DEPLETED URANIUM IN THE ENVIRONMENT – AN ISSUE OF CONCERN?

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Introduction

Natural uranium (U) occurs in soils in typical concentrations of a few parts per million. U-238 is the most abundant isotope in natural uranium (fraction by weight in natural uranium is 99.28%) and decays into other radioactive elements. A radioactive waste product of uranium enrichment is known as »depleted uranium« (DU) which is basically natural uranium in which the fissionable U-235 isotopic content has been reduced from 0.71% to 0.2% - 0.3%. It is practically pure alpha emitter, only selected (in-growth) daughter products are gamma and beta emitters. Comparison of radioactivities shows that the total activity in 1 mg of natural uranium is 25.28 Bq and in 1 mg of DU is 14.80 Bq. The radioactivity of DU is 60% of that of natural uranium. Currently in the USA alone, there are about 600.000 tonnes of DU in storage. DU is cheep and it is available in large quantities. It is widely used as ballast or counterbalances in ships and aircrafts, as radiation shielding and in non-nuclear civil applications requiring high density material.

Military Use of Depleted Uranium

The high velocity of DU ammunition permits to penetrate the reinforced armoring of tanks. DU is metallurgically similar to steel, thereby allowing similar production and processing techniques to be used. The very high density (18.9 g/cm^3) allows for much higher kinetic energy levels to be delivered to the target than an equivalent round made of, for example, steel. A secondary effect is that the DU on hiting the target ignites, thereby providing a pyrophoric effect within the target and forming an aerosol of DU particles and/or large fragments. The particle size of aerosol and the quantity depends on the impact angle, velocity and burning temperature. Alternative to DU is tungsten. Also, DU's density and physical properties make it ideal for use as armor plate. DU has been used in the USA Army systems for many years in both applications and over the past 20 years a number of weapon systems containing DU were developed and tested. Many other countries already have DU-containing weapon systems in their inventories as well.

The combination of design, high mass and high velocity allows the DU rounds to penetrate the target using the principles of hydrodynamic penetration. The pressures involved are so high that the armour of the target flows away from the DU penetrator. DU fragments are non-magnetic and will retain heat to the point where they will cause serious burns for three to four hours after firing.

The Gulf war was the first use of DU weapons (320 tons of DU) outside the test ranges. It was used also throughout Kosovo during approximately 100 NATO air strike missions. A total of approximately 31,000 rounds of DU ammunition was used in operation. The total weight of the amount used in Kosovo was estimated to be about 8-10 tons.

Health Effects of Depleted Uranium

The exposure pathways which will be described in this paper are those relevant to the use of DU munitions in a conflict situation. The main exposure pathways are:

- *External exposure* to radiation from DU munitions laying on the ground or buried under the surface after an attack, or loaded in a tank or from the DU in the armoured shielding of tanks.
- *Internal exposure radiation:*
- Inhalation of aerosols created from the impact of DU munitions.
- Ingestion of soil and dust containing DU from deposition of aerosols formed during the impact of DU munitions against a target or from oxidation of DU fragments buried under the surface.
- Ingestion of food and water containing DU which has been deposited directly (plants) or transferred to the terrestrial foodchain from the soil.
- Internal irradiation from DU fragments embedded in the human body.

The transport of a material that moves around a human body is simulated using biokinetic models. These models permit to estimate how much of the material that entered the body at the time of the intake is present in each organ at any time afterwards. In the case of radioactive material a dosimetric model is then applied to calculate how much of the energy released in each type of radiation is absorbed in each tissue or organ, and hence the radiation doses to the different organs.

The health hazards depend on:

a) the route of exposure (inhalation, ingestion or contact in wounds),
b) the magnitude of exposure,
c) the particle size and solubility of DU.

Effects due to external exposure would be limited to radiological effects, whilst the effects due to internal exposure include both radiological and chemical toxicity effects.

Chemical Effects: Uranium can be chemically toxic when large amounts enter and are retained in the body, absorbed in the blood, and carried to body tissues and organs. The severity of the toxic effect depends on the amount the blood absorbs, how that amount is distributed among the body's organs, and uranium's toxic effects in those organs. A considerable fraction of the DU ammunition can become aerosols. Typical composition of an aerosol after hitting a solid target is: ca. 61% U_3O_8 (moderately soluble compound), ca. 18% UO_2 (insoluble compound) and ca. 20% uranium amorphous oxides. In the war environment, DU entered the body through inhalation, ingestion, or wounds in the form of uranium metal and uranium oxides. Once disolved, uranium may react with biological molecules and, in the form of the uranyl ion, exerts its toxic effects. The kidney is the organ

most sensitive to chemical effects from excess uranium. Depending on the concentration of uranium in the kidney, these toxic effects may include damage and death of kidney cells, decreasing the kidney's ability to filter impurities from the blood.

- *DU oxides* could enter the human bodies when they are inhaled them from the air or when they are ingested as residues. About 95% of large inhaled particles deposit in the upper respiratory tract. Particles in the upper region of the respiratory system move by mucociliary transport to the gastro-intestinal tract. Insoluble uranium oxides can remain in the lungs for years, slowly absorbing into the blood and then being excreted in urine. Inhaled *uranium amorphous oxides* will rapidly transfer to blood and 98% of ingested uranium will be eliminated through the faeces and 2% absorbed by the intestine.
  Numerous studies of the effects of inhaled or ingested uranium in humans have not conclusively documented increased death rates or effects on the immune or nervous systems. Uranium miners have experienced an increased risk of lung cancer, which scientists agree is attributable to other substances, such as tobacco smoke, radon, and radon's short-lived radioactive decay products. Although animal studies have suggested possible liver injury from uranium, it has not been found in anyone exposed to DU, including workers at uranium testing sites. Particularly susceptible from damage from high doses of uranium is the kidney, where uranyl-carbonate complexes decompose in the acidic urine, causing health concerns. Uranium's toxic effects on the kidney resemble those caused by other heavy metals, such as lead or cadmium. Intake of 230 mg of moderately soluble DU aerosols is leading to a kidney concentration of 3 micro gram per gram of kidney (chemical toxicity limit).

- *Radiological Effects of Depleted Uranium.* DU and its decay products emit alpha, beta, and gamma radiation that can cause external and internal exposure to those who handle munitions. Natural and depleted uranium are much more likey to be chemical than radiation hazards. Recently, the American Agency for Toxic Substances and Disease Registry (ATSDR) concluded: »DU is radioactive but poses little radioactive danger because it gives off very small amount of radiation«. A small fraction of uranium taken into the body deposits in the skeleton but the results of scientific studies indicate DU does not cause bone cancer. In fact, scientists have never observed bone cancer in population exposed to any form of uranium, including enriched uranium.
  In a process called external radiation, depleted uranium outside the body can also expose people. The theoretical maximum whole body gamma irradiation dose rate from external exposure to DU is 0.0025 mSv per hour (mSv/hr). Dose rates   in this range might be experieced by a person surrounded by DU munitions. Dose from driving a fully loaded tank for 2000 hours will be equal to annual dose from natural background. It was estimated also that the dose rate to a person standing on the ground uniformly contaminated with 1 tonne DU per square kilometer or 1 gram per square metre of ground would be 0.01 mSv per year, which is 10% of the dose rate from uranium present in soil.
  The dose rate to skin from direct contact with DU is much higer, at about 2.5 mSv per hour, mostly from beta particles. A large proportion (98%) of this dose is attributable to beta radiation. DU's alpha radiation does not penetrate the dead layer of the skin. Only radiation emitting from the surface of DU is a factor, as the DU itself shields the internal emissions. Skin is relatively insensitive to radiation, so that even continuous

contact is unlikely to produce a radiation burn or other short-term effect. Such effects require doses of a few sieverts, delivered over a short period of time, but DU would need to be in contact for weeks to give such doses. There would, however, be expected to be an increased risk of skin cancer and such contact should be avoided. Direct skin contact with a DU penetrator for 400 hours would increase the risk of skin cancer by perhaps 40%.

In fires and during impact, DU forms mixtures of both soluble and insoluble oxide aerosols. The extent to which there are suspended particles in the air after an impact will depend on their sizes, on the way they are dispersed by the initiating events, and on local conditions. How far the particles reach within the respiratory system will depend on their size. Some of the particles may be very small, with diameters less than a micron. These may penetrate deep into the lungs. Fragmentation products will usually be bigger than a few microns. The coarse particles will not penetrate deep into the lungs and will be cleared by mucous flow and swallowed. If DU is in its soluble uranium oxides form, then any hazard would arise from inhaling the dust. The insoluble dust could be deposited in the lungs an over a very long period could be a contributory cause of lung cancer. The dose to the red bone marrow, which is considered to be the target organ for the induction of leukaemia, is about 100 times smaller than that to the lungs. *Since the lungs and red bone marrow are considered to be equally radiosensitive, one would expect fewer leukaemias than lung cancers.*

According to the United Nations Environment Programme Depleted Uranium Desk Assessment Group (UNEP 1999) the maximum endurable instantaneous intake by breathing is of the order of 1 g of dust, or 100 mg of depleted uranium. Assuming very dusty condition (100 mg per cubic metre) an individual would have to inhale dusty air that was heavily contaminated with DU for 10 hours to inhale 100 mg of DU. Inhalation of 1mg DU would typically lead to an effective dose of the order of 0.1 mSv and therefore, it is unlikely that anyone would incur an effective dose greater than 10 mSv from inhalation of DU. Intake leading to a dose of 1 mSv in a kidney is 32 mg of DU (activity is 480 Bq) during inhalation of moderately soluble DU aerosols, such as UO_3 and U_3O_8. For insoluble compounds, such as UO_2 this value is 11 mg. These values have been calculated with the biokinetic models currently recommended by the International Commission on Radiological Protection.

It can be concluded that despite of the fact that DU is radioactive material, DU is not a nuclear, radiological or chemical weapon; DU is used because of its high atomic mass and metallurgical properties.

External pathways are relatively minor contributors to dose unless DU is in contact with skin. Inhalation is a more significant exposure pathway than ingestion, because uranium is not readily transferred through the environment.

Potential Environmental Effects of Depleted Uranium

Uranium is a reactive metal, and, therefore, it is not present as free uranium in the environment. It combines with other elements in the environment to form uranium compounds. The solubility of these uranium compounds varies greatly. Uranium in the environment is mainly found as a uranium oxide, typically as UO2, which is insoluble compound. When DU rounds is fired DU metal will undergo dissolution in the

environment. Low levels of soluble uranium travel very slowly through the soil and are subject to adsorption as they pass through the soil. Transfer of soluble uranium with rainwater runoff is limited because of the low solubility of uranium in the rainwater. Contamination of ground water depends on depth of penetratio and proximity of water is probably most important factor.

DU contamination greater than 50 parts per million to be harmful to plants. In broad terms uranium in plants is the result of its uptake from the soil into roots and other parts of the plant. Uranium is, in general, not readily absorbed by plants from the soil; plant/soil concentration factors of uranium, which are used in models for the assessment of the radiological effects of radioactive material in the environment, are rather small, depending on the plant and the type of the soil and its characteristics.

Several potential pathways were indentified for DU introduction and transportation through the food chain that include small insects, reptiles, animals, and humans. Deer and rabbits could consume vegetation which has DU contaminated dust on its leaves; however, the dilution effects of obtaining food sources outside the impact area are highly influential. With this in mind it can be concluded that radiation doses from DU in the environment are relatively low from all exposure pathways.

## Analytical methods for determination of uranium and its radioisotopes

There are several very sensitive analytical methods for determination of uranium. Alpha spectrometry following radiochemical separation, isotope dilution mass spectrometry, inductively plasma mass spectrometry and neutron activation analysis are all used. However radiometric methods are time consuming, large samples are needed due to the low specific activity of uranium and are thus limited by reagent blanks. As well as isobaric interferences, mass spectrometric methods are also affected by blanks at ultratrace levels.

Neutron activation analysis

Neutron activation analysis offers important advantages for analysis of trace and minor elements due to its inherent characteristics such as matrix independence, virtual freedom from blank problems and specificity, as well as its totally independent nature as a nuclear-based method, rather than an electronically-based phenomenon as in all other chemical and spectroscopic methods. These inherent advantages make it attractive in quality control. Radiochemical neutron activation analysis (RNAA) is an excellent method for determining uranium and thorium and offers unique possibilities for their ultratrace analysis using selective radiochemical separations. An important source of error in RNAA is uncertainty in the chemical yield. Thus one of the most important stages relating to quality control in determination of uranium and thorium is the determination of the chemical yield.

In the last fifteen years, various radiochemical procedures for the determination of natural and man-made radionuclides were developed at the J.Stefan Institute, with emphasis on radioisotopic determination of the chemical yield.

Determination of uranium at trace levels by radiochemical neutron activation analysis is possible by two different approaches using the ^{239}U or ^{239}Np nuclides induced by neutron capture and β^- decay:

$$^{238}U(n,\gamma)^{239}U \ (t_{1/2}= 23.5 \ min) \rightarrow \ ^{239}Np \ (t_{1/2}= 2.35 \ d) \rightarrow .$$
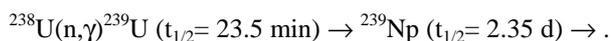
Determination of uranium via long-lived ^{239}Np daughter is based on separation by anion exchange. This nuclide allows use of lengthier dissolution procedures employing total decomposition with hydrofluoric acid. Conventional wet ashing might be subject to negative errors due to incomplete dissolution of any lithogenous or siliceous uranium-containing fraction. The chemical yield was determined by addition of ^{238}Np produced by neutron irradiation of ^{237}Np(n,γ)^{238}Np (t$_{1/2}$= 2.2 d).

Determination of uranium via short-lived nuclide ^{239}U offers better sensitivity. We have developed a very sensitive method for uranium determination using selective extraction with TBP. Addition of large amounts of carrier allows the chemical yield to be evaluated from the γ-spectrum of the isolated uranium fraction from the 187.5 keV γ peak of ^{235}U. This method was further improved by removing traces of radionuclides other than those of uranium, which is important if a well-type detector, with high efficiency but low resolution is to be used. ^{239}U nuclide allows determination of nanogram and picogram quantities of uranium.

Alpha spectrometry

Determination of the activity concentrations of the natural alpha emitting radioisotopes of uranium (^{234}U, ^{235}U, ^{238}U) and/or thorium (^{228}Th, ^{230}Th, ^{232}Th) and man-made radionuclides (^{241}Am, ^{238}Pu, $^{239+240}$Pu) in environmental samples by alpha spectrometry following their radiochemical separation is widely practised in radiochemical laboratories. Alpha spectrometry is an analytical technique, which gives very accurate values.
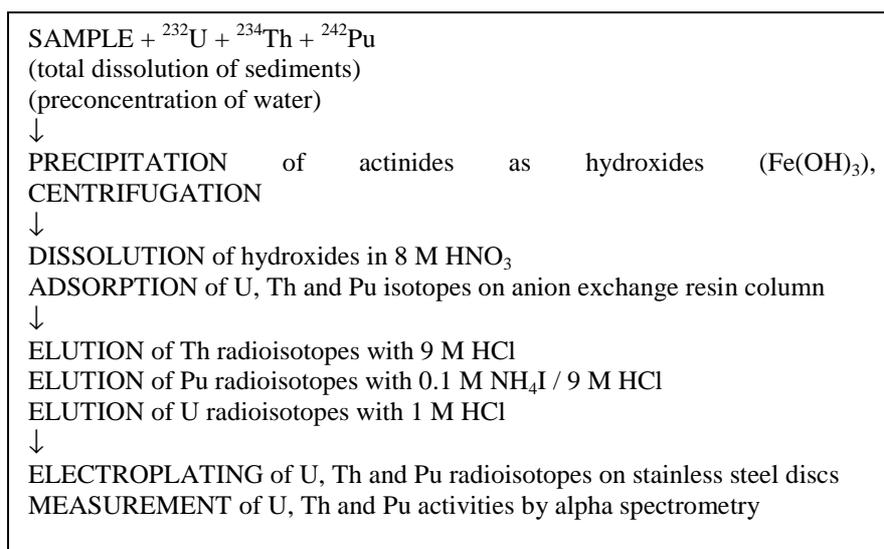
SAMPLE + ^{232}U + ^{234}Th + ^{242}Pu
(total dissolution of sediments)
(preconcentration of water)
↓
PRECIPITATION of actinides as hydroxides (Fe(OH)$_3$), CENTRIFUGATION
↓
DISSOLUTION of hydroxides in 8 M HNO$_3$
ADSORPTION of U, Th and Pu isotopes on anion exchange resin column
↓
ELUTION of Th radioisotopes with 9 M HCl
ELUTION of Pu radioisotopes with 0.1 M NH$_4$I / 9 M HCl
ELUTION of U radioisotopes with 1 M HCl
↓
ELECTROPLATING of U, Th and Pu radioisotopes on stainless steel discs
MEASUREMENT of U, Th and Pu activities by alpha spectrometry

Figure 1: Radioanalytical method for sequential separation and determination of uranium, thorium and plutonium radioisotopes by α-spectrometry.

Determination of radioisotopic composition by alpha spectrometry requires a very high degree of chemical purification because interference's give rise to self-absorption of the

alpha particle in the source, which is a critical point. The separation of radionuclides by means of anion exchange is a widespread method for analysis of environmental samples. To calculate the chemical yield in the separation procedure one or more quality control methods must be used: by adding some yield tracers (^{232}U, ^{234}Th, ^{243}Am, ^{242}Pu), internal standard method, when possible, both an analytical and a radiometric method, etc. Preparation of a weightless counting source for high resolution alpha spectroscopy is a common problem in alpha spectrometry.

We developed a sensitive and reliable method for sequential separation and determination of uranium, thorium and plutonium radioisotopes in the same sample aliquot. Figure 1 shows the radioanalytical procedure for sequential separation and determination of natural and man-made radionuclides.

The IAEA Fact Finding Mission to Kosovo, 16 – 19 August 1999

On request of the United Nations the IAEA carried out jointly with the UNEP/BTF a Fact Finding Mission to Kosovo shortly after the air strikes were ceased.

The purpose of mission was to conduct a preliminary fact finding mission in the context of the aims and objectives of the UNEP/Balkans Task Force (BTF) in order to assess the existing radiological situation in relevant locations in Kosovo, and to determine whether the expanded field study (sampling and measurements campaign) proposed by BTF related to depleted uranium (DU) in the environment is needed.

The team performed some preliminary measurements, such as, of absorbed dose rates in the air and surface alpha and beta contamination levels around the destroyed vehicles, at and along the roads the team travelled, and in Pristina around the damaged/destroyed Police Station and Post Office (checked partly inside as well). In addition to the measurements, two sets of smear samples were taken from the surfaces of the destroyed tanks for subsequent isotopic analysis. These samples were taken for the purposes of establishing "background" levels since the tanks were not attacked using DU weapons.

The mission found no elevated levels of radioactivity at the places indicated to the mission as possible target areas during the attacks. The analysis of smear samples from a few selected heavy military vehicles (tanks) also showed no radioactive contamination.

In October 1999 UNEP BTF (United Nations Environment Programme – Balkan Task Force) published a comprehensive report in which the DU was considered as a non-significant environmental and health-related issue.

Conclusions

- It may be concluded that the depleted uranium has no acute risk on the environmment.
- Theoretical cancer risk due to DU contamination is much smaller than that from natural radiation.
- However, in the future actions a reliable radioecological assessment and dose estimates should be carried out.
- Health-effect assessment should be made in heavily contaminated areas, if any.
- In the occupational hygiene area, monitoring before and after deplyment of personnel should be conducted, the DU activities should be properly documented, dose estimates should also be made jointly with reliable health statistics records.

Sources

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