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RISK ASSESSMENT APPLICATIONS FOR DETERMINING CLEANUP LIMITS FOR URANIUM IN TREATED AND UNTREATED SOILS

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ABSTRACT

Uranium-contaminated soils are present at various locations across the U.S. where uranium was processed for nuclear fuels or atomic weapons. Important issues relative to such contamination include the assessment of potential health risks associated with human exposures to the residual uranium and the determination of safe levels of uranium in soils that have been treated by a given technology. This paper discusses various risk assessment considerations that must be dealt with when developing cleanup limits for uranium in treated and untreated soils. Key issues addressed include alternative land use scenarios, potential exposure pathways, characterization of the bioavailability of uranium compounds in food and water, a brief overview of health risks associated with uranium and its daughter products as well as a summary of considerations for development of risk-based cleanup limits for uranium in soils.

INTRODUCTION

The principal goal in developing a cleanup limit or guideline for uranium in soil is the protection of human health. In order to achieve this goal, a safe dose or acceptable level of incremental cancer risk must be defined along with the exposure pathways that could bring an individual into direct or indirect contact with contaminated soil. An exposure scenario must also be specified that defines the relevant exposure parameters and assumptions that are used to represent the population at risk (e.g., breathing rates, amount of homegrown food consumed, duration of exposure, etc.). The cleanup limit is calculated as the maximum concentration of total uranium (or perhaps individual isotopes) in soil that does not produce combined inhalation, ingestion, external radiation, and dermal exposures that result in dose or risk levels in exceedence of established criteria.

Several issues must be considered when developing cleanup limits. The uranium decay chains of ^{235}U and ^{238}U contain daughter products that may constitute health hazards. Radon, a radioactive noble gas, is of special concern because it enters residences as a result of diffusive/advective transport from soils into the basements of houses. In order to restrict the long term buildup of this particular daughter product, it may be necessary to limit the levels of residual uranium in soil. Alternatively, foundation or basement venting technologies can be employed to reduce indoor exposures to radon.

Cleanup limits will also be sensitive to the exposure pathways and scenario(s) used to define human contacts with a radionuclide derived from soil, the nature of the dose-response relationship(s) of the radionculide, the physicochemical properties affecting its movement in the environment, and finally, properties of the contaminated landscape (e.g., depth to ground water, amount of rainfall, etc.). Another factor that must be considered when establishing such a limit is the background concentration of the radionuclide in host soils. Without knowledge of the background level of uranium in soil, it is not possible to evaluate whether or not a cleanup level is achieved.

Because there are so many parameters influencing the determination of soil cleanup limits, little progress has been made toward the development of generic cleanup standards, similar to standards for drinking water, that could be applied to contaminated sites across the country. The alternative is to develop cleanup limits that are site specific, using methodologies and supporting data that incorporate explicitly site-specific information and that focus on the key exposure pathway(s) of concern for each contaminant considered (1,2). Such methodologies should also specify the most important monitoring requirements for determining compliance with derived cleanup limit. For example, a concentration limit expressed as pCi radionuclide/g soil is ambiguous if no guidance is given regarding the chemical form of the radionuclide and pertinent soil properties. To the extent that soil-based exposures to uranium (and associated health risks) are sensitive to the areal extent and depth of contamination, it will also be necessary to incorporate these spatial factors in the development of cleanup limits. Moreover, the final monitoring guidance that is issued should be optimized so that only those measurements and statistical analyses that bear directly on compliance are required. This will help ensure that sampling and analysis costs are minimized.

ALTERNATIVE LAND-USE SCENARIOS AND ASSOCIATED EXPOSURE FACTORS

The types of land uses that are present or that may feasibly evolve in the future at a given location have a direct effect on the nature and magnitude of potential human exposures to residual uranium in soil. A residential land use, for example, might include exposure pathways involving the ingestion of vegetables grown in the contaminated soils of a home garden as well as the incidental ingestion of uranium contaminated soils/dusts. A commercial land use, in contrast, would not necessarily include these particular exposure pathways. The four primary land-uses considered in human health risk assessments are residential, agricultural, commercial/industrial, and recreational. Each type of land use has a suite of potential exposure pathways associated with it that must be identified and evaluated. Exposure pathways consist of four basic elements: (1) a source and a mechanism of contaminant release, (2) a retention or transport medium (e.g., soil, sediment, or water), (3) a point of potential human contact with the contaminated medium (i.e., an exposure point), and (4) an exposure route (e.g., ingestion) at the contact point (3). Environmental media that may be affected include air, surface water, sediment, soil, ground water, and biota. Paustenbach et al. (4) have recommended that analyses of soil contamination also address secondary scenarios relative to the protection of ground water, fish/wildlife, and surface-water/sediment quality. Figure 1 provides a conceptual diagram of soil-based exposure pathways that should be considered when assessing the health risks of ^{235}U and ^{238}U and their daughter products in soil.

PLACE FIG. 1 HERE

Several principal features that affect the nature and magnitude of exposures for each of the four basic land uses are summarized below.

Residential Scenario

Residential exposure scenarios should be used whenever there are occupied residences adjacent to the contaminated site. Under this scenario, residents are expected to be in frequent contact, either directly or indirectly, with uranium contaminated soil. The assumptions in this case account for daily exposure over the long term and generally result in the highest potential exposure and risk. Important exposure parameters that define this scenario include years of residence at a given house (i.e., exposure duration), exposure frequency (i.e., days per year in contact with contaminated media), fraction of food intake from a home garden, source of drinking water, and activity patterns (e.g., time spent outdoors as function of age).

Agricultural Scenario

Contaminated lands that are suitable for farming or that are in agricultural areas should be assessed for an agricultural land use in which exposures occur as a result of farm worker-related contacts with contaminated media as well as the consumption of home-grown foods. Exposure parameters for agricultural workers differ from the commercial/industrial worker because of differences in work habits, such as length of the work day or seasonal employment in the area. A farm-family scenario would include residential and worker exposure factors as well as consumption of home grown produce.

Commercial/Industrial Scenario

Under this scenario, workers are considered to be exposed to contaminated media within a commercial or industrial site. The exposure parameters apply to those individuals who work on or near the site. Exposure is based on assumptions relative to actual time spent on the site (8 hours a day for 250 days per year). The exposure duration at a given site should be specified as well as the nature of the commercial facility (e.g., outdoor operations or an office building).

Recreational Scenario

Exposures in this scenario are dependent on site-specific factors, such as the amount of time spent on or near the site where recreational activities occur. Hunting, fishing, biking, or other activities applicable to the site help define the scenario. Subsistence fishing may be considered in this scenario, or any of the other scenarios, if there is a possibility for the migration of uranium from a contaminated site to a local stream or lake.

ESTIMATION OF EXPOSURES TO URANIUM

It is important to understand the extent of human exposures to natural uranium via ingestion (of water, soil, biota), inhalation (of U particles in air and radon), and dermal contact (with soil and water) in order to compare these with exposures that might occur at contaminated sites (see Fig 1). In the subsections below, we discuss various factors that influence the magnitude of ingestion, inhalation, and dermal exposures to both ubiquitous and anthropogenic uranium in environmental media and foods.

Ingestion

The ingestion of uranium in foods and beverages is estimated as the sum of the products of the concentrations of uranium in various dietary items and the intake of those items. Fisenne et al. (5) used this approach to determine the ingestion of uranium isotopes by residents of New York City. They estimated that average intakes of ^{234}U, ^{235}U, and ^{238}U were 7.9×10^{-5}, 9.2×10^{-3}, and 1.3 μg/d, respectively. A pie chart of the different dietary sources of uranium is depicted in Figure 2. The measurements of the concentrations of uranium in various dietary items provides insight into the biotransfer processes influencing the movement of natural uranium in terrestrial and aquatic systems. For example, soil contaminants can be transferred to the above-ground portions of food-chain crops by root uptake as well as by ejection of contaminated soil particles to foliar surfaces. Root uptake of a substance is directly proportional to the solubility of the substance and therefore as solubility decreases, resuspension and rain splash become the important biotransfer mechanisms. In fact, the concentrations of ^{238}U that Fisenne et al. measured in leafy vegetables were a factor of three greater than for root vegetables, suggesting that soil derived particulate contamination of foliar surfaces is an important transfer process. Shepperd and Evenden (6) analyzed the relative importance of root uptake and soil deposition for uranium in spinach leaves and also concluded that direct soil contamination of leaves was the dominant biotransfer mechanism. Another interesting measurement reported by Fisenne et al. was the elevated concentration of ^{238}U in shellfish (1935 mBq/kg wet wt.) compared with finfish (13 mBq/kg wet wt.). The large difference in concentrations is attributed to the fact that shellfish are filter feeders and accumulate uranium that is sorbed to sediments.

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Inhalation

Uranium in the atmosphere is derived principally from the weathering of surficial soils. The movement of wind over the earth's surface results in the suspension of uranium bearing soil particles, which subsequently settle onto any exposed surfaces. Fisenne et al. reported concentrations of ^{234}U, ^{235}U, and ^{238}U in New York City air of 0.93, 0.036, and 0.94 $\mu Bq/m^3$. Inhalation accounted for about 0.1% of the total human intake of uranium isotopes, based on an elevated breathing rate of 20 m^3/d (the remaining 99.9% was derived from drinking water and foods). More realistic estimates of breathing rates(i.e., 11 to 15 m^3/d for females and males) would further decrease the estimated contribution of the inhalation route of exposure to uranium (7). However, even though inhalation represents a small fraction of overall uranium intake, Fisenne and Welford (8) found that the concentrations of uranium in lungs derived from autopsy cases from New York City displayed an increase with age. The age-dependent accumulation of uranium in lungs indicates that inhaled uranium is probably an insoluble uranium oxide (which has a long retention time in lungs) derived from the weathering and resuspension of surficial soils.

The decay chains of ^{235}U and ^{238}U include daughter products that also constitute a potential health hazard. The maximum hazard of the daughter products, however, does not occur until secular equilibrium is attained with the parent nuclides. A pivotal radionuclide in the decay chain of natural uranium is ^{226}Ra. Radium has been shown to cause bone cancer, and therefore ingestion of soil-derived radium in foods or drinking water warrants analysis in order to determine the magnitude of the associated health risks.

Of greatest interest, though, are two of radium's daughter products: ^{222}Rn and ^{214}Bi. Gamma radiation emitted from ^{214}Bi that is in secular equilibrium with ^{226}Ra in topsoil will result in increased external radiation exposures to those residing at a contaminated site. Radon gas (^{222}Rn) in near-surface soils produced by the decay of ^{226}Ra can migrate to the atmosphere due to diffusive and advective transport driven by concentration and pressure gradients, respectively, in near-surface soils.

Similarly, radon in soil adjacent to house foundations can also migrate into basement air and then to household air. There are many factors that influence the transport of radon from soils to indoor air, including: moisture content of soil, intrinsic soil permeability, pressure gradient, and type of foundation (9). Inhalation of radon and its short-lived daughter products present in indoor air poses an increased risk of lung cancer as a result of the irradiation of lung tissue by alpha particles emitted from those radionuclides.

Dermal Uptake

There is only limited information available on the uptake of uranium through the skin. It appears that systemic toxicity is correlated with the solubility of the uranium compounds.

For example, in animal studies a soluble species such as uranyl nitrate was actively transported across skin, causing toxic effects, but exposure to insoluble uranium oxide produced only slight weight loss (10). The magnitude of human exposures via the dermal route depend on several factors including; the concentration of uranium in the soil/dust deposited on skin surfaces, the areal extent of skin contamination, and the loading of soil/dust on skin. The actual uranium intake through the skin is a function of the permeability constant of the uranium species.

POTENTIAL HEALTH EFFECTS FROM EXPOSURES TO URANIUM

The principal health effects of concern associated with uranium are kidney toxicity and radiation-induced cancer in target tissues, including bone and lungs. The relative importance of these health effects depends a great deal on the route of exposure and the physicochemical properties of the uranium. Daughter products such as radium and radon also may constitute health hazards; however, for processed uranium compounds it could take thousands of years for decay products to reach levels which pose any health threat. In order to determine a "safe" level of uranium in soil, a methodology must provide estimates of direct and indirect exposures to uranium that: (1) protect against kidney damage, (2) result in cancer risks that are within acceptable levels, and (3) produce absorbed radiation doses that do not exceed applicable limits.

An important health effect is related to the biokinetic behavior of uranium compounds once inhaled or ingested. Kidney damage can occur as a result of exposure to soluble uranium. The nephrotoxicity is attributed to the heavy metal properties of uranium as opposed to the radiological component. The biouptake of uranium is dependent on the solubility of ingested or inhaled uranium and, thus, a related issue is the development of technique(s) for estimating the soluble/insoluble fractions of total uranium in treated and untreated soils. This is important because as the amount of bioavailable uranium in soil increases, the prevention of kidney toxicity becomes the primary health concern.

Other health effects are related to the radiological properties of uranium and its associated daughter products. The deposition and retention of uranium compounds in the respiratory tract is controlled principally by the particle-size distribution of the inhaled particles. As the aerodynamic diameter of the particles decreases, the removal efficiency for inhaled particles in the respiratory tract decreases, resulting in higher doses to lung tissue. Also, the dissolution rate of the uranium compound has an affect on the retention time in the respiratory tract. In addition to tissue damage and the induction of cancer to the lungs, uranium can also accumulate in bone tissue as a result of both the inhalation and ingestion exposure routes and pose a potential for risk of bone cancer. As previously mentioned, the radioactive decay products of ^{235}U and ^{238}U may also pose a cancer risk. Of greatest concern are the daughter products radium and radon.

SUMMARY

The development of risk-based cleanup limits for uranium in treated and untreated soils must take into consideration a host of human, environmental, and chemical specific parameters. This paper has discussed a few of the important issues related to risk assessment applications in the development of cleanup limits. In addition, Table 1 contains a summary of the various considerations in developing such limits. An important conclusion is that site-specific factors will have a strong influence on derived limits, and hence generic guidelines are not likely to serve as an effective tool for managing the health risks of such contamination.

The development and implementation of a site- and technology-specific cleanup limit will undoubtedly require that a host of input parameters be quantified, nevertheless, sensitivity analyses can be performed to reduce the set of parameters to the minimum necessary to yield defensible cleanup limits. Compliance monitoring approaches must be developed in conjunction with a cleanup limit to ensure that the required sampling and analysis of soils is consistent with the derived limits. Finally, there are clearly a number of uncertainties involved in each of the areas discussed above, and it is important to characterize the principal sources of variability in the defined cleanup limit.

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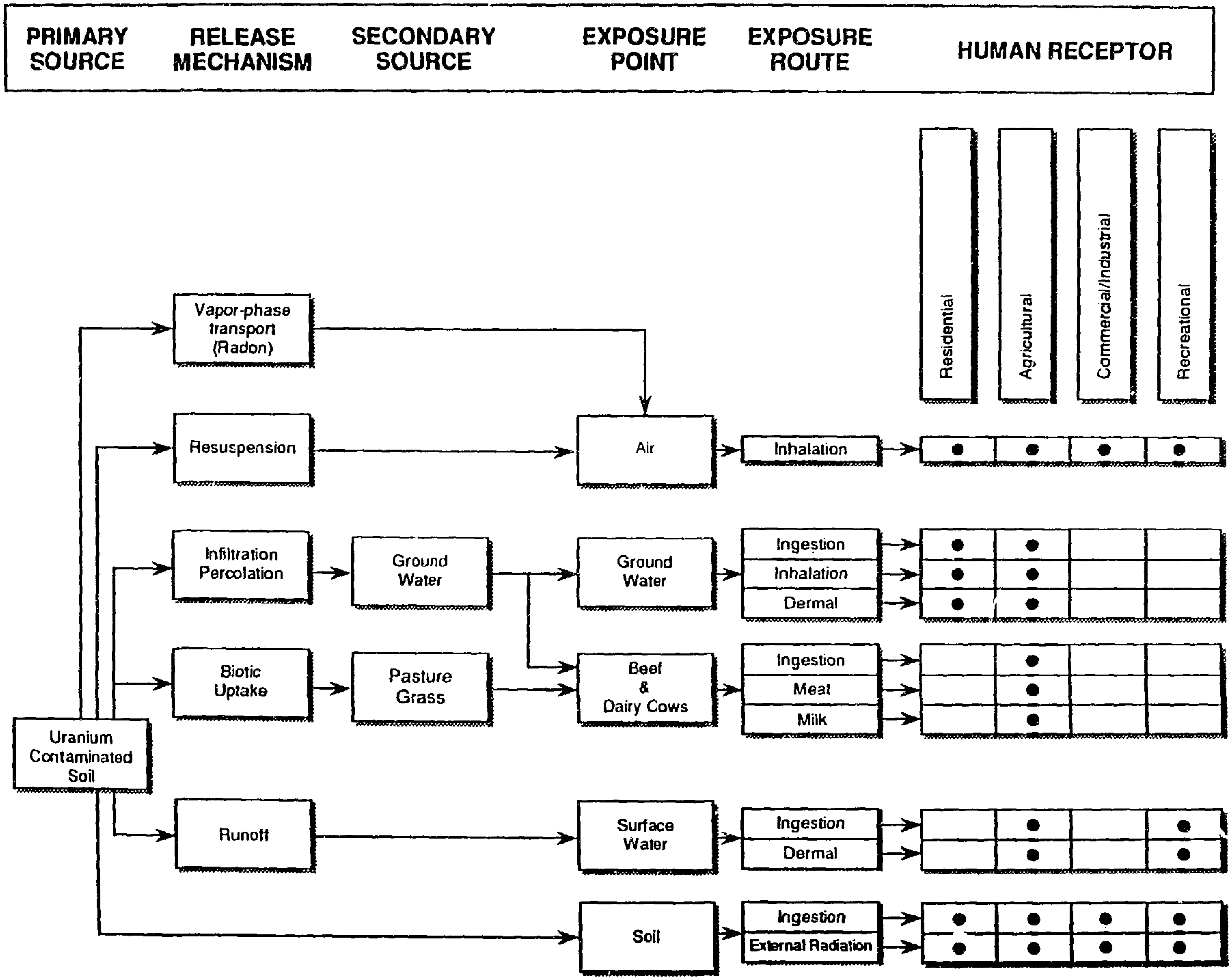

Figure 1. Overview of potential soil-based exposure pathways to uranium and its daughter products.

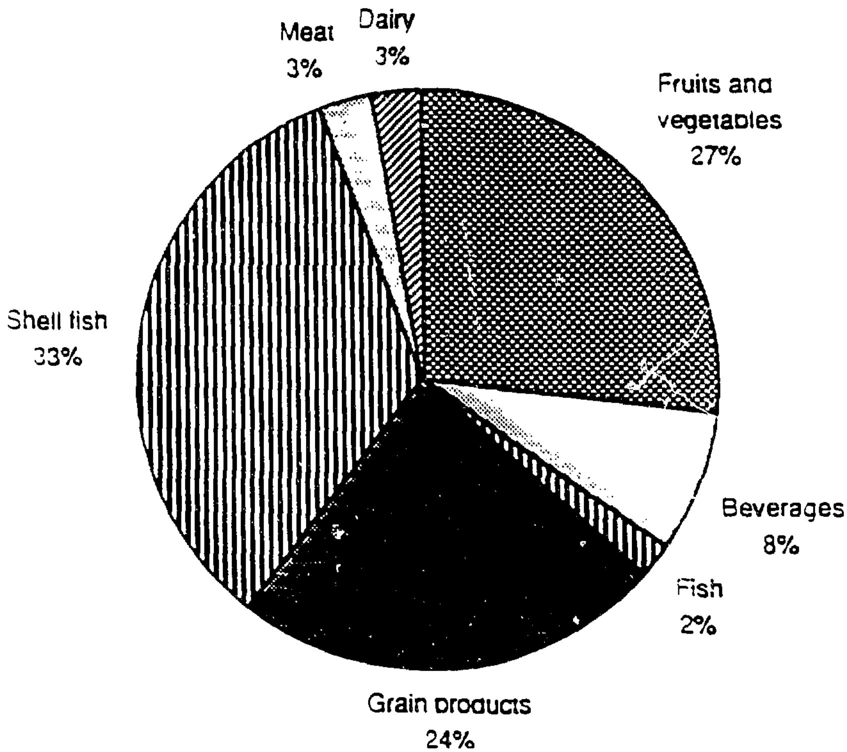

Figure 2. Sources of natural uranium in the average diet of residents of New York City (Fisenne, et al., 1987).

Table 1. Overview of various considerations affecting the development of risk-based cleanup limits for residual uranium in treated and untreated soils.

Type of parameter	Consideration
Human	Type of land use(s) likely to occur at a site Alternative exposure pathways Source(s) of drinking water Dose-response relationships Carcinogenicity (bone and lung) Kidney toxicity Human factors Mobility Consumption of contaminated foods Inhalation rates indoors/outdoors
Environmental	Depth to ground water Soil composition Leach rate of uranium species Annual rainfall Atmospheric dust loading Indoor/outdoor levels of airborne uranium
Chemical/Radiological	Isotopic composition of residual uranium Fraction of total uranium in soluble/insoluble species Potential for radon emanation Equilibrium or nonequilibrium between decay products
Monitoring	Spatial extent of contamination Depth of contamination Levels of statistical confidence on monitoring results Determination of background levels of radionuclides