AN AIR POLLUTION ASSESSMENT OF HYDROGEN FLUORIDE

AIR FORCE ENVIRONMENTAL TECHNICAL APPLICATIONS CENTER

MARCH 1976
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AN AIR POLLUTION ASSESSMENT
OF HYDROGEN FLUORIDE

By

Capt Richard W. Fisher

March 1976

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**Abstract:**

Only generalized conclusions about ground level concentrations (χ) of hydrogen fluoride (HF) are possible when the place and time of aerial release are unknown. HF is assumed to be a gas when released from altitudes of 3 km and 10 km and remains a gas during the entire diffusion process. Three line-source models are solved for χ/Q since the emission rate, Q, is unknown. H. Cramer's concentration model is the product of five terms, including an
edge effects term borrowed from Turner. Ground level dosages are also included. B. Turner's line-source model is a direct modification of Sutton's basic diffusion equations and uses vertical and horizontal standard wind deviations as calculated by Cramer. The third model is an empirical derivation from Air Force testing at the Cedar Hill Tower, Dallas, Texas. Results from these three models graphically depict the maximum ground concentrations of HF at 60 km downwind. Theoretically, when HF is released at 10 km altitude the maximum ground concentration occurs beyond 100 km, at which distance these equations become ineffectual. However, these estimated concentrations represent only the worst meteorological conditions. Therefore, only when the mixing depth is high will concentrated HF contamination occur.
The US Air Force Environmental Technical Applications Center (USAFETAC) prepared this report in answer to a request from the Argonne National Laboratory, Argonne, Illinois. The information is provided in support of the Air Force Weapons Laboratory (AFWL) Project 19008W15, "Environmental Implications of Airborne Hydrogen Fluoride (HF) Laser Operations."

Argonne National Laboratory requested USAFETAC support in the belief that USAFETAC maintains a working computer diffusion model for gases released from elevated line sources. USAFETAC does not have a working computer model; therefore, this report presents generalized conclusions based upon a thorough search of references that present information relevant to this problem.

In the event that this report is incorporated into another report by the requester or any other agency, request that USAFETAC be furnished a copy of the new report in all cases where such dissemination is not prohibited.

USAFETAC prepared this report for a specific purpose; therefore, any further application of this information should be undertaken with caution. Work on this report progressed under rigid constraints of resources including time, personnel, equipment, and data. Department of Defense agencies and their contractors should contact USAFETAC directly for aid in assessing the applicability of this material for their purposes. Other prospective users should contact professional environmental analysts in the National Oceanic and Atmospheric Administration (NOAA) or private industry for similar assessment service.
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Table 1. Estimated Ground Level Concentrations
AN AIR POLLUTION ASSESSMENT OF HYDROGEN FLUORIDE

Introduction

The purpose of this report is to estimate the diffusion characteristics and downwind concentrations of hydrogen fluoride (HF) after it is released from an airplane. The author conducted an exhaustive search of USAFETAC's in-house technical library to determine a method or methods adaptable to this problem.

Several authors have modified Sutton's basic diffusion equations for microscale analyses. Applications from three sources are included in this report. The most useful model, H. H. Cramer (3), offers a generalized concentration model for a finely divided particulate or gas using the Gaussian distribution functions. The second model, applied by D. B. Turner (9), rewrites Sutton's (1932) concentration equation into a simplified finite line source model including an edge effects term (EET). This term is also applied as the EET to Cramer's Generalized Model in this report. The third model is a reapplication of an equation by Smith and Hay (1961) using vertical turbulence intensity (7). The US Air Force empirically tested the diffusion characteristics of airborne substances near Cedar Hill, Texas and then appropriately modified the original equation. Under steady-state conditions, estimates can be made using these equations for distances of up to 100 kilometers. These line-source equations are briefly described in this report. However, the classical diffusion equations are not appropriate for unsteady-state diffusion estimates (5:17,19).
Assumptions

The state of the art of diffusion estimation and the limited amount of input data require that several assumptions be made. They are:

1. Diffusion in the alongwind or x-direction can be neglected when compared with a strong transport wind (5:13).

2. In the vertical direction HF assumes a statistical Gaussian distribution.

3. The emission is an instantaneous line source.

4. Homogeneous steady-state conditions exist (i.e., no space or time changes in wind or turbulence).

5. HF reflects perfectly at the surface (i.e., no ground absorption) and at the height of the mixing layer.

6. HF remains a gas.

7. No HF coalesces with water vapor, or washes or rains out.

8. HF has approximately the same molecular weight as air (i.e., no thermal buoyancy or settling velocity).

9. Atmospheric stability is neutral at all points downwind.

(11) The mean wind direction is normal to the airplane's flight path.

(12) No vertical wind shear at any point.

(13) The emission rate is constant.

Data Provided

We are given or can calculate several pieces of information.

Mean Wind Speed (m) = 20 m/s (45 mph)

Sampling will take place near the surface therefore

\[ z = 2 \text{ meters} \]

Four runs will be made:

<table>
<thead>
<tr>
<th>Run</th>
<th>Speed of Aircraft Mach (m/s)</th>
<th>Altitude of Release (H)</th>
<th>Length of Release (y)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1)</td>
<td>.5 (230)</td>
<td>3 km (10,000')</td>
<td>3,450 m</td>
</tr>
<tr>
<td>(2)</td>
<td>1.5 (760)</td>
<td>3 km</td>
<td>10,350 m</td>
</tr>
<tr>
<td>(3)</td>
<td>.5</td>
<td>10 km (33,000')</td>
<td>3,450 m</td>
</tr>
<tr>
<td>(4)</td>
<td>1.5</td>
<td>10 km</td>
<td>10,350 m</td>
</tr>
</tbody>
</table>

Summary

Hydrogen fluoride (HF) is a very stable substance in the atmosphere. Anhydrous HF or partially hydrolyzed HF is completely soluble in the presence of sufficient quantities of water vapor. Thus, in order to avoid coalescence and precipitation scavaging, no water vapor is
assumed to be present. At various temperature and pressure conditions present in the atmosphere HF may become a liquid, although it is conveniently assumed to always be a gas here. Gaseous HF is slightly lighter than air but for these computational purposes it will have the same mass. Thus, only mechanical mixing is considered in this report. Using the given and calculated data and the line source dispersion equations found in Appendix A, we can estimate ground-level and release-level concentration/emission rate values \((\chi/Q)\) in \(\text{sec/m}^3\) and dosages at pertinent distances downwind.

Table 1. Estimated Ground Level Concentrations (from Cramer (1))

<table>
<thead>
<tr>
<th>Concentrations (sec/m³)</th>
<th>Distances Downwind</th>
<th>60 km</th>
<th>100 km</th>
</tr>
</thead>
<tbody>
<tr>
<td>((\chi/Q)_3)</td>
<td></td>
<td>4.0x10⁻⁵ (max)</td>
<td>3.9x10⁻⁵</td>
</tr>
<tr>
<td>((\chi/Q)_{10})</td>
<td></td>
<td>8.0x10⁻⁷</td>
<td>4.0x10⁻⁶</td>
</tr>
<tr>
<td>Dosage₃</td>
<td></td>
<td>4.0x10⁻⁵ (max)</td>
<td>3.9x10⁻⁵</td>
</tr>
<tr>
<td>Dosage₁₀</td>
<td></td>
<td>2.8x10⁻⁸</td>
<td>4.0x10⁻⁶</td>
</tr>
</tbody>
</table>

*Subscripts denote release from 3 km and 10 km.

Maximum values for releases at 10 km altitude are given for 100 km downwind. The mathematical peak occurs beyond that distance but the model may not be accurate past 100 km. If the mixing is below the release height
of the HF gas, virtually no ground contamination will occur.

Consistent and accurate meteorological inputs are paramount for the successful application of any pollution concentration estimation method. Among the meteorological parameters, the mixing depths downwind from the release point are the most important factors in determining cloud expansion and ground contamination.

The assumptions made at the outset of this report make the expected concentrations extreme worst case figures. This conservatism means that under virtually all meteorological conditions, the expected concentrations will not exceed those given.

Discussion

Using Cramer's equation (3.21) we can calculate center line concentrations and graph the results for $\chi/Q$ values at the release height (Figure 1) or at the ground level when released from 3 km (Figure 2). When HF is released from 3 km, the maximum ground level concentration occurs 60 km downwind and the corresponding $\chi/Q$ is $3.22 \times 10^{-5}$ sec/m$^3$.

When HF gas is released from an altitude of 10 km, the maximum concentration theoretically occurs 200 km downwind and is about $10^{-5}$ sec/m$^3$. However, Cramer's steady-state equation is not valid beyond 100 km. At this distance $\chi/Q$ is about $5.2 \times 10^{-6}$ sec/m$^3$.

The EET (Figure A-4) is a function of the length of the spray line, $y$, which, when considering a constant emission time, varies directly with aircraft speed. Thus, at Mach 0.5, the concentration values at either edge of the 3450 meter spray line falls off by only 4% while at Mach 1.5, virtually no concentration loss can be noticed at the spray line's edge.
Figures 3 and 4 give the vertical term (VT) versus the distance downwind for a release altitude of 10 km. Similar calculations for using the edge effects term can be made for the release height.

Figure 5 illustrates the estimated center line dosage term versus distance downwind. This is interpreted as the amount of HF that passes a point during an entire spray episode. The EET (Figure A-4) is applicable to the dosage terms as well.

Figures 6 and 7 show the expected ground level concentration of HF when released from an altitude of 3 km and 10 km respectively.

Other investigations to determine the actual downwind concentration from elevated line sources have been conducted using empirical experiments. Among them was an Air Force test at the Cedar Hill, Texas television tower (7:171). The Air Force released traces of zinc cadmium sulfide from a low flying aircraft while samp'ers were placed at regular intervals downwind to 48 km. The experimenters related cloud expansion to meteorological parameters including vertical and horizontal turbulence and wind velocity. In a well developed turbulent layer, estimated ground level concentrations agreed well with the mathematical model listed below.

\[
\frac{C}{Q} = \frac{2}{3\sqrt{2\pi}} \exp \left(-\frac{H^2}{18x}\right)
\]  

(1)

"For releases above this turbulent layer, ground exposures were much more erratic than predicted by the model (1:171)."

When vertical turbulence is unity the released altitude concentration can be plotted for distances downwind (Figure 8). Cramer's VT, calculated above, can be used together with the above C/Q to estimate ground level concentrations.
Turner (9:41) developed an equation to estimate downwind concentrations at the ground from a finite line source. The wind must be normal to the spray line. The standard deviation in the vertical, $\sigma_z$, is taken from the calculations for Cramer's model.

Figure 9 is an illustration of the estimated downwind ground concentrations from Turner's equation:

\[
\frac{\chi}{Q} = \frac{2}{2\pi \sigma_z u} \exp \left[ -1/2 \left( -1/2 \frac{H}{\sigma_z} \right)^2 \right]
\]

(2)

For distances greater than 100 km downwind, in the mesoscale under turbulent conditions, or in the macroscale, the classical diffusion equations are not appropriate. Predictions of concentration distributions are more accurately made with synoptic forecasts of the movement of large air masses. USAFETAC does not have the capability to make large scale turbulence predictions at this time.
Figure 1. \( \chi/Q \) (Sec/m\(^3\)) at Altitude of Release.
Figure 2. Estimated Ground Level Concentration/Emission Rate ($\chi/Q$) Versus Distance Downwind When HF Gas is Released from an Altitude of 3 km.
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Figure 8. Concentration/Emission Rate, χ/Q, and Release Height versus Downwind Distance from Cedar Hill Experiment.
Figure 9. Concentration/Emission Rate, $\chi/Q$, at Ground versus Distance Downwind from Turner at Two Release Altitudes.
REFERENCES


Appendix A

CALCULATIONS FOR CRAMER'S GENERALIZED CONCENTRATION MODEL

Cramer (3:21) uses a generalized mathematical prediction model containing five terms to calculate downwind line source concentrations. The model is simply written:

\[ \text{Concentration } (\chi) = \text{CCT} \times \text{VT} \times \text{EET} \times \text{AT} \times \text{DT} \quad (A-1) \]

where

\[ \text{CCT} = \text{Centerline Concentration Term} \]

\[ = \frac{Q}{2\pi \sigma_z \sigma_x} \quad (A-2) \]

The emission rate, \( Q \), is unknown, therefore when we solve for \( \chi/Q \),

\[ \text{CCT} = \frac{1}{2\pi \sigma_z \sigma_x} \quad (A-3) \]

where

\[ \sigma_z = \text{standard deviation of vertical concentration distribution} \]

\[ = \sigma_E^*(x_{rz}) \frac{\chi + x_{z} - x_{rz} (1-\beta)^\beta}{\beta x_{rz}} \quad (A-4) \]

where

\[ \sigma_E^* = \sigma A/3 \quad (A-5) \]
where

\[ \sigma_A = \text{standard deviation of the wind azimuth (it can be interpolated from Figure A-1)} \]

\[ = 10 \text{ (using the median expected value, 50% at 20 m/s)} \]

Figure A-1. Relationship Between the Wind Speed 2 Meters Above the Ground and the 10 Minute Standard Deviation of Wind Azimuth Angle (\( \sigma_A \)) in the Daytime (3:52).

\[ x_{rz} = \text{distance downwind at which cloud stabilization occurs - experience shows that a conservative estimate is about 20 meters.} \]
\( \beta = \) vertical diffusion coefficient (3:63)

References sources that show this value approaches unity for elevated releases under neutral conditions.

\( x_z = \) vertical virtual distance - when the standard deviation of the vertical concentration distribution is small, this value

\[
= \frac{\sigma_Z R}{x_{Rz}} \tag{A-6}
\]

\( = \frac{1}{3.3} = 0.30 \)

\( x_{Rz} = \) distance downwind at which the standard deviation of the vertical concentration distribution is measured

\( = 0 \) (since we are interested only in the standard deviation at the source)

Thus, as an example, assume that we are interested in a distance 10 km (10^4 m) downwind,

Then

\[
\sigma_Z = .052(20) \frac{10^4 + 0.3 - 20(1 - 1)}{1(20)} \tag{A-7}
\]

\( = 500 \)

Figure A-2 plots all \( \sigma_Z \) for distances downwind to 100 km.
\( \sigma_x = \text{standard deviation of the downwind concentration distribution.} \)

\[ \text{thus,} \quad \frac{\text{CCT/}Q}{2\pi(500)(1)} = 3.14 \times 10^{-4} \text{ sec/m}^3 \quad (A-8) \]

for a release altitude of 3 km at a distance of 10 km downwind.

\[ \text{VT} = \text{vertical term} \]

\[ \begin{align*}
&= \exp \left[ -1/2 \left( \frac{H-z}{\sigma_z} \right)^2 \right] + \exp \left[ -1/2 \left( \frac{H+z}{\sigma_z} \right)^2 \right] + \\
&+ \exp \left[ -1/2 \left( \frac{2iH_m-H-z}{\sigma_z} \right)^2 \right] + \exp \left[ -1/2 \left( \frac{2iH_m+H+z}{\sigma_z} \right)^2 \right] \\
&+ \exp \left[ -1/2 \left( \frac{2iH_m+H-z}{\sigma_z} \right)^2 \right] + \exp \left[ -1/2 \left( \frac{2iH_m-H+z}{\sigma_z} \right)^2 \right] \quad (A-9)
\end{align*} \]

The vertical term refers to the expansion of the gas in the \( z \)-direction where for Run (1)

\[ H = \text{height of release} = 3000 \text{ m} \]
\[ z = \text{height of interest} = 2 \text{ m (ground level)} \]
\[ H_m = 3000 \text{ (see assumption (1))} \]
\[ \sigma_z = 500 \text{ m (at 10 km)} \]

for one iteration

\[ \begin{align*}
&\text{VT} = \exp \left[ -1/2 \left( \frac{3000-2}{500} \right)^2 \right] + \exp \left[ -1/2 \left( \frac{3000+2}{500} \right)^3 \right] + \\
&\exp \left[ -1/2 \left( \frac{6000-3000-2}{500} \right)^2 \right] + \exp \left[ -1/2 \left( \frac{6000-3000+2}{500} \right)^2 \right]
\end{align*} \]
\[ + \exp\left[-1/2\left(\frac{6000 + 3000 - 2}{500}\right)^2\right] + \exp\left[-1/2\left(\frac{6000 + 3000 + 2}{500}\right)^2\right] \]

\[ = \exp(-18) + \exp(-18) + \exp(-18) + \exp(-18) + \exp(-162) + \exp(-162) \quad (A-10) \]

\[ \text{VT} = 4 \exp(-18) + 2 \exp(-162) \quad (A-11) \]

\[ = 1.52 \times 10^{-8} \]

Only one iteration is used in the vertical term calculation because succeeding iterations become negligibly small relative to the total vertical term. Figure A-3 relates the vertical diffusion term with downwind distances.

\[ \text{EET} = \int_{p_1}^{p_2} \frac{1}{\sqrt{2\pi}} \exp (-1/2p^2) dp \]

\[ = -\exp(-1/2p^2) + \sum_{i=2}^{\infty} \left[ \frac{-p^{(2i-1)}}{i \prod_{n=2}^{i} (2n-3)} \right] \quad \text{(Turner (9:41)) (A-12)} \]

where

\[ p_1 = \frac{y_1}{\sigma_y} \quad \text{and} \quad p_2 = \frac{y_2}{\sigma_y} \]

when the spray line stretches from \( y_1 \) to \( y_2 \),

and

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\[
\sigma_y = \left[ \left( \sigma_a'(\tau) \right) \frac{x_{xy} (x+x_y-x_{xy} (1-\alpha))}{ax_{xy}} \right]^2 + \left( \frac{(\Delta C x)}{4.3} \right) \right]^{1/2}
\]

(A-13)

where

\[
\sigma_a'(\tau) = (\sigma_a'(\tau_0)) \left( \frac{\tau}{\tau_0} \right)^{1/5}
\]

(A-14)

where

\[
\tau = \text{emission time} = 15 \text{ seconds}
\]

\[
\tau_0 = \text{reference time} = 600 \text{ seconds}
\]

\[
\sigma_a'(\tau_0) = \text{standard deviation of the angle measured over reference time} (\tau_0)
\]

= 15° = .263 radians (from Cramer (3:53))

\[
\sigma_A'(\tau) = (0.263) \left( \frac{15}{600} \right)^{1/5} = 0.263(0.025)^{20}
\]

= 1.58x10^-4

\[
x_{xy} = \text{distance at which crosswind cloud stabilization occurs downwind from source.}
\]

= 40 meters (from Dettling (4:12))

\[
x = 10^4 \text{ meters (from example in Equation A-7)}
\]

\[
x_y = \text{crosswind virtual distance}
\]
\begin{equation}
\frac{\sigma_y}{\sigma_A(\tau)} = x_{Ry}
\tag{A-15}
\end{equation}

where

\(\sigma_y\) = standard deviation of the cross-wind distribution
\(x_{Ry}\) = distance at which the standard deviation of crosswind concentration is measured

\(a = \) crosswind diffusion coefficient
\(= 1\)

\(\Delta \theta = \) azimuth wind direction shear between ground level and release level

\begin{equation}
\Delta \theta = \frac{\Delta \theta}{\Delta z} (z_2 - z_1)
\tag{A-16}
\end{equation}

where

\(\frac{\Delta \theta}{\Delta z} = \) rate change of wind direction from surface to release height (radians/meters)

Since the HF is released well above the gradient level, the angle between the surface and gradient level is assumed to be 45° as estimated by Sutton (6:71).

\begin{equation}
= \frac{0.14 \text{ radians}}{3000 \text{ meters}}
\tag{A-17}
\end{equation}
\[ \Delta \theta' = 0.47 \times 10^{-4} \times (3000 - 2) \]
\[ = 0.14 \]

thus

\[ \sigma_y = (1.58 \times 10^{-4}(40) \left( \frac{10^4 + 1 - 40(1 - 1)}{40} \right)^{1/2} + \frac{1.4 \times 10^4}{4.3} \] 
\[ = 811 \text{ meters (for a distance 10 km downwind)} \]

Finally, to calculate the limits of the EET summation term

\[ y_{1,2} = \text{distance from centerline of concentration} \]
\[ = 1725 \text{ meters} \]

then

\[ p_{1,2} = \frac{y_{1,2}}{\sigma_y} = \frac{1725}{811} = 2.13 \]
\[ +2.13 \]

\[ \text{EET} = \int_{-2.13}^{2.13} \exp \left( -\frac{1}{2}(2.13)^2 \right) dp \] 

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.9668 (from Burrington (2:273)) for a sampling height of 2 m when HF is released from a height of 3 km.

Figure A-4 expresses EET relative to airplane speed at release time.

\[ \text{AT} = \text{cloud growth in the along wind direction} \]
\[ = 1 \text{ (using assumptions (1) and (13))} \]

\[ \text{DT} = \text{depletion term or the loss of material by decay processes} \]
\[ = 1 \text{ (using assumption (5))} \]
Figure A-2. Standard Deviation of Vertical Concentration Distribution ($\sigma_z$) Versus Distance Downwind.
Figure A-3. Vertical Term of Cramer's Concentration Equation for a Release Altitude of 3 Km.
Figure A-4. Edge Effects Term (EET) Versus Speed of Aircraft (Based Upon a 15-sec Release Time).