## Summary regarding air monitoring methods and results at Rocky Flats Plant Site

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The Department of Energy (DOE) and its contractors have previously presented detailed information describing the ambient air sampling program employed at Rocky Flats Plant Site. To summarize, ambient air samplers located on the plant site, at the plant perimeter, and in surrounding communities monitored airborne dispersion of radioactive materials from the Rocky Flats Plant Site into the surrounding environment. As of 1992, for example, these samplers were positioned at 23 locations on the plant site, at 14 locations around the plant boundary, and in 11 communities. The Colorado Department of Health (now the Colorado Department of Public Health and Environment, CDPHE) also maintained an independent sampling network with a different instrument design in and around the plant site to verify the Rocky Flats Plant Site data. The Rocky Flats (DOE) samplers were updated in the latter half of the 1990s, providing the ability to determine the contribution of smaller respirable particles in the particulate samples.

The samplers continued to use high-efficiency filters to capture ultra-fine and small particles that are known to be respirable by humans. Larger particles were captured separately to provide information about total airborne radioactive particles. The new sampler also improved slightly the ability of the sampler to capture larger particles, compared to the standard CDPHE samplers and the older Rocky Flats samplers. The data were reported in various public forums and in annual air emissions reports including a report to the U.S. Environmental Protection Agency (EPA) and the CDPHE to demonstrate DOE's compliance with Radioactive National Emission Standards for Hazardous Air Pollutants (Rad-NESHAPs).

The plutonium contamination that resided in the soils at Rocky Flats Plant Site (and still resides in very low concentrations in limited areas, such as downwind from the former 903 Pad site) is known to be attached to soil particles and to the extent it exists will be moved with that soil via soil disturbance and subsequent wind erosion events. Plutonium in the environment exists in an insoluble form and can present no significant potential hazard other than through the air pathway. All of the air monitoring data collected at the site demonstrates there to be no potential for significant emissions from the remaining surface soils, as defined by the established federal standards.

Filters used to collect the particles suspended in the air at Rocky Flats Plant Site were nearly 100% efficient, meaning that they captured nearly all of the airborne particles (including those from airborne soils and plant processes) in the sizes ranges that are significant for human health. Any small loss of efficiency would not change the results of any air monitoring analyses performed.

The air monitoring data collected at Rocky Flats Plant Site are well characterized with results from years of monitoring that suggest there is no significant airborne risk from residual plutonium in surface soils.

## **Detailed Discussion**

Previously presented information about the ambient air monitoring network and the particulate matter samplers employed at Rocky Flats Plant Site will not be presented in detail here. The information presented here is to reiterate what is known about the efficiency of the samplers to collect ultra-fine particles and to put that information in context with the standards established to be protective to members of the public.

Extremely small particles (typically those smaller than 0.01 micrometers [microns] diameter) are captured with near 100% efficiency by diffusion to the filter fibers, no matter what type of fibrous filter is employed, as a number of research papers indicate. This is true both of the samplers employed by DOE at the Rocky Flats site and the EPA "high volume" samplers used by CDPHE. J. Steffens and J. R. Coury, for example, note that: "*A number of studies during the past decade have advanced the theoretical understanding of fibrous filtration and some thorough reviews are available.* ... *These studies have shown that, within the sub-micrometer particle size range, diffusion is the dominant collection mechanism and particle bouncing/reentrainment are considered negligible.*" (From "Collection Efficiency of Some Fiber Filters Operating on the Removal of Nano-sized Aerosol Particles: I – Homogenous Fibers," *Separation and Purification Technology*, Vol. 58, Issue 1, December 2007, pp 99-105)

Particle sampling on filters occurs through several capture mechanisms. Chiu-Sen Wang and Yoshio Otani describe these mechanisms as follows: "As an aerosol stream approaches a fiber, particles may deposit on the fiber by the simultaneous action of several mechanisms, including inertial impaction, interception, Brownian motion [diffusion], gravitational settling, and electrostatic forces. The first four mechanisms ... are known as mechanical capture mechanisms. Inertial impaction occurs when a particle, by its inertia, departs from the original gas streamline and hits a fiber. Interception takes place because a particle has a finite size, which leads to deposition when it comes within one particle radius of the fiber surface, even though it stays on the original streamline. For particles smaller than a few tenths of a micrometer, the Brownian motion can be sufficiently strong to move them from the original streamline to a fiber. Gravitational setting can contribute to particle collection if the aerosol stream flows downward." ("Removal of Nanoparticles from Gas Streams by Fibrous Filters: A Review," Industrial & Engineering Chemistry Research, Vol. 52, Issue 1, 2013, pp 5-17)

Larger particles are captured primarily via the mechanisms of impaction and interception, depending on their relative size. The least efficient sampling is in the range where interception and diffusion have near equal efficacy, around 0.15 microns aerodynamic diameter. (The aerodynamic diameter is the diameter of a sphere with density of 1 gram per cubic centimeter that has the same terminal settling velocity under gravity as the airborne particle considered.) In this particular particle size range, the filters lose a small fraction of their efficiency. Note that the higher density plutonium particles by themselves would have a physical diameter about three times smaller than their aerodynamic diameter, with a consequent lower activity than would be estimated for a particle of true physical diamets.

It is also important to place the role of filter efficiency into a proper context. The filters employed in the ambient air samplers at Rocky Flats Plant Site typically have overall capture

efficiencies exceeding 99%. Whether we account for the additional <1% not sampled would have no impact on estimates of exposure and risk from the contaminants. In fact, the possible underestimate due to capture efficiency is less than any uncertainty introduced by the prescribed laboratory analysis protocols. From sampling that was performed by CDPHE at and around the Rocky Flats Plant Site, concentrations of plutonium were low enough that even with high volume sampling, there was not enough material to detect unless multiple-day samples were composited for analysis.

As is generally known, the majority of airborne plutonium at Rocky Flats Plant Site was derived from contaminated soils, reasonably estimated at more than 90% of total emissions at the site perimeter. Since other potential sources of emissions have been removed, any residual potentially airborne plutonium (100%) derives exclusively from contaminated soils. It is well documented that plutonium contaminants (mostly from the storage of contaminated cutting oils at the 903 Pad and subsequent soil contamination resulting from leaking metal drums) were dispersed around the 903 Pad during occasional high-wind events in the late 1960s before the remaining contaminated soil directly beneath the drum storage area was covered with an asphalt containment cover in 1969. Careful studies of these contaminants show that the plutonium became attached to the soil matrix and became airborne with those soils, not as separate plutonium particles. The typical size distribution of disturbed, airborne soil particles ranges from sub-micron to several tens of microns in aerodynamic diameter and is very efficiently captured in the types of air samplers that were deployed at the site. This understanding of the contaminant source further reinforces our confidence that airborne plutonium would not have been significantly under sampled even if the postulated low capture efficiency for ultra-fine particles were true.

Regarding the sampling systems themselves, in the mid to late 1990s, the ambient particulate matter samplers at Rocky Flats Plant Site were upgraded to characterize the size distribution of airborne materials, so that the respirable fraction of most concern in assessing exposure and dose could be distinguished from the larger-sized non-respirable fraction. It is well documented that plutonium dose would be derived almost exclusively through the inhalation of plutonium particles. When reporting the estimated exposure to a member of the public due to airborne radionuclide contamination to EPA and CDPHE, the site reported the exposure using the total suspended particulate result, even though only the respirable fraction would have contributed to that exposure (resulting in an approximately 1-2 fold over-reporting of exposure). In addition, the prescribed EPA method for estimating annual dose from airborne plutonium and other radionuclides at Rocky Flats Plant Site assumes the member of the public is immersed in the contaminated air for the entire year, at the point of maximum exposure, leading to additional conservatism in the present context of the site and its use.

Even so, the site levels were far below the requirements of the standard prescribed by 40 Code of Federal Regulations, Part 61, Subpart H (Rad-NESHAP) to limit exposure to any member of the public from DOE facilities (for example, during the period of active demolition and remediation which would have generated the greatest amount of airborne dust, annual dose was less than 3% of the Rad-NESHAP limit). After demonstrating that these levels were not exceeded for many years before and during the period of site demolition, including cleanup of many areas of soil contamination, and then for a period following that cleanup, sampling was terminated. Because

most potential sources of airborne plutonium emissions from the site, particularly the 903 Pad and surrounding lip area, have been removed and the area revegetated, plutonium emissions and subsequent dose are certainly less now than they were prior to completion of demolition and remediation. EPA has defined levels of airborne radionuclides that protect public health and the small amount of residual contamination in Rocky Flats soils would not result in airborne levels in excess of those limits.