INVESTIGATION OF THE SOLUBILITY OF YELLOWCAKE IN THE LUNG OF URANIUM MILL YELLOWCAKE WORKERS BY ASSAY FOR URANIUM IN URINE AND IN VIVO PHOTON MEASUREMENTS OF INTERNALLY DEPOSITED URANIUM COMPOUNDS

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INTRODUCTION

Evaluation of occupational inhalation exposure to uranium is routinely performed using the results of bioassay measurements for uranium in excreta and in vivo scintillation measurements for uranium deposited in the respiratory system of potentially exposed workers (1). Uranium toxicity is similar to other heavy metals, such as arsenic, lead, and mercury, and differs essentially in that uranium is an alpha particle-emitting radioactive material. Therefore, in order to establish an effective worker protection program at a uranium mill, both nephrotoxicity and the radiological hazard to the lung must be considered for the overall assessment of worker exposure.

Recent studies of the solubility of uranium in simulated lung fluid have demonstrated that yellowcake, the generic name of the end-product material from the uranium milling process, does not have a unique solubility classification (2). The solubility of yellowcake has been shown to depend upon the specific chemical separation process employed at the mill (3). That is to say, yellowcake solubility is influenced by the chemical form of the material, the method of chemical extraction of the uranium from the ore, and the temperature at which the yellowcake is calcined or dried. To further exacerbate the lack of a unique solubility classification for yellowcake, each mill will vary its own chemical separation process, depending upon the ore quality, and produce a yellowcake material with slightly different physiochemical characteristics including solubility.

The lack of a specific solubility classification for yellowcake impacts the worker protection program at its most fundamental level. It prohibits development of a simple method for interpreting bioassay and in vivo measurements for uranium which are performed to assess the occupational exposure of workers who come into contact with yellowcake. It is due to the question of the solubility of yellowcake in the human lung that in vivo scintillation measurements for uranium in the respiratory system must be performed as an adjunct to the assay for uranium in excreta.

METHOD

The United States Nuclear Regulatory Commission, Division of Safeguards, Fuelcycle, and Environmental Research is sponsoring a study at the Pacific Northwest Laboratory (PNL) to assess the solubility of yellowcake in the human lung as determined from measurements of workers who were accidentally exposed on the job at a uranium mill. Extensive evaluation of accidental human yellowcake exposures, using

sequential measurements of uranium in excreta and in the respiratory system, should provide data to characterize the solubility of the inhaled material.

Analysis of excreta samples from uranium mill workers routinely indicate the presence of microgram amounts of uranium resulting, presumably, from low-level, chronic exposure at the mill. It is not possible to absolutely identify the source of uranium in these samples as arising from an actual worker inhalation exposure or from external contamination, such as dust in the mill environment.

The first phase of this study is to determine baseline uranium measurements in excreta samples from yellowcake and other mill workers. Intercomparisons of routine assay procedures at the mills and the methods employed at PNL are performed to relate records of past measurements to the analysis of the current program. Urine and feces are obtained from workers at many different uranium mills. Management personnel from several uranium mills in Wyoming, Washington, Colorado and New Mexico have been very cooperative with this voluntary program and have enthusiastically supported the study. Their assistance in reporting potential overexposure incidents is paramount to the success of the second part of this study, i. e., measuring yellowcake solubility in the lungs of an overexposed worker.

In order to alleviate the potential for external contamination during sample collection and assay volunteer workers are transported to PNL for collection of excreta samples under more controlled circumstances than are available at the mill. The travel arrangements also provide a period of time for recently inhaled soluble airborne particulate to be eliminated prior to initiating sequential sample collections (4). In other words, this phase of the research is an attempt to collect excreta samples for uranium assay in order to characterize any chronic exposure that may exist at the mills.

Measurement of any insoluble uranium material deposited in the respiratory system is performed by simultaneously detecting photon emissions from ^{235}U, ^{234}Th, and x-rays from uranium, protactinium, and thorium (5). Two dual-crystal NaI(Tl)/CsI(Tl) scintillation detectors are placed on the anterior thorax of the worker while he lays prone in a shielded room at the laboratory whole body counting unit. An overabundance of the $\approx$16 keV x-rays in an in vivo measurement for uranium in the lung indicates that some fraction of the material is located on the surface of the worker. Evaluation of the internal uranium deposition must be adjusted to eliminate the influence of surface contamination on the worker. Prior in vivo measurements for uranium deposited in the lungs of uranium mill workers have been performed by another researcher without special regard for the presence of external contamination (6). Although, in that study, workers were required to shower and wear coveralls, the presence of uranium bearing dust and imbedded soil on the skin could not have been entirely eliminated by washing.

Once baseline reference values are established for uranium in excreta and in the lungs of uranium mill workers, measurements will be performed with an overexposed yellowcake worker. Uranium excretion from the acutely overexposed yellowcake worker will be compared to baseline values determined from "unexposed" mill workers. The temporal relationship between uranium in urine and feces and the change in the lung burden of the overexposed worker will provide a basis for

describing the solubility of the inhaled material.

It is unlikely that any one of the volunteers who are measured during the initial phase of the program will later become contaminated with yellowcake as a result of an accidental overexposure at the mill. For this reason a representative number of volunteers from several mills must be measured according to the aforementioned protocol. The millworkers will typically spend three to five days at PNL and need only collect excreta and be available for in vivo measurements each day.

The schedule for the overexposed worker will require at least two weeks time at the laboratory for initial sample collections. Immediately following the inhalation incident the worker will be transported by air to PNL so that the metabolism and translocation properties of the rapidly soluble, class D material can be studied. Excreta samples will be collected for several weeks after this initial period. Follow-up in vivo measurements for uranium in the lung will be performed in order to identify any insoluble uranium material remaining in the lung following the early clearance.

DISCUSSION

A unique application (7) (8) of the dual-crystal detector to the in vivo measurement for uranium in the lung provides a mechanism to distinguish external uranium contamination from that actually deposited in the lung tissue. The thin (3 mm) NaI(Tl) scintillator in the dual crystal detector can identify the presence of uranium on the skin by measuring the ≈ 16 keV x-rays from U, Pa, and Th. Using this same scintillator, the 63 keV and 93 keV ^{234}Th photons indicate the presence of ^{238}U. The thicker (5 cm) CsI(Tl) scintillator simultaneously detects the ^{235}U photons at 186 keV.

In order to take advantage of this technique, a surrogate thorax structure (phantom) is fabricated with a known amount of yellowcake uranium material deposited within the lungs. Two detectors are placed in contact with the surface of the anterior thorax, one detector centered over each lung. A standard ratio of the counts in the x-ray region to the number of counts in the ^{234}Th region is determined from a measure of the phantom. A similar ratio is calculated from an in vivo measurement of a potentially exposed subject. This ratio is first modified for chest wall attenuation and then compared to the surrogate thorax phantom. Detection of an abundance of 16 keV x-rays with the exposed subject indicates that skin contamination is present. The subject in vivo measurement is corrected to account for surface contamination so that the amount of uranium in the lung can be determined.

SUMMARY

Experience has shown that uranium has a low order of chemical toxicity in man and that other heavy metals, such as lead, arsenic, and mercury would produce severe injury at the same levels of exposure (9). However, without a precise knowledge of the solubility of yellowcake in the human lung its hazard to man from an inhalation exposure cannot be adequately determined.

Experiments to measure the solubility of yellowcake in simulated lung fluid indicate that the material exhibits some properties of each classification, i. e., D, W, and Y. This research study attempts

to measure the solubility of yellowcake in the human lung from analysis of the metabolic and translocation characteristics of the material in a worker who has been exposed at a uranium mill. The elimination of uranium from the body will be measured in urine and fecal samples collected from the exposed worker. In vivo scintillation measurements for uranium in the lung will be performed to determine the clearance of the material from the lung.

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