^{239,240}Pu and Inorganic Substances in Aerosols from the Vicinity of the Waste Isolation Pilot Plant: The Importance of Resuspension

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Abstract— Aerosol samples from the Carlsbad, New Mexico area were collected and analyzed as part of a monitoring program designed to characterize the spatial and temporal variations in the concentrations of plutonium and selected inorganic substances in the atmosphere surrounding the Waste Isolation Pilot Plant (WIPP). High-volume aerosol sampling was conducted at three sites, (1) On Site, ~0.1 km northwest (downwind) of the WIPP exhaust shaft; (2) Near Field, ~1 km northwest of the WIPP; and (3) Cactus Flats, ~19 km southeast (upwind) of the WIPP. At the Near Field and Cactus Flats sites, concurrent high-volume samples of total suspended particulate (TSP) and particulate matter less than 10 micrometers in diameter (PM_{10}) were collected, but only high-volume TSP samples were collected at the On Site location. ^{239,240}Pu was determined in these samples by alpha spectrometry following chemical separations, and these nuclides were quantified in all but one of the 141 samples analyzed. A separate set of aerosol samples was analyzed for major ions and trace elements as a complement to the ^{239,240}Pu data, using ion chromatography (IC)and inductively-coupled emission spectrometry and mass spectrometry for the analyses, respectively. For these studies, low-volume TSP, PM_{10} and $PM_{2.5}$ (particulate matter less than 2.5 micrometers in diameter) samples were collected at Near Field and Cactus Flats, but again only TSP samples were collected at On Site. The IC analyses were done on aqueous extracts of Teflon® filters while the elemental analyses were done on acid (HNO_3, HCl, and HF) digests of cellulose ester filters. Gravimetric determinations were made only for the high-volume radionuclide filters.

The ^{239,240}Pu activity concentrations (that is, the activities per unit volume of air sampled) in the aerosol samples were consistent with those presented in Lee et al. (1998), but they varied substantially with season for both TSP and PM_{10}. The highest activity concentrations of ^{239,240}Pu

generally occurred in spring: this was true at all stations and for both types of samples. Furthermore, the ^{239,240}Pu activity concentrations were comparable among the three sites, and therefore there was no evidence for elevated ^{239,240}Pu activities due to WIPP operations. The fraction of the ^{239,240}Pu activity concentrations in the PM_{10} samples relative to TSP (50-55%) was lower than the corresponding PM_{10}/TSP ratios of high-volume mass or several inorganic analytes (sulfate, aluminum or lead), thus indicating that ^{239,240}Pu tends to be associated with relatively large aerosol particles.

The aerosol mass loadings (mass of aerosols collected per unit volume of air) followed a seasonal pattern similar to that of the ^{239,240}Pu activity concentrations, and a distinct positive linear relationship existed between these two variables for all samples from the Near Field and Cactus Flats sites. However, there was a clear difference in this relationship at On Site where the TSP samples showed a higher mass to ^{239,240}Pu concentration ratio than samples from the other sites. This result implies that activities or processes occurring at or near the WIPP site produced aerosols that contributed to the mass loadings but contained less ^{239,240}Pu than ambient aerosols. A second inference we draw is that local sources of non-radioactive particulate material can be an important parameter affecting the mass-based plutonium activity densities. Therefore measurements of activity calculated with respect to both particulate mass and air volume are useful in understanding the factors controlling the dynamics of ^{239,240}Pu in the atmosphere. It is also worth noting that the volume-based activity concentrations likely encompass larger error terms (than activity density) owing to uncertainties in air flow volume measurements and possible losses of particulate material from filters during handling.

Even though time intervals for the radionuclide and inorganic samples were not precisely matched, the ^{239,240}Pu activity concentrations were significantly, albeit weakly, correlated with Al concentrations averaged over the same time intervals. Aluminum can be regarded as an indicator of mineral dust, and the observed correlation indicates that resuspension of soils is an important determinant of ^{239,240}Pu activities in aerosols. Comparisons between the aerosol and soils data show that the ^{239,240}Pu activity to Al concentration ratios for the aerosols (all data = 3.3×10^{-2} nBq ng^{-1}) was much higher than in soils (approximately 4.0×10^{-5}, Kirchner et al., *in press*). This difference can be explained by the preferential binding of ^{239,240}Pu to small soil dust particles that have large surface area to mass (and volume) ratios and also have higher aluminum contents than

larger particles (which include a considerable amount of quartz). These small soil particles are large compared with aerosols; and yet they stay suspended in the atmosphere much longer than the bulk of the resuspended soil particles which are rapidly re-deposited by gravitational settling.

In summary, the results of this study suggest that processes affecting the resuspension of $^{239,240}Pu$ and possibly other substances are of great importance and must be known and quantified if any impacts of operations at the WIPP are to be properly evaluated. This is an important consideration because any potential releases of radionuclides or other materials from the WIPP will be superimposed on the variability caused by the winds and other meteorological factors responsible for the resuspension and re-distribution of contaminants deposited independent of WIPP operations. The WIPP environmental monitoring studies demonstrate how mass-based activity concentrations and ancillary inorganic data can support interpretations concerning the dynamics of plutonium and other analytes of interest in the environment. Moreover, for purposes of monitoring contaminants in the atmosphere, there are advantages in interpreting data for the analytes of interest within the broader context of changes in composition of atmospheric particulate matter. This approach goes beyond simply documenting environmental levels, leading instead to an understanding of why those concentrations change.

Literature Cited:

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