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Application of the HGSYSTEM/UF_6 Model to Simulate Atmospheric Dispersion of UF_6 Releases from Uranium Enrichment Plants[1]

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Application of the HGSYSTEM/UF_6 Model to Simulate Atmospheric Dispersion of UF_6 Releases from Uranium Enrichment Plants[1]

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ABSTRACT

Uranium hexafluoride (UF_6) is a dense, reactive gas used in Gaseous Diffusion Plants (GDPs) to make uranium enriched in the ^{235}U isotope. Large quantities of UF_6 exist at the GDPs in the form of in-process gas and as a solid in storage cylinders; smaller amounts exist as hot liquid during transfer operations. If liquid UF_6 is released to the environment, it immediately flashes to a solid and a dense gas that reacts rapidly with water vapor in the air to form solid particles of uranyl fluoride (UO_2F_2) and hydrogen fluoride (HF) gas. Preliminary analyses were done on various accidental release scenarios to determine which scenarios must be considered in the safety analyses for the GDPs. These scenarios included gas releases due to failure of process equipment and liquid/gas releases resulting from a breach of transfer piping from a cylinder. A major goal of the calculations was to estimate the response time for mitigating actions in order to limit potential off-site consequences of these postulated releases.

The HGSYSTEM/UF_6 code was used to assess the consequences of these release scenarios. Inputs were developed from release calculations which included two-phase, choked flow followed by expansion to atmospheric pressure. Adjustments were made to account for variable release rates and multiple release points. Superpositioning of outputs and adjustments for exposure time were required to evaluate consequences based on health effects due to exposures to uranium and HF at a specific location.

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INTRODUCTION AND BACKGROUND

Uranium hexafluoride (UF_6) is a dense, reactive gas used in Gaseous Diffusion Plants (GDPs) where the uranium is enriched in the ^{235}U isotope. UF_6 is a solid at ambient temperature and has a triple point at 65°C and 152 kPa. It sublimes at 56°C and 1.0 atm (101 kPa) and forms a very dense gas (molecular weight is 352). Figure 1 shows the regions of gas, liquid, and solid phases as a function of temperature and pressure under equilibrium conditions [based on correlations given by Williams (1985)]. The gas reacts vigorously with water vapor (H_2O) to form solid particles of uranyl fluoride (UO_2F_2) and hydrogen fluoride (HF) gas according to the exothermic reaction

$$UF_6 + 2\,H_2O \rightarrow UO_2F_2 + 4\,HF\,.$$

Large quantities of UF_6 exist at the GDPs as in-process gas and as a solid in storage cylinders. Smaller amounts exist as feed gas and as liquid during analysis prior to feeding, withdrawal from the process, transfers between storage cylinders of different sizes, and cooling prior to storage. The solid UF_6 in the storage cylinders is in a very safe condition with respect to potential atmospheric releases. The cylinder pressure is generally below ambient pressure, and in the absence of a heat source, the solid sublimes and reacts slowly at ambient temperatures. Therefore, if a breach of the cylinder

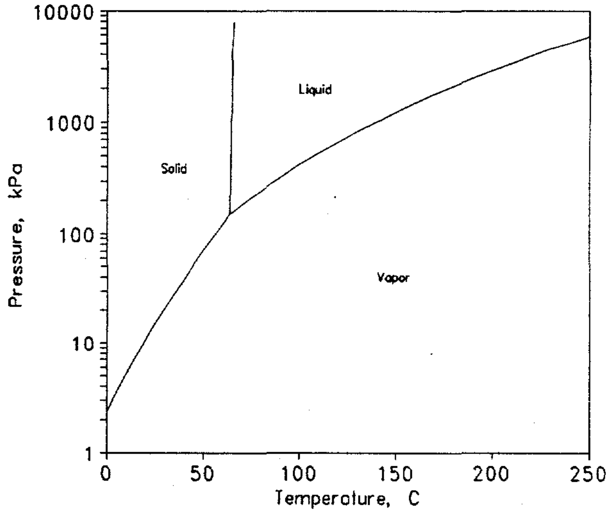

Figure 1. Phase Diagram for UF_6

containment were to occur, there would be ample time for mitigating actions. A cylinder containing liquid UF_6, however, is above ambient pressure, and a breach of containment could rapidly release a large quantity. If liquid UF_6 is released to the environment, it immediately flashes to a solid and a dense gas which reacts with ambient water vapor. Portions of the enrichment process operate above ambient pressure and a breach of containment could release a large quantity of gas.

Potential consequences from accidental releases of UF_6 have been a concern of the GDP operators as well as of the U.S. Department of Energy (DOE) and its predecessors, the Atomic Energy Commission and the Energy Research and Development Administration. Prior to 1977, simple Gaussian models were used to simulate the atmospheric dispersion of UO_2F_2 and HF resulting from UF_6 releases. From 1978 to 1985, DOE and its predecessors sponsored the development and improvement of a more detailed atmospheric dispersion model (Bloom, 1980; Bloom et al., 1989). This model (PLM89A) incorporated the following mechanisms:

1. the chemical reaction of UF_6 gas with water vapor along with the corresponding energy change;
2. the phase changes of H_2O, UF_6, and HF along with their corresponding energy changes;
3. positive, neutral, and negative buoyancy;
4. gravity spreading of a ground-hovering, negatively buoyant plume;
5. lift-off if a ground-hovering plume becomes positively buoyant; and
6. heat and mass transfer to ground.

The model was used extensively until about 1985 to analyze accident scenarios that were developed for earlier safety analyses reports for the GDPs. After 1985 no funding was available for further development or improvement, but some minor modifications were done to arrive at PLM89A.

About 1991 additional accident analyses for the GDPs were deemed necessary in response to new regulatory requirements. Since no significant work had been done on PLM89A for several years, Sykes and Lewellen (1992) were asked to examine existing atmospheric dispersion models that might serve as a basis for an improved UF_6 model. PLM89A was included in this survey, but Sykes and Lewellen determined it was not up-to-date in parameterizations of transport and diffusion, entrainment, and dense gas slumping. They recommended that either PLM89A be modified to include these updates or another model, HGSYSTEM (McFarlane et al., 1990), be modified by adding UF_6 chemistry. The choice was made to modify HGSYSTEM.

HGSYSTEM/UF_6

HGSYSTEM consisted of several submodels that simulated the release and atmospheric dispersion of HF, ideal dense gases, and aerosols. It produced results that compared well with experimental releases of HF and nonreactive, dense gases (Hanna et al., 1993) and was familiar to most of the atmospheric dispersion community. The primary change needed was to add the UF_6 chemical reaction with water vapor, including the component and energy changes resulting from the reaction. Other changes included the ability of a ground-hovering plume to lift off if it becomes positively buoyant, removal of gases and particles by dry and wet deposition, accounting for variations in concentration with averaging time and concentration fluctuations, effects of large buildings on constricting plume spread (canyon effect), parameterization of boundary layer meteorological variables using state-of-the-art algorithms, and estimating concentrations on building roofs and sides due to emissions from short vents.

Hanna et al. (1994) developed the HGSYSTEM/UF_6 model by making the above changes. HGSYSTEM/UF_6 consists of the following modules which can be chosen by the user:

- AEROPLUME/UF_6 is intended for steady-state, elevated jet releases and applies from the point of release until the resulting plume either strongly interacts with the ground or becomes passive (i.e., the density, velocity, and composition of the plumes approach that of ambient air). AEROPLUME/UF_6 has a submodule that calculates the temperature and size of the two-phase, expanded (to atmospheric pressure) jet arising from a pressurized liquid release.
- HEGADAS/UF_6 is intended for continuous, ground-based, dense gas plumes and applies to either area sources on the ground or the continuation of jet releases from the point where the plume strongly interacts with the ground. There are two versions: HEGADAS-S/UF_6 applies to steady-state releases and the continuation of AEROPLUME/UF_6 jets; HEGADAS-T/UF_6 applies to transient releases, including the continuation of a HEGABOX/UF_6 puff. HEGADAS-T/UF_6 is actually a series of HEGADAS-S/UF_6 calculations for different time periods.
- HEGABOX/UF_6 is intended for instantaneous, ground-based dense gas puffs. It applies until the gravity-slumping phase ends and then makes a transition to HEGADAS-T/UF_6.
- PGPLUME is intended for steady-state, passive plumes and applies to the continuation of jet releases (using AEROPLUME/UF_6) from the point where the plume becomes passive. PGPLUME is also used to approximate vertical and horizontal concentration profiles for AEROPLUME/UF_6.

In addition, a simple postprocessor was written to approximate the transient concentration response at a specified location due to a constant release rate of finite duration. This approximation is based on those used in PLM89A (Bloom et al., 1989, pp 3-8 to 3-16) and HGSYSTEM (McFarlane et al., 1990, pp 6.18 to 6.20). This postprocessor is used with both PGPLUME and HEGADAS-S. Use of this postprocessor avoids difficulties which are encountered when HEGADAS-T is used for short-term releases.

CONSEQUENCE CRITERIA

The two operating GDPs in the United States are located near Portsmouth, Ohio, and Paducah, Kentucky. A third GDP at Oak Ridge, Tennessee, ceased operating in 1985. The Paducah plant is located on relatively flat terrain; the Portsmouth plant is in a small basin surrounded by low hills. Both plants are surrounded by security fences, and access to the sites is restricted. Plant personnel and visitors are instructed about emergency protection procedures in case of an accident; therefore, the major concern in accident analysis is potential consequences to off-site personnel. The nearest location of off-site personnel to any accidental release of UF_6 gas would be at the site boundary. The shortest distance between such an accident location and the site boundary is approximately 1000 m. Therefore, the consequences were evaluated at 1000 m downwind from the release point.

Since UF_6 reacts rapidly with H_2O in the air, the health effects arising from accidental UF_6 releases are primarily due to inhalation of uranium and HF (McGuire, 1991). Most of the uranium isotopes involved in the GDP operations are only mildly radioactive, and the principal hazard from uranium is due to heavy metal poisoning rather than to radiation. UO_2F_2 is relatively soluble in the lungs where it is absorbed into the blood and migrates to the kidneys. Just and Emler (1984) summarized the data on health effects to the kidneys due to the inhalation of soluble uranium. They arrived at concentration-time products ($C \times t$, where C is concentration and t is time) that delineated the regions of different health effects. If concentration varies with time, $C \times t$ may be replaced by the integral of C dt over the duration of the exposure. For exposures less than 30 min, no health effects occur at $C \times t$ values less than 650 mg min/m^3; possible mild (reversible) health effects occur between 650 mg min/m^3 and 1,250 mg min/m^3; renal injury occurs at 1,250 mg min/m^3; and the value for 50 percent lethality is 35,000 mg min/m^3. Fisher et al. (1994) concluded that limiting an acute intake to less than 30 mg of soluble uranium by inhalation appears to provide sufficient protection for the average human. The total amount inhaled is equal to the product of the inhalation rate and $C \times t$. An inhalation rate of 0.0075 m^3/min corresponds to that of an average person at rest (ICRP, 1978, p 344ff). The 30-mg value, evaluated with the 0.0075 m^3/min inhalation rate, is the target value applied at 1000 m downwind to assess the relative hazard due to uranium.

Health effects due to inhalation of HF can be related to Emergency Response Planning Guides (ERPG) (AIHA, 1994). Concentrations below ERPG-2 for exposures up to 1 hour are considered to cause only mild (reversible) consequences. The ERPG-2 value for HF is 20 ppm or about 16.6 mg/m^3. For lethal exposures of HF, ten Berge et al. (1986) suggested an expression of the form

$$C^2\, t = \text{constant}\ .$$

McGuire (1991) assumed this same type of expression could be applied to non-lethal exposures, and we further generalized the expression to the following concentration-time integral:

$$\int C^2\, dt = \text{constant}\ .$$

For a relatively constant concentration, the value of the constant corresponding to ERPG-2 is about 16,500 mg^2 min/m^6. However, the concentration at any time should not exceed 57 mg/m^3, which corresponds to applying the above constant and integral to a constant concentration over a 5-min exposure time. These criteria are applied at 1000 m downwind as target values for assessing the relative hazard due to HF.

ACCIDENTAL RELEASE SCENARIOS

Four accidental release scenarios are presented here and represent some of the highest release rates that could occur at the GDPs. These scenarios also illustrate the calculations and adjustments needed to apply HGSYSTEM/UF_6 to estimate the consequences due to the releases.

The gaseous diffusion process consists of thousands of separation stages grouped into cells. The stream of UF_6 depleted in ^{235}U is called the "B" stream and the piping carrying this stream from stage to stage can be designated as a B-line. There are B-lines between cells within a building and between process buildings. At the highest operating levels, a large number of the B-lines are at pressures above ambient. The scenarios for UF_6 releases from the process involve potential breaches of these B-lines at high pressures.

Breach of a B-Line Between Buildings

A B-line carrying hot UF_6 gas between two process buildings has a 0.6-m diameter and runs 6.9 m above ground level. The gas is above atmospheric pressure and, if the pipe is severed, would emerge from the cut end at a rate of 100 kg/s. This release is assumed to be directly to the atmosphere with no building effects. Gas flow is not choked so that the pipe diameter and elevation, along with gas temperature and flow rate, are direct inputs to AEROPLUME/UF_6.

AEROPLUME/UF_6 does not calculate vertical or horizontal concentration distributions; however, if a transition is made to PGPLUME, the latter model calculates these distributions. To obtain a ground-level concentration from AEROPLUME/UF_6, a transition is forced at that location. The calculations are then stopped because the PGPLUME calculations downwind of the forced transition may not be valid. The transient response postprocessor is applied to the steady-state PGPLUME result to produce the estimate of concentration versus time at 1000 m downwind. The duration of the release corresponds to a response time for mitigating actions and is varied to determine the time that would limit the off-site consequences to acceptable values.

Release from a Punctured B-Line Within a Building

This scenario postulates a release of 8000 kg of UF_6 gas over a 5-min period due to the puncture of a B-line between cells within a building. The UF_6 mixes with air in the building and reacts with moisture in the air. The building ventilation system is assumed to continue to operate during the accident, so the release to the atmosphere is through a number of ventilation ducts on the roof and sides of the building. Because of in-building mixing, the sum of release rates from the ducts is lower and continues for a longer duration than the release from the B-line. A set of computer programs developed by Williams (1985) is used to estimate the mixing and reaction within the building. These programs also estimate the amount of the resulting mixture that is retained in the building (primarily due to deposition of UO_2F_2 particles) and the temperatures and rates of gases released through the ducts. The large volume of air in the buildings contains enough moisture to react with all the UF_6.

The gas released from the building consists primarily of air and is equivalent to a passive gas release. We use both a standard passive gas model (ISC2) (EPA, 1992) and PGPLUME to simulate the atmospheric dispersion. Since our principal location of concern is 1000 m downwind, each duct is not considered as a separate source but combined into one large source. The effect of the building wake for PGPLUME is estimated using expressions given by R.P. Hosker, Jr. in Randerson (1984, chap. 7, pp 251 to 302). ISC2 includes the effect of the building wake. The release rate varies continuously over a duration of 4000 s, and this rate is approximated by three constant rate periods: an initially high rate (4.8 kg/s uranium and 1.7 kg/s HF) for the first 300 s, a slightly reduced rate (3.6 kg/s uranium and 1.6 kg/s HF) for the next 600 s, and a much reduced rate (0.12 kg/s uranium and 0.11 kg/s HF) for the final 3100 s. The transient concentration postprocessor is applied to each of the three steady-state ISC2 or PGPLUME runs, and the results are superposed to produce the overall estimate of concentration versus time at 1000 m downwind. The ISC2 and PGPLUME results are comparable for this scenario.

Breach of Cylinder Transfer Line, Valve in 6:00 Position

During analysis prior to feeding and transfers between storage cylinders, a storage cylinder containing liquid UF_6 may be positioned such that the valve on the cylinder is towards the bottom of the cylinder and points downward (6:00 position). If the transfer line or the valve is breached, UF_6 liquid will be released under pressure and will flash to a solid and a gas.

Flow from the severed line is calculated with the CYLIND model, which accounts for the two-phase, choked flow conditions (Williams, 1985). For a cylinder containing liquid at 127° C and 100 psig (791 kPa), the calculated flow is 4.4 kg/s through a 1.9-cm diameter opening. This rate remains constant until the cylinder is almost empty. The largest storage cylinders hold about 12,700 kg of UF_6, so the duration of the release could be as long as 2900 s.

The release to the atmosphere consists of a jet containing gas and solid particles, but the flow is pointed downward. AEROPLUME/UF_6 does not work well with jets that point steeply downward; also, most of the momentum of the jet is dissipated when the jet hits the ground. Consequently, this release tends to form a ground-based, dense gas plume. HEGADAS-S/UF_6 is considered to be the appropriate model to use for this scenario. The footprint on the ground is arbitrarily set to 1 m × 0.8 m. Previous tests with different footprints indicated the results at 1000 m downwind were not sensitive to the choice of footprint. Results are mostly dependent on the flow rate. The transient concentration postprocessor is applied to the HEGADAS-S/UF_6 results to obtain concentration as a function of time at 1000 m downwind.

Breach of Cylinder Transfer Line, Valve in 12:00 Position

During withdrawal of product and tails from the process, transfers between storage cylinders of different sizes, and cooling prior to storage, UF_6 may exist as a liquid in a storage cylinder with the valve positioned near the top of the cylinder and pointing upward (12:00 position). If the transfer line or the valve is breached, UF_6 liquid will be released under pressure until the liquid level in the cylinder falls below the position of the valve. After that time, only vapor is released. The worst consequences will occur if the severed line is bent such that the release is directed horizontally, and this is assumed for this scenario.

This scenario involves a very complicated flow situation, but the CYLIND code (Williams, 1985) is capable of calculating this flow and is also used to calculate the release rate for this scenario. The liquid release duration is relatively short (about 160 s), the rate is constant (4.4 kg/s), and it is modeled as two-phase, choked flow as with the 6:00 case. The gas release portion is one-phase, choked flow and includes a heat balance to determine the evaporation rate of the liquid and the subsequent temperature and pressure of the gas in the cylinder. Because the temperature and pressure of the gas in the cylinder vary with time, the gas release rate also varies with time.

Since both the liquid and gas releases are jets, the AEROPLUME/UF_6 model is used for atmospheric dispersion calculations; however, the pressure at the opening is above ambient pressure because the flow is choked. The liquid release submodule in AEROPLUME/UF_6 is used to calculate the size and temperature of the expanded jet for the liquid release portion, but the gas release portion requires a separate calculation for these inputs to AEROPLUME/UF_6. This calculation assumes the pressure-volume-temperature behavior of UF_6 gas is ideal, which is a good approximation (Williams, 1985). Gas leaving the opening is assumed to expand (and accelerate) isentropically until it forms a shock wave with the pressure on the downstream side equal to ambient pressure. Inputs to AEROPLUME/UF_6 can then be calculated from the Rankine-Hugoniot relations for the shock wave and the isentropic flow expressions for an ideal gas (Roberson and Crowe, 1975, Chap. 12).

Following the initial liquid release period, the varying release rate of gas is approximated by three constant rate periods: 1.8 kg/s for 160 s, 1.5 kg/s for 880 s, and 1.0 kg/s for the final 1200 s. As with the breach of a B-line between buildings, PGPLUME is used to estimate the ground-level concentration at 1000 m downwind. The transient concentration postprocessor is then applied to each of the four steady-state runs, and the results are superposed to produce the overall estimate of concentration versus time at 1000 m downwind.

RESULTS OF CALCULATIONS

Results of calculations for the above scenarios are summarized in Table 1 in terms of release durations to limit consequences. Figure 2 shows the steady-state plume concentrations and plume height, resulting from a B-line break, as functions of distance. These are calculated by AEROPLUME/UF_6 and the concentrations are expressed as total uranium and total fluoride (in both HF and UO_2F_2). In spite of being hot (greater than 56° C), the plume initially falls towards the ground because of the high molecular weight of the UF_6. However, the plume soon changes composition (due to dilution and reaction) and heats up further (due to the reaction) so that it lifts off the ground and becomes airborne again between 500 and 600 m downwind. The airborne plume entrains air at a much faster rate than a ground-hovering plume, and this is reflected in a rapid decrease in the concentrations.

Table 1. Release Durations To Limit Consequences

Scenario	*Release Duration To Limit Consequences*	
	Based on Uranium	*Based on HF*
B-Line Between Buildings	1700 s	Not Estimated
B-line Within A Building	More than 1 hour	More than 1 hour
Cylinder Valve, 6:00	195 s	101 s
Cylinder Valve, 12:00	144 s	78 s

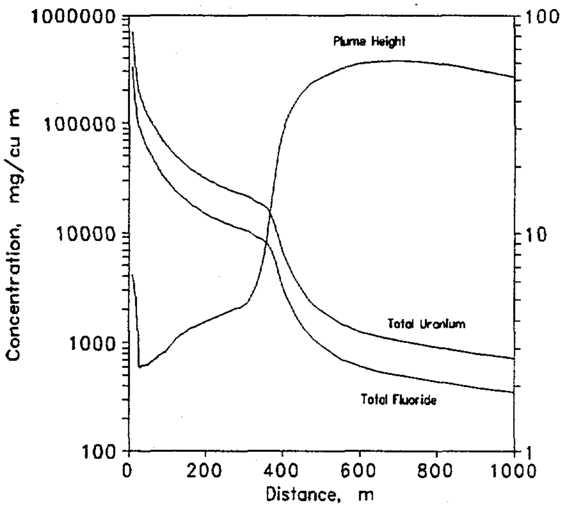

Figure 2 AEROPLUME/UF_6 Results for B-Line Between Buildings

Figure 3 shows ground-level concentrations of total uranium and total fluoride as a function of the time since the start of the release. These values are obtained from AEROPLUME/UF_6 results at 1000 m; the PGPLUME approximation to estimate the steady-state, ground-level concentration; and the transient concentration postprocessor to obtain the time variation. Since concentration is relatively unchanged over a long time period, obtaining the C × t product for estimating uranium uptake is simple. As Table 1 indicates, uptake will not exceed 30 mg if this release can be terminated within about 1700 s. The comparable value for HF was not calculated for this scenario.

Figure 4 shows the uranium concentrations at 1000 m downwind that are due to the puncture of a B-line within a building. The duration of this release within the building is only 5 min, but the 1000-m concentration extends beyond an hour due to the hold-up in the building. Values in Figure 4 are from PGPLUME runs and the transient postprocessor. A separate curve is shown for each of the three release durations, and the overall curve is the summation of the three. The C × t product is estimated by numerically integrating the overall curve. Comparable curves for HF are similar, and the $C^2 \times t$ product is also estimated by numerically integrating the overall HF curve. As indicated in Table 1, mitigation time to stop the release from the building is greater than 1 hour.

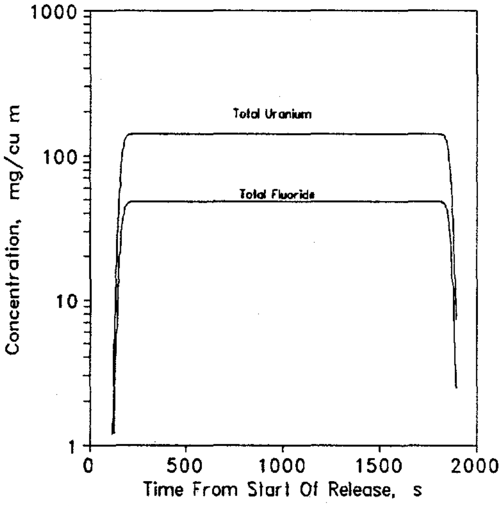

Figure 3. Ground-level Concentrations for a B-Line Between Buildings

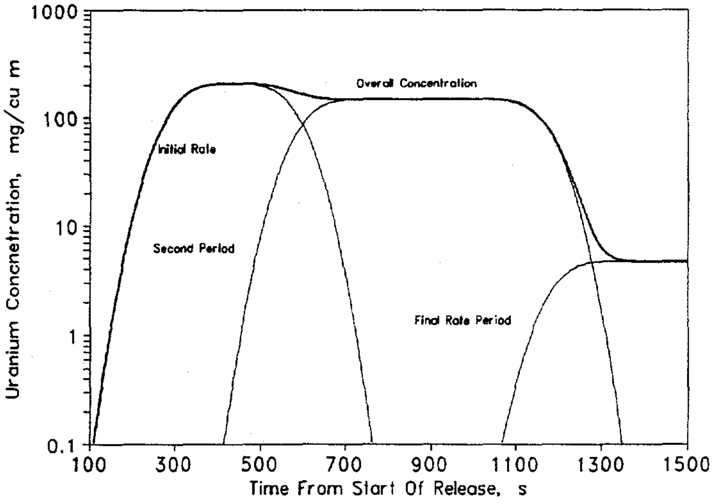

Figure 4. Uranium Concentrations from B-Line Within a Building

The results for a transfer line break with the valve in the 6:00 position are shown in Figures 5 and 6. The results in Figure 5 are calculated with HEGADAS-S/UF_6 and show a steady dilution of the uranium. Since HF arises from the chemical reaction, the HF concentration increases close to the release location, reaches a maximum where the reaction is about complete, and then dilutes in a manner similar to the uranium. The uranium concentration at 1000 m downwind (Figure 6) shows a typical bell-shaped curve as a function of time. As before, the C × t and $C^2 \times t$ products are obtained by numerical integration and used to arrive at the mitigation times in Table 1.

The results for a transfer line break with the valve in the 12:00 position as shown in Figure 7, are similar in appearance to those for the B-line within a building as shown in Figure 4. However, the results in Figure 7 are obtained from four AEROPLUME/UF_6 runs, corresponding to the liquid release rate followed by three gas rates. PGPLUME is only used to obtain ground-level concentrations at 1000 m. Superpositioning and numerical integration are used to obtain C × t and $C^2 \times t$ products as the B-line within a building. As shown in Table 1, the mitigation time would have to be about 78 s to prevent significant health effects due to HF.

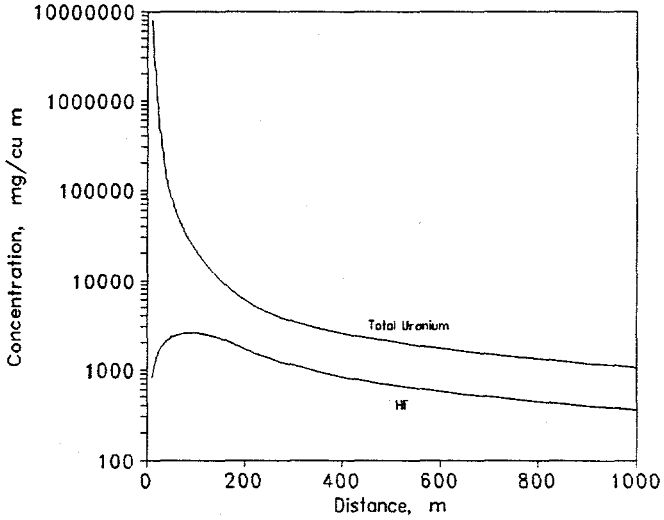

Figure 5. HEGADAS-S/UF_6 Results for Valve in 6:00 Position

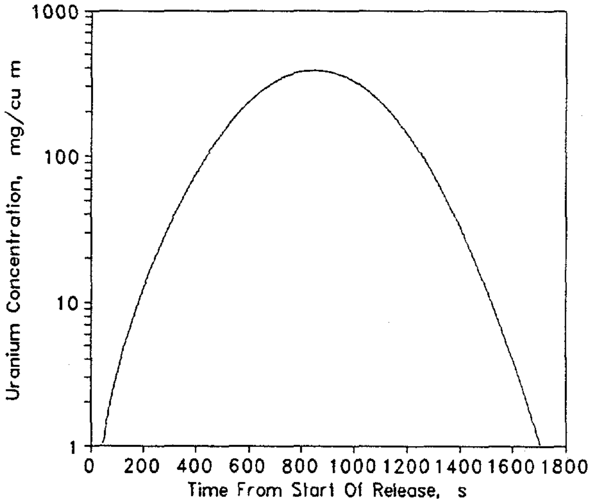

Figure 6. Uranium Concentration at 1000 m for Valve in 6:00 Position

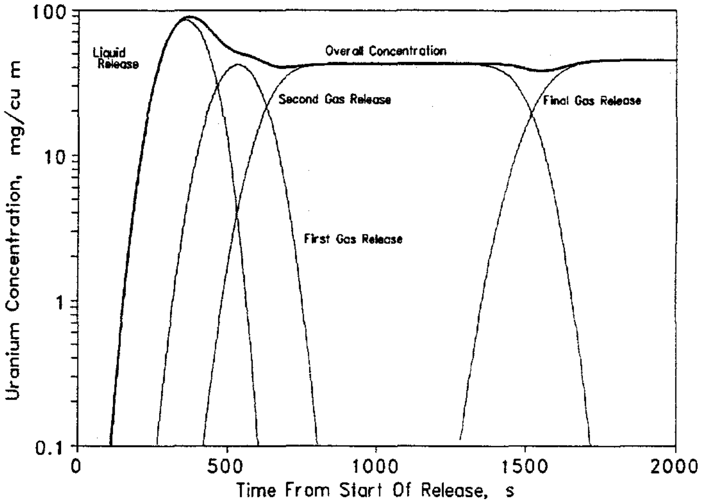

Figure 7. Uranium Concentrations at 1000 m, Valve in 12:00 Position

It is interesting to note from Table 1 that the health effects due to HF, rather than those due to uranium, control the mitigation response time. If the HF arises only from the chemical reaction of UF_6 with H_2O, the ratio of the uranium to HF concentrations (expressed as mg/m^3) is about 3.0 after all the UF_6 has reacted. However, the target value of the HF concentration increases proportionately to $t^{-1/2}$ while uranium increases proportionately to t^{-1}. It can be shown from these relationships that HF is more significant for exposures less than about 100 min.

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