APPENDIX C
ENVIRONMENTAL EFFECTS OF PLUTONIUM DIOXIDE

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This appendix addresses the potential impacts from plutonium dioxide (PuO_2) released to the environment, which could occur in any of the representative, low-probability accident scenarios described in Section 4. The health and environmental risks associated with plutonium (mainly Pu-238) dioxide were previously addressed in the Galileo and Ulysses EISs (NASA 1989b, NASA 1990). This appendix briefly describes the general physical and chemical characteristics, transport, and biological effects of plutonium dioxide.

C.1 PHYSICAL AND CHEMICAL CHARACTERISTICS OF PLUTONIUM DIOXIDE

Plutonium is one of the most widely studied elements in terms of chemistry and environmental behavior. Although its chemistry and oxidation states are quite diverse, the element's environmental mobility is very limited (INSRP 1989a).

The extent and magnitude of potential environmental impacts caused by PuO_2 releases depend on the mobility and availability of PuO_2 in the environment. The mobility and availability of PuO_2 in turn, are directly controlled by a number of physical and chemical parameters, including particle size, potential for suspension, deposition and resuspension, solubility, and oxidation state of any dissolved plutonium. These factors, in conjunction with the three potential exposure pathways (i.e., direct external exposure from ground-deposited material, ingestion, and inhalation), determine the potential impacts. Of the three pathways, only the potential for direct exposure from ground-deposited material or surface contact is not significant, because alpha radiation cannot pass through more than a few inches of air.

The size of plutonium dioxide particles can affect the rate of dissolution in water and the initial deposition and subsequent resuspension of particles in both air and water. The dissolution and the suspension and resuspension potential ultimately control the mobility and availability of PuO_2 to Plant and animal species including man. Generally speaking, larger particles have less potential for suspension and resuspension; as the particle size decreases, particles are more easily kept in suspension or resuspension.

A number of factors can affect the solubility of PuO_2 in water. Physiochemical parameters most important to the solubility of PuO_2 are the reactive surface area and oxidation state of plutonium and the water chemistry, including pH, reduction/oxidation potential, and temperature. The mass to surface area ratios of particles affect reactivity and solubility, with solubility being inversely related to particle size. The dissolution rate of the plutonium dioxide fuel is very small, ranging from 1.2 to 90 $\mu Ci/m^2/s$ (0.1 to 7.3 $\mu g/m^2/s$) in seawater and freshwater, respectively, based upon the dissolution rate per unit surface area of the fuel (NASA 1990, INSRP 1990). In general, PuO_2 is insoluble.

Chemically, PuO_2 is extremely resistant to dissolution, including dissolution in the environment as well as in lung or digestive fluids of the human body. PuO_2 is slowly removed from environmental pathways by sedimentation processes, percolation into soil, and other physical means.

Clays, organics, and other anionic constituents tend to bind most of the PuO_2 particles in the soil column. The binding of PuO_2 would occur in the first few centimeters of sediment, greatly reducing the concentration of this constituent with depth. This natural filtering of PuO_2 would probably reduce the concentrations in drinking water to levels below the Primary Drinking Water Standard of 4 mrem/year (NASA 1990).

It is also possible that surface water runoff containing PuO_2 could directly contaminate drinking water supplies from surface water bodies, because this type of contamination is greatest due to suspended PuO_2 particles and not from dissolved PuO_2. Filtering the surface water before chemical treatment would reduce the concentration of total plutonium to very low levels (NASA 1990).

C.2 MODELS USED TO STUDY TRANSPORT AND EFFECTS OF PLUTONIUM DIOXIDE

PuO_2 including its transport in the environment, its uptake in the human body by ingestion or inhalation, and its fate following uptake, has been studied in great detail over the last 50 years. Models have been developed to determine the radiation dose from plutonium transport and uptake (e.g., NUS 1982, ICRP 1977, ICRP 1979, ICRP 1990). When developing these pathway, dosimetry, and risk models, the following factors are usually considered:

1 . PuO_2 enters the environment and a fraction of it is transported via air, groundwater, surface water, or foodstuffs to humans. Because Pu-238 has a relatively long radioactive half-life (an initial quantity of Pu-238 will decay to 50 percent in 87.75 years), only a small fraction of it will be removed from the environment by radioactive decay.

2. A quantity of the PuO_2 is inhaled or ingested and a fraction of it is transferred to the bloodstream and then to organs within the human body, retained in the lungs, or excreted. It is assumed for this analysis that other entry mechanisms in the body, such as injection, are not significant.

3. The plutonium that resides in certain organs, principally the lungs, liver, and bone surfaces, is retained for a long period of time with a slow rate of excretion.

4. Alpha radiation, characteristic of Pu-238 irradiates nearby cells and cell components, such as chromosomes, and a fraction of the cells are killed or damaged. Most non-lethal damage is completely repairable by the cell.

5. A very small fraction of the damaged cells survive, undergo defective repair, mutate, and, may after many years delay, produce significant detrimental effects in humans, including cancer and genetic abnormalities. This fraction is the basis for the associated health risks discussed in the following sections.

Due to the extremely small amount of plutonium transported to and accumulated in the human body from exposure to a release from a postulated Cassini accident and the stochastic (random) nature of the detrimental effects produced in irradiated cells, it is not

possible to accurately predict the long-term effects to any one individual exposed during the postulated accident. However, it is possible to use the risk estimates experienced in a large exposed population to provide an estimate of the average risk to an individual (National Research Council 1988, National Research Council 1990). Detrimental effects, such as an increased rate of cancer, may possibly be predicted for a very large population, on the order of several millions of exposed individuals. From such estimates the average risk to a member of that population may also be calculated.

Dose equivalents to critical organs and tissues for all members of the general public exposed to the worst case postulated accident during the Cassini mission would be many orders of magnitude below those that produce acute effects, such as "radiation sickness," and even subtle acute effects, such as changes in blood chemistry, should not be detectable.

C.3 TRANSPORT OF PLUTONIUM DIOXIDE IN THE ENVIRONMENT

The transport mechanisms and pathways of any accidental releases of PuO_2 in the environment will depend on the mission phase and the subsequent environment in which the release occurs. For example, if the accident occurs near the launch pad, the concentrated release would primarily result in the formation of larger, more environmentally inert particles that would be deposited in a relatively small area within a short period of time. Conversely, should the release occur during the short-term reentry scenario, a widely dispersed cloud of smaller particles would gradually fall to Earth over a much larger area in much lower concentrations.

Because PuO_2 is so insoluble, the movement through the environment depends on physical processes. PuO_2 may be carried into the soil by a number of routes, including percolation of rainfall and subsequent leaching of particles into the soil, animal burrowing activity, and plowing or other disturbance of the soil by humans. Migration of the PuO_2 particles into the soil column is of concern, primarily because of the potential for PuO_2 to reach groundwater aquifers used as drinking water supplies. Once deposited on soil, however, PuO_2 appears to be extremely stable. Soil profile studies have shown that generally more than 95 percent of the PuO_2 from nuclear weapons fallout remained in the top 5 cm (2 in.) of surface soil (in undisturbed areas) for 10 to 20 years following deposition (DOE 1987b).

In the unlikely event of an accident, PuO_2 would be taken tip in the human body primarily via inhalation with ingestion of contaminated water and foodstuffs as a secondary means. These pathways and the generalized behavior of Pu-238 in the environment are described in the literature (e.g., Aarkrog 1977, Anspaugh et al. 1975, Pinder and Doswell 1985, Pinder et al. 1987, Yang and Nelson 1984).

In general, PuO_2 is insoluble and is poorly transported in aquatic, marine, and terrestrial environments. Most forms of plutonium, including PuO_2, are removed from biological pathways by processes such as sedimentation and fixation in soil. Only small amounts of material are concentrated by biological accumulation into most seafood, grazing animals, and other food products.

In marine and aquatic systems, larger particles will quickly settle to the bottom sediments; smaller silt-size particles may remain in suspension within the water column indefinitely. Smaller particles may not even break the water surface (due to surface tension), forming a thin layer on the water surface and subsequently transported to the shoreline by wind and wave action. Resuspension of smaller particles from the bottom can occur due to physical disturbance of the sediments by wave action and recreational use of the water bodies (e.g., swimming, boating, and fishing), as well as by the feeding activity of various marine and aquatic species. Particles of PuO_2, as a component of the bottom sediments, may also be transported toward and along the shoreline by wave action and currents in near-shore environments (NASA 1990).

PuO_2 entering into a water/sediment system would be preferentially taken out of solution and bound in saturated sediments in amounts on the order of 100,000 times greater than the amounts that would remain in the associated water column (NASA 1990).

Studies have indicated that the bioaccumulation in marine organisms can range from 2 to 3,000, depending on the type and density of seafood impacted (e.g., freshwater fish, saltwater fish, mollusca), the amount of radioactive material released, and the deposition area.

Parameters used for estimating the uptake from harvesting and consumption of agricultural products have been measured (Baes et al. 1984, Rupp 1980, Yang and Nelson 1984). These and similar agricultural and food consumption parameters and plutonium ingestion parameters (ICRP 1979) are used as the basis for estimating human doses via ingestion. For example, an analysis of Pu-238 contamination of orange trees shows that a total of only 1 percent of the plutonium intercepting the plants would be transported from field to market during the following 12 months of harvesting (Pinder et al. 1987). Most of this plutonium would adhere to the fruit's peel and would be removed prior to ingestion; uptake to the orange itself would be extremely small or nonexistent.

C.4 TRANSPORT AND DEPOSITION OF PLUTONIUM DIOXIDE IN THE HUMAN BODY

Plutonium dioxide that enters the human body by ingestion or inhalation has many possible fates, all of which have been studied in detail (ICRP 1979, ICRP 1986). The inhalation route is found to be approximately 1000 times as effective as ingestion in transporting plutonium to the blood, due to the short time of residency, the chemical properties of plutonium, and the physiological environment of the GI (gastro-intestinal) tract (ICRP 1979).

Ingested plutonium dioxide will quickly pass through the digestive system and be excreted with only a small quantity being absorbed via the mucosa into the blood stream. The fractional absorption of PuO_2 is estimated to average about 10^{-5} (i.e., about 1 part in 100,000 ingested would be absorbed) (ICRP 1979, ICRP 1986). The fractional absorption is based on the average individual. PuO_2 in the environment could become more soluble with time due to the use of fertilizers in gardening, chlorination in drinking water, and soluble forms in seawater. Dietary and physiological factors, such as fasting, may increase or decrease the fractional absorption.

Inhaled plutonium dioxide is transported to one or more portions of the respiratory system depending on the particle size. Generally, most particles larger than 5 micrometers are intercepted in the nasopharyngeal region and either expelled or swallowed to pass through the digestive tract; what is not absorbed, is then excreted. Particles smaller than about 5 micrometers are transported to and remain in the trachea, bronchi, or deep lung regions. Particles reaching the deep lung are cleared from the body much more slowly than those not entering the lung. For example, 1.4-micrometer particles are typically cleared from the lung at the rate of 40 percent in the first day and the remaining 60 percent are cleared in 500 days (ICRP 1979). Particles captured in the mucous lining of the upper respiratory tract are moved more rapidly to the pharynx, where they are swallowed. Once swallowed, they behave as if ingested.

Plutonium dioxide remaining in the lung will continuously irradiate lung tissue, and a small fraction will be transported over time directly to the blood or to lymph nodes and then to the blood. The fraction of plutonium transferred directly from lung tissues to the blood is believed to be about 1 percent of the amount retained in the lungs, depending on the size distribution of ultra-fine particles. Smaller particles are likely to form over time from larger particles due to the natural fragmentation processes associated with radioactive decay and may also be transferred to the blood. Approximately 15 percent of the plutonium initially deposited in the lungs will be transferred to the lymph nodes, and up to 90 percent will eventually be transferred to the blood (ICRP 1986).

Once PuO_2 has entered the blood via ingestion or inhalation, it circulates and is deposited primarily in the liver and skeletal system. It is currently accepted that plutonium transported by the blood is distributed to the following organs: 45 percent in the liver, 45 percent in the skeletal system, 0.035 percent in testes in males, and 0.011 percent in ovaries in females with a non-measurable amount crossing the placenta of pregnant women and entering the fetus. The remaining 10 percent of the activity in the blood is excreted through the kidneys and colon or deposited in other tissues (ICRP 1979, ICRP 1986).

The resident times in the liver, skeletal system, and gonads are quite long. Current estimates for 50 percent removal times for plutonium are 20 years for the liver, 50 years for the skeleton, and an infinite retention time for the gonads.

C.5 CANCER INDUCTION AND GENETIC EFFECTS

Several possible outcomes to the ejection of an alpha particle from a decaying Pu-238 nucleus may occur to a nearby cell:

1 . The alpha particle entirely misses the cell and has no damaging effect.

2. The alpha particle strikes the cell but does riot strike critical components within the cell, and the cell survives.

3. The alpha particle strikes a critical component of the cell and quickly kills it. The same may be true of striking and chemically changing water molecules, which act to damage critical cell structures.

4. The alpha particle or reaction products strike a portion of a chromosome within the cell, modifying the chromosome but not killing the cell. Most chromosome damage is correctly repaired, but a small fraction is incorrectly repaired (a mutation).

Generally, the last outcome, which has an extremely low probability of occurrence, is the most potentially harmful. This process may lead to the induction of cancer or genetic effects, which may be passed on to offspring (ICRP 1990). Damage may occur at any time following large uptakes of plutonium, and the detrimental effects may occur immediately or be delayed years after the initiating event in cell DNA. Because of the very small amount of activity ingested or inhaled by the average exposed individual and transported to internal tissues where effects may occur, however, the probability of this outcome for all accident scenarios is exceedingly small.