A dose reconstruction of air emissions from the Oak Ridge Gaseous Diffusion Plant, Oak Ridge TN, using historic air monitoring and emission data

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Abstract

As part of its mandated public health mission, ATSDR must determine whether historic air emissions from the Oak Ridge Gaseous Diffusion Plant represented a public health hazard to adjacent communities. ORGDP produced enriched uranium via gaseous or thermal diffusion of uranium hexafluoride (UF₆; which undergoes rapid atmospheric transformation into hydrogen fluoride and uranium oxides). The dose reconstruction was accomplished using established air dispersion models and site-specific meteorological and historic air monitoring data to verify the use of both the modelling tools and the emission estimates. Air concentrations and doses to radionuclides were estimated using the standard air dispersion models. Meteorological data are from multiple weather years for two on-site stations. The Department of Energy has been collecting environmental measurements in soil, air, and water since at least 1953. Two stations adjacent to ORGDP were sampled for airborne gross alpha particulates since at least the mid-1960s. With some simplifying assumptions, there is good agreement between the historic gross alpha concentrations and those predicted from dispersion modelling. When combined with health-protective exposure assumptions, the estimated emission data and dispersed air concentrations provide a reliable basis for the public health determinations at this site.

Keywords: uranium hexafluoride, historic monitoring, dispersion modelling, dose reconstruction, air emissions.
1 Introduction

In 1942, the federal government established the Oak Ridge Reservation (ORR) in Anderson and Roane counties in Tennessee (USA) as part of the Manhattan Project to research, develop, and produce special nuclear materials for nuclear weapons. The ORR included the K-25 and S-50 sites, which were created to enrich uranium. The overall goal of this dose reconstruction is to determine the potential public health hazard posed by historic uranium and fluoride air emissions from the K-25/S-50 facility. It will accomplish this goal by using the estimates of historic air emissions and established air dispersion models to reconstruct past doses to offsite residents. The estimated contaminant concentrations are compared with historic air monitoring data to ensure the adequacy of the dispersion models as a reliable basis for assessing potential offsite health hazards.

This dose reconstruction evaluates the potential health effects from K-25 and S-50 fluoride and uranium releases for the three communities with the highest potential exposures—Happy Valley, Sugar Grove, and Union/Lawnville (Figure 1). If historic exposures at these areas of maximum potential exposure are below levels likely to produce adverse public health effects it will be assumed that all other lesser exposures are also below levels of public health concern. The timeframe for the exposures evaluated are bounded by the operational history of the K-25/S-50 facility. Significant emissions from either S-50 or K-25 only occurred when those facilities were operating. S-50 operated for only a 12-month period in 1944 and 1945. K-25 operations began in 1945 and were discontinued in 1985. Consequently, this dose reconstruction will focus on potential off-site exposures (both chronic and acute) during the 1944 to 1985 time period for historic releases.

The primary airborne contaminant released from the K-25/S-50 sources was UF$_6$. UF$_6$ is a dense or heavy gas (heavier than air) at atmospheric temperatures and pressures. When released in the air, UF$_6$ reacts rapidly with atmospheric water to form hydrogen fluoride and uranyl fluoride and uranium oxide particulates. Initially, all of the UF$_6$ fed into the gaseous diffusion cascades was made from natural uranium. However, beginning in 1952 uranium that had been reprocessed from previously fissioned material (reactor tails) was introduced as UF$_6$ feed material and contained fission products and transuranic radionuclides including technetium 99 (Tc 99), neptunium 237 (Np 237) and very small quantities of plutonium 239 (Pu 239). Consequently, after 1952, airborne emissions from the K-25 facility also contained quantities of Tc 99 and Np 237 that are accounted for in airborne emission estimates. The very small quantities of Pu 239 account for less than one percent of the total radiation and are not included in subsequent radiological dose assessments.

Uranium emission estimates are from the 1959 to 1995 environmental reports, a historical summary [1], and the ChemRisk dose reconstruction (Task 6; [2]). Although the DOE and Task 6 release estimates were independently derived, they are based on the same underlying monitoring and incident release reports. The cumulative airborne uranium release, as estimated by DOE is about 8
percent larger than the Task 6 estimate and the highest annual releases (1961 and 1963), as estimated by DOE, are more than 30 percent larger than the Task 6 estimates for those years. Although the DOE estimate of total airborne uranium releases is more conservative than the Task 6 estimate, the Task 6 emissions data is more complete in that it includes the relative composition of the uranium isotopes (U 234, U 235, and U 238) for each of the release years. Subsequent analyses will use the DOE estimate of total uranium emissions and the Task 6 estimate of uranium isotope proportions.

The long-term or annual uranium release estimates represent the sum of individual release events for each year. One of the specific tasks of this dose reconstruction is to determine if any of the individual historic short-term release events represented an acute public health hazard to the adjacent communities. Specific acute releases are based on the four largest accidental releases specifically identified in the Chemrisk Task 6 report [2] or in the accident report records available to ATSDR (Material Release Reports 81-187, 252-344; various dates and authors). Although the available data are probably not complete, these records do include the years of highest production and annual emissions and are assumed to be representative of the most significant individual release events.

Chronic (or annual) doses to airborne radionuclides were estimated using the Clean Air Act Assessment Package—1988 (CAP88PC), a system developed by the EPA and the DOE [3]. CAP88PC uses a modified Gaussian plume equation
to estimate the average dispersion of radionuclides released from up to six sources. The CAP88PC model does not account for complex topography. The Radiological Assessment System for Consequence Analysis (RASCAL) 3.0 is the latest version of a computer model designed for the assessment of radiological accidents developed by the Nuclear Regulatory Commission (NRC). The relevant components of the RASCAL model for this analysis use Gaussian models to describe the atmospheric dispersion of radioactive effluents from nuclear facilities. Theoretical description of the model components is presented in the RASCAL user’s guide [4].

Based on a review of the available documents and emission reports, there does not appear to be a record of long-term fluoride emissions. There are however, 16 years of fluoride air monitoring records at a number of stations around the K-25 facility. These records measured actual airborne fluoride concentrations over seven day intervals and the reported results include annual averages and maximum seven-day concentrations for each station. These measured fluoride air concentrations will be used to assess historic fluoride air concentrations.

2 Results

2.1 Measured vs. Predicted Concentrations at Monitoring Locations

Figure 2 shows the predicted vs. measured gross alpha concentrations at the HP 35 and HP 33 locations (respectively) for the 1966 to 1983 time period. Each chart has three trend lines—the measured annual average gross alpha concentration at that location and the concentrations predicted using CAP88PC with the DOE and Task 6 release estimates (plus background). For the CAP88PC predictions, the HP 35 location is 2,000 m northeast of the center of the site (point of release) and the HP 33 location is 3,000 m south-southeast of the center of the site. Background concentrations, taken as the annual average of the remote stations (12 to 70 miles from ORR), are added to the CAP88PC concentrations based on K-25/S-50 emissions. Predicted gross alpha concentrations are the sum of the U-234, U-235, U-238, and Np-237 concentrations and all CAP88PC model runs use the 1999 meteorological data year.

This figure shows, even with the above simplifying assumptions, very good agreement between the historic measured gross alpha concentrations and those predicted using the CAP88 air dispersion model with K-25/S-50 air release estimates. The overall trends shown in Figure 2 shows that CAP88PC and the estimated emission rates adequately predict the environmental concentrations of radionuclides released from the K-25 facility. It should be noted that both predicted and measured gross alpha for the HP-35 location are about 1.5 to 2 times higher than the HP-33 location. This is expected, as HP-35 is in the dominant downwind direction relevant to releases from the K-25 sources. It is also significant to note that, at the higher concentration HP 35 location, the concentrations predicted from DOE-estimated emissions provide a somewhat
better agreement with measured gross alpha concentrations than the Task 6 emissions.

Measurements of airborne fluoride were collected at six locations around the perimeter of K-25 from 1971 to 1985 (Figure1). The sampling duration for these measurements was either 24 hour or six or seven-day collection periods and annual averages are reported for all years. All of the monitoring results are reported in the annual environmental monitoring reports for the respective years.

Figure 2: Measured annual average gross alpha concentration (pCi/m3) from monitoring location HP 35 and HP 33 vs. annual average gross alpha concentrations predicted using CAP88PC.

Figure 3 shows all of the K-25 perimeter fluoride annual average air concentrations (in ppb) relative to the uranium air emissions (in kg). The relationship between the uranium emissions and fluoride air concentrations for the 1971-1985 time period is used to predict the annual average fluoride air concentrations for those years before and after fluoride was measured. The
predicted fluoride concentrations were estimated using the FORECAST function in an EXCEL spreadsheet. The relationship is a linear regression between known fluoride air concentrations and known uranium emissions in kg (DOE estimate). The correlation coefficients for those relationships vary from about 0.5 to 0.7, which indicates moderately good agreement between annual uranium emissions and annual airborne fluoride concentrations.

Figure 3 shows the highest predicted yearly fluoride air concentration is about 6 ppb. Note that the maximum uranium emission in kg is for 1945 at S-50 and that the correlations are based on emissions and measurements from the K-25 facility. Nonetheless, predicted historic long term airborne fluoride concentrations at the K-25 perimeter locations are less than 6 ppb for the maximum K-25 release year (1958; 2711 kg U). Also, station F-2 had the highest annual average concentrations due to its downwind location. It is also worth noting that the fluoride monitoring location with the best correlation coefficient (0.74) was station F-6, which was approximately 5 miles upwind (NW) of K-25 and considered by DOE to be a background location [5].

![Graph showing Measured and Predicted Fluorides in Air at Selected Stations: Annual Averages](image)

Figure 3: Measured and predicted annual average fluoride air concentrations (ppb).

For both the short term (24 hour, 7-day, and 30-day) and annual average fluoride air concentrations, measured and predicted values at the F-1 to F-5 stations will be higher than areas of potential off-site exposure. The F-6 station is located about 8 km (~5 miles) northwest of the K-25 facility. The maximum measured short term fluoride concentration (24 hour) at the F-6 station was 10.9 ppb in 1976. The following evaluation of accidental releases using the
RASCAL3 air dispersion model further evaluates predicted HF concentrations at off-site areas.

2.2 Estimated HF and Uranium Doses and Concentrations

Estimated radiological doses and hydrogen fluoride concentrations from the 09/01/58 accidental UF₆ release are listed in Table 1. These values calculated using the RASCAL3 model are lung-specific equivalent doses in mrem. The 09/01/58 accidental release represents the largest single release event (1184 kg UF₆) that is included in the available documents. The doses/concentrations listed in Table 1 were estimated using worst-case meteorological and exposure assumptions. The UF₆ release scenario assumes emission from the 24 m high roof vents and that 100% of the UF₆ released was emitted to the atmosphere. These results assume worst-case conditions that result maximum offsite exposure (light winds, a stable atmosphere, and no precipitation). Also, these results ignore the effect of topography on plume dispersion such that doses/concentrations at SG are likely to be much lower due to the effect of the 100 m Black Oak Ridge (Figure 1).

Table 1.

<table>
<thead>
<tr>
<th>Distance</th>
<th>U Dose</th>
<th>U Inhaled</th>
<th>U Conc.</th>
<th>HF Conc.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5 km</td>
<td>942 mrem</td>
<td>9.7 mg</td>
<td>1,340 µg/m³</td>
<td>1,310 ppb</td>
</tr>
<tr>
<td>1.0 km</td>
<td>569 mrem</td>
<td>5.9 mg</td>
<td>833 µg/m³</td>
<td>2,680 ppb</td>
</tr>
<tr>
<td>1.5 km</td>
<td>67 mrem</td>
<td>0.7 mg</td>
<td>97 µg/m³</td>
<td>461 ppb</td>
</tr>
<tr>
<td>2.0 km</td>
<td>48 mrem</td>
<td>0.5 mg</td>
<td>69 µg/m³</td>
<td>267 ppb</td>
</tr>
<tr>
<td>2.57 km (SG)</td>
<td>34 mrem</td>
<td>0.4 mg</td>
<td>51 µg/m³</td>
<td>156 ppb</td>
</tr>
<tr>
<td>3.0 km</td>
<td>27 mrem</td>
<td>0.3 mg</td>
<td>38 µg/m³</td>
<td>108 ppb</td>
</tr>
<tr>
<td>4.32 km (U/L)</td>
<td>12 mrem</td>
<td>0.1 mg</td>
<td>17 µg/m³</td>
<td>27 ppb</td>
</tr>
<tr>
<td>5.0 km</td>
<td>8 mrem</td>
<td>0.1 mg</td>
<td>11 µg/m³</td>
<td>14 ppb</td>
</tr>
</tbody>
</table>

U Dose mrem—inhaled lung dose equivalent (acute exposure—1 hour)
U Inhaled mg—assumes inhalation rate of 1.2 m³/hr
U Conc. µg/m³—inhaled uranium dose in milligrams/inhalation volume m³
HF ppb—maximum one hour HF concentration in parts per billion

The estimated annual doses to people residing in the in the vicinity of the K-25/S-50 facility are presented in Table 2. These values are estimated using the previously described CAP88PC air dispersion/dose assessment model. Specific annual doses are calculated for the 1945 releases from S-50 and the 1963 releases from K-25. Doses from the S-50 release use 2002 data from the L-1209 meteorological tower. Doses from the K-25 release use 1999 data from the K-1208 meteorological tower (Figure 1). The whole body effective dose
equivalents, presented as “Individual Effective Dose Rate” in mrem/year, include all pathways and all radionuclides [3].

The doses in Table 2 are specifically calculated on the basis of the distance and direction of the exposure areas relative to the K-25 or S-50 sources. These locations are relative to the origin of the plumes (as shown in Figure 1). Note that the doses from the K-25 release, although based on higher annual emissions, are much lower than doses from the S-50 release. This is because the exposure areas for the K-25 release are approximately cross-wind of the source area while the HV exposure area is much closer to the S-50 source than other areas and the ULV area is downwind of a relatively strong SSW component. It should also be noted that exposures to the SG area are over-estimated due to the presence of Black Oak Ridge between the K-25 emission source and the SG exposure area. Black Oak Ridge is a steep-sided ridge with an elevation of approximately 100m (330ft) above the adjacent valleys. Dispersion from the CAP88PC model does not accommodate this type of complex topography. Consequently these doses are a health-protective over-estimate of likely doses to the SG community.

Table 2.

<table>
<thead>
<tr>
<th>Source</th>
<th>Exposure Area and Release Year</th>
<th>Individual Effective Dose Rate (mrem/yr)</th>
<th>Total U Annual Air Conc.* µg/m³</th>
<th>Fluoride Annual Air Conc. ppb**</th>
</tr>
</thead>
<tbody>
<tr>
<td>K-25</td>
<td>SG-1963</td>
<td>3</td>
<td>0.0011</td>
<td>&lt;5</td>
</tr>
<tr>
<td></td>
<td>ULV-1963</td>
<td>1</td>
<td>0.0003</td>
<td>&lt;5</td>
</tr>
<tr>
<td>S-50</td>
<td>HV-1945</td>
<td>14</td>
<td>0.02</td>
<td>&lt;5</td>
</tr>
<tr>
<td></td>
<td>ULV-1945</td>
<td>30</td>
<td>0.04</td>
<td>&lt;5</td>
</tr>
</tbody>
</table>

SG—Sugar Grove (2,570m NNW of K-25)
ULV—Union/Lawnville (4323m SSW of K-25; 2335m SSW of S-50)
HV—Happy Valley (1,447m ESE of S-50)

*CAP88PC model output is pCi/m³ for each uranium isotope at each location; those results are divided by specific activity for each isotope and summed (U-234, U-235, and U-238) for total U air concentration.

3 Conclusions

Short-term (1-hour average) off-site uranium air concentrations were modelled for the largest documented accidental release (September 1958). If people were exposed to the estimated short term uranium air concentrations, they would not have experienced adverse health effects due to the chemical effects of uranium exposure. Long-term exposure to airborne uranium also occurred during the years 1944 to 1985, as a result of elevated operational emissions. The highest
annual uranium release (as UF₆) occurred in 1963. The maximum estimated annual uranium air concentrations for this year (in an area of potential off-site exposure; ULV) is 0.04 µg/m³. Radiological doses from K-25/S-50 airborne releases for the largest documented accidental release and for the largest estimated annual release were estimated for the communities closest to the facilities. Historic exposure to airborne releases of ionizing radiation from the K-25/S-50 facility is not expected to cause any adverse health effects.

Historically, chronic (long-term) exposures to fluoride and hydrogen fluoride (HF) happened to people living around the K-25/S-50 facility as a result of releases during normal process operations; acute (short-term) HF exposures happened as a result of accidents or controlled releases. The most serious accident (September 1, 1958) may have created potential exposures in the Sugar Grove or Union/Lawnville communities. If a sensitive person was exposed to HF at the maximum estimated level for that accident, we expect, that person could have experienced minor adverse health effects; however, due to uncertainties (e.g., quantities released, modelling, locations of individuals at time of accidents), it cannot be determined if that accident posed a public health hazard to an individual. As with the long-term fluoride modelling, estimates from the accidental release are over-estimates of likely concentrations. Other accidental releases involved smaller quantities and probably did not affect the off-site communities.

Acknowledgements

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References


