USE OF REAL-TIME MEASUREMENTS FOR ESTIMATING RELEASE RATE

Shahryar Khajeh Najafi*, Ernie Gilbert
SAFER Systems, L.L.C., Camarillo, CA

There are several key parameters that influence the action taken by the emergency responder on the scene of a chemical spill: the location of the source; the type of chemical is involved; and how much chemical is being released. The objective of this paper is to present a methodology for using meteorological data and information from chemical sensors to obtain the requested information. The meteorological data is used to locate the source. The sensors feedback is used to calculate the release rate and identify the chemical. To reconstruct the emission rate versus time, a dispersion model which is capable of incorporating the real-time data has been utilized.
The proposed methodology was tested against field trials. The maximum error between the simulated and measured rate ranged from 20% to 90% for the trials. It was concluded that of all the parameters affecting the performance of release rate estimation algorithm, the near source phenomena of aerosolization and dilution at the source, and the plume meander are the most important parameters. A two-tier approach is suggested for the rate estimation depending on the source of emission. For high momentum releases (e.g. tank/pipe), the goal would be to find the size of the rupture, and for low momentum release (e.g. pool), the goal would be to find the emission rate.

Backcalculation, sensor, dispersion

INTRODUCTION

A typical chemical release event may involve several derailed railroad cars leaking unknown amounts of chemical or a plant process area engulfed in a toxic gas. In events like these, it is very difficult to locate the leak and to determine the amount of chemical being released. Initial release rate estimates are very challenging and even an expert responder can only guess. Response agencies who intend to warn people in harm's way and evacuate would therefore have a great demand for new method for quick and accurate estimation of release rate.

For accident within a plant, the release rate can be calculated by a process engineer from a mass balance around the leaking vessel/pipe using the information available to him from process control unit. This method works best if the release rate is substantial. In the case of small leaks, the release rate estimation is very difficult. For mobile sources (e.g. train derailment or truck accident) release rate estimation is very difficult during an actual event.

The objective of this paper is to present a methodology of inverting downwind concentration measurements using an appropriate dispersion model to reconstruct emission rate versus time. We call this method backcalculation. Utilizing weather data and the position of the deployed sensors, the source location can be identified. The unique response of sensors to a specific chemical would also determine their identity.

Using real-time concentration measurement to estimate the rate has been studied previously by Lehning et al. (1994) and Piccot et al. (1994, 1996). Lehning studied the possibility of inverting the downwind concentration measurements to calculate the total emission rate and distribution of a non-uniform area source. Piccot used the open-path Fourier

Transformation Infrared (FTIR) spectrometer to measure the emission from area and volume sources. They used a Gaussian model to obtain an integrated average concentration along the spectrometer's path and compared the result with the measured values. They showed good performance of the model when an ad hoc method for stability calculation was used. These methods are mostly suitable for measuring emissions from fugitive steady sources. The present study proposes a methodology that can be utilized during an emergency event.

The chemical sensors can be categorized into two groups: point detection sensors and scanning devices sensors. The scanning device sensors include: Lidar (Light Detection and Ranging), Dial (Differential Absoption Lidar), Fourier Transform Infrared (FTIR) spectrometers, thermal imaging. Points sensors include Photo-Ionization Detector (PID), Flame-Ionization Detector (fiD), electro-chemical, and paper tape. The backcalculation algorithm works best with the point sensors. Due to cost constraints, portable sensors which can be dropped into the path of the cloud during an episodic release are more practical. The portable sensors transmit, via radio wave, their geographic location (latitude/longitude), gas concentration and the time of that measurement to the backcalculation algorithm. The activities of the sensors can be observed on a GIS based mapping system. The sensors can also be mounted on vehicle (with positive pressure inside) which can be driven through the cloud for sampling and transmitting the results to the base.

One of the advantages of portable point sensors is their flexibility in terms of rapid deployment. The open-path instruments like FTIR need some time to set up and it is very hard to move them around. This makes point sensors more suitable for emergency events.

With the recent terrorist attack and vulnerability of chemical plants to such threats, chemical companies can not afford to ignore the importance of a combined sensors/ backcalculation set-up as a tool for early warning and mitigation. The sensor technology has matured to a point that the sensors are durable, affordable, and able to measure concentration in real time. Most of them have a global positioning system and can transmit information via radio wave. Rapid detection of release enables prompt implementation of corrective action and a response procedure.

DESCRIPTION OF THE MODEL

An integrated Lagrangian puff model is used for this study. The model allows for the incorporation of real-time weather and sensors data for backcalculation. It uses the 16 sectors wind rose (N, NNE, NE, E, …) where sectors are 22.5 degrees apart. The wind speed, direction, temperature, and solar radiation are measured. The polling frequency for these parameters is 3 seconds. The archived data which is used for backcalculation represent five minutes running average. The vertical and horizontal stabilities are calculated based on the 10 minutes running average of wind speed and wind direction. The polling frequency for the sensors is one second with an archived value of one minutes running average.

The dispersion model uses the Richardson Number criteria to invoke the Gaussian or dense gas sub models.

$$\mathrm{Ri} = \frac{\mathrm{g}\,\dfrac{(\rho_g - \rho_\mathrm{a})}{\rho_\mathrm{a}}\,\mathrm{H}}{U_*}$$

Where ρ_g, and ρ_a are the gas and air density, H is the height of element, and U_* is the friction velocity. Therefore, for a specific gas and weather data, the quantity of release which affects the height of the puff, would determine the type of the model to use.

OVERALL VIEW OF THE CONCEPT

Basically there are five major components in a backcalculation assessment:

1. Gas detection sensors
2. Meteorological measurement
3. Release location
4. Starting time of the release
5. Sophisticated dispersion model

To find the release location a reverse corridor is constructed from the position of each impacted sensor utilizing the weather data (Figure 1). The reverse corridor is a wedge drawn from the position of each sensor using the opposite wind direction measured by the meteorological tower. So if wind is blowing from the west the reverse corridor is drawn from the east. The angle of the corridor depends on the atmospheric stability.

To calculate the wedge angle, a downwind distance of one kilometre is selected (OA). The horizontal dispersion parameter (σ_y) is found from Pasquill-Gifford charts using appropriate stability curve. The wedge is then constructed by connecting the end of a line segment, which is perpendicular to the reverse mean wind direction, to the sensor location. The length of this line segment is 2.14 times the horizontal dispersion parameter. The factor of 2.14 sets the cloud width where the concentration falls to about one percent of the centreline value.

The intersection of those wedges would contain the most probable area for the source location. This technique is very useful in narrowing down the location of the source considering that most of the time the chemical presence is suggested by its smell and not its visibility.

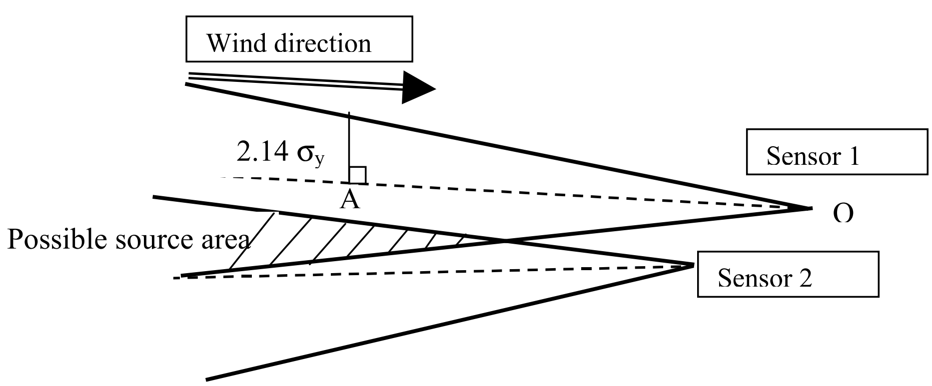


Figure 1. Construction of reverse corridor

The backcalculation uses a trial and error procedure (Figure 2) to estimate the rate. For each trial rate a concentration-time profile is predicted by the model at each sensor location (figure). Actual sensor information, namely, the time and measured concentration is compared against the model prediction. If there is a match within the convergence span in terms of time and concentration measurement for all the sensors, the predicted release rate is recorded. Otherwise, a new release rate is assumed. There are two loops for convergence; one loop converges on the time, the other converges on the concentration. A substantial change in measured concentration is an indication of a rate change. This process would render the rate vs. time profile for a transient source. The release rate is then fed to the dispersion model to render the final plume impact. This procedure is repeated as new information is received and updates are necessary.

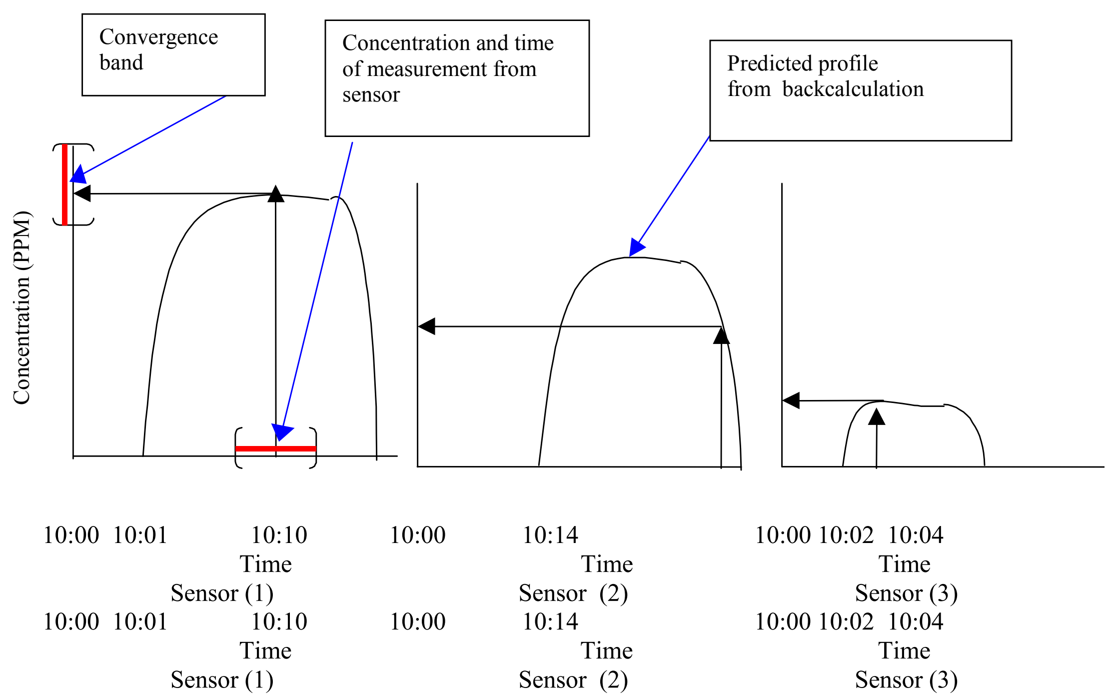
The greatest accuracy would be obtained if the sample is taken close to the plume centreline and away from its edges. To take this fact into account, a weight factor is assigned to each sensor due to its position with respect to plume centreline. The weight factor would adjust the convergence criteria for each sensor.

The convergence criteria is defined as:

$$(C_{meas} - C_{est})/C_{meas} < Tol/W_n$$

Where:

Cmeas = Measured concentration
Cest = Estimated concentration
Tol = Tolerance
Wn = Weight factor for each sensor ($0 < W_n < 1$)



10:00 10:01 10:10 10:00 10:14 10:00 10:02 10:04
Time
Sensor (1)
Time
Sensor (2)
Time
Sensor (3)

Figure 2. Comparison of model predictions against the sensors measurements

A global tolerance level of 10-3 is assumed, but this value is adjusted (increased) as the sample points get farther away from the mean wind direction. Therefore, different convergence criteria is used depending on the location of the sample point to the prevailing wind direction.

In practice, the wind direction would not be maintained steady; therefore, it is difficult to sample close to the plume centreline (which coincides with wind direction). For this reason the weight factor was devised. With a handheld GPS and a knowledge about wind direction, an emergency responder can have a good sense of placing the portable sensors. The sample points must be clear from any obstacles (e.g. buildings, fences, and trees, etc).

DETAIL DESCRIPTION OF THE PROCESS

Consider the dispersion of an accidental release of a chemical (Figure 3). The impact of the plume on each sensor differs due to its travel time and/or wind shift with respect to the position of each sensor. Therefore, some sensors may not begin reading a concentration until later in an event while other sensors may be impacted early on. Some sensors may go out of commission if they become saturated or reach a maximum upper limit of their reading range.

Assume the following hypothetical situation for the first few moments of a release. The inner area represent the lowest level we will model, but not necessary the lowest level that can be measured. The outer area represents the area where some level of the cloud may be monitored at a range below the level of concern. Assume sensors 1, 2, 3, 10 are fixed and sensors 4, 5, 6, 7, 8, 9 are portable ones.

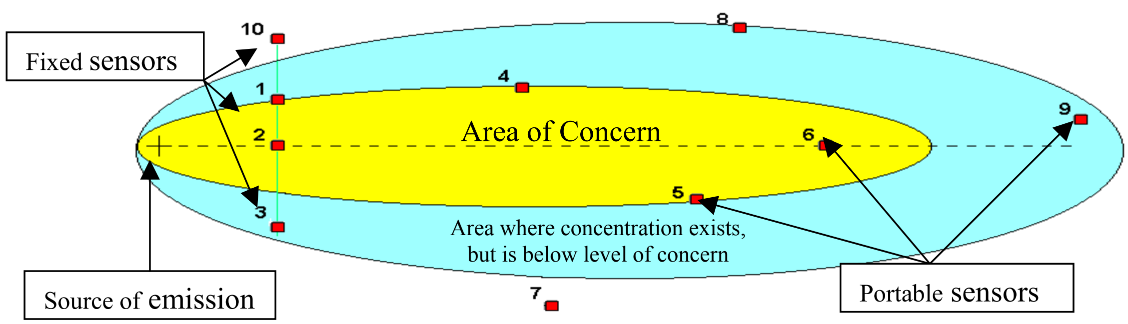


Figure 3. Plume/sensor impact

Fixed sensor along with deployed portable sensors start transmitting the following information to the back calculation module.

Some gas monitoring sensors take 50 seconds to start detecting the chemical (sensor lag), this lag is factored in time convergence loop. The starting time of an event is a necessary piece of information for backcalculation algorithm. Backcalculation can accept either an starting time of event from the user or can the starting time based on the position of the closest impacted sensor to the source and the wind speed.

Table 1. Sensor data communication to backcalculation

Sensor ID	Location	Concentration (ppm)	Time	Saturation (ppm)
1	X1, Y1, Z1	C1	t1	C1limit
2	X2, Y2, Z2	C2	t2	C2limit
.	…	…	…	…
N	Xn, Yn, Zn	Cn	tn	Cnlimit

Suppose the event starts at time T. Sensors 1 and 2 pick up the gas concentration T4 minute after the release. After T7 minute into the release, sensor 2 reaches its maximum concentration range. At T9 minutes, plume impacts sensors 3 and 4.

At T7 minutes the operator starts the program and sites the release. Sensors 1 and 2 are available at this time. Sensor 2 has reached it maximum concentration. However, valid readings are available from sensor 1 to time T7 minutes and for sensor 2 up to T6.

The sensor poling frequency is 1hz, but a one minute running average is used for the backcalculation which is consistent with one minute interval puff releases for the dispersion model. The release rate calculated for the T4 interval is the assumed for the T1 through T4 where there was no actual measurement.

At T + 10 minutes the program runs again. This time, it has built an array of data similar to table 2. For time T through T7 seconds, the sensors 1 and 2 are used for rate calculation. For time T7 to T8, sensor 1 is used. For time T8 through T9, sensors 1 and 4 are used for rate calculations.

The process continues as more sensors with valid reading participate. The following table is a sample (based on three sensors) of how the release rate is calculated and sorted in the ascending order of time before it is passed to the dispersion modeling for the calculation of the plume impact.

For the case when the responder gets to the scene of accident and the event has been in progress for a while, the starting time of calculation would be different than the starting time of the event. As might be expected, the release rate history from the actual time of accident to the calculated starting time based on the deployed sensors is lost.

COMPARISON WITH FIELD DATA

The Desert Tortoise sensors data is used to test the accuracy of backcalculation model. A series of four large-scale (15–60 m^3) pressurized ammonia spill test were conducted by the Lawrence Livermore National Laboratory, Goldwire et al. (1983). The tests, called the Desert Tortoise series, were conducted at Frenchman Flat in Nevada during August and September of 1983.

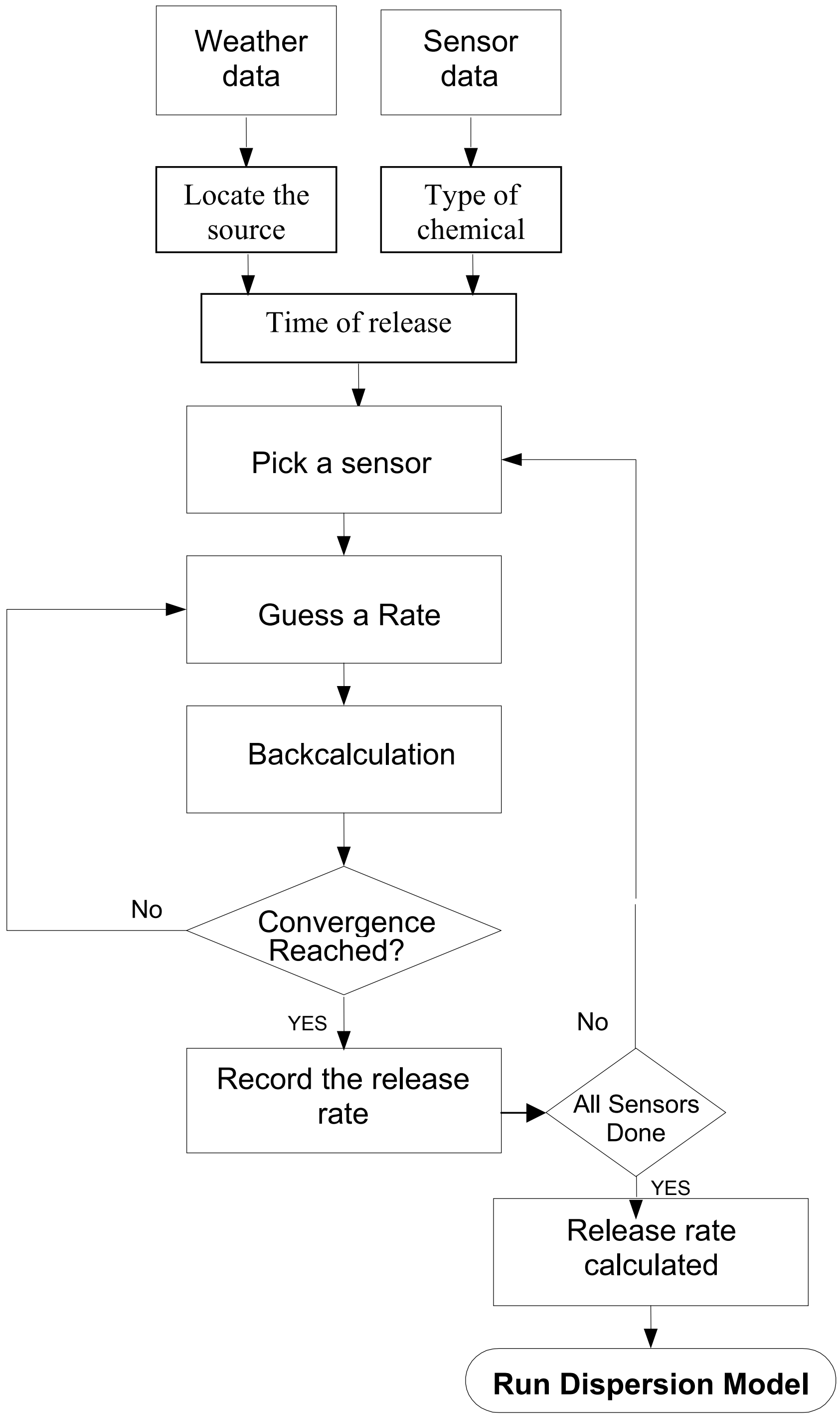


Figure 4. Algorithm for backcalculation

Table 2. Typical sensor/plume interaction behaviour

Time + (seconds)	Sensor 1	Sensor 2	Sensor 3	Sensor 4
T1	Fill in from 60	Fill in from 60	No reading	No reading
T2	Assumed Conc.	Assumed Conc.	No reading	No reading
T3	Assumed Conc.	Assumed Conc.	No reading	No reading
T4	First actual reading	First actual reading	No reading	No reading
T5	Valid reading	Valid reading	No reading	No reading
T6	Valid reading	Valid reading	No reading	No reading
T7	Valid reading	Maximum reading obtained	No reading	No reading
T8	Valid reading	Not used	No reading	Assumed Conc.
T9	Valid reading	Not used	Below range reading	Valid reading

Table 3. Release rate set up for dispersion calculation

Time (min) \ Sensor #	1	2	3
T1	R11	R12	R13
T2	R21	R22	R23
T3	R31	R32	R33
T4	R41	R42	R43

$\Longrightarrow$

Time (min)	Average release rate based on three sensors (lb/min)
T1	R1
T2	R2
T3	R3
T4	R4

Sensors were placed at 100 and 800 meters away from the source. At 100 meters, the sensors were placed 15 meters apart. At 800 meters, they were 100 meters apart. The seven sensors at 100 meters were numbered as G02 through G08, with G05 assumed to be on the centreline. There were five sensors at 800 meters (G20 through G24) at 800 meters, with G22 being on the centreline.

The following table shows the characteristics of the two tests chosen for this study:

Table 4. Field data for Desert Tortoise 1 and 4 trials

Test number	Desert Tortoise 1 (DT1)	Desert Tortoise 4 (DT4)
Chemical	Ammonia	Ammonia
Phase of release	Liquid	Liquid
Duration	126 (s)	381 (s)
Orifice diameter	3.19 (in)	3.72 (in)
Spill amount	10200 (kg)	41100 (kg)
Spill rate	4860 (kg/min)	6480 (kg/min)
Average wind speed @ 2 m	7.42 (m/s)	4.51 (m/s)
Average wind direction	223	229
Average directional variability	5.73	5.02
Relative humidity	13%	21%
Surface roughness	3 mm	3mm
Temperature	29 (c)	32 (c)
Stability class	D	E

The actual field sensor measurements were averaged over one minute intervals and the results were used to compare the model prediction against sensor readings. The one minute averaged concentration is used as input to the backcalculation. A sample of the process for DT4 for sensors at 100 and 800 meters is shown in Figures 13 and 14.

Two studies were performed:

a) Knowing the rate, compare the prediction of the model concentration-time profile with that measured by the sensors. This will show the goodness of the dispersion model utilized.
b) Knowing the measured concentration-time profile of each sensor, calculate the release rate. This will test the goodness of the backcalculation algorithm and the benefit of the weighing factors.

A) RELEASE RATE IS KNOWN

Liquid ammonia flashes to vapor, and aerosol upon release to the atmosphere. A fraction of the aerosol rains out to form a pool. The combined streams of vapor, aerosol, and emission from pool are dispersed by atmospheric flow. This process continues until the tank content is emptied or tank release and pool emission is mitigated.

The following graphs (Figures 5 & 6) show the calculated source for atmospheric dispersion. Comparing the total rate for dispersion with ammonia liquid discharge rate of 4860 kg/min for DT1 and 6480 kg/min for DT4 represent the effect of rainout. The initial cloud dilution due to jetting is not taken into consideration.

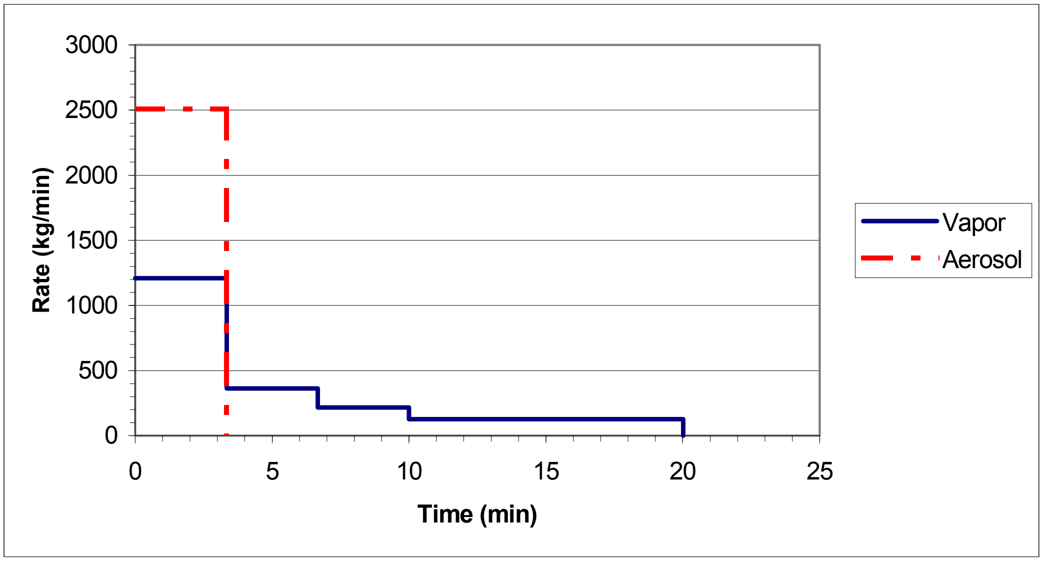


Figure 5. Source term for dispersion-DT1 field trial

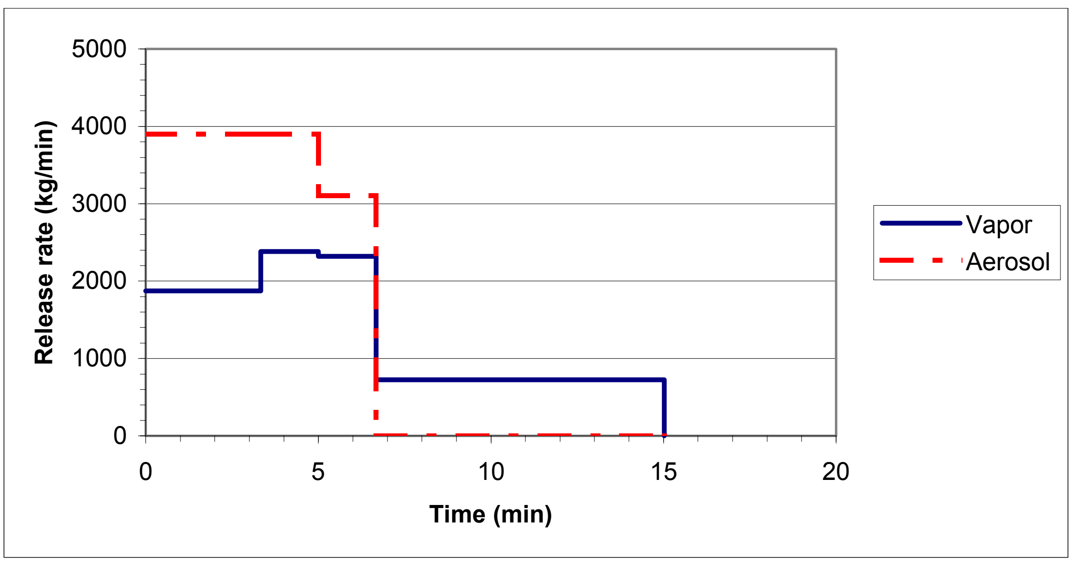


Figure 6. Source term for dispersion-DT4 field trial

The result of the dispersion model is overlaid against the sensor data for DT1. Figure 7 & 8 show the comparison at 100 and 800 meters. The assessment is performed for the centreline values only. There is no resemblance between the two plots at 100 meters downwind. The duration and the time of the maximum concentration is different. The percent error between the peak sensor concentration and that of the model is 34%.

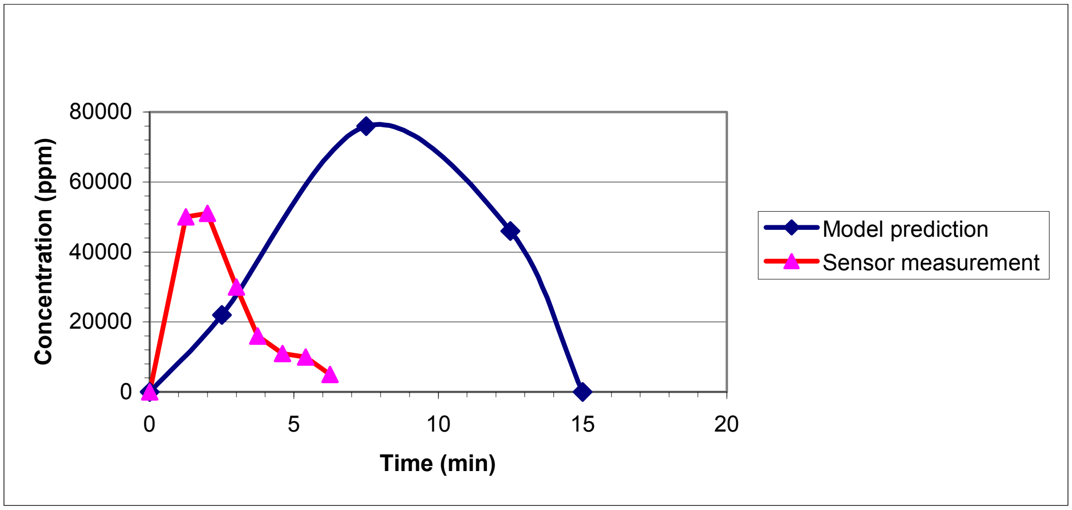


Figure 7. Predicted vs. measured centreline concentration profile at 100 meters-DT1

The comparison of the model with sensor measurement looks very good at the distance of 800 meters. The concentration profiles seem reasonably matching in their trend and occurrence of the maximum concentration. There is a 30% error between the model predicted peak concentration and the sensor measured values.

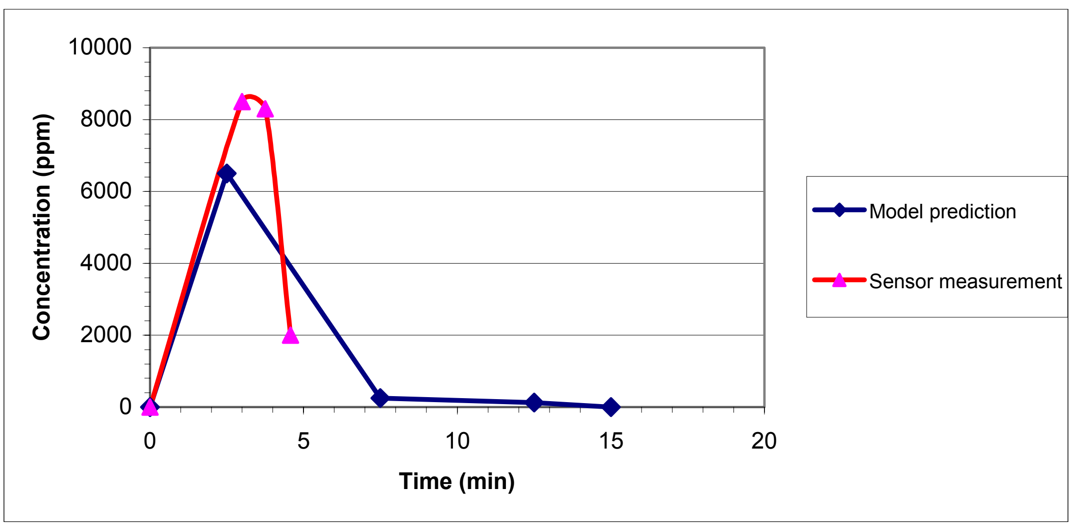


Figure 8. Predicted vs. measured centreline concentration profile at 800 meters-DT1

Comparison of the concentration profile predicted by the model against the measured values at 100 meters for DT4 is interesting. The slope of the concentration plots after the cloud arrival is identical, and the peak concentrations occur at the same time. However, there is 65% error between the measured and model peak concentration.

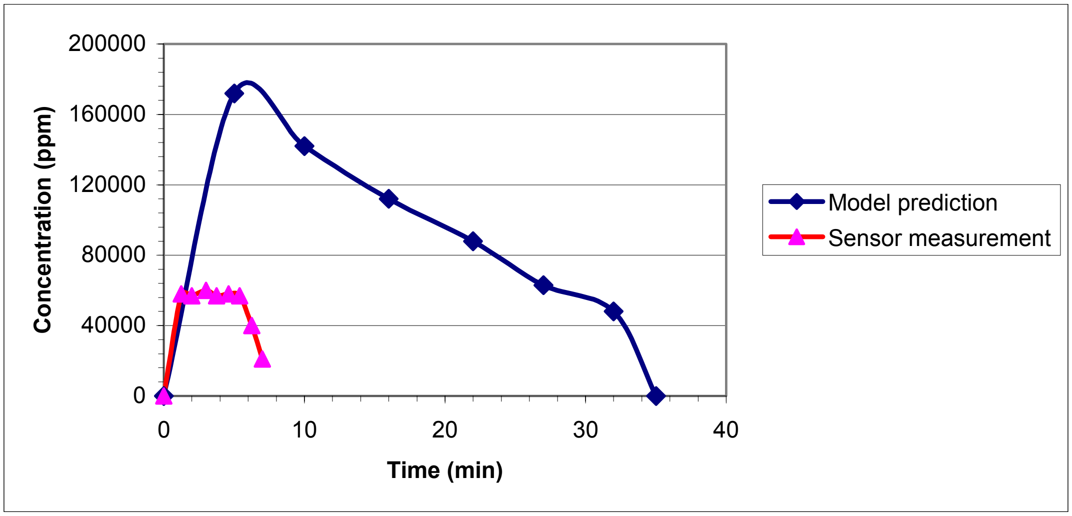


Figure 9. Predicted vs. measured centreline concentration profile at 100 meters-DT4

The concentration profiles seem reasonably matching in their trend and occurrence of the maximum concentration at 800 meters for DT4 experiment. The maximum concentration occurs at almost the same time and the percent error between the field data and model simulation is roughly 58%.

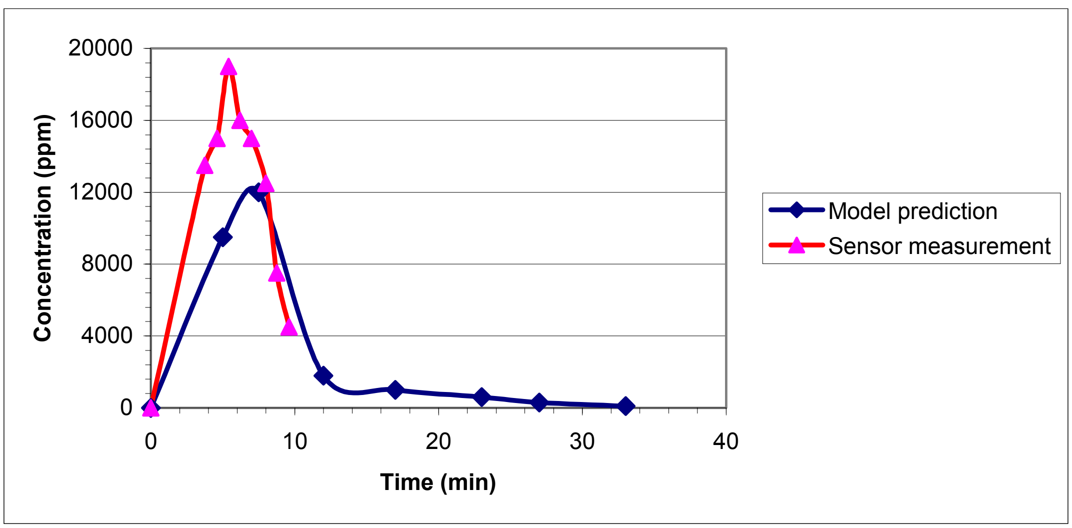


Figure 10. Predicted vs. measured centreline concentration profile at 800 meters-DT4

The model prediction is higher than the measured concentration at 100 meters for both DT1 and DT4 and this trend it reversed at 800 meters. This can be due to the jetting effect which causes the trial cloud to dilute faster than the model up to the point that jetting effect is dominant. At the end of the jetting zone the modelled cloud dilute faster due to its higher density than the trial cloud. The persistence of the jetting effect up to the 100 meters downwind was observed by LLNL personnel during the field trials.

B) RELEASE RATE IS UNKNOWN

The two field trials DT1 and DT4 were selected to gauge the behavior of the backcalculation model. The one minute average concentrations for all impacted sensors (Figures 13–14) along with their time of measurements were input to the backcalculation algorithm. The one minute averaged sensor data produced 16 data points for DT1 and 56 data points for DT4. These data were input to the backcalculation module for release rate estimation.

Duration of liquid release from the tank was 2 and 6 minutes for DT1 and DT4 respectively. The emission from the pool continued for several more minutes after the valve was closed. The total rate for dispersion was 3700 kg/min for DT1 and 5800 kg/min for DT4 trials.

To simulate the field trials with backcalculation algorithm the molecular weight of ammonia was modified. This adjustment was necessary to obtain identical initial density between the model and field trials. Figures 11–12, represent release rate estimates for both field trials. The symbols represent the estimated values, and the horizontal line is the plot of actual rate.

Two distinct regions are observed for the release estimation curves. An almost steady rate during the discharge of liquid ammonia from tank and a sharp drop in rate after the valve is closed and emission is solely from the shrinking pool. According to sensors data the pool content was depleted in just a few minutes. The maximum percent error between the predicted and measured values for the period where the tank is contributing is 20% for DT1 and 90% for DT4.

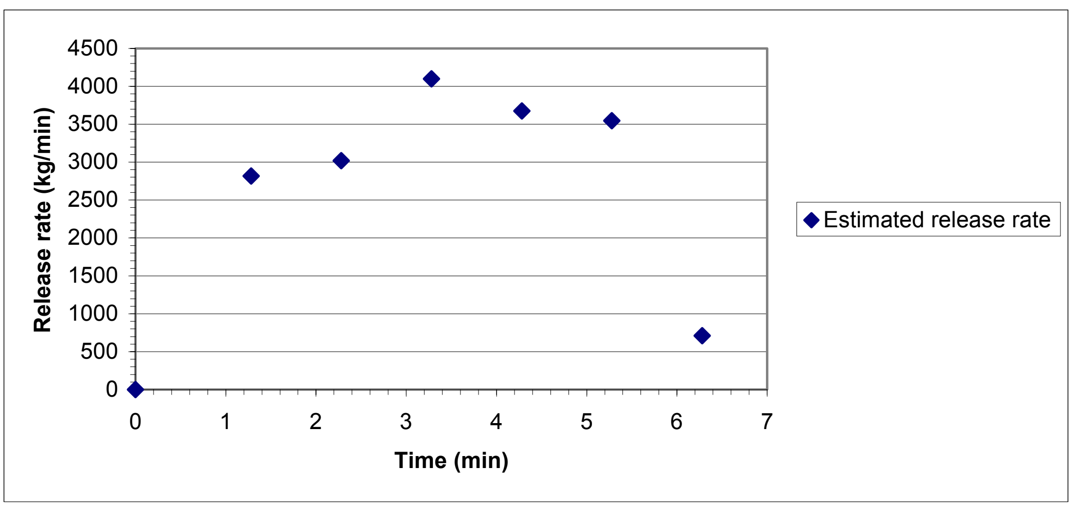


Figure 11. Model prediction against the actual release rate-DT1

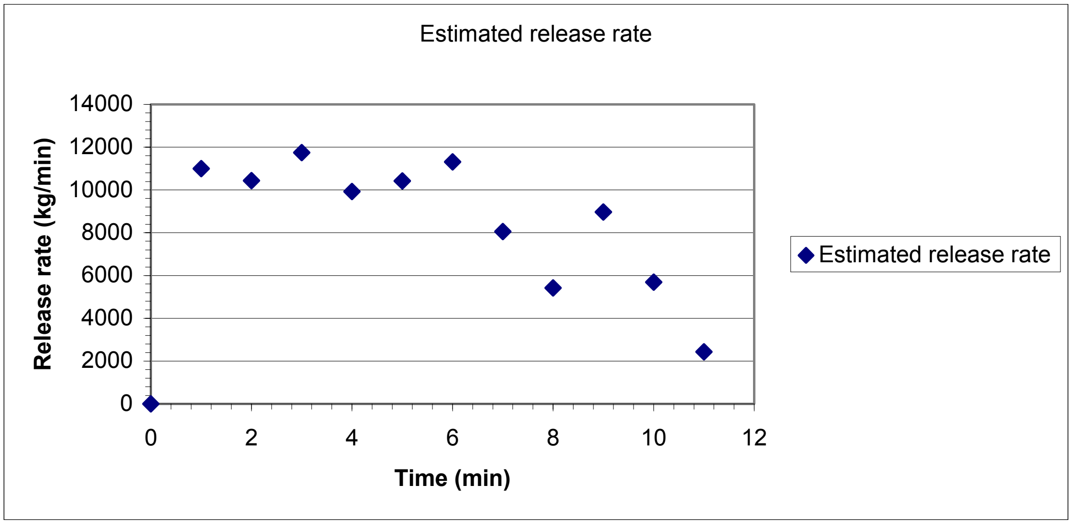


Figure 12. Model prediction against the actual release rate-DT4

One possible explanation for the magnitude of error is ignoring of the initial dilution at the source by the backcalculation algorithm. The liquid ammonia jetting out of vessel flashes to vapor and aerosol and is diluted by air entrainment. The effect of jetting is more pronounce in DT4 than DT1 due to higher release rate and temperature. The backcalculation algorithm assumes pure vapor and does not take into consideration the initial dilution of the cloud. This leads to overestimation of rate for both cases with DT4 having the poorest performance due to its higher dilution effect. This observation is an important piece of information for further enhancement to the backcalculation module.

The break up of error does definitely contain the plume meandering effect as well, although this is very hard to quantify in this study due to lack of information. However a test of sensitivity of the model to wind meander was performed. The sensors position was fixed and the mean wind direction was changed within fifteen degrees. The analysis of the simulation results indicated a model sensitivity of between 5% to 10% for a fifteen-degree plume meander.

CONCLUSION

This study was set out to accomplish two goals: (1) to develop a concept to obtain certain key parameters needed during an emergency response, and (2) using the field data to validate the concept. The novel idea of locating the source using weather data and impacted sensors location was introduced. The construction of several reverse corridors, which are based on the wind direction and stability, narrows the possible location of the chemical spill.

A backcalculation algorithm was presented which takes into account the spill location, time of release, and measured concentration to estimate the release rate. Two field data from Desert Tortoise (DT) series of tests, DT1 and DT4, were selected for validation purposes. It is recognized that there are many uncertainties involved in the rate estimation process: (1) averaging time of sensors, meteorological input, and model; (2) position of sensors with respect to the mean wind direction (cloud meandering); (3) near source phenomena (e.g. aerosolization) and initial dilution due to jetting effect.

Sensitivity analysis of the result indicates that 2, and 3 plays the most important part on the overall performance of the model. To reduce or dampen the effect of cloud meandering, several sensors should be deployed at any downwind distance where they are placed across the wind.

The near source phenomena can be included in the rate estimation model if two different parameters are used for trial and error calculations: (1) for high momentum releases from a tank and/or pipe the parameter of interest would be the size of the hole; (2) for other sources (e.g. pool only) the parameter of interest would be the emission rate. Following the above guidelines and using a good dispersion model, which can take into account the real-time measurements, can provide a good tool for rate estimation of chemical spills.

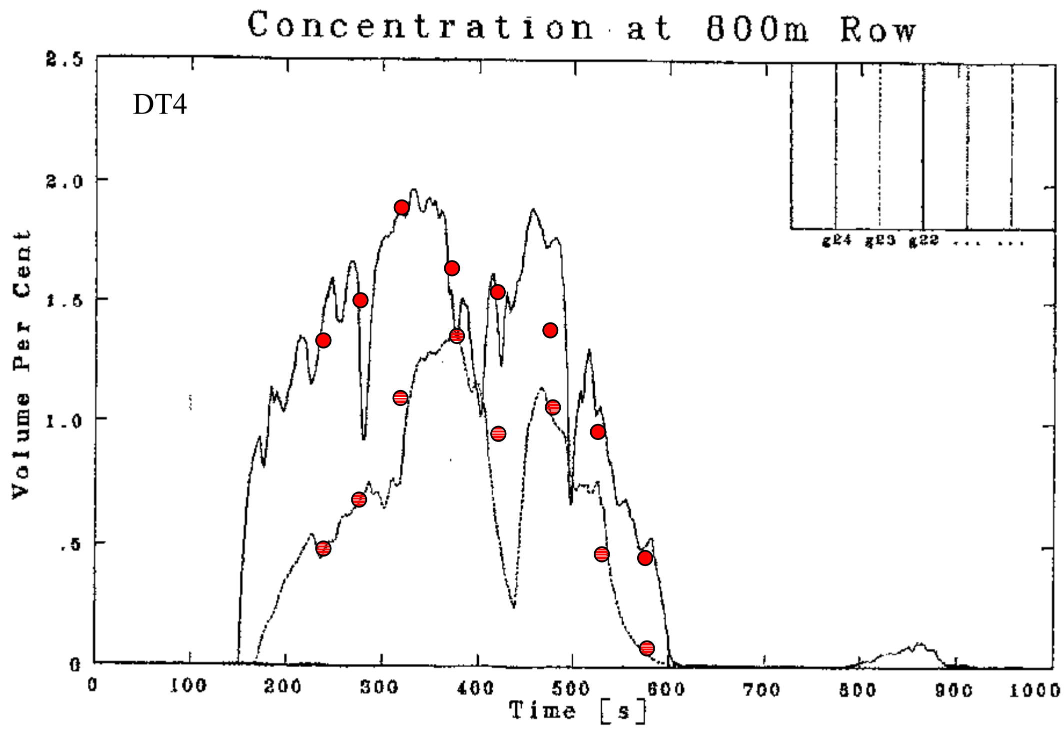
REFERENCES

M. Lehning, D. P. Y. Chang, D. R. Shonnard, and R. L. Bell, "An Inversion Algorithm for Determining Area-Source Emissions from Downwind Concentration Measurements", J. Air & Waste Manage. Assoc, 44: 1204–1213 (1994)

S. D. Piccot, S. S. Masemore, E. S. Ringler, S. Srinivasan, D. A. Kirchgessner, W. F. Herget, "Validation of a Method for Estimating Pollution Emission Rates From Area Sources Using Open-Path FTIR Spectroscopy and dispersion modeling techniques", J. Air & Waste Manage. Assoc, 44, 271, 279 (1994)

S. D. Piccot, S. S. Masemore, W.L. Bevan, E. S. Ringler, D. Bruce Harris "Field Assessment of a New Method for Estimating Emission Rates from Volume Sources Using Open-Path FTRI Spectroscopy", J. Air & Waste Manage. Assoc, 46, 159–171, (1996)

H. C. Goldwire, G. McRae, G. W.Johnson, D. L. Hipple, R. P. Koopman, J. W. McClure, L. K. Morris, R. T.Cederwall, "Desert Tortoise Seris Data Report, Pressurized Ammonia Spills", Lawrence Livermore National Laboratory, December 1985.



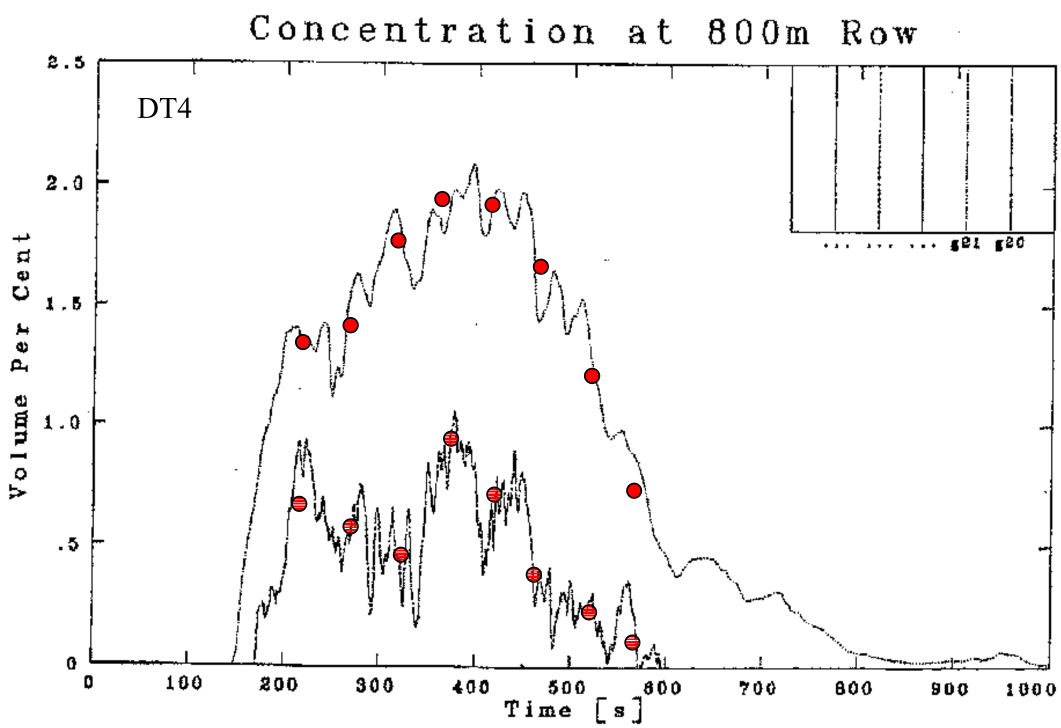


Figure 13. Sensor measurement at 800 meters-DT4 trial

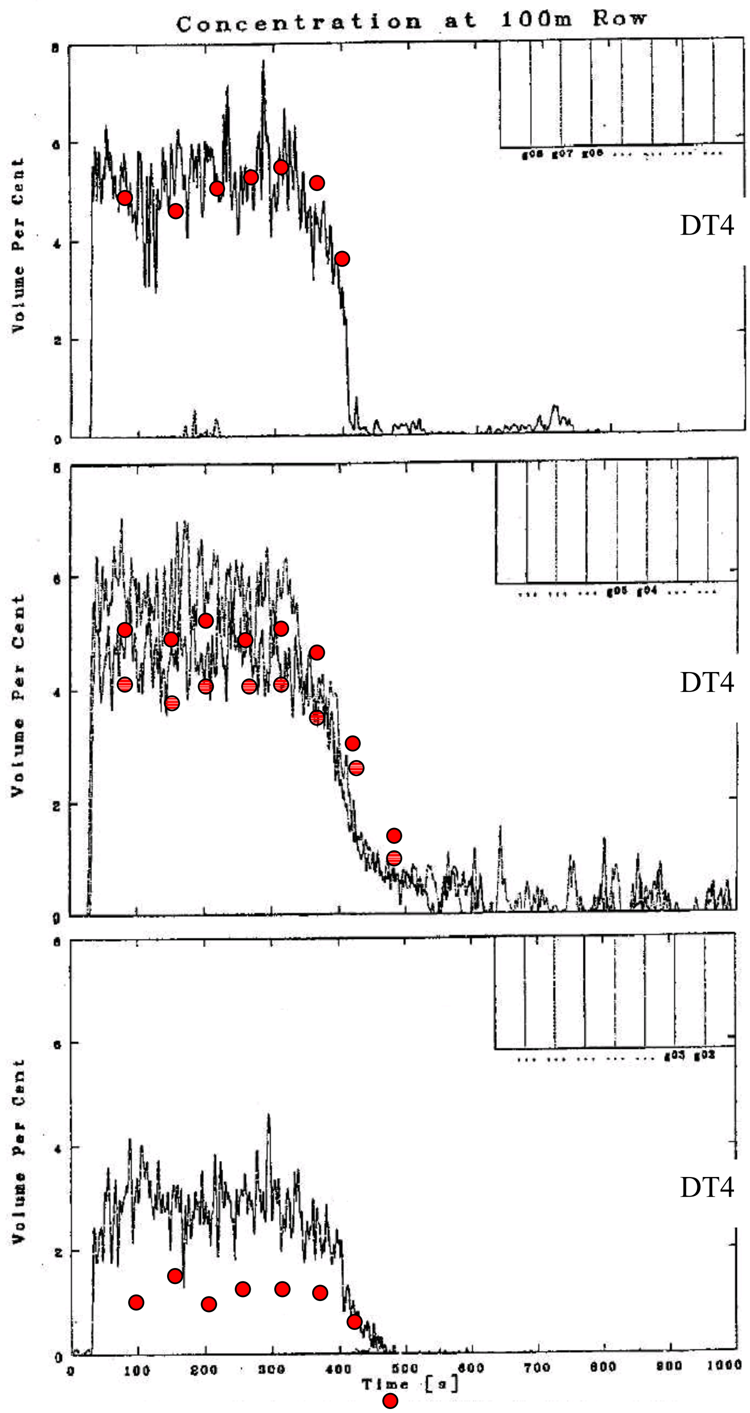


Figure 14. Sensor measurement at 800 meters-DT4 trial