A study of uranium excreted in urine

An assessment of protective measures taken by the German Army KFOR Contingent

Research report

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January 2001

GSF Report 3/01

(The views expressed in this report are the authors' own and do not necessarily reflect those of the Federal Ministry of Defense.)

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1. Introduction and definition of problem

In the course of Operation Allied Force from 22 March to 10 June 1999, NATO forces used depleted uranium (DU) ammunition to penetrate armored targets. In the interest of the members of the German Army KFOR Contingent serving in Kosovo, it is therefore necessary to clarify the question whether there is a connection between their stay and activities in this area, although some time after the military air operations, and possible adverse health effects caused by the incorporation of uranium which continues to be present in ammunition remains.

The question of a possible intake of uranium can in principle be answered by measuring the excretion of uranium in urine, since our knowledge of the metabolic behavior of incorporated uranium is sufficiently reliable. At the request of the Federal Ministry of Defense on 30 June 1999, the *GSF - National Research Center for Environment and Health, Neuherberg* submitted an offer to carry out the necessary research work; this offer was accepted by the FMOD in the proposed form. Additionally, the FMOD established a special task force (Biomonitoring Working Group) to support the study. Initial research work was performed on 30 November 1999.

This report summarizes and interprets the results obtained from research work carried out between 30 November 1999 and 17 December 2000. Its structure is as follows. Chapter 2 briefly describes the major properties of natural and depleted uranium, particularly its radiological characteristics. Chapter 3 contains a basic account of the metabolic processes of uranium, followed by a description of the chemical and radiological toxicity of uranium or depleted uranium in section 4. In order to assess the effects of an additional incorporation of uranium or depleted uranium, it is important to identify what the fraction of a certain measured uranium concentration in urine is attributable to natural sources. This issue is therefore addressed in detail in a separate section (section 5). The method used to measure the concentration of uranium in urine and the implications for interpreting the measurement results are described in section 6. Section 7 outlines the intended scope and the organizational sequence of investigations. For an overview of the results obtained thus far, see section 8, and for a discussion and evaluation of these results, see section 9. Section 10 contains a summary of the study, and section 11 a list of pertinent scientific publications. Tables listing individual measurement results have not been included for reasons of data confidentiality.

2. Depleted uranium

Uranium, with atomic number 92, is a natural element; it is a silver-gray heavy metal with an

average concentration of approximately 3 mg per kg in the earth's crust and is thus more common than, for example, mercury, silver or gold. **Natural uranium** exists as a mixture of three different isotopes, namely ^{238}U (99.2745%), ^{235}U (0.7200%) and ^{234}U (0.0055%) (see Table 1). ^{238}U and ^{235}U are primordial substances and the parent nuclides for two of the three natural decay chains (the third parent nuclide is ^{232}Th) that, via a series of radioactive daughter products, ultimately result in a stable lead isotope. Some of these daughter products may remain in the vicinity of the decaying atomic nucleus and must therefore be included in toxicity assessments.

For most nuclear technology applications, the relatively low proportion of fissionable ^{235}U (0.7200%) in natural uranium is insufficient and must therefore be enriched. After the isotope separation process, the remaining uranium is referred to as **depleted uranium** with regard to the abundance of ^{235}U (and ^{234}U as well). Depleted uranium normally contains 0.2 to 0.3% ^{235}U. The enrichment process disrupts the radioactive equilibrium of uranium and its progeny which normally prevails in an undisturbed environment. For this reason, radioactive daughter products other than the first short-lived daughter nuclides (^{234}Th and ^{234}Pa for ^{238}U; ^{231}Th for ^{235}U) will not be taken into consideration here.

Table 1: Radiological properties of natural uranium and depleted uranium

Isotope	Half-life (years)	Mass fraction (%)	Activity (mBq/µg U)	Activity ratio ^{234}U/^{238}U	Activity ratio ^{235}U/^{238}U
Natural uranium					
^{234}U	**2.450×10^5**	**0.0055**	**12.40**		
^{235}U	**7.037×10^8**	**0.7200**	**0.60**		
^{238}U	**4.468×10^9**	**99.2745**	**12.40**		
Total			**25.40**	**1.00**	**0.048**
Depleted uranium					
^{234}U	**2.450×10^5**	**0.0010**	**2.26**		
^{235}U	**7.037×10^8**	**0.2000**	**0.16**		
^{238}U	**4.468×10^9**	**99.8000**	**12.40**		
Total			**14.80**	**0.18**	**0.013**

The most important radiological properties of natural uranium and depleted uranium are listed in **Table 1**. The changed isotope composition also results in differences in the radioactivity of natural uranium and depleted uranium. The activity per unit mass of uranium is approximately 25.40 mBq/µg uranium for natural uranium and 14.80 mBq/µg uranium for depleted uranium (containing 0.2% ^{235}U). Thus, the specific activity of depleted uranium as well as its **radio**toxicity (see section 4) is about 40% lower than that of natural uranium.

The differences in the specific activity (per mass of the isotope) between the uranium isotopes are very large and reciprocal to the related half-lives. The specific activity of ^{235}U is six times, that of ^{234}U about 18000 times higher than that of ^{238}U (see **Table 2)**. All in all, however, both natural uranium and depleted uranium are low-level radioactive substances.

Table 2: **Specific activities of major (pure) uranium isotopes**

Isotope	Specific activity	
	(Bq/mg)	(µg/Bq)
^{234}U	2.3×10^{5}	4.3×10^{-6}
^{235}U	71.1	14.1
^{238}U	12.4	80.4

Although depleted uranium has favorable properties such as a very high density of 19.3 g/cm³ (lead: 11.3 g/cm³), its potential range of applications is considerably limited on account of its radioactivity. Depleted uranium is used not only in the **civilian** sector, for example as balance weight in aircraft and in the keel of racing yachts or as shielding material in clinical irradiation facilities, but also in the **military** sector as projectile cores. Due to the special physical properties of depleted uranium, these projectiles are characterized by a very high penetrating power, even for armored targets. When depleted uranium impacts with the armor, part of it (normally 10-30% but no more than 70%), depending on the type and thickness of the armor, is converted into aerosols and - due to its pyrophoric nature - immediately burns, forming low-soluble uranium oxides. They consist largely of "respirable" particles (<10 µm), i.e. they may be inhaled and may even settle in the lower respiratory tract.

Additionally, aerosol deposits on the ground can be resuspended by the wind and subsequently inhaled or ingested with food or water and thus enter the body. These processes and any

associated health risks will be thoroughly discussed in the following sections.

External radiation doses in the vicinity of DU are very low. The specific gamma dose rate of pure ^{238}U at a distance of 1 m is $8.6 \bullet 10^{-8}$ µSv/g • h. A point source of 1 kg ^{238}U generates an annual dose rate of no more than a few µSv within a distance of 1 m. This is approximately a thousandth of the natural radiation dose per year in Germany.

3. Metabolic behavior of uranium

As mentioned above, uranium is a naturally occurring radioactive element that has been present virtually everywhere in the earth's crust since the earth came into being. Depending on the geological substratum, the concentration of uranium in the soil ranges from approximately 1 mg/kg to 10 mg/kg of soil, with an average value of approximately 3 mg/kg. This is the reason why plant and animal food products and water, too, contain different levels of uranium (in Germany 0.0001 to 40 µg/l), depending on its solubility. This is why every individual ingests a certain amount every day which is later excreted. The behavior of a substance in the body (intake, uptake, distribution, excretion) is called **biokinetics**. Uranium, although not an essential element, largely follows other essential or toxic metals in its biokinetic behavior. Since the chemical and thus the biological behavior of uranium is the same for all isotopes, depleted and natural uranium exhibit the same biokinetic behavior.

On average, the total amount of uranium in the human body is approximately 56 µg (equivalent to 690 µBq ^{238}U) in non-exposed subjects [1]. With 32 µg or 56%, the skeleton accounts for the largest share, followed by muscle tissue (11 µg), fat (9 µg), blood (2 µg) and lung, liver and kidney each with less than 1 µg of uranium.

Uranium mainly enters the human body through the ingestion of food and water. The largest amounts, ranging from approximately 10 to 200 mBq per kg net weight, come from fresh vegetables, cereal products and certain kinds of fish [2]. In addition, certain types of mineral water also contain substantial amounts of uranium (0.001 to 500 mBq/l) [3] and may therefore account for a major share of uranium intake if consumed regularly. The average daily intake of uranium in different countries has been analyzed in several studies which have found values ranging between 11 and 18 mBq ^{238}U per day, which are equivalent to 0.9 to 1.5 µg of uranium [4]. Only about 2% of the ingested amount is actually transferred from the gastrointestinal tract into the systemic circulation, while the remainder passes through the gastrointestinal tract without being absorbed and is excreted with feces within a few days.

The intake of natural uranium by inhalation is negligible since the normal air concentration is approximately 0.5 µBq/m³. However, the importance of this route may increase in accidental or occupational exposures or in exposures to resuspended DU aerosols and dust. The fate of an inhaled particle largely depends on its physicochemical properties. Particle size determines the depth of penetration into the lung and the probability of deposition. The chemical solubility of the particle determines the clearance and solution mechanisms to which it is subsequently subjected, i.e. which portion is dissolved and absorbed into the blood and thus transferred to other parts of the body. As shown below, these properties play an important role in determining the resulting radiation dose. Uranium absorbed into the blood from the lungs is subject to the same metabolic changes as that which is absorbed systemically or intestinally.

Uranium absorbed into the systemic circulation of the body is mostly excreted through the renal system within a short time. The remainder is stored in different organs as described above and is also excreted through urine within a prolonged half-life [5]. Endogenous fecal excretion of uranium is only of minor importance. Permanent chronic intake will come close to an equilibrium between the uranium level in the body and renal excretion. Since 98% of ingested uranium passes through the gastrointestinal tract without being absorbed, fecal uranium excretion basically indicates merely the current level of uranium in the food. As this process will be completed within no more than 3 to 4 days, examinations of the feces can prove with reasonable certainty if uranium has recently been incorporated, but fail to detect any incorporation through inhalation. It can, however, be assumed that uranium excretion in urine is proportional to the uranium level in the body. **This is why renal uranium excretion is used in this study to detect incorporated uranium.**

ICRP Publication 69 provides a detailed description of the biokinetic model for systemic uranium [6]. In combination with the ICRP respiratory tract model (ICRP Publication 66) [7] it is now possible to predict uranium excretions in urine for different types of exposure within different periods of time after exposure. ICRP Publication 78 discusses the retention and excretion functions of the major radionuclides [8]. **Table 3** lists several uranium levels to be expected after different periods of time.

Table 3: **^{238}U excretions in urine (in Bq/d per unit intake) to be expected in different exposure scenarios**

Intake pathway	Time following exposure		
	1 d	30 d	180 d
Inhalation (acute; AMAD = 1 µm)			
Solubility type F (fast)	**1.8×10^{-1}**	**6.8×10^{-4}**	**(3.1×10^{-5})**
Solubility type M (moderate)	**2.3×10^{-2}**	**2.7×10^{-4}**	**6.5×10^{-5}**
Solubility type S (slow)	**7.0×10^{-4}**	**7.7×10^{-6}**	**3.3×10^{-6}**
Ingestion (acute; $f_1 = 0.02$)	**1.3×10^{-2}**	**4.8×10^{-5}**	**2.2×10^{-5}**
Direct absorption into blood	**6.5×10^{-1}**	**2.4×10^{-3}**	**1.1×10^{-4}**

As shown in **Table 3**, the fractions of uranium excreted in urine following the inhalation of low-soluble compounds remain considerably high even after long periods of time have elapsed since internalization. This means that even under these circumstances significant levels of internalized uranium can still be detected by measurements of uranium excretion in urine. Section 9 provides a detailed discussion of this fact in connection with the measurement values obtained in this study.

4. Toxicity of uranium

Potential health risks posed by depleted uranium and by uranium in general are attributable to two properties of uranium:

- uranium is a heavy metal and as such **chemically toxic** (as are other heavy metals such as tungsten, lead, cadmium and mercury);

- all uranium isotopes are **radioactive**, i.e. they emit high-energy radiation during their spontaneous transformation and this radiation may have adverse biological effects.

4.1 Chemical toxicity

The chemical toxicity of a heavy metal is identical for all isotopes of the element. Since natural and depleted uranium differ only in their isotopic composition, they share the same chemical

toxicity. Heavy metal compounds may cause a number of cytotoxic effects. For uranium, the most important oxyanions in biological systems are carbonate/bicarbonate compounds, e.g. $[UO_2(CO_3)_2]^2$ [9, 10]. While these compounds are stable at a neutral pH value (pH value of blood) and in this form are not very reactive, the highly reactive uranyl ion $[UO_2]^{2+}$ is released at low pH values (as for example in urine).

Consequently, the **kidney** is the main organ affected by the chemical toxicity of uranium. As is the case with mercury, cadmium and other heavy metals, increased uranium levels lead to a reduction of glomerular filtration, the tubular secretion of organic anions and the reabsorption of filtered glucose and amino acids in the proximal tubules [11].

The severity of damage depends on the uranium level. A single inhalation of 8 mg of soluble uranium is regarded as the threshold level for transient kidney toxicity; persistent damage can be caused by 40 mg.

Today, the U.S. uranium industry considers **0.05 mg of soluble uranium per cubic meter of air** and 0.25 mg of insoluble uranium **as safe limits for the chronic exposure of workers**. These values are expected to prevent uranium levels in the kidneys from exceeding 3 µg/g. Germany, too, has established a maximum allowable concentration (MAC) of 0.25 mg/m³. Publications from the early years of the uranium industry specify much higher exposure levels of up to 10 mg/m³ at which there occurred no detectable health problems among workers.

The levels of renal uranium excretion to be expected in different exposure scenarios have already been discussed in the previous section (see Table 3).

4.2 Radiotoxicity

The damage caused by **ionizing radiation** from radionuclide transformatios is largely a result of the energy absorbed by body tissues. Radiation doses are expressed in sievert (Sv), millisievert (mSv) or microsievert (µSv) (1 Sv = 1.000 mSv = 1.000.000 µSv). The **dose coefficient** (Sv/Bq) denotes the relationship between the activity intake of a radioactive substance (in Bq) and the radiation dose caused by this substance.

As mentioned above, dose calculations for uranium are based on the biokinetic model and the dosimetric respiratory tract model specified in ICRP Publications (publications 69 and 66, respectively). The age-dependent dose coefficients for the major radionuclides, for different intake pathways and chemical forms which are to be observed following revision of the Radiation Protection Order (*Strahlenschutzordnung*) are listed in the tables attached to Council

Directive 96/29/Euratom of 13 May 1996 (Official Journal of the European Communities of 29 June 1996, ISSN 0376-9453) [12]; they correspond to the values specified in the ICRP Publications 56, 67, 69, 71, 72 [13, 14, 6, 15, 16]. The dose coefficients for committed effective doses of relevant uranium isotopes are listed in **Table 4**. (For organ-specific uranium ingestion and inhalation dose coefficients, see ICRP Publications 69 and 71, respectively.)

Table 4: **Committed effective dose per unit intake (Sv/Bq) of ^{234}U, ^{235}U and ^{238}U for different routes of exposure (according to EURATOM Directive 96/29)**

Intake pathway	Radionuclide		
	^{234}U	^{235}U	^{238}U
Inhalation (acute; AMAD = 1 µm)			
Solubility class F (fast)	5.6×10^{-7}	5.2×10^{-7}	5.0×10^{-7}
Solubility class M (moderate)	3.5×10^{-6}	3.1×10^{-6}	2.9×10^{-6}
Solubility class S (slow)	9.4×10^{-6}	8.5×10^{-6}	8.0×10^{-6}
Ingestion (acute; $f_1 = 0.02$)	4.9×10^{-8}	4.7×10^{-8}	4.5×10^{-8}
Direct absorption into blood	2.3×10^{-6}	2.1×10^{-6}	2.0×10^{-6}

Radiotoxicity, as expressed by the value of the dose coefficients, depends in part on the radiation characteristics of the different nuclides and therefore varies for each uranium isotope. In addition, the resulting dose, as has been mentioned in several instances, also depends to a great extent on biokinetic parameters. This is illustrated in **Table 4**. The inhalation dose coefficient for low-soluble material (S class) exceeds that for readily soluble material (F class) by more than one order of magnitude.

Chronic and permanent ingestion of approximately 1.5 µg of natural uranium per day, a reasonable estimate of the average value prevailing in Germany (see section 3), will lead to an average effective dose of less than 1 µSv per year which is very low compared to dose levels (approx. 2.4 mSv) resulting from other natural sources.

When carrying out dose calculations for isotopic mixtures such as natural and depleted uranium, each isotope must be assessed on a pro rata basis. On account of its lower specific activity, depleted uranium will therefore be assigned a lower level of radiotoxicity than natural uranium.

As described in section 2 and shown in **Table 1**, the specific activity of depleted uranium is about 40% less than that of natural uranium (14.80 mBq/µg versus 25.40 mBq/µg, see Table 1).

As is the case with chemical toxicity, ionizing radiation, too, has a threshold level below which no symptoms of **acute radiation sickness** are observed. This threshold dose is approximately 1 Sv. A threshold level for the incidence of **delayed effects** resulting from radiation, such as a greater likelihood of contracting cancer or suffering genetic damage, is unknown. To be on the safe side, radiation protection experts assume that the incidence of delayed effects arising from low-dose radiation is proportionate to the dose received, regardless how small. Tests conducted on survivors of the first atomic bombs in Hiroshima and Nagasaki as well as comprehensive epidemiological data currently suggest an increase in the incidence of cancer and genetic defects of approx. 0.005% per mSv of the committed effective dose [17]. The current dose limit for the exposure of members of the general public from additional man-made radiation is 1 mSv per year. This type of exposure will at most cause an unmeasurable increase in the natural risk of cancer. Exposure to radon at home and from natural background sources has, however, been explicitly excluded.

In this context it should be mentioned that in Germany the average committed effective radiation dose resulting from **natural** sources (e.g. cosmic radiation, terrestrial radiation, radon, ^{40}K, etc.) is about 2.4 mSv per year. This value plus an average dose level of more than 1.5 mSv from medical applications (radiology and nuclear medicine diagnosis) brings us to an **average effective dose of more than 4 mSv per year for members of the general public**, a dose characterized by significant individual variability. The fact that we are invariably exposed to various radiation sources wherever we may live should be kept in mind when considering radiation exposure in general. These exposures add up to an average value of about 10 µSv per day.

5. Renal uranium excretion in non-exposed subjects

As mentioned in section 3, measurements of uranium excretion in urine, in contrast to feces, provide a reliable basis for detecting any prior DU incorporation. Direct measurements of uranium concentrations in the body are highly insensitive, since the detection limits of this method are ≥ 200 Bq, in other words 15 mg of uranium, owing to the low levels of gamma radiation emitted during uranium decay. For this reason, renal uranium excretion will be discussed in detail in this section.

Until recently, only limited data were available on the uranium content in the bodies of non-

exposed individuals, the resulting excretion levels in urine and the relationship of these values to age and sex, since collecting this data by radiometric procedures such as alpha-ray spectrometry required a great deal of time and effort. Recent developments in the field of mass spectrometry as described in section 6 now make it possible for us to obtain reliable reference data on uranium excretion in non-exposed individuals. Meaningful interpretations of measurements and distinctions between additional incorporation and the contribution of natural sources are impossible unless such reference data are available.

More than 200 male and female healthy volunteers, from 7 to 84 years of age and from different regions throughout Germany took part in research concerning uranium excretion in the urine of non-exposed individuals. The results of these studies [3, 18, 19, 20] can be summarized as follows:

1. There are no differences between men and women with regard to renal uranium excretion per day.

2. On average, the measured values increase with age.

3. The variation range increases significantly as people grow older. Whereas the expected low excretion levels were obtained for most children, much higher values were recorded for some adults. Interestingly, very low levels were also found in higher age groups, i.e. excretion levels are, in fact, spread over two orders of magnitude. This behavior is, however, not only typical of uranium, but can also be observed in other long-lived radionuclides such as ^{226}Ra and ^{232}Th as well as in non-radioactive heavy metals.

An exploration study enabled us to derive from the values obtained for the group of healthy volunteers an age-dependent average value and an upper empirical value of renal uranium excretion in individuals who have not knowingly been exposed to additional radiation. This result is illustrated in **Figure 1**. However, we also found a number of values above this typical reference range.

Based on these results, the average value for 20-year-olds is 15 ng/d and approximately 30 ng/d for 50-year.olds; the upper limits of the reference ranges are 30 ng/d and 70 ng/d, respectively. It is important to note that the upper limits of the reference range must not be misinterpreted as threshold levels for health risks. Instead, they define the range of values obtained for the majority of (although not all) healthy volunteers. In the light of current knowledge on uranium toxicity, a hazardous level of incorporation would result in excretions

exceeding these values by more than one order of magnitude. The tables listing the results of measurements for members of the KFOR contingent (section 8) also include the age-adjusted average values and the upper reference values of non-exposed individuals for purposes of comparison.

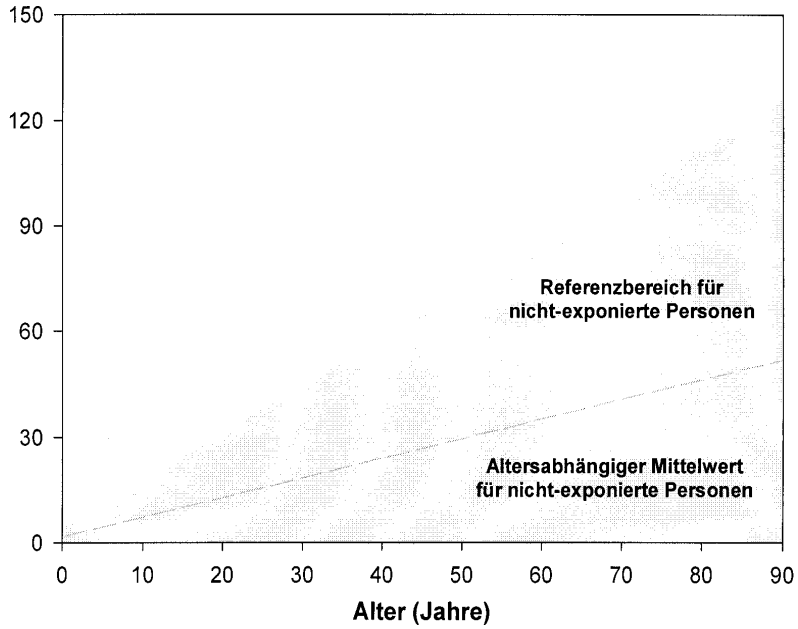

Figure 1: Uranium excretion in urine: age-dependent mean value and reference range for non-exposed subjects	
Original text	Translation
Referenzbereich für nicht-exponierte Personen	Reference range for non-exposed persons
Altersabhängiger Mittelwert für nicht-exponierte Personen	Age-dependent mean for non-exposed persons
Alter (Jahre)	Age (years)

The large variation in levels of excretion is to a high degree attributable to individual dietary habits. In the case of measurement values that are significantly above the reference range, a thorough nutritional analysis should therefore be performed first, in particular with respect to the consumption and supplier of mineral water. There are also cases, however, in which the daily excretion of one and the same person shows considerable variations that go beyond different eating habits; the causes of such variations have yet to be clarified in detail. Examples of such cases are represented in **Figure 2,** which also illustrates the influence of a regular consumption

of mineral water.

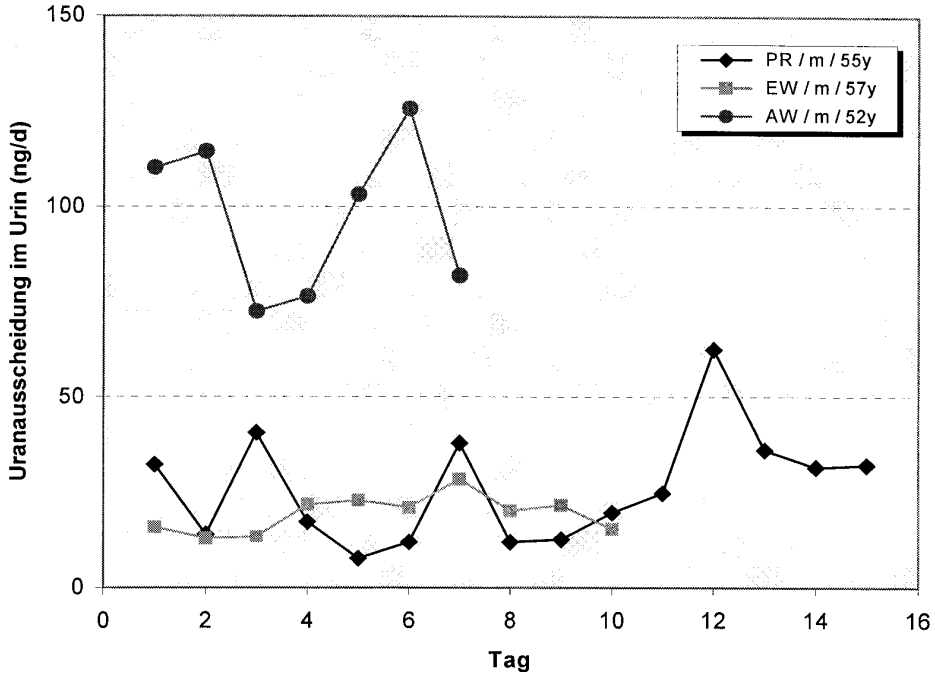

Figure 2: Variations in the daily uranium excretion of three non-exposed persons on subsequent days; AW: approx. 2 l of mineral water (uranium content approx. 10 µg/l) per day	
Original text	Translation
Uran-Ausscheidung im Urin (ng/d)	Urinary uranium excretion (ng/d)
Alter (Jahre)	Age (years)

The individual variation range of excretion levels can differ considerably, as shown in **Figure 3**. On the assumption that the measured values follow a Gaussian distribution, there are great differences in the individual variation ranges of measurement values, i.e. in the standard deviations. To avoid false-positive conclusions drawn from increased excretion levels, multiple examinations should be performed whenever possible, ideally on subsequent days.

A comparison of the data we collected with values published in literature on this subject partly shows marked differences in some cases. According to ICRP Publication 23, the renal excretion of ^{238}U in non-exposed persons should reach levels of between 600 and 6000 µBq per day (this corresponds to 49 and 490 ng of uranium per day). By contrast, the vast majority of the values measured in this study are below 49 ng/day. On the other hand, our values are in broad agreement with the results of an Indian study [22] which found a mean excretion level of 12

ng/day. All other published studies are limited to relatively small numbers of cases and/or are based on outdated analytic methods.

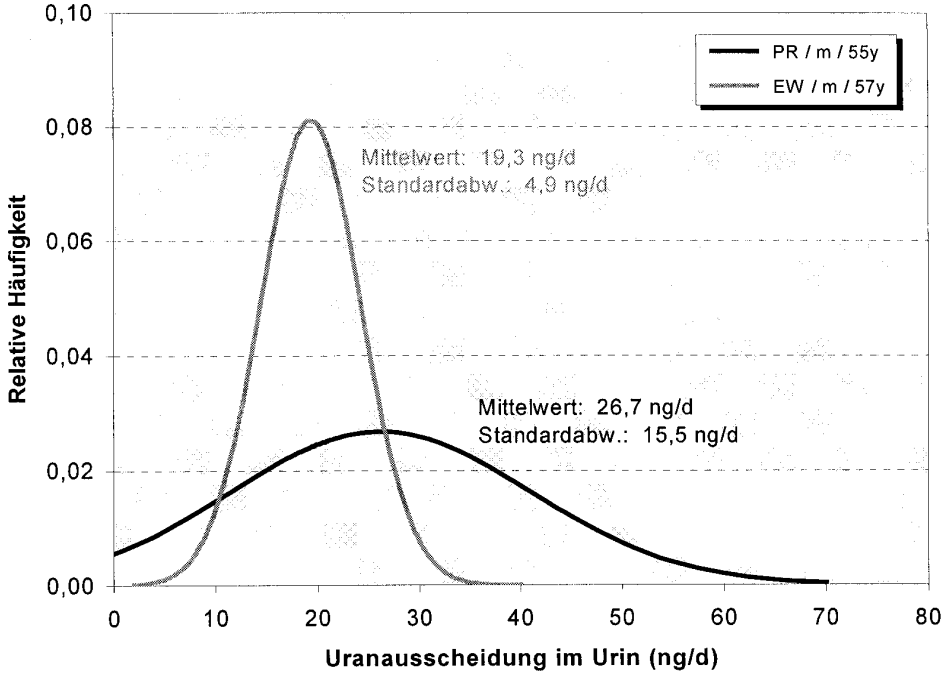

Figure 3: Frequency distribution of n measurement values of daily uranium excretion in two non-exposed persons; (subject PR: n = 15; subject EW: n = 10).	
Original text	Translation
relative Häufigkeit	relative frequency
Uranausscheidung im Urin (ng/d)	urinary uranium excretion
Mittelwert	mean value
Standardabw.	standard deviation

In summary, the method we used to determine renal uranium excretion in non-exposed persons and the analysis of its variations constitute a reliable basis for the evaluation of possible cases of DU incorporation among the members of the German Army KFOR Contingent.

6. Measuring methods applied

In the past, the standard method for determining uranium in excretions was alpha ray spectrometry. This method involves a great expenditure of time and effort, and its detection

limits are too high for the objective of this study. The development of mass spectrometric methods, in particular the ICP-MS (*inductively coupled plasma mass spectrometry*), has made it possible to perform simpler and faster excretion measurements with a higher degree of accuracy. This method has been sufficiently validated and is thus considered to be an established approach. Details of this method are described in [23] and [24]. For this reason, the essential characteristics of how uranium levels in urine are determined by ICP-MS will only be briefly summarized here:

1. A special sample preparation is not required;

2. The detection limit for uranium in urine samples is about 1 ng/liter. Reference values for non-exposed persons can thus also be determined for the first time;

3. The accuracy and reproducibility are good;

4. The calibration curve shows a good linearity of the procedure for a large range of concentration.

In the course of the establishment of this measuring method, further developments in equipment technology have led to a new generation known as high-resolution ICP-MS which has considerably better mass resolution power. This sector field ICP-MS makes it possible to exclude possible interferences between isotope masses to be measured and matrix components in the masses of uranium isotopes and achieves a level of sensitivity which is more than ten times higher than previous methods. This also allows a quantitative determination of isotope ^{235}U in addition to that of ^{238}U. It will thus in principle be possible to assess whether the uranium under study has a natural isotope composition or whether it is "enriched" or "depleted" uranium. Further developments are necessary, however, before we can reliably determine the isotope ratio $^{235}U/^{238}U$ in urine samples with uranium levels in the reference range of healthy persons.

Conducted to a limited extent as a supporting measure to assess exposure conditions, the measurement of uranium concentrations in soil samples involved a far greater analytical and engineering effort. First the soil samples were dried in a heating cabinet at 50 °C for three days. After that, they were rendered soluble with aqua regia according to DIN 38406. This was done by moistening 1 g of soil with high-purity water (MilliQ), mixing it with 10 ml of aqua regia and letting it stand at room temperature overnight (at least 12 hours). The mixture was then boiled under reflux for three hours, filtered off and washed three times with aqua regia. The filtrate was

then made up to 100 with high-purity water; the measurement was then performed using ICP-MS as described above.

7. Check for uranium contamination

7.1 Study groups

Past, present and future contingents were or will be examined in order to evaluate the protective measures taken by the German Army KFOR Contingent to prevent soldiers from incorporating the uranium which NATO used in its ammunition in Kosovo. When this study was launched, past German Army KFOR Contingents had already returned home from Kosovo, others were still serving in this area, and future contingents were preparing for deployment. Accordingly, the study was structured as follows:

a) Study group 1

Due to the high inter-individual and intra-individual variability of uranium excretion, one study group of German KFOR soldiers was examined both **before** and approximately two months **after** deployment began in Kosovo. This group originally included 50 persons, only 43 of whom were examined. Depending on their duties in the area of operations, these persons were assigned to one of two subgroups:

(i) soldiers who, on account of their duties in the area of operations, were likely to come into contact with ammunition or destroyed weapons and equipment or with high levels of dust on the battlefield so that they might have been contaminated with DU and thereby could have incorporated ^{238}U;

(ii) soldiers who were primarily employed in the field camp or hospital in the area of operations; soldiers in this group had living conditions comparable to those of group (i) but a very low risk of ^{238}U contamination (control group).

b) Study group 2: soldiers in the area of operations

This group included members of the German Army KFOR Contingent who were already serving in Kosovo when this study was launched and who, on account of their duties in the area of operations, had come into contact with DU-contaminated objects and were suspected of being contaminated with DU or having incorporated ^{238}U. A total of 38 soldiers from this group were examined.

c) Study group 3: soldiers from past contingents

This group consisted of members of past contingents who, on account of their duties in the area of operations, had come into contact with objects possibly contaminated with DU and, after their return home, were suspected of being contaminated with or of having incorporated depleted uranium. As was expected, the examinations of these persons have not yet been completed and will be continued in the future as required. At the time this report was written, a total of 41 persons had been examined. At present, another 15 persons from this group are being examined.

7.2 Environmental samples

Soil and water samples were analyzed on a smaller scale with a view to assessing the environment where the soldiers may have received exposure. For this purpose, soil samples were taken at selected sites on the basis of the procedure described below (see Figure 4) and analyzed (see section 6).

Untreated water is taken from a municipal cistern located outside the field camp in Prizren and fed through existing pipelines to a water treatment plant. In this plant, the water passes through a precoat filter and is chlorinated. This water is then fed through pipelines to the Prizren field camp where it is used in all camp areas. Four water samples were taken from different water supply points and analyzed in the same way as the urine samples.

7.3 Examination procedure

Participants from group 1 were examined **before** and approximately two months **after their** deployment in Kosovo began. When the soldiers were preparing for deployment, a group of approximately 50 persons gave 24-hour urine collections on two successive days; these samples were analyzed for uranium. The Biomonitoring Working Group of the Bundeswehr was responsible for organizing the collection of urine samples. The collection vessels with relevant instructions and the sample containers were provided by the GSF. These measures were repeated in the same way two months after the soldiers had begun their deployment in Kosovo.

In the same way, examinations involving participants from groups 2 and 3 have been and will be carried out in individual cases at the request of unit surgeons or the Biomonitoring Working Group of the Bundeswehr.

7.4 Measurement

A total number of 271 measurements have thus far been conducted on volunteers from groups 1 to 3. Each member of group 1 was examined four times and members of the other groups were usually examined twice.

As described in section 6, the GSF employed a validated method based on ICP-MS (inductively coupled plasma mass spectrometry) for determining uranium in the urine. All measurement results as well as an interpretation of the data obtained were submitted to the Biomonitoring Working Group.

8. Results

Uranium excretion was measured in the urine of 43 participants from **study group 1** during the preparatory training they received **prior to their deployment** in Kosovo. Urine was collected in the period from 24 April 2000 to 9 May 2000. Most collections, however, were carried out on 8 May 2000 and 9 May 2000. 41 participants took part in two 24-hour urine collections, 2 participants in only one collection. The collection of urine was organized by the Working Group of the Federal Ministry of Defense. An aliquot of 100 ml was taken from each collection, acidified as required and sent to the GSF for analysis.

Figure 5 shows the results of the single measurements (**Figure 5a)** and the mean values which were determined when two analyses had been conducted **(Figure 5b)**. In addition, the Working Group received separate tables giving the names of the participants and their individual measurement results. The total volumes of urine collected during the 24 hours were taken from case histories or the labels on the sample containers. The column entitled "uranium" shows the amounts of uranium excreted per day derived from the concentration measurements and the figures for the total amounts of urine collected. These columns have been highlighted to make them easier to read. Section 9 provides a discussion and an interpretation of the results.

In the second half of September 2000, participants from **study group 1** were examined once again **during** their stay in Kosovo. Urine was collected from 14 September 2000 to 1 October 2000. As already described above, double collections were performed on two successive days in 33 cases; one participant gave only one 24-hour urine sample. Urine collection was organized by the Working Group of the Federal Ministry of Defense in the area of operations, and the samples were flown to the GSF on 2 October 2000 for analysis. **Figure 6** shows the

results of the single measurements (**Figure 6a)** and the mean values determined on the basis of the two analyses **(Figure 6b)**.

38 members of the German Army KFOR Contingent who were already serving in Kosovo when the study was launched **(study group 2)** provided urine samples for an analysis of uranium urine excretion **during** their stay in the area of operations. The samples were collected from 28 March 2000 to 3 April 2000. Two analyses were carried out in 27 cases and one analysis in 11 cases. Urine collection was organized by the Working Group of the Federal Ministry of Defense and all samples were shipped to the GSF for analysis in early April 2000.

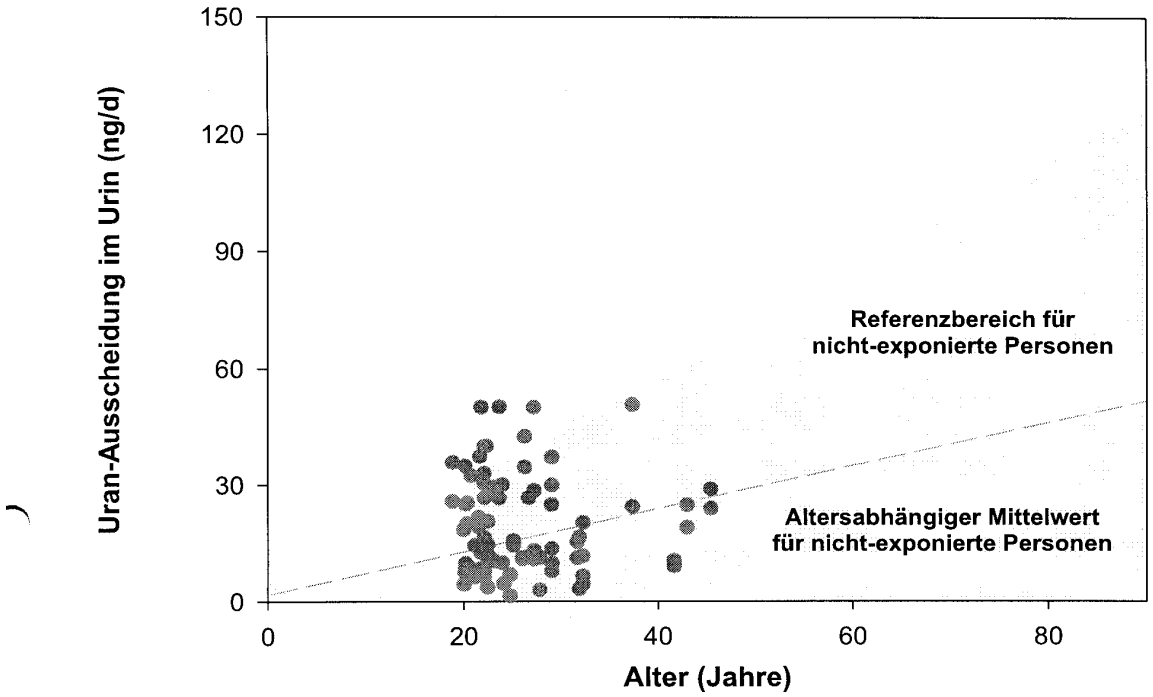

Figure 5a: Urinary uranium excretion in study group 1 (prior to deployment; results of single measurements)	
Original text	Translation
Referenzbereich für nicht-exponierte Personen	Reference range for non-exposed persons
Altersabhängiger Mittelwert für nicht-exponierte Personen	Age-dependent mean for non-exposed persons
Alter (Jahre)	Age (years)
Uran-Ausscheidung im Urin (ng/d)	Urinary uranium excretion (ng/d)

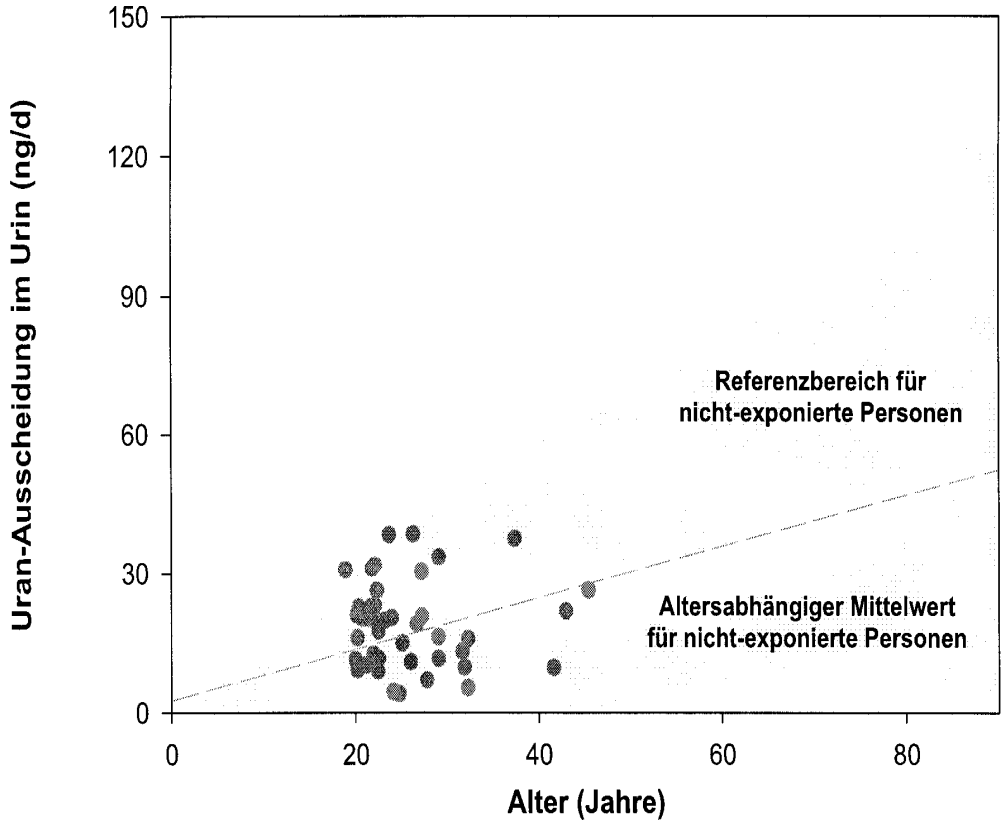

Figure 5b: Urinary uranium excretion in study group 1 (prior to deployment; mean values based on two investigations)	
Original text	Translation
Referenzbereich für nicht-exponierte Personen	Reference range for non-exposed persons
Altersabhängiger Mittelwert für nicht-exponierte Personen	Age-dependent mean for non-exposed persons
Alter (Jahre)	Age (years)
Uran-Ausscheidung im Urin (ng/d)	Urinary uranium excretion (ng/d)

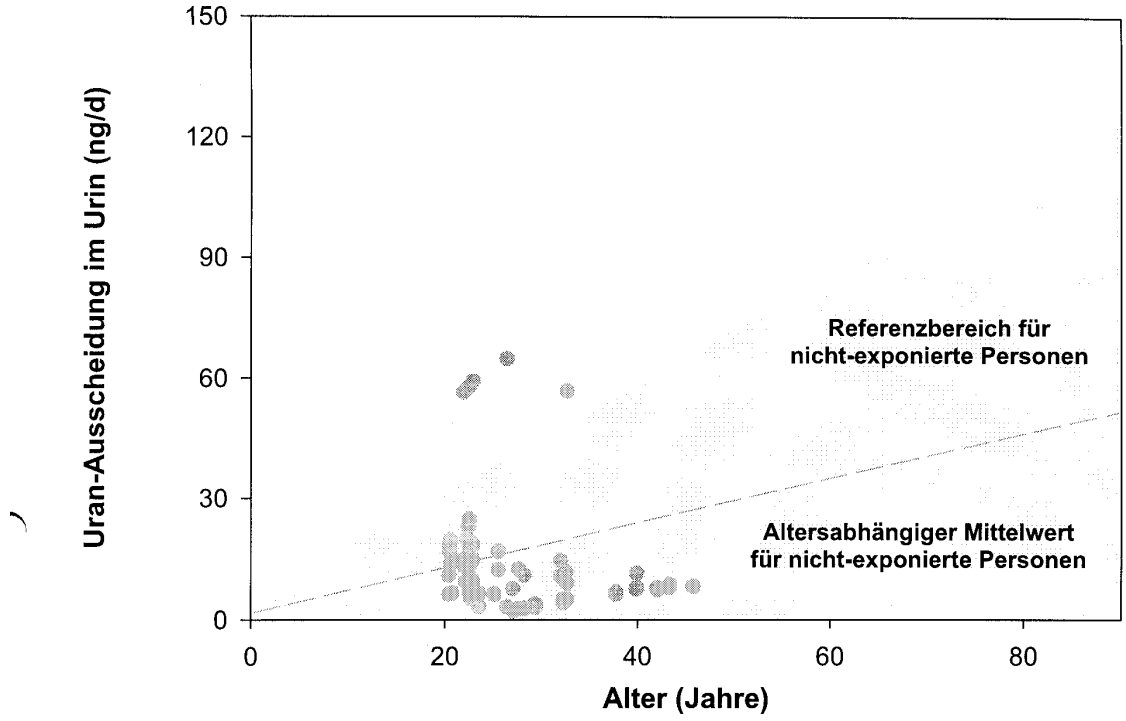

Figure 6a: Urinary uranium excretion in study group 1 (during deployment; results of single measurements)	
Original text	Translation
Referenzbereich für nicht-exponierte Personen	Reference range for non-exposed persons
Altersabhängiger Mittelwert für nicht-exponierte Personen	Age-dependent mean for non-exposed persons
Alter (Jahre)	Age (years)
Uran-Ausscheidung im Urin (ng/d)	Urinary uranium excretion (ng/d)

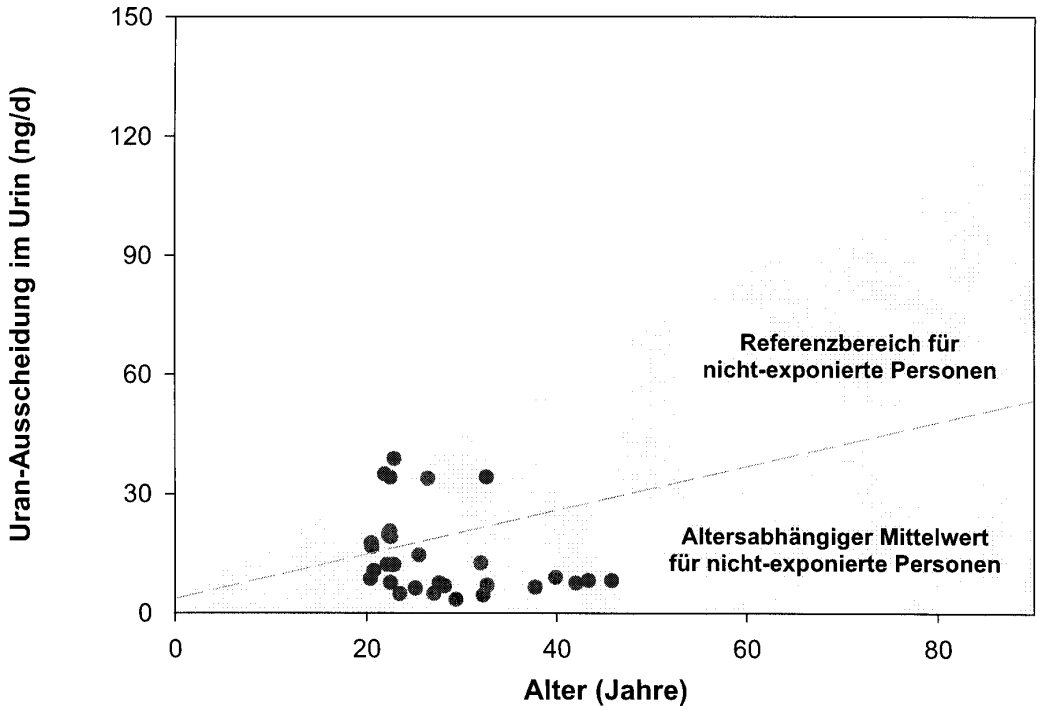

Figure 6b: Urinary uranium excretion in study group 1 (during deployment; mean values based on two investigations)	
Original text	Translation
Referenzbereich für nicht-exponierte Personen	Reference range for non-exposed persons
Altersabhängiger Mittelwert für nicht-exponierte Personen	Age-dependent mean for non-exposed persons
Alter (Jahre)	Age (years)
Uran-Ausscheidung im Urin (ng/d)	Urinary uranium excretion (ng/d)

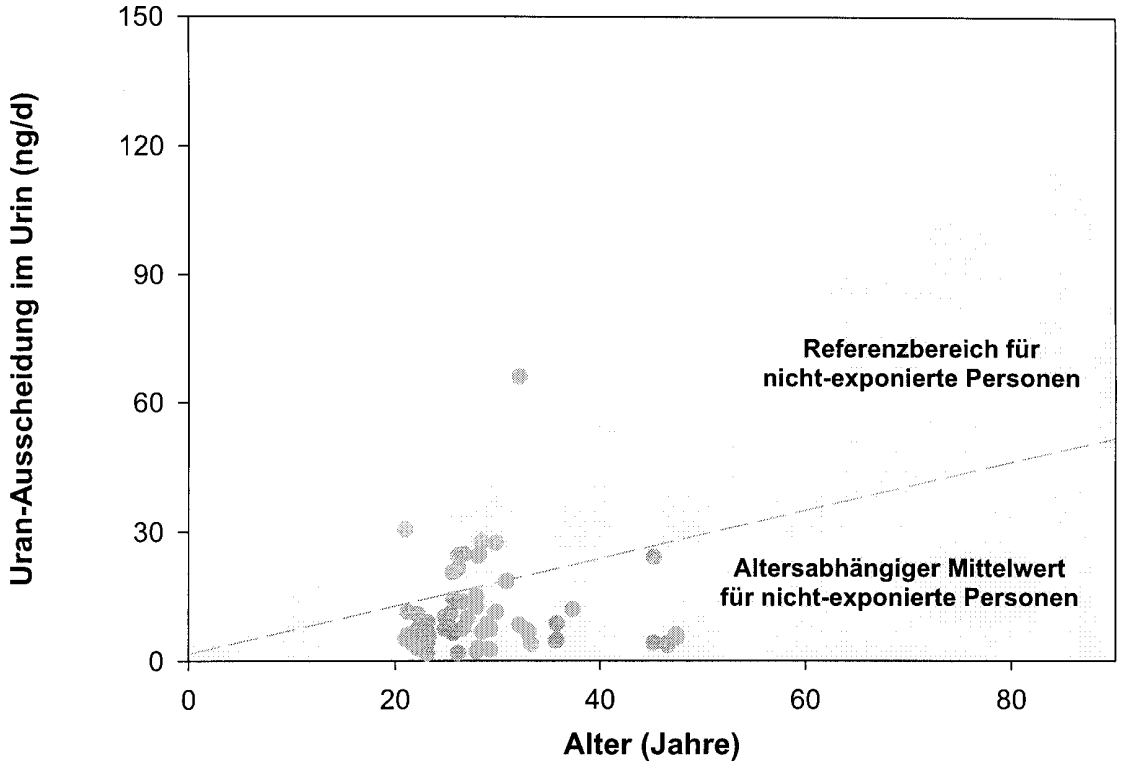

Figure 7a: Urinary uranium excretion in study group 2 (during deployment; results of single measurements)	
Original text	Translation
Referenzbereich für nicht-exponierte Personen	Reference range for non-exposed persons
Altersabhängiger Mittelwert für nicht-exponierte Personen	Age-dependent mean for non-exposed persons
Alter (Jahre)	Age (years)
Uran-Ausscheidung im Urin (ng/d)	Urinary uranium excretion (ng/d)

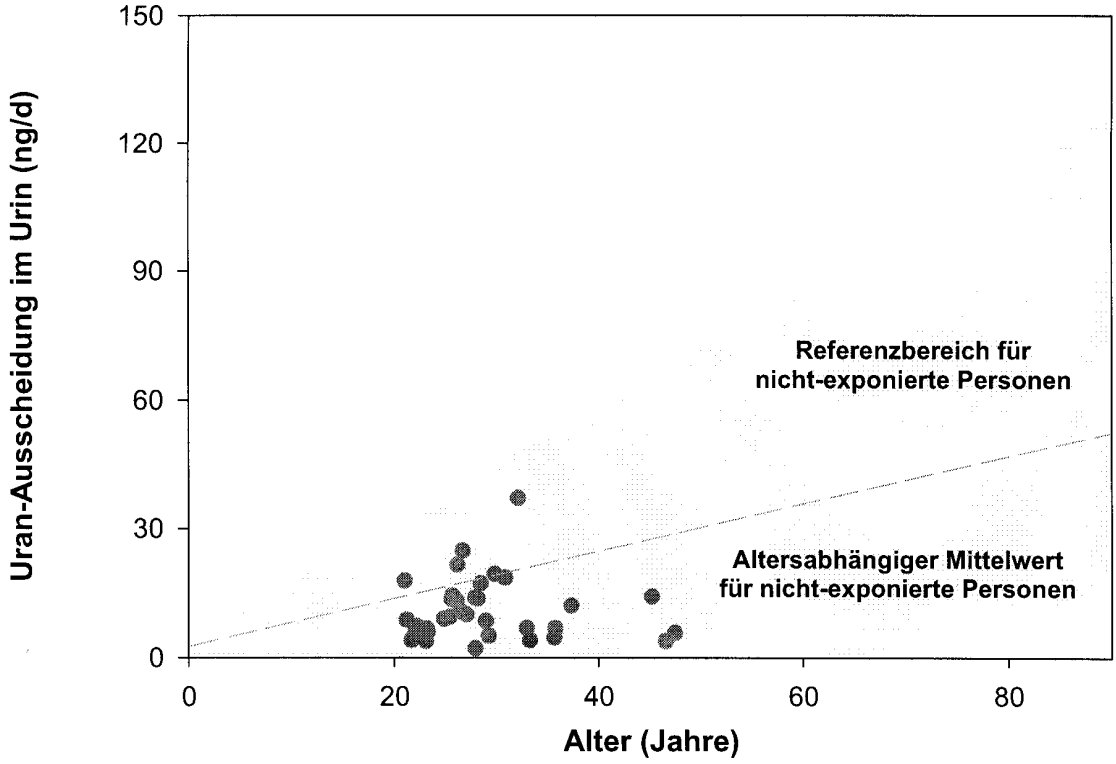

Figure 7b: Urinary uranium excretion in study group 2 (during deployment; mean values based on two investigations)	
Original text	Translation
Referenzbereich für nicht-exponierte Personen	Reference range for non-exposed persons
Altersabhängiger Mittelwert für nicht-exponierte Personen	Age-dependent mean for non-exposed persons
Alter (Jahre)	Age (years)
Uran-Ausscheidung im Urin (ng/d)	Urinary uranium excretion (ng/d)

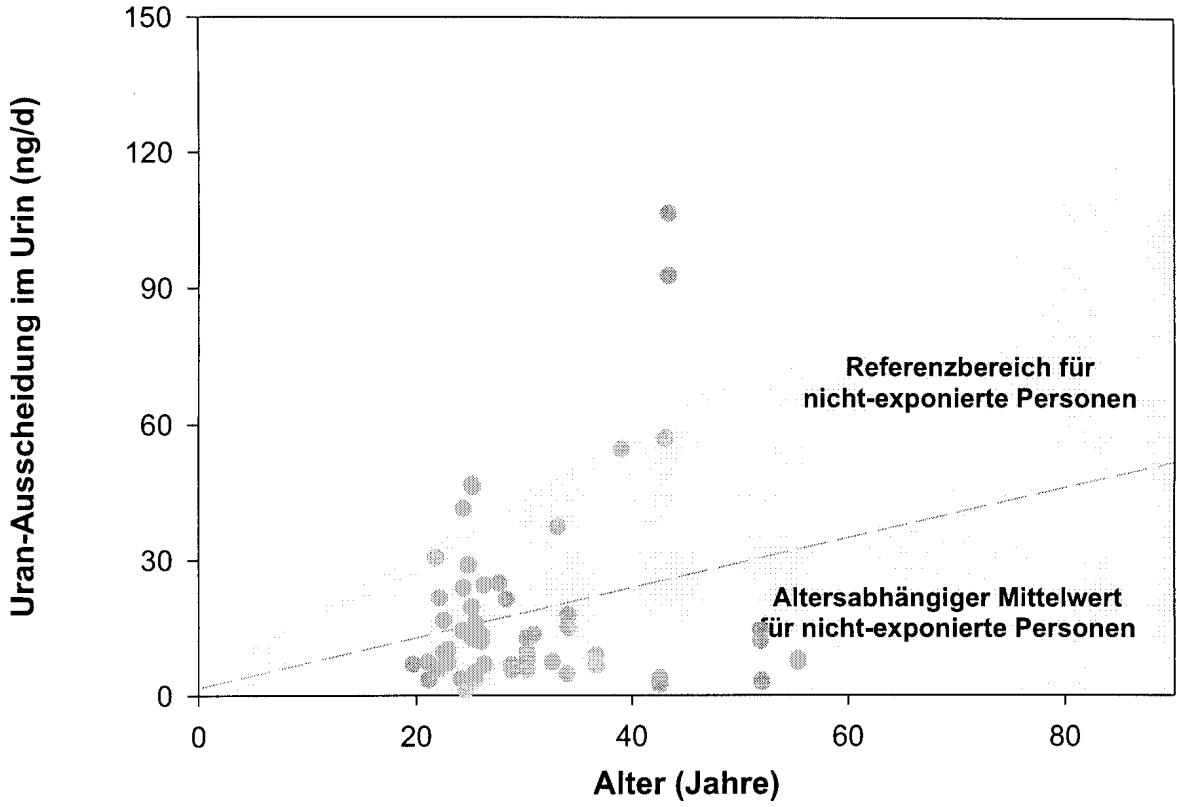

Figure 8a: Urinary uranium excretion in study group 3 (after deployment; results of single measurements)	
Original text	Translation
Referenzbereich für nicht-exponierte Personen	Reference range for non-exposed persons
Altersabhängiger Mittelwert für nicht-exponierte Personen	Age-dependent mean for non-exposed persons
Alter (Jahre)	Age (years)
Uran-Ausscheidung im Urin (ng/d)	Urinary uranium excretion (ng/d)

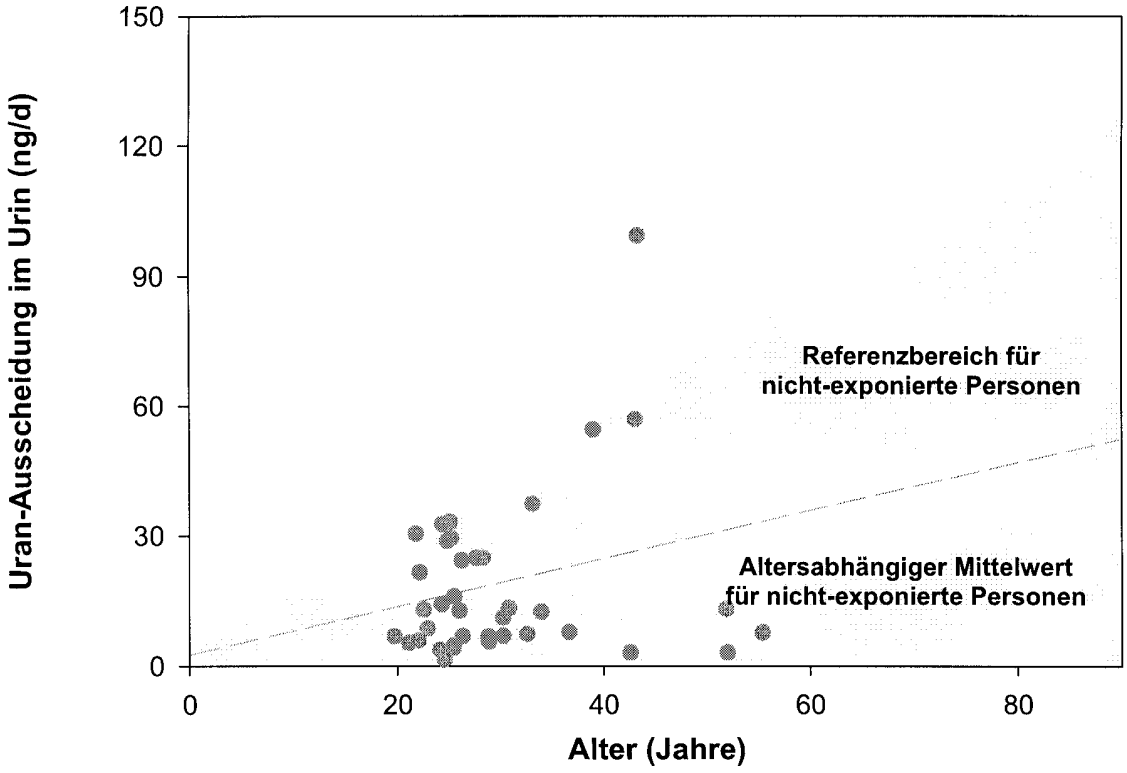

Figure 8b: Urinary uranium excretion in study group 1 (after deployment; mean values based on two investigations)	
Original text	Translation
Referenzbereich für nicht-exponierte Personen	Reference range for non-exposed persons
Altersabhängiger Mittelwert für nicht-exponierte Personen	Age-dependent mean for non-exposed persons
Alter (Jahre)	Age (years)
Uran-Ausscheidung im Urin (ng/d)	Urinary uranium excretion (ng/d)

The results obtained for study group 2 are presented in **Figure 7.** The results of the single measurements are shown in **Figure 7a** and the mean values determined on the basis of the two investigations in **Figure 7b**.

Urine samples from 41 members of **past** contingents **(study group 3),** who had already returned home by the time the study was conducted, have thus far been analyzed for uranium. 20 participants each provided two or three samples for analysis; the other 21 participants only provided one sample each which was sent to the GSF for analysis. The participants from this study group gave urine samples at the request of the responsible unit surgeon. The material needed for the investigations was provided by the GSF. Urine collections were carried out at the

garrisons and in most cases sent by mail or courier to the GSF. The results are presented in **Figure 8** in the same way as the other study group data: **Figure 8a** shows the results of the single measurements and **Figure 8b** the mean values of the two or three investigations.

The analysis of the changes in urinary uranium excretion shown by the participants from study group 1 allows more important conclusions to be drawn than do the results of the single measurements. This is because the analyses of changes are unaffected by differences between individuals and draw attention to the more interesting changes within each individual. For this reason, the results obtained for study group 1 are shown once again in **Figure 9**. The left side of this figure shows the urinary uranium excretion measured before deployment began in Kosovo, and the right side shows the values determined during the stay in Kosovo. The different colors show which of the two subgroups the soldiers belonged to (**red**: higher-risk group, **green**: control group). In addition, this figure includes the age-dependent mean value and reference range. This figure clearly shows that all the values of urinary uranium excretion measured in this group both before and after deployment correspond to the reference range exhibited by non-exposed persons.

A statistical analysis of these values was performed as well. The results are shown in **Table 5**. The mean value of urinary uranium excretion measured prior to deployment was 17.9 ng/d for the higher-risk group and 17.5 ng/d for the control group. The two values are almost identical and correspond to the mean value for non-exposed persons of the same age. During deployment, a mean value of 19.3 ng/d was measured for the higher-risk group and 12.0 ng/d for the control group. The corresponding values for the entire group were 17.7 ng/d before deployment and 15.9 ng/d during deployment. These changes were tested using a paired t-test for samples with unequal variances on the basis of the null hypothesis. The resulting t-values and significance levels for the two-sided test are given in the right columns of Table 5. **No** statistically significant differences in the excretion rates were found either for the entire group or for one of the two subgroups.

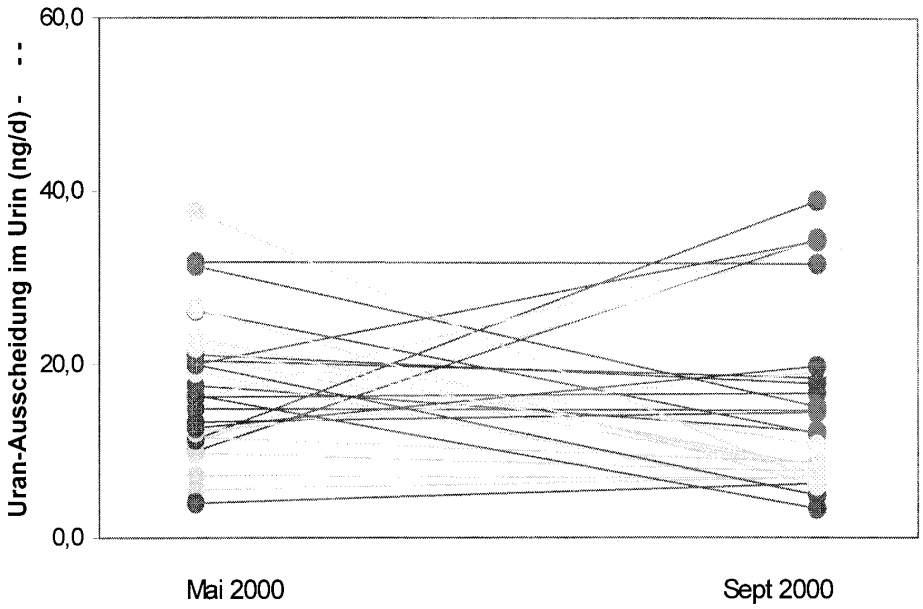

Figure 9: Changes in urinary uranium excretion measured in participants from study group 1: left side: prior to deployment – right side: during deployment red: higher-risk group – green: control group	
Original text	Translation
Uran-Ausscheidung im Urin (ng/d)	Urinary uranium excretion (ng/d)
Referenz-Bereich	Reference range
Mai	May

Table 5: Statistical parameters for urinary uranium excretion (ng/d) in participants from study group 1

	Mean value prior to deployment	SD prior to deployment	Mean value during deployment	SD during deployment	t	p*)
Higher-risk group	**17.9**	**7.4**	**19.3**	**11.5**	**0.407**	**n. s.**
Control group	**17.5**	**8.6**	**12.0**	**9.9**	**1.378**	**n. s.**
total	**17.7**	**7.9**	**15.9**	**11.2**	**0.704**	**n. s.**

*) significance level (n. s. = not significant); SD = statistical standard deviation

The results of the analyses involving participants from study group 3 have been and will be submitted to the responsible unit surgeons who are to inform the soldiers accordingly. The measurement results for the participants from groups 1 and 2 were presented to the Biomonitoring Working Group which has informed these soldiers about the findings.

9. Interpretation of the results

When two or more analyses were conducted, it was confirmed that significant differences in urinary uranium excretion can occur in one and the same individual (see Tables A1-A4). Although in most of these cases the two analyses were conducted on two successive days, measurement results differed widely in some cases. As was explained in section 2, these differences are too great to be attributable to the different types of food consumed by the soldiers. The mechanisms involved have yet to be determined. Additional uranium incorporation in the period between the two analyses, however, can be ruled out for all study groups, not least because the statistical distribution of the values shows differences **in both directions**. These uncertainties can be reduced when a mean value is determined.

As expected, the measurement results obtained for study group 1 **prior to** deployment closely correspond to the reference values for non-exposed persons. **Figure 5a** shows the results of the single measurements. It is not surprising that some data are outside the reference range since the distribution of measurement results for normal persons always includes some extreme "normal" values. As described above, it is possible to reduce the overall variation of measurement results and to correct extreme values by determining mean values on the basis of multiple measurements. **Figure 5b** clearly shows that measurement results correspond even more closely to the reference range when this method is applied. It should be mentioned once again, however, that one and the same person can exhibit significant differences in excretion values (see Figure 3). **Figure 6** shows the results of the analyses performed for study group 1 during deployment. **Figure 6a** gives the results of the single measurements and **Figure 6b** the mean values obtained on the basis of two measurements.

Figure 7 shows the results of the measurements carried out for study group 2 (**during** deployment). **Figure 7a** gives the results of the single measurements and **Figure 7b** the mean values derived from the two measurements. **In no case do these analyses suggest an additional incorporation of DU by any of the persons examined.**

When soldiers from past KFOR contingents (study group 3) were examined, it had to be taken into consideration that the intervals between their deployment in Kosovo and the time of

measurement were different. In some cases, several months had even elapsed. Nevertheless, the significant intakes of DU, especially intakes which may have adverse effects on health, would be expected to result in increased excretion values even after an extended period of time.

The results obtained for this study group are shown in **Figures 8a** and **8b**. **In no case do these measurement results suggest an incorporation by any one of the subjects examined.** Since multiple measurements were performed for only 14 of the 34 participants, variance would be even lower than suggested by Figure 8b if mean values could have been determined for all subjects. The only slightly increased value measured for one soldier was explained by his consumption of a particular brand of mineral water.

Assuming the purely hypothetical scenario that the complete urinary uranium excretion measured after approximately 30 days was a result of the inhalation of highly insoluble uranium particles, a measured value of 30 ng/d, which corresponds to the upper threshold for persons of this age, would result in a committed effective dose of approximately 2 mSv (over the next fifty years). After about one year, i.e. the time that has actually elapsed, this dose would be approximately 0.4 mSv. Compared with a normal lifelong radiation exposure of approximately 2.4 mSv per year, such a dose can be regarded as negligible. A causal relationship between such a dose and the occurrence of malignant diseases can in all probability be ruled out.

Similar conclusions can also be drawn from a different perspective. An additional dose of 1 mSv (the current annual dose limit for members of the general public) from DU incorporation would presuppose the intake values given in Table 6. After 1 day, 30 days or 180 days, these values would lead to the urinary uranium excretion specified in Table 6. A dose of 1 mSv, for example, would presuppose an acute ingestion of approximately 22,000 Bq of ^{238}U or an acute inhalation of 125 Bq of a relatively insoluble ^{238}U compound.

In summary, none of the available analyses for uranium excreted in urine suggests that any the subjects examined had incorporated DU.

Table 6: Intake of ^{238}U which would lead to an additional radiation dose of 1mSv and the corresponding ^{238}U urinary excretion per day to be expected on the basis of these intake levels

Intake pathway	Intake (Bq of ^{238}U)	Urinary ^{238}U excretion per day 1d	30 d	180 d
Inhalation (acute; AMAD = 1µm)				
Solubility class F *(fast)*	**2000**	**360 Bq (= 28.9 mg)**	**1.4 Bq (= 0.11 mg)**	**62 mBq (= 5.0 µg)**
Solubility class M *(moderate)*	**345**	**7.9 Bq (= 0.64 mg)**	**93 mBq (= 7.5 µg)**	**22 mBq (= 1.8 µg)**
Solubility class S *(slow)*	**125**	**17 mBq (= 1.4 µg)**	**0.19 mBq (= 15 ng)**	**83 µBq (= 7 ng)**
Ingestion (acute; f_1 = 0.02)	**22222**	**289 Bq (= 23.2 mg)**	**1.1 Bq (= 86 µg)**	**0.49 Bq (= 39 µg)**
Direct absorption into blood	**500**	**325 Bq (= 26.1 mg)**	**1.1 Bq (= 96 µg)**	**55 mBq (= 4.4 µg)**

10. Summary

This study was conducted to determine whether members of the German Army KFOR Contingent were exposed to health risks associated with the incorporation of depleted uranium (DU) during the time they served in Kosovo. For this purpose, urinary uranium excretion was measured and the results were compared with reference values determined for non-exposed persons. The analyses were performed by means of high-resolution ICP-MS (inductively coupled plasma mass spectrometry).

A considerable number of measurements based on a large group of non-exposed persons provided a reliable basis for evaluating individual measurement results.

The samples used for determining urinary uranium excretion were obtained from members of past and present contingents. A selected study group including approximately 50 soldiers were examined before and approximately two months after deployment began.

A total of 122 soldiers (43 participants from the study group, 38 participants presently serving in Kosovo, 41 participants from earlier contingents) have thus far been examined. In most cases, more than one analysis was performed (usually on successive days).

None of the available analyses for uranium excreted in urine suggests that any of the soldiers examined had incorporated DU.

Acknowledgement

The authors would like to thank Professor Dr. P. Schramel and his staff at the GSF - Institute for Ecological Chemistry for the excellent trace element analyses they conducted.

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