

1 **Calculation of internal dose from ingested soil-derived uranium in humans – Application of a new method**

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16 **CONCISE AND INFORMATIVE TITLE**

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23 **ABSTRACT**

24 The aim of the present study was to determine the internal dose in humans after the ingestion of soil highly
25 contaminated with uranium. Therefore, an *in vitro* solubility assay was performed to estimate the bioaccessibility of
26 uranium for two types of soil. Based on the results, the corresponding bioavailabilities were assessed by using a
27 recently published method. Finally, these bioavailability data were used together with the biokinetic model of
28 uranium to assess the internal doses for a hypothetical but realistic scenario characterized by a daily ingestion of 10
29 mg of soil over 1 year. The investigated soil samples were from two former uranium mining sites of Germany with
30 ²³⁸U concentrations of about 460 mg/kg and 550 mg/kg. For these soils, the bioavailabilities of ²³⁸U were quantified
31 as 0.18% and 0.28% (geometric mean) with 2.5th percentiles of 0.02% and 0.03%, and 97.5th percentiles of 1.48%
32 and 2.34%, respectively. The corresponding calculated annual committed effective doses for the assumed scenario
33 were 0.4 μSv and 0.6 μSv (GM) with 2.5th percentiles of 0.2 μSv and 0.3 μSv, and 97.5th percentiles of 1.6 μSv and
34 3.0 μSv, respectively. These annual committed effective doses are similar to those from natural uranium intake by
35 food and drinking water, which is estimated to be 0.5 μSv. Based on the present experimental data and the selected
36 ingestion scenario, the investigated soils - although highly contaminated with uranium - are not expected to pose any
37 major health risk to humans related to radiation.

38 INTRODUCTION

39 Uranium is the heaviest naturally occurring element. It occurs ubiquitously in soils at concentrations of about 3
40 mg/kg (Bleise et al. 2003) and comprises three isotopes with percentages by mole fraction of 0.0054% (^{234}U), 0.72%
41 (^{235}U) and 99.27% (^{238}U) (Berglund and Wieser 2011). All three isotopes are alpha emitters with half-lives of
42 245,500 years (^{234}U), 704,000,000 years (^{235}U) and 4,468,000,000 years (^{238}U) (ICRP 2008), respectively. The
43 corresponding percentages by radioactivity of naturally occurring uranium are about 49.2% (^{234}U), 2.2% (^{235}U) and
44 48.6% (^{238}U), respectively (Mkandawire 2013).

45 Elevated uranium concentrations in soils are mostly of anthropogenic nature. In agriculture, for example, uranium-
46 contaminated phosphate fertilizers are the main source of uranium contamination of soils. About 14,000 tons of
47 uranium were deposited between 1951 and 2011 on agricultural land in Germany, which equals about 1 kg of
48 uranium per hectare (Schnug and Lottermoser 2013). Uranium mining is another source of potential uranium
49 contamination of soils (Brugge and Buchner 2011). The global uranium production has increased from about 36,000
50 tons in 2002 to 60,000 tons in 2013, whereby the top five uranium producers in 2013 were Kazakhstan, Canada,
51 Australia, Niger, and Namibia (WNA 2014). Even for remediated former uranium mining sites elevated uranium
52 concentrations are reported, since these sites are re-contaminated due to natural processes like capillary rise of
53 contaminated ground water (Langella et al. 2014). A third notable source of environmental uranium contamination is
54 by the military use of depleted uranium (DU) penetrators, leading to DU dust formation after impact (Bleise et al.
55 2003).

56 The unintended ingestion of small amounts of soils by humans via various routes is observed all over the world
57 (Abrahams 2002, Sing and Sing 2010) Thereby, the average ingestion rate of soil by adults is assumed to be about
58 10 mg/day (Stanek et al. 1997). Since uranium ubiquitously occurs in soil, soil ingestion is always accompanied by
59 the ingestion of uranium. To estimate the resulting internal dose, the bioavailability (f_1) of soil-derived uranium has
60 to be assessed. The bioavailability (f_1) is the fraction of uranium which is absorbed from the human alimentary tract
61 into the circulatory system. In practice, the bioavailability of uranium from highly contaminated soils is not directly
62 assessed by human soil ingestion studies, but is indirectly assessed by *in vitro* solubility assays. However, by these
63 assays only the bioaccessibility (DF) of soil-derived uranium can directly be estimated. The bioaccessibility (DF)
64 quantifies the fraction of soil-derived uranium in human alimentary tract, which is soluble and therefore potentially

65 available for absorption. Consequently, the bioavailability is usually estimated based on bioaccessibility data.
66 However, there are different solubility assays and different estimation methods described in the literature which can
67 lead to varying estimated bioavailabilities for one particular soil of up to three orders of magnitude (Träber et al.
68 2014). In response to that, Träber et al. 2014 recently reported a solubility assay-specific factor (f_A^{sol}) (Fig. 1), which
69 was based on a human study and by which more reliable data on the bioavailability of soil-derived uranium can be
70 deduced from the bioaccessibility data. Using this method, only a solubility assay has to be performed e.g. for a
71 highly uranium-contaminated soil to receive more reliable data on its bioavailability.

72 The aim of the present study was to estimate the internal dose in humans after a potential ingestion of soils highly
73 contaminated with uranium by applying the recently published method (Träber et al. 2014). Two types of soil highly
74 contaminated with uranium and additionally one pure fertilizer were investigated. Thereby more reliable data on the
75 uptake of uranium in humans were obtained from highly contaminated soils than previously available.
76 Consequently, more reliable data on the internal dose enhancement can be obtained for the risk assessment of
77 potential ingestion scenarios.

78 MATERIALS AND METHODS

79 Samples

80 In the present study, two types of soil and additionally one fertilizer were analyzed. The soil sample “Gauern” was
81 selected from a former uranium mining site in the east of Thuringia, Germany. It was taken from the surface (0-10
82 cm) of a hot spot, a supposed former ore terminal, near the former heap “Gauernhalde”. The soil sample “E1” was
83 taken from a heap of a former uranium mining site (Coschütz/Gittersee) near Dresden in Saxony, Germany. Both
84 soil samples were sieved at 2 mm. The fertilizer “Blaukorn NovaTec“ (COMPO Gesellschaft GmbH & Co. KG,
85 Germany), with an indicated mass fraction of P₂O₅ of 7%, was bought at retail.

86 For the analyses of total soil-derived ²³⁸U 250.0 mg of each soil was mixed with 1.5 mL of HNO₃ (65%), 4.5 mL of
87 HCl (30%) and 1 mL HF (40%). The mixture was digested in a Multiwave 3000 microwave device (Anton Paar,
88 Austria); power: ramp for 5 min up to 1400 W, hold for 30 min at 1400 W and cooling down for 20 min. Thereafter
89 6 mL of H₃BO₃ were added to neutralize free fluorides and the solution was placed a second time in the Multiwave
90 system; power: ramp for 5 min up to 1400 W, hold for 15 min and cooling down for 15 min. For the analyses of
91 total fertilizer-derived ²³⁸U 118.1 mg fertilizer was mixed with 1.0 mL of HNO₃ (65%) and heated at 160 °C
92 overnight under pressure (Schramel et al. 1980). All solutions were stored at 4 °C until measurement of ²³⁸U by
93 using inductively coupled plasma mass spectrometry (ICP-MS, see below).

94 Determination of bioaccessibility (DF) and bioavailability (f_A)

95 The bioaccessibility (DF) of the soil-derived ²³⁸U and of the fertilizer-derived ²³⁸U in the relevant part of the
96 alimentary tract, which is the intestine (Frelon et al. 2007), was estimated by an *in vitro* solubility assay. In
97 accordance to the previous study, the same *in vitro* solubility assay based on the German DIN 19738 (DIN 2000)
98 was performed; the assay is described in detail elsewhere (Träber et al. 2014).

99 Briefly, 2 g of soil or fertilizer was incubated under physiological conditions, using an artificial gastric fluid with a
100 pH of 2 followed by the addition of an artificial intestinal fluid with a pH of 7.5. After 8 h of incubation an aliquot
101 was withdrawn, centrifuged at 5000 rpm (Hettich Universal 32R) and filtered at 0.2 µm (sterile filter, Millipore). All
102 experiments were repeated three times independently. The solutions were stored at 4 °C until measurement of ²³⁸U
103 using ICP-MS (see below).

104 The bioaccessibility (DF) was calculated as the percentage of soluble ^{238}U based on the total concentration of soil-
105 derived ^{238}U or fertilizer-derived ^{238}U .

106 The sample-specific bioavailabilities were calculated for the two soil samples and the fertilizer by the previously
107 published relation Eq. (1) (Höllriegl et al. 2010).

$$108 \quad f_A = f_A^{\text{sol}} \text{DF} \quad (1)$$

109 Note that for the current study the bioavailability is denoted as f_A , since the notation of the bioavailability has
110 changed by the International Commission on Radiological Protection (ICRP) from f_1 to f_A (ICRP 2006). DF was
111 derived from the applied solubility assay whereas the f_A^{sol} factor was directly adopted from the previous work being
112 0.53% (geometric mean, GM) and ranging from 0.06% (2.5th percentile) to 4.43% (97.5th percentile) (Träber et al.
113 2014). The f_A^{sol} factor quantifies the fraction of bioaccessible uranium which is absorbed into the circulatory system.
114 It is emphasized here again that the data on f_A^{sol} are based on human data.

115 **Measurement of ^{238}U by ICP-MS**

116 For the analysis of ^{238}U a NexIon ICP-MS instrument (Perkin-Elmer, Rodgau-Jügesheim, Germany) in standard
117 mode was used. The samples of the solubility assay were diluted between 1:2 and 1:100 with diluted nitric acid (5%,
118 final concentration). The samples of the microwave-assisted digested soils were diluted 1:2 with diluted nitric acid
119 (3 %, final concentration). An internal standard solution (1 $\mu\text{g/L}$ ^{193}Ir , final concentration) was added to each sample
120 to correct for matrix interferences. For each sample three replicates were measured. Sample transport to nebulizer
121 was realized by a peristaltic pump at a flow rate of 0.5 mL/min. Sample introduction to ICP-MS was performed by a
122 Meinhard nebulizer fitting into a cyclone spray chamber. A uranium stock standard solution of 1 g/L purchased and
123 certified by SPEX (USA) was used for calibration. Uranium was determined at $m/z = 238$. RF power was 1250 W,
124 nebulizer gas (Ar) was daily optimized and usually set to 0.92 L/min. Plasma gas: Ar, 15 L/min. Auxiliary gas: 0.8
125 L/min, dwell time 300 ms, 3 readings per replicate. The instrument was calibrated using a 7-point calibration
126 between blank and 2000 ng/L. After ten measurements regularly three blank determinations and a control
127 determination of a certified standard were performed. Calculation of results was carried out on a computerized lab-
128 data management system, relating the sample measurements to calibration curves, blank determinations and control

129 standards. The detection limit, calculated as blank + 3 times the blank standard deviation (SD) was 1.5 ng/L, the
130 limit of quantification (LOQ) calculated as blank + 10 x SD was 4.5 ng/L.

131 **Biokinetic model**

132 To model the biokinetics of ingested soil-derived ^{238}U , the systemic model for uranium (ICRP 1995a) and the human
133 alimentary tract model (HATM) (ICRP 2006) were coupled. These two models were connected by the alimentary
134 tract transfer rate, which was quantified in the present study for two soils and one fertilizer. For internal dose
135 assessment of ^{238}U , the radiologically relevant progeny ^{234}Th , ^{234}Pa , and $^{234\text{m}}\text{Pa}$ were also taken into account (ICRP
136 1979). Similar to the parent ^{238}U , the systemic models of thorium, protactinium and protactinium (meta) as decay
137 products, which were published by ICRP in Publication 71 in Annex C (ICRP 1995b), were also coupled to the
138 human alimentary tract model. Thereby, each systemic model of a progeny was connected to one human alimentary
139 tract model. The corresponding alimentary tract transfer rates were adopted from ICRP Publication 100 (ICRP
140 2006). The resulting four ingestion models (Fig. 2) were interconnected in accordance with the ^{238}U decay series by
141 the corresponding decay constants (ICRP 2008).

142 As the biokinetic models of different radionuclides are independent, their transfer rates and especially their
143 compartment structures are not necessarily identical. For a proper interconnection of biokinetic models with varying
144 compartment structures, like the biokinetic model of uranium and the biokinetic model of thorium as a progeny, two
145 approaches are proposed by ICRP (ICRP 1995b). By the first approach the biokinetics of a radionuclide of a chain
146 are calculated by using the biokinetic descriptions given by ICRP (ICRP 1995b). Thereafter, necessary, non-existing
147 compartments representing source regions receive a portion of nuclear transformations which are partitioned by
148 mass fraction from the so-called “Other” tissue. This “Other” tissue represents all systemic tissues, which are not
149 explicitly specified in a biokinetic model. In the present work, however, the second approach was applied because
150 this approach will be adopted by the forthcoming ICRP Publications on “Occupational Intakes of Radionuclides,
151 Part 1”. By this approach, prior to biokinetic modeling, the biokinetic model of a radionuclide of a chain is expanded
152 for the necessary, non-existing compartments and transfer rates, respectively. In the present work, only the
153 biokinetic model for uranium had to be expanded for the compartments gonads, cortical marrow, and trabecular
154 marrow, to match with its progeny biokinetic model of thorium. The structures and transfer rates of the protactinium

155 model and the protactinium (meta) model were assigned to the biokinetic model of thorium (ICRP 1995a, ICRP
156 1995b).

157 As an example, for the biokinetic model of ^{238}U the transfer rate from the blood compartment to the newly created
158 cortical marrow compartment is calculated from the corresponding transfer rate of the so-called “Other” tissue
159 compartment by its mass-fraction. The transfer rate from the blood compartment to the “Other” tissue compartment
160 is reduced accordingly. Since the uranium model contains three soft tissue compartments with different transfer
161 rates, three new cortical marrow compartments were integrated into the uranium model. As a part of a decay series,
162 all three cortical marrow compartments were connected to the single cortical marrow compartment of thorium by
163 their decay constant. Finally, nine additional compartments were integrated into the biokinetic model of the parent
164 radionuclide ^{238}U .

165 Beside the transfer rates of the systemic model, the transfer rates for “total diet” of the HATM model were adopted
166 from ICRP (ICRP 2006) for male and female, which resulted in two sex-specific biokinetic models for ^{238}U . In
167 addition, sex-specific biokinetic models for ^{234}U and ^{235}U with their corresponding progeny were implemented.
168 Thereby the radiologically relevant progeny of ^{235}U is only ^{231}Th , whereas ^{234}U has no progeny with relevant
169 dosimetric contribution (ICRP 1979).

170 With these six models, the sex-specific biokinetics of the three naturally occurring isotopes of uranium and their
171 progeny are described by a system of first-order linear ordinary differential equations, which were numerically
172 solved by using the commercially available software SAAM II (Barrett et al. 1998) (The Epsilon Group VA, USA).
173 For internal dose assessment of adults, the integrated activity of the ingested uranium and its progeny in all
174 compartments over a 50-year period was calculated.

175 **Calculation of the committed effective dose**

176 The committed equivalent dose (H_T) and the committed effective dose (E) were calculated based on the time-
177 integrated activity (\tilde{A}) in so-called source regions (r_S) and radiation-weighted factors (S_w) and the appropriate tissue-
178 weighting factors (w_T) (Bolch et al. 2009, ICRP 1989). In the present calculation, only adults were considered for
179 the internal dose calculation because only the f_A^{sol} value for adults was established (Träber et al. 2014).

180 The committed equivalent dose ($H_{T,sex}$) for female and male was calculated by Eq. (2) as the sum of a radionuclide
181 and its progeny (N):

$$182 \quad H_{T,sex} = \sum_N \sum_{r_S} \tilde{A}(r_S, T_{50}, sex, N) S_w(r_T \leftarrow r_S, sex, N) \quad (2)$$

183 Where $\tilde{A}(r_S, T_{50}, sex, N)$ is the cumulated activity (\tilde{A}) of a radionuclide or progeny (N) in a source region (r_S) over
184 50 years (T_{50}), which is sex-specific (sex); \tilde{A} was calculated by the biokinetic models as described above.

185 $S_w(r_T \leftarrow r_S, sex, N)$ is the radiation-weighted S factor calculated for a radionuclide or progeny for both sexes (sex)
186 by Eq. (3).

$$187 \quad S_w(r_T \leftarrow r_S, sex, N) = \sum_R w_R S(r_T \leftarrow r_S, E_R, sex, N) \quad (3)$$

188 Where w_R is the radiation weighting factor and $S(r_T \leftarrow r_S, E_R, sex, N)$ is the specific energy of a radiation type R
189 (E_R), which is absorbed in a target organ (r_T) emitted from a source region (r_S), per nuclear transformation of a
190 radionuclide or its progeny (N). S_w was calculated as the sum of all radiation types per nuclear transformation of a
191 radionuclide or its progeny (N) by using the SEECAL program (Oak Ridge National Laboratory, Oak Ridge, TN,
192 USA). Since S_w factors are not yet available for a few organs like the prostate, the “splitting rule” in the treatment
193 for remainder tissues was applied in the current work as recommended in ICRP Publication 60 (ICRP 1991).
194 Accordingly, the appropriate radiation weighting factors (w_R) and tissue weighting factors (w_T) were adopted from
195 ICRP Publication 60 (ICRP 1991).

196 Finally the committed effective dose (E) was calculated by Eq. (4) by averaging the effective dose of male and
197 female (ICRP 2007):

$$198 \quad E = \sum_T w_T \left(\frac{H_{T,male} + H_{T,female}}{2} \right) \quad (4)$$

199 **Dose calculation for ingestion scenarios**

200 By the introduced committed effective dose calculation (see above), sample-specific ingestion effective dose
201 coefficients were assessed by adopting the corresponding sample-specific alimentary tract transfer rates to the
202 biokinetic models and assuming a single uptake of 1 Bq of ^{234}U , ^{235}U or ^{238}U .

203 Based on the sample-specific ingestion effective dose coefficients the committed effective dose can be simply
204 obtained for different ingestion scenarios by Eq. (5) (Simon 1998).

$$205 \quad D_{soil} = \sum_i C_{soil,i} \times I_{soil} \times ED \times DC_i \quad (5)$$

206 D_{soil} committed effective dose from soil-derived radionuclides (Sv)

207 $C_{soil,i}$ average concentration of radionuclide i in soil (Bq/g)

208 I_{soil} average daily ingestion of soil during the exposure period (g/day)

209 ED exposure duration (d)

210 DC_i ingestion effective dose coefficients of radionuclide i (Sv/Bq)

211 **RESULTS AND DISCUSSION**

212 **Concentration of ^{238}U in samples**

213 The concentration of ^{238}U in the three samples “Gauern”, “E1” and “Fertilizer” was determined (see Table 1). The
214 soil samples “Gauern” and “E1” revealed elevated concentrations for ^{238}U of about two orders of magnitude
215 compared to the average concentration of ^{238}U in soils of about 3 mg/kg (Bleise et al. 2003).

216 **Bioavailability (f_A) of soil and fertilizer samples**

217 To calculate the sample-specific f_A values for ^{238}U , first the bioaccessibilities (DF) for ^{238}U of all three samples were
218 determined by the mentioned solubility assay. The results are given in Fig. 3 based on the corresponding total
219 concentrations of ^{238}U (Table 1).

220 The sample “Fertilizer” showed the lowest bioaccessibility for ^{238}U of about 24%, while the two soil samples
221 revealed higher bioaccessibilities of about 33% and 53%, respectively. In comparison, the bioaccessibility of the
222 previously examined healing soil with a uranium concentration of about 2.6 mg/kg was below 10% (Träber et al.
223 2014). The different bioaccessibilities among the healing soil and the here investigated soil samples might be a
224 result of the different mining processes. Whereas healing soil is a pure natural product, soils from uranium mining
225 sites are intensively chemically processed (leaching) to dissolve more uranium. This might also increase the
226 bioaccessibility of uranium of these processed soils. Apart from that, different particle sizes of soil samples may also
227 explain different bioaccessibilities; for samples with similar uranium concentrations, smaller particle sizes are
228 accompanied by a larger total surface by which more uranium is accessible for dissolution (Jovanovic et al. 2012).

229 Based on the determined bioaccessibilities (DF), the sample-specific bioavailabilities (f_A) were calculated by Eq.
230 (1). The results are given in Table 2 and reveal bioavailabilities between 0.13% and 0.28% (GM). These data are
231 similar to ICRP data, by which a bioavailability of uranium of 0.2% for relatively insoluble compounds is assumed
232 (ICRP 2006). In Table 2 the 2.5th percentile and the 97.5th percentile of the bioavailabilities are also given to cover a
233 95% confidence interval.

234 **Committed effective doses**

235 The committed effective dose was estimated by Eq. (5) for all three samples, for a conceivable exposure scenario by
236 which 10 mg of soil or fertilizer are daily ingested over one year. For that, besides the average daily ingestion (I_{soil})
237 of soil or fertilizer and the exposure duration (ED), the sample-specific ingestion dose coefficients (DC_i) of the
238 radionuclides ^{234}U , ^{235}U , and ^{238}U are needed. Therefore, the sample-specific alimentary tract transfer rates (Table 3)
239 were calculated from the sample-specific bioavailability data (Table 2) (ICRP 1997) and applied to the used
240 biokinetic models. The resulting sample-specific ingestion effective dose coefficients (DC_i) of the radionuclides
241 ^{234}U , ^{235}U , and ^{238}U are given in Table 4. For the dose calculation using Eq. 5, the data on the sample-specific
242 average concentrations ($c_{soil,i}$) of the radionuclides ^{234}U , ^{235}U and ^{238}U are also needed. The sample-specific activities
243 of ^{238}U are based on our measurements (Table 1) whereas the proportional sample-specific activities of ^{234}U and ^{235}U
244 are based on literature data (Berglund and Wieser 2011). The resulting sample-specific average concentrations
245 ($c_{soil,i}$) of the radionuclides ^{234}U , ^{235}U and ^{238}U are given in Table 5.

246 Our assumption of 10 mg of soil or fertilizer that are daily ingested over 1 year is assumed to be a realistic worst
247 case scenario. The investigated uranium-contaminated soil sample E1, for example, was from a heap of a former
248 uranium mining site nearby the city of Dresden, Germany. In the worst case scenario, the whole amount of daily
249 ingested soil or fertilizer (10 mg) is assumed to be from a uranium or phosphate mining site.

250 The sum of the calculated sample-specific annual committed effective doses of the isotopes ^{234}U , ^{235}U and ^{238}U and
251 their radiologically relevant progeny are given in Table 6. The soil sample “Gauern” revealed the highest total
252 concentration of uranium (553 mg/kg, Table 1) as well as the highest bioaccessibility of uranium (53%, Fig. 3) and
253 therefore the highest annual committed effective dose among all samples, with about 0.6 μSv (GM) ranging from
254 0.3 μSv (2.5th percentile) to 3.0 μSv (97.5th percentile). Besides, a daily ingestion of 10 mg of the soil sample
255 “Gauern” would equal a daily ingestion of 5.57 μg of uranium. These results are similar to those from the daily
256 intake of 1.25 μg uranium by food and drinking water, which is estimated to be 0.5 μSv for adults (UNSCEAR
257 2000). Furthermore, the calculated annual committed effective dose of about 3.0 μSv (97.5th percentile) for the
258 assumed scenario is about three orders of magnitude lower than the average annual natural background effective
259 dose of 2.4 mSv (UNSCEAR 2008).

260 The present results are not appropriate to be applied to children who are expected to exhibit a two- to tenfold
261 increased soil ingestion rate compared to adults (Stanek et al. 2012, UNSCEAR 2013). As reported by ICRP (ICRP

262 1995a), the committed effective dose coefficients of uranium for children are 1.5 to 2.7 times greater than that of
263 adults; this increase is based on an assumed bioavailability of uranium of 2% for adults and children. Moreover, a
264 2.4-fold increase of the bioavailability of uranium can be concluded from recent data for children aged between 1
265 and 7 years compared to adults (Chen et al. 2011). Therefore, an increased effective dose of about one to two orders
266 of magnitude might be considered for children.

267 **Quality assurance of dose calculations**

268 The calculated effective dose coefficients of the ingested naturally occurring isotopes ^{234}U , ^{235}U and ^{238}U , and their
269 radiologically relevant progeny, were compared with the effective dose coefficients given by ICRP (ICRP 1995a)
270 (Table 7), based on an exemplary intake of 1 Bq of ^{234}U , ^{235}U or ^{238}U and an alimentary tract transfer factor for
271 uranium of 2% (ICRP 1995a). As reported by ICRP, the difference of both approaches for treatment of decay
272 products in the dose calculation are less than 5% (ICRP 1995b). From Table 7 it is evident that the effective doses
273 calculated in the present work are not more than 4% different, for all three isotopes, from those given by ICRP. It is
274 implied that the present method of dose calculation is consistent with that proposed by ICRP.

275 **CONCLUSION**

276 Based on the experimental data and the assumption of a daily soil or fertilizer ingestion of 10 mg over 1 year,
277 neither the uranium-contaminated fertilizer nor the investigated highly uranium-contaminated soils are expected to
278 pose any major health risk to humans related to radiation. It is worth to note that the present results are based on
279 values for the f_A^{sol} factor, which were derived from a study on healthy volunteers aged between 22 and 55 years
280 (Träber et al. 2014). Therefore, the low health risk refers only to adults and not to children who are expected to
281 exhibit an increased soil ingestion rate and a higher bioavailability for uranium as well as a higher committed
282 effective dose coefficient.

283

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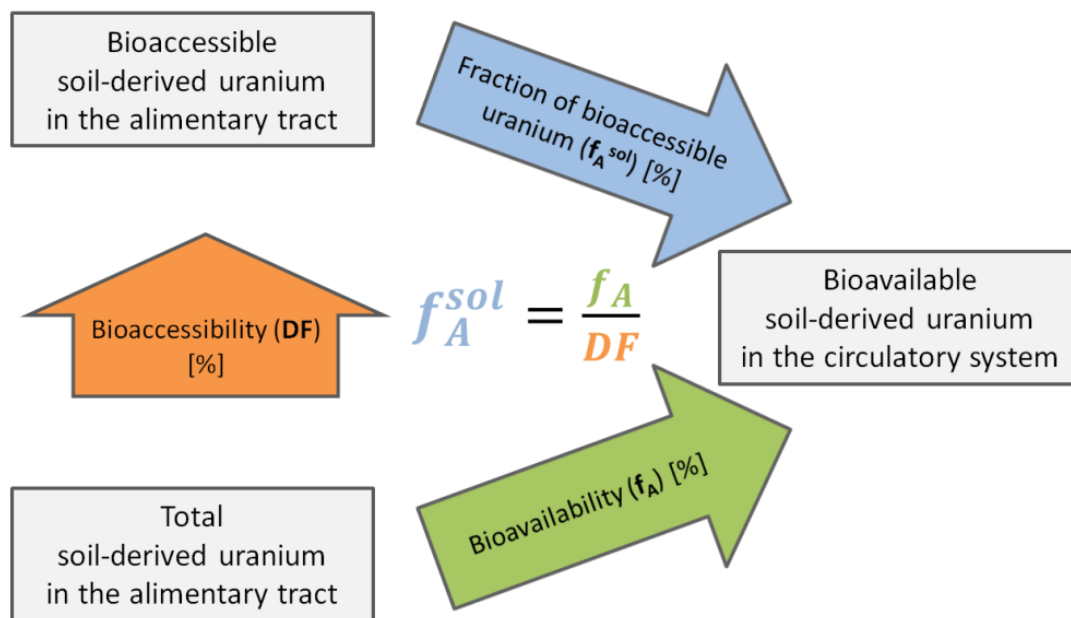
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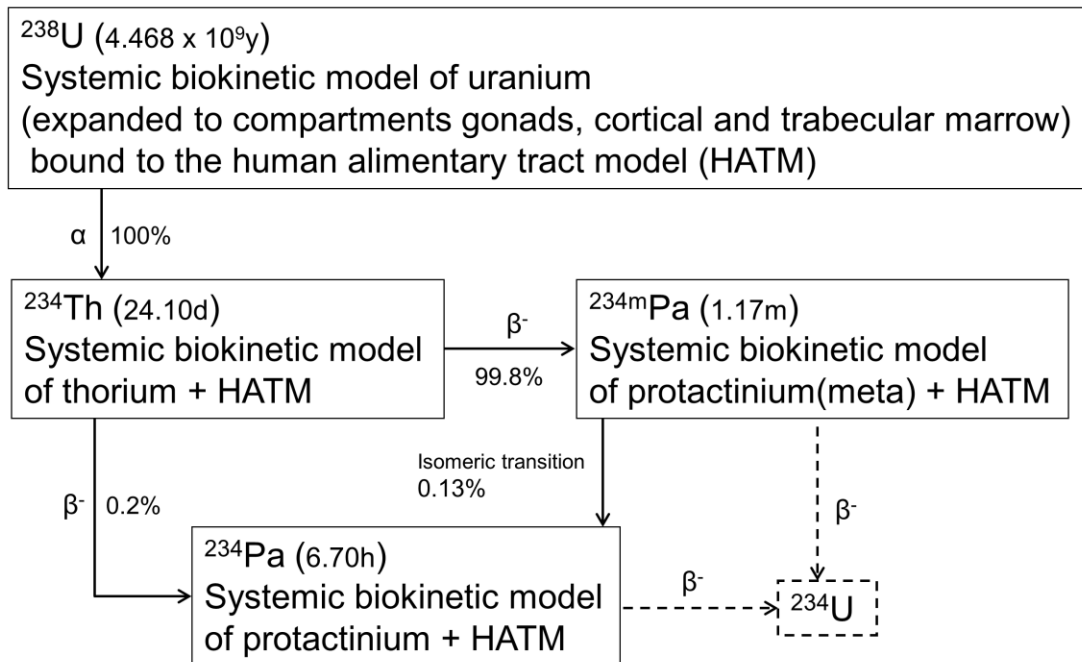
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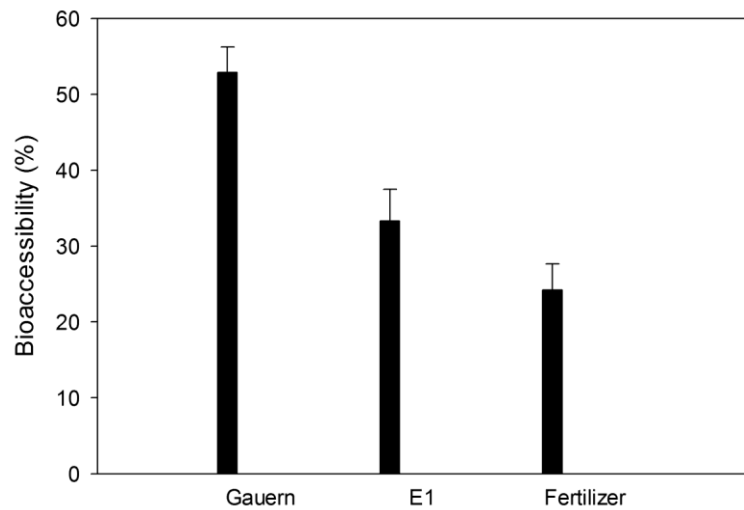


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356 **Fig 1** Scheme of the relation of bioavailability (f_A), bioaccessibility (DF) and the f_A^{sol} factor. The figure is
 357 reprinted (adapted) with permission from Träber SC, Höllriegl V, Li WB, Czeslik U, Rühm W, Oeh U,
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360
 361 **Fig 2** Interconnection of the biokinetic models (for ingestion) of uranium and its radiologically relevant progeny
 362 ²³⁴Th, ^{234m}Pa and ²³⁴Pa (half-lives given in brackets)



363

364 **Fig 3** Bioaccessibility (DF) of soil-derived (Gauern, E1) and fertilizer-derived (Fertilizer) ^{238}U in artificial
365 gastrointestinal fluid (mean \pm SD, n=3)

366 **Table 1** Concentration of ^{238}U in soil samples (Gauern, E1) and fertilizer

	Total concentration of ^{238}U (mean \pm SD) in mg/kg
Gauern	553 \pm 9
E1	456 \pm 3
Fertilizer	23.3 \pm 0.5

367 SD - standard deviation of three measurements per sample

368 **Table 2** Sample-specific bioavailabilities f_A of ^{238}U

	GM (%) ^A	P _{2.5th} (%) ^B	P _{97.5th} (%) ^C
Gauern	0.28	0.03	2.34
E1	0.18	0.02	1.48
Fertilizer	0.13	0.01	1.07

369 ^AGeometric mean, ^B2.5th percentile, ^C97.5th percentile

370 **Table 3** Sample-specific alimentary tract transfer rates

	GM (d ⁻¹) ^A	P _{2.5th} (d ⁻¹) ^B	P _{97.5th} (d ⁻¹) ^C
Gauern	1.69×10 ⁻²	1.90×10 ⁻³	1.44×10 ⁻¹
E1	1.06×10 ⁻²	1.20×10 ⁻³	8.99×10 ⁻²
Fertilizer	7.70×10 ⁻³	8.70×10 ⁻⁴	6.49×10 ⁻²

371 ^AGeometric mean, ^B2.5th percentile, ^C97.5th percentile

372 **Table 4** Sample-specific committed effective dose coefficients (ingestion)

	GM (Sv/Bq) ^A	P _{2.5th} (Sv/Bq) ^B	P _{97.5th} (Sv/Bq) ^C
Guern			
²³⁴ U	1.21×10 ⁻⁸	6.30×10 ⁻⁹	6.02×10 ⁻⁸
²³⁵ U	1.15×10 ⁻⁸	6.18×10 ⁻⁹	5.61×10 ⁻⁸
²³⁸ U	1.09×10 ⁻⁸	5.73×10 ⁻⁹	5.41×10 ⁻⁸
E1			
²³⁴ U	9.68×10 ⁻⁹	6.03×10 ⁻⁹	4.00×10 ⁻⁸
²³⁵ U	9.31×10 ⁻⁹	5.93×10 ⁻⁹	3.74×10 ⁻⁸
²³⁸ U	8.76×10 ⁻⁹	5.48×10 ⁻⁹	3.60×10 ⁻⁸
Fertilizer			
²³⁴ U	8.55×10 ⁻⁹	5.90×10 ⁻⁹	3.05×10 ⁻⁸
²³⁵ U	8.26×10 ⁻⁹	5.81×10 ⁻⁹	2.86×10 ⁻⁸
²³⁸ U	7.74×10 ⁻⁹	5.37×10 ⁻⁹	2.75×10 ⁻⁸

373 ^AGeometric mean, ^B2.5th percentile, ^C97.5th percentile

374 **Table 5** Sample-specific activities of ^{234}U , ^{235}U and ^{238}U

	^{234}U (Bq/g)	^{235}U (Bq/g)	^{238}U (Bq/g)
Gauern	6.93	3.21×10^{-1}	6.88
E1	5.72	2.65×10^{-1}	5.67
Fertilizer	2.92×10^{-1}	1.35×10^{-2}	2.90×10^{-1}

375 ^AGeometric mean, ^B2.5th percentile, ^C97.5th percentile

376 **Table 6** Sample-specific committed effective doses (ingestion of 0.01 g over 1 year)

	GM (Sv) ^A	P _{2.5th} (Sv) ^B	P _{97.5th} (Sv) ^C
Gauern			
²³⁴ U	3.06×10 ⁻⁷	1.59×10 ⁻⁷	1.52×10 ⁻⁶
²³⁵ U	1.35×10 ⁻⁸	7.24×10 ⁻⁹	6.57×10 ⁻⁸
²³⁸ U	2.74×10 ⁻⁷	1.44×10 ⁻⁷	1.36×10 ⁻⁶
Σ	5.94×10 ⁻⁷	3.11×10 ⁻⁷	2.95×10 ⁻⁶
E1			
²³⁴ U	2.02×10 ⁻⁷	1.26×10 ⁻⁷	8.34×10 ⁻⁷
²³⁵ U	8.99×10 ⁻⁹	5.73×10 ⁻⁹	3.61×10 ⁻⁸
²³⁸ U	1.81×10 ⁻⁷	1.14×10 ⁻⁷	7.45×10 ⁻⁷
Σ	3.92×10 ⁻⁷	2.45×10 ⁻⁷	1.62×10 ⁻⁶
Fertilizer			
²³⁴ U	9.12×10 ⁻⁹	6.29×10 ⁻⁹	3.25×10 ⁻⁸
²³⁵ U	4.08×10 ⁻¹⁰	2.87×10 ⁻⁶	1.41×10 ⁻⁹
²³⁸ U	8.20×10 ⁻⁹	5.68×10 ⁻⁹	2.91×10 ⁻⁸
Σ	1.77×10 ⁻⁸	1.23×10 ⁻⁸	6.30×10 ⁻⁸

377 ^AGeometric mean, ^B2.5th percentile, ^C97.5th percentile, Σ is the sum of the sample-specific committed effective doses
378 of the isotopes ²³⁴U, ²³⁵U and ²³⁸U and their radiologically relevant progeny

379 **Table 7** Committed effective dose coefficients (ingestion)

	ICRP (ICRP 1995a) (Sv/Bq)	Present method (Sv/Bq)
²³⁴ U	5.0×10^{-8}	5.2×10^{-8}
²³⁵ U	4.7×10^{-8}	4.9×10^{-8}
²³⁸ U	4.5×10^{-8}	4.7×10^{-8}

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