ANALYSIS OF 'HIGH-FIRED' PLUTONIUM OXIDE IN TISSUES OF EXPOSED WORKERS

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The United States Transuranium and Uranium Registries (USTUR) studies actinide (Pu, Am, and U) biokinetics and tissue dosimetry by following up former nuclear workers with documented internal deposition of these radioactive elements. The USTUR research relies heavily upon postmortem autopsy findings and radiochemical analyses of donated tissues. The use of hydrofluoric (HF) acid is critical for the analyses of samples contaminated with highly insoluble plutonium material - 'high-fired' plutonium oxide ($^{239}PuO_2$). ^{239}Pu activity concentrations were measured in lungs and thoracic lymph nodes from 291 occupationally exposed individuals. Plutonium insoluble material was present in ~50% of the analyzed respiratory tract tissues collected by the USTUR.

INTRODUCTION

The mission of the United States Transuranium and Uranium Registries (USTUR) is to study the uptake, translocation, retention and excretion (biokinetics), and tissue dosimetry of U, Pu, Am, and other actinides in occupationally exposed volunteer Registrants (tissue donors). These individuals were exposed to various types of radioactive material, including highly insoluble 'high-fired' aerosol particles. The USTUR research relies heavily upon postmortem autopsy findings and radiochemical analyses of tissues. Tissue analysis provides data on actinide distribution, retention, and radiation dose estimation from internally deposited radionuclides. The USTUR Radiochemistry Laboratory analyzes from 500 to 600 human tissue samples annually.

At the USTUR, HNO_3-HCl-HF acid mixture is routinely used for soft tissue sample digestion, including respiratory tract tissues, where insoluble forms of Pu material and silica particles are likely present. The HNO_3-HCl (reverse aqua regia) is used for bone samples digestion, where Pu is incorporated in a soluble form, and HF cannot be used due to formation of CaF_2 precipitate with a bone matrix. Sample leaching with HNO_3-HCl on a hot plate is often used as a method of choice for Pu analyses in environmental samples. Recent results of the Mixed Analyte Performance Evaluation Program (MAPEP) exercise for actinide analyses in soil revealed that a substantial number of participants failed on ^{239}Pu analysis due to its presence in a 'high-fired' oxide form.

ANALYSIS METHODS AND RESULTS

Analysis of 'high-fired' $^{239}PuO_2$

The USTUR performed a study to verify standard analytical protocol used by the Registries for ^{238}Pu and ^{239}Pu analyses in human tissue samples. The reference soil (MAPEP-11-MaS24) certified for ^{239}Pu, ^{241}Am, ^{234}U, and ^{238}U concentrations and plutonium in the form of 'high-fired' $^{239}PuO_2$ was used. Samples of 0.5, 1.0, and 2.0 g were collected in triplicate and were microwave digested at a control temperature of 200 ºC and monitored pressure of 40 bar for 20 minutes using concentrated HNO_3-HCl-HF or HNO_3-HCl acid mixtures. After the digestion, samples were spiked with ^{242}Pu, ^{243}Am, and ^{232}U tracers and actinide separation was carried out using TEVA-TRU-DGA extraction chromatographic column consisting of 1-ml cartridges. Following electrodeposition, activities of Pu, Am, and U fractions were measured by α-spectrometry.

Regardless of sample size, complete soil digestion was achieved with HNO_3-HCl-HF, while HNO_3-HCl yielded insoluble residue.

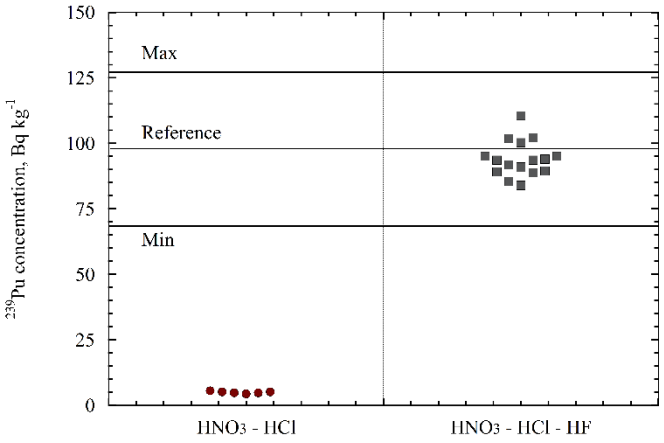

Fig.1. Results of 'high-fired' $^{239}PuO_2$ analyses in MAPEP soil.

Figure 1 demonstrates that only 5% of the initial ^{239}Pu was dissolved and recovered from soil matrix using HNO_3-HCl (10:3 ml), while HNO_3-HCl-HF (9:3:4) allowed complete dissolution of 'high-fired' plutonium oxide.

^{239}Pu in the Respiratory Tract Tissues

Respiratory tissue samples from 291 voluntary donors were analyzed using standard USTUR analytical protocol. The activity concentrations of ^{239}Pu were measured in 288 lung tissues and 265 thoracic lymph nodes (LNTH). The ^{239}Pu activity concentrations in the lungs ranged from 0.55 mBq kg^{-1} to 7.23 kBq kg^{-1} (geometric mean: 1.4 Bq kg^{-1}); in LNTH from 1.79 mBq kg^{-1} to 68.4 kBq kg^{-1} (geometric mean: 20.8 Bq kg^{-1}). Figure 2 summarizes ^{239}Pu activity concentration distribution in the lungs and LNTH of exposed workers.

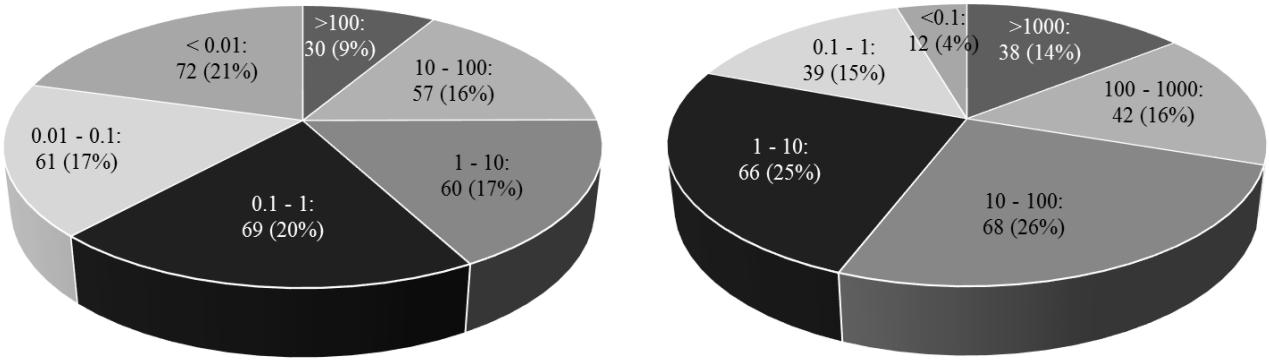

Fig.2. ^{239}Pu concentrations (Bq kg^{-1}) in the lungs (left) and thoracic lymph nodes (right) of exposed workers.

The LNTH-to-lung activity concentration ratios were calculated for 257 cases. The ratios ranged from 0.07 to 561, with geometric mean of 14. In 48% of cases, this ratio was greater than 19, the value predicted by ICRP Human Reparatory Tract Model for inhalation of insoluble, type S, material.[1]

CONCLUSIONS

The use of hydrofluoric (HF) acid is critical for analyses of samples contaminated with highly insoluble plutonium material such as 'high-fired' plutonium oxide ($^{239}PuO_2$). ^{239}Pu activity concentrations were measured in the lungs and LNTH from 291 occupationally exposed individuals. Plutonium insoluble material was present in ~50% of the analyzed tissue samples.

ACKNOWLEDGMENTS

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REFERENCES

1. International Commission on Radiological Protection, *Dose coefficients for intakes of radionuclides by workers*. ICRP Publication 68, Pergamon Press, Amsterdam (1994).