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Controlled Experiments for Dense Gas Diffusion - Experimental Design and Execution, Model Comparison

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ABSTRACT

An experimental baseline CO_2 release experiment at the DOE Spill Test Facility on the Nevada Test Site in Southern Nevada is described. This experiment was unique in its use of CO_2 as a surrogate gas representative of a variety of specific chemicals. Introductory discussion places the experiment in historical perspective. CO_2 was selected as a surrogate gas to provide a data base suitable for evaluation of model scenarios involving a variety of specific dense gases. Releases were conducted under baseline conditions including a simulated "evaporating pool" release over flat unobstructed terrain. The experiment design and setup are described, including design rationale and quality assurance methods employed. Design conditions included moderately low wind speed, stable atmospheric conditions. Four releases were performed, two of which were during near-neutral conditions and two during slightly stable conditions. Resulting experimental data are summarized. These include CO_2 cloud characteristics measured at 40 m downwind from the release point. Experiment success and effectiveness is discussed in terms of mass balance analyses. For Tests 1, 3, and 4 the measured mass accounted for at least 90% of the released mass. Measured values for Test 2 accounted for only 60% of the released mass. Data usefulness is examined through a preliminary comparison of experimental results with simulations performed using the SLAB and DEGADIS dense gas models.

This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

INTRODUCTION

Several large-scale experimental releases of denser-than-air gases have been performed in recent years. These programs were designed and carried out primarily to quantify potential hazards of specific chemicals such as liquefied natural gas (LNG), ammonia (NH_3), hydrogen fluoride (HF), and nitrogen tetroxide (N_2O_4). Historical dense gas dispersion field experiments are described in a review by Havens (1992). They were performed over a wide range of release rates, initial densities, and meteorological conditions. They provided data sufficient for a reasonable test of models, were representative of "dense gas" effects, and were sufficiently large scale to test the important attributes that models required for practical prediction of dense gas dispersion. However, there are important limitations on the utility of the current field test data base:

1. The data (and their use for model validation) are limited to description of dispersion over level, unobstructed terrain.

2. There are important uncertainties in the specification of the source conditions, including the release areas and rates, release momentum effects, and specification of the properties of the aerosol and the density vs. concentration relation.

3. The data are of questionable value for testing the submodel descriptions of the effect of density stratification (dense gas effect) on dispersion.

4. There are very few data for releases under stable meteorological conditions; such conditions are a principal concern in accident consequence assessment.

The Clean Air Act (CAA) Amendments of 1990 direct the Environmental Protection Agency (EPA) to coordinate an experimental and analytical research program at the Hazardous Chemicals Spill Test Facility (STF) operated by the Department of Energy (DOE) at the Nevada Test Site (NTS) near Mercury, Nevada. Although the STF is ideally suited for large-scale testing of flammability as well as toxicity hazards, CAA provisions currently are assumed to apply primarily to toxic dense gas hazards, which are typically associated with much lower concentrations than those associated with flammability hazards. The STF, previously the Liquefied

Gaseous Fuels Spill Test Facility (LGFSTF), has been the site of several test programs involving releases of HF (Goldfish series), N_2O_4 (Eagle Series) and NH_3 (Desert Tortoise Series).

The Chemical Hazards of Atmospheric Releases Research (CHARR) Steering Committee was formed to recommend and prioritize research tasks to be carried out at the STF. These Research tasks should address limitations of existing field test data, discussed above, and meet CAA requirements for research at the STF.

The CHARR Steering Committee recommended a DOE/EPA long-term research program at STF that targets data limitations in the areas of flow over uneven and obstructed terrain, specification of source conditions, dense gas effects, and stable meteorological conditions. The CHARR steering committee also recommended that data should be acquired using surrogate gases with appropriate physical characteristics, principally density, that can be more generally applied than data acquired using very specific gases, and that a series of baseline experiments should be conducted with a surrogate gas and with idealized conditions of flat, level terrain without obstacles. Baseline experiments would provide data sets useful to the world community for evaluating models and would provide a comparative basis for evaluating the effects on dispersion of non-ideal conditions such as uneven terrain with obstructions.

This paper describes the design and execution of a baseline experiment at STF. The experiment differs from previous test releases in its use of carbon dioxide (CO_2) as a surrogate gas with density characteristics that make it applicable to a wide variety of dense gases. Subsequent sections of this paper describe the experiment design, setup, execution, and some preliminary modeling results using data from CO_2 releases over flat unobstructed terrain. The work was funded by EPA and conducted by the Desert Research Institute (DRI) and Western Research Institute (WRI).

PURPOSE AND GOALS OF EXPERIMENT

The experiment reported here represents the first use of the STF for highly controlled releases of heavy gases under stable to neutral atmospheric conditions. The purpose of the experiment was to develop a data set for use in characterizing the source term component of mathematical models, that is, to capture sufficient data close to the emission point to allow for computation of a mass balance. CO_2 was selected as the gas to be used because:

- CO_2 possesses dispersion properties similar to those of other heavy gases.
- Large quantities of CO_2 can be obtained for reasonable costs and can be safely transported and handled.

- CO_2 measurement methods are well established and can be efficiently implemented.

- High concentrations of CO_2 present a hazard to workers in industries where it is used.

- CO_2 is of relatively low toxicity, inert and chemically stable.

Specific goals of the experiment were to:

- Design and test a release, monitoring, and data management system for 0.1 to 1 ton releases of CO_2.

- Acquire a data base of specified accuracy, precision, and validity suitable for data analysis and modeling of CO_2 releases in flat terrain under stable atmospheric conditions.

- Establish a mass balance between the amount of CO_2 released and measured downwind fluxes of CO_2 through the atmosphere.

EXPERIMENT DESIGN AND PREPARATION

Site Description

The STF is located on the dry lake bed in Frenchman Flat on the NTS, approximately 100 km northwest of Las Vegas. The dry lake provides a smooth surface with a roughness length of about 0.2 mm. The site is at an elevation of 939 m above mean sea level (MSL) and extends 1 km to the north and west, 2 km to the south, and 3 km to the east of the STF. Terrain slopes gradually downward toward the dry lake bed, which is in the southeast quadrant of the valley. The surrounding mountains are located 8 to 20 km from the dry lake at elevations ranging from 1500 to 2000 m MSL.

The STF lies near the southern edge of the Great Basin. Its climatology is similar to that of the middle elevations of the southwestern desert area of the US. Precipitation occurs during winter when northern storm tracks move southward and in summer when moisture from the south causes thunderstorms (monsoons). Annual precipitation totals about 100 mm (4 in). During summer months, southwest winds predominate the hours from late morning to sundown with the wind direction being in the SSW to WSW sector more than 60% of the time. Nocturnal and early morning

winds are influenced by the surrounding terrain and become light and variable during the hours after sunset. During winter months, southwest winds still occur from late morning to sundown but with frequencies in the 30 to 40% range. The winter storm pattern results in northerly winds about half the time. Terrain continues to control the nocturnal winds during the winter when storms are not influencing the area.

The NTS is a controlled-access area operated by the U.S. Department of Energy (DOE). As such, access by the public is restricted to a minimum distance of 25 km from the STF. On the NTS, procedures are in place to restrict access near the STF during experiments. The nearest major population center is Las Vegas.

The STF includes a command and operations center, fast-response data acquisition systems, a wind tunnel, and a tank farm capable of automated high-pressure releases. It is available for government, commercial, and academic organizations to conduct tests on the release and mitigation of hazardous materials and has been used in the past to understand phenomena related to large-scale spills of liquefied petroleum, chlorine, hydrofluoric acid, and a number of other chemicals.

Experiment Design

To accomplish the objectives of the experiment, it was necessary to choose a period during which atmospheric conditions were stable while maintaining a consistent enough wind direction to transport the released gas towards a fixed array of sensors and samplers. The permit issued by the state of Nevada allowing the release of hazardous materials at the STF also required that any releases be done before sunset.

Meteorological Requirements

Data collected onsite prior to the experiment and near the site from NTS weather stations showed that afternoon and evening winds generally follow a regular pattern during summer. Southwesterly winds (wind directions centered around 225°) typically set up by 1200 PST, reaching maximum speeds of 4 to 8 m/s during midafternoon. At 30 to 60 minutes before sunset, the speed typically begins to decrease abruptly, reaching speeds near 1 m/s within a 50 to 80 minute period. The southwesterly wind direction persists as the speed decreases for as much as 1 hr later. Eventually, the winds become light and variable. This decrease in wind speed coincides with the change in atmospheric stability from slightly unstable or neutral to stable as the surface heat flux reverses from positive to negative.

The regularity of the decrease in wind speed at sunset and the persistence of southwesterly winds during this decrease established a target release window. The deciding factor for the release time was the decrease in wind speed to 3 m/s in conjunction with the consistent direction toward the sampling array. A release

duration of 5 minutes was selected to maintain nearly stationary atmospheric conditions during the release period.

Release Method for CO_2 and SF_6

Consideration of available models of dense gas dispersion placed the following requirements on the release system for CO_2: 1) the gas had to be released with low momentum (i.e., not as a jet), 2) the release rate had to be constant in the range of 1 to 2 kg/s, and 3) the released material had to be in the gas phase and nearly isothermal. These requirements were met by designing a release method with sufficient residence time between depressurization (release from the tank) and release to the atmosphere for the CO_2 to completely vaporize and reach ambient temperature. In addition to CO_2, a tracer gas, sulfur hexafluoride (SF_6), was released at the same time at a fixed rate as a secondary test to evaluate the use of tracers in characterizing the dispersion of dense gases.

CO_2 was obtained in a refrigerated six-ton tank at 2 °F (-17 °C) and 2103 Pa (305 psig), (Airco Gases, City of Industry). The CO_2 passed through a 15 kW heater and into an insulated 1012 ft^3 (28.7 m^3) surge tank at a temperature of ~85 °F (29 °C). A 4 in (0.1 m) diameter, 60 m long pipe connected the surge tank to a 6 in (0.2 m) control valve placed near the release point. The control valve was activated remotely to start and end a release. A baffled discharge chamber (BDC) was connected to the outlet of the control valve to provide an initial dampening of the flow discharge. An 18 in (0.5 m) diameter flexible hose was connected from the BDC to the bottom of a 1 m^3 box that was buried flush with the ground. The 1 m^3 box had additional baffles to further reduce the momentum of the released gas. Gaseous CO_2 was released from the top of the 1 m^3 box at ground level. The ground within 3 m of the discharge box was restored to approximately the same surface roughness as the terrain of the dry lake bed.

SF_6 was introduced to the CO_2 gas stream between the control valve and the BDC with the BDC providing a homogeneous mixing of CO_2 and SF_6. A Tylan Model 260 mass flow controller maintained a constant flow rate of 0.9 g/s of SF_6 from a 10-lb cylinder from Scott Environmental. The release of SF_6 coincided with the release of CO_2.

Thermocouples in the release line ahead of the control valve, behind the control valve, in the box, and in the area immediately surrounding the release point quantified temperatures at these locations during each release. Temperature and pressure were also measured inside the surge tank. The CO_2 flow rate was calculated from these quantities. The SF_6 flow rate was measured and recorded from the mass flow controller.

Measurement Methods

CO_2 concentrations were measured by 1) collection of air samples in Tedlar bags and subsequent analysis (Horiba CO_2 analyzer, model 355098-2) and 2) real-time sensors (Nova Analytical Systems, Inc., micro CO_2 sensor, model DEO, 1% and 10% ranges). Bag samples were used for mass balance determinations for the CO_2 releases. The continuous sensors were used to detect the presence of the CO_2 cloud and its instantaneous concentrations. Bag sampling was controlled remotely and was coordinated with the release time. Air samples were collected from the time of release to the time that all CO_2 had passed the sensor array. Flow rates for sample collection were set to collect about 12 l of air. Data from the continuous sensors were collected every 2 seconds by the STF data acquisition system (DAS).

Bag samples were analyzed for SF_6 concentrations at the same time that CO_2 was measured. The SF_6 measurement system consisted of a Varian gas chromatograph with electron capture detector, a Hewlett-Packard integrator (model 3390A), and a reactor sampling train to remove oxygen while retaining SF_6. A Campbell Scientific data logger, model CR-10, controlled sample injection and data collection.

Bag samplers and sensors were deployed downwind of the release point along a line perpendicular to the prevailing wind direction of 225°. Preliminary modeling for the anticipated release rate and meteorological conditions estimated that concentrations along a line 40 m from the release point (the 40 m arc) would be within range of the continuous sensors. Accounting for variations in wind direction and plume spread, bag samples were collected along the center line, at 3.5° on either side of the centerline, and at several locations 5° to 6° apart, outside the arc. The outside samples were 20° from the centerline. In distances from the centerline, these locations were 0 m,±2.5 m,±6 m,±10 m, and ±15 m. Preliminary modeling results indicated the best heights for defining the cloud mass to be 0.10 m, 0.33 m, 0.67 m, 1.00 m, and 1.40 m above the ground. A total of 43 bag samplers was deployed along the 40 m arc while 2 bag samplers were placed upwind of the release point to collect background samples. Also placed along the 40 m arc were 37 continuous sensors, with 30 sensors collocated with bag samplers. The remaining 7 continuous sensors were placed along the 40 m arc at locations outside the anticipated cloud to check for possibly wider and higher clouds at ±30 m from the centerline, ±22 m from the centerline, and on the centerline at 2 m above ground. The 10% sensors were located at the lower two height levels within the inner five towers.

Wind speed and direction were measured (R.M. Young Wind Monitor-RE, model 5701) along the release centerline 5 m upwind of the 40-m arc at a height of 0.5 m above the ground. Data from these sensors were collected by the STF DAS with other continuous sensor data for use in subsequent mass balance calculations.

A sonic anemometer/thermometer (Applied Technology, Inc., model SAT-211/3K) was placed 40 m upwind of the release point to make direct measurements of heat and momentum fluxes. Wind speed component and temperature data were

collected at a rate of 10 Hz and averages, variances, and covariances were computed in 1-minute blocks by a dedicated DAS.

Meteorological measurements were also made at 8 levels on a 24-m tower located 112 m south-southwest of the release point. Measurements included wind speed and direction, temperature, relative humidity, solar radiation, net radiation, and soil temperature. The intent of the tower has been to provide a climatology of the atmosphere near the ground in the vicinity of the STF. Data were collected as continuous 5-minute averages with a dedicated DAS and as instantaneous values during the release period with the STF DAS.

Data Management

Continuous data from the real-time CO_2 sensors, the wind speed and direction sensor at the array, the meteorological tower, the temperature and flow sensors for the releases, and the control sensors, were collected by the STF DAS. Data were collected every 2 seconds, with meteorological data updated every 10 seconds, for approximately an hour before each release to 10 minutes after each release. Raw data from the STF DAS, including header information, were transferred in comma delimited format to diskettes for verification and analysis on IBM-compatible personal computers. Following the experiment, continuous CO_2 data were corrected for calibration drifts and for background readings. Continuous meteorological data were entered into Excel spread sheets for editing. Because of intermittent data collection problems, some meteorological data required deletion.

CO_2 and SF_6 concentrations from bag samples were saved directly in dBase IV data bases. In addition to concentrations, the data bases contained bag ID, sample location, and replicate analyses. After each test, preliminary data were plotted to check that the systems were operating. Following the experiment, calibration data provided minor corrections to the integrated CO_2 and SF_6 concentrations.

Quality Assurance

Quality assurance (QA) procedures for this experiment were defined in a QA Plan (DRI/WRI, 1993) to ensure that data of known and acceptable quality were collected. Written protocols defined operating procedures for each major measurement system. Independent field audits were performed to verify the CO_2 and SF_6 measurement systems. Meteorological instruments were purchased new and calibrated just prior to the experiment.

Data accuracy was determined by challenging each measurement system with blind audit standards prepared and administered by an independent QA Officer. Data precision was determined using replicate measurements.

To assess data accuracy, blind samples of SF_6 and CO_2 in Tedlar bags were introduced to each measurement system. The samples were made from standards of

SF_6 and CO_2 purchased from Scott-Marrin, Inc. (Riverside, CA), traceable to the National Institute of Standards and Technology (NIST). Standards were diluted in each audit bag with ultra high purity nitrogen or air, using calibrated mass flow controllers. Nominal concentrations of SF_6 were verified by analysis using gas chromatography with an electron capture detector (GC/ECD), using 60 m x 0.32 mm i.d. DB-1 capillary column (J&W Scientific Co.). Chromatographic temperatures were programmed: for -60°C for 3 min and then raised 6°C/min to 30°C. CO_2 standards were verified using a Nickel based methanizer, coupled with a GC/flame ionization detector (FID). A 30 m x 0.52 mm i.d. GS-Q capillary column (J&W Scientific Co) at 40°C was used for CO_2 analyses.

Two SF_6 standards were prepared in N_2 and in air at 67 pptv. Four CO_2 standards were prepared in N_2 at 10.12%, 0.459%, 0.304% and 0.081% concentrations.

Accuracy was calculated as a percent difference between the audit standard and found value, according to the equation:

$$A = 100 \cdot \frac{Y-X}{X} \qquad (1)$$

where :

X = audit target value (nominal)
Y = found value (measured)

The percent differences between the audit standard in nitrogen and air and the SF_6 system were -0.6 and -0.7, respectively. The percent difference between the three audit standards and the CO_2 bag measurement system was -2, 0, and 1. Nine continuous sensors were audited with the CO_2 standards. The maximum difference was -19% and the minimum was 2% (mean difference of -7 ± 11).

Precision for CO_2, as shown in Table 1, was evaluated by measurement of sample replicates, measurement of repeated instrument spans, and determination of the standard deviation of four one-minute instrument averages. Reported CO_2 concentrations for each sample were based on the mean of four one-minute averages (except those samples that had erratic readings or fewer averages because of small sample volume). For each test, the average of the standard deviations gave a precision of the measurement. Measurement bias based on sample replicates and instrument spans also is given in Table 1.

TABLE 1
Summary QC Data for CO_2

Test	*Parameter*	*Bias*	*Precision*	*n*
1	Sample Replicates	-4 %	9%	5
	Four One Minute Averages	-----	1 ppmv	43
	Instrument Spans	<1%	<1%	4
2	Sample Replicates	<1%	1%	5
	Four One Minute Averages	-----	2 ppmv	42
	Instrument Spans	<1%	<1%	4
3	Sample Replicates	< 1%	2	6
	Four One Minute Averages	-----	3 ppmv	42
	Instrument Spans	< 1%	<1%	4
4	Sample Replicates	-3%	3%	5
	Four One Minute Averages	-----	1 ppmv	43
	Instrument Spans	<1%	<1%	6

Note: Bias is based on the mean of the percent differences between replicates (or target versus found span values) and precision is based on one standard deviation of the mean. The precision of the four one-minute means is based on mean values in ppmv.

Precision for SF_6 (Table 2) was determined from sample replicates and repeated instrument spans. For Test 1, some bags were analyzed the next day so replicate analyses were repeated the next day to evaluate aging effects. The next day replicate analyses suggest a negative bias (decrease in bag SF_6 concentration) and greater variability (higher standard deviation of the mean). All bags after Test 1 were analyzed immediately after the release.

TABLE 2
Summary QC Data for SF_6

Test	*Parameter*	*Bias (%)*[1]	*Precision (%)*[1]
1	Replicate day 1	0	1
	Replicate day 2	-8	15
	Replicate both days	-4	11
	156 pptv span (both days)	-6	4
2	Replicates	0	1
	156 pptv span	-5	1
3	Replicates	0	2
	156 pptv span	0	3
4	Replicates	2	6
	156 pptv span	0	1

[1] Bias based on the mean of the percent differences between replicates (or target versus found span values) and precision based on one standard deviation of the mean.

RESULTS

At the time this manuscript was prepared, data from the experiment had been validated but results are considered preliminary. This section provides an overview of a portion of the data analysis performed to date.

Meteorology

A summary of the meteorological conditions during each release is provided in Table 3. Values reported are averages for periods during which the CO_2 plume was in steady state. For Tests 1, 3 and 4, the averaging period begins one minute after the start of the release and ends at the time the release ends for Test 1 and one minute before the end of the release for Tests 3 and 4. For the short release of Test 2 (70 seconds), the averaging period is for the one-minute period beginning ten seconds after the start of the release.

As seen from the stability classes, releases targeted to occur under stable conditions (E-F) were performed under neutral (D) and neutral to stable (D-E) conditions, where stability classes were estimated from graphs by Golder (1972). At the STF, chance plays a large role in obtaining very stable conditions while maintaining the target wind direction. As wind speeds decrease and stable conditions develop, the required wind direction (225°) becomes more variable, as discussed previously. Therefore, it was necessary to compromise between triggering the release at the slowest possible wind speed (an indication of increasing stability) and limiting plume meander (so as not to miss the downwind sensor array).

Chemical Release

The amount of CO_2 released in each test is the difference between the mass calculated before and after each test. The volume of the CO_2 before the release includes the volume of a 30 ton surge tank (1012 ft^3, 28.7 m^3), while the volume after the release includes the volume of the tank plus the release line (1025 ft^3, 29.0 m^3). The ideal gas law was used to calculate the mass concentration using an atmospheric pressure P_{atm}= 90.32 Pa (13.1 psia), appropriate for the elevation of the STF at 939 m above mean sea level. A summary of the release parameters for each test is provided in Table 4.

Plume Measurements

This section gives results of the CO_2 and SF_6 measurements. For bag samples, all but three were successfully collected (98% recovery) during the four tests. Failure of the three bags was related to insufficient sample collection attributed to failed solenoid valves or loose sample container lids.

TABLE 3
Summary Meteorology During Releases

Parameter	*Test 1*		*Test 2*		*Test 3*		*Test 4*	
Pasquill Stability Class	D		D-E		D-E		D	
Tower Height (m)	WS (m/s)	WD (deg)	WS (m/s)	WD (deg)	WS (m/s)	WD (deg)	WS (m/s)	WD (deg)
24.0 m	6.17	228	4.84	---[1]	5.36	---	8.32	---
16.0 m	6.18	---	4.39	---	5.06	---	7.85	---
8.0 m	6.03	---	3.78	---	4.44	---	7.03	219
4.0 m	5.70	231	3.37	237	4.06	234	6.65	218
2.0 m	5.26	224	3.00	233	3.61	229	6.08	214
1.0 m	4.75	---	2.67	236	3.19	232	5.49	215
0.5 m	4.32	226	2.35	233	2.93	---	4.97	213
0.25 m	3.99	227	2.01	236	2.64	233	4.61	216
40 m Arc Height=0.5 m	3.98	231	2.76	232	2.95	232	4.94	218
Ambient Temperature at 1 m (° C)	31.4		31.4		33.5		33.7	
Ambient Pressure (mb)	903		903		903		902	
Ambient Relative Humidity (%)	10		12		9		8	

[1]Missing Data

TABLE 4
Chemical Release Summary

Parameter		Test 1	Test 2	Test 3	Test 4
		7/22/93	7/26/93	7/27/93	7/28/93
CO_2 Release	Start:	18:55:01	19:41:01	19:30:00	19:46:00
	Stop:	18:58:00	19:42:11	19:34:25	19:50:25
SF_6 Release	Start:	18:55:04	19:41:06	19:30:06	19:46:06
	Stop:	18:58:04	19:42:16	19:34:28	19:50:30
Bag Sampler	Start:	18:55:08	19:41:08	19:30:09	19:46:08
	Stop:	19:00:01	19:44:39	19:35:41	19:52:01
CO_2/SF_6 Release Duration (s)		179	70	265	265
CO_2 Mass Released (kg)		81.9	112.9	171.9	165.0
CO_2 Release Rate (kg/s)		0.458	1.613	0.649	0.623
SF_6 Mass Released (mg)		163.4	63.6	238.2	240.0
SF_6 Release Rate (mg/s)		0.908	0.909	0.909	0.909

During Test 1, preliminary evaluation of the release indicated the volume was less than the target amount and analysis of the bags would not be necessary. The bags were analyzed nevertheless, if only to provide a shakedown of laboratory operations. However, a portion of the bags were not analyzed until the following morning. Tests on aging SF_6 standards in bags indicated a decrease in concentration with time for some bags. Replicate analyses performed the day after the release (Table 2) followed a similar trend indicated by a higher negative bias and poorer precision. Bags of Tests 2, 3 and 4 were analyzed within four hours of collection. A summary of results of bag data for CO_2 and SF_6 is presented in Figures 1 and 2 respectively.

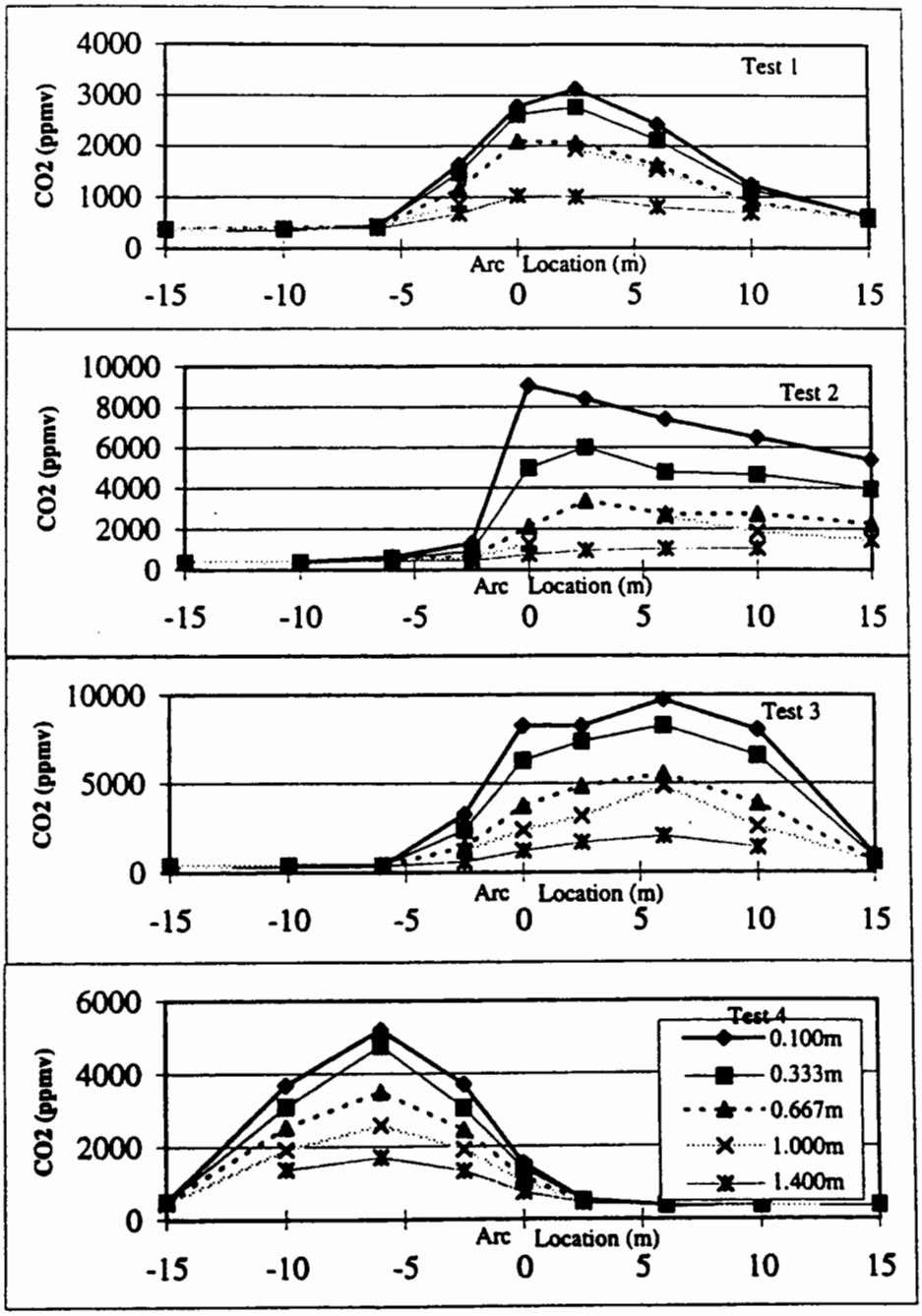

Figure 1. Summary of Integrated CO_2 Data for 5 Heights 40m Downwind of Release.

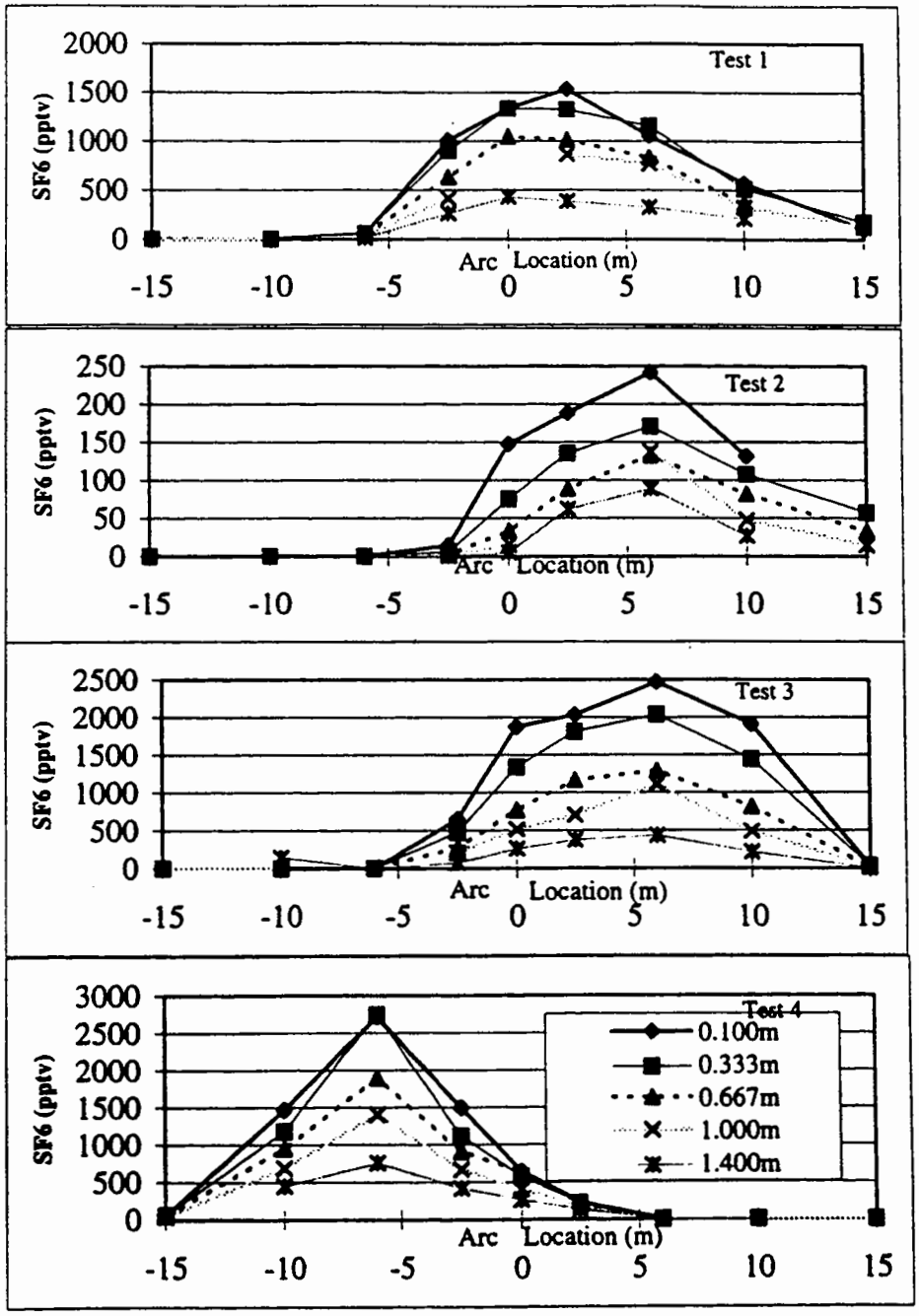

Figure 2. Summary of Integrated SF_6 Data for 5 Heights 40m Downwind of Release.

For continuous measurements of CO_2, the sensors generally operated successfully during the four tests. Maximum CO_2 concentrations recorded for Tests 1-4 were 10,500 ppmv (1%), 54,000 ppmv (5.4 %), 21,000 ppmv (2.1%) and 16,000 ppmv (1.6%), respectively. In regard to the adequacy of the measurement ranges of each sensor (0-1% and 0- 10%), three sensors saturated (i.e., they were exposed to a concentration that exceeded the maximum range) in Test 2, one in Test 3 and two in Test 4 (all 0-1% units). One unit in Test 3 recorded an anomalous trace and the data are considered suspect.

A comparison between continuous and bag integrated CO_2 generally indicates good agreement between the two measurement systems. Background CO_2 (approximately 366 ppmv) was measured for each test and subtracted from the bag samples and an average was obtained from the continuous sensors for the same period the bags were open. Linear regressions of all valid collocated bag and sensor pairs for Tests 1-4 indicate correlation coefficients of 0.9797, 0.9180, 0.9883, and 0.9873, respectively.

A comparison of bag CO_2, averaged sensor CO_2 and bag SF_6 data, at a height of 0.1 m above ground, by arc location, is provided in Figure 3. Generally there is good agreement between bag and averaged sensor data and the concentrations of SF_6 are proportional to CO_2. Test 2 SF_6 data levels were much lower than those in the other tests. No anomalies were noted in the SF_6 sampling and analysis systems, therefore the decrease in downwind concentrations was related to the release system.

Chemical Mass Balance

The effectiveness of the sensor and bag systems was determined by comparing the integrated fluxes of CO_2 and SF_6 measured at the 40-m arc to the amount of released material. The ratio of detected to released mass, or mass balance, is an indicator of the collective accuracy of the total system and the sufficiency of the wind speed and concentration arrays to characterize the total mass of a release.

The mass of CO_2 released was calculated as described previously, using temperature and pressure data from the tank and line collected before, after, and during each test.

The amount of released SF_6 was determined from the flow reading of the mass flow controller that maintained a constant flow rate during release. Flow voltages were collected by the STF DAS and converted to mass flow rates from calibration factors developed for SF_6. The total amount of released SF_6 was calculated from mass flow rate in mg/sec for time of the release.

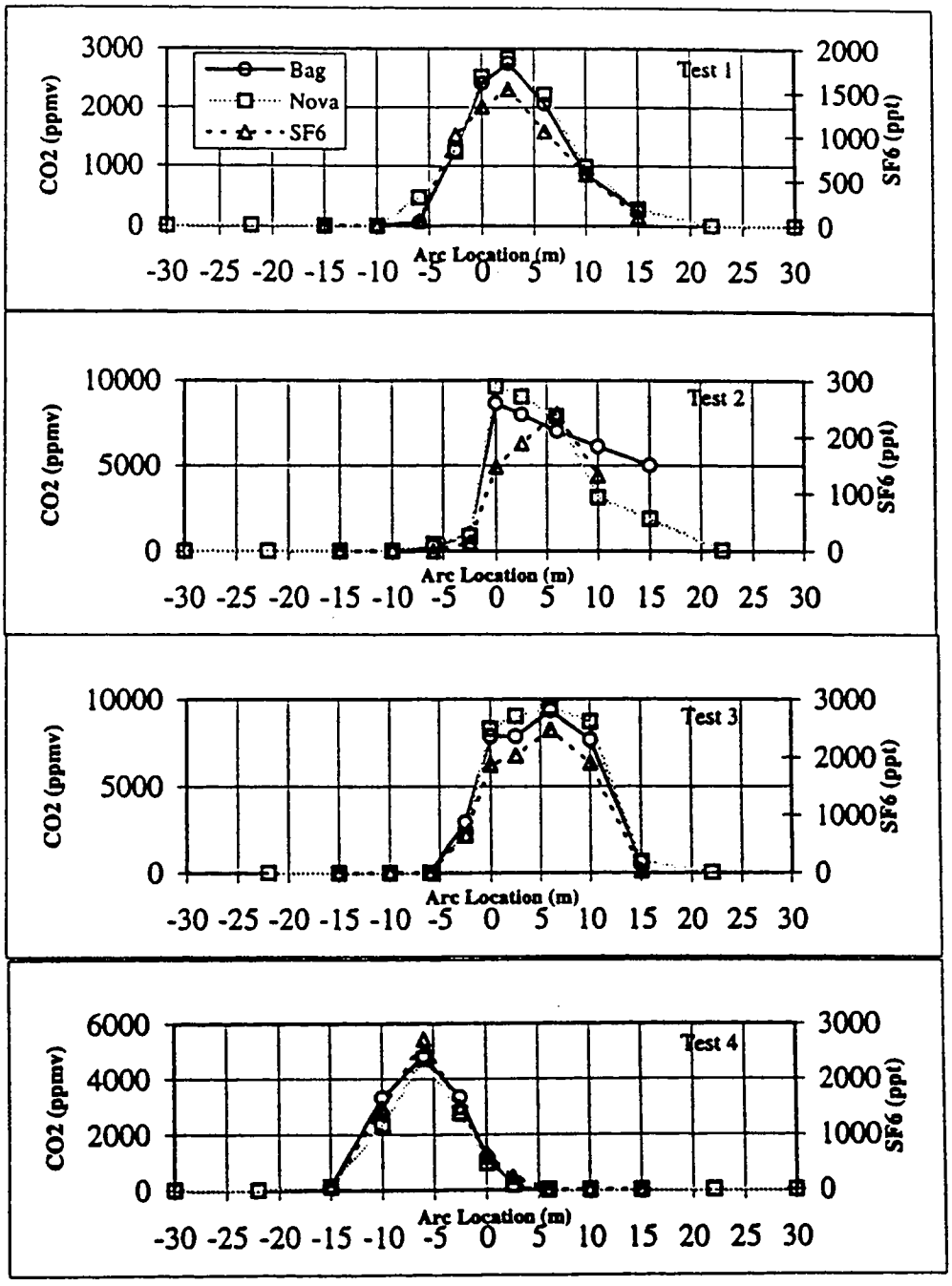

Figure 3. Comparison of CO_2 and SF_6 Data at the 0.1 m height.

The amount of mass passing through the 40-m arc of the sampling array was determined from direct measurements by the CO_2 sensors, integrated measurements of CO_2 and SF_6 in bags, and wind speed measurements from the meteorological tower. Over the period of sampling, the combined air and released gases defined a volume with cross-wind height and width defined by the dispersion of the released gas and along-wind length defined by the average wind speed. The integration or summation of the concentrations of the released gases in the volume, converted to density using ambient pressure and temperature, gave the amount of mass for the released gases in the volume.

The integration of the mass of released gases measured at the 40-m array was accomplished in the following steps:

1. Average concentrations for continuous CO_2 sensors and average wind speeds for sensors on meteorological tower were calculated for the time that bags collected air.

2. Background concentrations for sensors and bag samples were subtracted from all data. For the sensors, the background was determined from measurements when the plume was not present. For the bag samples, the background was determined from upwind samples and from samples not affected by the plume.

3. Missing gas concentrations for sensors and bags were estimated by interpolation. The scheme started at the level nearest ground and worked in an upward direction. Missing concentrations were estimated by assuming that the ratio of the missing measurement to that of its nearest neighbor at the same height equals the ratio of concentrations at the same horizontal locations at the next lower height. For missing values equidistant between two points, the averages of the concentrations on both sides of the missing value were used.

4. Cross-wind integrated concentrations (CIC) were calculated with a linear interpolation of concentration across the array. Each measured concentration was assumed to be constant over a horizontal distance defined by the midpoints between measurements. The CIC was the sum of the products of the individual concentrations and their horizontal spacing.

5. Average horizontal wind speeds were determined at the level of the concentration measurements for the period of bag sampling. Wind speeds at 0.333, 0.667, and 1.4 m above the ground were calculated using a geometric interpolation from data collected on the meteorological tower at 0.25, 0.5, 1, and 2 m above the ground. The interpolation for u_h at level z_h with u_1 at z_1 and u_2 at z_2 was given by:

$$u(z_h) = u(z_1)\left(\frac{u(z_2)}{u(z_1)}\right)^{\ln(z_h/z_1)/\ln(z_2/z_1)} \quad (2)$$

6. The average horizontal wind speed at the level nearest the ground (0.1 m) was determined by extrapolating the logarithmic wind profile from 0.25 m using a z_0 of 0.0002 m:

$$u(z) = u(0.25)\left(\frac{\ln(z/z_0)}{\ln(0.25/z_0)}\right) \quad (3)$$

7. The volume flux of released gas was computed at each vertical level by multiplying the CIC at each level by the wind speed for that level. The vertically integrated flux was calculated with a linear interpolation of flux in the vertical direction. At each level, the flux was assumed to be constant over a vertical distance defined by the midpoints between measurements. To account for the gas that was above the measurement array, the flux was assumed to decrease exponentially with increasing height. The concentrations at levels 0.667 and 1.4 m were used to determine the rate of decrease with height. The total flux of released gas was calculated as the sum of the products of the individual fluxes and their vertical spacing.

8. The total mass measured by the sampling array was calculated from the total volume flux using the density of CO_2 and SF_6 at the pressure and temperature conditions during each test.

Chemical mass balance results are summarized in Table 5. Results of CO_2 for both continuous sensors and integrated bag samples were reasonably good. For Tests 1, 3, and 4, the measured mass accounted for at least 90% of the released mass. Measured values for Test 2 accounted for only about 60% of the mass. SF_6 results were not as good. For Tests 1 and 4, the measured mass was more than the released mass. For Test 3, the measured mass accounted for only about 50% of the released mass. Results of Test 2 indicate that there may have been a problem with the release system at that time.

TABLE 5
Chemical Mass Balance Summary

Parameter	Test 1	Test 2	Test 3	Test 4
	7/22/93	7/26/93	7/27/93	7/28/93
CO_2 Mass Released (kg)	81.9	112.9	171.9	165.0
CO_2 Mass Measured-Continuous (kg)	93.0	59.9	161.0	163.1
CO_2 Mass Measured-Integrated (kg)	77.4	73.2	155.3	159.6
CO_2 Mass Balance- Continuous (%)	114	53	94	99
CO_2 Mass Balance-Integrated (%)	95	65	91	97
SF_6 Mass Released (mg)	163.4	63.6	238.2	240.0
SF_6 Mass Measured (mg)	171.9	9.4	126.9	280.0
SF_6 Mass Balance (%)	105	15	53	117

MODEL COMPARISON

The remainder of this paper describes an application of the dense gas models SLAB and DEGADIS to two of the Experiment 1 releases. The SLAB (Ermak, 1990) and DEGADIS (Spicer and Havens, 1989) models were selected because of their popularity and wide use. They are both included as Alternative Air Quality Models in EPA's Guideline on Air Quality Models (U.S. EPA, 1994).

Model Inputs

SLAB and DEGADIS were used to simulate Tests 3 and 4. Test 3 occurred during Pasquill stability Class D-E (slightly stable) conditions and Test 4 occurred during class D (neutral) conditions. Meteorological conditions during these tests are detailed in Table 3. Actual model inputs are tabulated in Table 6. Both Tests 3 and 4 consisted of releases lasting 265 seconds. Wind speeds and wind directions are

averages over the 3-minute (180-second) period beginning 60 seconds after CO_2 releases began, and ending 25 seconds before the releases ended (wind direction was used to align model output with the sensor array, as described below). DEGADIS was run in the isothermal steady state mode, and the last 4 items in Table 6 were used to define the air/contaminant density profile. A linear profile was assumed, from molar fraction equal to 0.0 to molar fraction equal to 1.0. SLAB was run in the evaporating pool mode. It was assumed that the temperature of CO_2 at the moment of release was the same as the ambient air temperature.

Model estimates and corresponding experimental data are for the nominal "40 m" sensor array. This array is actually 38.5 m from the center of the release pit. DEGADIS does not allow designation of specific discrete receptors, but rather calculates receptor locations internally. The DEGADIS- generated receptor distance closest to the nominal 40 m array was 38.3 m downwind for Test 3 and 38.5 m for Test 4. A distance of 38.5 m was defined for both SLAB simulations as the maximum modeled downwind distance. Also, the sensor array was centered on an axis oriented at 225°, for prevailing southwest wind directions. The actual average wind direction during experimental releases was off the 225° line by varying amounts. For Test 3, the actual average wind direction was 231°, 6° off the sensor array axis and for Test 4 the array wind direction was 215°, 10° off the array axis. For presentations, model results were thus shifted laterally to line up modeled and experimental plume center lines. For these reasons, actual downwind distances are not exactly the same for modeled and experimental conditions, but are within about 1 meter.

Model Results and Comparison with Experimental Data

Three different experimental data sets were used for model comparisons, corresponding to different averaging times:

1. Integrated Bag Samples. Bag sample concentrations were adjusted to represent the actual CO_2 release time rather than the total bag sampler time (see Table 4).

2. Three minute averages from the Nova sensors, for the same periods for which wind data were averaged.

3. Maximum 30-second end-to-end average concentrations during the same 3-minute averaging period.

TABLE 6
Model Inputs Used for SLAB and DEGADIS Simulations

Parameter	*Test 3*	*Test 4*
Common Parameters		
CO_2 Release Rate (kg/s)	0.649	0.623
Wind Speed @ 1.0 m agl (m/s)	3.19	5.49
Wind Direction (deg)	231	215
Ambient Temperature (°K)	306	306
Ambient Pressure (mb)	903	903
Relative Humidity (%)	9	8
Surface Roughness (mm)	0.17	0.17
Pasquill Stability Category	E	D
Averaging Time (sec)	30	30
Source Area (m^2)	1.0	1.0
CO_2 Physical Properties(SLAB)		
Vapor Heat Capacity (J/kg-K)	853	853
Heat of Vaporization (J/kg)	573,500	573,500
Liquid Specific Heat (J/kg/K)	1,276	1,276
Liquid Density (kg/m^3)	1,564.3	1,564.3
CO_2 Physical Properties (DEGADIS)		
Mean Heat Capacity Constant (J/kg-Mol/K)	4,241	4,241
Power for Contaminant Heat Capacity Equation	1.0	1.0
Contaminant Concentration for Molar Fraction = 0.0 (kg/m^3)	0.0	0.0
Air/Contaminant Mixture Density for Molar Fraction = 0.0 (kg/m^3)	1.0512	1.0512
Contaminant Concentration for Molar Fraction = 1.0	1.5969	1.5969
Air/Contaminant Mixture Density for Molar Fraction = 1.0	1.5969	1.5969

Background values of 360 ppm were subtracted from bag sample concentrations. Background values based on upwind measurements were subtracted from Nova sensor data.

DEGADIS model output consists of ground-level (height = 0.0 m) centerline concentration plume half-widths, parameters S_y and S_z (Sigma Y and Sigma Z, not the same as Gaussion Plume Sigmas), and a power-law wind velocity profile. These values were used, with equations v-71 (pp. 38-39) of the DEGADIS User's Guide (Spicer and Havens, 1989) to extrapolate crosswind profiles to heights above ground level. The SLAB model includes height above ground as a model input variable. Separate SLAB model runs were made for each sensor height.

Trinity Consultants, Inc. software was used for DEGADIS model runs and Bowman Environmental Engineering software was used for SLAB model runs. Trinity and Bowman software enhancements consist primarily of menu-driven data entry software.

The following results are qualitative. More detailed analyses, using performance measures such as recommended in by EPA (1984) and used by Hanna et al (1991), and a wider selection of models, should be made.

Maximum Concentrations

Figure 4 depicts maximum concentrations. SLAB and DEGADIS results are shown, along with experimental data as described above. For the slightly stable Test 3 case, there is a tendency toward overprediction by both models at low cloud heights (near 0.1 m agl) and underprediction at high cloud heights (1.0 to 1.4 m agl). Both models appear to skew the vertical distribution, i.e. underestimate cloud thickness. Near ground level, where concentrations are highest, DEGADIS overpredicts by about a factor of 2. For Test 4, the neutral case, the same tendencies are evident toward overprediction at low cloud heights and underprediction at high cloud heights by DEGADIS. The SLAB results appear to agree better with experimental results than do the DEGADIS results.

Cloud Widths

Figures 5 and 6 show cross-wind profiles of CO_2 concentration at cloud heights of 0.1 m and 0.667 m for Tests 3 and 4.

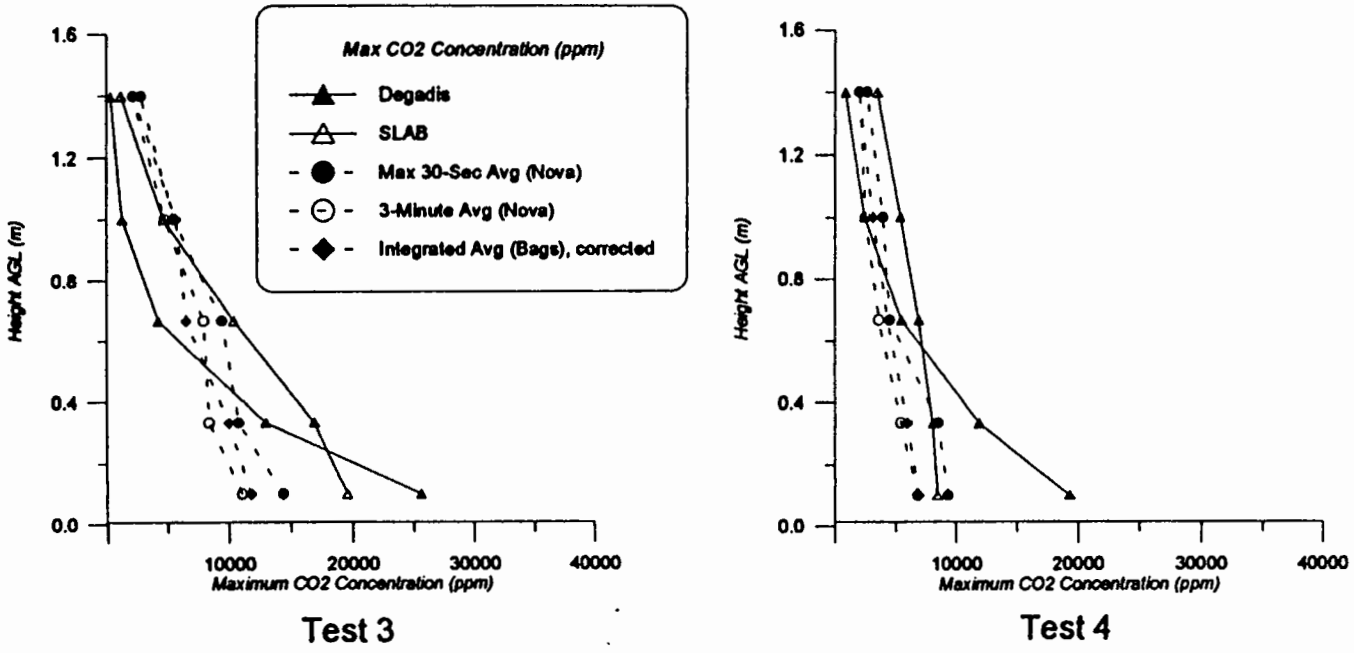

Figure 4. Modeleded and Observed Vertical Profiles of Maximum CO_2 Concentrations at 40 m, Surface Roughness = 0.17 mm

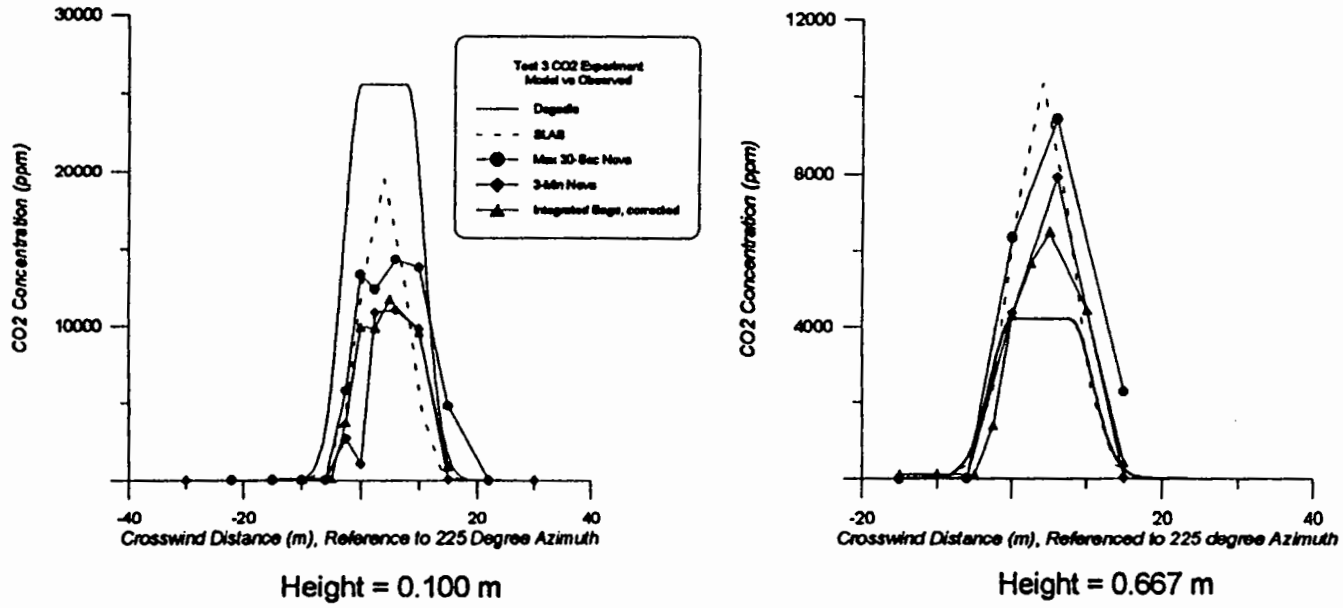

Figure 5. Modeled and Observed Crosswind Profiles of CO_2 Concentrations at 40 m Downwind for Two Heights, Test 3, Surface Roughness 0.17 mm

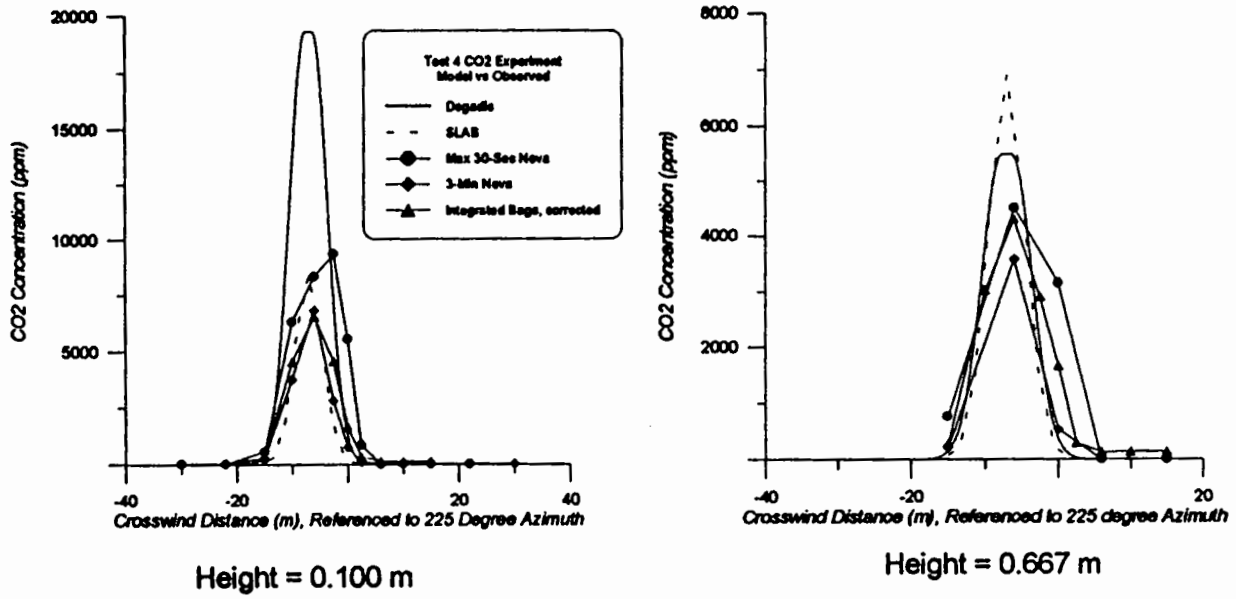

Figure 6. Modeled and Observed Crosswind Profiles of CO_2 Concentrations at 40 m Downwind for Two Heights, Test 4, Surface Roughness 0.17 mm

For the slightly stable Test 3 case, DEGADIS appears to slightly overpredict cloud width at low cloud heights, while SLAB is more in line with experimental results. At mid-heights, represented by the 0.667 m height, both SLAB and DEGADIS are in good agreement with experimental results. For the neutral Test 4 case, both models are in good agreement with experimental results.

SUMMARY AND CONCLUSION

In July, 1993 a baseline experiment involving a series of four CO_2 releases was performed at the DOE Spill Test Facility in Nevada. Specific goals of the experiment were to design and test a release, monitoring, and data management system and to acquire a data base suitable for model testing. The data collected during the 1993 experiment began to fill data gaps in two important areas. First, releases were performed during stable atmospheric conditions, albeit only slightly stable conditions. Previous wind tunnel and field experiments have amassed data to evaluate dense gas dispersion models for flow over flat or sloping terrain during neutral and unstable atmospheric conditions. However, there is a paucity of data for model development and evaluation for low wind speed, stable atmospheric conditions. Second, the meteorological data collected at eight levels on the 24 m tower and from a fast response sonic anemometer provide ample on-site boundary layer characterization during each release. During the rest of the time meteorological data collection continues at a less intense rate to document on-site boundary-layer climatology; this will be valuable for optimizing future field experiments, such as experiments designed to capture data during more stable conditions.

The design conditions for the CO_2 releases were moderately low wind speed, stable atmospheric conditions. Tests 1 and 4 were actually performed during near-neutral conditions with winds about 5 m/s, while Tests 2 and 3 were performed during slightly stable conditions with winds about 3 m/s. While the Spill Test Facility provides an ideal location for performing releases of hazardous materials because of its remote location, variety of terrain, and relatively predictable meteorology, chance still plays a part in attempting a release during stable conditions. Analysis of the meteorological data shows that stable atmospheric conditions develop shortly before sunset as the wind speed begins to drop. The regularity of the near-sunset drop in wind speed and the persistence of southwesterly winds during the drop makes it relatively easy to target release times for various atmospheric stabilities. The 0.08 m/s per minute drop in wind speed is not a serious problem for release durations of 5 minutes or less. However, for much longer releases this non-stationarity complicates data analysis.

Dense gas dispersion models were used to design the monitoring array for a CO_2 release rate of about 1 kg/s. The primary sampling arc at 40 m downwind was instrumented with integrated bag samplers and continuous CO_2 analyzers at 5 levels in the vertical and out to ±30 m in the lateral. CO_2 gas was released from an insulated 1012 ft^3 (28.7 m^3) surge tank through a 4 in (0.1 m) pipe into a 1 m^3 baffled discharge chamber, successfully releasing a steady, low momentum, nearly isothermal dense gas. SF_6 was injected into the CO_2 stream at 0.9 mg/s flow rate. Vertical and lateral distributions of SF_6 compare well with distributions of CO_2, suggesting that the SF_6 was well mixed in the CO_2 stream. At 40 m downwind the plume clearly exhibits dense gas behavior, with width-to-depth ratios ranging from 10 to 30. A comparison of the continuous CO_2 analyzers with bag samples revealed good agreement. Mass flux estimates at the 40 m arc, excluding Test 2, averaged 90% ±7% of the released mass. Mass flux estimates for SF_6 were not in as good agreement as CO_2.

Preliminary comparisons of measured CO_2 and modeled concentrations for Tests 3 and 4 show that the SLAB and DEGADIS models tend to overpredict ground level concentrations and underpredict the cloud depth, especially for the more stable test (Test 3). Both models are in much better agreement with observations at upper plume elevations. For Test 4, SLAB estimates are in excellent agreement with the vertical profile observations. Comparisons of the lateral concentration profiles and model estimates at two heights show that both models do well at characterizing the lateral spread of the dense cloud.

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16. ABSTRACT

An Experimental baseline CO2 release experiment at the DOE Spill Test Facility on the Nevada Test Site in Southern Nevada is described. This experiment was unique in its use of CO2 as a surrogate gas representative of a variety of specific chemicals. Introductory discussion places the experiment in historical perspective. CO2 was selected as a surrogate gas to provide a data base suitable for evaluation of model scenarios involving a variety of specific dense gases. Releases were conducted under baseline conditions including a simulated "evaporating pool" release over flat unobstructed terrain. The experiment design and setup are described, including design rationale and quality assurance methods employed. Design conditions included moderately low wind speed, stable atmospheric conditions. Four releases were performed, two of which were during near-neutral conditions and two during slightly stable conditions. Resulting experimental data are summarized. These include CO2 cloud characteristics measured at 40 m downwind from the release point. Experiment success and effectiveness is discussed in terms of mass balance analyses. For Test 1, 3, and 4 the measured mass accounted for at least 90% of the released mass. Measured values for Test 2 accounted for only 60% of the released mass. Data usefulness is examined through a preliminary comparison of experimental results with simulations performed using the SLAB and DEGADIS dense gas models.

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