Uranium Concentration in Typical Pakistani Diet

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INTRODUCTION

Naturally occurring radionuclides are the largest contributors of radiation doses to human beings in most countries. These radionuclides reach the human body through the food chain and accumulate in critical organs, e.g., uranium in the lungs and kidneys, thorium in the lungs, liver and skeleton, and potassium in the muscles. They, if present in large amounts, pose radiation damage in the respective organs, cause physiological disorders, develop various types of diseases/cancers and increase the mortality rate.1

Uranium is a naturally occurring element, which is radiologically and chemically toxic. It occurs in the earth’s crust by 2–3 parts per million. It is present in most rocks, soils and rivers/seawater. This element contains 99.28% of 238U (half-life: 4.5 × 109 years), 0.72% of 235U (half-life: 7.01 × 108 years) and 0.006% of 234U (half-life: 2.5 × 105 years). These isotopes and their progeny emit alpha, beta, and low-energy gamma rays and normally do not pose any external hazard, but are a cause of concern if inhaled or ingested, and contribute significant radiation and chemical hazards.

For a realistic estimation of the radiation dose to various populations, a determination of the ingestion of uranium through foodstuffs is essential. In Pakistan, no published data on uranium intake through the diet has been available. Its concentration in the typical Pakistani diet was measured through an IAEA-RCA Project, entitled “Ingestion and organ content of trace elements of importance in radiological protection.” The results are presented in this article.

MATERIALS AND METHODS

Collection and preparation of total diet samples

To prepare typical Pakistani diets, food items of daily consumption (such as rice, flour, meat, vegetable, pulses, fruit, milk, egg, oil, etc.) were collected from 12 major districts/cities of the country (i.e. Abbottabad, Chakwal, Faisalabad, Hyderabad, Islamabad, Karachi, Mardan, Murree, Muzafarabad, Peshawar, Sialkot, Sargodha), using the market basket method. The edible portions of collected food samples were separated and washed with tap water. These individual foods were weighed, oven dried and pulverized separately in a commercially available grinder (Braun GmbH, Kronberg, Germany) with stainless-steel blades. Daily diets were prepared by mixing these dried food items according to the criteria of National Institute of Health, Pakistan1, as listed in Table 1. After mixing, a simulated diet was prepared from major cities and districts of the country by the market basket method, from which daily diets were prepared. These diet samples were analyzed using inductively coupled plasma mass spectrometry (ICP-MS) at the National Institute of Radiological Sciences, Japan, the regional Central Reference Laboratory of the Reference Asian Man Project. The measured values of the uranium content were found to vary from 2.3 ng (g dry)–1 to 11 ng (g dry)–1. The geometric mean concentration and geometric standard deviation were 4.5 ×101.7 ng (g dry)–1. This leads to a daily dietary intake of 2.6 ×101.7 µg d–1 or 33 ×101.7 mBq d–1, which is approximately 40% higher than the ICRP value. The measured value, i.e. 33 mBq d–1, contributes 12 Bq y–1 to annual intake of 238U activity and 0.54 µSv to the committed effective dose to the adult population. This is a very small fraction of the ICRP annual effective dose limit of 1 mSv for the general public. Therefore, it would pose no significant health hazard.

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prepared by heating the mixture in a microwave oven for 3 min. It was again oven dried, ground and stored in airtight jars. Water, tea, fish, and spices (i.e. garlic, pepper, salt, turmeric, etc.) were not included in these diets, because tea, fish and spices are only a small fraction of whole diet, and would not much affect the daily estimates, while water intake would be studied separately.

Chemical analysis

The uranium concentration was determined using the ICP-MS technique by following the Shiraishi et al. method at NIRS, Chiba, Japan. In this work, the digestion procedure was slightly modified. Instead of using an ash sample, an aliquot of a dried and mixed powder sample weighing 0.5–1 g was digested in a mixture of 5 ml ultrapure nitric acid (Tamapure-AA-100, Tama Chemicals, Tokyo, Japan), 0.5 ml perchloric acid (Tamapure-AA-100), and 0.5 ml hydrofluoric acid using a microwave digestion system (Model MLS-1200 MEGA, Milestone, Bergamot, Italy). The digested sample solution was evaporated to dryness in a 50-ml PTF beaker. The residue was completely decomposed with a mixture of acids, if any colored residue remained. The residue was dissolved in 10% ultrapure nitric acid and kept in a sample bottle until analysis.

The uranium concentrations were measured three times in triplicate samples by an ICP-MS instrument (Model 4500 Series, Yokogawa Analytical Systems, Tokyo, Japan) using bismuth as an internal standard. The measurements conditions were as follows: an ICP plasma torch had a microwave frequency of 27.12 MHz and an output of 1.2 kW; the position of ion sampling was 7.5 mm above the load coil; the sample uptake rate was 0.4 ml min⁻¹; and the total Ar gas flow rate was 16.9 liter min⁻¹. The scanning mass range was 209–239 m/z.

Quality control

The uranium analysis for NIST SRM 1486 Bone Meal, SRM 1548 Total Diet and SRM 1548a Typical Diet (NIST, Gaithersburg, MD, U. S. A.) was carried out, and the results are given in Table 2. The obtained values for these three reference materials were in agreement with the certified or reference values within 6.3, 18 and 32%, respectively. The larger deviations in the reference diet materials were considered to be one order of magnitude lower uranium concentrations than that of Bone Meal.

RESULTS AND DISCUSSION

The uranium concentration was determined in 12 typical Pakistani diets, prepared from various foodstuffs collected from major districts/cities of Pakistan; the results are presented in Table 3. A statistical analysis of the data shows that the uranium concentration in our diet varied from 2.3 to 11 ng (g dry⁻¹). The concentration is plotted as a function of the frequency distribution in Fig. 1 along with a Gaussian fit to the data. The fitted curve shows that the data are skewed (1.46) to the right with the degree of kurtosis being −0.10.

### Table 1. Typical composition of the daily diet of a common Pakistani man.

<table>
<thead>
<tr>
<th>Food group</th>
<th>Fresh weight (g d⁻¹)</th>
<th>Food type</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cereals</td>
<td>500</td>
<td>Wheat (roti, paratha, weaning food, rusk, biscuits, rice, corn, etc.)</td>
</tr>
<tr>
<td>Milk</td>
<td>161</td>
<td>Cow, buffalo, commercial milk products, lassi, yoghurt</td>
</tr>
<tr>
<td>Roots</td>
<td>40</td>
<td>Potatoes, other root vegetables</td>
</tr>
<tr>
<td>Pulses</td>
<td>43</td>
<td>Pulses, gram</td>
</tr>
<tr>
<td>Meat</td>
<td>39</td>
<td>Beef, mutton, poultry</td>
</tr>
<tr>
<td>Fish</td>
<td>6</td>
<td>Any kind</td>
</tr>
<tr>
<td>Eggs</td>
<td>7</td>
<td>Any kind</td>
</tr>
<tr>
<td>Oils</td>
<td>33</td>
<td>Ghee and other vegetable oils</td>
</tr>
<tr>
<td>Vegetables</td>
<td>91</td>
<td>All vegetables except roots</td>
</tr>
<tr>
<td>Fruits</td>
<td>8</td>
<td>Any kind</td>
</tr>
<tr>
<td>Sugar</td>
<td>39</td>
<td>Any kind</td>
</tr>
<tr>
<td>Total</td>
<td>967</td>
<td></td>
</tr>
</tbody>
</table>

### Table 2. Analysis of NIST Standard Reference Materials (SRM) for uranium.

<table>
<thead>
<tr>
<th>Specific SRM</th>
<th>Present work mean ± S.D. (µg g⁻¹)</th>
<th>Certified value mean ± S.D. (µg g⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SRM 1486 Bone Meal</td>
<td>20 ± 0.6</td>
<td>19 ± 1.2</td>
</tr>
<tr>
<td>SRM 1548 Total Diet</td>
<td>2.6 ± 0.3</td>
<td>2.2 ± 1.2</td>
</tr>
<tr>
<td>SRM 1548a Typical Diet</td>
<td>2.9 ± 0.01</td>
<td>2.2 ± 1.4</td>
</tr>
</tbody>
</table>

### Table 3. Details of a statistical analysis for uranium concentrations and daily intakes from the Pakistani diet.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Concentration (ng g dry⁻¹)</th>
<th>Daily intake (µg d⁻¹)</th>
<th>Daily activity intake (mBq d⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Geometric mean ± G.S.D.</td>
<td>4.5 ± 1.7</td>
<td>2.6 ± 1.7</td>
<td>33 ± 1.7</td>
</tr>
<tr>
<td>Arithmetic mean ± S.D.</td>
<td>5.2 ± 3.0</td>
<td>3.1 ± 1.8</td>
<td>38 ± 22</td>
</tr>
<tr>
<td>Median</td>
<td>3.7</td>
<td>2.2</td>
<td>27</td>
</tr>
<tr>
<td>Range</td>
<td>2.3–11</td>
<td>1.4–6.7</td>
<td>17–82</td>
</tr>
<tr>
<td>95% confidence interval</td>
<td>1.5–13.3</td>
<td>0.9–8.0</td>
<td>11–98</td>
</tr>
<tr>
<td>Skewness</td>
<td>1.46</td>
<td>1.49</td>
<td>1.49</td>
</tr>
<tr>
<td>Kurtosis</td>
<td>−0.10</td>
<td>−0.07</td>
<td>−0.07</td>
</tr>
</tbody>
</table>

Notes:
- †Number of samples 12.
The use of an appropriate mean for such data was determined by plotting the uranium concentration as a function of the percentage of the sampling frequency on the log-normal scale in Fig. 2. A straight line with a correlation coefficient of 0.975 predicts that the data follow a log-normal distribution. Therefore, the use of the geometric mean was assumed as being an appropriate representation of the data. The geometric mean (G.M.) and geometric standard deviation (G.S.D.) were calculated. The G.M. × G.S.D. was 4.5 × 1.7 ng (g dry)–1. These values lie well within the 95% confidence interval of 1.5–13.3 ng (g dry)–1.

The daily dietary uranium intake by the Pakistani population was calculated by multiplying the measured concentration on a dry-weight basis with the daily intake of diet in dry weight; the results are presented in Table 3. It was found that the measured uranium concentration (G.M. × G.S.D.) in daily diets was 2.6 × 1.7 µg d–1. It varied from 1.4 to 6.7 µg d–1 and was well within the 95% confidence interval of 0.9–8.0 µg d–1.

The daily intake of uranium based on the measured value of geometric mean was also determined in terms of the 238U activity. It was calculated by using the conversion rate for 238U (1 µg = 12.3 mBq). The estimated activity is 33 mBq with a G.S.D. value of 1.7, which lies within the 95% confidence interval of 11–98 mBq.

The measured daily dietary intake (2.6 µg d–1) by the Pakistani population was compared with the ICRP value of 1.9 µg d–1. The daily intake determined in this work was 1.4-times larger than the ICRP value for a 70 kg man.

The ingestion of 238U activity was compared with the median values of published data, as listed in Table 4. The comparison shows that the median intake of the 238U activity, as determined in the present work (27 mBq), is larger than the world average of 14 mBq d–1, but lies within the reported range of 6.6–62 mBq. This difference could be due to the variation in the composition of the diet and the natural environment among different countries.

The higher trends of uranium in Pakistani food might be due to following facts:

**Sampling techniques i.e. Market Basket Method:** It has been observed that the measured concentrations of uranium and thorium in market basket studies are on the higher side than those reported for the duplicate portion method. An underestimation of up to 20% has been observed for the Duplicate Portion Method.

**Composition of diet:** Grains and leafy vegetables are observed...
to be the largest contributors of terrestrial activity, i.e. $^{238}\text{U}$ and $^{232}\text{Th}$ concentrations, in food.  

As shown in Table 1, the major constituents of Pakistani daily diet are cereal (52%), milk products (17%) and vegetables (14%). Cereal and vegetables constitute 66% of the total daily intake, which could be the cause of the comparatively high values.

The extensive use of phosphate fertilizers in agriculture may also be responsible for the slightly high levels of the uranium contents in Pakistani diet because most of the diet consumed is home grown.

The annual intake of $^{238}\text{U}$ activity was calculated to be 12 Bq from the measured value of 33 mBq d$^{-1}$ as the geometric mean. The committed effective dose due to the ingestion of $^{238}\text{U}$ was estimated using the observed annual intake of 12 Bq and an ICRP dose coefficient of $4.5 \times 10^{-8} \text{Sv Bq}^{-1}$ for adult members of the public. It was found to be 0.54 $\mu$Sv, which is only a minor fraction of 1 mSv of the ICRP annual dose limit for the general public. UNSCEAR estimates the annual intake and committed effective dose from the ingestion of $^{238}\text{U}$ in the adults to be 5.7 Bq and 0.25 $\mu$Sv, respectively, using the same effective dose coefficient.

**CONCLUSION**

Analytical data on the baseline dietary intake of uranium in the Pakistani male adult has been obtained. The uranium concentration varied from 2.3 to 11 ng (g dry)$^{-1}$. The range of daily intake was 1.4 to 6.7 $\mu$g d$^{-1}$ and the daily activity intake was 17–82 mBq d$^{-1}$. The geometrical mean daily mass and activity intakes were 2.6 $\mu$g d$^{-1}$ and 33 mBq d$^{-1}$, respectively. The median daily activity intake, 27 mBq d$^{-1}$, seems to be considerably higher than the worldwide medians value of 14 mBq d$^{-1}$. It is shown that the daily activity intake is 33 mBq d$^{-1}$, or an associating annual intake of 12 Bq y$^{-1}$ contributes 0.54 $\mu$Sv of the committed effective dose in Pakistani adults. This constitutes only a very small fraction of the ICRP annual effective dose limit of 1 mSv for the general public. It is concluded that it would pose no significant health hazard.

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**APPENDIX**

In the sampling technique employed in the project, a total diet that is typical of a particular country, area or population group is constructed by the market basket method. Foods are purchased and prepared for consumption before analysis. The construction of a diet is often based on household food surveys and is identical to those consumed by individuals or groups of individuals over a period of time. This method does not assess the variability of intakes by individuals or reveal extreme intakes, but provides a useful basis for comparing individual studies. If continued over an appreciable time, it can expose trends in trace-element intake. Another technique known as Duplicate Portion Study (DPS), is also used as a second choice.

**REFERENCES**


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