no feedback in the system.

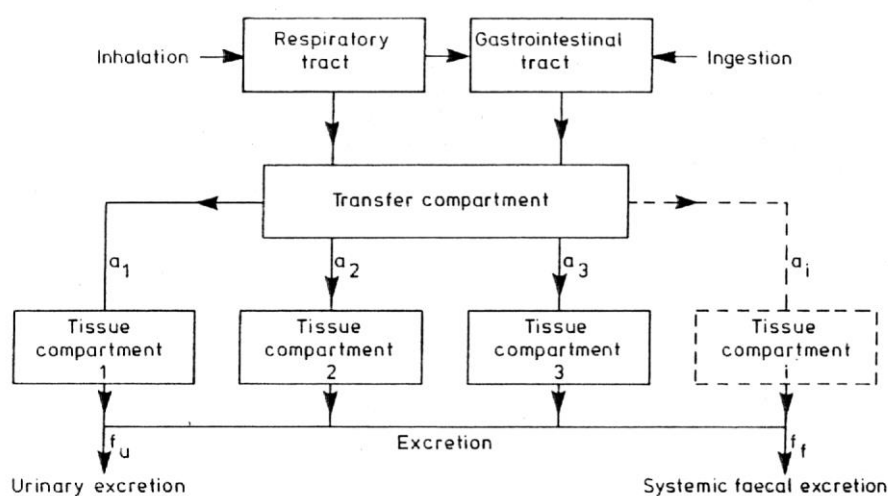


Fig.2 General model of the ICRP to represent the kinetics of radionuclides in body compartments (exceptions are noted in the metabolic data for individual elements).

The model of the gastrointestinal tract (GI) consists of the compartments of the stomach, small intestine, lower colon, and upper colon which are passed by the nuclide sequentially with assumed biological half-lives (fig.3). The absorption from the gut by the body liquids occurs in the small intestine according to an absorption fraction  $f_1$  which represents the portion of the whole substance which is resorbed.

The biokinetic model of the ICRP publication No.30 was developed in order to derive limits for the incorporation of radionuclides for workers, i.e. adults. The dose coefficients committed by the EU (Council Directive 96/29/EURATOM) were developed in the later ICRP publications Nos.56, 67, 69, and 71. They include values for persons of the normal population for the effective dose and for different organs and tissues, regarded for age classes 3 months, 1 year, 5 years, 10 years, 15 years, adults. The values for adults differ in several cases from the estimates for workers. Sex specific alterations are not considered. Data for embryos and fetuses were published in the year 2001 (ICRP No.88).

The methodology of the ICRP shows that the dose coefficients are not derived in a conservative mode. Conservatism would mean that the predominant part of the occurring exposures would be included in a way that no underestimations of doses could be possible. Instead of that a kind of median estimation is aspired.

Many parameters are involved in the estimation of a dose after incorporation of a radionuclide which show great variations in individuals and many uncertainties because of lacking knowledge. Table 1 shows a selection of presently valid dose coefficients.

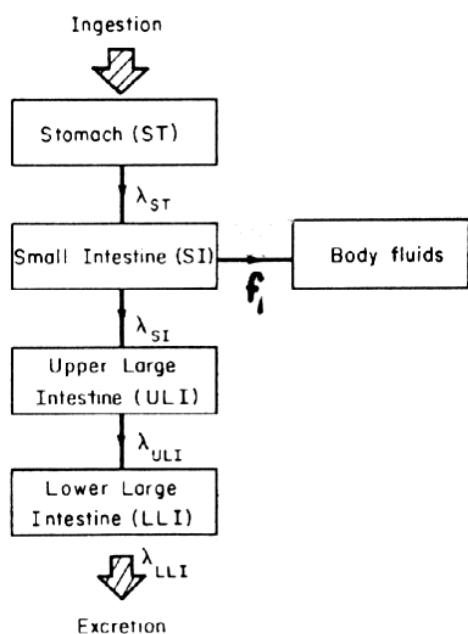


Fig.3

ICRP model of the ingestion pathway

Generally, it must be stated, that the use of the ICRP dose coefficients is suffering from the following problems:

- the dose coefficients are derived without confidence limits
- the biokinetic models of the ICRP are of such a high complexity that the user is not able to evaluate their reliability

Additionally, the effective dose may be higher in reality because of the high Relative Biological Effectiveness (RBE) of alpha-irradiation which is not estimated conservatively by the weighting factor  $w_R$  of 20 of the ICRP in comparison to X- and gamma-rays.

### Dose estimation in the case of aerosol inhalation

The uptake of radionuclides by inhalation is considered to be relevant for workers in nuclear establishments and in nuclear medicine, furthermore for the population after nuclear accidents and from contaminations by Radon in the environment.

Radioactive nuclides may be inhaled although they are not gaseous. This is the case for solid substances if they occur as aerosols. The ICRP divides the respiratory tract in five anatomical regions (fig.4): the anterior nasal region (Extrathoracic  $ET_1$ ), the region of the naso-oropharynx/larynx ( $ET_2$ ), the bronchi (BB), the bronchioles (bb), and the alveolar interstitium (AI). The distribution of the radionuclide to these regions depends on the spectrum of particle sizes characterized by an activity median aerodynamic diameter (AMAD,  $\mu\text{m}$ ) which is gained by randomising according to the radioactivity.

The anatomical regions except ET<sub>1</sub> contain several compartments from where the substance is transferred to the GI tract. The relative volume of the different compartments and the half-lives of the transfer depend on the physico-chemical properties of the aerosol.

**Tab.1** Dose coefficients for selected radionuclides after inhalation and ingestion by adults (members of the public) and in children  $\leq 1$  y, committed for cases of unknown chemical of the compound

Effective dose by EU Directive 96/29/EURATOM in Sv/Bq

Particulate aerosol size for inhalation of particles of diameter 1 AMAD (1  $\mu\text{m}$ )

The time given in the first column is the (physical) half-life of the nuclide

Nuclide	Type of decay	Origin	Adults		Children $\leq 1$ y	
			Inhalation Sv/Bq	Ingestion Sv/Bq	Inhalation Sv/Bq	Ingestion Sv/Bq
K-40 1.26 $10^9$ y	$\beta$	natural	$2.1 \cdot 10^{-9}$	$6.2 \cdot 10^{-9}$	$2.4 \cdot 10^{-8}$	$6.2 \cdot 10^{-8}$
Co-60 5.27 y	$\beta$	neutron activation	$3.1 \cdot 10^{-8}$	$3.4 \cdot 10^{-9}$	$9.2 \cdot 10^{-8}$	$5.4 \cdot 10^{-8}$
Sr-90 29.1 y	$\beta$	fission product	$1.6 \cdot 10^{-7}$	$2.8 \cdot 10^{-8}$	$4.2 \cdot 10^{-7}$	$2.3 \cdot 10^{-7}$
Cs-137 30.0 y	$\beta$	fission product	$3.9 \cdot 10^{-8}$	$1.3 \cdot 10^{-8}$	$1.1 \cdot 10^{-7}$	$2.1 \cdot 10^{-8}$
U-238 4.47 $10^9$ y	$\alpha$	natural	$8.0 \cdot 10^{-6}$ S $f_1=0.002$	$4.5 \cdot 10^{-8}$ $f_1=0.020$	$2.9 \cdot 10^{-5}$ S $f_1=0.020$	$3.4 \cdot 10^{-7}$ $f_1=0.040$
U-235 7.04 $10^8$ y	$\alpha$	natural	$8.5 \cdot 10^{-6}$ S $f_1=0.002$	$4.7 \cdot 10^{-8}$ $f_1=0.020$	$3.0 \cdot 10^{-5}$ S $f_1=0.020$	$3.5 \cdot 10^{-7}$ $f_1=0.040$
Pu-239 24,100 y	$\alpha$	breeding product	$1.2 \cdot 10^{-4}$	$2.5 \cdot 10^{-7}$	$2.1 \cdot 10^{-4}$	$4.2 \cdot 10^{-6}$

### The respiratory tract model of the ICRP

Fig.5 shows the respiratory tract model in a revised version published in 1994 (ICRP No.66). Contrary to the formerly applied simulation (ICRP No.30) which intended to result in a mean lung dose the different radiation sensitivity of the lung tissues are now considered. The tissue of the compartment ET<sub>2</sub> in the extrathoracic region is weighted with a sensitivity factor of 1. Compared to that the compartment BB of the bronchial region is considered to be less sensitive by a factor of 3 and also the tissues of bb and AI. The pulmonary lymph nodes where radioactive particles can be stored are considered to have only 1/1000 of relative sensitivity, and also the tissue of the anterior nasal region ET<sub>1</sub>.

Particles which enter the extrathoracic region ET will be transported quickly by the superficial fluids (fast). In the thoracic part they are divided between the bronchial compartments BB and bb where a relatively fast transport occurs by the cilia, and the compartment AI where the passage is slow.

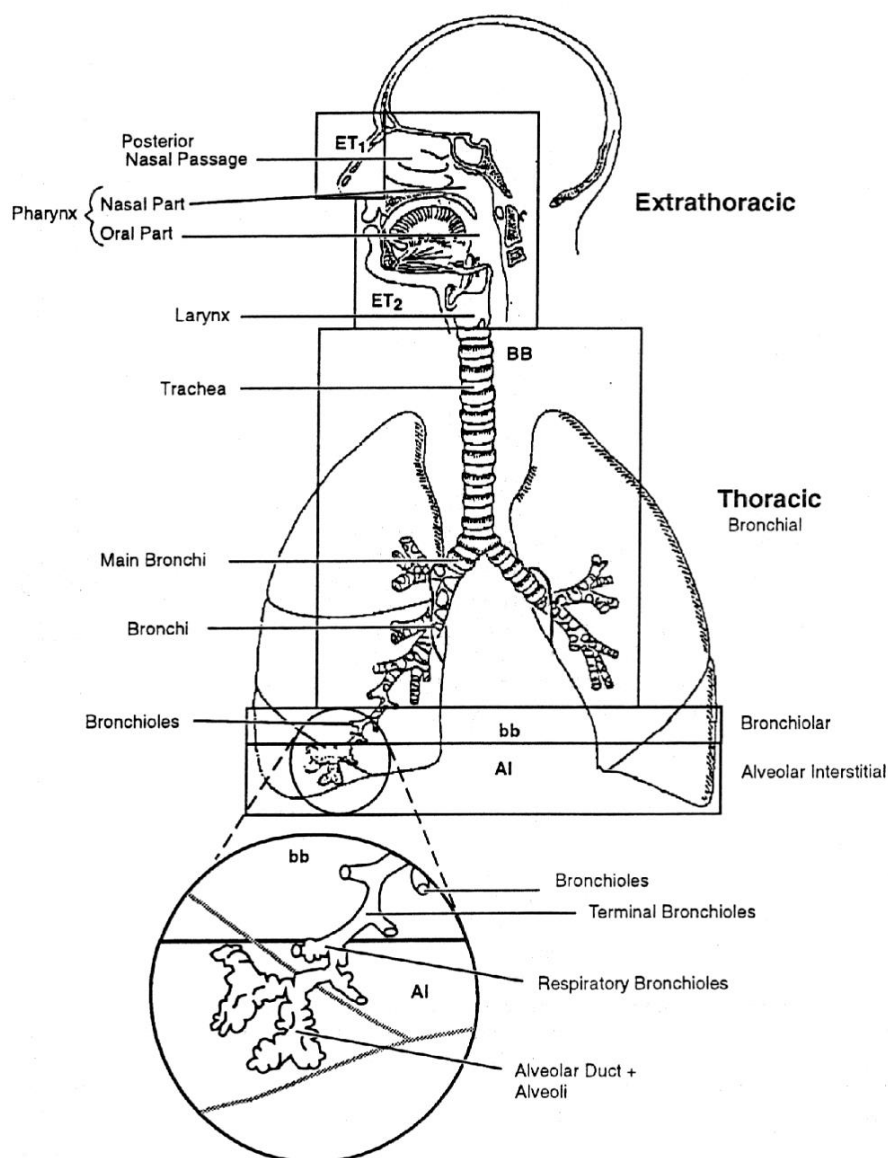
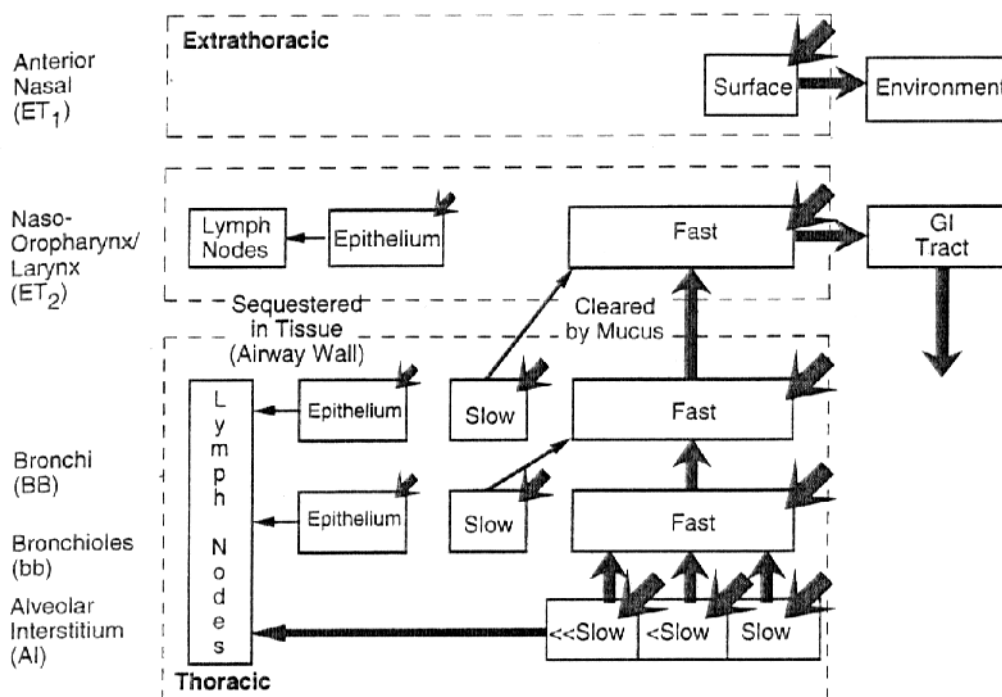


Fig.4 Respiratory tract

The transfer of radioactive aerosols which are inhaled as solid particles to the blood and the following compartments of the body is very difficult to simulate in a quantitative way, because the deposition in the five regions of the respiratory tract depends not only on the size of the particles but also from the respiratory volume and frequency. The transfer is also influenced by the solubility of the substance which also depends on the particle size. The particle size is important for the elimination from the lungs following the deposition. Breathing through the mouth or the nose is relevant, too.

The weighted compartment doses are accumulated to the lung dose equivalent.

The efficacy of the cilia in transporting the deposited particles in each region is discussed controversially (ICRP No.66). The model assumes that half of the particles deposited in BB and bb are subjected to a „slow“ clearance. The likelihood that a particle is cleared slowly by the mucociliary system is assumed to depend on its physical size.



**Fig.5** Respiratory tract model of the ICRP

Material which is deposited in the AI region is divided between 3 compartments (AI<sub>1</sub>-AI<sub>3</sub>), each showing a slow but specific rate of transport.

According to these modifications further qualities have to be considered: the physical mode of the inhaled substance, i.e. gaseous or as aerosol, and then the distribution of sizes; the chemical compounds and their solubility in the lungs. Because these parameters are not generally known in practice, there is a new discussion in the scientific literature about the resulting reliability of the inhalation dose coefficients derived in such a manner.

The former grouping of the radioactive substances in the classes D (days), W (weeks), and Y (years) was not adopted in the new lung dosimetry. Introduced were the classes F (fast), M (moderate), and S (slow) which are now related only to the absorption by the blood in the lungs. The transfer factors to the gastro-intestinal tract and to the lymph nodes are regarded as similar for all classes.

The description of the new lung model and its parameters is very complex and represents a book of 482 pages (ICRP No.66). It is doubted by critics that the higher complexity of the model is accompanied by a higher reliability of the dose coefficients.

Applying the model, Roy has derived confidence regions for the inhalation dose coefficients by varying the particle size distributions and the dissolution within the proposed range. According to the quantity of the confidence interval he divides the nuclides into 4 classes which are considered each to enclose 90 % of the occurring cases. In the third class the confidence interval lies in a region where the lower and the upper limit of the confidence interval differ by factors between 16 and 64. The fourth class has even higher ranges.

Co 60 in the case of inhalation is adjoined by Roy to the second class (factor between the lowest and the highest value of the 90 % confidence limit 4-16) deriving the effective dose, and to the third class for the lung dose. It must be noticed that these variations are originated by systematic differences, therefore they can be relevant for whole contaminated populations and

not only for single individuals. Additionally, there are individual variations because of physiological parameters as breathing frequency and lung volume, which are regarded in ICRP No.71.

Regarding the latter Roy has derived the category one for Co 60 and the effective dose (factor < 4), altogether the factor between the lowest and the highest value for the possible effective dose after the inhalation of the same amount of Co 60 would lie in the region <16 until <64.

For the inhalation of U-238 in unknown modifications the author also offers considerations, see below.

In consequence to the many discussions in the scientific literature which had been initiated by their revised lung model the ICRP published a supporting guidance in 2002. The purpose is to adopt the model in situations *‘where using specific information can give more accurate or more reliable assessments of intake and/or dose than using reference values....’*

Besides the mentioned problems of the lung model there exists another uncertainty in the dose estimation for alpha-emitting particles, because a process of fragmentation must be considered after the deposition of the aerosols (Diel 1983, Fleischer 1977, Stradling 1978). It was observed after the deposition of  $^{238}\text{PuO}_2$  particles of diameters < 5  $\mu\text{m}$  that the fraction of the smallest particles ( $\leq 0.001\mu\text{m}$ ) increases significantly in the course of time. Therefore the solubility and the transport to the adjoining compartments increases. This effect is explained by the recoil because of the alpha-emission.

### **Dosimetry in the case of ingestion: the absorption fraction**

The dose by ingestion of radionuclides is assumed to be relevant e.g. for the exposure of the population by the normal operation of nuclear establishments.

The absorption of the nuclides from the small intestine by the blood is the most important parameter in estimating the doses (fig.3). The absorption fraction or “gut factor”  $f_1$  is proportional to the organ dose except for the colon and the other excretion organs. There are only few investigations of the absorption in humans. A criticism of the ICRP estimations is done by Fischer et al.:

*„While there are complete data sets available for only few nuclides as e.g. I 131 it is necessary for the most substances to refer to animal experiments or to a few single experiences in humans. Therefore the problem exists that a mean absorption is estimated by the few data without knowing the individual variances of the absorption. Also the dependency of the biokinetic reaction from other parameters as the simultaneous uptake of different substances or the varying metabolism of different compounds of the same substance are not regarded.*

*While the different chemical behaviour is only considered by division into soluble and insoluble compounds in ICRP No.2 there are further distinctions for some nuclides (Co, Sr, Mo, and others) in ICRP No.30.*

*The statistical uncertainty of the absorption fraction remains unconsidered in the recommendations of the ICRP, although it is known that the absorption of radionuclides can be described by statistical distributions.“*

The latter was also not altered in the later publications of the ICRP. But there were supplemented age dependencies to the absorption fractions and to the half-lives of the retention in the certain compartments of the body. Whether these assumptions lead, however, to more conservative dose values must be doubted, because it is shown in the literature that the age de-

pendent differences of the biokinetic parameters result in a range of 2 orders of magnitude for the estimated dose (Taylor 1992) which is not reflected in the documented coefficients.

Fischer et al. have undertaken a study about the absorption of cobalt in the literature. Their results for adults and further ones cited from the German Institute of Energetic and Environmental Research (Steinhilber-Schwab 1978) are to be found in table 2. They are compared there with the valid and former assumed  $f_1$  values of the ICRP and also that derived by the NEA Expert Group on gastrointestinal absorption (NEA/OECD 1988). Compared to the recommended ICRP value for adults, the values of the other authors are higher up to 7 fold.

**Tab.2** Absorption of cobalt in adults

Source	Chemical compound and amount	Fraction $f_1$
ICRP No.67		0.1
(ICRP No.30)		(0.3)
NEA/OECD 1988		0.3
Fischer et al.	organically-complex bound Co as vitamin B <sub>12</sub>	0.68 ± 0.15
	inorganic Co:	
	low amount (≤ 2 µg)	0.12 ± 0.16
	high amount (≥ 1 mg)	0.36 ± 0.22
	Co in foodstuffs (6 µg - 8 µg)	0.36 - 0.15
Steinhilber et al.	inorganic Co	0.29 - 0.97
	vitamin bound Co (vitamin B <sub>12</sub> )	0.06 - 0.92

Therefore, it must be concluded that a conservative dose estimation was not chosen by the ICRP.

A further example for great uncertainties in the knowledge about absorption is given by the element plutonium. For adults, an absorption fraction  $f_1 = 10^{-4}$  was adopted in ICRP No.30 for unknown and mixed Pu compounds. It was increased in ICRP No.48 by a factor of 5. Because of altered assumptions about the retention, however, the ingestion dose for Pu 239 in adults results in a lower value than before (see table 1).

In order to consider the retention of the radionuclides for dose estimation the ICRP assumes that the absorption of the radioactivity from the upper intestine by the body fluids (fig.3) is followed by a parallel influx to several compartments without feedback. These tissues and organs are also divided in a different number of compartments depending on the nuclide. It is assumed that the excretion rate from each compartment is constant and the temporal decrease is proportional to the respectively existing radioactivity. This is described by a chain of differential equations for which the ICRP has published solutions in ICRP No.30.



The biological half-lives and the absorption fractions which are used in the compartment models can at best be only represented by median values. The results of the calculations will therefore be also median values at best, without any confidence limits. Thus, the correct estimation of an individual dose is not really possible. The calculations are also not suitable to estimate the maximum possible dose of a population for a given environmental contamination. This could only be done by a „worst case“ consideration.

### **Dosimetry for the offspring**

The early stages of life are known to be most sensitive for radiation. It was found that fetuses in the first trimester were more sensitive than in the later stages, and it is assumed that fetuses are more sensitive than newborns and the latter more than older sucklings.

It is a basic problem to simulate the early 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 No.128, Prosser 1994, Taylor 1991).

The ICRP has published, therefore, dose coefficients for embryos and fetuses not before the year 2001 (ICRP No.88). They are given in Sv/Bq for the offspring dose (embryo, fetus, and newborn) in dependency of the incorporated activity of the mother. The effective dose is derived and the brain dose (because of the experience in Hiroshima and Nagasaki where the central nervous system was found to be most radiation-sensitive between the 8<sup>th</sup> and 15<sup>th</sup> week of development). They are considered for single uptakes at several stages of incorporation and also for chronic uptakes. The mothers are divided in persons from the normal population and in workers.

The ICRP develops the dose coefficients as follows:

1. The dose to the embryo from conception until the end of the 8<sup>th</sup> week is taken to be the same as to the maternal uterus.
2. For the fetus, from the 9<sup>th</sup> week after conception until birth, the dose is estimated by similar biometric and biokinetic modeling 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. The U.S. National Council of Radiation Protection and Measurements, who has published a report about the exposure of embryos and fetuses in 1998, draws especially attention to the fact that several of the considered structures are smaller than the pathways of the ionizing particles.

In experiments in mice where pregnant and newborn animals were injected 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 1992). This high fetal sensitivity was found to be specific for alpha-irradiation. Chronical gamma exposure which was applied for comparison showed much lower effect. An RBE between 250 and 360 was esti-

mated in this system for alpha-rays (Jiang 1994) which is more than tenfold of the value which is thought to be a conservative approach by the ICRP ( $w_R = 20$ ).

These results could be most relevant in evaluating the effects by contaminations of the environment. The authors had undertaken the studies because of the undissolved problem of the Sellafield leukaemias.

In NCRP No.128, a problem is also seen in the extraembryonal deposition of radionuclides in the uterus - especially in the yolk sac - which was measured for the actinides Pu, Am, Np, and Cm (Sikov 1992). Because the first stages of the haemopoietic system are generated in the yolk sac and also germ cells, these stem cells which later migrate into the embryo are affected. The exposure of such stem cells was also discussed in connexion with the observed leukaemia clusters near nuclear establishments (Sikov 1992, Stather 1992).

Especially the yolk sac showed high concentrations of Pu in the animal studies with monkeys and rodents (Morgan 1992, Sikov 1992, Stather 1992). The already mentioned damages of the haemopoietic system in mice by Pu 239 in the early development were observed also for a range of very low incorporation doses (Mason 1989). Therefore, a consideration of the embryonic exposure by this pathway is demanded which was, however, not fulfilled by the ICRP.

### Dose estimation after uranium inhalation

The inhalation of the nuclide is assumed to be the critical pathway for U incorporation, because a much faster and more effective elimination from the body is observed in the case of ingestion. The dose coefficients of the U isotopes for inhalation are therefore higher compared to the case of ingestion for about two orders of magnitude (Table 1). The inhalation dose coefficients are also derived for a particle size of 1 AMAD (1  $\mu\text{m}$ ) diameter. The assumed properties of uranium compounds are shown in Table 3.

**Tab.3** Compounds, lung absorption types, and  $f_1$  values acc. to ICRP (1995) for the calculation of inhalation dose coefficients for uranium

Absorption type	$f_1$	Compound
F (fast clearance)	0.020	Highly soluble compounds: Most hexavalent compounds as pure $\text{UO}_3$ ; $\text{UO}_2(\text{NO}_3)_2$ instilled intratracheally
M (moderate clearance)	0.020	Less soluble compounds: $\text{UO}_3$ hydrated $\text{UO}_2(\text{NO}_3)_2$ in aqueous solution, $\text{U}_3\text{O}_8$ sometimes Hexavalent compounds other than F
S (slow clearance)	0.002	Highly insoluble compounds: as $\text{UO}_2$ $\text{U}_3\text{O}_8$ sometimes

The descendants of uranium contributing to the dose are shown in Tables A, B (Appendix).

The committed dose factors by the European Union Council Directive 96/29 EURATOM for ingested or inhaled uranium by members of the public of different ages are also given in Ta-

bles C, D (Appendix). For inhalation of uranium in absence of specific knowledge about the chemical form the absorption type M is committed by the ICRP (1995), for ingestion

Because of the generation process by nuclear decay the activity of an descendant in the chain can not exceed the mother's activity.

The overwhelming part of the radiation emitted from the nuclides in the natural U-238 series is emitted from the nuclides that follow after U-234. Compared with the sum of the energy of the alpha radiation emitted per transformation from all isotopes in the U-238 series, the nuclides that follow after U-234 emit about 89 % of the alpha energy, about 58 % of the beta energy, and about 98.6 % of the gamma energy (Table A).

In natural composition the isotope U-238 is accompanied by U-235 which amounts to 0.7 % of the U-238 weight. The emitted radiation energy of the U-235 chain is therefore much lower than from U-238 and its daughters.

The composition of depleted uranium (DU) is quite different. Its origin is the enrichment process of chemically pured uranium for nuclear fuel, separated from all daughter products beyond U-234. After enrichment of the fissile U-235 isotope (usually to 4 % for common light water nuclear power plants) there remains uranium as an depleted by-product. It is also possible that DU originates from the reprocessing of burned nuclear fuel.

The composition of DU in Table 4 considers pure uranium. Th-234 and Pa-234 are the short-lived descendants of U-238 and therefore in activity equilibrium, Th-231 is the short-lived daughter of U-235 (Tables A, B).

**Tab.4** Composition of DU from nuclear fuel production (UNEP 2002)

Isotope	Share	Specific activity Bq/mg DU
U-238	99.8000%	12.27
U-235	0.2000%	0.16
U-234	0.0010%	2.29
Th-234	Traces	12.27
Pa-234	Traces	12.27
Th-231	Traces	0.16
		<b>Sum 39.42</b>

Uranium occurs naturally in the +2,+3,+4,+5, and +6 valence states, but it is most commonly found in the hexavalent form. In nature, hexavalent uranium is commonly associated with oxygen as the uranyl ion,  $\text{UO}_2^{2+}$ . The different isotopes of uranium are chemically identical and thus exert the same chemical and toxicological effects.

Metallic DU reacts chemically in the same way as metallic uranium, which is considered to be a reactive material. The general chemical character of uranium is that of a strong reducing agent, particularly in aqueous systems. In air at room temperature, solid uranium metal oxidises slowly. It first assumes a golden-yellow colour. As the oxidation proceeds, the film becomes darker, and at the end of three to four weeks the metal appears black (UNEP 2002).

Upon oxidation, uranium metal forms  $\text{UO}_2$ . Significant oxidation of  $\text{UO}_2$  does not occur except at temperatures above 275 °C. Uranium oxides are sparingly soluble but in a moist environment will gradually form hydrated oxides. Under such conditions, the addition of 0.75 % titanium to DU metal as used in penetrators appears to slow the oxidation rate.

Microbial action can speed the corrosion of uranium. The corrosion rate is controlled by several variables, including the oxygen content, presence of water, size of metal particles, presence of protecting coatings and the salinity of any water present. The principal factor controlling corrosion is the size of the particles. Small particles of uranium metal, produced by abrasion and fragmentation, corrode rapidly. Large masses of uranium metal corrode very slowly. In the long term, all uranium metal will oxidise to  $\text{U}^{4+}$  and  $\text{U}^{6+}$ . Studies carried out on penetrators collected by the UNEP DU mission to Kosovo in 2000 showed that impact on the ground causes numerous fine cracks in penetrators. This favors corrosion and dissolution. Rapid dissolution has been further confirmed by studies made on penetrators collected during the UNEP mission to Serbia and Montenegro in 2001 (UNEP 2002).

DU, particularly as a powder, is a pyrophore, which means that it can ignite spontaneously at temperatures of 600-700 °C. When DU burns, the high temperatures oxidise the uranium metal to a series of complex oxides, predominately triuranium octaoxide ( $\text{U}_3\text{O}_8$ ), but also uranium dioxide ( $\text{UO}_2$ ) and uranium trioxide ( $\text{UO}_3$ ).

The dose originated by inhaled DU will therefore strongly depend on the particle sizes and their distribution as well as the different oxidation states of the metal, further on the additional radioactive tracers and possible relics of reprocessing.

Discussing the ICRP respiratory track model for uranium, the author Roy has found the following dependencies: variations of the particle size (the assumed ICRP reference value is for diameter 1  $\mu\text{m}$ ) between 0.3 – 5  $\mu\text{m}$  lead to category 1-2 for the effective doses, lung, and bone surface doses for all ages, that means the relation between the lower and the upper limit goes up to 16. Changing the dissolution type from F, M to highly insoluble compounds of type S (Table 3) has an enormous influence on the lung doses.

Bolch et al. (2003) have studied the influence of the particle size and deposition fraction in the respiratory tract model for the  $^{238}\text{U}$  compounds  $\text{UO}_2$  and  $\text{U}_3\text{O}_8$  which are assumed to be highly insoluble (Tab.3). They chose the integral number of nuclear disintegrations  $\text{U}(\text{s})$  in various lung compartments as a measure of exposure to these tissues. The results were again expressed by the relation of the upper to the lower limit of the 95 % confidence interval. For particle sizes  $<5 \mu\text{m}$  uncertainties in  $\text{U}(\text{s})$  were highest within the BB (seq) tissues, i.e. where the sequestering of particles occurs in the bronchi (Figs 4, 5). The limits of the confidence interval differ by factors of 20-60. As the particle size approaches 10  $\mu\text{m}$  in size, uncertainties in  $\text{U}(\text{s})$  within the three thoracic tissue regions approach a factor of 1000 and are dominated by corresponding uncertainties in particle deposition.

In order to reach more transparency about the confidence limits of the uranium dose estimation when it has passed the lungs, an introduction to the modeling assumptions for this element is given by Leggett (1994). Uranium is primarily considered as similar to the alkaline earths. The ICRP has developed a special biokinetic model for this group of elements (Ca, Sr, Ba, Ra) because of their bone-seeking behaviour. A specialty of the model is the division of the bones in several compartments.

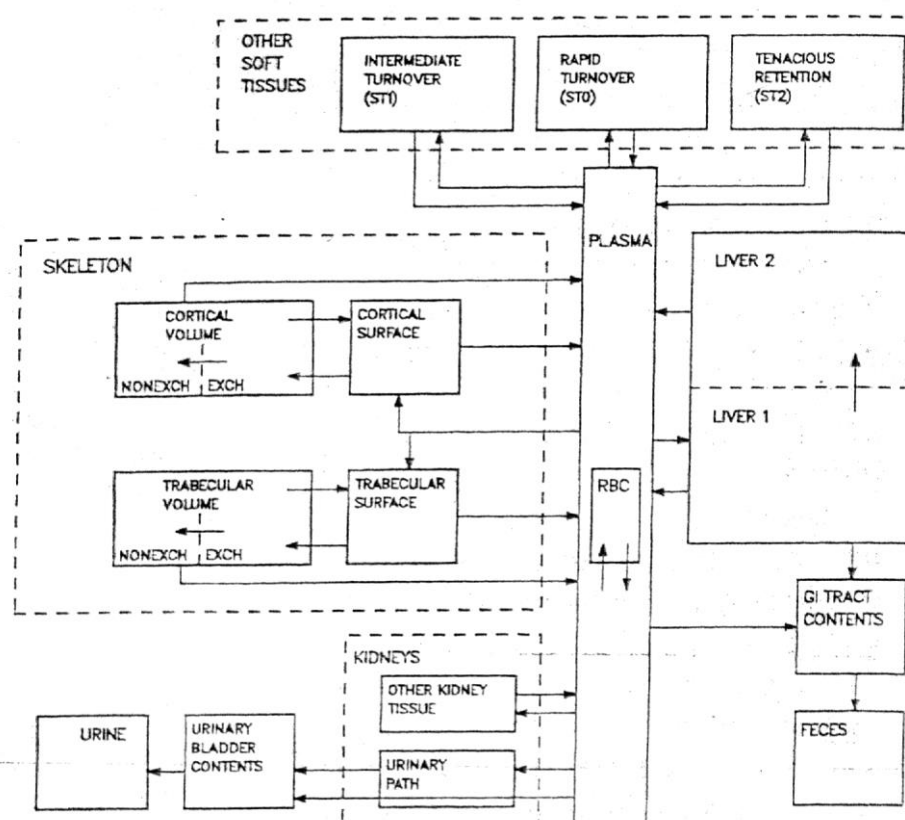
The biokinetic model for uranium used in ICRP Publication 69 (1995) based on the work of Leggett is shown in Fig.6. In a later publication he discusses the biokinetic models in general and also the variability of the occurring parameters for uranium.

A range of confidence limits for evaluating the exposures of children and adults from DU was not yet derived in the literature, also not for fetuses.

## Conclusions

The dose coefficients of the ICRP have been adopted by the national governments in every stage of their development without restriction. Only in the last years there were increasing demands on confidence limits in the scientific literature.

The chain of uncertain approaches is already beginning with the physical modeling of the organs and tissues because of the individual variation of the compartment shape and mass (which is the denominator of the dose term). Examples for children and adults showed variations up to a factor of 7 (Fischer et al. 1982). Even higher uncertainties are generated by the different metabolic turnovers of the substance in the system which can lead to dose underestimations of 2 orders of magnitude for several radionuclides.



**Fig.6** ICRP biokinetic model for uranium

It is usually claimed by the advocates of the ICRP concepts that the models produce a conservative result, they maintain that many unfavourable assumptions are included. There is, f. i., a possible overestimation for those adults who are older than 18 years at the time of incorporation, because a period of further 52 years of lifetime is included in the dose coefficient. This is, however, realistic in individual cases, and if the effective half-lives of the nuclides are short compared to the lifetime this effect is neglectable and not suitable for compensating deficits in the simulation of the biochemical or biokinetic behaviour.

It has not been proven by the ICRP that a more confidential dose estimate is gained for the collective dose by averaging about a great number of individuals using the recommended dose coefficient, this cannot be guaranteed by the choice of the data and because of the possibility of systematic errors in the modeling. In many cases, a conservative procedure would demand a maximum estimate of the possible exposures because of lacking knowledge.

Special problems arise with the use of the ICRP dose coefficients for embryos and fetuses because the most uncertain estimates are evidently applied to the most sensitive systems. Based on the placenta concentrations after incorporation by the mother the uncertainties of the adult transfer simulation are transmitted. The structures in the early development stages are hardly to simulate because they are very small and rapidly changing. Therefore, not only the assumed metabolism of the nuclides but also the physical model is questionable. Enrichments and inhomogenous distributions of particle emitters in the structures and the neighbouring tissues are not considered even if they have been found in animal experiments as in the case of plutonium.

The ICRP dose coefficients should therefore be applied with caution. They are not sufficiently exact to exclude radiation effects by environmental contaminations.

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**Appendix****Tab.A** Uranium-238 series (ICRP, 1983)

Nuclide	Type of decay	Half-life	Average emitted energy per transformation		
			Alpha energy MeV	Beta energy MeV	Gamma energy MeV
U-238 ↓	$\alpha$	4.469 10 <sup>9</sup> y	4.26	0.010	0.001
Th-234 ↓	$\beta$	24.1 d	-	0.059	0.009
Pa-234m (99.84 %) + Pa-234 (0.16%) ↓	$\beta$ $\beta$	1.17 min 6.7 h	-	0.820	0.013
U-234 ↓	$\alpha$	2.45 10 <sup>5</sup> y	4.84	0.013	0.002
Th-230 ↓	$\alpha$	7.54 10 <sup>4</sup> y	4.74	-	0.002
Ra-226 ↓	$\alpha$	1600 y	4.86	-	0.007
Rn-222 ↓	$\alpha$	3.824 d	5.59	-	-
Po-218 ↓	$\alpha$ (99%) + $\beta$ (0.02%)	3.05 min	6.11	-	-
At-218 (0.02%) +	$\alpha$	1.6 s	6.82	0.04	-
Pb-214 (99.98%) ↓	$\beta$	26.8 min	-	0.291	0.284
Bi-214 ↓	$\beta$ (99%) + $\alpha$ (0.04%)	19.9 min	-	0.648	1.46
Po-214 (99.98%) +	$\alpha$	1.64 10 <sup>-4</sup> s	7.83	-	-
Tl-210 (0.02%) ↓	$\beta$	1.3 min	-	-	-
Pb-210 ↓	$\beta$	22.3 y	-	0.020	0.047
Bi-210 ↓	$\beta$	5.01 d	-	0.389	-
Po-210 ↓	$\alpha$	138.4 d	5.40	-	-
Pb-206		Stable			

d days      h hours      min minutes      s seconds      y years

**Tab.B** Uranium-235 series (ICRP, 1983)

Nuclide	Type of decay	Half-life	Average emitted energy per transformation		
			Alpha energy MeV	Beta energy MeV	Gamma energy MeV
U-235 ↓	$\alpha$	7.04 10 <sup>8</sup> y	4.47	0.048	0.154
Th-231 ↓	$\beta$	25.52 h	-	0.163	0.026
Pa-231 ↓	$\beta$	3.28 10 <sup>4</sup> y	-	0.063	0.048
Ac-227 ↓	$\alpha$ (1.38%)+ $\beta$ (98.6%)	21.77 y	0.069	0.016	0.069
Th-227 (98.6%) + Fr-223 (1.38%) ↓	$\alpha$ $\beta$	18.72 d 21.8 min	5.95	0.046 0.391	0.106 0.059
Ra-223 ↓	$\alpha$	11.43 d	5.75	0.075	0.133
Rn-219 ↓	$\alpha$	3.96 s	6.88	-	0.058
Po-215 ↓	$\alpha$	1.78 10 <sup>-3</sup> s	7.52	-	-
Pb-211 ↓	$\beta$	36.1 min	-	0.454	0.053
Bi-211 ↓	$\alpha$ (99.7%)+ $\beta$ (0.28%)	2.14 min	6.68	-	0.047
Po-211 (0.28%) + Tl-207 (99.7%) ↓	$\alpha$ $\beta$	0.516 s 4.77 min	0.021 -	- 0.492	- -
Pb-207		Stable			



**Tab.C** Committed effective dose per unit intake via inhalation (Sv/Bq)  $H_T$  for members of the public**Uranium-238**

Type	Age ≤ 1 y		Age	1-2 y	2-7 y	7-12 y	12-17 y	> 17 y
	$f_1$	$H_T$	$f_1$	$H_T$	$H_T$	$H_T$	$H_T$	$H_T$
F	0.040	$1.9 \cdot 10^{-6}$	0.020	$1.3 \cdot 10^{-6}$	$8.2 \cdot 10^{-7}$	$7.3 \cdot 10^{-7}$	$7.4 \cdot 10^{-7}$	$5.0 \cdot 10^{-7}$
M	0.040	$1.2 \cdot 10^{-5}$	0.020	$9.4 \cdot 10^{-6}$	$5.9 \cdot 10^{-6}$	$4.0 \cdot 10^{-6}$	$3.4 \cdot 10^{-6}$	$2.9 \cdot 10^{-6}$
S	0.020	$2.9 \cdot 10^{-5}$	0.002	$2.5 \cdot 10^{-5}$	$1.6 \cdot 10^{-5}$	$1.0 \cdot 10^{-5}$	$8.7 \cdot 10^{-6}$	$8.0 \cdot 10^{-6}$

**Uranium-234**

Type	Age ≤ 1 y		Age	1-2 y	2-7 y	7-12 y	12-17 y	> 17 y
	$f_1$	$H_T$	$f_1$	$H_T$	$H_T$	$H_T$	$H_T$	$H_T$
F	0.040	$2.1 \cdot 10^{-6}$	0.020	$1.4 \cdot 10^{-6}$	$9.0 \cdot 10^{-7}$	$8.0 \cdot 10^{-7}$	$8.2 \cdot 10^{-7}$	$5.6 \cdot 10^{-7}$
M	0.040	$1.5 \cdot 10^{-5}$	0.020	$1.1 \cdot 10^{-5}$	$7.0 \cdot 10^{-6}$	$4.8 \cdot 10^{-6}$	$4.2 \cdot 10^{-6}$	$3.5 \cdot 10^{-6}$
S	0.020	$3.3 \cdot 10^{-5}$	0.002	$2.9 \cdot 10^{-5}$	$1.9 \cdot 10^{-5}$	$1.2 \cdot 10^{-5}$	$1.0 \cdot 10^{-5}$	$9.4 \cdot 10^{-6}$

**Uranium-235**

Type	Age ≤ 1 y		Age	1-2 y	2-7 y	7-12 y	12-17 y	> 17 y
	$f_1$	$H_T$	$f_1$	$H_T$	$H_T$	$H_T$	$H_T$	$H_T$
F	0.040	$2.0 \cdot 10^{-6}$	0.020	$1.3 \cdot 10^{-6}$	$8.5 \cdot 10^{-7}$	$7.5 \cdot 10^{-7}$	$7.7 \cdot 10^{-7}$	$5.2 \cdot 10^{-7}$
M	0.040	$1.3 \cdot 10^{-5}$	0.020	$1.0 \cdot 10^{-5}$	$6.3 \cdot 10^{-6}$	$4.3 \cdot 10^{-6}$	$3.7 \cdot 10^{-6}$	$3.1 \cdot 10^{-6}$
S	0.020	$3.0 \cdot 10^{-5}$	0.002	$2.6 \cdot 10^{-5}$	$1.7 \cdot 10^{-5}$	$1.1 \cdot 10^{-5}$	$9.2 \cdot 10^{-6}$	$8.5 \cdot 10^{-6}$

**Tab.D** Committed effective dose per unit intake via ingestion (Sv/Bq)  $H_T$  for members of the public**Uranium-238**

Age ≤ 1 y		Age	1-2 y	2-7 y	7-12 y	12-17 y	> 17 y
$f_1$ for ≤ 1 y	$H_T$	$f_1$	$H_T$	$H_T$	$H_T$	$H_T$	$H_T$
0.040	$3.4 \cdot 10^{-7}$	0.020	$1.2 \cdot 10^{-7}$	$8.0 \cdot 10^{-8}$	$6.8 \cdot 10^{-8}$	$6.7 \cdot 10^{-8}$	$4.5 \cdot 10^{-8}$

**Uranium-234**

Age ≤ 1 y		Age	1-2 y	2-7 y	7-12 y	12-17 y	> 17 y
$f_1$ for ≤ 1 y	$H_T$	$f_1$	$H_T$	$H_T$	$H_T$	$H_T$	$H_T$
0.040	$3.7 \cdot 10^{-7}$	0.020	$1.3 \cdot 10^{-7}$	$8.8 \cdot 10^{-8}$	$7.4 \cdot 10^{-8}$	$7.4 \cdot 10^{-8}$	$4.9 \cdot 10^{-8}$

**Uranium-235**

Age ≤ 1 y		Age	1-2 y	2-7 y	7-12 y	12-17 y	> 17 y
$f_1$ for ≤ 1 y	$H_T$	$f_1$	$H_T$	$H_T$	$H_T$	$H_T$	$H_T$
0.040	$3.5 \cdot 10^{-7}$	0.020	$1.3 \cdot 10^{-7}$	$8.5 \cdot 10^{-8}$	$7.1 \cdot 10^{-8}$	$7.0 \cdot 10^{-8}$	$4.7 \cdot 10^{-8}$