Mitigation of HF releases

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The hazards of hydrogen fluoride (HF) have long been recognized and industry performance reflects sound operating practices. However, full-scale industry-sponsored HF release tests conducted at the US Department of Energy Test Site in 1986 caused concern in view of the toxicity of HF. Ambient impacts were greater than anticipated, and diking, a primary mitigation technique, proved ineffective for releases of pressurized superheated HF. In partial response to these new technical data, an *ad-hoc* three-component industry cooperative hydrogen fluoride (HF) mitigation and assessment programme (ICHMAP) was begun in late-1987 to study and test techniques for mitigating accidental releases of HF and alkylation unit acid (AUA), and to enhance capabilities to estimate ambient impacts from such releases. The programme's mitigation components have recently been completed while work on the impact assessment component is nearing completion. The purpose of this article is to describe the programme and to summarize the objective, scope of work, structure, and conclusions from the two mitigation components. The objectives and scope of work of the impact assessment component are also briefly described. Detailed and summary reports for each programme component will shortly be available through the US Department of Commerce, National Technical Information Service (NTIS) of Springfield, Virginia. ICHMAP member companies, collaboratively and individually, screened numerous potential mitigation techniques for accidental HF releases including techniques such as total enclosures with exhaust scrubbers, liquid containment dikes, foam application, water sprays, vapour barriers, and surface tension modifying agents. Water sprays and vapour barriers were judged to be the most promising techniques. Accordingly, the ICHMAP mitigation components consisted of both water spray and vapour barrier programmes. The water spray programme investigated the effectiveness of water or augmented water application systems in mitigating accidental releases of both HF and AUA. The vapour barrier programme assessed the effectiveness of vapour barriers in delaying and diluting accidental releases of heavier-than-air HF vapour clouds in an industrial setting and determined what impact such barriers might have on the consequences of an unconfined vapour cloud explosion.

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Industry performance reflects the fact that sound practices for handling HF are in place. HF hazards are well known and operating practices have been aimed at minimizing the possibility of a release and mitigating the effects of a release should it occur. These practices have been continually monitored and improved to maximize safety protection based on the technical data available. This industry programme has been aimed at further improvements based on new technical data, primarily those derived from the HF release tests conducted by Amoco and Lawrence Livermore National Laboratories in the summer of 1986.

Prior to 1986, accidental releases of pressurized, superheated HF were commonly thought to form liquid pools. This led to the concept that the ambient impacts of accidental HF releases could be mitigated by liquid containment dikes.

In the summer of 1986, Amoco, Allied-Signal, DuPont, Mobil, and Lawrence Livermore National Laboratories conducted a series of atmospheric HF release tests at the Department of Energy (D.O.E.) test site in Mercury, Nevada (code-named the Goldfish series). The major conclusions from these tests were that releases of pressurized, superheated HF did not pool, negating the benefit of liquid containment dikes, and that the resultant ambient impacts were greater than anticipated because the vapour clouds advected downwind consisted of flashed HF vapour and entrained HF liquid as a finely dispersed aerosol. In addition, the Goldfish tests demonstrated through three limited tests that water sprays were at least partially effective in mitigating such releases of HF.

In mid-1987, ICHMAP was formed. This programme, which was formally established in December 1987, had as its broad objectives the development of information to better design and implement effective

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mitigation techniques for accidental releases of HF and the development of a computer tool to better estimate ambient impacts from accidental HF releases[1]. The programme was structured with a limited charter and life and was open to any interested company on a cost-shared basis.

A total of 20 companies participated in the ICHMAP. These included both HF producers and consumers. Participating companies included Allied-Signal, Amoco, Ashland, BP, Chevron, Conoco/Dupont, Dow, Elf Aquitaine, Exxon, Kerr-McGee, Marathon, Mobil, Phillips, Saras, Shell Internationale, Sun/Suncor, Tenneco, Texaco, Unocal, and 3M.

The three programme components and their major objectives are summarized as follows:

- water spray component: investigate the effectiveness of water or augmented water application systems in mitigating accidental releases of both HF and AUA
- vapour barrier component: assess the effectiveness of vapour barriers in delaying and diluting accidental releases of heavier-than-air HF vapour clouds in an industrial setting, and determine what impact such barriers might have on the consequences of an unconfined vapour cloud explosion
- impact assessment component: develop validated ambient impact assessment computer models for calculating release rates and jet and plume dispersion of accidental releases of HF

This paper will summarize the work conducted as part of the water spray and vapour barrier components, and will briefly describe the objectives and scope of work of the impact assessment component. The impact assessment component is nearing completion and will be reported on in other technical papers and ICHMAP detailed reports.

Water spray component

The overall objective of the water spray component was to investigate the effectiveness of water or augmented water application (via either sprays or monitors) in mitigating accidental releases of both HF and AUA as a function of flow conditions, HF acid and water spray properties, and geometric factors. Augmented water refers to water plus additives such as sodium bicarbonate, caustic, or surfactant. This work was largely a follow-on to the 1986 Amoco HF spill tests, i.e. the Goldfish series. Goldfish included three water spray tests that demonstrated 35 to 55% HF removal efficiency. These tests provided valuable information, but were limited in scope and detail and left several key questions unanswered[2].

To meet the objective of this component, a series of bench-scale laboratory and larger-scale field tests were conducted in controlled humidity/wind speed flow chambers. The bench-scale experiments identified the key variables for testing in the larger-scale field tests. In the larger-scale field tests, HF and AUA removal efficiencies were measured as a function of variables such as water to HF liquid ratio, water application method (sprays, fire monitors), water additives (neutralizing agents, surfactants), distance of the water application to the release point, water application droplet size, ambient relative humidity, etc.

It was outside the scope of this work to generate generic design criteria for water mitigation systems, since each application depends on many local factors. However, the conclusions from this work can provide guidance to those responsible for designing mitigation systems. To this end, two separate computer models were developed to assist in assessing the effect of various water application design parameters on HF removal efficiency.

Overview of laboratory test programme

The objective of the small-scale laboratory test programme was to help define the test matrix for the larger scale field testing, to evaluate design criteria for a larger flow chamber, and to gain operating experience with HF and AUA for the larger unit[3,4].

In the laboratory, a 6 inch wide by 12 inch high by 40 inch long totally enclosed flow chamber made of clear Lexan was set-up under an exhaust hood. Air flowed from the inlet of the chamber to an external scrubber. Anhydrous HF or AUA was released through an orifice at the upstream end of the chamber at a rate of approximately 150 $ml\,min^{-1}$ for 10 min. The acid flashed to vapour and aerosol at the exit of the orifice and was contacted with either water or augmented water via a pair of spray nozzles. The acidic water was collected and analysed for HF content. The unscrubbed HF was removed in the external scrubber.

Among the key variables identified, the water flow rate at a constant acid flow rate was clearly the dominant variable. In addition, HF removal efficiency was found to increase as the droplet size of the water spray decreased. Among the various water additives tested, which included $NaHCO_3$, $CaCl_2$, NaOH, and surfactant, only NaOH showed a marginal increase in removal efficiency compared with pure water.

Overview of field test programme

A series of 87 large-scale field tests were conducted in which the effect of major variables on the HF removal efficiency of water or augmented water, applied via either sprays or a fire monitor, was determined. This test series also included seven tests in which aerosol particle sizes were measured.

The 87 tests were conducted at the US Department of Energy's Liquefied Gaseous Fuels Spill Test Facility, located in the Frenchman Flats area of the Mercury, Nevada test site. The test series has been designated as the Hawk series. *Figure 1* shows the entire HF water spray facility.

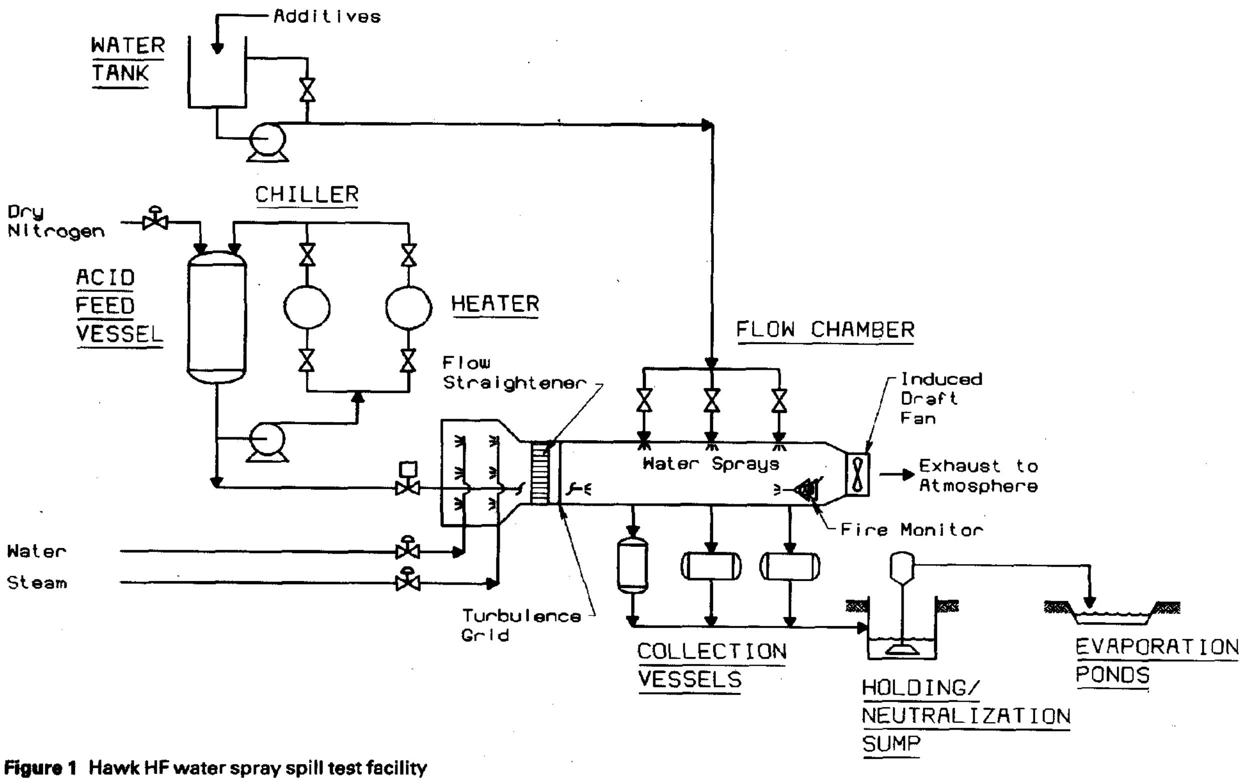

Figure 1 Hawk HF water spray spill test facility

Pressurized, superheated HF, either in pure anhydrous form or in the form of AUA, was released at controlled pressure and temperature into a totally enclosed flow chamber (140′ long, 16′ high, 8′ wide) and contacted with water or augmented water. The inside of the flow chamber during a typical Hawk downflow water spray test is shown in *Figure 2*. *Figure 3* shows a typical Hawk fire monitor test.

In the chamber, water was applied via either sprays or a fire monitor. The HF was released from a square-edged orifice at a nominal rate of approximately 5 gpm. The flow chamber was kept under negative pressure via an induced draft fan. It included an inlet flow straightener to provide uniform air flow by decoupling the chamber from ambient wind speed variations and a turbulence grid to provide a known level of turbulence. Steam and water spray grids at the chamber inlet allowed for control of both air humidity and temperature. Acidic water was collected in three drums located beneath the flow chamber and was analysed for HF concentration. In addition, HF concentrations within the flow chamber were measured using integrated filter samplers. The acidic water was routed to a sump, neutralized in evaporation ponds and disposed of at a waste disposal site. Temperature was recorded at various positions within the flow chamber. In addition to releases of pressurized HF, several tests were conducted to assess the HF removal efficiency of water sprays for HF pool evaporation releases.

The impact of the following variables on HF removal efficiency was determined:

- water-to-HF liquid volume ratio
- water spray application geometry: number of sprays per header; spray header distance from the release point; spray orientation (upflow and downflow); spray header elevation above the release point; dual spray headers in series; water spray droplet size
- water application via a fire monitor: application pattern (fog or jet); monitor distance from the release point
- acid type (anhydrous HF or AUA)
- acid temperature, pressure
- water additives ($NaHCO_3$, surfactant, NaOH)
- ambient relative humidity and wind speed
- steam as an acid jet dispersant

In addition to measuring HF removal efficiency, seven tests were conducted in which the size of the HF aerosol particles (liquid HF entrained with the flashed vapour) was measured using Insitec Corporation's Particle Counter Sizer Velocimeter (PCSV). The PCSV is a laser-based light scattering instrument that makes direct in-line particle size measurements.

The major conclusions from the large-scale field tests are as follows:

- Water-to-HF liquid volume ratio was identified as the major variable affecting removal efficiency.

Figure 2 Representative Hawk upflow water spray test

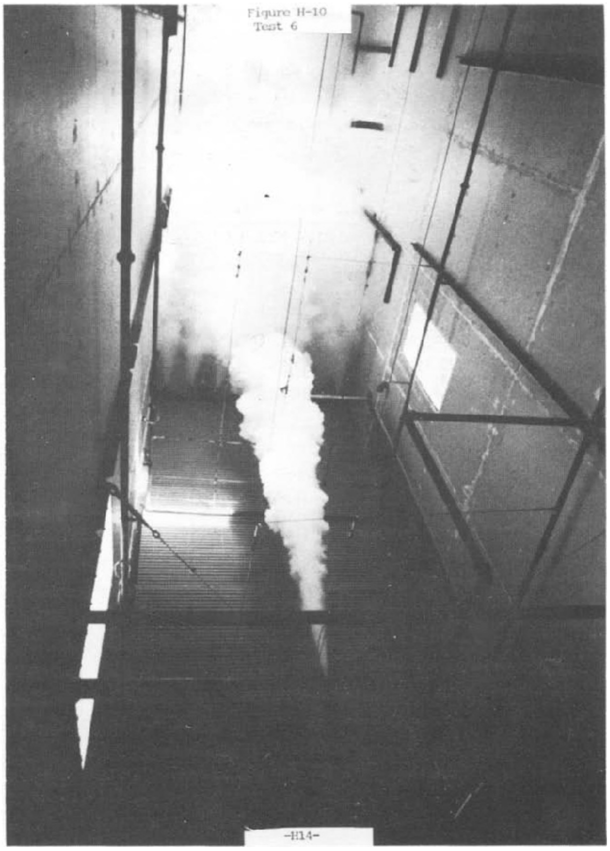

Figure 3 Representative Hawk fire monitor water spray test

- HF removals of 25 to 90+% were demonstrated at water-to-HF liquid volume ratios of 6/1 to 40+/1. *Figure 4* shows HF removal efficiency versus water-to-HF liquid volume ratio for the base case (downflow sprays at 8′ elevation located 16′ from release point, 320 micron Sauter mean water spray droplet diameter, anhydrous HF).
- As seen in *Figure 4*, fire monitors demonstrated HF removals nearly as high as those demonstrated with water sprays.
- Water additives (neutralizing agents, surfactants, etc.) and steam as a jet dispersant had little measurable effect on removal efficiency.
- Water spray removal efficiency increased with: decreasing water spray droplet size (320 to 160 μm); increasing distance between the spray nozzles (2′ or 3′ nozzle spacing versus 1′ in the base case); decreasing spray header elevation above the release point (16′ versus 8′ in the base case).
- Dual water sprays (i.e., two spray headers in series, separated by approximately 15′) had little measurable effect on HF removal efficiency at a constant total water-to-HF liquid volume ratio.
- Upflow water sprays provided higher removals than did downflow water sprays.
- There was essentially no difference in HF removal efficiencies between HF and AUA for a constant water-to-total contained HF acid liquid volume ratio.
- Wind speed and relative humidity had little measurable effect on removal efficiency under the conditions tested.
- A fire monitor provided better HF removals when operated with a coarse droplet jet pattern aimed directly at the release point from a short distance

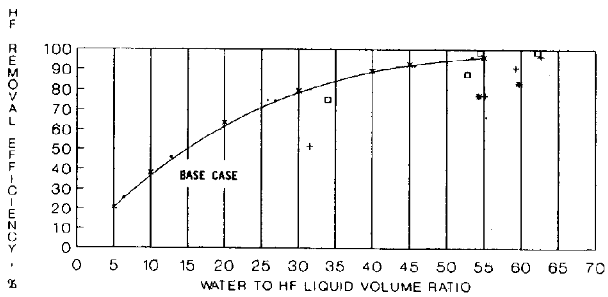

Figure 4 Water spray removal of HF base case (sprays) and monitors. (Base: downflow sprays at 8 ft elevation, 15 ft from release, 350 μm water spray droplet size). •, base case (sprays); *, jet monitor at 50 ft; +, fog monitor at 20 ft; □, jet monitor + plate

(as opposed to a wide fog pattern or a jet applied from a greater distance).

- The aerosol produced at the exit of the release orifice was predominantly submicron in size. The aerosol particle size was unaffected by acid pressure, acid type, acid temperature, or relative humidity.
- HF storage pressure appeared to affect HF removals, but adequate data were not collected to isolate the potential effect of other variables.

Aerosol droplet size measurements

In addition to the water application mitigation tests, a series of tests were conducted in the large-scale flow chamber to measure the size of the aerosol droplets produced when pressurized, superheated HF or AUA is released into the atmosphere. A newly developed *in-situ* optical instrument (PCSV-P, a single particle counting instrument based on absolute light scattering) was used to obtain on-line measurements of the aerosol droplet sizes in a total of 86 data sets[5].

To briefly summarize the results, the PCSV-P measured predominantly submicron liquid aerosol droplets at the exit of the sharp-edged discharge orifice. The aerosol droplets subsequently grew to larger particles at the outlet of the flow chamber. The presence of submicron particles at the orifice exit point was probably due to formation of aerosol by the process of flash atomization break-up as opposed to conventional shear spray break-up. Temperature, humidity, release pressure, radial position in the plume, and type of acid did not appear to have any significant effect on the measured size distributions for the range of conditions investigated. The increase in droplet size at the exit of the chamber is likely to be due to the condensation of ambient moisture resulting from contact with the cold HF plume.

Application of test results

In applying the results of this work to conditions different from those prevailing in the field tests, due account must be taken of the dynamic interaction between the water spray and the HF cloud. Work has recently been completed by the ICHMAP's Water Spray Subcommittee to characterize these interactions. Two separate computer models have been developed to assess the effect of various design parameters such as water-to-HF liquid volume ratio, distance of the water application from the release point, water application method (i.e., upflow or downflow sprays, fire monitors), on overall HF removal efficiency. A supplementary report on this work is planned for release in mid-1990.

In developing a water mitigation system design, it is imperative that the HF leak be detected as quickly as possible and that large amounts of water be applied as rapidly as possible to provide a high water-to-HF volume ratio. At present, there is no proven, reliable HF-specific leak detector available. However, work is progressing rapidly in several companies to develop such a device. This work includes development of both point detectors and area detectors. In the absence of such a device, industry experience has shown that remote TV surveillance via multiple cameras, perhaps coupled with contrast video (i.e., imaging), can be an effective method to rapidly detect HF leaks.

Damage tests with fire monitors

If the mitigation water is applied by fire monitors, a potential concern is damage to the unit resulting from rapid activation and application of high pressure, large-volume water monitors. To help assessment of the extent of this concern, Exxon conducted a series of fire monitor damage tests at a mothballed process unit in Sarnia, Canada.

A series of tests were conducted in which water was applied via fire monitors at rates of 2000, 4000, and 6000 gpm at distances of 7, 15, and 25 ft from the process unit. The target area included pressure gauges, small-bore piping, control valve stations, and conduit. Water was applied via both a wide angle fog pattern and a straight jet pattern. The water delivery pressure was approximately 100 psig.

In the most severe test, water was applied in a narrow jet pattern at a rate of 6000 gpm at a distance of 7 ft from the target area. The resulting damage to the target area was minimal and consisted mainly of broken pressure gauge glasses, dented insulation, and some loosening of conduit from its supports. The overall integrity of the target area was not affected, i.e. there was no broken piping or instrument tubing.

Vapour barrier component

The primary objective of the vapour barrier component was to assess the effectiveness of vapour barriers in delaying and diluting accidental releases of heavier-than-air (HTA) HF vapour clouds in an industrial setting. Because such barriers might be applied in an HF alkylation plant where there is also the potential for flammable releases, another objective was to determine what impact, if any, such vapour barriers might have on the consequences of an unconfined vapour cloud explosion. Finally, a third objective was to quantify the impact that existing obstacles in an industrial plant (e.g., tanks, pipe racks, towers) might have on the dilution of a HTA vapour cloud.

Review of previous related work (Phase I)[6]

The major objective of this phase was to analyse existing field and laboratory data sets relevant to obstacle/vapour barrier impacts on HTA cloud dilution and transport time. Other objectives included assessing the need to conduct more detailed wind tunnel studies to better quantify the impact of vapour barriers on dilution and transport time and determining how HTA releases of HF could be simulated in such wind tunnel studies. Two independent studies were conducted[7,8].

The following major conclusions were found:

- Vapour barriers and obstables can reduce near-field concentrations, but the level of concentration reductions tended to decrease with increasing downwind distance.
- Adequate data were not available to completely assess far-field impacts.
- Near-field cloud arrival times tended to increase as a result of barriers and obstacles.
- HF can be accurately modelled in a wind tunnel using an appropriately selected simulant gas. The procedure developed to simulate HF in a wind tunnel was used for the Phase III wind tunnel simulations.

Flammability concerns (Phase II)
Accidental releases of hydrocarbons as well as HF are possible in an HF alkylation plant. Therefore, if vapour barriers are considered for mitigating HTA HF releases in an HF alkylation plant, the potential impact of such barriers on flammable releases needs to be considered. The objective of Phase II was to determine the impact a vapour barrier might have on flammable vapour cloud explosion overpressures. This component phase was conducted by Christian Michelsen Institute (CMI)[9,10].

To quantify the impact of a vapour barrier on flammable cloud explosive overpressures, the FLACS code (a proprietary model developed by CMI) was used to simulate the release of $7.6\,\mathrm{kg\,s^{-1}}$ of iso-butane. This release rate was specified by the ICHMAP Vapour Barrier Subcommittee based on the results of several alkylation plant hazard and operability reviews as being a representative rate for a large accidental hydrocarbon release from an HF alkylation plant. Peak overpressures within the alkylation plant were estimated, both with and without a vapour box. Sensitivity runs were conducted to assess the impact of vapour box volume and height.

The following major conclusions were found:

- Peak explosion overpressure is, for a given plant geometry, primarily dependent on the height of a flammable vapour cloud.
- A vapour box will result in appreciable increases in peak overpressure, primarily through its effect on cloud height.
- Decreasing the height of a vapour box and/or using explosion vents in the box walls will decrease peak overpressures, which will however still be appreciably greater than for the no-box base case.
- Peak overpressures will increase as the perimeter of the vapour box decreases.

It should be pointed out that Phase II did not assess certain other important factors such as the impact of a vapour box on the probability of an UVCE.

Wind tunnel modelling (Phase III)
Cermak Peterka Petersen Inc. (CPP) conducted a series of over 200 simulations in a boundary layer wind tunnel (75′ by 8.5′ by 12′) to quantify the impact of vapour barriers and obstacles on the dilution and transport time of simulated HTA HF clouds. These simulations examined the effect of both uncontrollable and controllable variables on barrier performance. The simulations are valid for aerosol-type HF releases, but are not applicable to pool evaporation or vapour HF releases.

Concentrations were measured at scaled distances equivalent to full-scale distances of up to 3000 m for both barrier/no barrier and obstacle/no obstacle cases at ground-level and at various elevations. Concentration reduction factors and cloud arrival, departure, and duration times were calculated. The concentration reduction factor is defined as the base case concentration divided by the concentration with obstacles. The base case for the industrial plant obstacle simulations without barriers was flat, unobstructed grassland. The base case for the barrier simulations was the plant obstacles without barriers. In addition, mass flux integrations were performed for several vapour box configurations to quantify the amount of vapour leaving the box as a function of time.

Site configurations simulated included both a heavily-obstructed and a moderately-obstructed process unit. The uniform surface roughness upwind and downwind of the process units was typical of grassland. Sensitivity cases were run to assess the impact of larger upwind and downwind surface roughness typical of an urban setting.

To help validate the results of the wind tunnel simulations, several selected data sets from the Thorney Island and the 1986 Amoco Goldfish HF full-scale field trials were simulated. The results compared favourably with the field data. Predicted concentrations were within a factor of two of field-observed concentrations.

Figure 5 shows the various vapour fences (i.e., 2-dimensional vapour barriers) that were tested. As stated previously, sensitivity runs to determine the effect of fence height and fence distance from the release point were conducted. *Figure 6* shows one type of vapour box tested. This box surrounds not only the release point, but also the unit from which the release is assumed to emanate. The openings at the bottom of the box were intended to represent openings over which one might install water sprays to scrub-out HF (i.e., a hybrid mitigation system). Tests were run with the doors both closed and open. The intent of the open door tests was to determine the amount of gas that would flow out of the vapour box through the openings. Sensitivity runs were conducted to determine the effect of vapour box height, construction (e.g., permeable or solid, with and without flow spoilers at the top), and placement relevant to upwind plant obstacles.

The following major conclusions were found:

- The impact of any barrier on cloud dilution and delay is extremely dependent on: the release scenario (e.g., release rate, duration, density decay as a

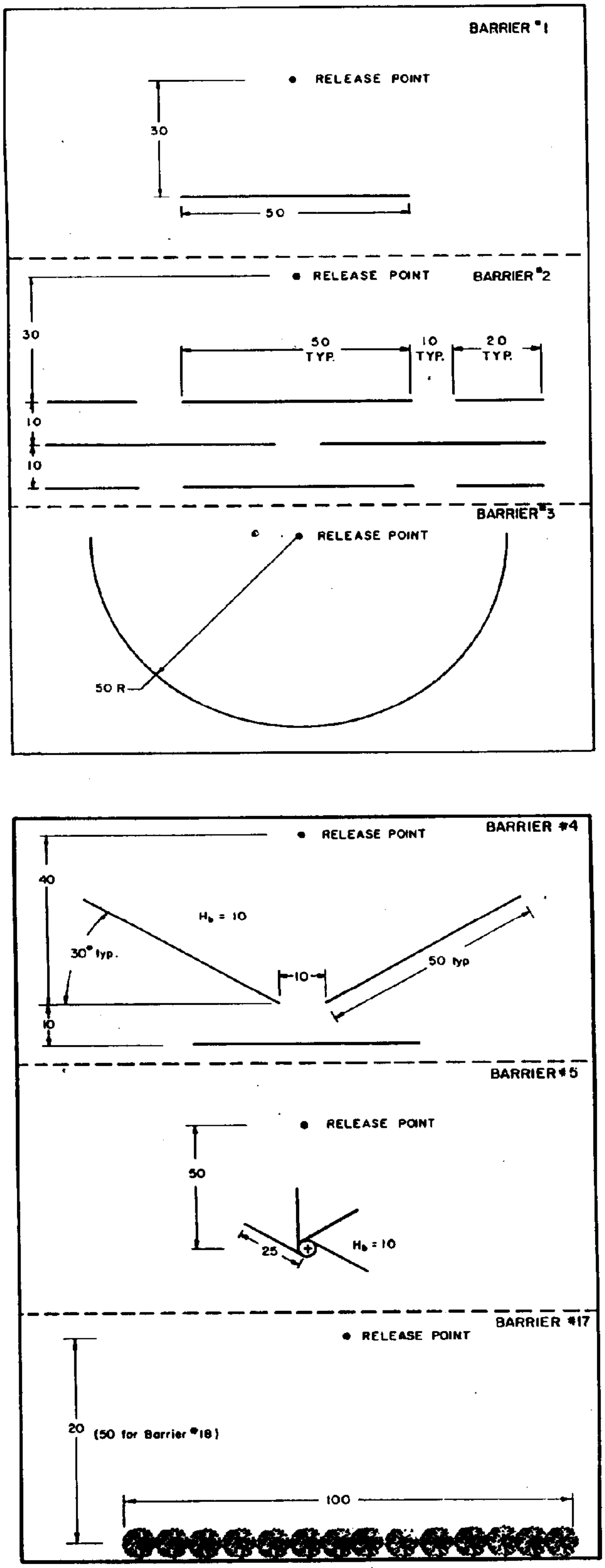

Figure 5 Vapour fences tested in wind tunnel. (Barrier height 10 m, all dimensions in m)

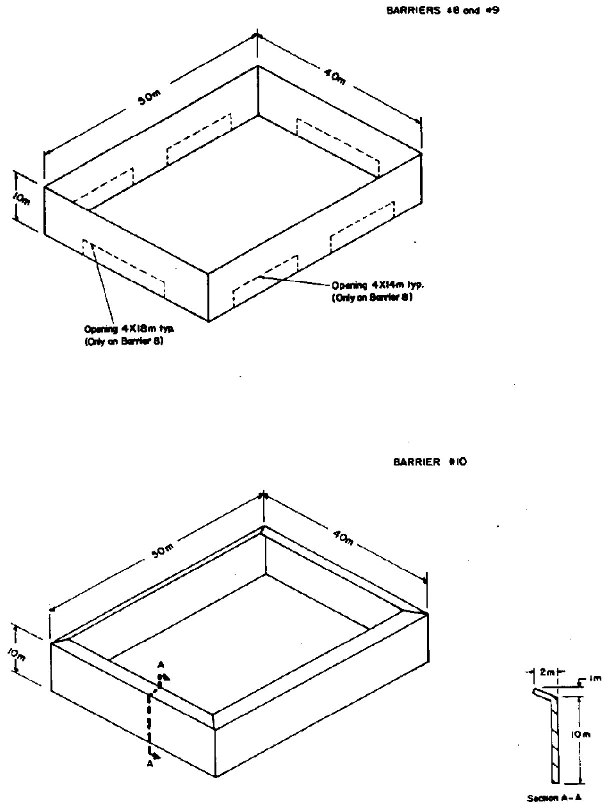

Figure 6 Vapour boxes tested in wind tunnel. (Barriers 11 and 12 similar to barrier 10, except dimensions are 28 × 38 m and 52 × 65 m, respectively)

function of air entrainment, wind direction); the specifics of the site (e.g., nature of upwind plant obstacles, surface roughness); and the design of the barrier (e.g., height, distance from upwind plant obstacles).

- As seen in *Figure 7*, existing plant obstacles (e.g. tanks or pipe racks) will decrease near-field concentrations compared with flat, unobstructed grassland by factors ranging from 3 to 25, but concentrations will return to the grassland base case within approximately 3000 m of the release point.
- Two-dimensional vapour barriers (i.e., fences) will typically: reduce near-field concentrations by factors ranging from 2 to 9 (see *Figures 8a* and *8b*), but concentrations will return to the base case no-barrier levels within approximately 1000 m of the release point; and not delay appreciably a HTA cloud.
- Three dimensional vapour barriers (i.e., open-top enclosures) will typically: reduce near-field concentrations by factors ranging from to 4 to 15 (see *Figures 9a* and *9b*); reduce far-field concentrations by affecting a reduction in the rate of gas advected

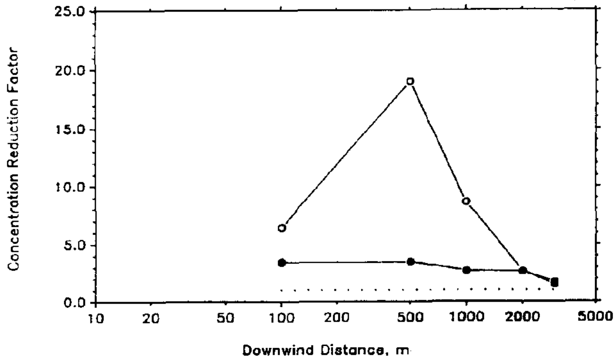

Figure 7 Observed concentration reduction factors – plant obstacles: ○, heavily obstructed; ●, moderately obstructed. U_{wind} at 10 m = 5 m s^{-1}; $Q_{release}$ = 6.3 kg s^{-1}; release duration = 60 s; receptor location = 1 m above grade at cloud centreline; initial density ratio = 1.33 (air = 1.0); flat grassland as base case

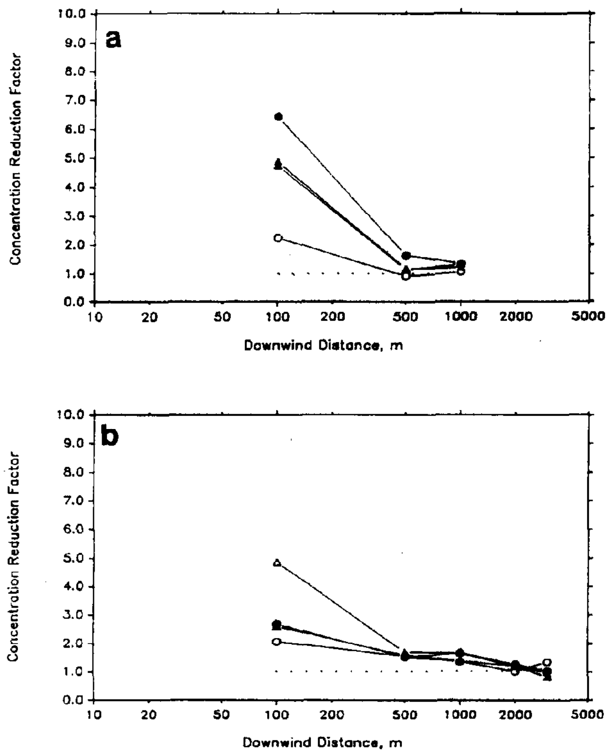

Figure 8 Observed concentration reduction factors – vapour fences: a, heavily; and b, moderately obstructed units. ○, Barrier 1; ● barrier 2; △, barrier 3; ▲, barrier 4. Details as in *Figure 7*

downwind; as seen in *Figures 9a* and *9b*, far-field concentration reduction factors ranging from 1 to 4 were observed with the degree of reduction being dependent on the release rate and duration and the barrier volume; increase both the arrival time and duration of a HTA cloud, with the amount of delay dependent on the nature of obstacles upwind of the barrier; cloud arrival times were delayed by about

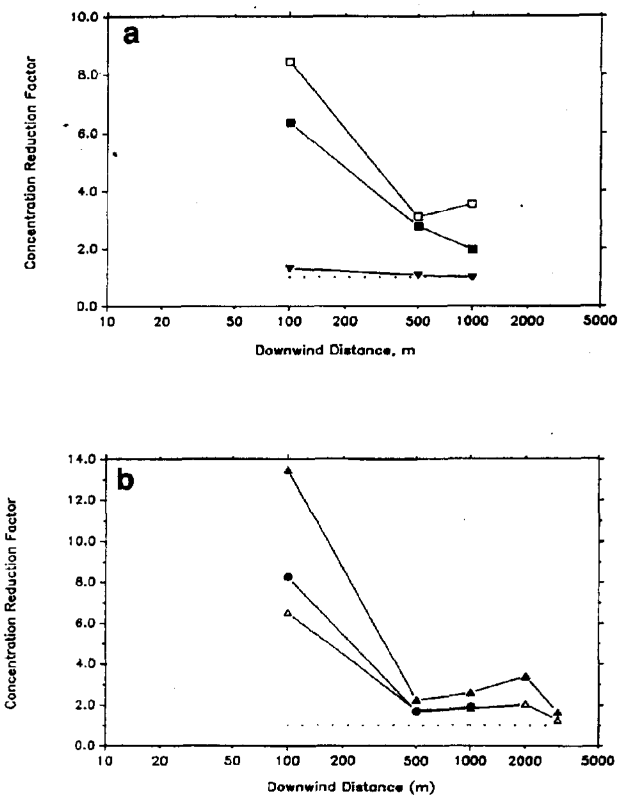

Figure 9 Observed concentration reduction factors – vapour boxes: a, heavily obstructed unit, ▼, vapour box 8, ■, vapour box 9, □, vapour box 10; b, moderately obstructed unit; ●, 5 m high box; △, 10 m high box; ▲, 20 m high box. Details as in *Figure 7* except: $Q_{release}$ = 6.3 kg s^{-1} (6A) and 25.3 kg s^{-1} (6B); release duration = 180 s (6A) and 240 s (6B); and upwind process unit as base case

500 s with a heavily obstructed process unit upwind of the barrier and were delayed by <50 s with a moderately obstructed process unit upwind of the barrier.

Ambient impact assessment component

The objective of the Ambient Impact Assessment Component was to develop a PC-based computer model that would better estimate downwind concentrations from an accidental release of HF as well as the effectiveness of mitigation systems. The three major areas in which substantial uncertainties existed in generally available ambient impact predictive tools were identified as:

- Modelling the complex thermodynamics of HF/H_2O/air mixtures (including aerosol) and their effects on cloud density
- Treatment of a wide range of surface roughness conditions (including possible multiple surface roughness conditions)
- Jet flow and air entrainment for pressurized elevated releases of HF, followed by transition to ground-based dense-gas dispersion.

This component was conducted in two phases. In the first phase, several widely available ambient impact assessment models were screened and evaluated to determine which model would be most appropriately modified to meet the objectives outlined above. Shell's HEGADAS model was selected.

In the second phase, which is nearing completion, the HEGADAS model was modified to incorporate the thermodynamic behaviour of dense HF/H_2O/air mixtures. Also, the method used by HEGADAS to represent the effects of surface roughness conditions on dense gas behaviour was validated against available data (including the wind tunnel data generated by CPP as part of the vapour barrier component). This method was then modified to include a new formulation for gravity spreading effects on dense gas dispersion, and a jet/plume model was developed that provides a unified representation of the stages of the flow from a pressurized release, through elevated plume dispersion, to the touchdown of dense plumes and transition to ground-based dispersion. To provide a complete source and dispersion modelling package, the programmes being developed also include a source release rate model, a pool evaporation model, and a far-field Gaussian plume model.

This component is nearing completion and it is anticipated that its deliverables (a PC-based programme, a technical reference manual, and a users guide) will be available shortly.

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