THE EFFECT OF OXIDATION STATE ON THE ABSORPTION OF INGESTED OR INHALED PLUTONIUM

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Larsen and Oldham found that chlorine, at the concentrations found in municiple water supplies, can oxidize quadrivalent plutonium to its hexavalent state (1). Since studies in this laboratory had shown one thousand times more ^{239}Pu(VI) was absorbed than ^{239}Pu(IV) (2), they suggested that the maximum permissible concentrations (MPC), apparently based on data from Pu(IV), should be lowered.

Our initial experiments performed with ^{238}Pu(VI) nitrate did not support those earlier results. This suggested that the conditions of fasting and oxidation used in those studies may have been responsible. Absorption of plutonium either by gavage or inhalation was compared in fasting and nonfasting rats to determine if the intestinal contents influence absorption of plutonium that was injected intragastrically or swallowed as a result of clearance from the lungs.

METHODS AND MATERIALS

Wistar female rats weighing about 200 g received plutonium nitrate (pH2) by gavage or by nose-only exposure from a nitric acid aerosol generated by a Lovelace nebulizer (3). Fasted rats were deprived of food 18 hours before Pu gavage and for 72 hours following it. Animals exposed by inhalation were fasted either before, or both before and after treatment with Pu. Excreta was collected daily from gavaged rats for four days.

All animals were killed five days after treatment. Femurs were removed from all animals and the total skeletons analyzed from many with which a femur factor was derived to determine total bone Pu content. The skin and GI tracts were discarded. Carcass values were determined by a summation of bone and soft tissue, excluding the liver and lung values. High lung values in gavaged animals indicated poor injections and the data from these animals was rejected.

The ^{239}Pu was purified by anion exchange on a Dowex MSA-1 resin and oxidized to its hexavalent state by passing a stream of O_3, O_2 and Ar through a 0.4M HNO_3 solution for six hours. It was shown to be 100% ^{239}Pu(VI) by spectrophotometric analysis. Solutions that included the holding oxident $K_2Cr_2O_7$ were made 0.015 M by dilution. Gavaged animals received 1.0 ml of a solution containing 0.5 mg ^{239}Pu, 97% of which was filterable through a 0.01 μm filter at the time of treatment. The dose administered by gavage was 30 μCi and by inhalation 5 μCi.

Plutonium was analyzed by a modification of the Keough and Powers method (4). Carbon-free aliquots in mixture of 1.0% boric acid were mixed in a scintillation solution containing 1,4 bis-2-[5 phenyloxazolyl] benzene, 2,5,-diphenyloxazole (PPO), Triton X-100 toluene and water.

RESULTS

The data obtained by Weeks et al (2) after intragastric administration of $^{239}Pu(VI)$ are shown in Figure 1 along with data that we obtained simulating conditions used by those investigators. Other

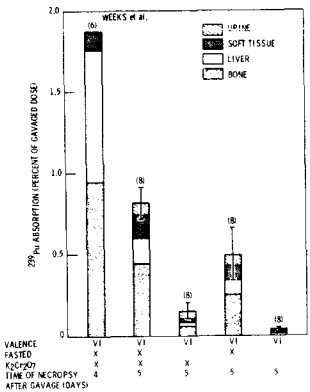

Figure 1. Absorption of $^{239}Pu(VI)$ by fasted and unfasted rats after intragastric administration of ^{239}Pu nitrate.

groups are included to show the effect that ad libitum feeding and an absence of the holding oxident, $K_2Cr_2O_7$ had on ^{239}Pu absorption from the GI tract. The fasting period lasted from 18 hours before gavage until 72 hours thereafter. Our absorption data under those conditions amounted to about half that of Weeks, possibly because their solutions were more acid, pH 1 versus pH 2, and their $K_2Cr_2O_7$ may have been more concentrated. Feeding reduced absorption 18-fold and the combination of feeding and elimination of dichromate from the solution reduced it about 26-fold.

Results obtained by exposing groups of rats to aerosols of either $^{239}Pu(IV)$ or $^{239}Pu(VI)$ (Figure 2) indicate that there was increased retention of plutonium after exposure to $^{239}Pu(VI)$ in comparison to $^{239}Pu(IV)$. The absence of food either before, or both before and after the inhalation exposure had no effect on the amount of ^{239}Pu retained by the liver and carcass.

DISCUSSION

Plutonium in its hexavalent state may under certain conditions be more readily absorbed from the GI tract. However, the experimental conditions in which increased uptake occurred, i.e. fasting for 90 hr, high acidity (pH 1-2) and a dose of 130 μCi/kg, are unlikely to occur in human exposure. The oxidation state may also influence absorption after inhalation because of the large fraction entering the bowel as a result of swallowing Pu cleared from the

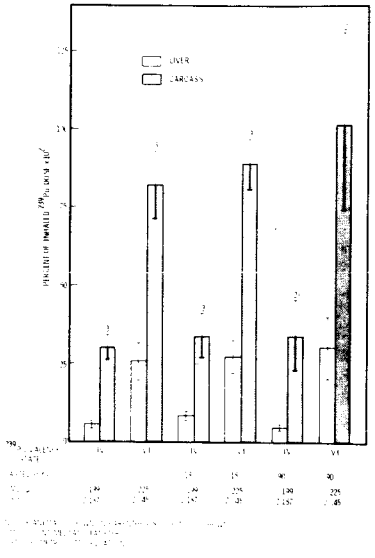

Figure 2. Retention of ^{239}Pu by fasted and unfasted rats after inhalation of either $^{239}Pu(IV)$ or $^{239}Pu(VI)$ nitrate.

lung. Although the Pu dose inhaled was lower than the dose gavaged (25 μCi/kg) some of it probably entered the stomach in its hexavalent state. Absorption from the GI tract was not higher than when $^{239}Pu(IV)$ was inhaled, even when the intestinal contents were depleted by fasting. The absence of an effect of food deprivation suggests that the increased absorption of plutonium was due to translocation of $^{239}Pu(VI)$ from the lung and not to an increase in transport across the GI tract.

This data support the observation made in the gavage studies that the MPC for drinking water, apparently based on the absorption of Pu(IV), is adequate for Pu(VI) ingestion in quantities that may be expected in the environment. Absorption of Pu(VI) from the lung, however, is higher than that of Pu(IV).

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