BNL 28349

Presented at the Symposium on Plumes
and Visibility: Measurements and Model
Components, Grand Canyon, Arizona
Oct. 10-14, 1980
Also submitted to
Atmospheric Environment

MASTER

CONF-801114--8

Conversion Rates in Power Plant Plumes Based on Filter Pack Data
Part II: The Oil Fired Northport Plume

Robert W. Garber, Joseph Forrest and Leonard Newman

Environmental Chemistry Division
Department of Energy and Environment
Brookhaven National Laboratory
Upton, New York 11973 U.S.A.

*This work was done under Contract DE-AC02-76CH00016 with the United States Department of Energy and was jointly sponsored by the Electric Power Research Institute, the Empire State Electric Energy Research Corporation and the Long Island Lighting Company.

DISCLAIMER

Abstract

More than 60 airborne plume studies were conducted at a large oil fired power station during a 3-1/2 year period. These studies were conducted to determine the typical rate of formation of sulfate in the plume and the conditions which most influence these atmospheric processes. The power plant chosen for this program is located in the northeast region of the U.S. and during the course of these studies a typical variety of meteorological conditions were encountered. The diurnal variation was also extensively studied.

Plume sulfate rarely accounted for more than 5% of the total plume sulfur even for plume travel times of up to 4 hours. For most experiments more than half of the plume sulfate was that emitted from the power plant units. The rate of atmospheric oxidation of sulfur dioxide to sulfate was not readily discernible due to the low rate of conversion and the relatively high amount of the sulfate emitted. The results reported in this paper generally indicate an apparent oxidation rate of less than 1% per hour. A diurnal influence or effects due to changes in various meteorological conditions are difficult to detect. However the large data set permits us to conclude that either higher temperatures, higher partial pressures, or mid-day periods can give rise to oxidation rates two to three times higher than the average.

Introduction

Anthropogenic activities are responsible for the daily release into the atmosphere of nearly one half million tons of sulfur oxides. Fossil-fuel combustion for electric power production is the principal activity producing these emissions. The environmental impact of such atmospheric emissions is not well understood and has been of public concern for many years. In response to this concern several large-scale field studies have been conducted at coal-fired power plants to aid in the understanding of the environmental problems (e.g. Forrest et al., 1980).

Oil-fired power plants release nearly 100,000 tons of sulfur oxides into the atmosphere each day. The chemical composition of the emissions from these plants are markedly different from those of coal-fired plants, and if heterogeneous reactions are involved, then conceivably the atmospheric behavior of sulfur oxides could differ as well. Although oil-fired power plants are predominantly located in the most heavily populated regions of the United States, only a few short-term field studies of their plume chemistry have been conducted (Forrest and Newman, 1978; Forrest et al., 1979; Newman et al., 1975). In light of the paucity of relevant information, Brookhaven National Laboratory proposed a comprehensive study of the emissions from a large oil-fired power station located on Long Island, New York with emphasis upon investigating sulfur oxide reactions. This program was jointly accepted and sponsored by the Electric Power Research Institute (EPRI), the Empire State Electric Energy Research Corporation (ESEERCO) and the Long Island Lighting Company (LILCO). The measurements for this program were started in the fall of 1976 and completed in early February 1980.

Experimental

Plant description

The Northport Power Station of the Long Island Lighting Company is located on the north shore of Long Island along Long Island Sound. At present power is generated by four 375 MW tangentially fired boilers, three of which use No. 6 grade residual fuel oil of ~ 2.4% sulfur and one uses fuel of < 0.5% sulfur. Each unit is equipped with electrostatic precipitators having a design collection efficiency of 98%. The flue gases from each boiler are emitted via separate 183 m stacks

Although plant operating conditions varied from day to day, in general at least two units were operating at near full load for all experiments. The plant conditions are summarized in Table 1. During some of the earlier experiments, Nos. 1-7 emissions from Unit No. 2 were controlled by a cyclone separator. During experiment Nos. 9-36 Unit No. 2 operated without any particle removal device.

Sampling

A Cessna 206 was utilized for experiments 1-23, and was equipped with a sampling package consisting of an air scoop, a filter cassette assembly, a high volume blower and a Sign-X SO_2 conductivity analyzer. Subsequent samples were taken with the BNL Atmospheric Sciences aircraft, a Britten-Norman Islander, cruising at ~ 75 knots. The instrument package for this plane was previously described (Forrest et al., 1980). Samples were collected with a 12.5 cm diam. high volume assembly consisting of a quartz prefilter to collect particulate matter and two K_2CO_3-impregnated cellulose filters to absorb SO_2 (Forrest et al., 1979).

Plume traverses were made at selected locations, e.g., 0.5, 1, 5 and 20 miles downwind or further when possible. Multiple traverses across the plume centerline to accumulate sufficient sample for analysis required 20-40 minutes flight time at each location. Background samples upwind of the stack or alongside the plume were collected for each experiment.

Temperature and relative humidity measurements were made aloft for each flight and ranged quite widely, from -9° to 31°C and 20-95% respectively. Flights were conducted during varying periods of the day to evaluate temporal and insolation effects. Additional data collected for each experiment included wind speeds obtained from pilot balloons, meteorological summaries from the National Weather Service, and power plant operating conditions. Detailed information is available (Garber et al., 1980).

Analysis

Potassium carbonate-impregnated filters were analyzed for collected SO_2 during experiments 1-48 by a ^{110}Ag technique (Forrest and Newman, 1977). Subsequent filters were analyzed by ion chromatography (Small et al., 1975).

The ^{110}Ag procedure was employed for measuring particulate sulfate on the quartz filters from experiments 1-20. For experiments 21-47, the sulfate extracted from the quartz filters was determined by an automated methylthymol blue procedure (Adamski and Villard, 1975). Subsequent to experiment 47, extracted sulfate was analyzed by ion chromatography. Ammonia was determined by an automated sodium phenoxide technique (Bolleter et al., 1961). Particulate samples were also processed for the determination of total titratable strong acid using coulometrically-generated base (Tanner et al., 1977).

Observations

Sixty-six airborne plume studies were attempted for this program, sixty-two of which yielded plume data. Ratios of particulate to total sulfur (fraction of SO_2 oxidized) were plotted as a function of time from emission (calculated from the wind speeds and the distance from the source). Background subtractions for SO_2 and particulate sulfur were applied to all measurements. The data for all experiments are presented in Fig. 1.

A logarithmic scale is employed for the abscissa to avoid overcrowding of the plot with data at the low plume age region at which time most of the samples were taken. It can be seen that with few exceptions, virtually all points lie within a bracket of fraction oxidized ranging between 0 and 0.04. Several values have negative fractions, attributable to normal analytical error or arising from background corrections variable between the point of measurement and the downwind plume location. The data, when plotted on a linear scale, produced an oxidation rate for SO_2 of 0.5% per hours (Figure 2). Shown in this and subsequent figures is the best fit least squares regression line along with the 95% confidence limits presented as dashed curves.

The data were then subjected to a detailed examination by first establishing as a reasonable criterion for the acceptance of a measurement the requirement that the net plume concentrations of either sulfate or sulfur dioxide be at least equal to that of the background correction. Those samples (30% of the total) which failed to meet this prerequisite (thus by definition eliminating negative values) were eliminated. A 16 km, 45 min sample of Exp. 57 (particulate S/total S = 0.29), although meeting the above criterion, was discarded because of the strong likelihood that the aircraft was not in the plume during sampling, as evidenced by an SO_2 concentration of but 14.9 $\mu g/m^3$. Table 2 lists the meteorological and plume data for the acceptable runs. A scattergram was generated for this subset as described above and is shown in Figure 3. When transposed to a linear time scale (Figure 4) the plot yielded a least square fit with a correlation coefficient of 0.31 (95% confidence level = 0.16) and giving an oxidation rate for SO_2 of 0.9% per hour.

We explored the possibility that two apparent outlying values for SO_2 conversion, Exp. 37 at 32 km, 268 min and Exp. 43 at 1.6 km, 5.4 min, each with a particulate S/total S of 0.15, could tend to markedly skew the subset scattergram slope. A new scattergram was generated, omitting these two points, which yielded a least squares fit with a correlation coefficient of 0.12 (95% confidence level = 0.16) but now giving an oxidation rate for SO_2 of only 0.30% per hour (Figure 5).

Plume studies conducted by BNL and other researchers have implicated, in some locations, several meteorological parameters such as temperature, H_2O partial pressure and insolation as being associated with the oxidation rate of atmospheric SO_2. We have attempted to gain further insight into these relationships from our data by subdividing our subset into various classifications derived from these parameters. Two series of scattergrams were generated with and without the two apparent outlying values. Shown in the following figures is the subset without the outlying values. Included in the analysis of the fraction of SO_2 converted is a subdivision with respect to wind speeds of <8 and >8 meters per second (Figures 6 and 7). In examining the data for seasonal effects, flights were separated into 3 categories which roughly correspond to winter, spring and fall, and summer seasons by examining the relationship between SO_2 oxidation and the temperature intervals of <5°, 5-13°, and >13°C (Figures 8, 9 and 10). The effect of relative humidity was explored by forming groups of <75% and >75% R.H. (Figures 11 and 12). As a possible more meaningful indicator of atmospheric moisture content, the partial pressure of water in the atmosphere during each run was calculated and two subsets were formed, <5.4 and >5.4 mm (Figures 13 and 14). Solar radiation as an indicator of photochemistry, was examined from the data which was divided into three time regimes. All nighttime experiments were placed in Time Code 1 and combined with Time Code 2 which consisted of daylight runs obtained during the early morning and late afternoon or evening (Figure 15). Examined separately (Figure 16) was Time Code 3 which contained those runs made during the hours of maximum anticipated insolation, 10 A.M. to 2 P.M.

Discussion of Results

In reviewing the results of all experiments and Figure 3, we observe that the extent of oxidation is quite low, largely confined to 1-4% and rarely exceeding 5%. Most of the oxidation occurs during the early history of the plume with very little apparent conversion taking place downwind.

As a means of summarizing our attempts to stratify the data on the basis of meteorological parameters, we show in Table 3 a compilation obtained from the previous figures of the "best fit" SO_2 oxidation rates for each of the various meteorological categories that were investigated. Two groups are listed: Group I includes all samples of the subset, while Group II (plots for which are sown in Figures 5 to 16) excludes the samples from Experiment 37 at 268 minutes and Experiment 43 at 5 minutes. As can be seen from Table 3, the average conversion rate for all samples was 0.9% per hour for Group I and 0.3% per hour for Group II. The fact that two data points could so vastly affect the slope of the scattergram plots led us to strongly suspect their validity. Although no plausible reason could be advanced for exclusion of Exp. 43 at 1.6 km, 5.4 min, some doubt definitely exists concerning the location of the plume traverse for Exp. 37 at 32 km, 268 min. Notwithstanding a background SO_2 concentration of ~ zero, the plume SO_2 concentration for this location of but ~ 9 $\mu g\ m^{-3}$ creates a large uncertainty as to whether the aircraft actually sampled the plume. In light of these doubts, we place greater confidence in the data set which excludes both these points.

The reader should observe (Figure 5) that most of the data points fall outside the 95% confidence limits. Consequently the absolute value of the rate (0.3% per hour) is not well established but it can be concluded that the oxidation rate on average is certainly less than 1% per hour. This rate and the others shown in Table 3 are generally consistent with those observed at other locations, namely 0.5-2% per hour. The Northport power plant typically emits a plume having an aerosol sulfur to total sulfur ratio of 0.5 to 2.5% (Dietz, 1980). This agrees with the average value ~ 1.8%, which is obtained from the zero time intercept of Figure 5. At these levels the sulfate emitted during combustion accounts for the major portion of plume sulfate observed during the first few hours after emission, and tends to overwhelm our ability to detect specific trends within that base. Nevertheless, examination of Table 3 reveals that, under certain conditions, enhanced oxidation seems to be taking place. In Group II, for temperatures greater than 13°C, partial pressures greater than 5.4 mm and periods of maximum insolation (Time Code 3), we observe that the oxidation rates exceed that for the average of all samples by factors of 2 to 3. The correlation coefficients for the scattergram plots in Figures 10, 14 and 16 from which these rates were derived were 0.13, 0.16 and 0.20; 95% confidence levels for these sets are 0.24, 0.22 and 0.20 respectively. Although the rates derived from the Group I subset are higher (influenced by the two questioned points) we nevertheless observe a similar trend in that the rates for each of these parameters are significantly higher than the average value of the 0.90% per hour for all samples in the group.

One should recognize that not many plumes were trackable beyond two hours duration, largely because of emphasis upon mid-day experiments when unstable conditions usually prevail. Also, the peculiar geography of Long Island, a narrow strip of land surrounded on three sides by large water bodies, did not lend itself to long distance tracking. Consequently, the scattergram slopes and oxidation rates derived therefrom are strongly influenced by a few points. In view of the fairly large spread for the 95% confidence limits of these rates, a firm statement relating specific meteorological parameters with sulfate formation could not be absolutely justified. At best, and only because more than sixty experiments were performed, can we claim that the data implies some positive relationship; a more definitive claim would be extremely tenuous.

The acid and ammonium contents of the collected aerosol were also measured. The data showed a marked scatter but, in general, the plume aerosol was slightly acid. However, a number of samples indicated an aerosol composition which was distinctly alkaline. The aerosol sulfate also had a larger ammonium ion component, suggesting possible compositions varying from ammonium sulfate [$(NH_4)_2SO_4$] to ammonium acid sulfate (NH_4HSO_4). The average composition of the sulfate aerosol as indicated by the ammonium to sulfate ratios and the acid to sulfate ratios aproached that of Letovacite $[(NH_4)_3H(SO_4)_2]$. Both the acidity and ammonium data are currently being subjected to a more critical examination to evaluate possible trends and relationships.

Conclusions

The experimental data from the airborne experiments yield the following information about the Northport plume:

1. Primary sulfate accounts for the major portion of plume sulfate during the first few hours after emission.

2. With few exceptions, virtually all measured ratios of plume sulfate to total sulfur lie within the range of 0-5% for plume ages up to 200 minutes.

3. The oxidation rate was essentially unmeasurable but is certainly less than 1% per hour.

4. The data imply, but not very conclusively, a positive relationship between sulfate formation with temperature, partial pressure and insolation. Firm conclusions and the reasons therefore are not justified.

5. The sulfate aerosol of the plume is generally acidic with a high ammonium ion content. Compositions varying from ammonium acid sulfate through Letovacite to ammonium sulfate are possible. Some aerosol samples were distinctly alkaline.

Acknowledgments

We wish to acknowledge the efforts of Daniel Leahy, Mary Phillips, Seymour Fink and Robert Brown of Brookhaven National Laboratory in taking plume samples and monitoring airborne instruments. We are grateful to the Analytical Group for performing the chemical analyses and to Joyce Tichler for processing the real time data. Thanks go to the Electric Power Research Institute, the Empire State Electric Energy Research Corporation and the Long Island Lighting Company for sponsorship and support.

References

1. Adamski J.M. and Villard S.P. (1975). Application of methylthymol blue sulfate method to water and wastewater analysis. Analyt. Chem. 47, 1191-1194.

2. Bolleter W.T., Bushman C.J. and Tidewell P.W. (1961). Spectrophotometric determination of ammonia as indophenol. Analyt. Chem. 33, 592-594.

3. Dietz R.N. (1980). An extensive study of an oil fired power plant. Final Report, Vol. I Breech Measurements. Brookhaven National Laboratory (in press).

4. Forrest J. and Newman L. (1977). Silver-110 microgram sulfate analysis for the short time resolution of ambient levels of sulfur aerosol. Analyt. Chem. 49, 1579-1584.

5. Forrest J. and Newman L. (1978). Oxidation of sulfur dioxide in power plant plumes. AIChE Symposium Series 74 (175), 48-53.

6. Forrest J., Garber R. and Newman L. (1979). Formation of sulfate, ammonium and nitrate in an oil-fired power plant plume. Atmospheric Environment 13, 1287-1297.

7. Forrest J., Garber R. and Newman L. (1980). Conversion rates in power plant plumes based on filter pack data: Part I - The coal fired Cumberland plume. Presented at Symposium on plumes and visibility: measurements and model components. Grand Canyon, Arizona, Nov. 10-14. (Submitted to Atmospheric Environment).

8. Garber R., Forrest J. and Newman L. (1980). An extensive study of an oil-fired power plant. Final Report, Vol. II Plume Measurements. Brookhaven National Laboratory (in press).

9. Newman L., Forrest J. and Manowitz B. (1975). The application of an isotopic ratio technique to a study of the atmospheric oxidation of sulfur dioxide in the plume from an oil-fired power plant. Atmospheric Environment 9, 959-968.

10. Small H., Stevens T.J. and Bauman W.C. (1975). Novel ion exchange chromatographic method using conductimetric detection. Analyt. Chem. 47, 1801-1809.

11. Tanner R.L., Cederwall R., Garber R., Leahy D., Marlow W., Meyers R., Phillips M. and Newman L. (1977). Separation and analysis of aerosol sulfate species at ambient concentrations. Atmospheric Environment 11, 958-966.

LIST OF FIGURES

Table 1. Plant operating conditions.

Experiment No.	Generating units in service	Particle removal system
1-7	1,3	Electrostatic precipitators
	2	Cyclone
9-36	1,3	Electrostatic precipitators
	2	None
37-48	1,3,4	Electrostatic precipitators
49-66	1,2,3,4	Electrostatic precipitators

Table 2. Meteorological and plume data.

Exp.	Location km	Age min.	Temp. °C	Rel. Hum. %	Part. Press. mm	Time* Code	SO_2 µg m^{-3}	SO_4^{2-} µg m^{-3}	Partic. S / Total S
1	0.8	1.3	4.0	44	2.7	1	283	3.38	0.008
2	0.4	3.4	-1.0	71	3.2	1	547	21.8	0.026
	1.6	13.4					447	10.5	0.015
	9.7	81					76.5	7.82	0.064
	29	242					53.2	2.17	0.027
3	0.4	0.8	5.0	46	3.0	1	918	25.2	0.018
	1.6	3.4					220	7.74	0.023
4	0.6	1.9	5.0	58	3.8	2	1337	15.6	0.008
5	0.8	4.5	4.0	82	5.0	2	2288	21.7	0.006
	1.6	8.9					1212	15.7	0.009
	4.8	27					1993	30.9	0.010
	16	89					956	20.2	0.014
6	0.4	1.3	2.0	61	3.2	1	4572	49.9	0.007
	1.6	5.4					940	9.24	0.007
7	0.4	1.3	4.4	68	4.3	2	2195	61.4	0.018
	4.8	16.1					211	7.89	0.024
9	1.6	3.0	-2.0	63	2.5	2	836	37.5	0.029
10	1.6	1.9	5.0	43	2.8	1	957	27.3	0.019
	4.8	5.7					208	6.98	0.022
	4.8	5.7					765	21.0	0.018
	16.1	19.2					442	27.4	0.040
11	1.6	2.7	3.0	55	3.1	2	2165	55.1	0.017
	4.8	8.0					1171	32.4	0.018
	16.1	26.8					451	12.6	0.018
12	1.6	2.4	-1.0	60	2.5	1	3937	146	0.024
	4.8	7.3					3122	125	0.026
	16.1	24.4					4825	177	0.024
	40	61					2587	80.8	0.020
	64	98					2193	65.7	0.020
	97	146					736	24.5	0.022
	129	195					2245	61.1	0.018
13	1.6	3.4	1.0	59	2.9	1	1343	50.0	0.024
	4.8	10.1					849	23.9	0.018
	16.1	33.5					493	13.2	0.017

Table 2. Meteorological and plume data, cont.

Exp.	Location km	Age min.	Temp. ^{o}C	Rel. Hum. %	Part. Press. mm	Time* Code	SO_2 $\mu g\ m^{-3}$	SO_4^{2-} $\mu g\ m^{-3}$	$\frac{\text{Partic. S}}{\text{Total S}}$
14	1.6	1.5	-1.0	43	1.8	1	2969	69.9	0.015
	4.8	4.5					1589	23.1	0.010
	16.1	14.9					856	16.2	0.012
	40.2	37.3					975	15.5	0.011
15	1.6	3.0	10.5	32	3.0	3	904	17.6	0.013
	4.8	8.9					961	17.5	0.012
	16.1	29.8					420	10.5	0.016
	40.2	74.5					420	11.8	0.018
	77.3	143					150	7.67	0.033
16	1.6	3.4	11.0	56	5.5	3	3664	144	0.026
	4.8	10.1					940	27.8	0.019
	16.1	33.5					405	18.2	0.029
	40.2	83.9					273	12.5	0.030
17	1.6	1.5	22.0	61	12.1	1	2909	42.6	0.010
	1.6	1.5					2149	38.2	0.012
	4.8	4.5					2357	88.6	0.024
18	0.8	2.7	24.0	62	13.9	3	1069	28.7	0.018
	1.6	5.4					1674	33.9	0.013
	1.6	5.4					493	11.2	0.015
	4.8	16.1					267	8.00	0.020
	16.1	53.7					375	12.1	0.021
19	0.8	3.8	14.0	88	10.6	3	3976	56.0	0.009
	1.6	7.7					544	28.3	0.034
	4.8	23.0					251	16.3	0.042
	16.1	76.7					107	1.45	0.009
20	1.6	4.5	22.0	28	5.5	3	729	17.2	0.016
	1.6	4.5					535	4.46	0.006
	4.8	13.4					141	3.61	0.017
	4.8	13.4					121	3.95	0.021
21	0.8	1.7	24.0	75	16.8	3	5827	181	0.020
	0.8	1.7					638	29.1	0.029
	4.8	10.1					438	36.0	0.052
	4.8	10.1					1059	14.5	0.009
23	0.8	1.9	31.5	36	12.5	3	3287	56.3	0.011
	0.8	1.9					1814	25.6	0.009
	0.8	1.9					2210	35.6	0.011
	0.8	1.9					2504	46.0	0.012
	4.8	11.5					286	9.76	0.022

Table 2. Meteorological and plume data, cont.

Exp.	Location km	Age min.	Temp. °C	Rel. Hum. %	Part. Press. mm	Time* Code	SO_2 µg m^{-3}	SO_4^{2-} µg m^{-3}	Partic. S / Total S
24	0.8	2.7	22.0	61	12.1	2	3485	63.6	0.012
	1.6	5.4					1824	32.5	0.012
	4.8	16.1					1256	20.5	0.011
25	0.8	1.5	22.0	54	10.7	1	1675	33.6	0.013
	1.6	3.0					829	23.2	0.018
26	0.8	2.7	22.0	61	12.1	3	1094	29.6	0.018
	4.0	13.4					125	3.33	0.017
27	0.8	2.7	21.0	44	8.2	1	2965	56.5	0.013
	4.8	16.1					336	6.86	0.013
28	0.8	2.7	9.0	95	8.2	3	2746	49.6	0.012
	2.4	8.0					629	15.8	0.016
29	0.8	4.5	9.0	95	8.2	3	2715	57.6	0.014
	8.0	44.7					447	13.9	0.020
30	0.2	0.7	-5.0	53	1.6	2	1005	27.3	0.018
	0.8	2.7					1700	61.0	0.023
	1.6	5.4					985	25.7	0.017
31	0.2	0.7	-1.0	60	2.5	3	13885	285	0.013
	0.2	0.7					13046	313	0.016
	0.8	2.7					2725	93.4	0.022
	1.6	5.4					4068	83.4	0.013
	8.0	26.8					1001	20.9	0.014
32	0.2	0.9	1.0	51	2.5	3	8030	24.6	0.002
	0.2	0.9					11364	274	0.016
	0.4	1.7					5211	97.6	0.012
	0.4	1.7					5836	138	0.016
	0.8	3.4					4089	101	0.016
	1.6	6.7					2589	48.7	0.012
33	0.4	1.3	-1.5	42	1.7	3	4522	107	0.015
	1.6	5.4					809	25.3	0.020
34	0.8	1.9	-4.5	53	1.7	3	2898	61.7	0.014
	1.6	3.8					596	13.3	0.015
	6.4	15.3					87.9	4.10	0.030
35	0.5	1.3	-8.8	54	1.2	3	1511	7.23	0.003
	1.6	3.8					267	3.71	0.009
	4.8	11.5					155	2.41	0.010
	11.3	26.8					121	1.97	0.011

Table 2. Meteorological and plume data, cont.

Exp.	Location km	Age min.	Temp. °C	Rel. Hum. %	Part. Press. mm	Time* Code	SO_2 µg m^{-3}	SO_4^{2-} µg m^{-3}	Partic. S / Total S
36	0.4	0.7	10.0	60	5.5	3	1824	23.7	0.009
	0.8	1.3					881	5.12	0.004
37	1.6	13.4	21.0	50	9.4	3	626	8.61	0.009
	32.2	268					8.7	2.30	0.149
39	1.6	2.7	22.0	60	11.9	3	151	4.47	0.019
40	0.8	6.7	24.0	55	12.3	3	1748	45.1	0.017
	0.8	6.7					1796	42.4	0.015
	4.8	40.2					1020	32.6	0.021
	4.8	40.2					873	42.3	0.031
41	0.8	2.2	24.0	50	11.2	3	479	15.3	0.021
42	0.8	2.2	25.0	50	11.9	3	410	17.9	0.028
	0.8	2.2					206	31.9	0.093
43	1.6	5.4	23.0	45	9.5	3	33.6	8.85	0.149
45	3.2	10.7	28.0	60	17.0	3	115	7.26	0.040
46	3.2	8.9	23.0	33	12.7	3	173	6.12	0.023
	27.4	76.0					50.4	3.37	0.043
48	1.6	3.8	18.0	60	9.3	3	279	30.2	0.067
49	1.6	4.5	19.0	68	11.2	2	851	37.1	0.028
50	1.6	4.5	18.0	60	9.3	3	651	47.3	0.046
	19.3	53.7					284	31.3	0.068
51	1.6	13.4	13.1	50	5.7	3	437	26.4	0.039
52	1.6	4.1	18.0	48	7.4	3	430	8.61	0.013
53	1.6	4.9	13.0	40	4.5	3	128	1.40	0.007
56	1.6	6.0	17.4	50	7.5	3	190	4.99	0.017
57	1.6	4.5	19.0	40	6.6	3	266	29.7	0.069
58	1.6	6.7	16.0	40	5.4	3	389	3.12	0.005
	16.1	67.1					68.7	2.17	0.021
60	1.6	1.9	−5.8	25	0.7	1	468	27.5	0.038
	8.0	9.6					87.1	6.50	0.047

Table 2. Meteorological and plume data, cont.

Exp.	Location km	Age min.	Temp. °C	Rel. Hum. %	Part. Press. mm	Time* Code	SO_2 µg m^{-3}	SO_4^{2-} µg m^{-3}	Partic. S / Total S
61	32.2	76.7	-7.9	20	0.5	3	66.0	3.14	0.031
62	37.0	103	-6.8	20	0.5	1	766	2.77	0.002
63	0.8	2.6	-3.9	35	1.2	3	1217	7.98	0.004
	11.3	36.1					44.1	2.57	0.037
64	1.6	2.4	-3.9	35	1.2	1	286	11.5	0.026
	35.4	52.2					50.7	1.83	0.023
65	0.8	1.3	-2.0	40	1.6	2	150	2.98	0.013
66	8.0	12.4	-4.0	30	1.0	1	728	4.78	0.004

* Time codes: 1 - Nighttime
2 - Daylight, early morning or late afternoon
3 - Midday, 10 a.m. to 2 p.m.

Table 3. SO_2 Oxidation Rates, % per Hour

Parameter	Group I[(a)]	Group II[(b)]
All Samples	0.90	0.30
Wind Speed <8 m sec^{-1}	1.38	0.48
Wind speed >8 m sec^{-1}	0.12	0.12
Temp < 5°C	0.24	0.24
5 < Temp. °C <13	0.84	0.84
Temp. > 13°C	2.3	0.72
Rel. Humidity < 75%	0.96	0.36
Rel. Humidity > 75%	-0.48	-0.48
Partial Press. <5.4 mm	0.30	0.30
Partial Press >5.4 mm	2.3	0.84
T Code 1 and 2	0.18	0.18
T Code 3	1.9	0.78

[a] Includes all samples in subset

[b] As in (a), but excluding Exp. 37, 268 minutes and Exp. 43, 5 minutes.

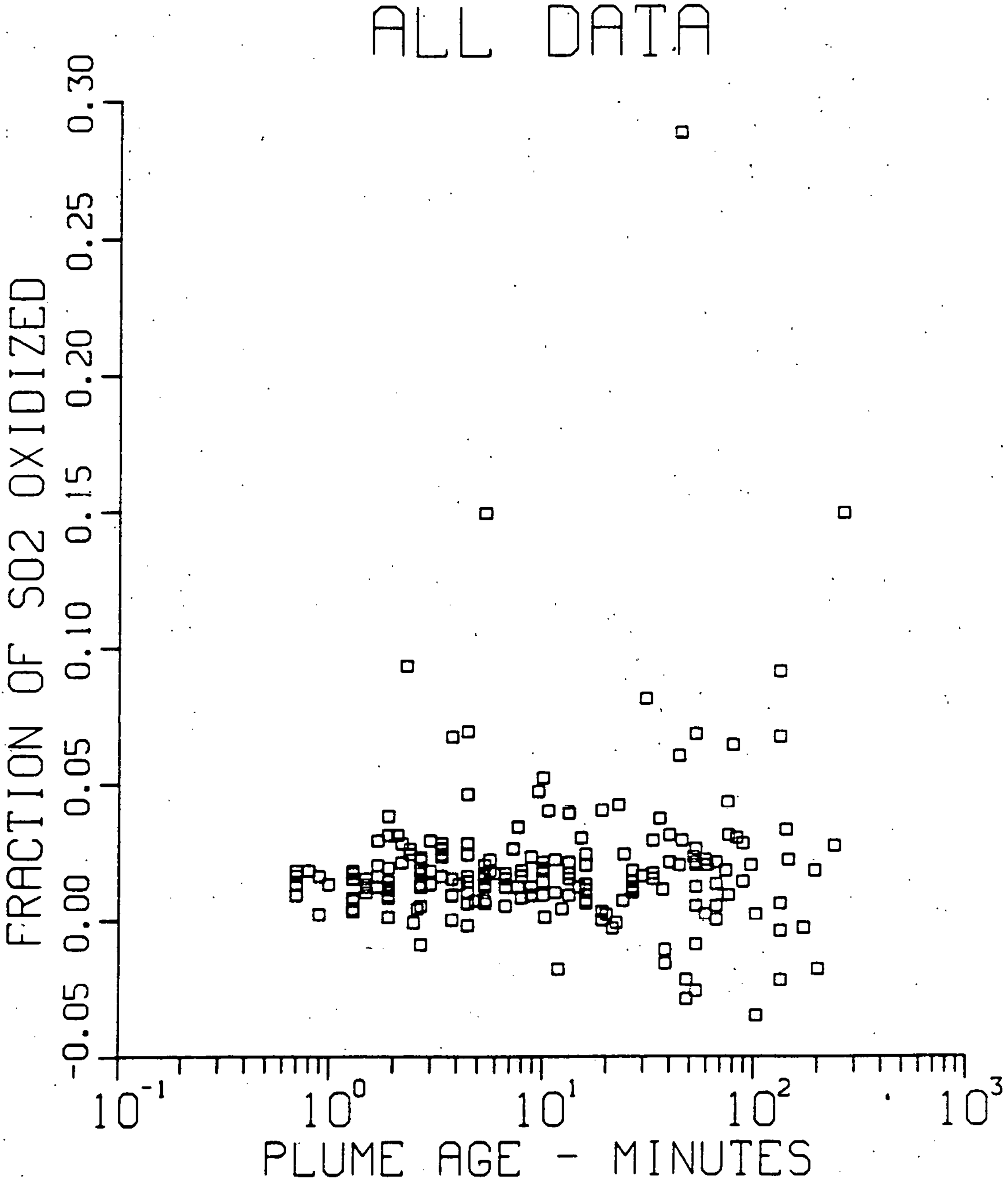
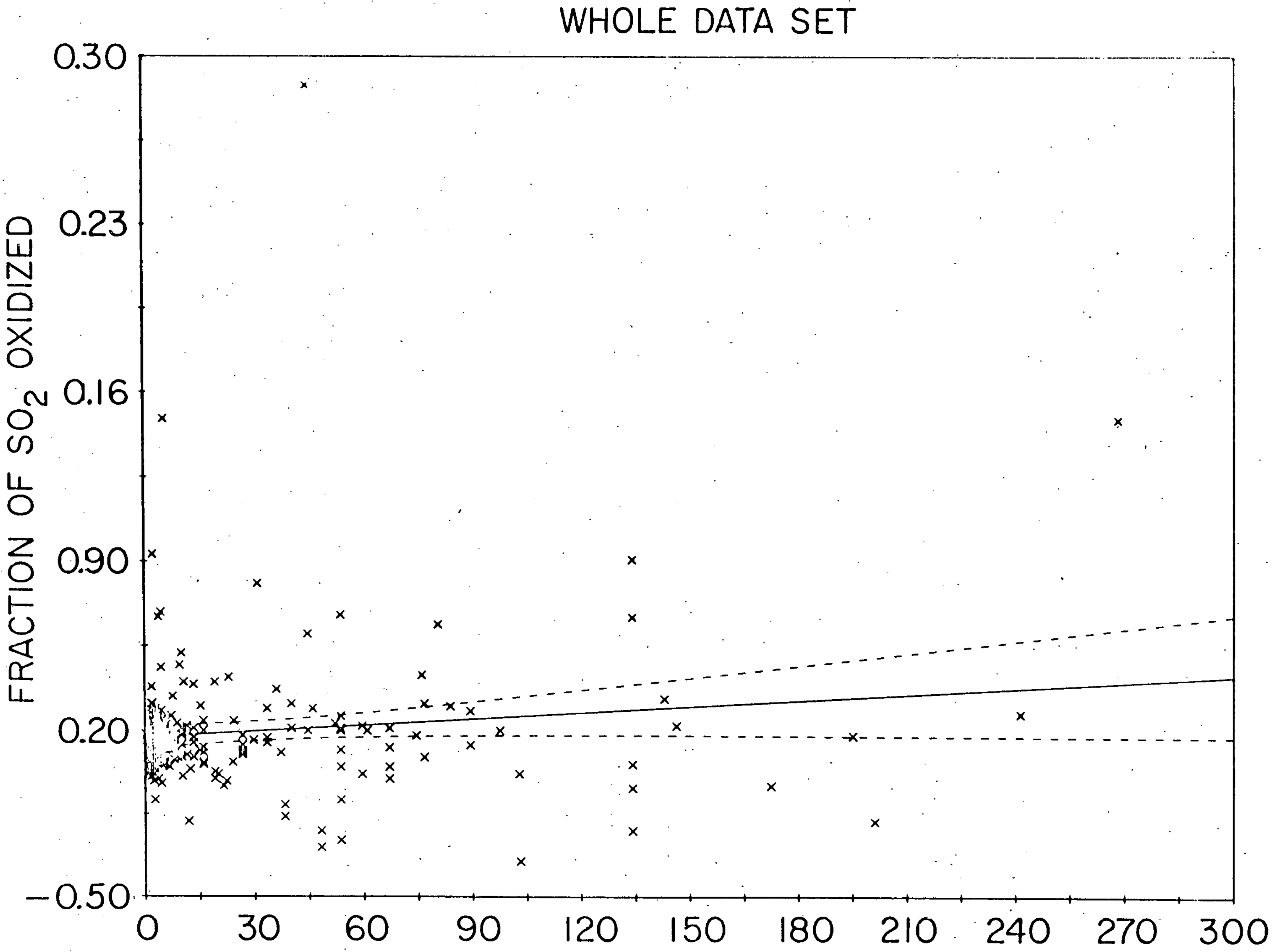

ALL DATA
FRACTION OF SO2 OXIDIZED
0.30
0.25
0.20
0.15
0.10
0.05
0.00
-0.05
-0.0
10^{-1}
10^{0}
10^{1}
10^{2}
10^{3}
PLUME AGE - MINUTES

FIG. 1



WHOLE DATA SET
FRACTION OF SO_2 OXIDIZED
0.30
0.23
0.16
0.90
0.20
−0.50
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG. 2

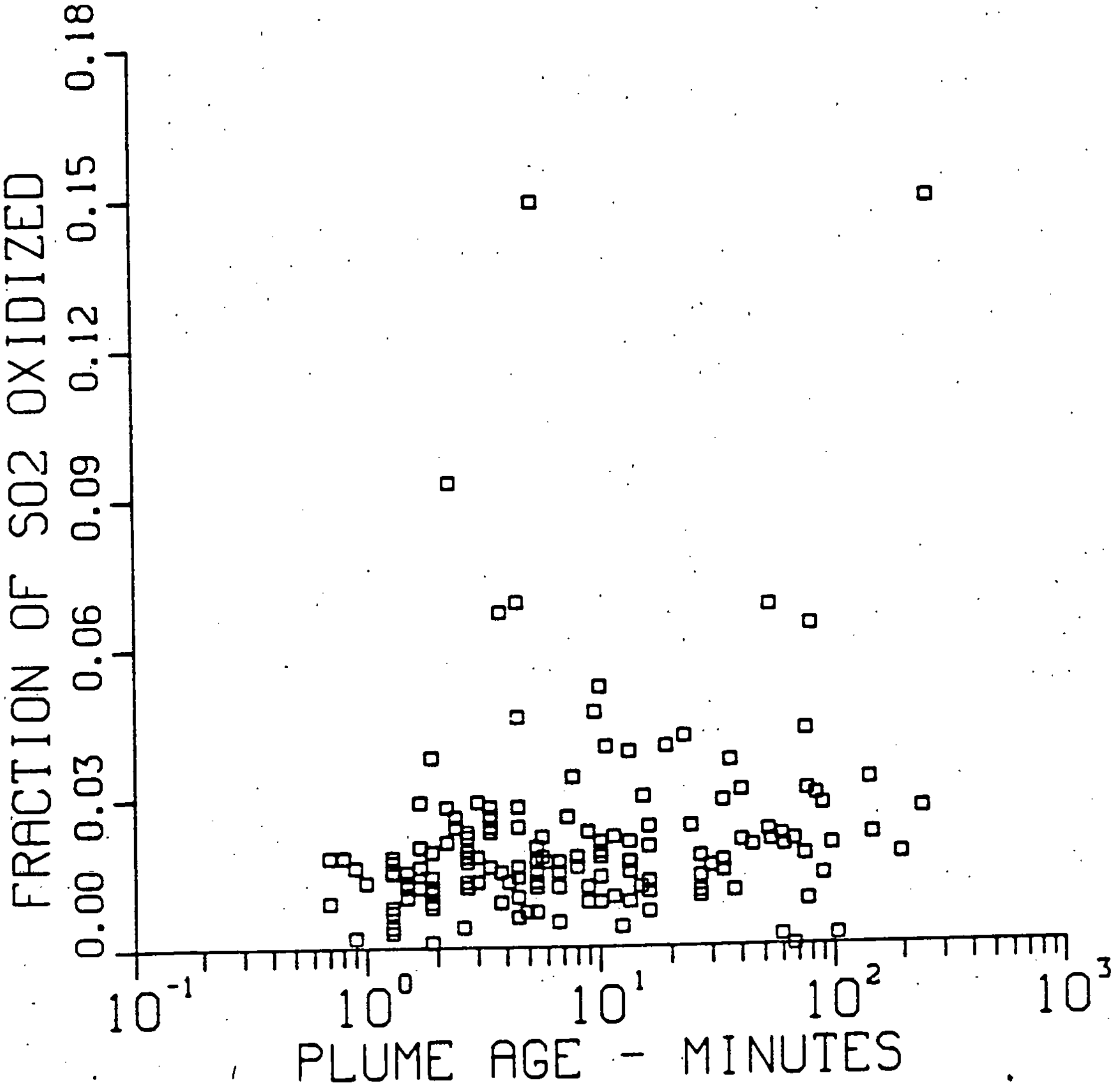
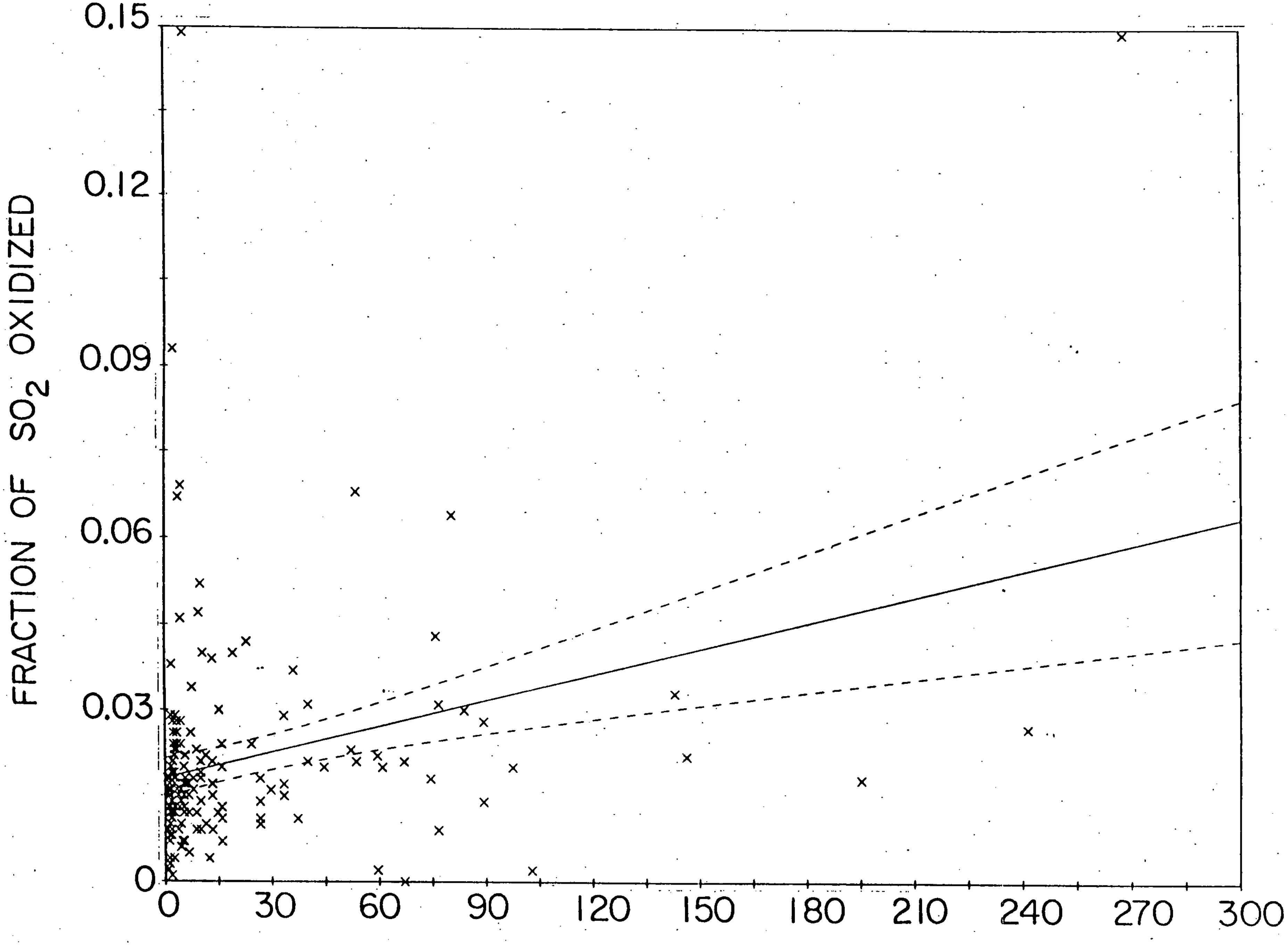

FRACTION OF SO2 OXIDIZED
0.00
0.03
0.06
0.09
0.12
0.15
0.18
10^{-1}
10^{0}
10^{1}
10^{2}
10^{3}
PLUME AGE - MINUTES

FIG. 3



0.15
0.12
0.09
0.06
0.03
0
FRACTION OF SO_2 OXIDIZED
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG. 4

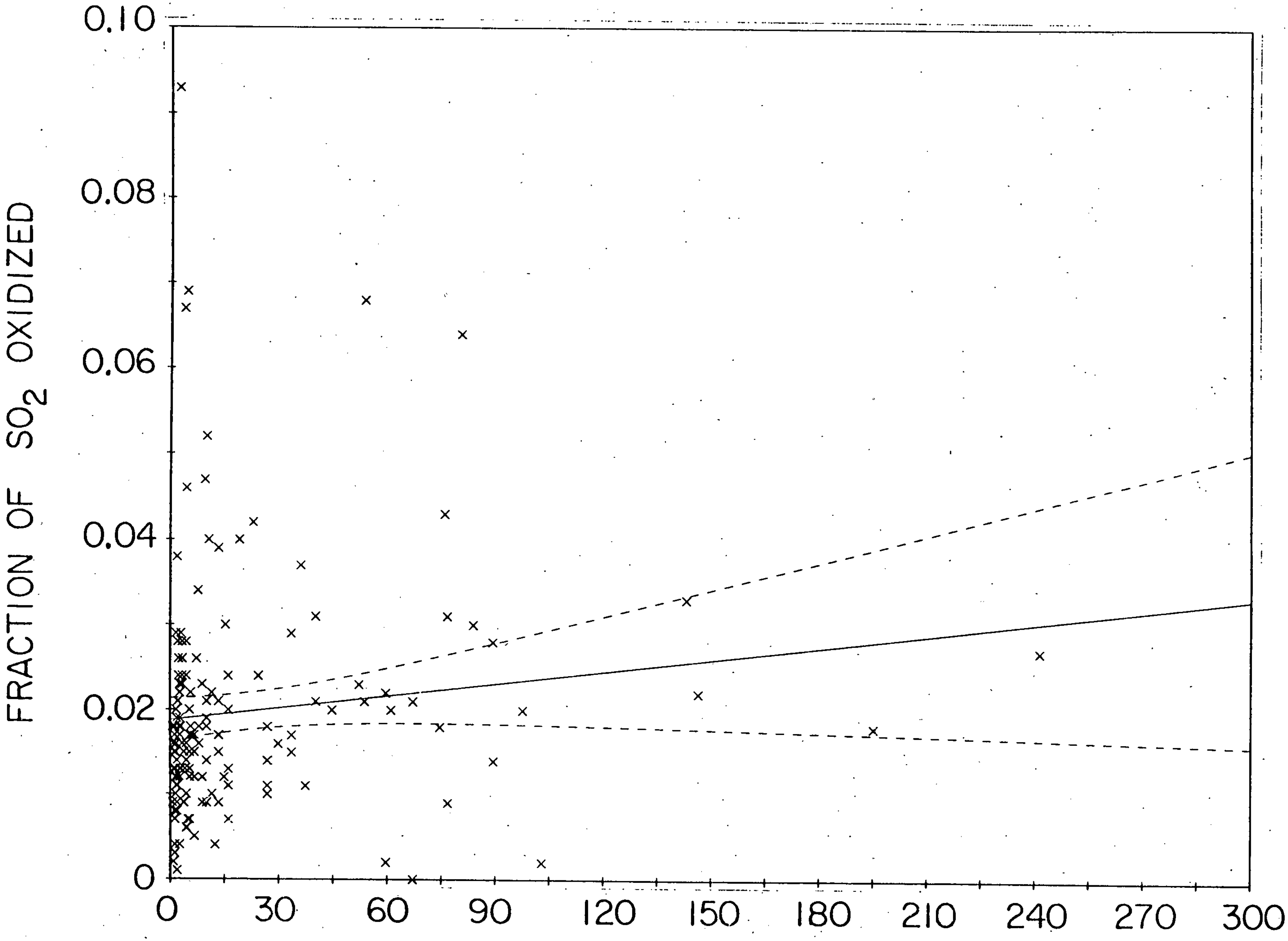


FIG. 5 PLUME AGE (minutes)

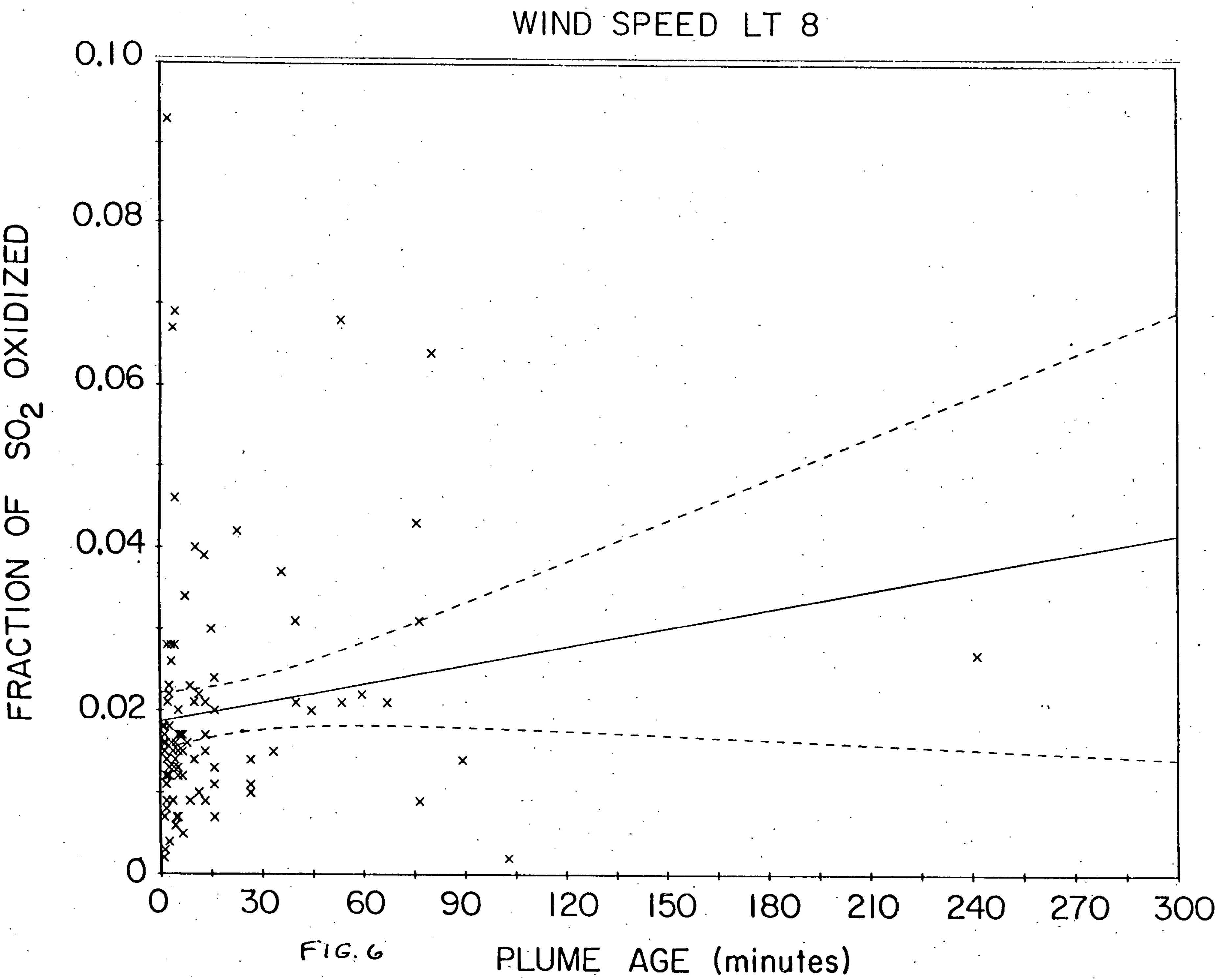
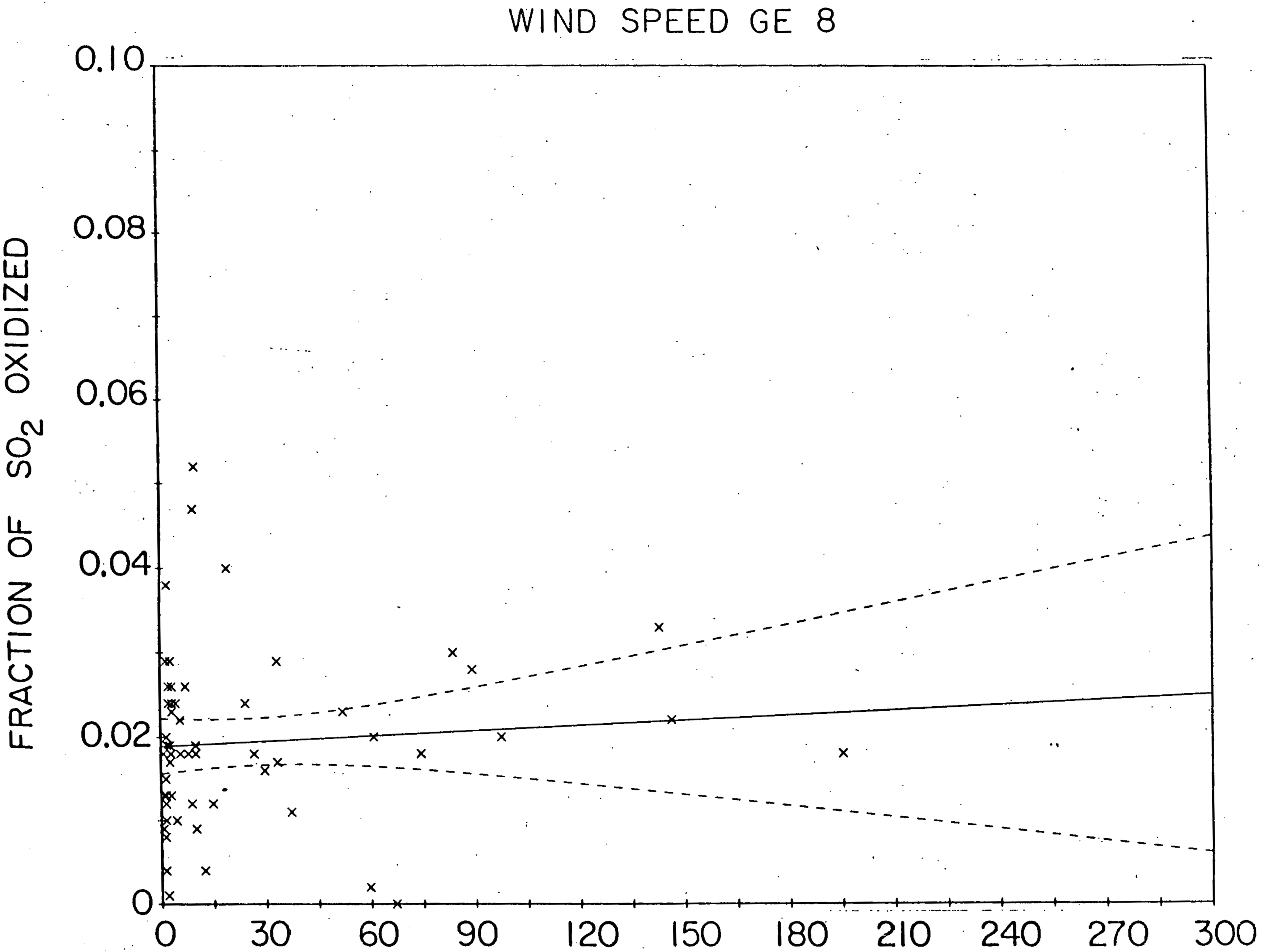

WIND SPEED LT 8
FRACTION OF SO_2 OXIDIZED
0.10
0.08
0.06
0.04
0.02
0
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG. 6



WIND SPEED GE 8
FRACTION OF SO_2 OXIDIZED
0.10
0.08
0.06
0.04
0.02
0
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG. 7

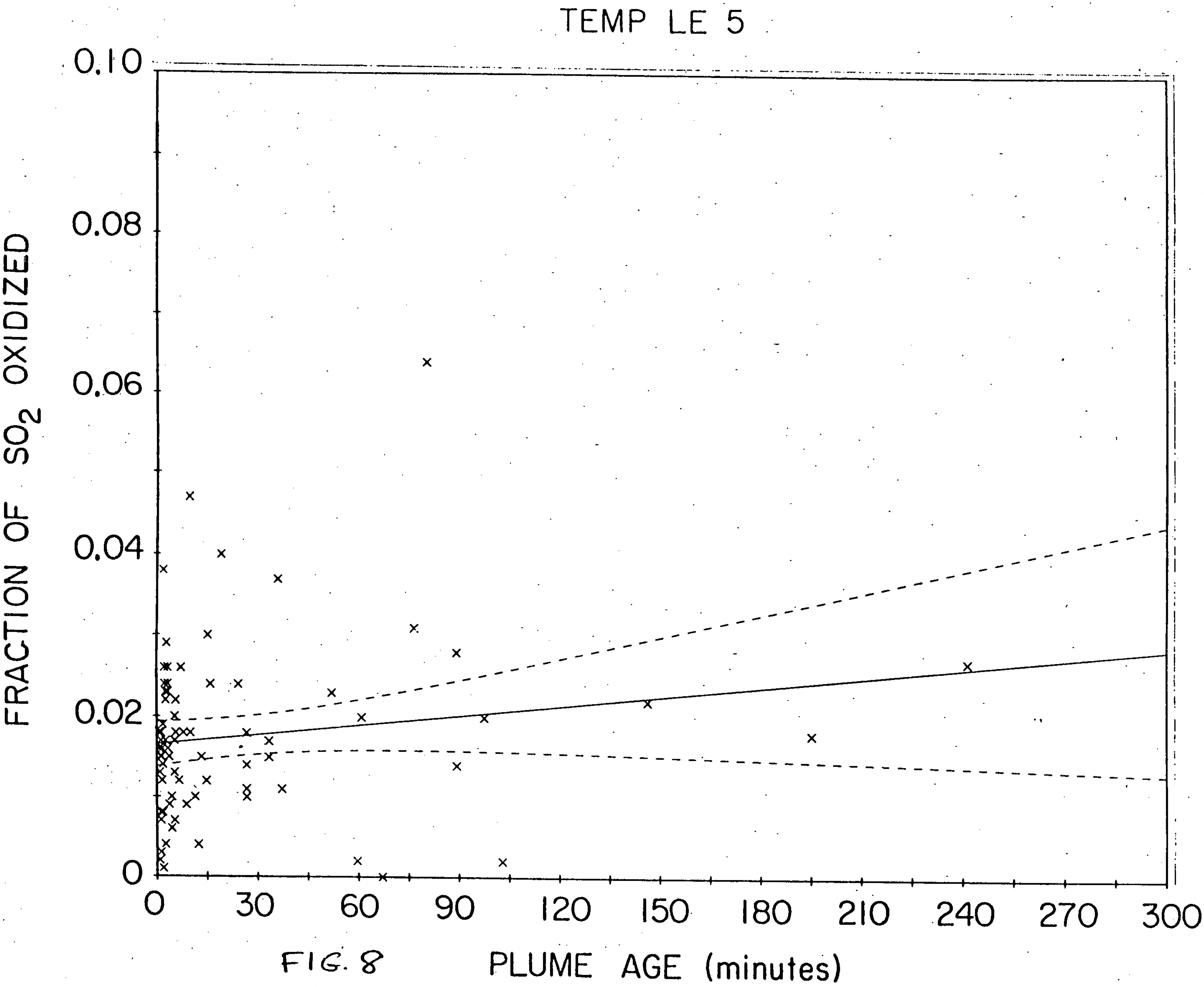

TEMP LE 5
FRACTION OF SO_2 OXIDIZED
0.10
0.08
0.06
0.04
0.02
0
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG. 8

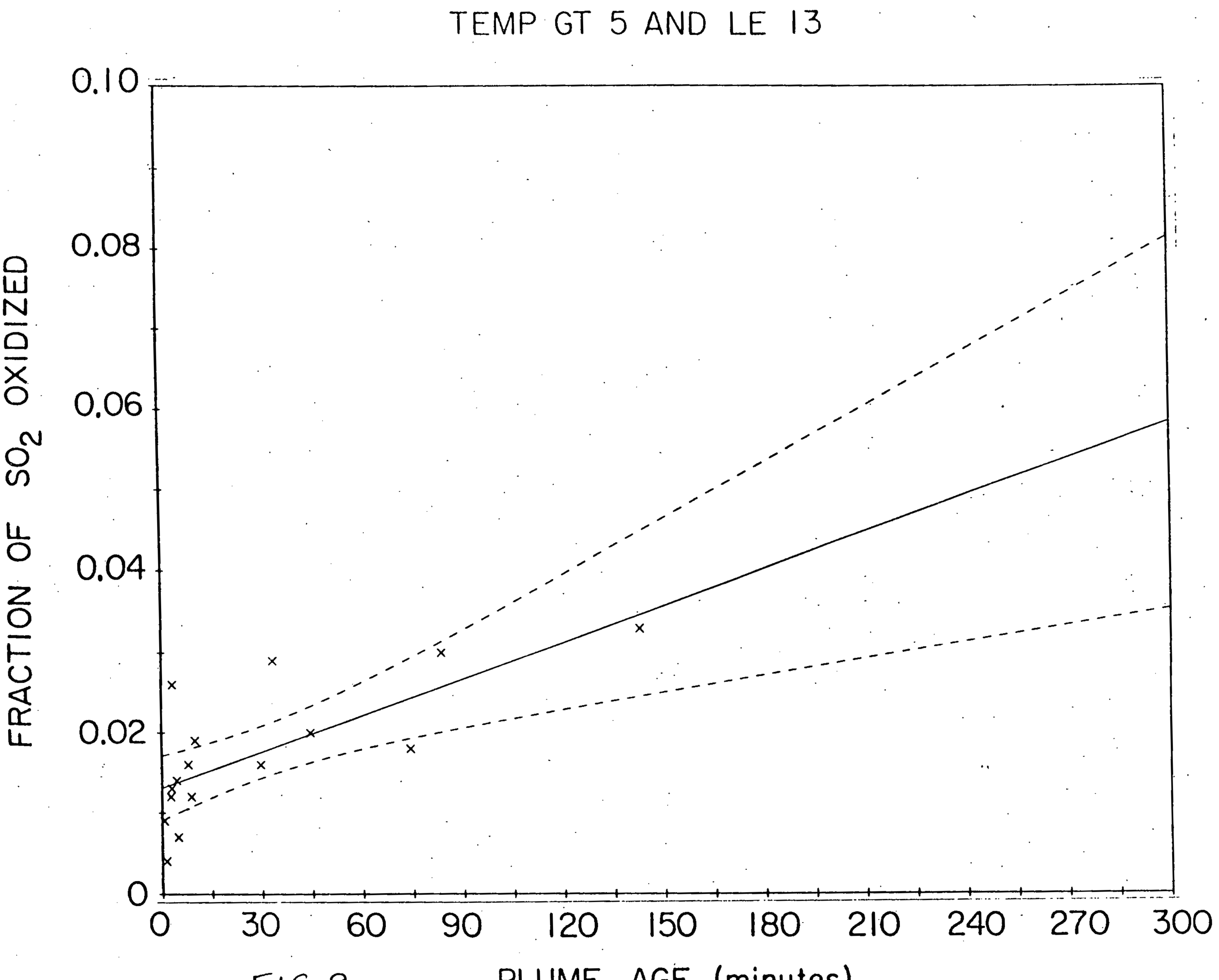
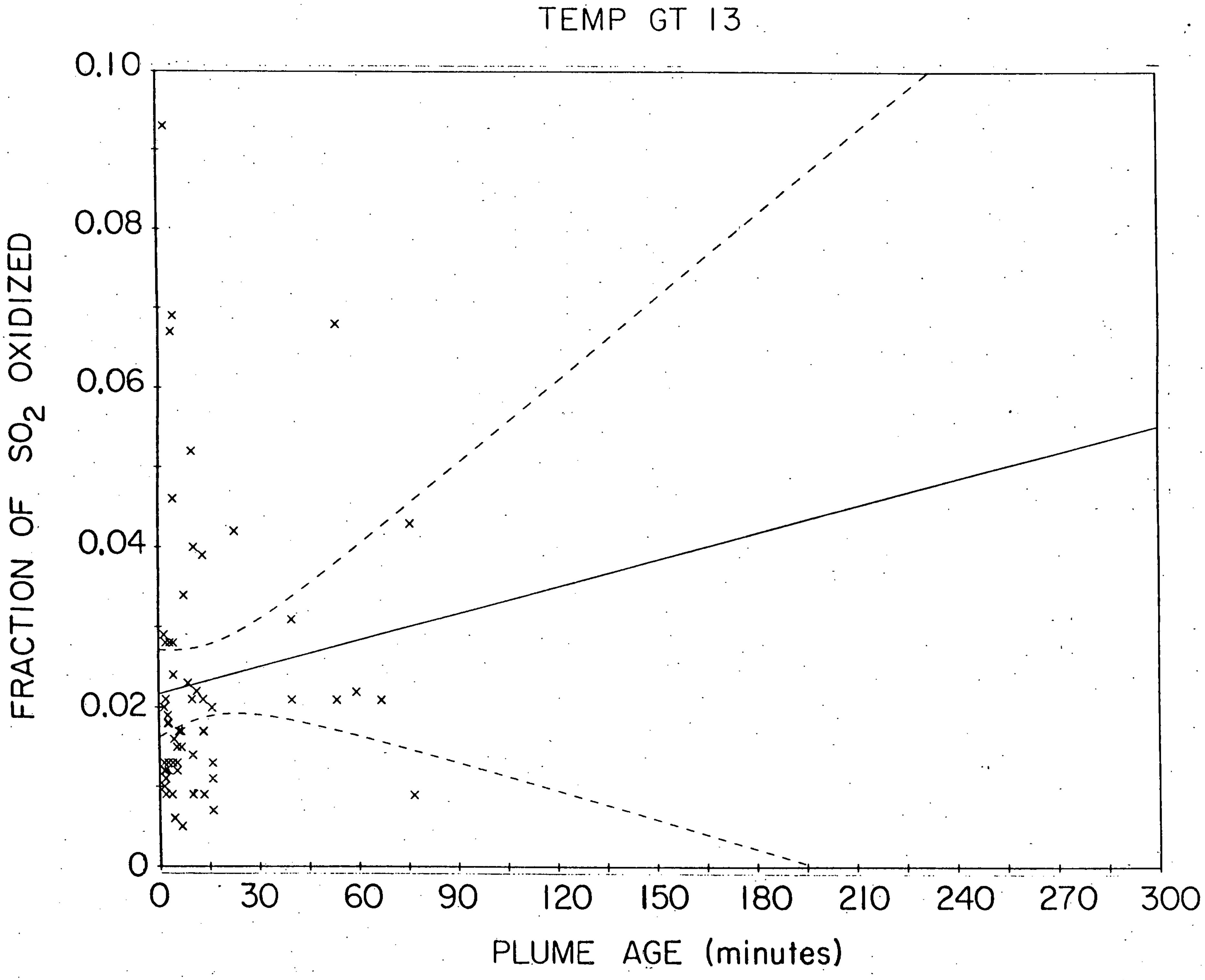

TEMP GT 5 AND LE 13
FRACTION OF SO_2 OXIDIZED
0.10
0.08
0.06
0.04
0.02
0
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG. 9



TEMP GT 13
FRACTION OF SO_2 OXIDIZED
0.10
0.08
0.06
0.04
0.02
0
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG. 10

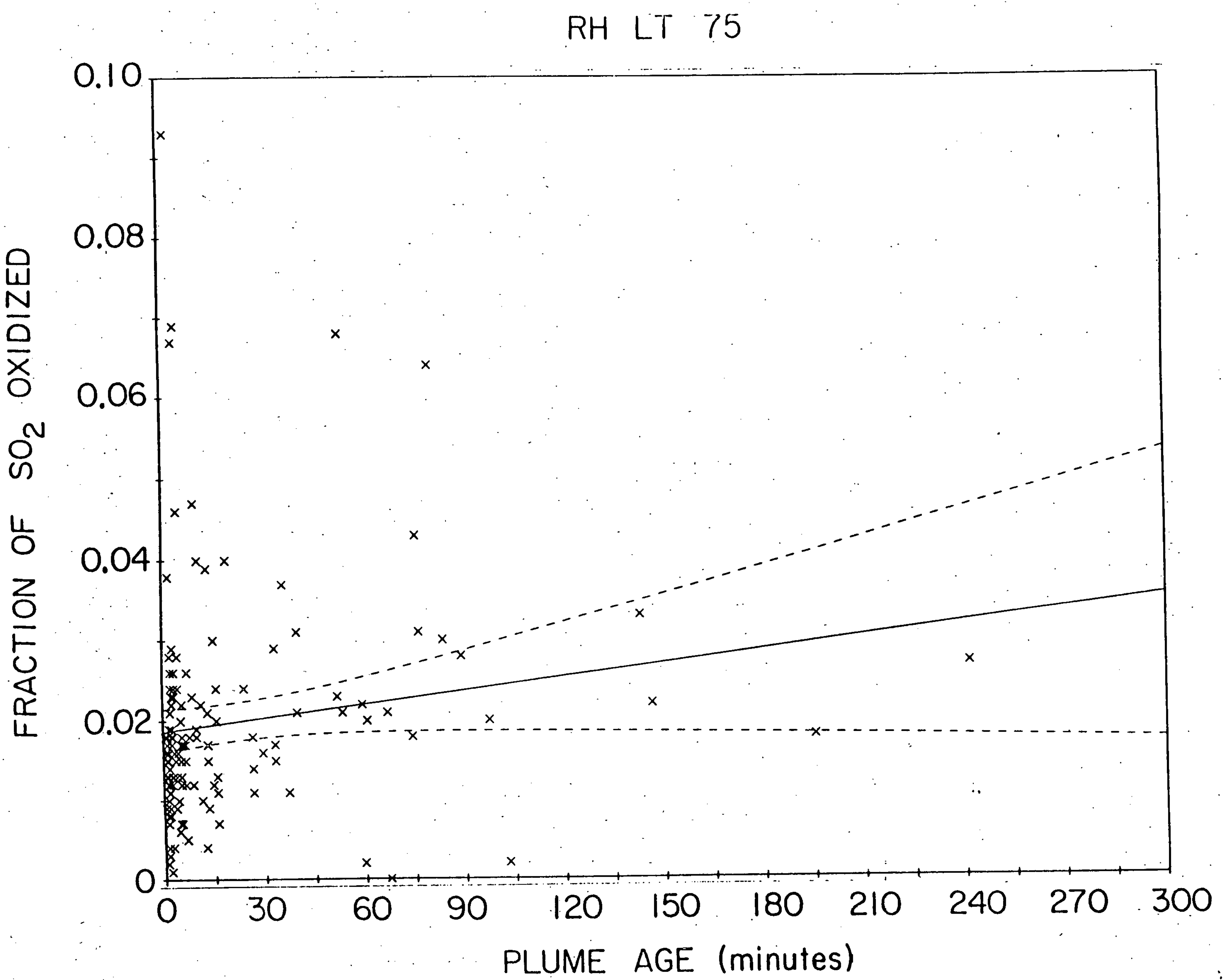

RH LT 75
FRACTION OF SO_2 OXIDIZED
0.10
0.08
0.06
0.04
0.02
0
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG. 11

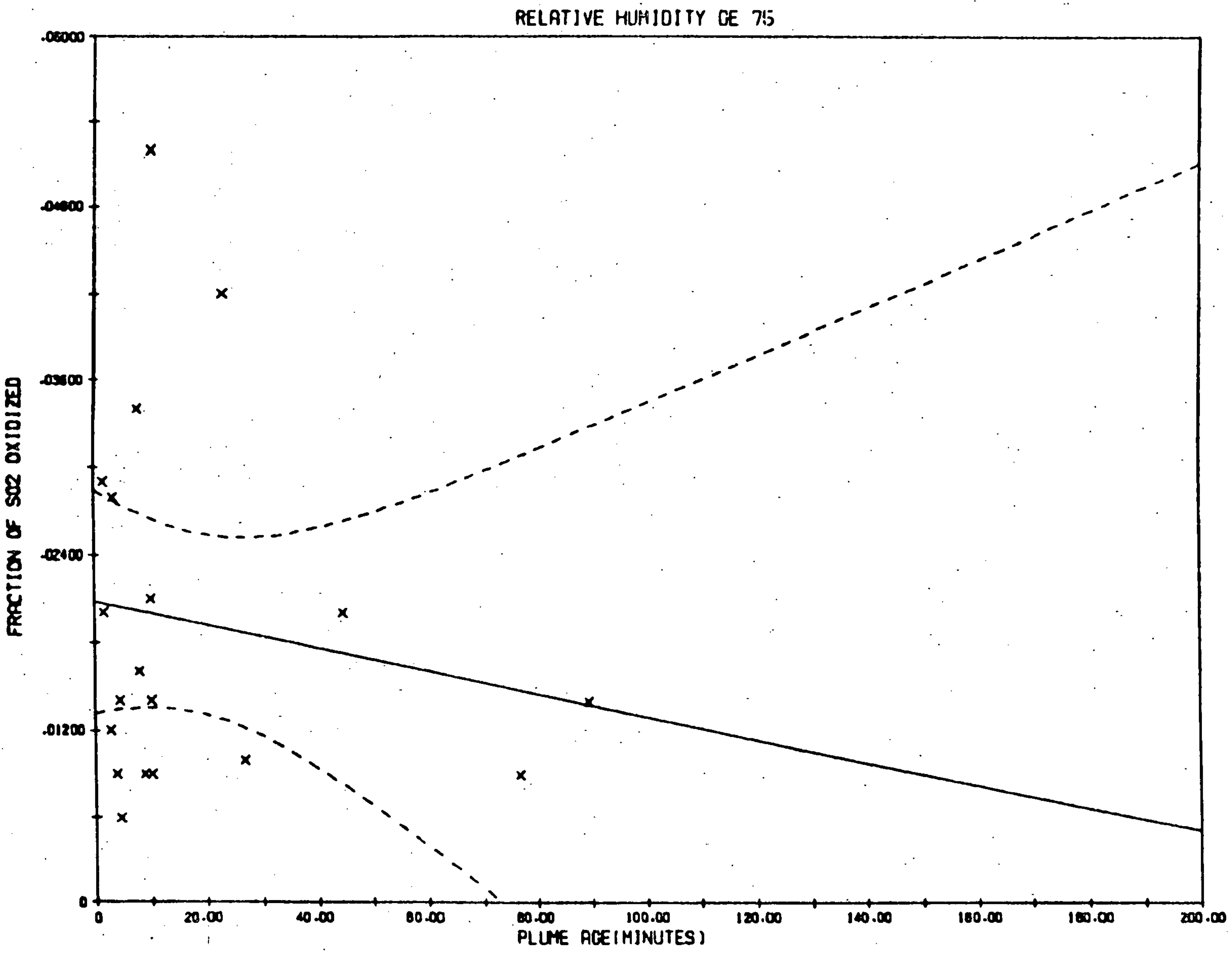

RELATIVE HUMIDITY GE 75
FRACTION OF SO2 OXIDIZED
.06000
.04800
.03600
.02400
.01200
0
0
20.00
40.00
60.00
80.00
100.00
120.00
140.00
160.00
180.00
200.00
PLUME AGE(MINUTES)

FIG. 12

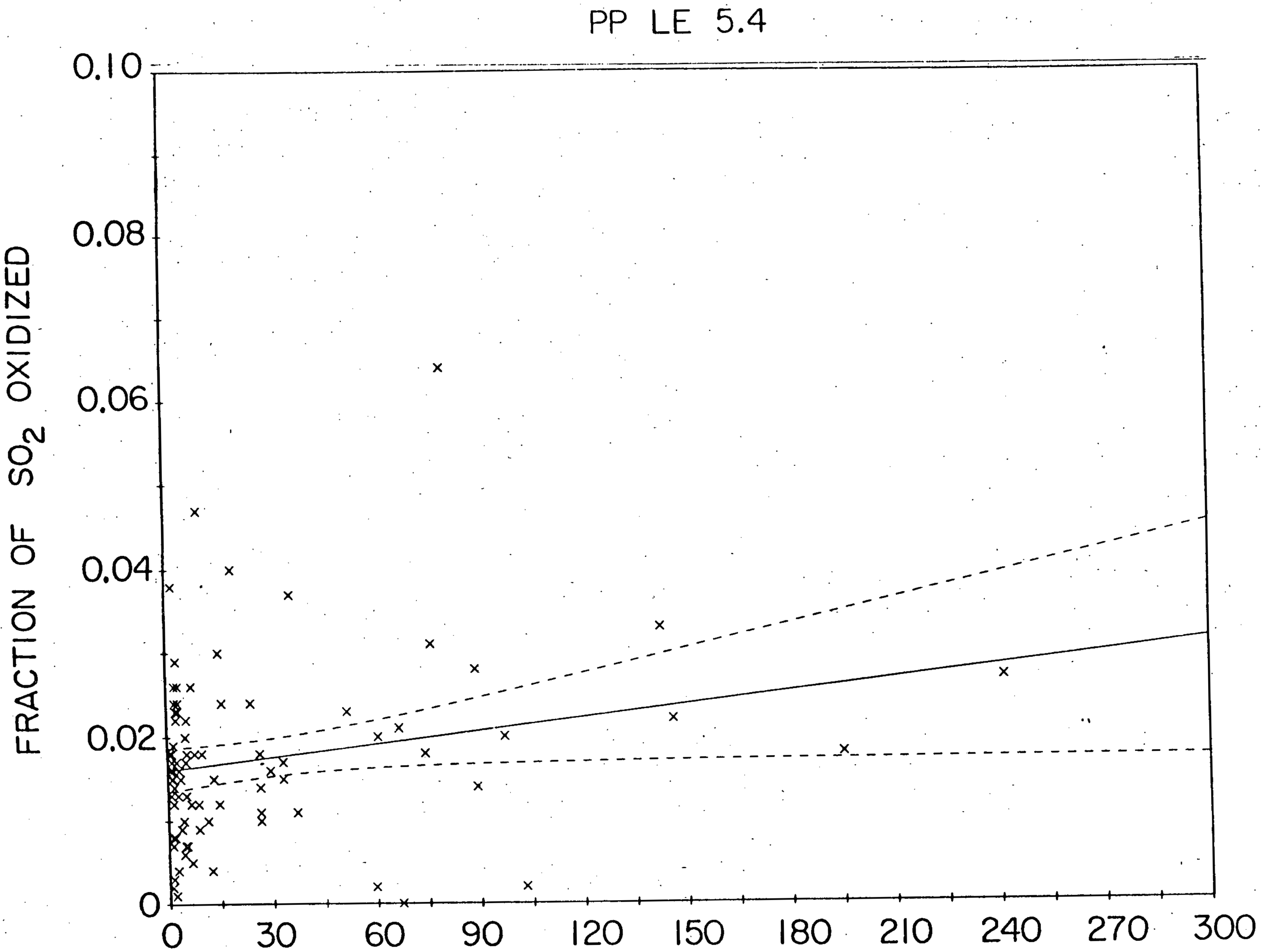

PP LE 5.4
FRACTION OF SO_2 OXIDIZED
0.10
0.08
0.06
0.04
0.02
0
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG. 13

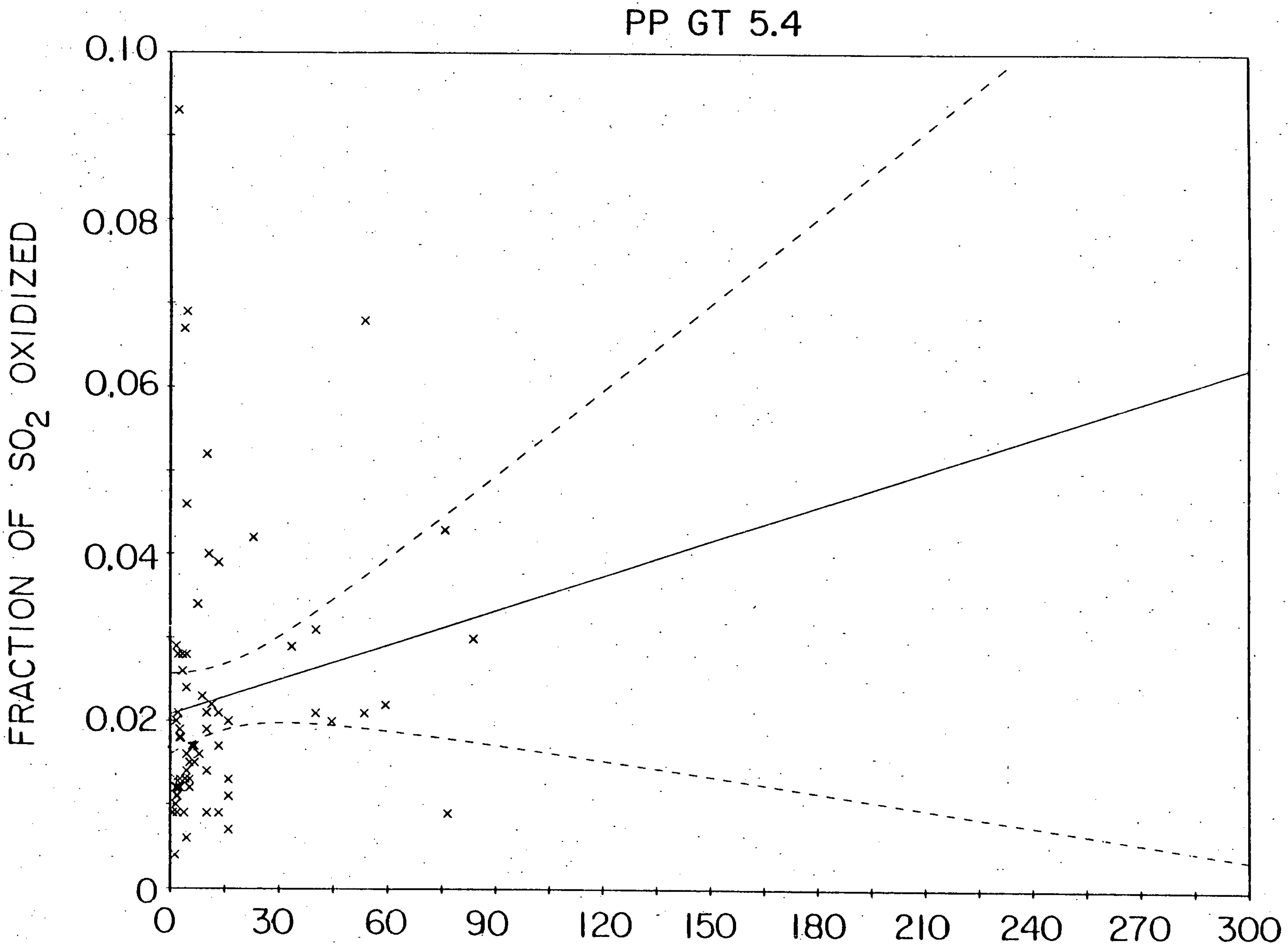


FIG. 14 PLUME AGE (minutes)

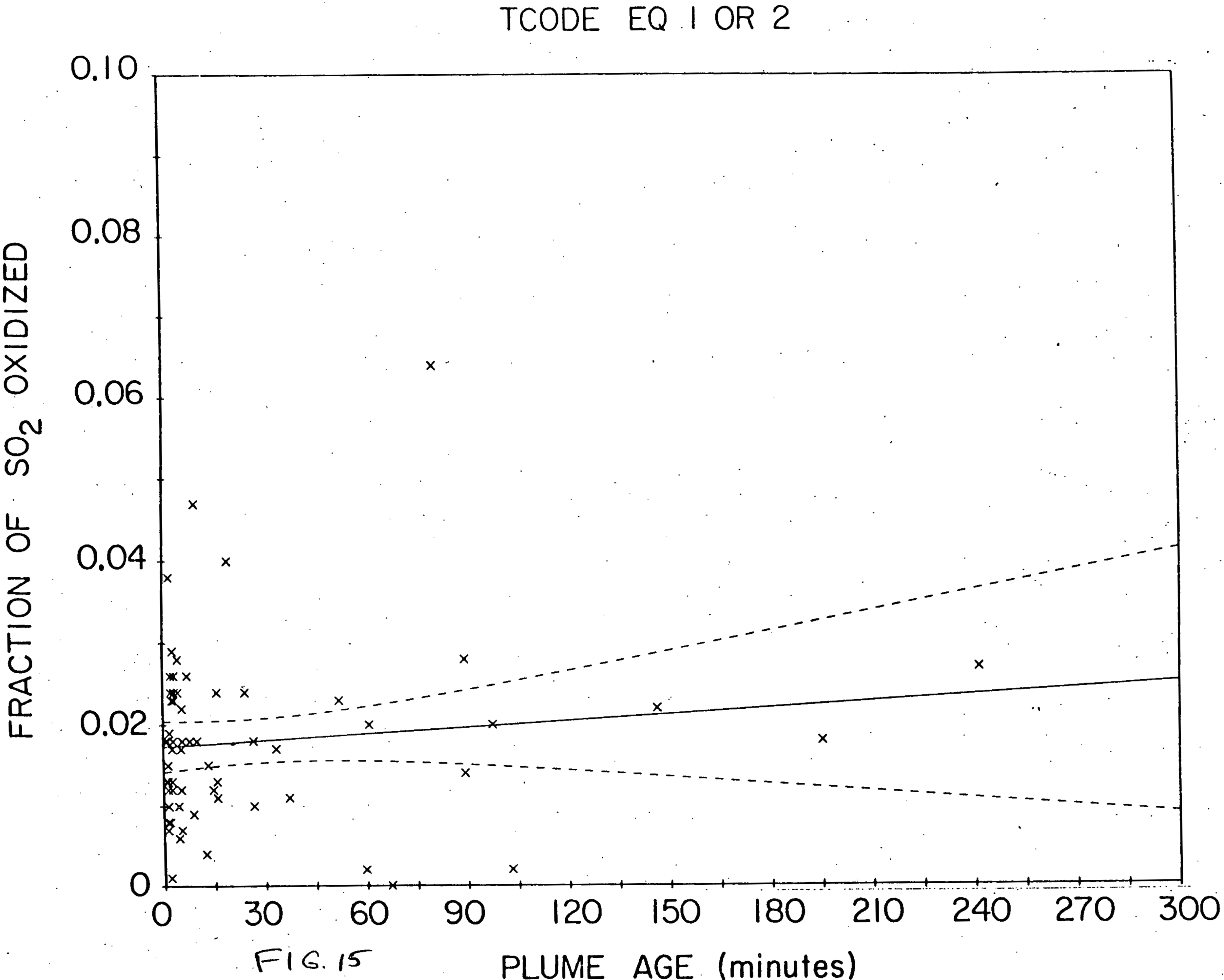

TCODE EQ 1 OR 2
FRACTION OF SO_2 OXIDIZED
0.10
0.08
0.06
0.04
0.02
0
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG. 15

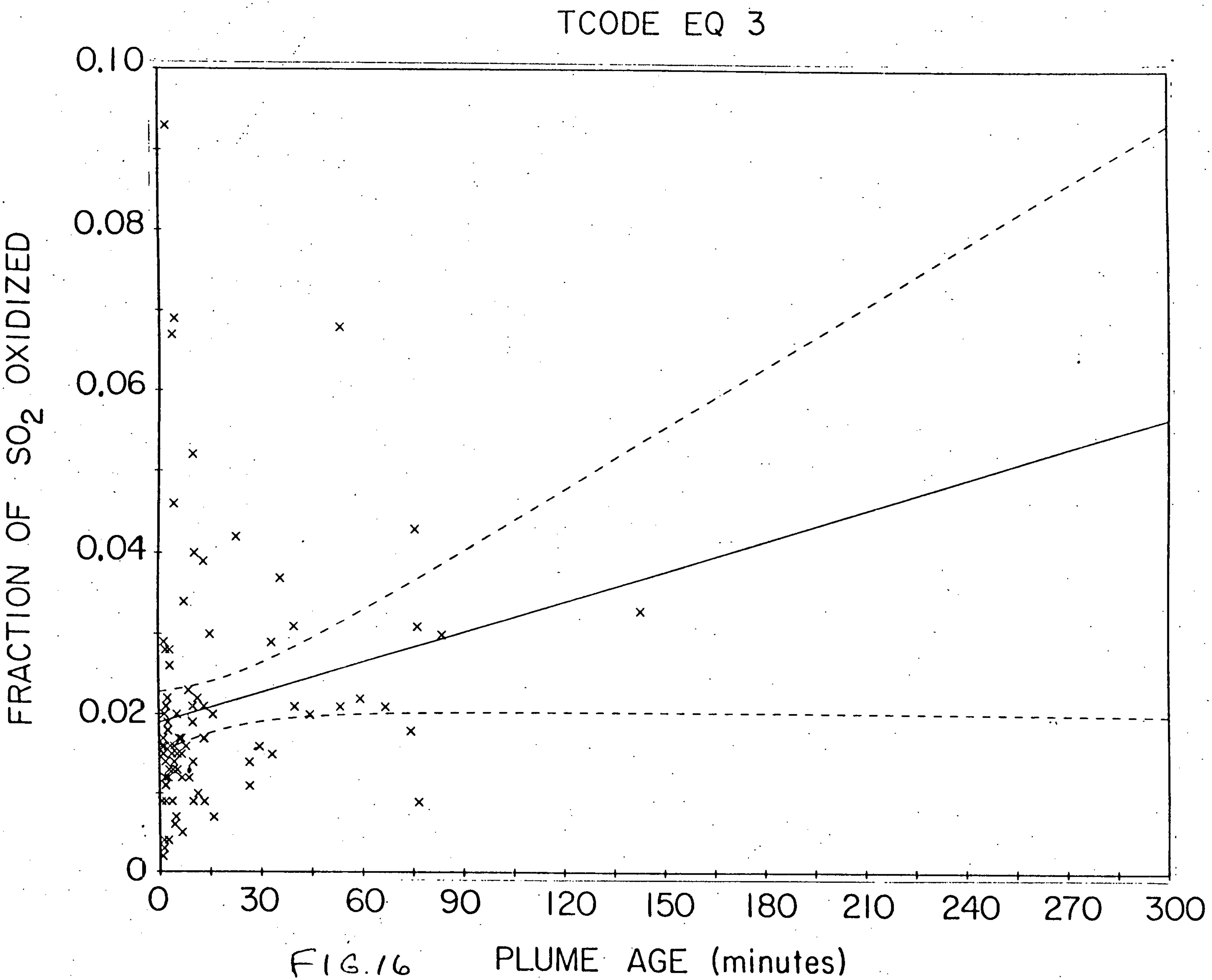

TCODE EQ 3
FRACTION OF SO_2 OXIDIZED
0.10
0.08
0.06
0.04
0.02
0
0
30
60
90
120
150
180
210
240
270
300
PLUME AGE (minutes)

FIG.16