UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

APR 24 1986

MEMORANDUM FOR: John G. Davis, Director
Office of Nuclear Material Safety
and Safeguards

FROM: Robert B. Minogue, Director
Office of Nuclear Regulatory Research

SUBJECT: RESEARCH INFORMATION LETTER # 145 CONCENTRATIONS OF URANIUM AND THORIUM ISOTOPES IN URANIUM MILLERS' AND MINERS' TISSUES

INTRODUCTION

This letter transmits the results of a research project funded by RES to investigate the validity of the special intake limit for uranium ore dust that has been specified in 10 CFR Part 20 since the early 1960's. This limit was based on an assumption that thorium, a daughter product of uranium, is not preferentially retained in the lung in accordance with the ICRP lung model in use at that time. In the derivation of the special ore-dust limit it was assumed that the retention function used for uranium was also applicable to thorium. The special limit is a factor of 3.3 larger than the ICRP-2 limit for the ore-dust mixture. The results reported here indicate that the thorium daughter products are not preferentially retained in the human lung following the deposition of uranium, and that adequate protection is provided by the special ore-dust limit.

These conclusions arise from the results of measurements of the concentrations of alpha-emitting isotopes of uranium and thorium in the lungs of 14 former uranium miners (and in the soft tissues and bones of three former uranium miners and two former uranium millers). The research project was initiated in response to a request from the former Office of Standards Development, RR-OSD-79-5, for human data relating to AEC/NRC special MPC for uranium ore dust. The work was performed by Drs. MacDonald E. Wrenn and Narayani P. Singh of the Radiobiology Division of the University of Utah. The report describing the study entitled, "Concentrations of Uranium and Thorium Isotopes in Uranium Millers' and Miners' Tissues," NUREG/CR-4382, has been transmitted to your staff.

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BACKGROUND

Uranium ore dust is a mixture of several different radioactive nuclides, primarily those of the uranium-238 decay chain. The most important nuclides from a radiological health viewpoint are the alpha particle emitters with long half-lives. In determining the maximum permissible concentrations (MPC) in air, the accepted procedure for a mixture involves use of a formula to combine the individual MPC's. In 1960, while developing regulations currently in 10 CFR Part 20, the Atomic Energy Commission (AEC) staff did not believe that the thorium in uranium ore dust was as hazardous as calculation by the mixture technique showed. It was thought that the behavior of thorium in ore dust in the lung would be similar to that of uranium; i.e., its half-life in the lung would be more like 120 days rather than 4 years as prescribed in the ICRP-2 model.

In order to verify this assumption, the AEC initiated studies at Battelle Pacific Northwest Laboratories involving inhalation of uranium ore dust by animals. The experiments on three mammalian species all showed a rapid separation of thorium and uranium, with thorium remaining in the lung for much longer times. These results cast doubt on the validity of the AEC's special uranium ore dust standard.

In 1973, Dr. Victor Archer of the National Institute for Occupational Safety and Health reported the results of an epidemiological study of uranium mill workers. He found excess deaths due to malignant disease of the lymphatic and hematopoietic tissue, other than leukemia. Archer speculated that these deaths may have resulted from irradiation of the pulmonary lymph nodes by thorium-230.

In 1979, the Office of Standards Development requested a study to determine the uranium and thorium content of tissues obtained at autopsy from former miners and millers. The results, which provide evidence for answering the question regarding the half-lives of uranium and thorium in the lung, are presented here.

In 1983, NRC published NUREG-0941, "The NRC's Limit on Intake of Uranium Ore Dust." This report evaluated other aspects of this question, including the results of numerous biological, physical, and chemical studies related to the MPC for uranium ore dust. It was found that ore dust particles collected from the air in uranium mills were very large (10 micron activity median aerodynamic diameter). Particles this large are trapped in the upper regions of the respiratory tract, subsequently swallowed, and then rapidly excreted; relatively few are deposited in the pulmonary region of the lung where they would be subject to long-term retention. In this report it was concluded that the AEC's underestimate of the half-life of thorium in the lung was

counterbalanced by its overestimate of deposition of ore dust in the deep lung so that the special limit for uranium ore dust was adequate. The results published in NUREG/CR-4382 also confirm the conclusion that the present regulatory limit for ore dust provides adequate protection.

RESULTS AND DISCUSSION

The concentrations of U-238, U-234, U-235 and Th-228, Th-230, Th-232 in tissues of former uranium miners and millers are given in the tables in NUREG/CR-4382.

In the set of uranium miners' lungs the concentration of U-238 ranged from 1.6 to 311 pCi/kg wet weight with an average of 75 pCi/kg. The concentration of Th-230 ranged from 3.7 to 306 pCi/kg wet weight with an average of 79 pCi/kg. The concentration of U-234 ranged from 2.5 to 325 pCi/kg wet weight with an average of 80 pCi/kg. From these measurements it is apparent that thorium was not preferentially retained.

The tissue sets collected from three former uranium miners and from two former uranium millers were not totally consistent. In one miner the concentrations of U-234 and U-238 were highest in lung, followed by kidney. In the other miner the concentrations of these two isotopes were highest in kidney, followed by lung. The concentrations of these two uranium isotopes among other soft tissues such as liver, gonads, heart, and spleen were comparable in both miners. In the first miller, the concentration of U-238 was highest in lymph nodes, followed by lung. Other soft tissues, such as kidney, liver, and gonad contained similar concentrations. In the second, the concentration of U-238 was also highest in lymph nodes, followed by lung. The other soft tissues contained lower concentrations. In the first miller, the concentration of Th-230 was highest in lymph nodes, followed by lung, the same being found for the second miller. These results for the millers show that their pulmonary macrophages were highly efficient in transporting ore dust particles out of the lung and into the lymph nodes.

In all cases the relative concentrations of U-238 and U-234 in lungs were close to equilibrium. Similarly, the ratio of Th-230/U-234 was close to 1. When the time elapsed between the last exposure to ore dust and death is sufficiently long, the concentrations of uranium and thorium remaining in the lung are related to their biological half-lives. This condition has been met for the miners and millers in this study. Therefore, because the thorium and uranium activities were equal at the time of intake and at the time of measurement, their elimination rates must also have been equal.

CONCLUSIONS AND RECOMMENDATIONS

The results described above indicate that retention of thorium and uranium by the human lung are similar. Thus the theory originally adopted by the AEC

appears to be supported, and the lungs of the experimental animals appear not to be representative of the human lung.

A thorough staff investigation of every important aspect of this question has led to the conclusion that adequate protection is provided by the special uranium ore-dust limit presently in 10 CFR 20, (1×10^{-10} uCi/ml), when evaluated from the viewpoint of former ICRP models, parameters, and calculational methods.

The proposed revision to 10 CFR 20 specifies a derived air concentration (DAC) for uranium ore dust of 6×10^{-11} uCi/ml. This value was derived from the DAC's from ICRP-30 for each of the constituents of ore dust and does not involve NRC staff assumptions regarding lung retention of thorium. The resulting 40% reduction in the DAC may result in public comments regarding the influence of thorium on the new standard. If so, the results of this research effort will be used in the resolution of the comments.

All research on this project has been completed. For further information on this study, please contact Dr. Judith D. Foulke of the Radiation Risk Assessment and Management Branch (427-4563).

Robert B. Minogue

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Office of Nuclear Regulatory Research

APPENDIX

METHODOLOGY

Whole lungs from eight uranium miners were collected at autopsy; specimens were taken for detailed histological studies by a collaborator. Three sets of tissues from former uranium miners and two sets from former uranium millers were obtained at autopsy from a second source. Finally, five sets of tissues were obtained at autopsy from members of the general public in order to determine background concentrations of the alpha-emitting isotopes of uranium and thorium.

Weighed amounts of tissue spiked with U-232 and Th-229 tracers were wet ashed by adding enough HNO_3 to immerse the tissues, followed by slow heating. Additions of H_2SO_4 and H_2O_2 completed the removal of organic materials; HF was added if necessary, to dissolve silica.

After wet ashings, the residue was dissolved in 1:1 HNO_3 and 1:3 HCl. Then 0.2ml of iron carrier was added and the solution heated. Uranium and thorium were co-precipitated by adding concentrated NH_4OH. The solution was then boiled for 15 minutes, cooled, and centrifuged. The supernatant was discarded and the precipitate dissolved in dilute HNO_3. After removal of sulfate ions, the precipitate was dissolved in concentrated HCl.

The tissue solution was transferred into a polypropylene centrifuge tube. An equal volume of trilauryl amine (TLA) in xylene pre-equilibrated with 10 M HCl was added to the tube, shaken for ten minutes, centrifuged and transferred to a separatory funnel. The aqueous phase was mixed with an equal volume of 20% TLA solution and the extraction repeated once more. Uranium was extracted into the organic phase, leaving thorium in the aqueous phase.

Since the aqueous phases obtained after the back-extraction of uranium also contained iron, isopropyl ether was used to transfer iron to the organic phase and leave uranium in the aqueous phase.

The aqueous phase containing the thorium was evaporated to dryness. The residue was dissolved in HNO_3 and transferred to a centrifuge tube. An equal volume of 20% TLA pre-equilibrated with 4M HNO_3 was added. The mixture was shaken for ten minutes, centrifuged and transferred into a separatory funnel. After several back-extractions, the thorium was found in the aqueous phase.

Sodium bisulfate solution in 9M H_2SO_4 was added to the solutions containing uranium and thorium. These solutions were heated strongly with occasional additions of HNO_3 to remove the last traces of organic matter and evaporated to dryness. The residue was dissolved in $(NH_4)_2SO_4$ solution and transferred into the plating cell. Uranium and thorium were electrodeposited onto platinum planchets at a constant current of 1.2 ampere for 1 hour. The platinum planchets were then counted alpha-spectrometrically.

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