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An Investigation of Concentration Fluctuations during a Tracer Study at Rafnes

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SUMMARY

Twelve tracer tests were conducted in June 1990 to characterize the dispersion of gases from leaks in the VCM area of the Rafnes petrochemical factory. Sulfur hexafluoride gas was released at a continuous rate from 2-5 points throughout the process, while 15-min air samples were collected along cross-wind traverses. Gas chromatography was used to analyze the samples for mean SF_6 concentrations, and during nine of the twelve tests, a new fast-response SF_6 analyzer was used to measure near-instantaneous concentrations at a fixed point near the sampling array. Main results from the concentration fluctuation data are summarized as follows:

- In most of the Rafnes tests, the dominant source of SF_6 measured at the fast-response analyzer was the northernmost release point.

- The time series with the lowest mean concentrations (and the highest peak-to-mean ratios) were measured when the analyzer was located near an edge of the mean plume, while the highest mean concentrations (and lowest peak-to-mean ratios) were measured when the analyzer was located near the mean plume centerline.

- The peak-to-mean ratio was less than 3.0 during most of the Rafnes tests, except when the receptor was located near the mean plume edges, in which case, the ratio was as large as 13.4.

- The concentration fluctuation intensities were 0.22-0.29 for the near-centerline data, and the intensities were 0.87-2.44 for the cases in which the analyzer was located near an edge of the mean plume. These values were lower than usually observed amid non-complex terrain and for isolated sources.

- Approximately 65% of the tests had intermittency factors greater than 0.90, where the highest was 1.00 and the lowest was 0.736. These values are higher than observed during studies amid simple terrain and for isolated sources.

- The concentration probability distributions nearly collapse for the time series collected in similar regions of the mean plume. In particular, the distributions were similar when the analyzer was located near the centerline of the mean plume (Tests 3,4,5, and 8), and when the analyzer was located near the fringes of the mean plume (Tests 6,7,11, and 12).

- In general, there were few differences in the concentration statistics and in the concentration distributions between sequential 15-min periods (i.e., the A and B tests), but the time scales varied considerably in some cases.

- In one experiment, Test 7, the prevailing winds were not channelled along the major axis of the large factory buildings; instead, the winds blew across an open, hilly area of trees and grassland. In this case, meander was the dominant source of concentration fluctuations at the fast-response analyzer, and the magnitude of the high-frequency fluctuations was smaller than the magnitude of the high-frequency fluctuations in the other tests.

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AN INVESTIGATION OF CONCENTRATION FLUCTUATIONS DURING A TRACER STUDY AT RAFNES

1 INTRODUCTION

Twelve tracer experiments were conducted in June 1990 to investigate diffuse emissions of hydrocarbons (HC) at the Norsk Hydro Rafnes petrochemical plant in southeastern Norway. Sulfur hexafluoride (SF_6) and bromotrifluoromethane ($CBrF_3$) were released from single and/or multiple points within the factory. The mean dispersion of each tracer gas was characterized from 15-min average concentrations in air samples collected within an array using portable, automatic syringe samplers. Samples for the diffuse hydrocarbons were collected using ATP tubes, and the mean concentrations were measured using flame ionization analysis. In addition, a new fast-response SF_6 analyzer was used to characterize the time fluctuations of the instantaneous exposure at a fixed receptor. (This study was the first field campaign in which NILU operated the new analyzer).

Analyses of the 15-min average data and the HC emission estimates are presented elsewhere (Tønnesen, 1990). In this document, only the instantaneous concentration data from the Rafnes Study are examined. Until recently, instruments were not available to measure concentration fluctuations on a near-instantaneous time scale, and thus, to date, few experimental data exist. Therefore, the Rafnes data set and, importantly, all of future NILU data sets will be extremely valuable to the developing field of research concerning concentration fluctuations.

Included in this paper are background information on the current status of concentration fluctuation research (Section 2), a description of the experimental procedure for the study at Rafnes (Section 3), and results from the instantaneous SF_6 data collected during the Rafnes experiments (Section 4). In an appendix are listings of the computer programs for processing and analyzing the time series of concentration data.

2 BACKGROUND ON CONCENTRATION FLUCTUATIONS

Until recently, most of the research in the field of atmospheric dispersion have focused upon modeling and monitoring mean concentrations (i.e., usually 1-hr averages) downwind of a contaminant source. Unfortunately, a mean concentration averaged over minutes or hours is not adequate to estimate potential damage to a receptor which is sensitive to instantaneous, high concentrations of the contaminant. In particular, downwind of a narrow meandering plume, a receptor receives intermittent doses of the pollutant (separated by periods of uncontaminanted air) as the plume sweeps back-and-forth amid large-scale wind direction changes. Also, the concentration distribution within an instantaneous plume may be irregular as a result of internal mixing processes; thus even in the absence of horizontal meander, the concentration time series at a downwind receptor near the ground will contain concentration fluctuations with time scales reflecting the internal mixing and vertical wind motions.

To illustrate this point, Figure 1 shows two hypothetical cases of dispersion of a pollutant from a small source. Case 1 represents traditional mean plume theory in which wind conditions are assumed to be stationary over an averaging period of, say, 15 minutes. The plume centerline follows the direction of the mean wind, and the plume spreads in the crosswind direction as a function of travel time from the source. If the 15-min average concentrations are measured by samplers oriented along a line perpendicular to the plume centerline (i.e. Receptors 1-5), the mean plume profile will be Gaussian as shown with the maximum concentration measured at the centerline. Likewise, if the instantaneous concentrations are also measured throughout the 15-min averaging period, the time series at each receptor will consist of concentrations which fluctuate about a steady mean concentration, where the high-frequency fluctuations are a function of internal mixing within the plume (i.e., refer to the time series depicted in the figure for Receptor 2).

Case 2, on the other hand, represents a more realistic view of dispersion in which the instantaneous winds are non-stationary. Shown in black is a snapshot of an instantaneous plume which spreads relative to the instantaenous plume axis and meanders relative to the mean plume axis. Throughout the 15-min averaging period, the instantaneous plume meanders back-and-forth across the line of receptors; thus the nature of the instantaneous exposure at any single receptor is a function of the concentrations within the instantaneous plume in addition to nature of the meander. The mean wind direction is the same as in Case 1, and as shown in the lower left-hand figure, the time-averaged plume is the same as for Case 1. However, as shown in the lower right-hand figure, the time series at Receptor 2 is quite different from the time series of Case 1. In Case 2, the peak concentrations are much higher than the mean, and the low-frequency component of the exposure is the result of the instantaneous plume meandering back-and-forth, while the high-frequency component is due to internal mixing within the plume.

Mean-field theory is not sufficient to describe pollutant impact at such time scales, nor under conditions in which meander is present. Little is known, theoretically or experimentally, about the transport and dispersion of plumes on the near-instantaneous time scales. It has only been during the last decade that experiments have been conducted in laboratory and field settings. These include the studies of Brown (1987), Deardorff and Willis (1984), Fackrell and Robins (1982a,b), Hanna (1984b), Jones (1979, 1983), Lamb et al. (1985), Lewellen and Sykes (1986), Mylne (1990), Netterville (1979), Peterson et al. (1988, 1990), Peterson (1989), Ramsdell and Hinds (1971), Robins (1979), Sawford (1985a,b), Storebø (1983), Storebø et al. (1983), and Wilson et al. (1985). During most of these

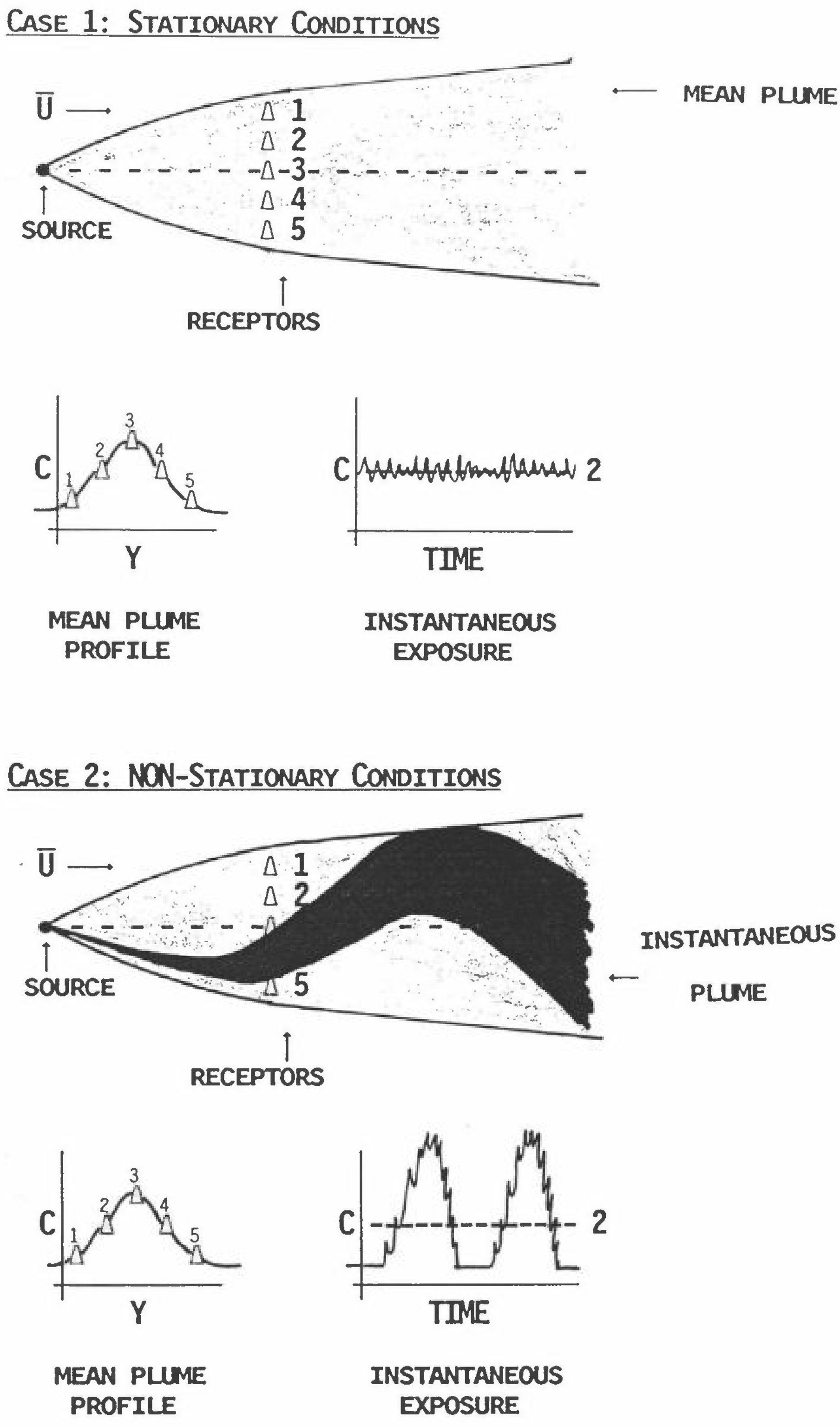

Figure 1: Concentration variations within the mean plume.

studies, a single point source was used rather than multiple point sources, line sources, or area sources.

In general, a statistical approach has been used to analyze the concentration time series from the above-mentioned studies. A few simple concentration statistics (i.e., intermittency factor, intensity, and peak-to-mean) have been related to the location of the receptor with respect to the source and with respect to the centerline of the time-averaged plume. These statistics are defined and described as follows.

The intermittency factor is defined as the fraction of time the receptor is impacted by the plume (in other words, the fraction of a concentration time series in which the concentration is non-zero). Several factors contribute to the intermittent nature of the concentration exposure, including downwind distance, source height, and plume buoyancy. According to Wilson et al. (1985), wind tunnel experiments revealed an increase of the intermittency factor (measured on the centerline of the mean plume) from approximately 0.15 (at a downstream distance of 4.3 source heights) to approximately 0.7 (at 15 source heights). In the convective tow tank studies of Deardorff and Willis (1984), centerline intermittency factors increased from near-source observations of 0.0 and 0.5, in buoyant and non-buoyant plumes, respectively, to downwind values of approximately 1.0. Across the mean plume, the intermittency factor decreases from a maximum at the centerline to a minimum near the fringes. In wind tunnel studies where the flow blows predominately in one direction, intermittency factors were 1.0 and 0.6 near the plume centerline for ground-level and elevated sources, respectively, and decreased to approximately 0.1 at crosswind distances of $2\sigma_y$ (Robins et al., 1979; Wilson et al., 1985). During full-scale experiments, typical intermittency factors ranged from 0.5 to 0.8 near the plume axis and from 0.1 to 0.3 at crosswind distances near $2\sigma_y$ (Hanna, 1984b; Sawford, 1985a; Lamb et al. 1985; Peterson et al., 1988).

The concentration fluctuation intensity for a time series is defined as the ratio of the root-mean-square concentration (i.e., the standard deviation) to the mean concentration. (Concentration intensity for concentration measurements is equivalent to turbulence intensity for velocity measurements). In general, concentration fluctuation intensity is at a minimum, typically less than 1.0, on the plume axis and increases with crosswind distance to a value of order 10 near the plume edges (Lamb et al., 1985; Ramsdell and Hinds, 1971; Hanna, 1984b; Sawford, 1985a,b; Lewellen and Sykes, 1985; Mylne, 1990). For wind tunnel tests, concentration intensity on the plume centerline decreased with downwind distance, and typical centerline intensities were near 5 at 4.3 source heights downwind, and near 2 at 15 source heights downwind. Likewise, the intensities near the plume edges were approximately 15 and 5 for the same downwind distances (Wilson et al., 1985).

The peak-to-mean ratio is usually calculated as the concentration at the 99th-percentile of the cumulative probability distribution divided by the mean concentration of the time series. Csanady (1967) reported values of 2 to 10 for receptors near the plume centerline and values of 30 to 100 near the plume edges with a tendency to decrease with increasing travel time. Jones (1983) observed peak-to-mean ratios ranging between 30 and 150 during a near-source field experiment, while Peterson et al. (1988) reported values of 3 and 16 for tests conducted under stable and convective conditions, respectively, where source-to-receptor distances in both cases were near 200 m.

Time series concentration data have also been analyzed in terms of the frequency distributions, and the general observations have been that the distributions are approximately exponential at receptors located near the fringes of the mean plume, and approximately normal at receptors located near the centerline of the mean plume (Sawford et al., 1985; Hanna, 1984; Lamb et al., 1985). Unfortunately in most cases, the actual distributions are more complicated than any of the simple (i.e., normal, log-normal, or exponential) distribution forms, but little effort has been made to try to explain and understand the 'kinks' and trends in the distributions. Peterson et al. (1990) addressed this topic to some extent, and showed how kinks in the distributions are a function of receptor location for plumes which meander periodically. Especially near an isolated source, meander is one of the most important factors which determine the nature of the instantaneous concentration exposure at a fixed receptor; thus it is critical that we strive to better understand the factors which determine the relationships between the frequency distributions of the wind and the concentration distributions at a single point.

Most of the experiments performed to date have been conducted in wind tunnels and amid simple, flat terrain and grasslands, and as mentioned earlier, most of the concentrations have been measured downwind of single, isolated point sources. Sawford (1985b) showed that the contributions of two sources at a given measuring point are only partially correlated and that the correlation is strongly a function of separation distance and downwind distance. Thus, it is important to improve our understanding of instantaneous dispersion from multiple point sources, in particular in areas such as petrochemical plants, because leakages of hazardous gases may occur throughout the process. The instantaneous concentration exposure at a fixed receptor will be affected by the spatial structure of the turbulence in addition to the separation distance of the leaks. The Rafnes data set discussed below is unique because it represents one of the first known collections of concentration fluctuation measurements amid an industrial complex and from multiple point sources.

3 EXPERIMENTAL PROCEDURE

Tables 1, 2, and 3 describe the experimental conditions and the release conditions during ten of the twelve tracer experiments performed at Rafnes during the period 19-06-1990 through 22-06-

1990. Tests 1, 9, and 10 are not described because instantaneous tracer data were not available to analyze from these periods for the following reasons: during Test 1, SF_6 was released from a single elevated point source which did not impact the ground near the location of the fast-response-analyzer; and during Tests 9 and 10, the instantaneous SF_6 data were not stored because of a problem with the data acquisition system.

Tests were conducted during morning, afternoon, and evening hours. The winds generally blew from the southeast, but during Test 7 and during Tests 11 and 12, the winds were were from the northeast and east, respectively. The mean wind speeds were 2.8-7.4 m/s, where the lowest mean wind speeds occurred during Test 11, and the highest, during Test 2. The standard deviation of the wind direction fluctuations varied between 1.8 deg and 8.4 deg, with the lowest observed in Test 4, and the highest, in Test 7. The release flowrates of SF_6 were the same at all release points, but the total release rate in each test ranged from 3.91 g/min to 9.77 g/min, where the number of release points in a single experiment ranged between two and five (i.e., see Table 2 for coordinates). The height of each release point was either ground level or 15 m above the ground, and as shown in Table 3, the distance between each release point and the location of the fast-response SF_6 analyzer ranged from 121 m to 285 m.

Figure 2 shows the layout of the factory, including the VCM area, the chlorine (KLOR) area, and the ethylene (ETHYLEN) area. The meteorological tower (denoted by the letter T) was positioned along the wharf (Kai 2), and the winds were measured at the 10-m level. Also shown are the major buildings in the vicinity of the SF_6 release, the roadways (denoted by the dashed lines), all release points used in the experiments (denoted by the symbols *), sampler locations (denoted by the triangles) for the 15-min average concentrations near the fast-response analyzer, and three locations for the fast-response analyzer (denoted by the plus signs). Note: during all but two

Table 1: Test conditions.

TEST	DATE	TIME	WD	WS	σ_θ	Q	Xr	Yr
			deg	m/s	deg	g/min		
2A	190690	1745-1800	144	5.6	3.6	9.77	5.00	15.740
2B	190690	1800-1815	141	7.4	3.6	9.77	5.00	15.740
3A	200690	1105-1120	129	5.6	2.1	9.77	5.00	15.740
3B	200690	1120-1135	129	6.3	2.1	9.77	5.00	15.740
4A	200690	1310-1325	130	6.8	1.8	3.91	5.00	15.740
4B	200690	1325-1340	129	7.2	2.1	3.91	5.00	15.740
5A	200690	1615-1730	130	6.3	3.6	7.82	5.00	15.740
5B	200690	1630-1645	135	7.0	3.0	7.82	5.00	15.740
6A	200690	1720-1735	133	6.3	3.6	3.91	5.00	15.740
6B	200690	1735-1750	144	6.7	3.6	3.91	5.00	15.740
7A	210690	1015-1030	40	3.9	8.4	7.82	4.83	15.565
7B	210690	1030-1045	40	3.9	7.2	7.82	4.83	15.565
8A	210690	1550-1605	135	3.9	3.6	5.86	5.00	15.740
8B	210690	1605-1620	133	3.7	2.4	5.86	5.00	15.740
11A	220690	1045-1100	92	2.8	6.0	5.86	5.00	15.740
11B	220690	1100-1115	99	3.4	6.0	5.86	5.00	15.740
12A	220690	1235-1250	90	4.9	5.4	5.86	4.94	15.770

where:

WD=wind direction Q=SF_6 release rate

WS=wind speed Xr,Yr=coordinates of analyzer location

σ_θ=standard deviation of horizontal wind fluctuations

Table 2: Release Coordinates.

TEST	X_1	Y_1	X_2	Y_2	X_3	Y_3	X_4	Y_4	X_5	Y_5
2*	5.050	15.500	5.075	15.540	5.055	15.565	5.100	15.645	5.075	15.645
3*	5.050	15.500	5.075	15.540	5.055	15.565	5.100	15.645	5.075	15.645
4	5.100	15.645	5.075	15.645	-	-	-	-	-	-
5	5.095	15.645	5.030	15.500	5.075	15.540	5.055	15.565	-	-
6	5.075	15.540	5.055	15.565	-	-	-	-	-	-
7	5.070	15.490	5.075	15.550	5.055	15.565	5.095	15.645	-	-
8*	5.050	15.550	5.030	15.500	5.095	15.645	-	-	-	-
11*	5.050	15.550	5.030	15.500	5.095	15.645	-	-	-	-
12*	5.050	15.550	5.030	15.500	5.095	15.645	-	-	-	-

where:

X_n, Y_n are the coordinates of the release point n, and * denotes tests in which X_1, Y_1 was at a height of 15 m. All other release heights were ground-level).

Table 3: Source-to-Receptor Distances.

TEST	D_1	D_2	D_3	D_4	D_5
	(m)	(m)	(m)	(m)	(m)
2	245	214	183	138	121
3	245	214	183	138	121
4	138	121	- -	- -	- -
5	134	242	214	183	- -
6	214	183	- -	- -	- -
7	251	245	225	277	- -
8	196	242	134	- -	- -
11	196	242	134	- -	- -
12	246	285	199	- -	- -

where:

D_n is the source-to-receptor distance between release point n and the fast-response SF_6 analyzer.

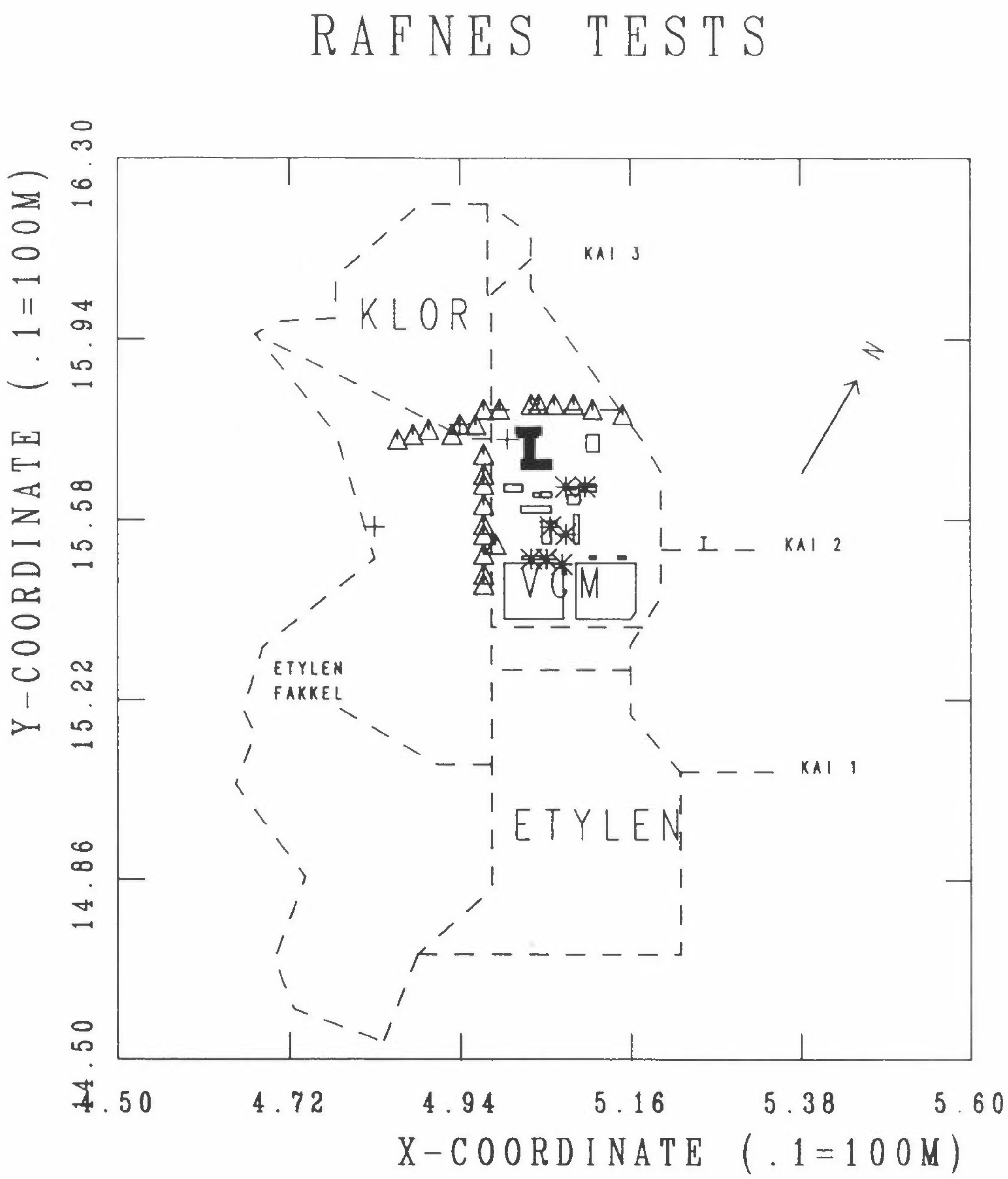

Figure 2: A site map showing the layout of the Rafnes petrochemical factory, including the VCM area, the chlorine (KLOR) area, and the ethylene (ETHYLEN) area. The symbols are described as follows: the meteorologiacl tower (T), roadway (---), SF_6 release points (*), syringe samplers (Δ), and the fast response SF_6 analyzer(+).

tests (Tests 7 and 12) the fast-response analyzer was located in the parking lot of the Verksted-Kantine-Kontrollrom (VKK) building which is shaded black in the figure.

The 15-min average samples were collected using portable, automatic syringe samplers. Samples were analyzed within 24 hours using gas chromatography. For more information on sampling and analytical techniques at NILU, see Heggen and Sivertsen (1983).

The NILU fast-response analyzer and the data acquisition system were installed in the back of a van. The analyzer was manufactured by Scientech, Inc., and the design is based upon the design of Benner and Lamb (1985). The instrument response time of this analyzer is 0.4 s, and the detection limits range from approximately 12 ppt at three times the instrument noise level to greater than 10000 ppt. During the Rafnes study, polyethylene tubing, 155 cm in length, was used as a sampling line, and the delay time through the tubing and the analyzer was 20 s. Calibrations were performed using standards of 220 ppt and 4800 ppt, and baseline checks were performed using a cylinder of pure air. Raw data were collected at a rate of 1 Hz using a Squirrel Data Acquisition system. The data were transferred to a portable PC for storage.

Data processing was performed on the NILU Norsk Data computer. The response of the fast-response SF_6 analyzer obeys a power-law releation between voltage and concentration, and the power-law coefficients were calculated from the calibration data. After converting the time series data to real units, the 15-minute blocks were selected which corresponded to the sampling periods of the syringe samplers. The data processing program used in the Rafnes study is listed in Appendix A.

4 RESULTS

Figures 3-11 contain the results for each test period in which data were available, including a site map showing the location of the fast-response analyzer with respect to the layout of the Rafnes plant and with respect to the release points and sampling points, a traverse showing a crosswind concentration profile of the 15-min average plume, the concentration time series measured at a fixed receptor, the probability distribution of the near-instantaneous concentration data, and the autocorrelogram of the time series.

On the site maps, the symbols are as referred to in Figure 1: the fast-response analyzer (+), the release points (*), and the 15-min average syringe samplers (Δ). The 15-min average plume is depicted, where the number of x's above each sampler represents the magnitude of the concentration (in intervals of 200 ppt), while the orientation of the x's correspond to the direction of the mean wind. In addition, the mean wind speed and direction are shown on each site map, where an arrow points in the direction the mean wind, and the length of the arrow represents the mean wind speed. The magnitude of the wind speed is also noted, in addition to an arrow which points north (N).

The crosswind profiles of the 15-min average plumes are also on a separate figure, where distance on the x-axis refers to the distance along the sampling traverse, and concentration on the y-axis is in units of micrograms/cubic meter. Note: left-to-right on the concentration profile always corresponds to left-to-right on the sampling traverse, with the exception of Test 7, where left-to-right on the concentration profile corresponds to lower-to-upper on the sampling traverse oriented parallel to the roadway. In the figure, the triangle symbols denote the measured 15-min average concentrations, whereas the smooth curve represents a Gaussian best-fit to the data.

Measurements from the fast-response analyzer are shown in the concentration time series plots. The sampling rate was 1Hz, thus the data for each 15-min period consist of 900 points. The time average concentration is shown on the graph as a horizontal, dashed line. The magnitude of the mean concentration (and the standard deviation) are also indicated in the figure.

In the final two figures, the concentration time series were analyzed in terms of the cumulative probability distributions and the autocorrelograms. Probability in this work is defined as the probability that the time series contained values less than or equal to the concentration on the y-axis. (These probability distributions were constructed using the fortran program PROBSQ listed in the appendix). Several concentration fluctuation statistics may be inferred from the probability distribution, including the intermittency factor and the peak-to-mean ratio: the intermittency factor is the probability that the concentrations are greater than zero, and the peak-to-mean ratio is the mean-normalized concentration corresponding to the 99th percentile.

The autocorrelograms were produced using AUTOSQ (also listed in the appendix), where the x-axis is lag time, and the y-axis is the autocorrelation coefficient. Autocorrelation analysis is a useful means of evaluating important time scales in a concentration time series, and at least three time scales may be apparent in the autocorrelograms. First, the high-frequency concentration fluctuations cause the correlation coefficient to fall off rapidly (with increasing lag time) from an initial value of 1.0. Second, because concentrations are correlated within the plume, the correlation coefficient continues to decrease with increasing lag time, but more slowly than the initial, sharp fall-off from the high-frequency fluctuations. Third, as the concentrations correlate and de-correlate during multiple plume events, meander time scales may be defined as the lag time between peaks.

Table 4 contains the concentration fluctuation statistics for each 15-min period during the Rafnes Study, including the mean concentration (C), the standard deviation (σ_c), the concentration intensity (i), the intermittency factor (I), the absolute maximum concentration (C_{max}), the concentration at the 99th-percentile (C_{99}), the concentration at the 50th-percentile (C_{50}), the peak-to-mean ratio as defined from C_{99} (P/M), and the ratio of C_{50} to the mean concentration (C_{50}/C). The final column in Table 4 corresponds to the mean concentration normalized by the SF_6 release rate (C/Q). The results from this study are discussed below: first, as an overview of these statistics, and second, in detail on a case-by-case basis to try to explain and understand the time series data.

Overview

The mean concentration during the test periods ranged between 54 and 2788 ppt, with the lowest mean observed in Test 12A, and the highest in Test 2A. Variability about the mean in each test is represented by the intensity, and with the exceptions of Tests 6A and 12A, the intensity was less than 1.0 for all cases. The lowest intensities were observed during the tests in which the intermittency factor was 1.0, while the highest intensities were observed during the tests in which the intermittency factor was less than 1.0. The highest concentrations, both absolute and at the 99th percentile, were observed during Test 2, and the lowest during Test 12. The difference between the absolute maximum concentration and the concentration at the 99th percentile reflects the slope of the concentration distribution at the upper limit, and as seen in the data, the difference ranged between 62 ppt in Test 2A and 764 ppt in Test 4A. The highest peak-to-mean ratio occurred during Test 12A, but during approximately 59% of the tests, the peak-to-mean ratio was less than 2.0. If the ratio of the concentration at the 50th percentile to the mean concentration is less than unity,

Table 4: Concentration Fluctuation Statistics.

TEST	C	σ_c	i	I	C_{max}	C_{99}	C_{50}	P/M	C_{50}/C	C/Q
	ppt	ppt			ppt	ppt	ppt			ppt/g/min
2A	2788	1081	0.39	.987	4503	4441	3076	1.6	1.1	285
2B	2511	1144	0.46	.998	4307	4162	2823	1.7	1.1	257
3A	1788	453	0.25	1.00	3177	2836	1782	1.6	1.0	183
3B	1727	461	0.27	1.00	2890	2739	1718	1.6	.99	177
4A	1501	381	0.25	1.00	3425	2661	1492	1.8	.99	384
4B	1713	384	0.22	1.00	2780	2573	1689	1.5	.99	438
5A	1351	393	0.29	.999	2665	2456	1293	1.8	.96	173
5B	1095	513	0.47	.967	2321	2183	1179	1.1	1.1	140
6A	249	312	1.25	.824	2472	1869	114	2.0	.46	64
6B	552	490	0.89	.923	2337	1953	458	3.5	.83	141
7A	457	434	0.95	.736	1611	1384	333	3.0	.73	58
7B	450	409	0.91	.740	1339	1217	341	2.7	.76	58
8A	2061	587	0.28	1.00	4181	3647	2009	1.8	.97	352
8B	2037	516	0.25	1.00	3499	3186	2141	1.6	1.1	348
11A	616	533	0.87	.899	2239	1990	481	3.2	.78	105
11B	486	430	0.88	.857	2230	1642	383	3.4	.79	83
12A	54	132	2.44	.808	979	721	12	13.4	.22	9

where:

C=mean concentration
σ_c=standard deviation
i=concentration fluctuation intensity
I=intermittency factor
C_{max}=absolute maximum concentration
C_{99}, C_{50}=concentrations at 99th- and 50th-percentiles
P/M=peak-to-mean ratio (C_{99}/C)
C/Q=mean concentration normalized by the SF_6 release rate

it is likely that the distribution is weighted toward low concentrations, and a ratio greater than unity indicates a distribution that is weighted toward high concentrations. Again, 59% of the experiments had ratios which were approximately 1.0, whereas the remaining ratios were between 0.22 and 0.83. The normalized mean concentration (C/Q) is included in the table because the release rate varied among the experiments, and C/Q was lowest during Test 12A, and highest during Tests 4 and 8.

<u>Test 2</u>

During Test 2, the mean winds blew from the southeast, and the mean wind speed increased from 5.6 m/s during experiment 2A to 7.4 m/s during experiment 2B. As shown in Figure 3, the release configuration consisted of five point sources, four of which were at ground level, and one of which was elevated (z=15 m). The maximum 15-min average concentration along the sampling arc was approximately 26% higher during Test 2A than during Test 2B, but the position and overall shape of the mean plumes were quite similar.

The mean concentration and the peak concentrations measured at the fast-response analyzer were also higher during Test 2A than during 2B. The low-frequency component in Test 2A is likely the result of wind motions which caused the instantaneous plume to meander across the measuring point (i.e., see Peterson et al., 1990), and it is significant that the lower concentrations of the meander component do not reach zero because this suggests that the scales of the meandering motions were not larger than the scale of the instantaneous plume.

A low-frequency component is also seen in the concentration data for Test 2B, but the effect is less visible amid several sharp transitions in which the concentration fluctuated within a few seconds over a range of approximately 3600 ppt. Sharp transitions such as those measured in Test 2B are not believed to be to be typical features, at least not for isolated plumes

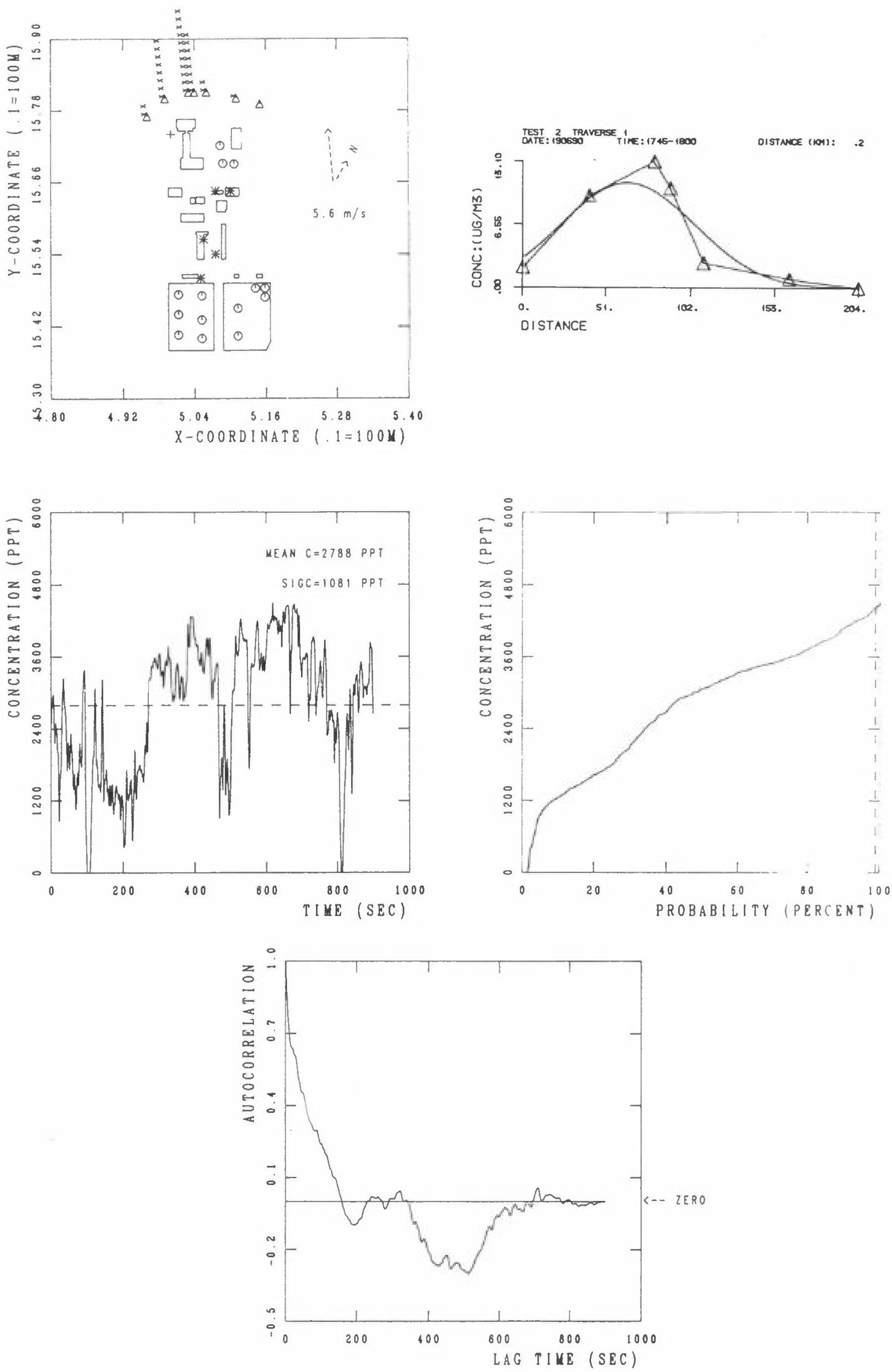

Figure 3a: Tracer results from Rafnes Test 2A, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

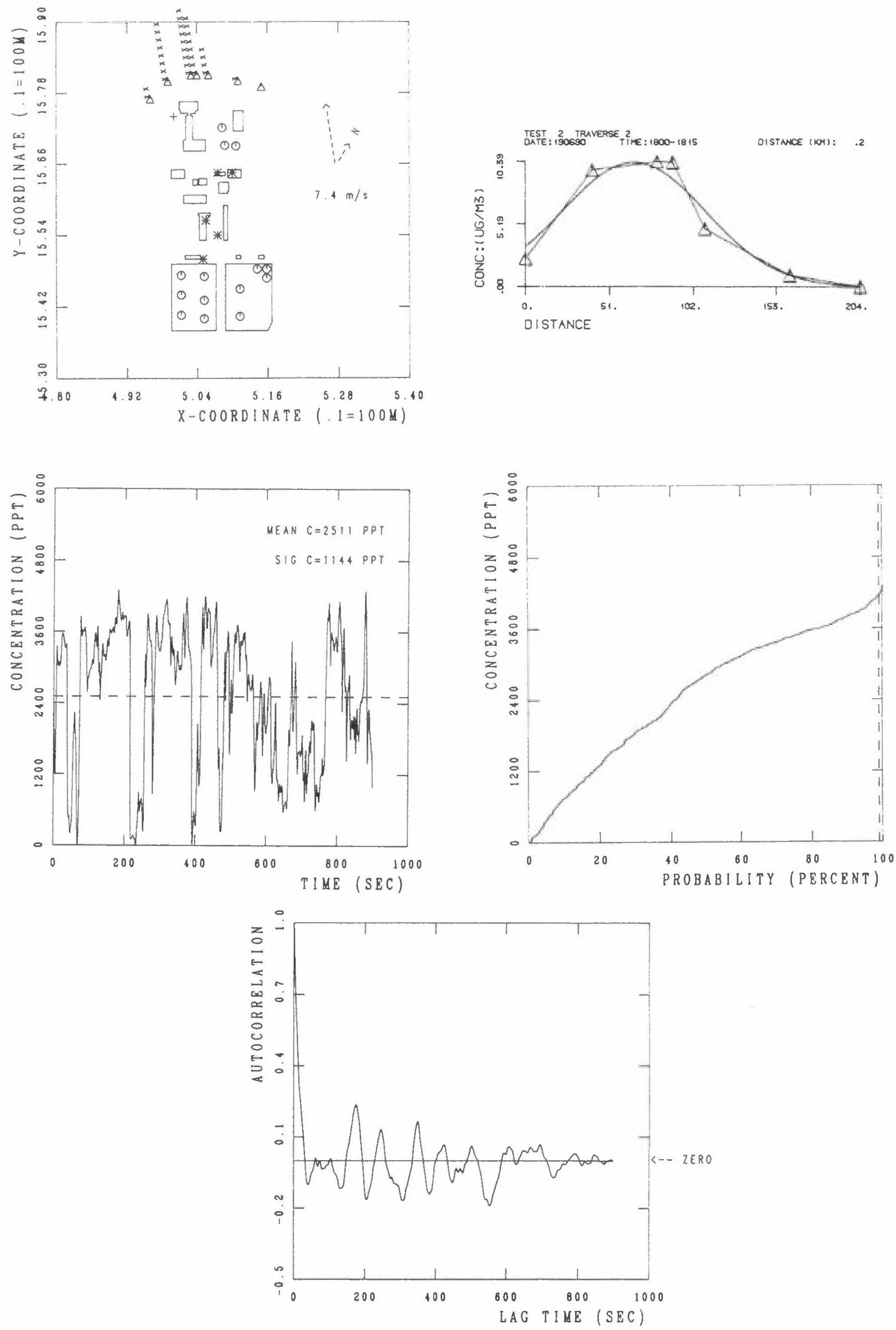

Figure 3b: Tracer results from Rafnes Test 2B, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

amid non-complex terrain. In this case, it is uncertain as to the origin of the dramatic fluctuations, but it is possible that the low concentrations were the result of ocassional bursts of clean air from flow which was being channelled down the roadway. Also, considering the irregular size and shape of the VKK building, it is also possible that temporal characteristics of the separation of the flow about the VKK building may have been affected by the instantaneous wind fluctuations. For example, perhaps there was a critical approach angle between the wind and the orientation of the buildings such that when this critical angle was exceeded, most of the SF_6-laden flow was transported along the northeastern of the VKK building, instead of along the southwestern side where the analyzer was located. (Unfortunately, without detailed wind data to describe the instantaneous wind field, it is not possible to test these theories in more detail).

The cumulative frequency distributions for the concentration time series of Tests 2A and 2B are steep and, in fact, nearly linear. Both distributions are more flat than distributions for Gaussian (white noise) data (i.e., in such case, and for the mean and standard deviation statistics measured, the concentrations at the 99th percentile, or $C+3\sigma_c$ would have been approximately 6000 ppt in both Tests 2A and 2B). In Test 2A, the slope is sharp at concentrations lower than 1000 ppt because concentrations near zero were rarely observed. The 'kinks' in the distributions are the result of the same portion of the plume getting sampled amid the meander cycles rather than getting a random sampling over the whole region of the plume. With the exception of the lower tails, the probability distributions in Test 2A and 2B are nearly identical, so it is likely that the concentration distribution within the instantaneous plume did not change much within the 30-min period. However, as seen in the autocorrelograms, the time scales of the exposure were quite different. For example, the dominant meander time scales were approximately 300s and 700s in Test 2A, but 100s and 200s in Test 2B. This illustrates the importance of reporting time series data, not only in terms of the concentration statistics and probability distributions, but also in terms of the auto-correlations.

Test 3

Figure 4 shows the results for Test 3. The release configuration was the same as during Test 2, but the meteorological conditions were not identical: the mean wind direction was 12-15 deg more easterly; and the magnitudes of the wind fluctuations (as estimated by σ_θ) were approximately one-half the magnitudes during Test 2.

The locations of the mean plumes in Test 3A and 3B, as compared to the locations in Test 2, reflect the more easterly winds. Meteological conditions were nearly identical during the two 15-min periods, 3A and 3B, and the profiles of the mean plumes were also nearly identical. Furthermore, the statistics of the concentration time series at the fast-response analyzer were also nearly the same during Tests 3A and 3B; although as in Test 2, the higher mean and peak concentrations occurred during the 15-min period in which the mean wind speed was the lowest (Test 3A).

There was very little plume meander during Test 3. Although a low-frequency component is seen in the concentration time series, the amplitude of the meander was small such that the concentrations were rarely less than or greater than 1/2 or 2 times the mean concentration, and the amplitude of the largest low-frequency fluctuations was approximately one-half the the amplitude of the largest low-frequency oscillations during Test 2. Similar to the comparison of the wind direction fluctuations, the magnitudes of the concentration fluctuations (as estimated by σ_c) were approximately 1/2 as large as the concentration fluctuations observed during Test 2.

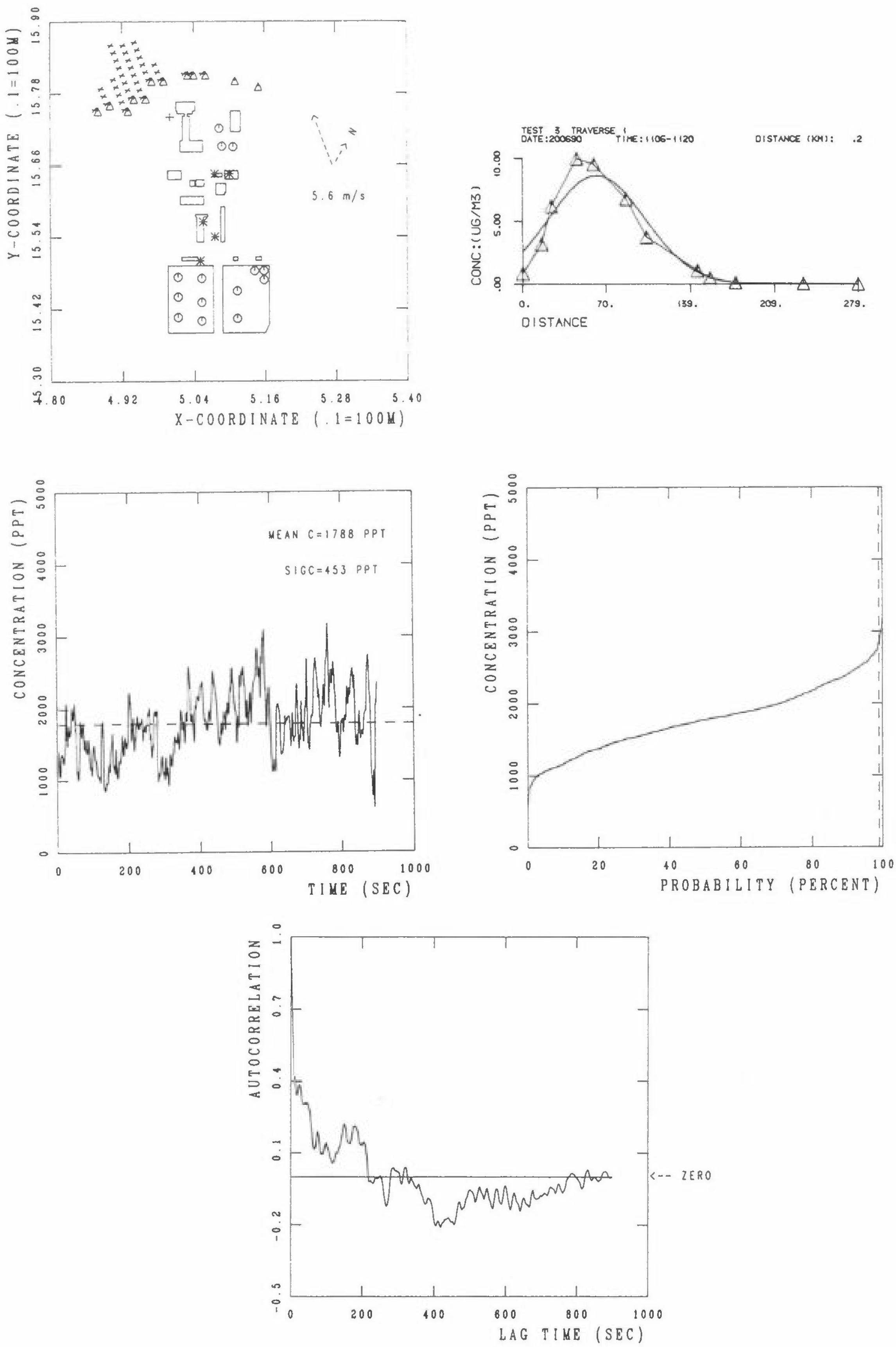

Figure 4a: Tracer results from Rafnes Test 3A, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

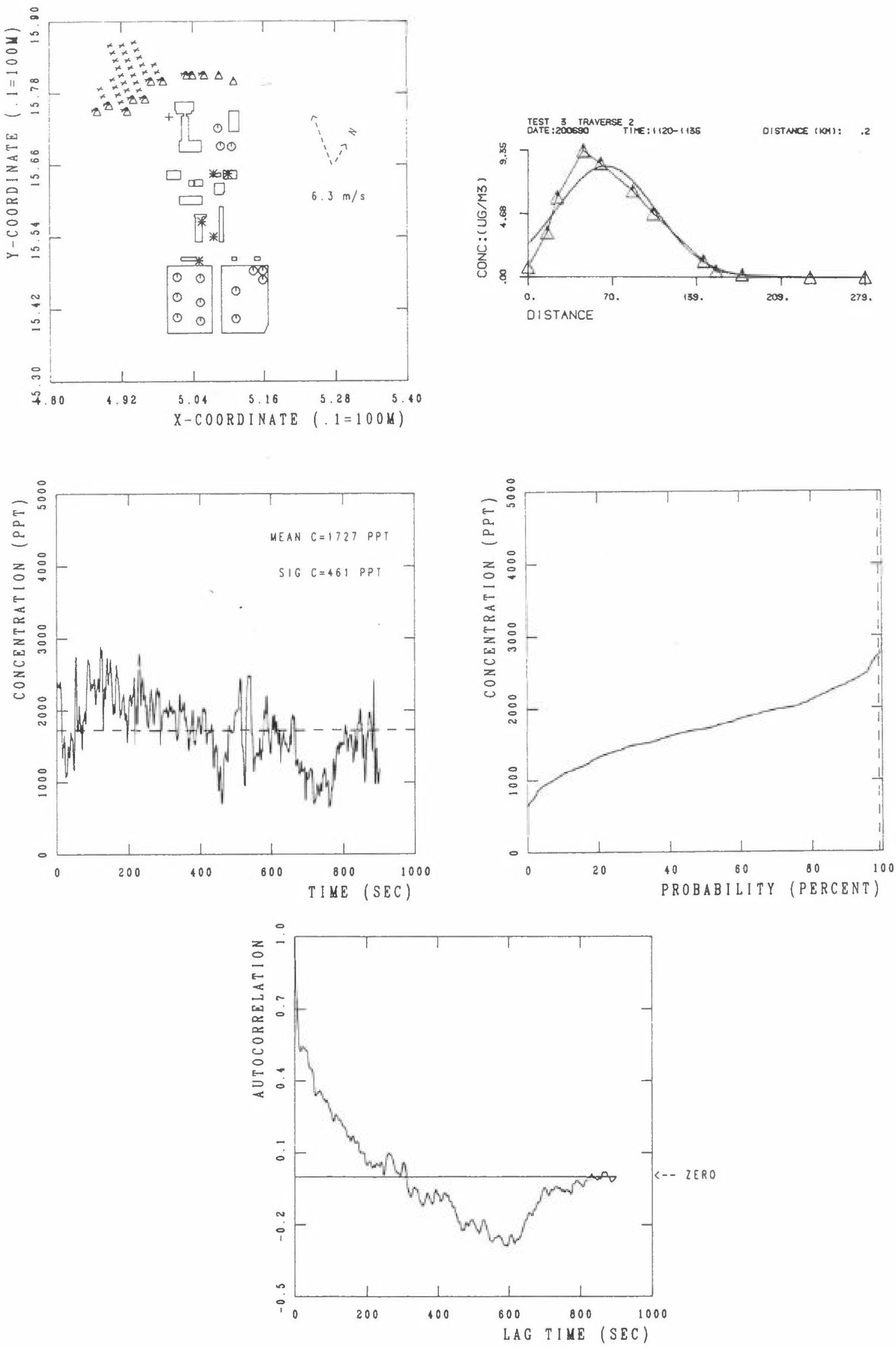

Figure 4b: Tracer results from Rafnes Test 3B, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

The concentration probability distributions for the data in Test 3 are flatter than the distributions in Test 2, and although the distributions are closer to distributions for random (white noise) data, the concentrations at the 1-percentile and 99th percentile are still higher and lower, respectively, than Gaussian distributions would indicate (i.e., if Gaussian, the concentrations at the 1-percentile would have been approximately 300-400 ppt at $C-3\sigma_c$, and the concentrations at the 99th percentile would have been approximately 3100 ppt at $C+3\sigma_c$).

Dominant meander time scales are not seen in the autocorrelograms for the concentration data in Test 3 as they were in Test 2. Some correlation is evident as a peak in 3A at a time scale of approximately 175 s, but the higher-frequency fluctuations (i.e., those fluctuations with time scales of order 20 s) are evident throughout the autocorrelogram and, thus, are not dominated by the effects of the larger-scales. Likewise, in Test 3B, the high-frequency fluctuations are a prominant feature on the autocorrelogram, unlike for example, in Test 2 where the high-frequency fluctuations of time scales in the 20-second range were smoothed out in the autocorrelograms.

Test 4

As seen in Figure 5, conditions during Test 4 were nearly the same as the conditions during Test 3, except the mean speeds were slightly higher, and the source configuration consisted of two rather than five release points. The mean concentration measured along the centerline of the mean plume was slightly lower in Test 4A than in 4B (i.e., approximately 60 ppt lower), and the mean concentration calculated from the fast-response data was also lower in Test 4A (i.e., about 200 ppt lower). Otherwise, the shape and location of the mean plumes were nearly identical for the periods during 4A and 4B.

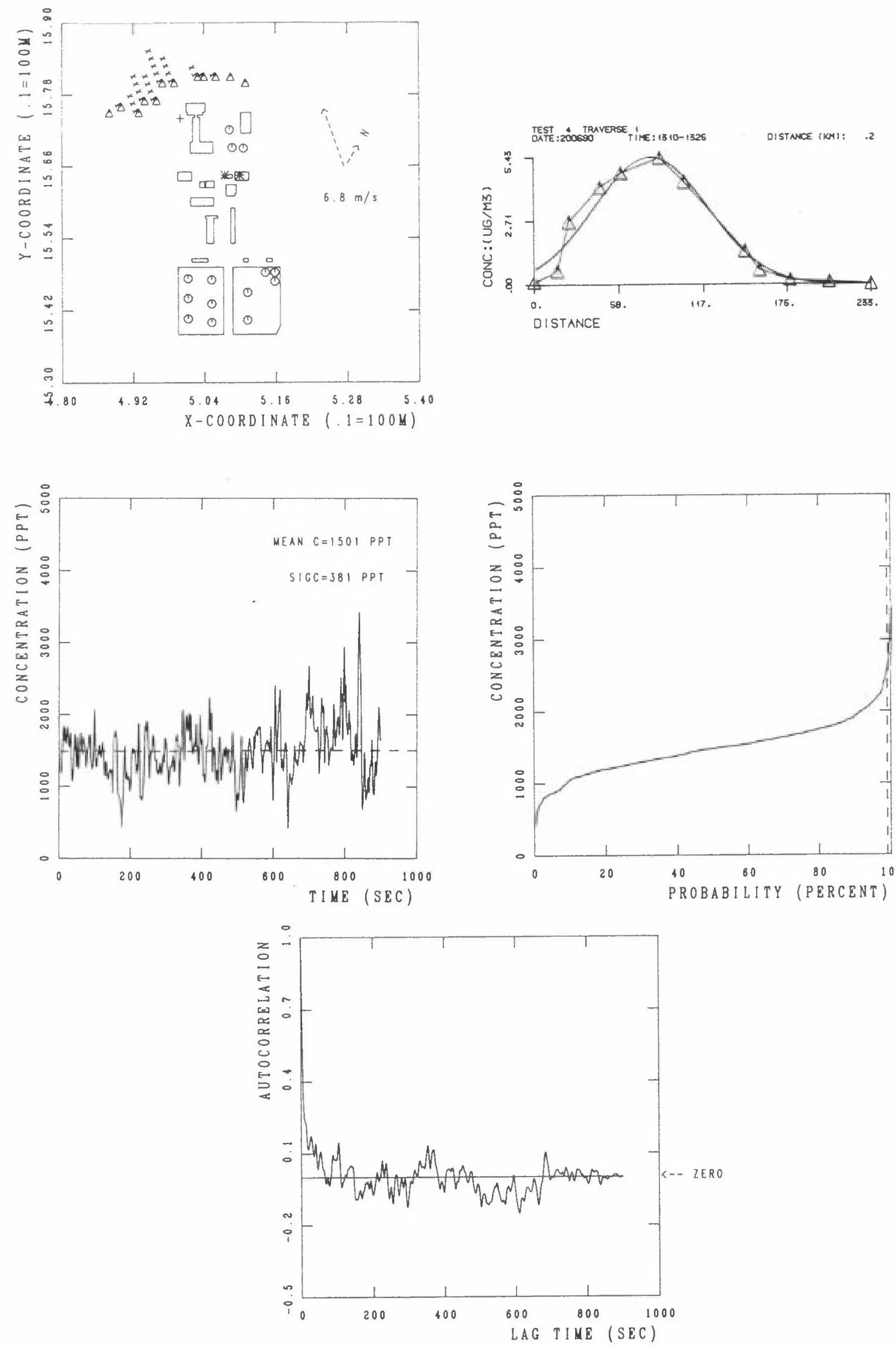

Figure 5A: Tracer results from Rafnes Test 4A, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

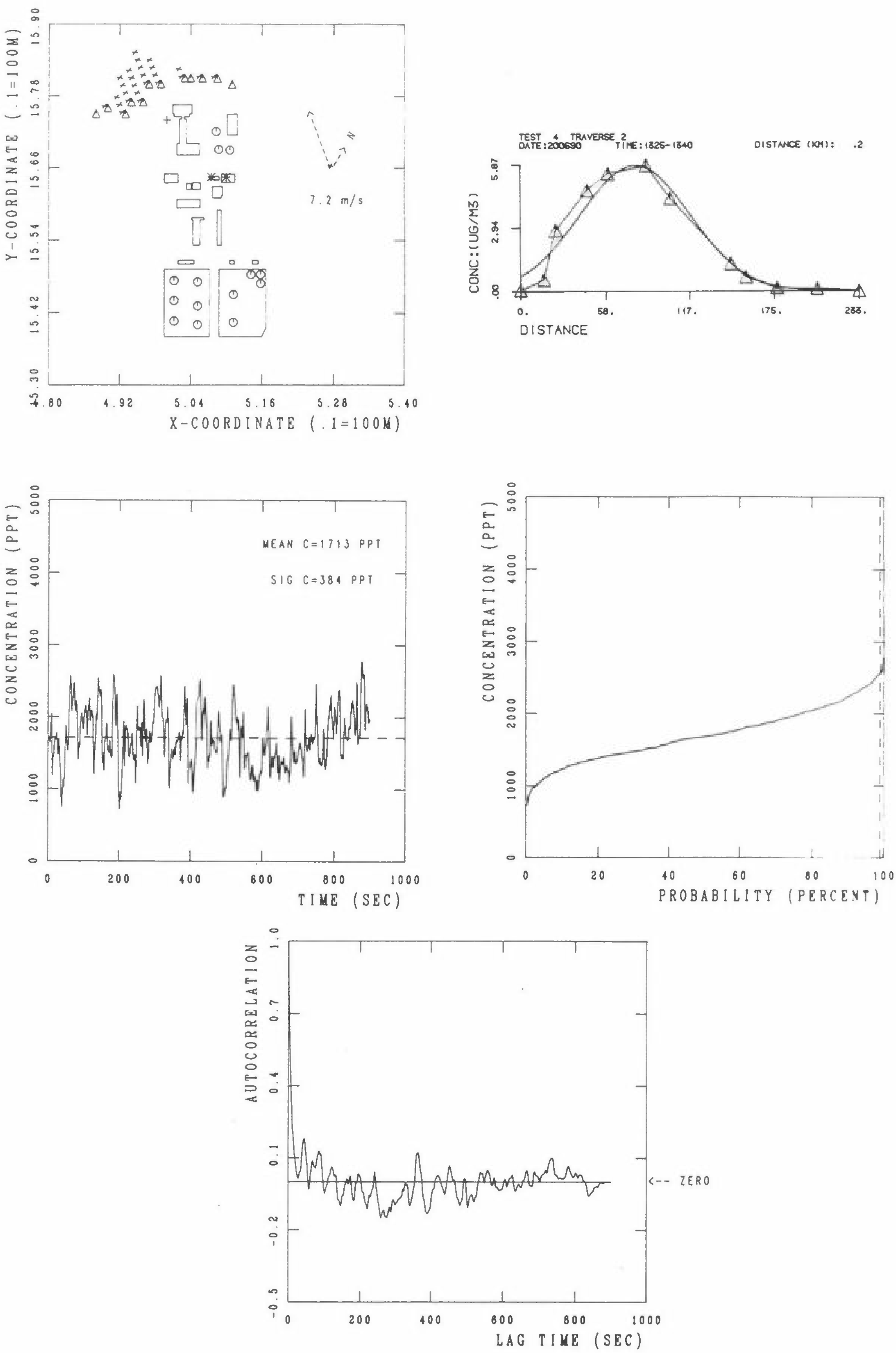

Figure 5B: Tracer results from Rafnes Test 4B, including a site map, the 15-min average plume profile along the sampling he traverse, tconcentration time series, the cumulative probability distribution, and the autocorrelogram.

The time series for test 4 are very similar to the time series for test 3. Although only 2/5 of the tracer in Test 3 was being released in Test 4, the magnitude of the concentrations were nearly the same in both cases, and the variability of the fluctuations was only slightly higher during Test 3 (i.e., σ_c was about 70 ppt greater). The wind conditions were nearly the same during Tests 3 and 4. This suggests that emissions from the furthest three release points (i.e., the three southernmost release points) probably had minor roles in determining the nature of the time series during Test 3.

Once again, the concentration probability distributions were very flat, and the distributions from Tests 3A, 3B, and 4B nearly collapse to a single curve, while the distribution of 4A falls slightly lower for concentrations greater than approximately 1250 ppt. However, as apparent in the autocorrellograms, the exposure during Test 4 had a weaker in-plume component than during Test 3; that is, the autocorrelation coefficient in Test 4 does not decay to a value less than zero as gradually as it does in Test 3, and most of the energetic fluctuations were of time scales 20-40 s. Especially at receptors located within a few hundred meters of a source, it is difficult to obtain concentration fluctuation data which are not dominated by large meander effects. Thus, the data from these tests are quite interesting as they contain information concerning in-plume variability, rather than meander-induced variability. Furthermore, the data for Test 4 are significant because the source configuration in this test consisted of releases in a single area of the plant in which hydrocarbon leaks are quite likely, and because meteorological conditions were relatively adverse, i.e., moderate wind speeds with very low cross-wind turbulence. In addition, because of the proximity to the control room and to the kantine, the analyzer was located in an area of the plant in which people may be directly exposed to the leaking gases. Thus the data from Test 4 may be used to examine potential health effects as a function of magnitude and duration of the concentrations during an accidental release of, say, deadly or toxic gases from an isolated point in the process.

Test 5

Figure 6 contains the results from Test 5. During this period, the winds were more variable than during Tests 3 and 4 (i.e., σ_θ was approximately a factor of 1.6 higher). Whereas the mean wind speeds in Test 5 were approximately the same as the mean wind speeds in Test 4, the wind direction in 5B was 5 degrees more southerly. SF_6 was released from four points in a source configuration which was the same as during Tests 2 and 3, with the exception of having only one of the two northern release points used in Test 4.

The profiles of the mean plumes were wider and more flat than during the other tests, and the mean concentrations on the centerline of the mean plumes were lower. In fact, the centerline concentrations during Test 5 were approximately the same as the centerline concentrations during Test 4 which had a total release rate of one-half as much.

During Test 5A, the mean winds nearly identical to the conditions in Test 4A, and once again, the concentration time series was extremely steady. However, the magnitude of the time-average concentration and the peak concentrations were 150 ppt and 205 ppt, respectively, lower than during Test 4A: an observation which may reflect an increase in the width of the instantaneous plume with increasing σ_θ. The mean concentration concentration in Test 5B is lower than in Test 5A, probably because the wind direction shifted such that the instantaneous plume meandered away from the sampling point during the experiment. This is seen in the time series of Test 5B where, after approximately 10 min, zero and near-zero concentrations were detected for a period of about 200 s before returning to the

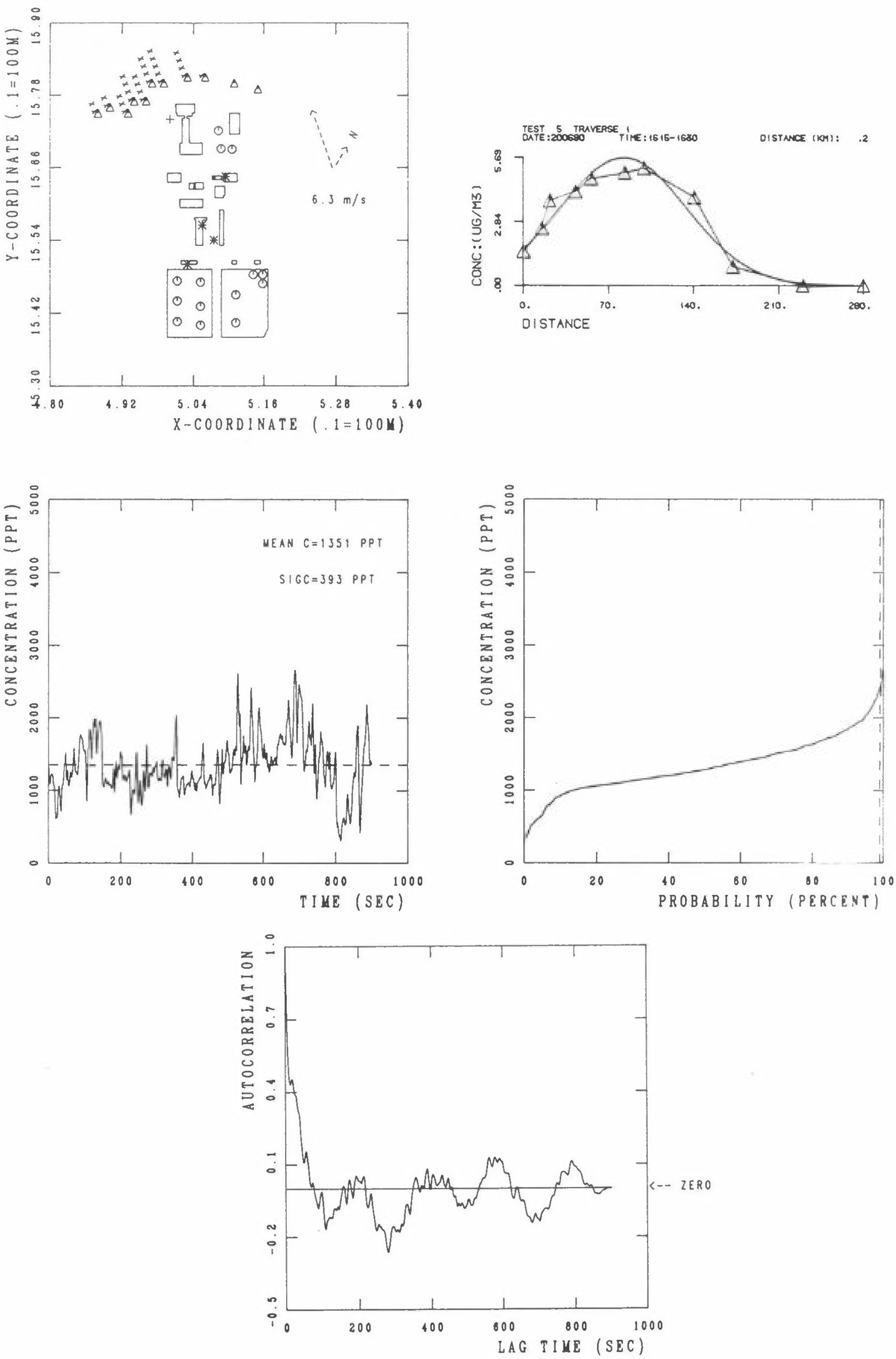

Figure 6A: Tracer results from Rafnes Test 5A, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

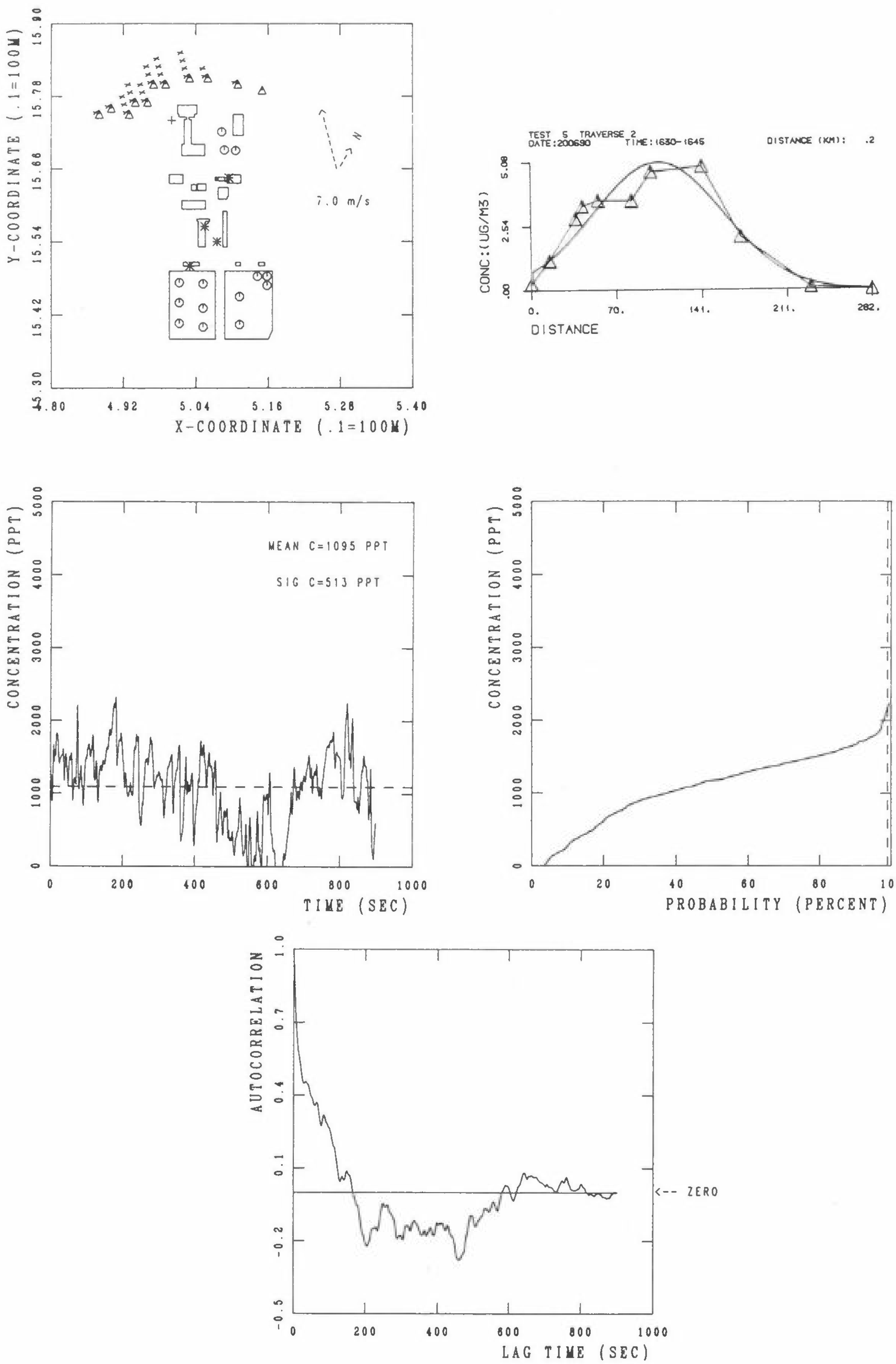

Figure 6B: Tracer results from Rafnes Test 5B, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probality distribution, and the auto-correlogram.

initial plume orientation. This plume meander probably coincided with southerly winds which caused the the mean wind direction during 5B to be 5 deg greater than during 5A.

Once again, the concentration probability distribution was very flat for the data collected during Test 5A, while during Test 5B, the effect of the meander is apparent in the lower quarter of the curve. That is, the steeper slope at low concentrations was the result of the relatively short period in which the low concentrations at the plume edge were sampled. The upper portion of the curves, however, for concentrations greater than approximately 1000 ppt, the curves nearly collapse and, thus, the structure of the instantaneous plume probably did not vary much within the 30-min period.

The autocorrelograms from Tests 5A and 5B illustrate the difference in dominant time scales during the two periods. A periodic motion with a time scale of approximately 200 s exists in the data for Test 5A, whereas for Test 5B, the larger-scale meander event caused the relatively weak secondary peak in the autocorrelation coefficient at approximately 600 s. Furthermore, the profiles of the mean plumes may be qualitatively explained on the basis of the nature of the concentration time series and the features in these autocorrelograms. The concentrations in Test 5A did not fluctuate much from the mean, thus the scales of the motions which caused the instantaneous plume to meander were smaller than the plume itself. In such cases, and when the meander is periodic rather than random, the profile of the mean plume may be either bimodal, or simply more flat than the Gaussian form as observed in Test 5A (the effects of periodic meander are discussed further in Peterson, 1989 and Peterson et al., 1990). However, when the time scale of a meander motion is long compared to the sampling time of the experiments, the receptors will not be exposed to an even sampling of the plume distribution; thus the mean plume may reveal a skewed sampling of the concentration distribution, and this is clearly illustrated in the concentration time series and in the non-Gaussian profile of the mean plume during Test 5B.

Test 6

During Test 6, the mean wind conditions were nearly the same as during Test 5, but as shown in Figure 7, only two of the four source points were used. These two source points were approximately 200 m from the fast-response analyzer (as compared to the two source points used in Test 4 which were approximately 125 m from the analyzer).

The time series in Test 6 are interesting because they were measured nearer to an 'edge' of the mean plume than in the other cases, and thus the concentrations were lower, and the intensities and peak-to-mean ratios were higher. It is important to note that the difference between the results for Test 6A and the results for Test 6B corresponded to a mean wind direction shift of 11 deg. The mean concentration during period 6A was approximately one-half the mean concentration during 6B, and in fact, concentrations greater than the mean were only observed periodically in correlated plume events during 6A, whereas during 6B, the signal was stronger and more stationary. This suggests that the meander component of the wind had a shorter time scale during Test 6A than during Test 6B, and because the amplitude of the concentration events in 6A appears to increase in time, the wind probably also had a low-frequency component which was longer than the sampling period. This explanation is supported by the profiles of the mean plume, where the profile is much flatter near the centerline in Test 6A than in Test 6B.

The concentration probability distributions in this test are unlike the distributions in Tests 2-5. The distributions are highly skewed toward low concentrations where the slopes of the curves are quite flat up to probabilities of about 80%, and the probability of concentrations greater than, say, 1000 ppt is

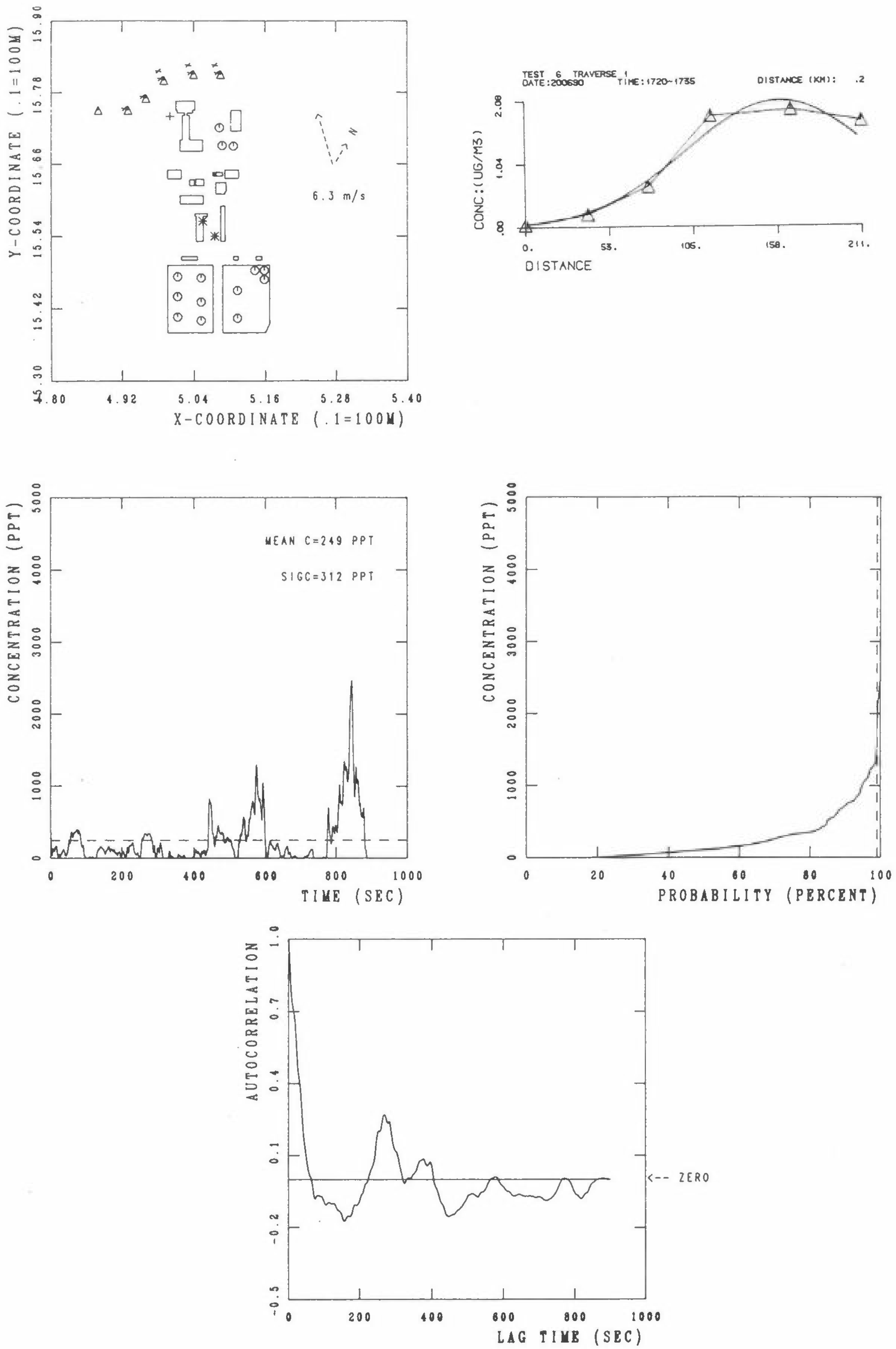

Figure 7A: Tracer results from Rafnes Test 6A, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

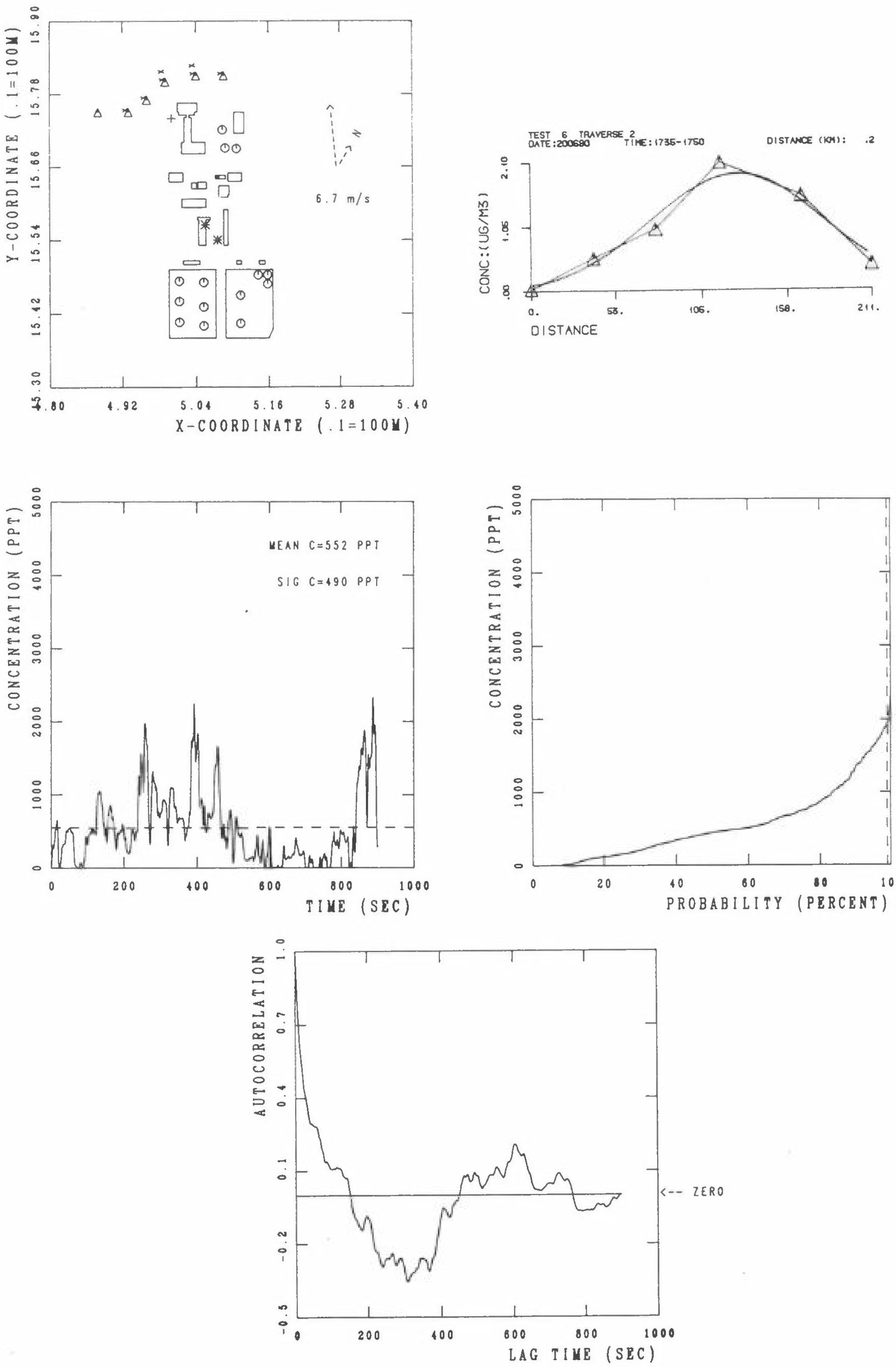

Figure 7B: Tracer results from Rafnes Test 6B, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

only ≈5-15%, as compared to ≈70-96% during Tests 2-5. These differences reflect the fact that the edge of the mean plume was sampled during Test 6 rather than near the centerline in the other tests.

The meander time scales, however, in the autocorrelograms are not unlike the meander time scales seen before: in Test 6A, the dominant time scale is about 200-300 s, and in Test 6B, the time scale is about 600 s. Thus, the nature of the meander probably did not change significantly between, for example, Test 5 and Test 6, and the major differences between the two periods are the results of the source-receptor configurations.

Test 7

The winds blew from the northeast during Test 7 as opposed to a southeasterly direction during Tests 2-6. Conditions for this experiment were not optimium because of high background levels of SF_6 and because of the location of the fast-response analyzer with respect to the sampling array. The high background levels were caused by an upwind SF_6 source located at a smelter, 5-6 km across the fjord. From the point-of-view of the fast-response analyzer data, the traverse of syringe samplers was approximately half-way between the source points and the analyzer; thus, the profile of the mean plume near the analyzer is not known. However, the concentrations in the Rafnes plume were higher than the background concentrations (i.e., the centerline mean concentration was 5-9 times higher than the background concentration), and the concentrations within the instantaneous plume were clearly resolved above the background using the fast-response analyzer. Furthermore, the data from this test are quite interesting because they illustrate the effects of plume meander where, in this case, the transport and growth of the plume was influenced more by turbulence induced from terrain effects, rather than in other cases where the plume was influenced more by turbulence and channelling effects caused by presence of large buildings.

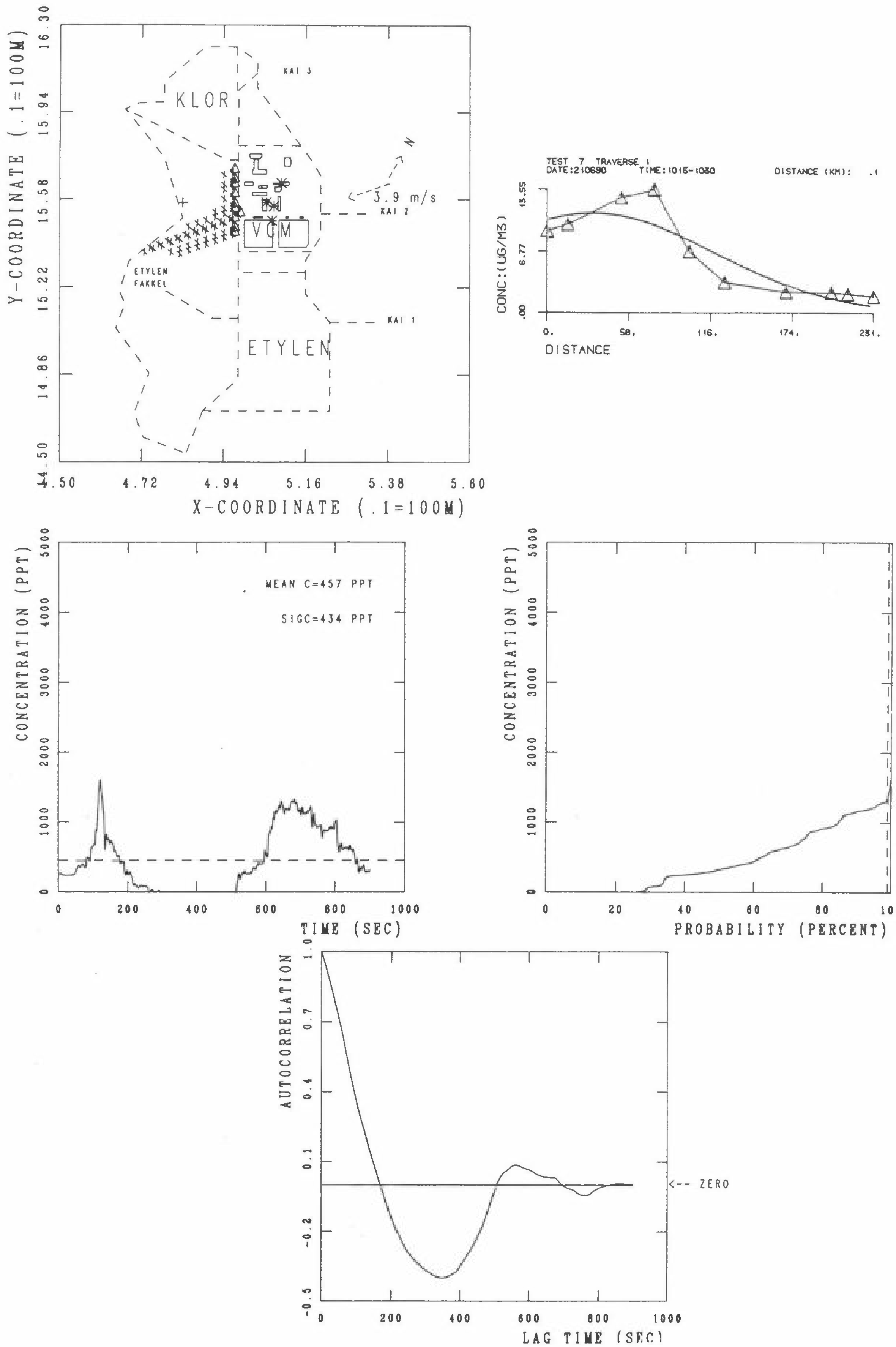

Figure 8A: Tracer results from Rafnes Test 7A, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

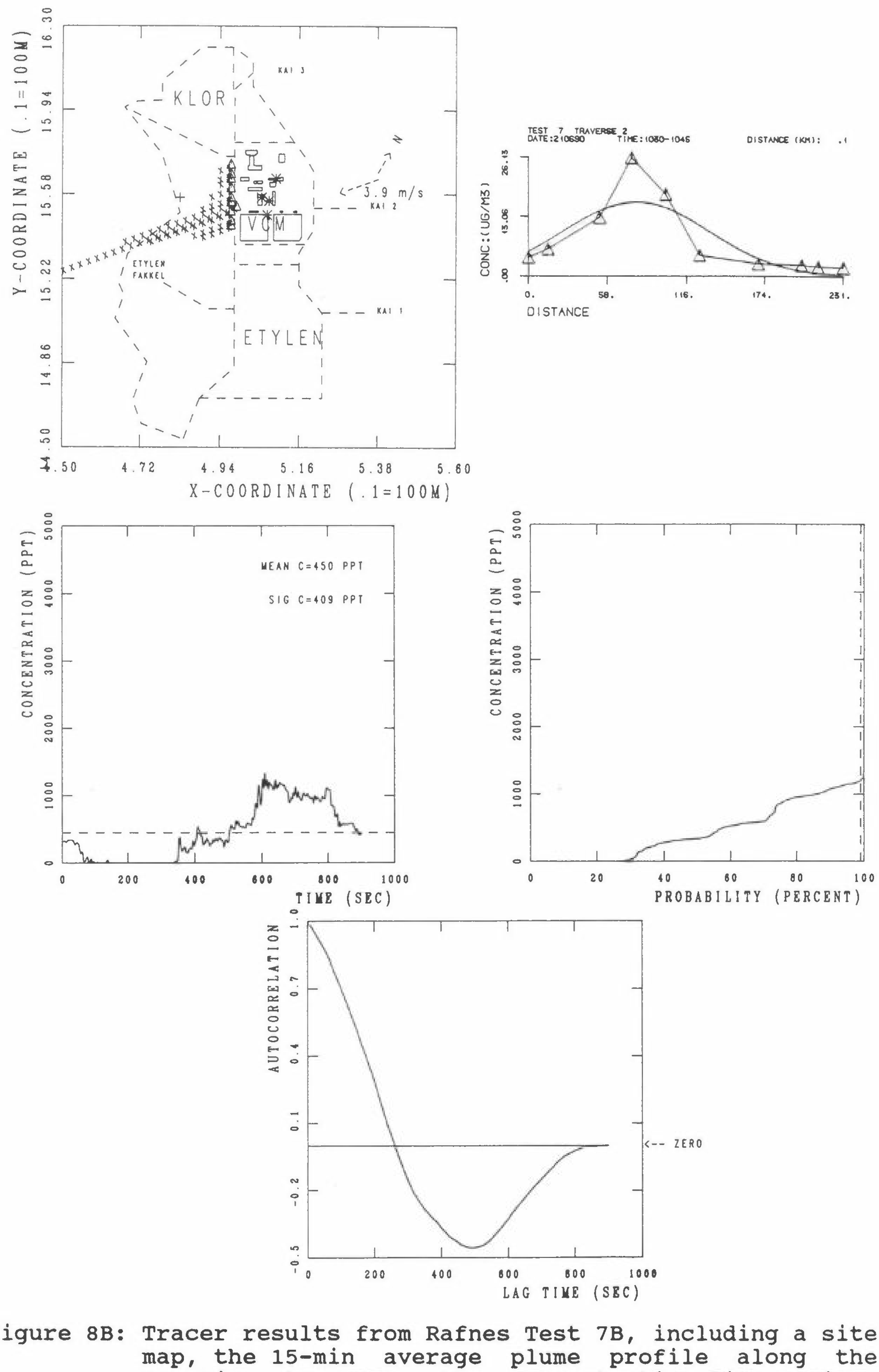

Figure 8B: Tracer results from Rafnes Test 7B, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

First, the time-averaged meteorological conditions were unlike those of the prior tests, not only in terms of the direction of the winds, but also the nature of the turbulence. In particular, the mean winds speeds were 1/3 to 1/2 as strong, and the standard deviation of the wind direction was 2.0-4.7 times larger. During Test 7, the mean plume measured along the roadway was wider than the extent of the sampling arc during both periods 7A and 7B. It was widest during 7A which suggests that the instantaneous plume probably meandered more during that period, and this agrees with the fact that the standard deviation of the wind azimuth was larger during Test 7A. The magnitude of the concentration along the centerline of the mean plume also reflects this, i.e., the magnitude was approximately twice as large during Test 7B as during Test 7A. (One thing to keep in mind concerning the plume profiles measured along the roadway is that the roadway was not alligned perpendicular to the mean wind direction; thus the measured concencentration distributions may be more skewed than the actual concentration distributions).

The concentration time series are also indicative of plume meander: the exposures consisted of intermittent periods of zero and non-zero concentrations as the plume meandered back-and-forth across the receptor. (Note: the background concentration was subtracted from the time series). As compared to the data of Tests 2-6, the effect of the meander is more dramatic. That is, in Tests 7A and 7B, the low-frequency component of was the main source of concentration fluctuations at the analyzer, whereas the high-frequency fluctuations were small in magnitude and contributed little to the overall variance of the time series. The fact that the high-frequency fluctuations were smaller in magnitude during this experiment than during other tests may reflect the in-plume mixing processes as a function of travel time from the source. For example, the source-to-receptor distance between the fast-response analyzer and the nearest release point was 183 m in Test 6, and 225 m in Test 7, whereas the respective mean wind speeds were 6.3 m/s and 3.9 m/s. Therefore, a lower-estimate of travel time between the source and the receptor is approximately 29 s for Test 6, and approximately 68 s for Test 7, and since the tracer within the instantaneous plume in Test 7 had approximately twice as much time to be thoroughly mixed before it reached the analyzer, it is not surprizing that more of the high-frequency fluctuations had been smoothed out. Also, in the other tests, the roughness elements between the source and the receptor consisted of a complex configuration of buildings and irregularly-shaped structures, whereas in Test 7, grassy hills and trees were the dominant roughness elements of the fetch between the source and the analyzer.

With the exception of the fine-scale kinks, the concentration probability distributions for Tests 7A and Tests 7B nearly collapse to a single curve; thus the overall exposure of the tracer did not change much at the analyzer within the half-hour period, and this suggests that the growth of the instantaneous plume probably did not change much during the two experiments. The nature of the meander, however, was different as seen in the autocorrelograms where the plume meandered with a time scale of approximately 600 s during Test 7A, but during Test 7B, the time scale of the meander was longer than the sampling period. Thus the concentration time series also support the results from the time-averaged plume profiles and from the wind data because all data indicate that the instantaneous plume meandered more during Test 7A than during 7B.

It is important to note that the mean concentration and all of the concentration fluctuation statistics listed in Table 4 are nearly identical for the time series data of Test 7A and 7B, and likewise, the concentration probability distributions are also nearly the same. However, the nature of the exposure was not the same in the two experiments, and once again, it illustrates the importance of reporting time scales for concentration fluctuation data. For some toxic pollutants, the life or health of a plant or an animal may be at risk when the organism is exposed to instantaneous concentrations above some threshold level, but assuming the organism has biological recovery processes, the overall damage will be a function of the time scale of these recovery processes in addition to the duration and time scales of the plume events (Griffiths and Megson, 1984). In such cases, the damage may be worse if the plume slowly meanders over the organism as, for example, in Test 7B, than if the organism is exposed to the same amount of pollutant but in two events, as in 7A, such that the plant or animal has a recovery period between events. This is a simple, hypothetical example, but it stresses the need to improve our understanding of instantaneous exposure at a fixed receptor as a function of the mean dispersion, the instantaneous dispersion of the instantaneous plume, and the meander of the wind field.

Test 8

Results from Test 8 are shown in Figure 9. The winds were from the southeast during periods 8A and 8B, and as in Test 7, the mean wind speeds (3.9 m/s and 3.7 m/s) were lower than during Tests 2-6, but the standard deviations of the wind azimuth (3.6 deg and 2.4 deg) were approximately the same as in Tests 2-6. The tracer was released from three of the four points used in Test 5, one of which was elevated (Z=15 m). The mean wind direction was also approximately the same as during Test 5, but the mean wind speeds in Test 8 were lower by a factor of 0.5.

Regarding the mean plume, the 15-min average distributions along the sampling traverses in Test 8 were similar in shape and location to the distributions measured in Test 5B, but the ranges of the concentrations were higher such that the highest concentration along the traverse in Test 8A was ≈40% higher than in Test 5B, and in Test 8A, ≈20% higher than in 5B.

Except for higher concentrations near the left end of the traverse, and a slight shift of the maximum concentrations to the right, the mean plume distributions during Test 8 were very

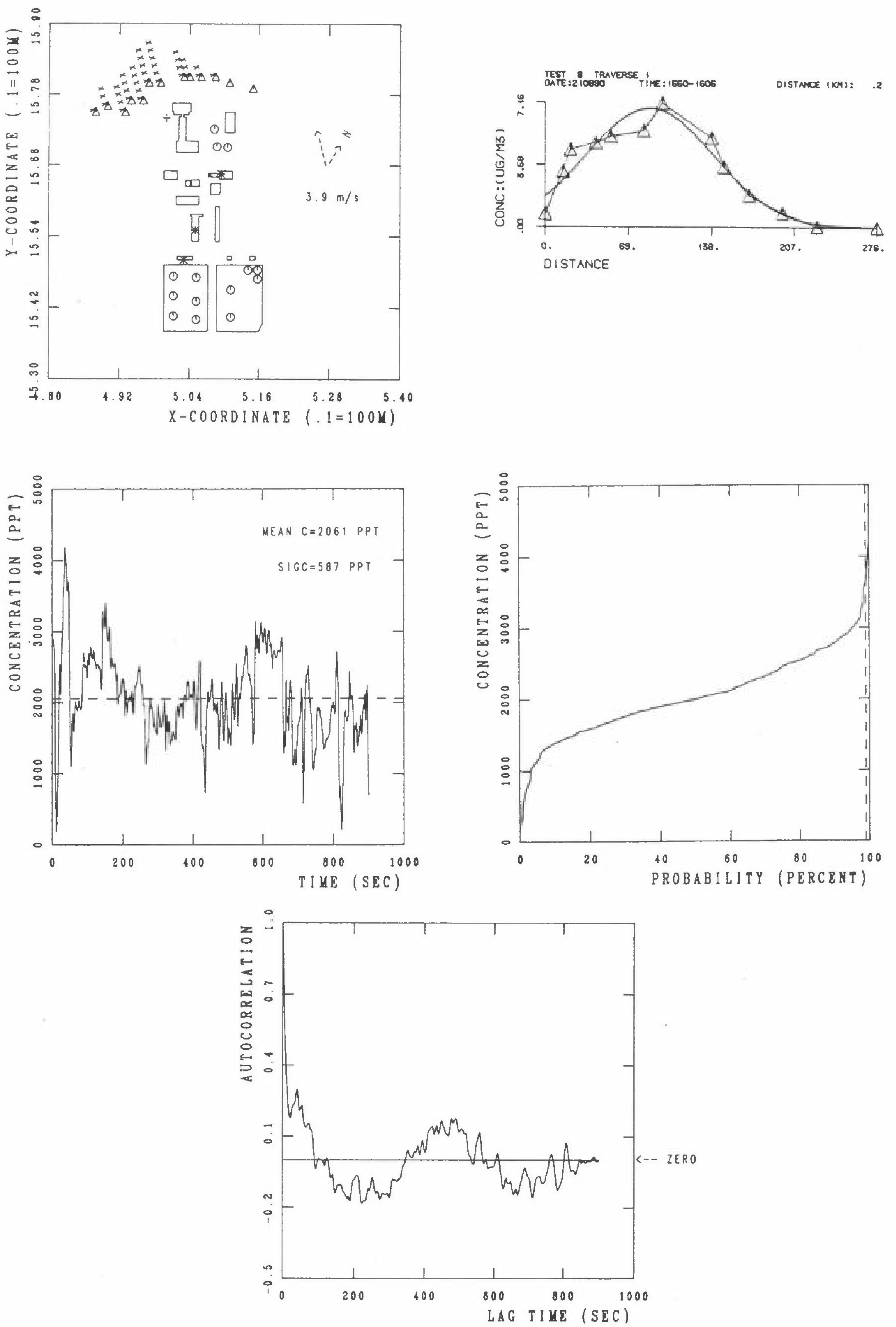

Figure 9A: Tracer results from Rafnes Test 8A, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

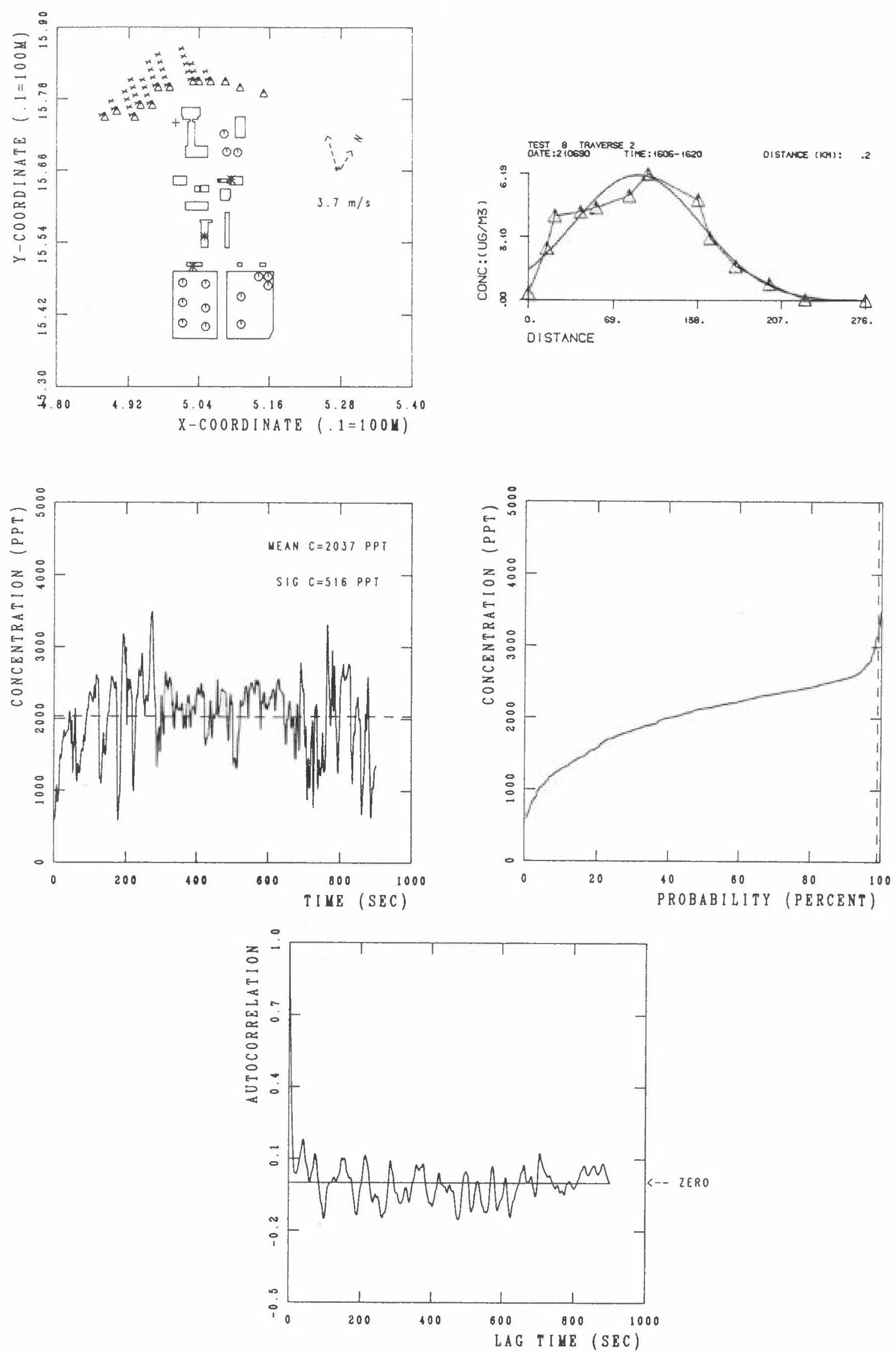

Figure 9B: Tracer results from Rafnes Test 8B, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

similar to the mean plume distributions during Test 4. The discrepancy between the profiles near the left end of the traverse can be explained in terms of the source configurations because during Test 4, SF_6 was not released from the southern release points. Likewise, the maximum concentrations downwind of the northernmost source point in Test 8 occurred to the right of the maximum in Test 4 because the winds were slightly more southerly (3-6 deg). It is significant that these mean plume profiles are similar because, in Test 4, the release rate of SF_6 from the northernmost source points was double the release rate at the northernmost point in Test 8, and the mean wind speed was approximately twice as high. The similarities between the mean plume profiles for Tests 8 and 4 also suggest that the plume from the elevated source in Test 8 probably did not impact the ground in the vicinity of the traverse or, hence, in the vicinity of the fast-response analyzer. Furthermore, it is important that the contributions from the northernmost and southernmost sources were still identifiable in the mean plume profiles because this suggests that, at least at this downwind distance, the instantaneous plumes from the northernmost and southernmost release points had not combined into a single instantaneous plume, and the reason that the mean concentrations downwind of the southern source were smaller than the mean concentrations downwind of the northern source is because the transport distance from the southern source was on the order of two-times farther than from the northern source.

In terms of the data from the fast-response analyzer, the mean concentration from the concentration time series was only slightly higher during Test 8A than during Test 8B, but both were nearly a factor of two greater than the mean concentration from the time series of Test 5B. This is significant considering the fact that the amount of tracer released during Test 8 was 25% less than the amount released during Test 5, and furthermore, because one of the releases was elevated, the tracer available for detection near the surface may have been even less at the near-source location of the fast-response analyzer. Therefore, the higher concentration levels at the analyzer and along the sampling traverse must have been the result of the lower wind speeds.

The results from this test are also interesting because they reveal insight on the rate of growth of the instantaneous plume. Considering the source configuration, the direction of the mean winds, and, of course, assuming that the plume from the elevated source was not continually impacting the fast-response analyzer at this near-source distance, the major source of SF_6 detected at the analyzer was probably the northernmost release point. Remembering that SF_6 was only released from the northernmost points during Test 4, it is interesting that there was little difference between the instantaneous time series measured at the analyzer during Tests 4 and 8. The magnitudes of the mean concentrations from the time series were approximately 300-500 ppt higher during Test 8, and likewise, the standard deviations were also higher, but the other concentration fluctuation statistics were nearly identical among Tests 4A, 4B, 8A, and 8B. If this suggests that the crosswind concentration distributions of the instantaneous plume from the northernmost release point were also quite similar in both cases, it also implies that the rate of growth of the instantaneous plume was approximately two-times faster during Test 4 than during Test 8 because although the release rate was two-times greater, the transport time was also a factor of two higher. (To study this further, simultaneous turbulence data are necessary to investigate the temporal and spatial characteristics of the turbulence which corresponded to the growth of the instantaneous plumes, but, unfortunately, such data are not available for the Rafnes Study. If experiments of this type are conducted in the future, it is critical to include real-time wind measurements near the inlet to the SF_6 analyzer and, if at all possible, near the source).

The shapes of the concentration probability distributions for Tests 8A and 8B were nearly identical to the shapes of the probability distributions for Tests 5B, 4A, and 4B. The curves from the various experiments were offset in the vertical scale by approximately the difference between the mean concentrations, and for Tests 8A and 8B, the slopes were steeper at extreme high and low concentrations.

The autocorrelogram for Test 8A shows a variety of time scales as a result of the irratic nature of the time series, but a dominant peak occurrs at approximately 500 s, and a secondary time scale occurres at approximately 25 s. On the other hand, for Test 8B, very little large scale meander is evident, and the dominant time scale of the fluctuations is about 25 s. These time scales are not unlike the time scales observed in the other tests. Without extensive turbulence data, it is impossible to determine whether these 25 s fluctuations are the result intermediate-scale meander, or whether they are simply the result of the mixing processes within the plume. However, it is significant that the time scales are approximately the same under wind speeds which are not necessarily the same because it suggests that the scales of the structures causing the fluctuations are smaller under lower wind speeds. In particular, a structure causing a 25-sec fluctuation in Test 8B should be about 60% as large as a structure causing a 25-s fluctuation in Test 4B, but as mentioned before, simultaneous wind data are necessary to quantitatively study this further because very little is known theoretically or experimentally about the internal structure nor the behavior of plumes on near-instantaneous time scales.

Test 11

The results shown in Figure 10 correspond to the experiments performed during Test 11 in which the winds were from the east. The source configuration was the same as during Test 8, but the mean wind speeds were slightly lower, and the standard deviations of the wind azimuth fluctuations were larger.

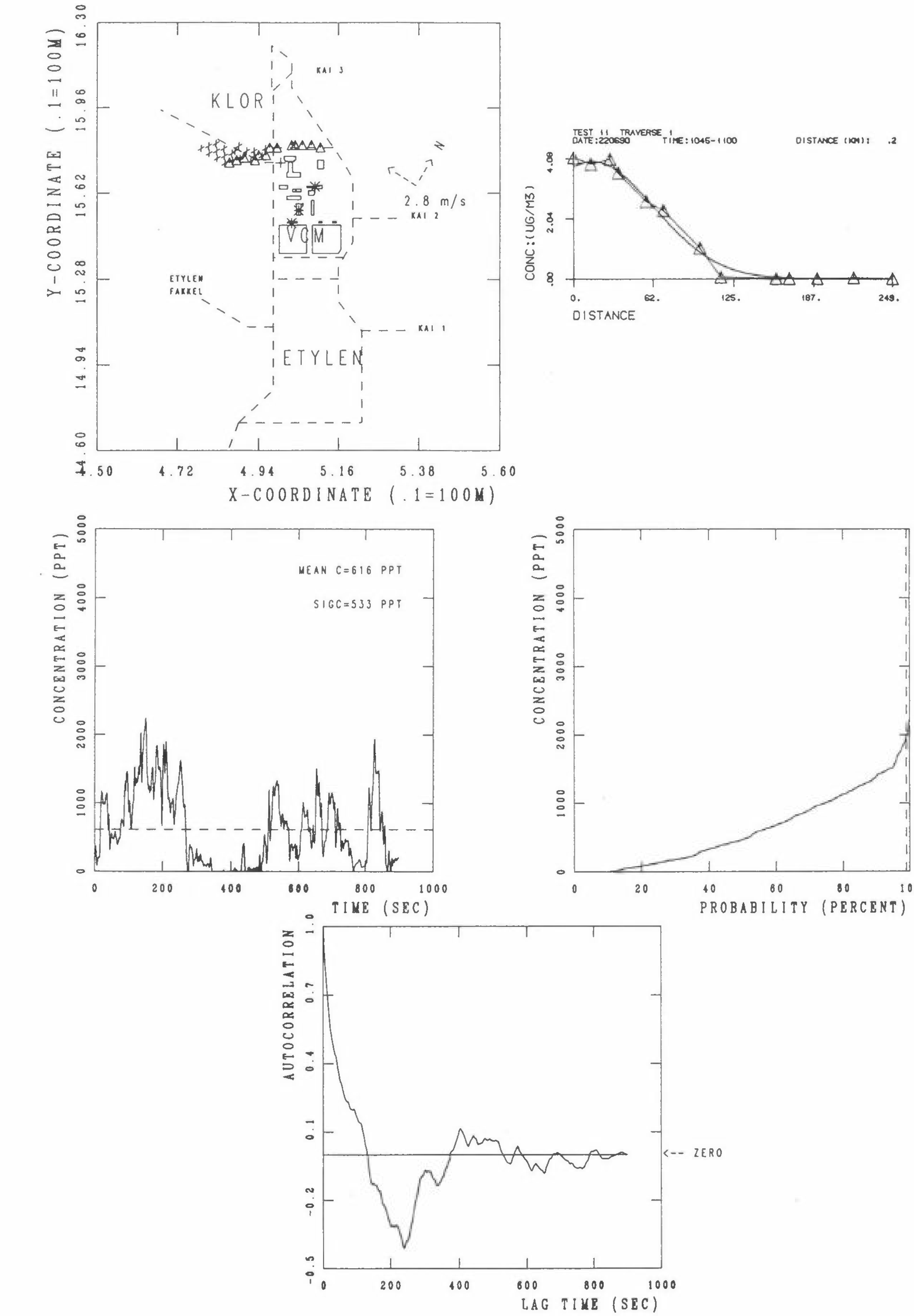

Figure 10A: Tracer results from Rafnes Test 11A, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

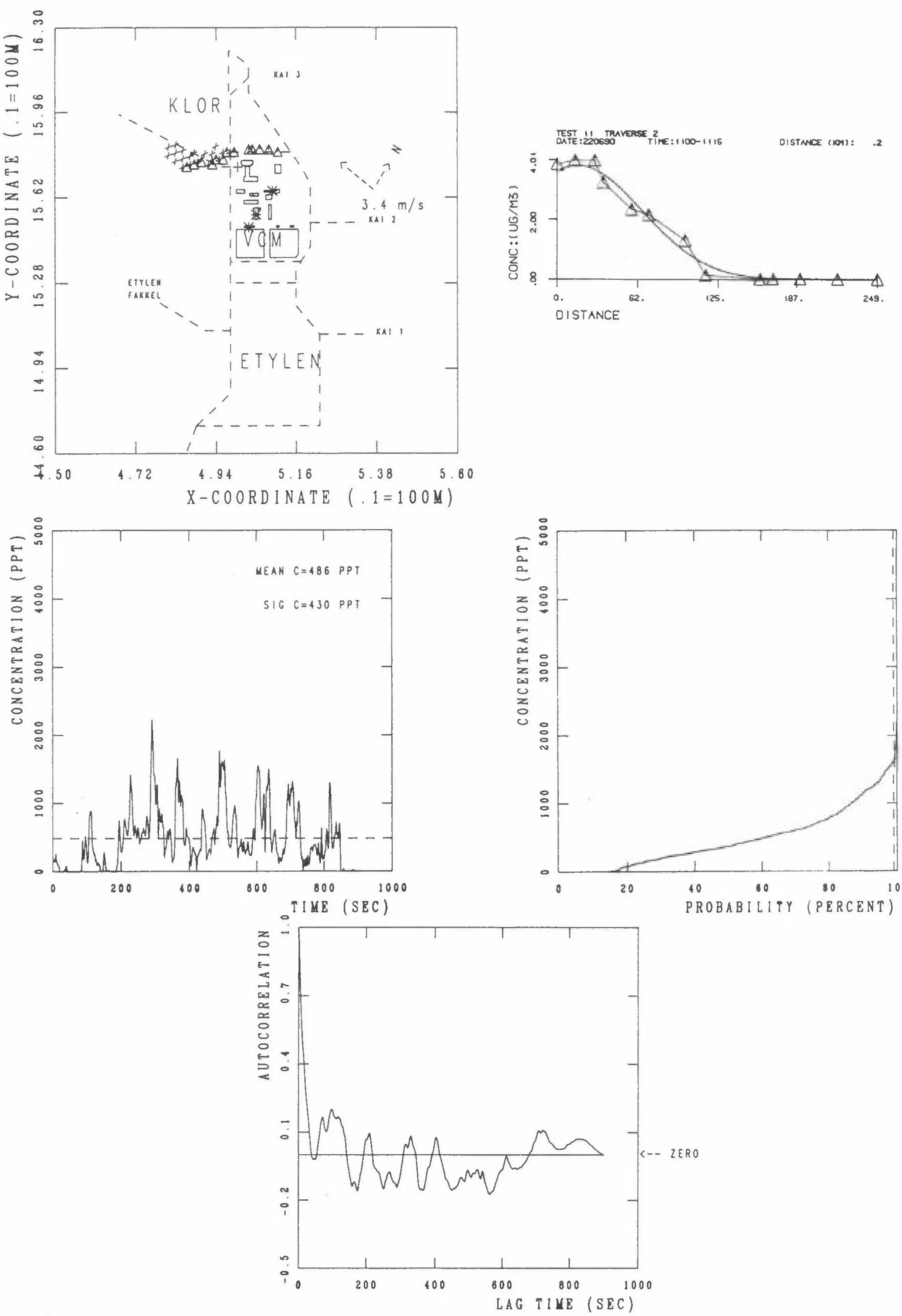

Figure 10B: Tracer results from Rafnes Test 11B, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

The sampling array captured approximately one-half of the mean plume in both 15-min periods. Although the mean wind direction was 7 deg more easterly during Test 11A, the profiles of the mean plume were nearly identical in 11A and 11B.

The fast-response analyzer was located near the northernmost edge of the mean plume. The time series, the concentration fluctuation statistics, and the concentration probability distributions were similar to the results from Test 6 which, in fact, also represented data collected near an edge of the mean plume. The low-frequency component in Test 11A indicates that the plume meandered more than in Test 11B, and from the auto-correlograms, the meander time scale during Test 11A was about 400 s, whereas during Test 11B, the dominant time scale was approximately 100 s.

Test 12

Figure 11 shows the results from Test 12A. (Test 12B is not included because the fast-response analyzer was used as a mobile unit during this period, rather than a stationary unit). The mean winds blew from the east, 2-7 deg more easterly than during Test 11; the wind speed was 1.5-2.1 m/s higher; and the standard deviation of the wind azimuth fluctuations was 0.6 deg smaller.

Because of the easterly winds during Test 12, even less of the mean plume was sampled by the sampling array than during Test 12. Likewise, the fast-response analyzer was located even closer to the edge of the mean plume. The mean concentration at the analyzer was 54 ppt, but as seen in the time series, most of the exposure was attributed to a single concentration event, centered about 500 s. The peak concentration within this event was a factor of two or three lower than the peak concentrations during Test 11; thus it is likely that the analyzer only sampled a fringe of the instantaneous plume.

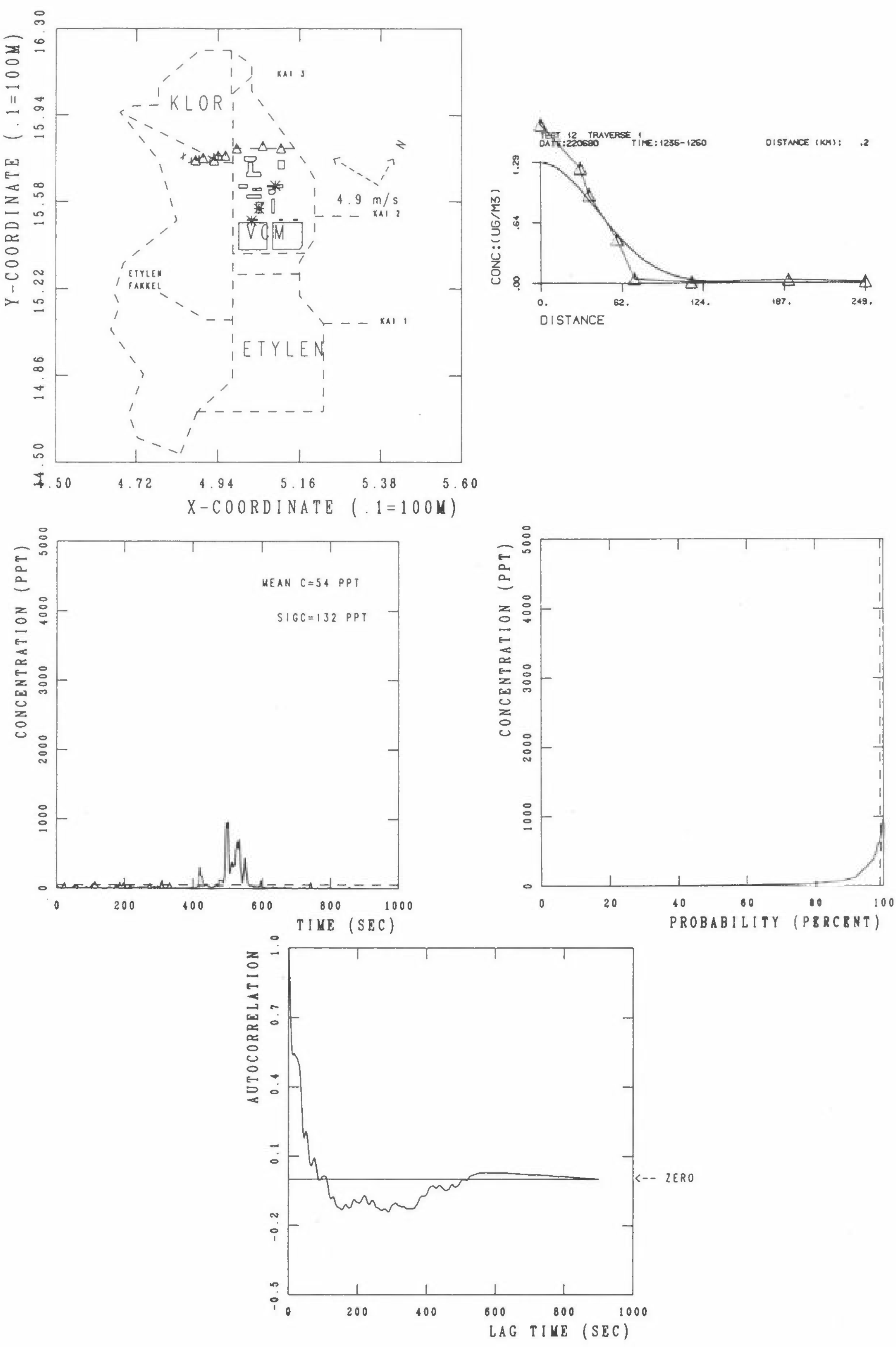

Figure 11: Tracer results from Rafnes Test 12A, including a site map, the 15-min average plume profile along the sampling traverse, the concentration time series, the cumulative probability distribution, and the autocorrelogram.

The concentration probability distribution is similar to the other distributions for data collected near the edge of the mean plume (Tests 6 and 11), except the slope of the curve is extremely flat for concentrations between 0 ppt and approximately 100 ppt, and the range of concentrations is less than 1000 ppt.

5 CONCLUSIONS

Twelve tracer tests were conducted in June 1990 to characterize the dispersion of gases from leaks in the VCM area of the Rafnes petrochemical factory. Sulfur hexafluoride gas was released at a continuous rate from 2-5 points throughout the process, while 15-min air samples were collected along cross-wind traverses. Gas chromatography was used to analyze the samples for mean SF_6 concentrations, and each test consisted of two 15-min experiments which were performed sequentially.

During nine of the twelve tests, a new fast-response SF_6 analyzer was used to measure near-instantaneous concentrations at a fixed point near the sampling array. Few data of this type are available elsewhere, and because this is the first data set acquired by NILU, this report may serve as a quide for analyzing and interpreting future data sets. Suggested below are a few important items to examine (and a few basic questions to ask) when analyzing the time series concentration data:

- The location of the analyzer with respect to the time-averaged plume.
  (Was it near the centerline? Near the edge?)

- The location of the analyzer with respect to the source.
  (Was it near the source? Within 200 m ? 1 km?)

- The magnitude and range of the concentrations.
  (Were they barely above the lower detection limit of the analyzer, or did they encompass a good portion of the detector range?)

- The time-average concentration and the standard deviation.

- The intermittency factor.
  (Was SF_6 detected throughout the sampling period, or was the exposure intermittent?)

- The concentration fluctuation statistics, including the intensity, and the peak-to-mean ratio.
  (Was the intensity >1? Was the peak-to-mean ratio >3?)

- The nature of the low-frequency component.
  (Did the low-frequency component extend to concentrations near or including zero?)

- The concentration probability distribution.
  (What is the form of the distribution? How steep is the curve, in particular, at high concentrations?)

- The time scales from the autocorrelograms.
  (Was the exposure periodic? How fast does the autocorrelation coefficient fall to zero? Do large-scale fluctuations or small-scale fluctuations dominate the autocorrelogram?)

With these items in mind, some important results from the Rafnes data set are as follows:

- In most of the Rafnes tests, the dominant source of SF_6 measured at the fast-response analyzer was the northernmost release point.

- As expected, the time series with the lowest mean concentrations (and the highest peak-to-mean ratios) were measured when the analyzer was located near an edge of the mean plume, while the highest mean concentrations (and lowest peak-to-mean ratios) were measured when the analyzer was located near the mean plume centerline.

- The peak-to-mean ratio was less than 3.0 during most of the Rafnes tests, except when the receptor was located near the mean plume edges, in which case, the ratio was as large as 13.4.

- The intensities were 0.22-0.29 for the near-centerline data, and in general, these are smaller than commonly observed at similar downwind distances for isolated plumes amid simpler terrain. The presence of large buildings probably induced a channelling effect in the advective flow, and thus the freedom of the instantaneous plume to meander was reduced. Also, it is possible that the overlapping of instantaneous plumes smoothed out some of the crosswind variation in the concentration distributionn.

- The intensities were 0.87-2.44 for the cases in which the analyzer was located near an edge of the mean plume. These values were also lower than usually observed, once again, probably as a result of the channelling effects.

- Likewise, the intermittency factors were larger than normal. Approximately 65% of the tests had intermittency factors greater than 0.90, and the lowest was 0.736. An intermittency factor of 1.0 has been seldom seen elsewhere for isolated plumes at a downwind distance of 200 m, except under very stable conditions because the instantaneous plume is still quite narrow at these distances and, thus, subject to meandering (Peterson, 1989). Again, the reason for the high intermittency factors is probably dispersion effects of buildings and the multiple point sources.

- The concentration probability distributions nearly collapse for the time series collected in similar regions of the mean plume. In particular, the distributions were similar when the analyzer was located near the centerline of the mean plume (Tests 3,4,5, and 8), and when the analyzer was located near the fringes of the mean plume (Tests 6,7,11, and 12).

- In general, there were few differences in the concentration statistics and in the concentration distributions between sequential 15-min periods (i.e., the A and B tests), but the time scales varied considerably in some cases. This suggests that the form of instantaneous plume did not vary much during a 30-min period, but the temporal characteristics of the meander did vary. This is important if the damage to a receptor is a function of both concentration and duration of exposure.

- In Test 7, the prevailing winds were not channelled along the major axis of the large factory buildings; instead, the winds blew across an open, hilly area of trees and grassland. In this case, meander was the dominant source of concentration fluctuations at the fast-response analyzer, and the magnitude of the high-frequency fluctuations was smaller than the magnitude of the high-frequency fluctuations in the other tests.

6 CONCLUDING REMARKS

Much work needs to be done to improve our understanding of concentration fluctuations, not only in terms of expanding the data bases to include instantaneous concentration measurements amid different meteorological conditions, source-to-receptor configurations, and terrain/roughness types, but also in terms of data analysis techniques. The most common technique to study the time series data has been purely statistical regarding the mean, the variance, the intensity, the intermittency factor, and the peak-to-mean ratio. Sawford (1987) has taken the statistical approach one step further by examining the 'conditional' statistics (i.e., the statistics for only the non-zero portion of the time series), and Hanna (1989) has examined the time series data using spectral analysis. Both of these approaches are useful in analyzing the data and are suggested approaches to take when analyzing future data sets; however, as yet, neither were used on the Rafnes data set. Without a detailed knowledge of the instantaneous wind field, at least at the location of the analyzer, it is difficult, if not impossible, to properly interpret the results, no matter how sophisticated the data handling routines become. As a result, much of the discussion in this report has been merely qualitative in terms of the nature of the concentration fluctuations and possible causes of the fluctuations. Ideally, we would like to be able to quantitatively describe concentration fluctuations as a function of the instantaneous wind fluctuations, the source-to receptor configuration, and the roughness. Thus, in conclusion, with the new fast-response analyzer, NILU has the opportunity to be at the forefront of concentration fluctuation research, but to take full advantage of the opportunity, the need for good, simultaneous wind data should not be overlooked.

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APPENDIX

Data processing and analysis programs

```
C THIS IS A SIMPLE PROGRAM WHICH READS LBF-3 SF6 DATA FROM A
C SQUIRREL FILE AND SUBTRACTS THE BASELINE AND CONVERTS TO
C CONCENTRATION UNITS
      DIMENSION NHR(10800),NMIN(10800),NSEC(10800)
      DIMENSION VOLT(10800),CONC(10800)
      COMMON NHR,NMIN,NSEC,VOLT,CONC
      CHARACTER*3 NM
      CHARACTER*8 NAME

      OPEN(UNIT=6,FILE='D22C:CMT',STATUS='NEW')

      WRITE(6,6000)
 6000 FORMAT('**********************************************')
      WRITE(6,6001)
 6001 FORMAT('**************SF6 DATA PROCESSING*************')
      WRITE(6,6002)
 6002 FORMAT('**********************************************')
      WRITE(1,6004)
 6004 FORMAT(' PLEASE INPUT YOUR SQUIRREL FILE NAME: ',/)
      READ(1,*) NAME
      WRITE(6,6005)NAME
 6005 FORMAT(' SQUIRREL FILE NAME: ',A8)
C
C INITIALIZE THE NUMBER AND SIZE OF DATA FILES TO BE READ IN
C SQUIRREL FORMAT (NOTE THE MAXIMUM NUMBER OF FILES IS FOUR,
C BUT THIS MAY BE EASILY CHANGED)

      N1=0;N2=0;N3=0;N4=0

      WRITE(1,1)
 1    FORMAT(' INPUT THE NUMBER OF DATA FILES TO BE READ: ',/)
      READ(1,*)NFILES
      WRITE(6,5001)NFILES
 5001 FORMAT(' THE NUMBER OF DATA FILES : ',I5)
      WRITE(1,5)
 5    FORMAT(' INPUT THE TOTAL NUMBER READINGS IN FILE 1:',/)
      READ(1,*)N1
      WRITE(6,5002)N1
 5002 FORMAT(' THE NUMBER OF READINGS IN FILE 1:',I7)
      OPEN(UNIT=5,FILE='D900622C:DATA',STATUS='OLD')
      DO 20 I=1,N1
      READ(5,30)ND,NM,NY,NHR(I),NMIN(I),NSEC(I),VOLT(I)
 30   FORMAT(1X,I2,1X,A3,1X,I2,1X,I2,1X,I2,1X,I2,16X,F7.3)
 20   CONTINUE
      CLOSE(5)
      IF(NFILES.EQ.1)GOTO2
      WRITE(1,6)
 6    FORMAT(' INPUT THE TOTAL NUMBER READINGS IN FILE 2: ',/)
      READ(1,*)N2
      WRITE(6,5003)N2
 5003 FORMAT(' THE NUMBER OF READINGS IN FILE 2: ',I7)
      OPEN(UNIT=5,FILE='D800622C:DATA',STATUS='OLD')
      DO 21 I=N1+1,N1+N2
      READ(5,30)ND,NM,NY,NHR(I),NMIN(I),NSEC(I),VOLT(I)
 21   CONTINUE
      CLOSE(5)
```

```
      IF(NFILES.EQ.2)GOTO2
      WRITE(1,7)
 7    FORMAT(' INPUT THE TOTAL NUMBER READINGS IN FILE 3:',/)
      READ(1,*)N3
      WRITE(6,5004)N3
 5004 FORMAT(' THE NUMBER OF READINGS IN FILE 1:',I7)
      OPEN(UNIT=5,FILE='D700622C:DATA',STATUS='OLD')
      DO 22 I=N1+1+N2,N1+N2+N3
      READ(5,30)ND,NM,NY,NHR(I),NMIN(I),NSEC(I),VOLT(I)
 22   CONTINUE
      CLOSE(5)
      IF(NFILES.EQ.3)GOTO2
      WRITE(1,8)
 8    FORMAT(' INPUT THE TOTAL NUMBER READINGS IN FILE 4:',/)
      READ(1,*)N4
      WRITE(6,5005)N4
 5005 FORMAT(' THE NUMBER OF READINGS IN FILE 1:',I7)
      OPEN(UNIT=5,FILE='D600622C:DATA',STATUS='OLD')
      DO 23 I=N1+1+N2+N3,N1+N2+N3+N4
      READ(5,30)ND,NM,NY,NHR(I),NMIN(I),NSEC(I),VOLT(I)
 23   CONTINUE
      CLOSE(5)


      LL=-1;M=0
 2    N=N1+N2+N3+N4

C
C THE FOLLOWING SECTION WRITES PROMPT RESPONSES TO THE SCREEN
C AND ALSO INTO A COMMENT FILE (:CMT)
C
      WRITE(1,10)ND,NM,NY
 10   FORMAT(' DATE OF TEST: ',1X,I2,1X,A3,1X,I2)
      WRITE(6,5006)ND,NM,NY
 5006 FORMAT(' DATE OF TEST: ',1X,I2,1X,A3,1X,I2)
      WRITE(1,40)NHR(1),NMIN(1),NSEC(1)
      WRITE(6,5007)NHR(1),NMIN(1),NSEC(1)
 5007 FORMAT(' START TIME: ',I2,':',I2,':',I2)
 40   FORMAT(' START TIME: ',I2,':',I2,':',I2)
      WRITE(1,50)NHR(N),NMIN(N),NSEC(N)
      WRITE(6,5008)NHR(N),NMIN(N),NSEC(N)
 50   FORMAT(' END TIME: ',I2,':',I2,':',I2)
 5008 FORMAT(' END TIME: ',I2,':',I2,':',I2)
      WRITE(1,51)N
 51   FORMAT(' TOTAL NUMBER OF SCANS: ',I10)
      WRITE(6,5009)N
 5009 FORMAT(' TOTAL NUMBER OF SCANS: ',I10)
C
C SUBTRACT BASELINE FOR SELECTED SEGMENTS OF THE FILE
C
      WRITE(1,60)
 60   FORMAT(' INPUT NUMBER OF SEGMENTS FOR BASELINE CALC. ',/)
      READ(1,*)NSEG
      WRITE(1,61)NSEG
      WRITE(6,61)NSEG
 61   FORMAT(' THERE WILL BE ',I5,' BASELINE SEGMENTS ')
```

```
C
      WRITE(1,130)
 130  FORMAT(' WHAT ARE POWER-LAW COEFFICIENTS (a AND b)?',/)
      READ(1,*)A,B
      WRITE(1,129)A,B
      WRITE(6,129)A,B
 129  FORMAT(' POWER-LAW COEFFICIENTS ARE: ',1X,F8.2,2X,F8.4)

      K=1;II=0 ;SUM1=0.0;SUM2=0.0;SUMSQ1=0.0;SUMSQ2=0.0
      OPEN(UNIT=2,FILE='P800622C:DATA',STATUS='NEW')
      DO 70 J=1,NSEG
      WRITE(1,80)J,NHR(K),NMIN(K),NSEC(K)
 80   FORMAT(/,' SEGMENT ',I2,' BEGINS AT ',I2,':',I2,':',I2)
      WRITE(6,80)J,NHR(K),NMIN(K),NSEC(K)
      WRITE(1,90)
 90   FORMAT(' INPUT NUMBER OF READINGS FOR THIS SEGMENT: ',/)
      READ(1,*)NREAD
      K=K+NREAD
      WRITE(1,100)J,NHR(K-1),NMIN(K-1),NSEC(K-1)
      WRITE(6,100)J,NHR(K-1),NMIN(K-1),NSEC(K-1)
 100  FORMAT(' SEGMENT ',I2,' ENDS AT ',I2,':',I2,':',I2)
      WRITE(1,110)
 110  FORMAT(' INPUT SLOPE, Y-INT. AND NOISE OF BASELINE: ',/)
      READ(1,*)SLOPE,YINT,XNOISE
      WRITE(6,5011)J
 5011 FORMAT(' SEGMENT=',I2)
      WRITE(6,5015)SLOPE,YINT,XNOISE
 5015 FORMAT(' SLOPE= ',F12.9,' YINT= ',F4.2,' NOISE= ',F6.4)
      L=0
      DO 120 I=K-NREAD,K-1
      L=L+1;II=II+1
      YB=SLOPE*REAL(L)+YINT+3.*XNOISE
      ADVOLT=VOLT(I)-YB
      IF(ADVOLT.LT.0.0)ADVOLT=0.0
      CONC(I)=A*ADVOLT**B
      WRITE(2,121)I,CONC(I)
 120  CONTINUE
  70  CONTINUE
      NTESTS=0
      WRITE(1,1131)
 1131 FORMAT(' HOW MANY TRACER TESTS IN THIS DATA BLOCK? ',/)
      READ(1,*)NTESTS
      WRITE(6,5012)NTESTS
 5012 FORMAT(' THERE ARE ',I5,'TRACER TESTS IN DATA BLOCK')
      LL=-1;M=0
      DO 1132 KK=1,NTESTS
      SUM1=0.0;SUM2=0.0;SUMSQ1=0.0;SUMSQ2=0.0
      LL=LL+4;M=M+4
      WRITE(1,1133)KK
      WRITE(6,1133)KK
 1133 FORMAT(' TEST NUMBER ',I5)
      WRITE(1,131)
 131  FORMAT(' WHEN DOES THE FIRST SYRINGE BEGIN ? (HR:MIN:S)',/)
      READ(1,3331)NNHR,NNMIN,NNSEC
 3331 FORMAT(I2,1X,I2,1X,I2)
```

```
      WRITE(1,128)NNHR,NNMIN,NNSEC
      WRITE(6,128)NNHR,NNMIN,NNSEC
 128  FORMAT(' FIRST SYRINGE BEGINS AT: ',I2,':',I2,':',I2)
      DIFFHR=REAL(NNHR)-NHR(1)
      DIFFMIN=REAL(NNMIN)-NMIN(1)
      DIFFSEC=REAL(NNSEC)-NSEC(1)
      DIFF=DIFFHR*3600.+DIFFMIN*60.+DIFFSEC*1.0
      IDIFF=DIFF
C     IDIFFMAX=IDIFF+1800-1

      IDIFFMAX=N

      IDIFFMIN=IDIFF+900
      WRITE(1,2228)IDIFF
 2228 FORMAT(' TIME INTERVAL FROM BEGINNING OF FILE (S): ',I7)
      WRITE(6,2228)IDIFF
      IIII=0;IIIII=0
      IF(KK.GT.1)GOTO4434
      OPEN(UNIT=3,FILE='P22C1:DATA',STATUS='NEW')
      OPEN(UNIT=4,FILE='P22C2:DATA',STATUS='NEW')
      GOTO4435
 4434 OPEN(UNIT=3,FILE='P22CA1:DATA',STATUS='NEW')
      OPEN(UNIT=4,FILE='P22CA2:DATA',STATUS='NEW')
 4435 DO 1120 II=1,N
 1134 IF(II.LT.IDIFF)GOTO1120
      IF(II.GT.IDIFFMAX)GOTO1120
      IF(II.GE.IDIFFMIN)GOTO122
      SUM1=SUM1+CONC(II)
      IIII=IIII+1
      WRITE(3,121)IIII,CONC(II)
      GOTO1120
 122  SUM2=SUM2+CONC(II)
      IIIII=IIIII+1
      WRITE(4,121)IIIII,CONC(II)
 121  FORMAT(I6,',',F9.0)
 1120  CONTINUE
      XMEAN1=SUM1/REAL(IIII);XMEAN2=SUM2/REAL(IIIII)
      DO 125 I=IDIFF,IDIFF+899
      SUMSQ1=SUMSQ1+(CONC(I)-XMEAN1)**2
 125  CONTINUE
      DO 126 I=IDIFFMIN,IDIFFMAX
      SUMSQ2=SUMSQ2+(CONC(I)-XMEAN2)**2
 126  CONTINUE
      STD1=SQRT(SUMSQ1/REAL(IIII))
      STD2=SQRT(SUMSQ2/REAL(IIIII))
      WRITE(1,2223)IIII
      WRITE(6,2223)IIII
 2223 FORMAT(' COUNTS FOR SRYINGE A : ', 2X,I8)
      WRITE(1,123)XMEAN1
      WRITE(6,123)XMEAN1
 123  FORMAT(' AVERAGE CONC. FOR SYRINGE A: ',1X,F8.0,'PPT')
      WRITE(1,124)STD1
      WRITE(6,124)STD1
```

```
124  FORMAT(' CONC. STD. DEV. FOR SYRINGE A: ',1X,F8.0,'PPT')
     WRITE(1,9223)IIIII
     WRITE(6,9223)IIIII
9223 FORMAT(' COUNTS FOR SRYINGE B : ', 2X,I8)
     WRITE(1,9123)XMEAN2
     WRITE(6,9123)XMEAN2
9123 FORMAT(' AVERAGE CONC. FOR SYRINGE B: ',1X,F8.0,'PPT')
     WRITE(1,9124)STD2
     WRITE(6,9124)STD2
9124 FORMAT(' CONC. STD. DEV. FOR SYRINGE B: ',1X,F8.0,'PPT')
     CLOSE(3)
     CLOSE(4)
1132 CONTINUE
     CLOSE(2)
     CLOSE(6)
     END
```

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ABSTRACT (max. 300 characters, 7 lines)
During tracer experiments to quantify leakages of hydrocarbons, a fast response analyzer (response time 0.4 sec) for SF_6 was tested in the field. The analyzer provided adequate cover of concentration fluctuations within the mean tracer plume. The peak to mean ratio within the plumes were mostly less than 3.0, while the concentration fluctuation intensities were 0.2-0.3.

* Kategorier:	Åpen - kan bestilles fra NILU	A
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