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SOME STUDIES ON STACK EMISSIONS FROM LIGNITE - FIRED POWER PLANTS

BY

G. H. Gronhoyd, P. H. Tufte, and S. J. Selle



*Good results for
SO₂, NO_x and
particulate emissions
and stack gas
flow rates*

LIGNITE COMBUSTION
AP-42 Section 1.7
Reference Number
2

4/93

For presentation at the 1973 Lignite Symposium
May 9-10, 1973
Grand Forks, North Dakota

NO_x:

$$\text{ppm} \times \frac{46}{32.5 \times 10^6} = \frac{\text{lb NO}_x}{\text{SCF}} = 1.2 \times 10^{-7} \text{ ppm}$$

NO_x calculations: (as 110%)

For pulverized coal units:

$$\left[\left(430 \text{ ppm} \right) \left(1.2 \times 10^{-7} \right) \right] \frac{\text{lb NO}_x}{\text{SCF}} \times \frac{112 \text{ SCF}}{\text{lb liquid}} \times \frac{2000 \text{ lb}}{\text{ton}}$$

$$= 11.6 \text{ lb/ton (average)}$$

range from 8.1 - 14 lb/ton

For cyclone fired plants:

$$(625)(1.2 \times 10^{-7})(112)(2000) = 16.8 \text{ lb/ton}$$

ranging from 15 - 18.8 lb/ton

Spreader ~~stacker~~ results not considered typical and should not be used.

SOME STUDIES ON STACK EMISSIONS FROM
LIGNITE-FIRED POWERPLANTS

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by

G. H. Gronhova^{1/}, P. H. Tufte^{2/}, and S. J. Sello^{3/}

INTRODUCTION

National stack emission limits for new stationary sources of greater than 250 MM Btu/hr input were promulgated by the Environmental Protection Agency in 1971 (2)^{4/}. The standards include limits for SO₂, NO_x and particulate matter and are applicable to all plants constructed or significantly modified after August 17, 1971. In addition, there are state, and sometimes local, regulations pertaining to emissions from new and existing powerplants.

Little information is available in the literature on the quantity and quality of stack emissions from boilers burning North Dakota lignite. For this reason NO_x limits for lignite-fired plants were not specified in the National Emission Standards. Lignite from the Northern Great Plains Province is currently used in plants totalling about 1,200 MW capacity, and plants now under construction will increase the total to 2,000 MW by 1975. During the past 8 years, but most actively since 1970, the Bureau of Mines has conducted field tests in which data on furnace exit and stack emissions from lignite-fired plants has been obtained. This report presents data on SO₂, SO₃, NO_x and solid particulate emissions obtained in these field tests. These data should be of value to those responsible for the design, procurement or operation of air pollution control devices for lignite-burning plants.

POWERPLANTS TESTED

Tests were conducted at three pulverized coal (pc)-fired plants, one cyclone-fired plant, and one spreader-stoker-fired plant, each burning lignite from a different mine in North Dakota. Information on each of the plants tested and the source of lignite burned is given in table 1. This report includes data from a total of 46 test days, 39 of which were with pc-firing.

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^{4/} Underlined numbers in parentheses refer to items in the list of references at the end of this report.

ACKNOWLEDGMENT

The authors wish to acknowledge the cooperation of Otter Tail Power Co., Basin Electric Coop., Central Power Electric Coop., and Minnesota Power Coop. in permitting us to conduct these tests at their plants. Without their support the tests would not have been possible.

CALCULATION METHODS AND TEST PROCEDURES

General

The principal emphasis of the tests reported here was on the measurement of SO_2 emissions and their relationship to sulfur level in the coal burned. For such a study a complete ash and sulfur balance on the boilers tested would have been desirable. However, the difficulty and expense involved in collecting, weighing and sampling large quantities of ash (15-20 tons per hour for a 200-MW unit) makes a complete sulfur balance quite impractical. These SO_2 studies are, therefore, based solely on sampling of the coal feed to the boiler and analysis of SO_2 in the flue gas.

Calculation Method

The SO_2 emission per million Btu input, and the pct of input sulfur in the lignite that is emitted as SO_2 , can be calculated from the coal and the flue gas analyses without knowing the weights of coal burned or the flue gas produced. Given the carbon, sulfur, and heating value of the coal and the Orsat analysis of the flue gas, a balance on the carbon, assuming that all of the carbon in the coal appears as CO_2 and CO in the flue gas, yields the following equations:

$$\text{lb } \text{SO}_2/\text{MM Btu} = 5.33 \times \frac{C}{(\text{CO}_2 + \text{CO})} \left(\frac{\text{ppm}}{\text{HHV}} \right)$$

$$\text{S.E.} = 2.67 \times 10^{-3} \times \frac{C}{(\text{CO}_2 + \text{CO})} \left(\frac{\text{ppm}}{S} \right)$$

where:

ppm = parts per million of SO_2 in the flue gas, volume per volume on a dry basis;

$\text{CO}_2 + \text{CO}$ = pct in flue gas from Orsat analysis;

C = pct carbon from the ultimate analysis;

HHV = higher heating value of the coal;

S.E. = pct of input sulfur in the coal emitted as SO_2 .

A typical North Dakota lignite (2) has 65 pct carbon, 10 pct ash, and 10,800 Btu on a dry basis. Using these values the lb SO₂/MM Btu can be calculated for various sulfur levels. These data along with corresponding ppm SO₂ in the flue gas, based on 30 pct excess air, have been plotted in figure 1. For each sulfur level the percentage of SO₂ removal required to meet the 1.2 lb/MM Btu national emission limit is given by the lines of pct removal. For example, (illustrated by the dashed lines on the chart) a lignite having 1 pct sulfur content on a dry basis would have an equivalent SO₂ in the flue gas of 1.85 lb/MM Btu or 837 ppm. A 37-pct reduction in SO₂ would be required to meet the national emission limit.

The NO_x emission rate reported as NO₂ can be calculated by the same method as for SO₂, as given below:

$$\text{lb NO}_x/\text{MM Btu} = 3.83 \times \frac{C}{(\text{CO}_2 + \text{CO})} \left(\frac{\text{ppm}}{\text{HHV}} \right)$$

where:

ppm = parts per million of NO_x in the flue gas, volume per volume on a dry basis.

Coal Sampling and Analysis

In sampling the lignite feed to the boilers during these tests, the objective was to collect a representative sample which would give an accurate value of the sulfur input during the SO₂ sampling period. The coal handling equipment and the facilities for sampling varied from plant to plant. The usual sampling procedure was to collect grab samples from each coal feeder at 1/2-hour intervals and combine these samples for determination of average sulfur content.

Special tests were run at both the Leland Olds and Milton R. Young plants to determine the variability of sulfur content in the lignite. ~~At the Leland Olds plant~~, grab samples were collected at four 15-minute intervals simultaneously from the two storage silos that were physically farthest apart. Each sample was split into four aliquots, two of which were analyzed by one laboratory and two by another. The results of the sulfur determinations at the two laboratories are shown in table 2 and the average of the four determinations for each sample are plotted in figure 2. The relative differences in sulfur contents of lignite between feeders at any given time or from a given feeder in a 15-minute interval approached 100 pct. Within- and between-laboratory results were usually within the ASTM tolerances of 0.05 and 0.10 percentage points, respectively. Similar results were found for the Milton R. Young plant.

These results show the difficulty in obtaining a representative coal sample for sulfur balances. This is a factor which is too often overlooked in powerplant testing. Variations in results for the SO_2 tests reported here could be attributed, in part, to non-representative coal samples despite the emphasis placed on coal sampling in all tests.

SO_3 , SO_2 Sampling Procedure

Sulfur trioxide and sulfur dioxide determinations were made by single point sampling in the flue gas ducts in each of the boilers tested. Usually the samples were collected in the duct between the boiler exit and the air heater, although some were collected after the air heater, and in one case samples were collected from the stack.

The apparatus used for SO_3 and SO_2 determination was similar to that described by Lisle and Sensenbaugh (3). The flue gas is drawn through a condenser maintained at 140° to 194° F where the SO_3 is selectively condensed and collected. The flue gas then passes through a bubbler containing a 3 pct H_2O_2 solution. The resulting sulfuric acid is titrated to a methylpurple endpoint using a standard NaOH solution.

Orsat analyses were usually taken before and after each 30-minute test for SO_2 . All of the SO_2 , SO_3 and NO_x values have been normalized to a 30-pct excess air basis for ease of comparison.

NO_x Sampling Procedure

The NO_x sampling was usually done at or near the SO_2 sampling location. The procedure used was the Phenoldisulfonic acid method, specified as "Method 7" by the EPA in the Federal Register (2). Because the test is more difficult to perform than the SO_2 test, fewer determinations were made.

Particulate Sampling and Study

During most of the tests, samples of fly ash were collected for determination of chemical analysis, particle size distribution, and, in later tests, for electrical resistivity. The fly ash samples were obtained from mechanical dust collector hoppers, pilot or commercial electrostatic precipitator hoppers, or from aspirated flue gas samples. Particle size analysis was done by the Bahco method and laboratory electrical resistivity measurements were by a method similar to that given in ASME PTC-28 (1). In the later tests two "in-situ" methods were used for resistivity studies. Details of these test procedures have been published (4).

RESULTS AND DISCUSSION

Summary of SO₃ Results

A total of 51 determinations of SO₃ content were made at the various lignite-burning plants. In most of these tests a trace of SO₃ was detected but in no case did it exceed 1 ppm, even with SO₂ levels as high as 1,395 ppm. These results differ considerably from those published for Eastern bituminous coals where it is normal for the SO₃ content to be 1-2 pct of the SO₂ content. The lack of free SO₃ in the flue gas from lignite-fired boilers is believed to be due to the presence of reactive alkali in the fly ash. Any SO₃ produced rapidly reacts with the alkali in the fly ash to produce sulfates.

Summary of SO₂ Results

Data on the coal analysis, ash analysis and SO₂ in the flue gas for all of the 46 test days covered in this report are given in table 3. Included are data collected during 39 test days with pc firing, 5 days with cyclone firing, and 2 days with spreader-stoker firing. Figure 3 shows a frequency distribution of pounds of SO₂/MM Btu input for the test days which include all variations in coal analysis, ash analysis, and firing method. For 17 of the test days, the SO₂ emissions levels were between 1.0 and 1.5 lb/MM Btu and for another 16 they were between 1.5 and 2.0 lb/MM Btu. For 13 test days, the SO₂ emissions were below the 1.2 lb/MM Btu national limit. There were only 7 test days in which the SO₂ was above 2 lb/MM Btu, at which level a 40-pct reduction would be required to meet the 1.2 lb/MM Btu level.

Figure 4 shows a frequency distribution of the test days that the percentage of sulfur in the lignite emitted as SO₂ (referred to as S.E.) was in a given range. Results are shown separately for each method of firing. In 29 of the 39 test days with pc-firing, S.E. was in the range of 60 to 90 pct. The minimum value was 48 pct and the maximum was 100 pct. Some of the reasons for these variations will be discussed in later sections.

With cyclone firing, S.E. was in the 80-100 pct range for 4 days and in the 70-80 pct range for 1 day. Other things being equal, one would expect less retention of the sulfur with the ash in a cyclone furnace because of the smaller percentage of ash which is carried through the boiler as fly ash. That portion of the ash that is melted to slag contains very little sulfur.

Two test days were on a spreader-stoker-fired plant. In one test, S.E. was 29 pct and in the other it was 42 pct. The lignites burned during these tests were very low in sulfur, 0.59 and 0.56 on a dry basis, and the ash had high sodium oxide contents of 10.9 and 8.6 pct. These factors combined to produce the very low emission

rates which cannot be assumed to be typical for spreader-stoker-firing. The grate ash from a spreader-stoker unit would not be expected to retain much sulfur and the percentage of ash carried over is less than for pc-firing, so less sulfur retention would be expected.

Prediction of SO₂ Emissions

The retention of sulfur in lignite fly ash is due to the presence of alkali constituents calcium, magnesium, sodium and potassium in reactive form. Calcium and sodium sulfates have been identified in lignite fly ash by X-ray diffraction in significant amounts. Water solubility stoichiometric data suggests all soluble sodium and potassium in the fly ashes are present as sulfates, while only a portion of the soluble calcium is as sulfate. The water solubility of magnesium in the fly ash is highly pH sensitive and sulfate in solution does not increase as magnesium dissolution increases with lowered pH, suggesting there is little MgSO₄ in a typical lignite fly ash. Since potassium is generally present in lignites in very small amounts, the alkalis most responsible for SO₂ retention in lignite fly ash are calcium and sodium.

Not all calcium and sodium is available for reaction with SO₂. Electron microprobe analyses of individual fly ash particles has indicated a significant CaO and Na₂O content in most glassy silicate and aluminum silicate particles appearing in lignite fly ash. These particles results from intimate contact of very fine silica and clay with organically-bound calcium and sodium in burning coal particles. This type of particle accounts for the significant fraction of calcium and sodium which remains insoluble in water, even in solutions having low pH. It is believed that the alkali appearing in these insoluble glassy particles is not reactive towards SO₂. Thus inherent silica and clay could be expected to reduce the fraction of the ash that is active in SO₂ pickup.

Field test data from pc-fired units were examined using statistical regression to determine whether correlations existed between ash composition, sulfur content, and the pct sulfur emitted as SO₂. The bivariate relationships between individual constituents sodium, calcium, silicon, aluminum, total ash and sulfur contents in the coal, and pct sulfur emitted as SO₂ are shown in figure 5. The most clearly discernible trends are the increased pct emitted with increased SiO₂ and Al₂O₃ in the coal on a dry basis. The effects of increased sodium and calcium are to decrease pct emitted, however, the correlations are very poor, being practically non-existent for calcium. No correlation between sulfur and pct emitted is seen. A slight increase in pct emitted with increased ash content is indicated, perhaps relatable to increased silica and clay contents in high ash coals.

To improve on the prediction of pct sulfur emitted, multiple linear regression was employed. The basic model tested was of the form:

$$(1) \text{ S.E.} = C_1 (\text{Element A})^W \times (\text{Element B})^X + C_2 (\text{Element C})^Y \times (\text{Element D})^Z + \dots$$

where Elements A, B, C, D, ---, were SiO_2 , Al_2O_3 , Fe_2O_3 , CaO , MgO , Na_2O and S tested in a multiplicity of combinations.

W, X, Y, Z, ---, were exponents ranging from -2 to +2, tested in a multiplicity of combinations.

The most useful correlation developed for the pc-fired test data combined good predictability and simplicity.

$$(2) \text{ S.E.} = -12.7 \frac{\text{CaO}}{\text{Al}_2\text{O}_3} - 48.1 \frac{\text{Na}_2\text{O}}{\text{SiO}_2} + 110.1$$

$$R^2 = 0.711$$

A plot of actual versus predicted pct emission is shown in figure 6.

The R^2 indicates 71 pct of the variance in the data is explained by the correlation, a significant improvement over individual predictors. An R^2 of 100 pct would indicate perfect agreement between the data and the equation.

The effect of each variable on S.E. can be illustrated by considering the change in S.E. with a 1 pct increase in each ash component on a dry coal basis. This would be roughly equivalent to a 10 pct increase on a pct of ash basis. A 1 pct increase in Na_2O (dry coal basis), or a 10 pct increase in the ash, would cause a decrease of 20 pct in S.E. A 1 pct increase in CaO would cause a 10 pct decrease in S.E., a 1 pct decrease in SiO_2 would cause a 6 pct increase in S.E., and a 1 pct increase in Al_2O_3 would cause a 10 pct increase in S.E. It should be noted that in other regression relations developed, the effect of sodium relative to calcium may be increased to a 3:1 ratio. The generally greater effect of sodium reflects its greater reactivity toward SO_2 .

Thus, one may conclude that the prediction formula confirms quantitatively the evidence presented initially that alkalis are responsible for SO_2 removal and silicates and clays would retard removals by tying up alkalis. The ratio of CaO to Al_2O_3 suggests that CaO reacts preferentially with clay, while Na_2O would react more readily with silica as mechanism for decreasing alkali reactivity.

The fact that the model suggests that S.E. is not a function of sulfur level deserves some discussion. Firstly, it should be emphasized that sulfur was considered in all possible combinations in the model, as were the other elements. In no instance did the inclusion of sulfur in the prediction increase R^2 more than a trivial

amount. Secondly, if one considers that S.E. has coal sulfur content as a divisor mathematically (that is, $S.E. \approx \frac{\text{ppm SO}_2 \text{ emitted}}{S \text{ in coal}}$), and then multiplies both sides of equation (2) by pct S dry coal, the apparent dilemma is resolved. S.E. then becomes absolute sulfur emitted, expressed as a function of the ratio of the alkali clay silica ratio and sulfur. It can then be seen that the model suggests sulfur retention is relatable to a first order alkali sulfur reaction scheme.

In figure 6 some data not included in the regression are included as a basis for evaluation. These are data for 1972 Hoot Lake, Beulah lignite tests. All other points on the plot were used to develop the relationship. The correlation explains the Hoot Lake points about as well as those originally included in the regression.

Effect of Sodium on SO₂ Emissions

Sodium has been shown to have the greatest effect of any element on pct sulfur emitted from lignite-fired boilers. This was demonstrated by tests run at the Hoot Lake plant in 1972 in which low and high sodium lignites from one mine were tested for succeeding weeks. The sodium content was the only substantial variable in the tests (see Analysis in table 3) and any effect can be clearly attributed to the sodium change. The results from three test days on each coal is shown in figure 7. The ppm SO₂ decreased from about 800 ppm to about 590 ppm when the sodium was increased from 0.9 to 6.1 pct. The disadvantage of the high sodium content is, of course, the greatly increased ash fouling rate.

Effects of Boiler Load on SO₂ Emissions

Sulfur balance studies conducted on plants burning German Brown coals have shown that the pct of sulfur retained in the ash is greatly influenced by boiler load and gas residence time (6). Most tests reported in this study were at 85 to 110 pct of rated load as shown in table 3. The one exception is at the Leland Olds plant where load was reduced to 55 pct for two test days. The results show no significant change in SO₂ emission as compared to the higher load tests.

Sulfur Retention in Various Ash Fractions

During most of the field tests, samples of ash were collected from various locations in the boiler for chemical and physical analyses. Results of chemical analysis of selected ashes from the various boilers are shown in table 5. Fly ash samples from both the Leland Olds and Hoot Lake plants were collected during special tests in which a pilot electrostatic precipitator was operating on a side stream of flue gas. In both cases the electrostatic precipitator dust is considerably enriched in SO₃ as compared to the cyclone dust collector sample.

In tests conducted at the Hoot Lake plant in 1965-66, four sodium levels in the lignite were studied and fly ash samples aspirated from the flue gas ahead of the air heater in each test. As shown in table 5, the SO_2 contents of ash ranged from 6.8 pct with the lowest sodium lignite to 28.3 pct with the highest sodium lignite. During the high sodium tests, considerable ash was being retained in the furnace because of the extreme ash fouling condition.

The results for the cyclone-fired plant show that again the aspirated fly ash from the furnace exit contained considerably more SO_2 than the fly ash caught in the mechanical dust collector. The bottom slag had only 0.2 pct SO_2 .

Tests conducted on a spreader-stoker-fired boiler during operation on high sodium lignite showed that the dust collector ash had 2.9 pct SO_2 but that the aspirated dust at the dust collector outlet had 38.0 pct SO_2 and 30.5 pct Na_2O . The grate ash had 1.0 pct SO_2 .

To summarize these data, the results show wide variation in the amount of sulfur retained with the ash, depending upon alkali content and temperature history of the ash. The fine fly ash fractions are most enriched in SO_2 , however, without ash balance data one cannot determine the quantity of input sulfur retained in the various fractions. Bottom slag from a cyclone and grate ash from a spreader-stoker retain little sulfur.

Summary of NO_x Results

Figure 8 shows the results of NO_x determinations at the various plants. Shown are the daily averages^x corrected to 30 pct excess air. The levels for the pc-fired plants varied from about 300 to 520 ppm. For the cyclone-fired plant, the levels varied from 570 to 690 ppm, significantly higher, as would be expected from the higher combustion temperatures. The value of 230 ppm for the spreader-stoker plant is surprisingly low. These values can be compared with a limit of 0.7 lb/MM Btu (about 550 ppm) for bituminous coal-fired boilers as specified by the national emission standards. At the present time lignite-fired boilers are specifically exempted from an NO_x limit in the national emissions standards.

Fly Ash Size Distribution

Two of the most important factors affecting design of fly ash control equipment are the fly ash loading and particle size distribution in the flue gas exiting from the boiler. Determination of dust loading was not a part of these studies, but size distribution was determined on fly ash samples aspirated from boiler exit ducts during some of the tests.

Figure 9 shows Bahco size analysis of fly ash extracted from the boiler exit flue gas at a pc-fired, cyclone-fired and spreader-stoker-fired boiler. As expected the spreader-stoker fly ash was coarsest, having a mass median diameter of 44 microns with 18 pct below 10 microns. It has been stated in the German literature on Brown coal firing that cyclone-firing produced a finer fly ash than pc-firing. This was not confirmed by tests run at Milton R. Young and Hoot Lake plants in 1972 as shown in figure 9. The mass median size for the cyclone-fired boiler was 18.5 microns with 33 pct below 10 microns and for the pc-fired boiler the mass median size was 7.8 microns with 54 pct below 10 microns. It should be noted that the cyclone-fired boiler was burning a very low sodium coal during these tests and it is possible that increased volatilization might increase the fine fraction if high alkali lignites were used.

Electrical Resistivity of Fly Ash

One of the most important factors affecting the electrostatic precipitation of fly ash is the electrical resistivity of the ash. A series of laboratory resistivity tests was run on the fly ash from a number of low sulfur Western coals. The results are described in an earlier paper (4). A computer study of the results in that work indicated that the ratio $(CaO + MgO)/(Na_2O + SO_3)$ in the fly ash was the best indicator of resistivity for those samples, with resistivity varying directly as the ratio.

In more recent work, the emphasis has been on "in situ" field resistivity determinations. The effect of sodium in the fly ash is demonstrated further by data from the 1972 Hoot Lake tests, shown in figure 10. The low sodium ash showed a significantly higher (100 X at 350° F) resistivity than the high sodium ash for both the laboratory and "in situ" results. The results also confirm the applicability of the ratio described above, as the higher resistivity fly ash shows the higher value for $(CaO + MgO)/(Na_2O + SO_3)$. The values determined fall within the range for satisfactory precipitator operation despite the low sulfur content of the lignite burned. This is confirmed by the satisfactory performance of the ESP units on the Hoot Lake plant since their startup in the spring of 1972.

SUMMARY AND CONCLUSIONS

Over the past few years, the Bureau of Mines has collected data on stack emissions from various lignite-fired powerplants. Included were tests on pc-fired, cyclone-fired and spreader-stoker-fired boilers operating on lignites having considerable variation in ash content and analysis. Most emphasis was on the study of SO_2 emissions and how they relate to input sulfur level, firing method, ash analysis and other factors. Limited data was collected on NO_x emissions from the various plants. Chemical and physical properties of fly ash as a function of firing method were determined and electrical

resistivity studies of the fly ashes were made. A method is shown for calculating SO_2 and NO_x emission levels and pct of input sulfur emitted as SO_2 without knowing the weights of coal burned or flue gas produced.

The principal conclusions are:

1. The 1.2 lb/MM Btu SO_2 emission limit was exceeded in 33 of the 46 test days. However, with a 40 pct reduction most of the tests would have been under the limit.
2. Significant amounts of sulfur were retained by the ash in most of the tests. The percentage sulfur retained varied from 73 to 0 pct with most of the results being between 10 and 40 pct. By comparison it is commonly stated that retention for bituminous coals rarely exceeds 5 pct.
3. Insufficient data are available to definitely establish a difference in sulfur retention between firing methods, although there are some indications that the retention is less with cyclone firing than with pc firing.
4. The percentage of sulfur retention on ash was not closely reproducible and some of the variation is probably a result of variability in the input sulfur. The results show the extreme importance of adequate coal sampling if the sulfur balance data is to be meaningful.
5. The SO_2 emission is strongly influenced by the sodium content of the coal. Increasing the sodium from 0.9 to 6.1 pct in the ash reduced the SO_2 from 800 to 590 ppm.
6. Sulfur retention in the ash can be predicted from the chemical composition of the ash with some degree of confidence. The computer-derived relationship shows that increasing CaO and Na_2O increases sulfur retention while clay and silica in the ash tend to decrease sulfur retention.
7. Boiler load does not greatly influence SO_2 emissions, based on two tests at 55 pct of rated load on a pc-fired unit.
8. NO_x emissions are greater for cyclone-fired than for pc-fired units. 3
9. The fly ash particle size at the boiler outlet when burning low-sodium lignite is considerably greater for cyclone-firing than for pc-firing.
10. As predicted by a correlation developed in laboratory tests increasing the sodium content of the ash from 0.9 to 6.1 pct reduced resistivity by a factor of 100 in field tests.

REFERENCES

1. American Society of Mechanical Engineers. Determining the Properties of Fine Particulate Matter. ASME PTC-26. New York, N.Y., 1965, pp. 15-17.
2. Environmental Protection Agency. Standards of Performance for New Stationary Sources. Federal Register, v. 36, No. 247, Dec. 23, 1971, pp. 24876-24895.
3. Lisle, E. S., and J. D. Sensenbaugh. The Determination of Sulfur Trioxide and Acid Dew Point in Flue Gas. Combustion, v. 36, No. 7, January 1965, pp. 12-15.
4. Selle, S. J., P. H. Tufte, and G. H. Gronhovd. A Study of the Electrical Resistivity of Fly Ashes from Low-Sulfur Western Coals Using Various Methods. Pres. at Air Pollution Control Assoc. meeting, Miami Beach, Fla., June 18-20, 1972, APCA Preprint 72-107, 31 pp.
5. Sondreal, E. A., W. R. Kube, and J. L. Elder. Analysis of the Northern Great Plains Province Lignites and Their Ash: A Study of Variability. BuMines RI 7158, 1968, 94 pp.
6. Speich, P. Schwefelbilanzuntersuchungen an Braunkohlenkesselanlagen. (Investigations of Sulfur Balance at Lignite-Fired Boiler Plants.) Braunkohle, v. 17, No. 9, September 1965, pp. 364-371.

TABLE 1. - Powerplants included in the study

Plant name:	Hoot Lake	Leland Olds	Wm. J. Neal	Milton R. Young	F. P. Wood
Plant location:	Fergus Falls, Minnesota	Stanton, N.Dak.	Voltaire, N.Dak.	Center, N.Dak.	Grand Forks, N.Dak.
Company:	Otter Tail Power Co.	Basin Electric Power Coop.	Central Power Electric Coop.	Minnkota Power Coop.	Minnkota Power Coop.
Size of installations tested, MW:	50	215	20	235	12
Type of firing:	Pulverized coal	Pulverized coal	Pulverized coal	Cyclone burner	Spreader stoker
Burner location:	Tangential	Front & rear walls	Front wall	Front wall
Source of lignite:	Beulah mine and Gascoyne mine of Knife River Coal Mining Co.	Glenharold mine of Consolidation Coal Co.	Velva mine of Consolidation Coal Co.	Center mine of Baukol-Moonan Inc.	Larson mine of Baukol- Moonan Inc.
Dates of tests:	1965, 1966, 1970, 1972	1970, 1971	1971	1970, 1971, 1972	1971, 1972
Type fly ash collector	Cyclone 1/ pilot ESP	Cyclone pilot ESP	Cyclone	Cyclone	Cyclone
during tests:	Commercial ESP				

1/ ESP = electrostatic precipitator.

TABLE 2. - Sulfur variations at Leland Olds plant

<u>Time, - minutes</u>	<u>Silo</u>	<u>Lab. G</u>		<u>Lab. P.</u>		<u>Av.</u>
0	A	1.49	1.34	1.33	1.41	1.39
0	K	0.76	0.82	0.93	0.82	0.83
15	A	0.79	0.77	0.82	0.85	0.81
15	K	1.42	1.25	1.55	1.56	1.45
30	A	0.96	0.94	0.96	0.96	0.95
30	K	0.91	0.91	0.95	0.94	0.93
45	A	0.90	0.94	0.97	1.01	0.95
45	K	1.37	1.37	1.46	1.46	1.42

TABLE 3. - Summary data on SO₂ emissions tests at lignite-fired plants

Test date	Boiler load, pct of rated	Lignite analysis, percent				Heating value, Btu/lb	Coal ash analysis, pct				No. of tests	SO ₂ in flue gas		Pct of input sulfur emitted
		Proximate analysis, percent			SiO ₂		Al ₂ O ₃	CaO	Na ₂ O	Av. SO ₂ ppm at 30 pct excess air		lb SO ₂ per 10 ⁶ Btu		
		Moisture	Ash	Sulfur									Carbon	
PC-Fired Plants														
Boat Lake Plant - Beulah Lignite														
7/25/65	80	35.6	10.3	1.09	65.4	10,830	14.2	9.8	20.1	8.2	4	420	0.93	48
7/28/65	82	35.9	10.1	1.03	65.4	10,840	14.4	10.2	21.1	9.0	4	450	1.00	52
8/15/65	82	36.7	9.9	1.15	65.4	10,900	17.0	11.4	26.0	1.7	2	800	1.76	84
8/13/65	84	36.6	10.2	1.10	65.8	10,970	19.7	12.0	26.3	1.6	2	830	1.83	92
9/7/65	102	36.1	11.9	1.36	NA ^{1/}	10,570	17.6	11.0	23.0	4.8	2	820	1.87	73
5/2/66	84	36.1	11.8	1.52	64.4	10,600	20.8	10.5	23.3	5.3	2	740	1.65	50
5/3/66	84	36.3	10.8	1.31	64.8	10,720	19.7	10.9	23.1	5.6	4	700	1.55	63
5/4/66	84	36.3	10.8	1.25	65.0	10,710	19.5	10.9	21.6	5.8	4	710	1.58	68
5/5/66	84	36.1	11.8	1.54	64.6	10,740	17.4	9.2	22.0	5.4	4	840	1.86	65
5/6/66	84	35.8	11.8	1.33	65.1	10,690	19.4	10.7	22.6	5.5	4	840	1.88	76
7/9/70	104	34.4	10.4	1.32	65.2	10,910	16.7	10.9	25.2	7.8	2	610	1.34	59
7/10/70	104	34.9	9.8	1.10	65.5	10,910	16.5	10.6	24.9	3.8	3	630	1.37	69
8/11/70	112	34.3	9.8	0.92	64.1	10,670	14.6	10.6	21.2	7.5	1	515	1.14	66
8/12/70	112	34.5	9.8	0.97	64.1	10,670	16.9	9.9	21.5	3.8	8	585	1.29	71
8/13/70	110	34.0	9.8	1.06	64.1	10,670	14.4	10.9	24.2	5.1	3	575	1.27	64
9/12/72	100	35.4	10.1	1.11	65.2	10,780	17.9	12.0	26.5	1.1	5	680	1.51	74
9/13/72	100	35.8	9.9	1.21	64.6	10,740	17.3	12.0	26.2	1.0	13	770	1.70	75
9/14/72	100	35.8	10.0	1.14	64.8	10,780	18.7	11.6	25.1	0.9	13	820	1.81	86
9/15/72	100	35.2	9.5	1.03	65.4	10,800	18.9	12.1	28.2	0.9	10	780	1.74	91
9/18/72	84	35.7	10.9	1.14	63.9	10,530	20.6	11.5	20.3	6.1	12	600	1.34	61
9/19/72	84	35.7	10.3	1.03	64.5	10,680	19.7	11.2	20.0	6.1	22	570	1.27	66
9/20/72	84	35.2	10.4	1.12	64.4	10,650	20.1	11.5	21.2	6.1	9	550	1.22	59
Boat Lake Plant - Gascoyne Lignite														
7/27/70	96	40.1	14.8	1.52	61.3	10,300	32.4	11.6	15.9	3.1	1	1,120	2.45	83
7/28/70	106	41.1	14.8	1.33	61.3	10,300	28.1	12.7	18.9	5.8	4	1,170	2.56	93
7/29/70	108	41.2	14.8	1.38	61.3	10,300	37.1	13.1	16.4	3.0	6	1,360	2.97	100
7/30/70	110	41.8	14.8	1.68	61.3	10,300	32.0	13.1	17.0	3.5	3	1,325	2.90	89
7/31/70	108	41.7	14.8	1.68	61.3	10,300	34.3	13.8	16.7	2.1	3	1,395	3.05	94
8/4/70	110	40.0	14.8	1.71	61.3	10,300	32.3	10.3	15.6	2.0	3	1,300	2.84	86

^{1/} Not available.

TABLE 3. - Summary data on SO₂ emissions tests at lignite-fired plants - Continued

Test date	Boiler load, pct of rated	Lignite analysis, percent				Heating value, Btu/lb	Coal ash analysis, pct				No. of tests	SO ₂ in flue gas		Pct of input sulfur emitted
		Moisture-free basis					SiO ₂	Al ₂ O ₃	CaO	Na ₂ O		Av. SO ₂ ppm at 30 pct excess air	lb SO ₂ per M Btu	
		Moisture	Ash	Sulfur	Carbon									
Iceland Olds Plant - Glenharold Lignite														
8/25/70	93	36.7	10.4	0.63	63.1	10,580	30.7	11.0	20.2	8.8	8	425	0.93	78
8/25/70	93	37.8	11.5	0.65	62.9	10,570	31.4	11.8	17.0	8.0	4	470	1.03	84
8/27/70	84	35.2	11.1	0.61	62.7	10,560	37.5	13.6	16.2	7.7	16	470	1.02	89
9/9/70	56	36.8	11.7	0.85	62.5	10,580	35.1	13.6	17.7	6.0	16	655	1.42	89
9/10/70	55	36.4	11.7	0.84	62.1	10,470	35.7	13.8	17.2	7.0	4	590	1.28	80
10/29/70	92	36.8	12.7	0.75	62.3	10,425	34.3	13.4	18.7	6.2	6	520	1.14	79
10/29/70	100	38.1	12.1	0.74	63.6	10,700	33.0	12.2	19.2	7.8	12	510	1.11	81
4/14/71	100	34.3	13.2	1.09	62.9	10,550	32.8	11.2	17.0	5.1	5	660	1.44	70
Wm. J. Neal Plant - Velva Lignite														
7/27/71	100	NA ^{1/}	9.1	0.57	64.7	10,920	38.1	13.0	22.6	0.9	7	355	0.77	74
7/28/71	100	NA ^{1/}	9.1	0.70	64.7	10,920	46.0	13.6	16.4	0.8	9	525	1.14	89
7/29/71	100	NA ^{1/}	9.1	0.66	64.7	10,920	34.1	14.5	23.4	0.9	12	455	0.99	82
Cyclone-Fired Plant														
Milton R. Young Plant - Baukol-Noonan (Center) Lignite														
11/24/70	106	36.5	11.9	0.90	63.9	10,490	30.9	12.7	19.4	0.7	6	765	1.71	100
4/16/71	92	37.3	12.5	0.93	62.9	10,580	29.1	13.3	21.8	0.8	10	735	1.61	91
9/6/72	106	36.5	20.6	1.11	55.9	9,270	45.8	14.0	11.9	0.4	5	930	2.02	86
9/7/72	106	37.1	19.4	0.97	56.7	9,270	44.8	14.3	13.8	0.4	17	785	1.76	82
9/8/72	106	38.2	13.8	1.10	59.2	9,550	29.9	10.9	19.4	0.7	17	765	1.68	73
Spreader Stoker-Fired Plant														
Franklin P. Wood Plant - Baukol-Noonan (Larson) Lignite														
11/17/71	100	35.0	10.7	0.59	65.7	10,940	32.5	14.1	15.6	10.9	8	130	0.29	27
12/15/72	100	33.0	12.5	0.54	64.7	10,880	32.8	14.4	16.8	8.6	18	194	0.42	43
/ Not available.														

^{1/} Not available.

TABLE 4. - Partial analysis of selected ash samples from
field tests of lignite-fired boilers

	Component, pct			
	CaO	MgO	Na ₂ O	SO ₂
<u>PC-Fired Plants</u>				
Hoot Lake plant, 8/12/70:				
Mechanical dust collector	28.8	9.1	4.2	4.5
Pilot ESP	26.4	8.9	5.3	8.7
Leland Olds plant, 8/28/70:				
Mechanical dust collector	19.9	6.0	6.9	2.3
Pilot ESP	20.1	5.8	7.9	6.0
Hoot Lake plant, 1965-1966:				
Samples aspirated from boiler				
exit duct:				
Test No. 1	32.9	10.3	2.3	6.8
Test No. 2	28.5	8.6	7.2	11.6
Test No. 3	26.2	9.1	9.4	14.1
Test No. 4	20.1	9.3	16.3	28.3
<u>Cyclone-Fired Plant</u>				
Milton R. Young Plant, 1970:				
Mechanical dust collector	32.4	8.8	1.4	3.9
Aspirated from gas to dust collector	31.5	8.4	1.4	6.7
Bottom slag	31.8	9.5	0.3	0.2
<u>Spreader-Stoker-Fired Plant</u>				
Franklin P. Wood Plant, 1971:				
Mechanical dust collector	23.5	6.4	11.8	2.9
Aspirated from dust collector exit gas	3.8	0.9	30.5	38.0
Grate ash	19.3	4.8	8.8	1.0

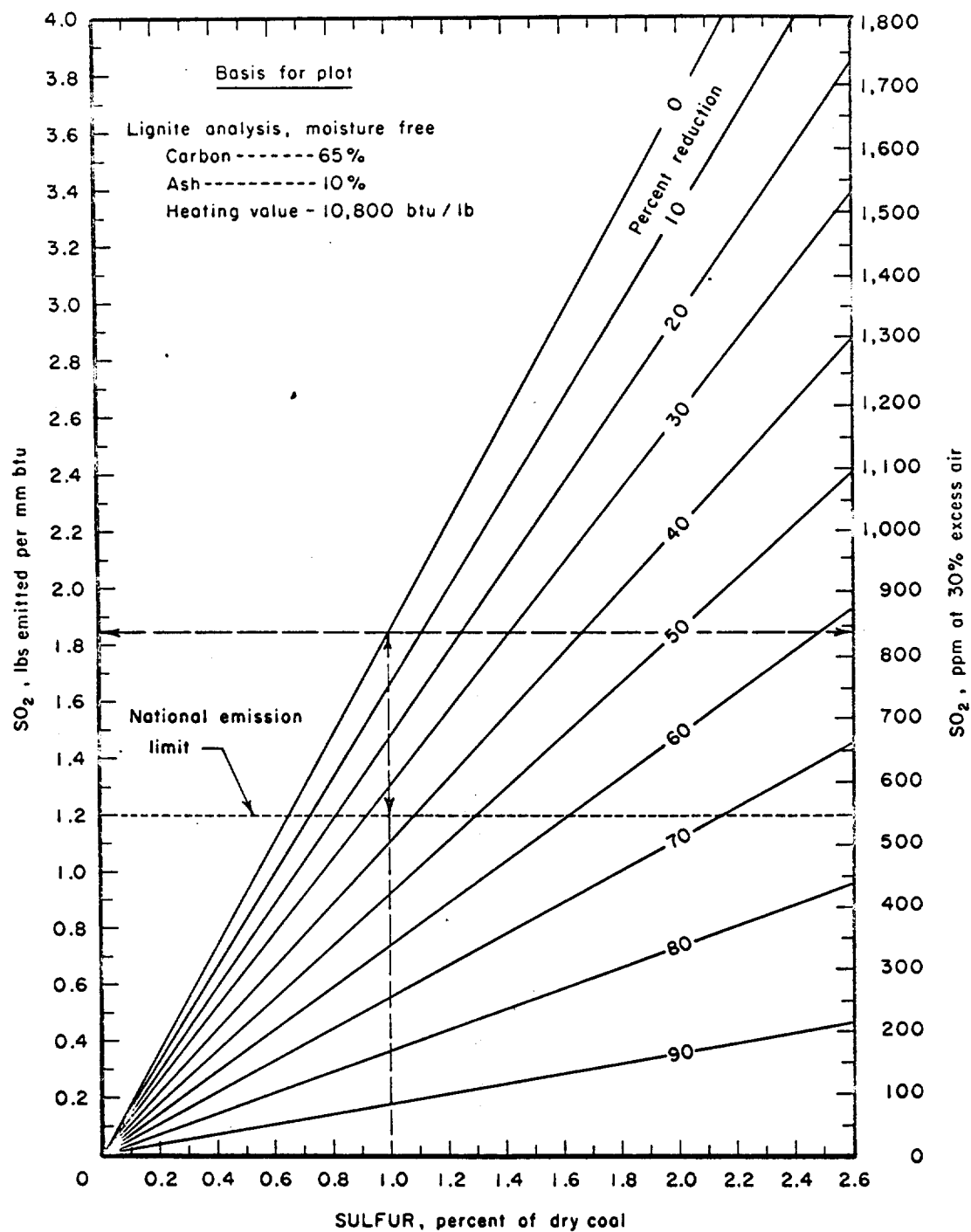


FIGURE 1. - Chart for determination of sulfur emissions and percentage removal required to meet national emission limit.

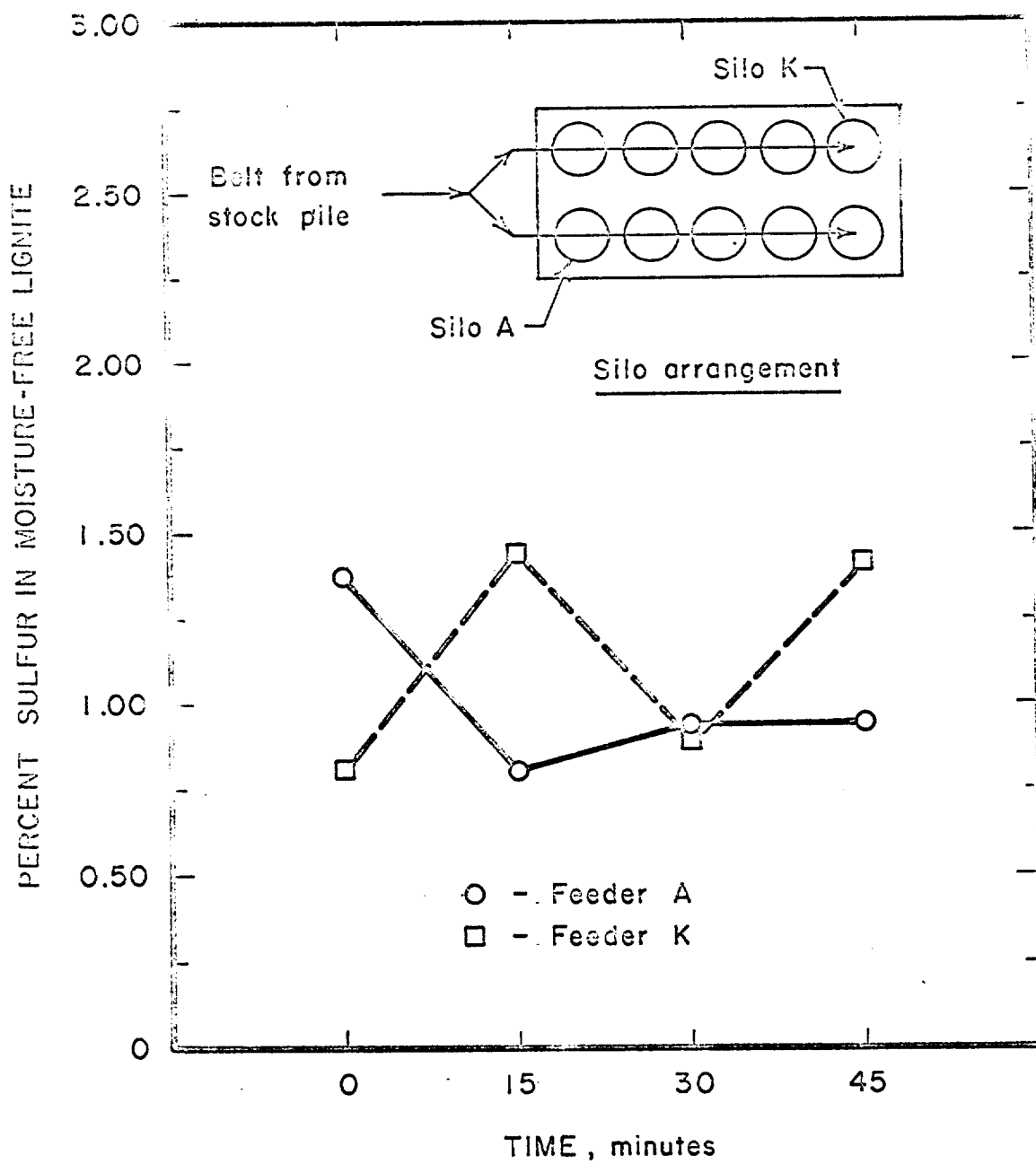


FIGURE 2. - Sulfur variation on lignite with time and sampling point.

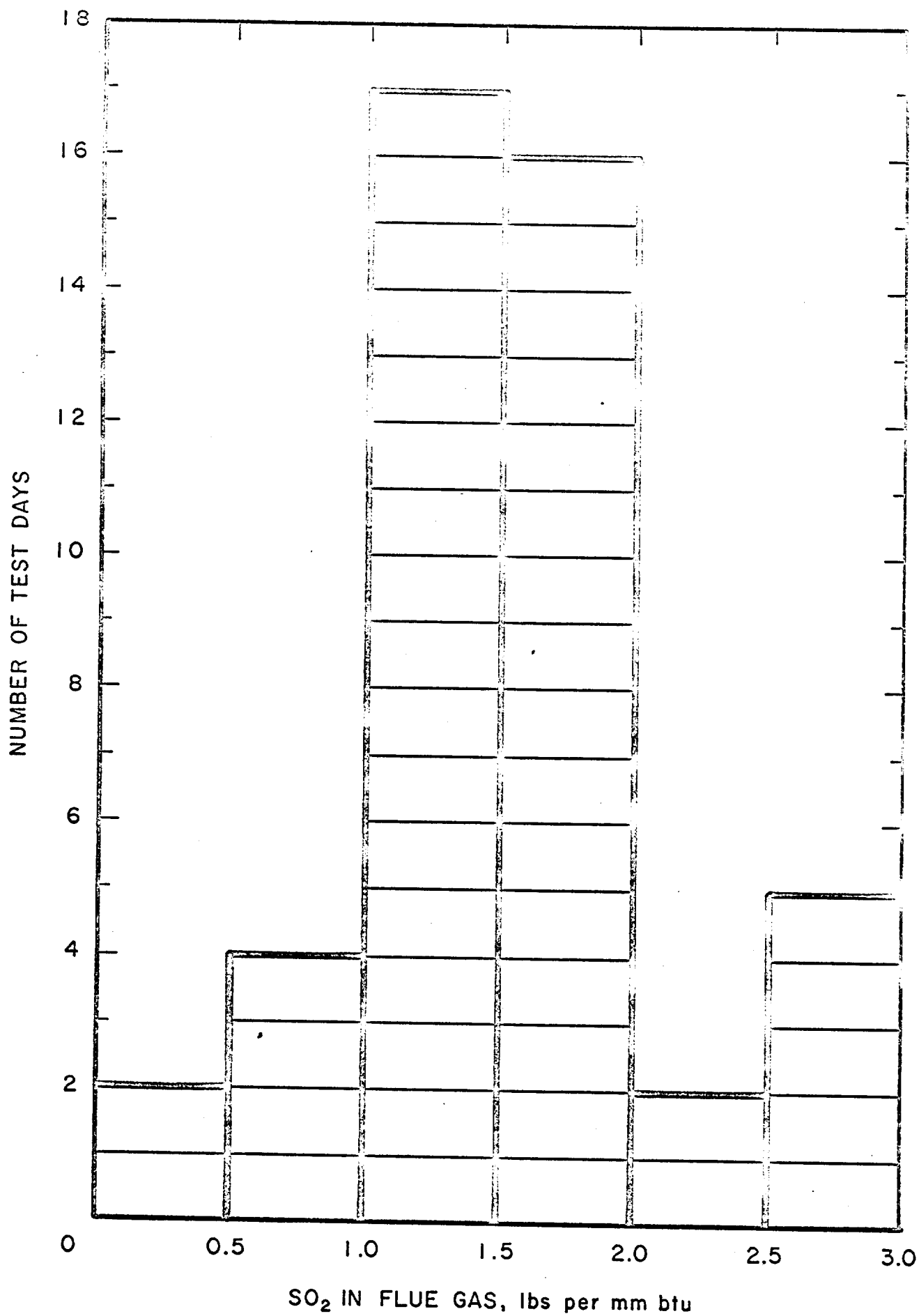


FIGURE 3. - Frequency distribution of SO₂ emission level.

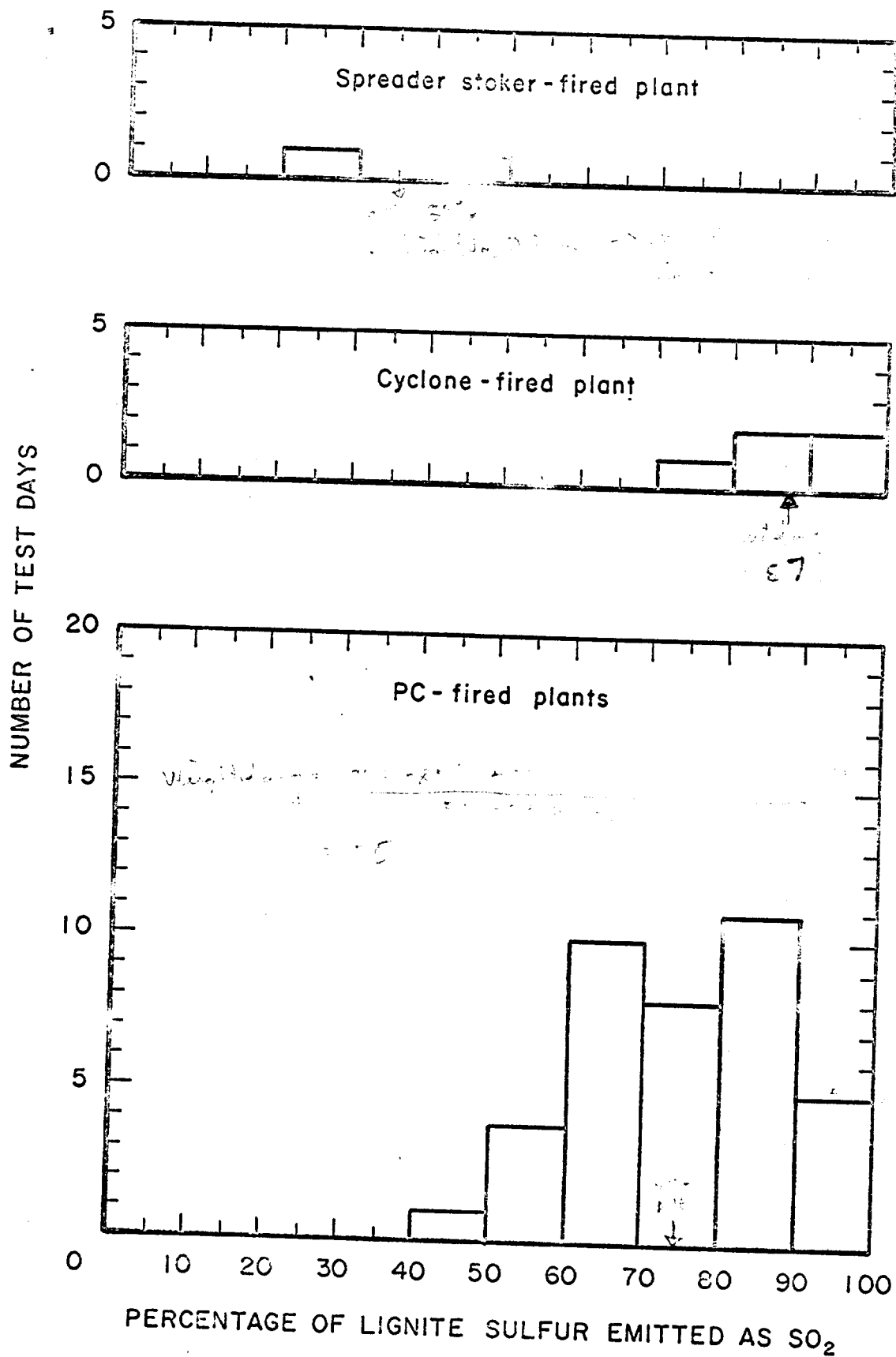


FIGURE 4. - Frequency distribution of sulfur in lignite emitted as SO₂ for various firing methods.

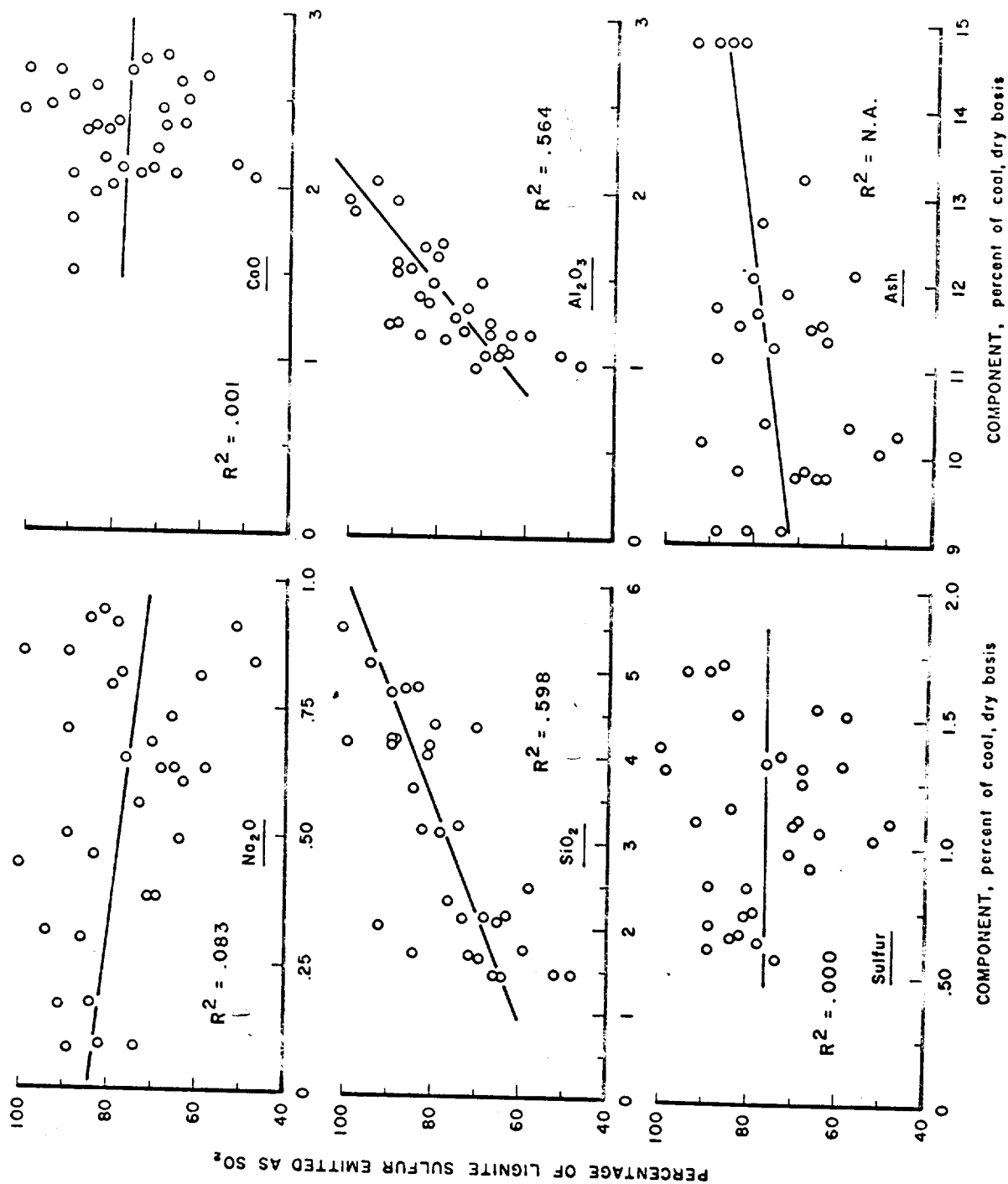


FIGURE 5. - Correlation of coal and ash analysis with S.F. for pe-fired plants.

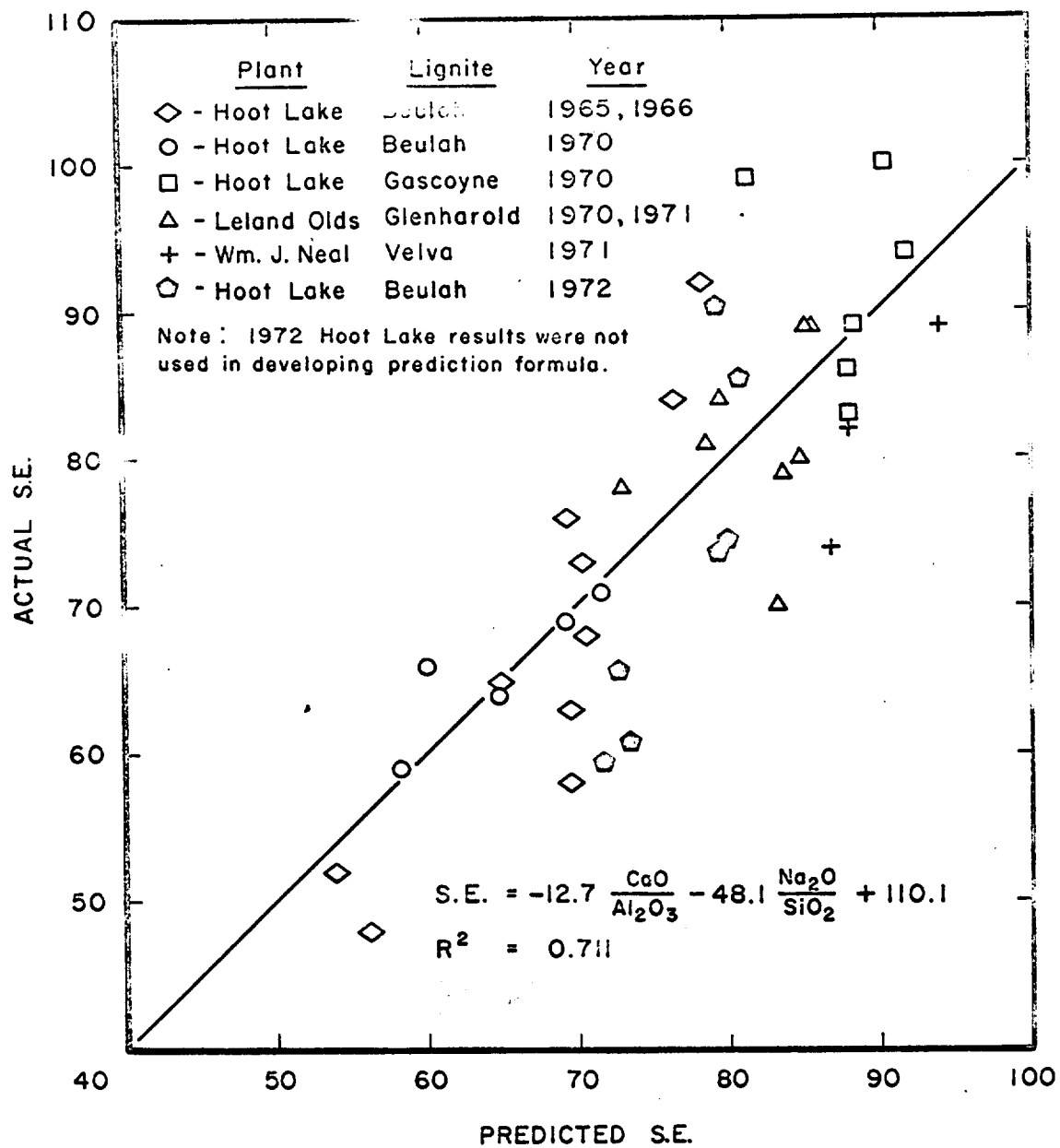


FIGURE 6. - Predicted versus actual S.E. for pc-fired plants.

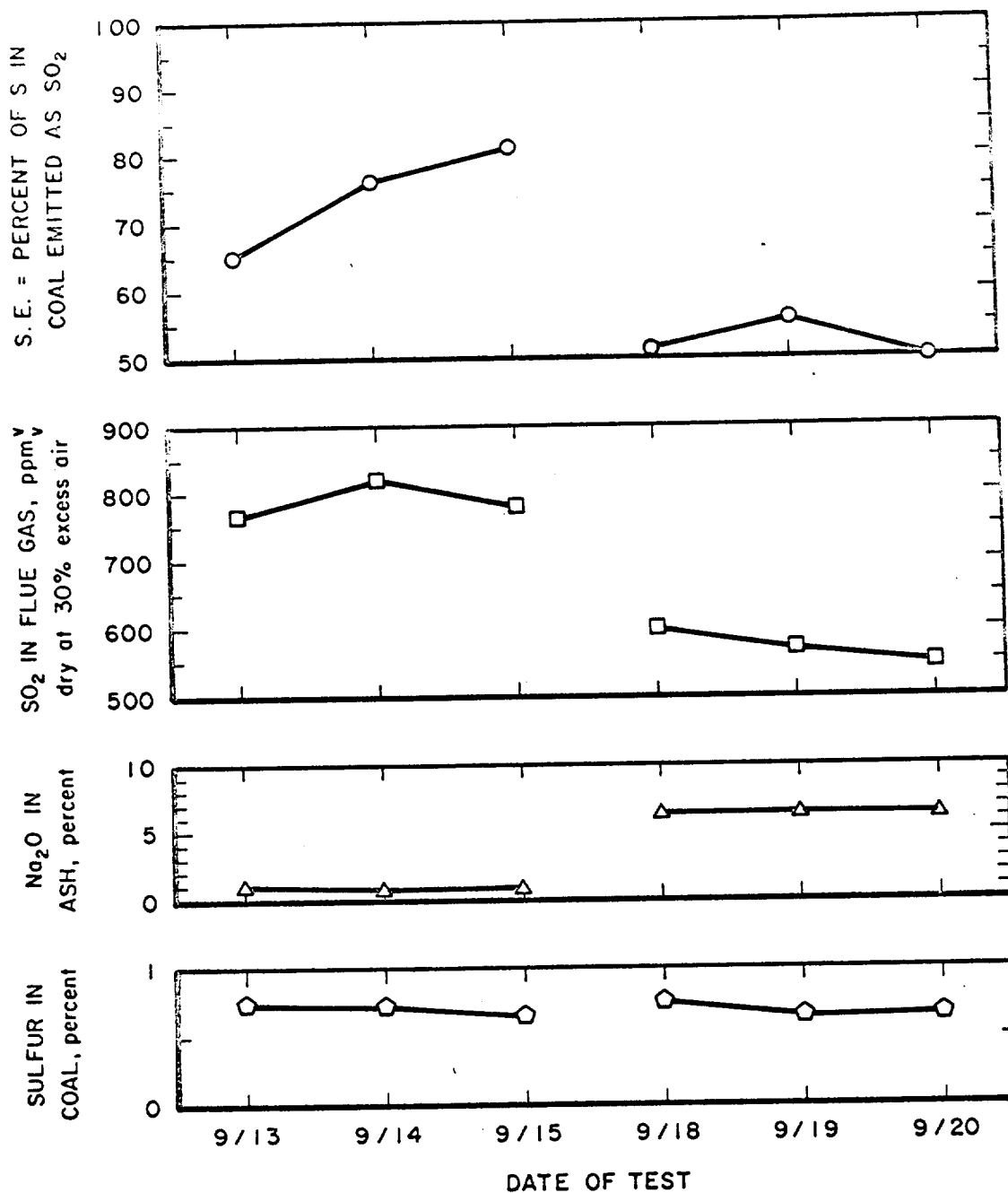


FIGURE 7. - Effect of sodium content of lignite on SO₂ emissions.
Hoot Lake tests, 1972.

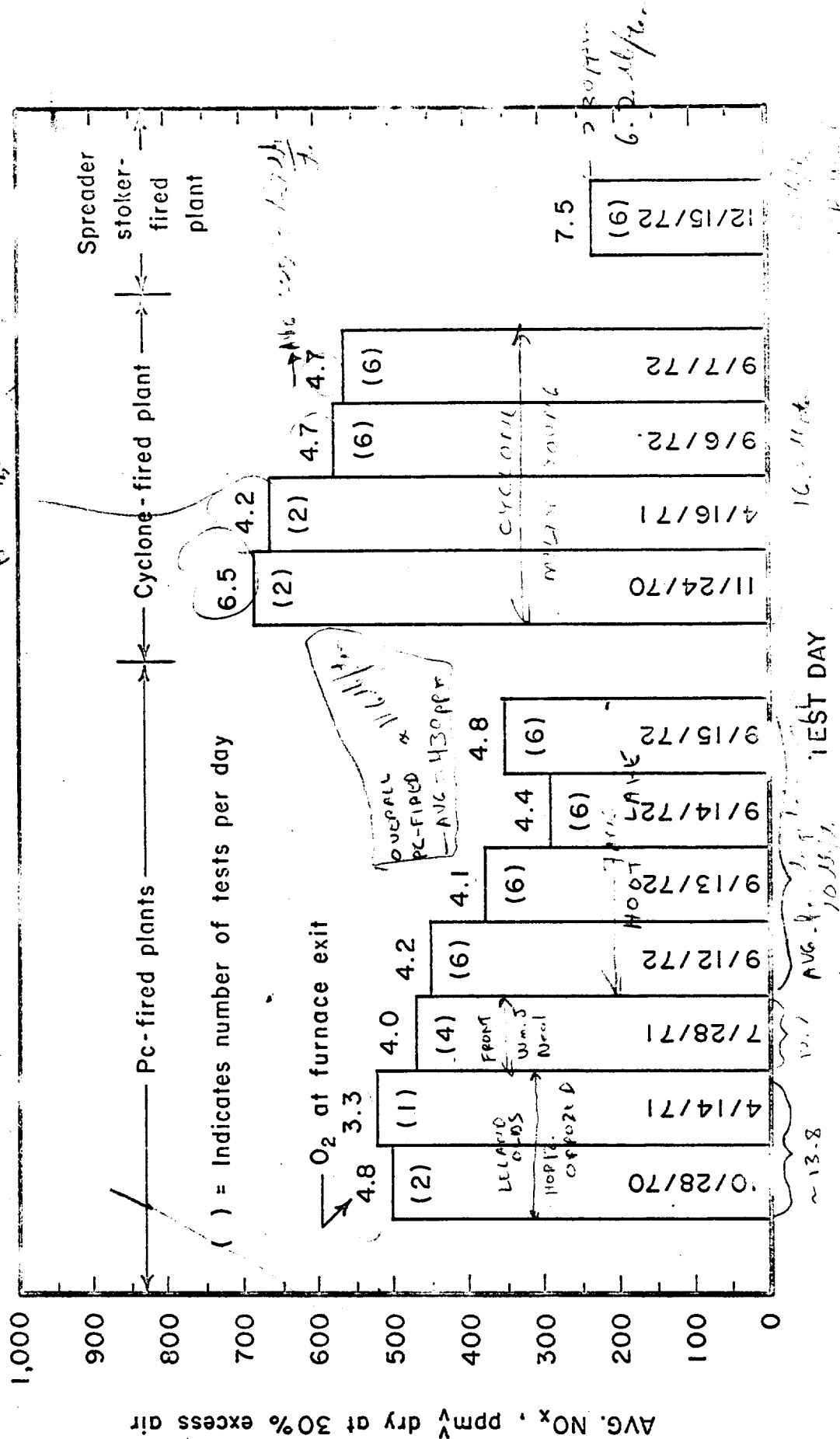


FIGURE 8. - NO_x emissions from various lignite-fired plants. Boiler loads varied from 92 to 106 pct of rated.

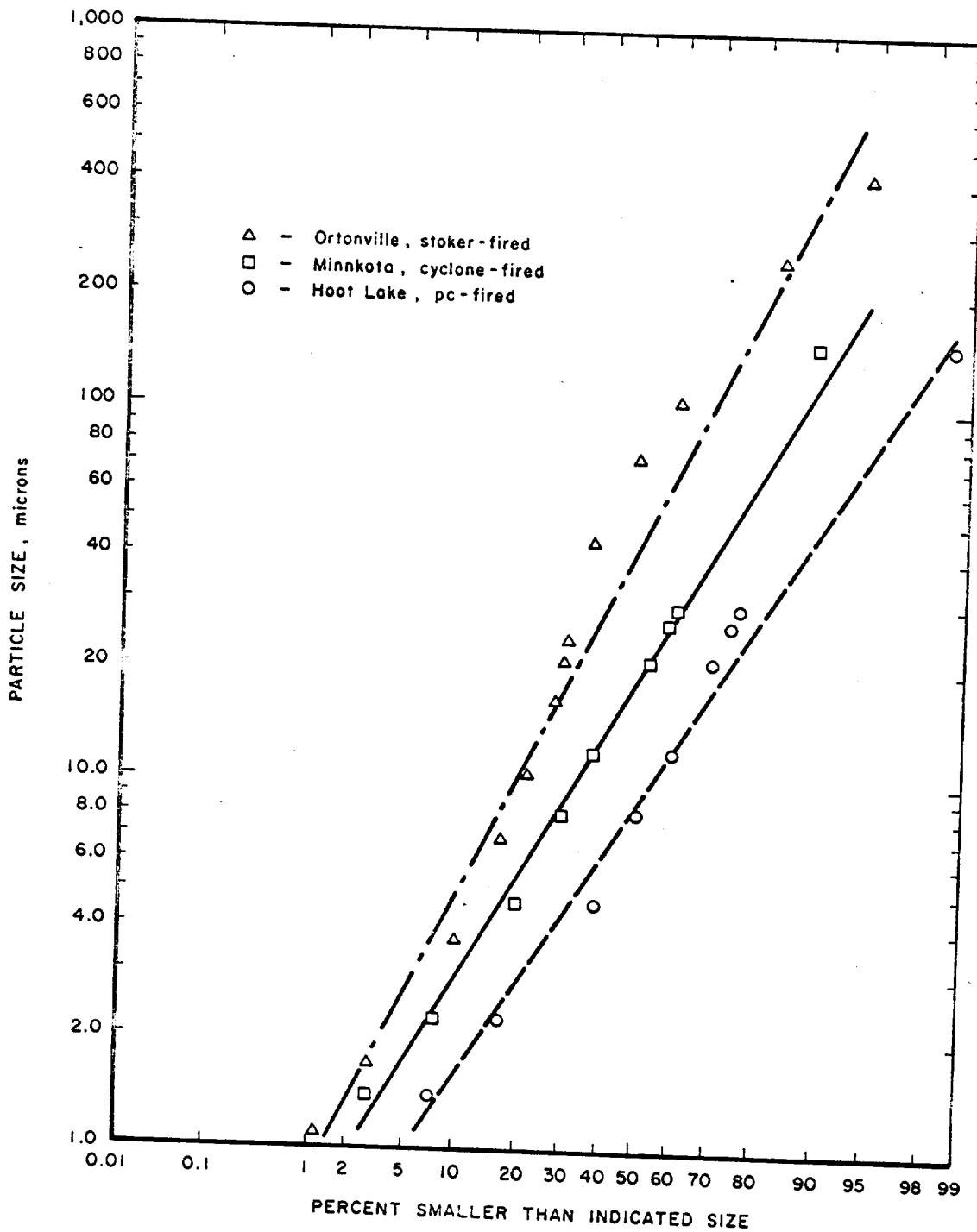


FIGURE 9. - Size analysis (by Bahco method) of fly ash samples aspirated from boiler exit duct. (Spreader-stoker data courtesy of Otter Tail Power Co.)

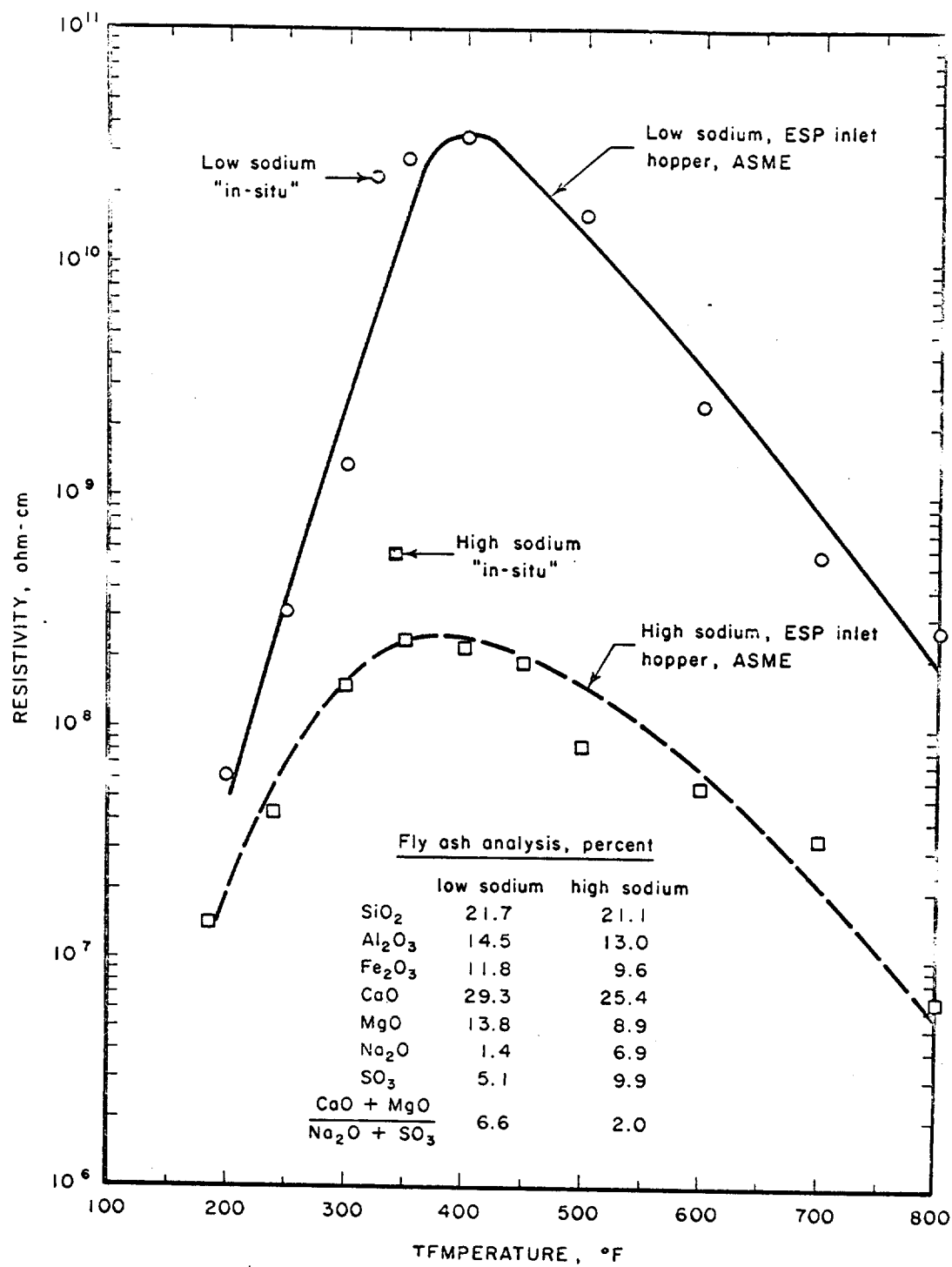


FIGURE 10. - Effect of sodium content on electrical resistivity of lignite fly ash.

Na2 O
Ash % by weight

Emission Factor
lb SOx /ton coal
S = % sulfur wet basis

0.4	33.6	82
0.4	33.7	86
0.7	29.2	73
0.7	39.9	100
0.8	36.6	91
0.8	35.6	89
0.9	36.5	91
0.9	29.5	74
0.9	34.2	86
0.9	32.8	82
1	30.2	75
1.1	29.3	74
1.6	36.5	92
1.7	33.4	84
2	34.2	86
2.1	37.4	94
3	40	100
3.1	33.2	83
3.5	35.5	89
3.8	28.4	71
3.8	27.2	69
4.8	29.1	73
5.1	25.6	64
5.1	27.9	70
5.3	23.1	58
5.4	25.9	65
5.5	30.2	76
5.6	25.4	63
5.8	27.1	68
5.8	39.7	99
6	35.3	89
6.1	24.8	61
6.1	26.3	66
6.1	22.4	59
6.2	31.7	79
7	31.9	80
7.5	26.4	66
7.7	35.3	89
7.8	22.2	59
7.8	32.1	81
8	33.5	84
8.2	18.4	48
8.6	16.9	43
8.8	31.2	78
9	21	52
10.9	10.9	27

29.9

22.65

2.6

Regression Output:

Constant	35.80859
Std Err of Y Est	5.004715
R Squared	0.375080
No. of Observations	46
Degrees of Freedom	44
X Coefficient(s)	-1.30198

where S is the percent sulfur in the coal, 40 S is seen as 100% conversion of fuel sulfur into SO_x . This conversion did not vary significantly between different boiler types or boiler loads. Rather, a correlation was clear (See Figure 1) between sulfur conversion and Na_2O content in the ash. Consequently, the results for SO_x emissions (regardless of boiler) were calculated to be the following:

$Na_2O < 4\%$ of ash	89.6% conversion of S into SO_x (36 S)
$4\% \leq Na_2O < 8.5\%$	68.6% (27 S)
$Na_2O > 8.5$	38.7% (15 S)

Note that while a high Na_2O content coal will have lower SO_x emissions, it will also have greater ash fouling tendencies.

1.3 NO_x Emissions

Nitrogen Oxides are formed by two basic processes: thermal fixation of atmospheric N_2 and combustion of fuel-bound nitrogen. In thermal fixation, the N_2 from the combustion air is converted to the atomic species (heat dependent), which in turn reacts with available oxygen (excess air dependent) to form NO_x . The combustion process involves the reaction of fuel nitrogen with oxygen in the air (excess air dependent). It is believed that the thermal fixation process is predominant at high temperatures, while the combustion process can be significant at lower temperatures. In this respect, the large NO_x emissions associated with the cyclone boiler ($17 \frac{lb}{ton}$) can be traced to the high heat of combustion. For PC fired boilers the emissions were greater for Front and Opposed Wall fired ($14 \frac{lb}{ton}$) than for tangentially fired boilers ($8 \frac{lb}{ton}$). This is obviously due to differences in the flame configuration; tangentially fired flames do not interact as much, rather they radiate outwardly to the cooling surfaces of the boiler. Due to lack of data for Other Stokers, these were assumed to be similar to spreader stokers ($6 \frac{lb}{ton}$).

DATA SUMMARY FOR ALL PLANTS
PULVERIZED COAL-FIRED (1b/ton)

Plant	Control	Particulates	Ash (%)	SO _x	Na ₂ O (% of Ash)	NO _x	Reference
Big Brown	ESP					7.4	8
Station	Cyclone	5.8A	7.5	38.S		17.2	5
eland Olds	Cyclone					11.2	8
n. J. Neal	Cyclone		7.5	32.5S	7.1 ^{OK}	14.3	2
oot Lake	Cyclone,		9.1	32.8S	0.86	13.2	2
	ESP			26.0S	6.3	10.3	2
		5 A	6.5	33.7S	2.2		2
		8 A	6.5	25 S	8.2		6
				46.8S	1.6		6
verage \bar{x}		6.3A		33.5		12.3	

CYCLONE BOILER (1b/ton fuel)

Plant	Control	Particulates	Ash (%)	SO _x	Na ₂ O (% of Ash)	NO _x	Reference
R. Young I	Cyclone		9.8	34.6S	0.6	17.4	2
						12.2	8
		6.5 A	7	34.0S	1.2	18.6	5
eland Olds II 2		5.59A	9.13	34.3S	1.2	18.8	5
	5.3	5.21A	9.13				12
		5.10A	9.25				12
ilton Young I		6.46A	7.47				12
	6.2	6.27A	7.89				12
		5.89A	9.59				12
ilton Young II		9.7 A	7.52				12
	8.6	9.9 A	6.31				12
		6.07A	7.48				12
verage	6.7	6.66A		34.3S		16.8	

Spreader
Controlled

ESP 17

SPREADER STOKERS (1b/ton)

Plant	Control	Particulates	Ash (%)	SO _x	Na ₂ O (% of Ash)	NO _x	Reference
Heskett #1 ✓ B	Cyclone *	6.2 A ^{Con}					5
Heskett #2 ✓ B	Cyclone *	7.8 A ^{Con}					5
F.P. Wood	Cyclone *	3.8 A	7	12.4 S ^{CB}	9.8	0	5
			7.5	14 S ^D	9.8	6.4 ^D	2
Jamestown ✓ D	Mech. Coll.	10.9 A ^{Vn}	5.4	20 S ^D	11.6	5.6 ^D	5
		9.7 A ^{Con}	6.1				5
Devil's L. #2 ✓	Cent. Coll.	4.7 A ^{Con}	8.77				5
North Dakota ✓		4.6 A ¹					5
Average \bar{x}		6.8 A		15.5 S		6	

¹ Based on one test assuming an ash content of 8%

Plant not identified

.57

.09

OTHER STOKERS (1b/ton)

Plant	Control	Particulates	Ash (%)	SO _x	Na ₂ O (% of Ash)	NO _x	Reference
Devil's L. #1 ✓	Cent. Coll.	2.9 A ^{Con}	6.59				5
Beulah #1 ✓	No Control *	2.5 A	8.6				5
Beulah #3 ✓	No Control *	3.6 A	8.6				5
North Dakota	<u>Underfeed</u>	2.5 A ²					5
Average \bar{x}		2.9 A					

² Average of two test assuming an ash content of 8% in lieu of unavailable test reports.
(Reports have been requested August, 1981)

σ

ω

Plant not identified

1.1 Particulate Emissions

Particulate emissions were directly related to the ash content of the coal, and were thus given in terms of percent ash content. From this calculation:

$$\frac{A \text{ lb ash}}{100 \text{ lb coal}} \times \frac{2000 \text{ lb}}{\text{ton}} = \frac{20 A \text{ lb ash}}{\text{ton coal}}$$

Where A is the percent ash in the coal, 20 A is seen as 100 percent emission of the ash as particulates. Due to lack of data, flyash reinjection was not given as a distinct category. This undoubtedly introduced some error, because of the lack of uniform firing practices. Of the particular boiler types, the greatest particulate emissions were from PC and Spreader Stokers ($7 A \frac{\text{lb}}{\text{ton}}$). This is believed to be due to the fact that a lot of coal is burned in suspension, and thus, there is a considerable amount of flyash of air contact. Cyclones exhibited slightly lower particulate emissions, based on 10 tests. Other stokers had the least emissions, (3 A), probably because of their smaller boiler size and because the ash is stationary with respect to the bed. Most particulates were controlled by cyclone collectors. Although most of these had 80 to 90 percent design efficiencies, a typical value of 63 percent was assumed when actual tested efficiencies were not given. This latter efficiency represents a measured efficiency of several existing cyclones and is felt to be a better estimate of the control potential of this kind of collector.

1.2 SO_x EMISSIONS

Sulfur Oxide emissions result from the oxidation of sulfur in the coal. From this calculation:

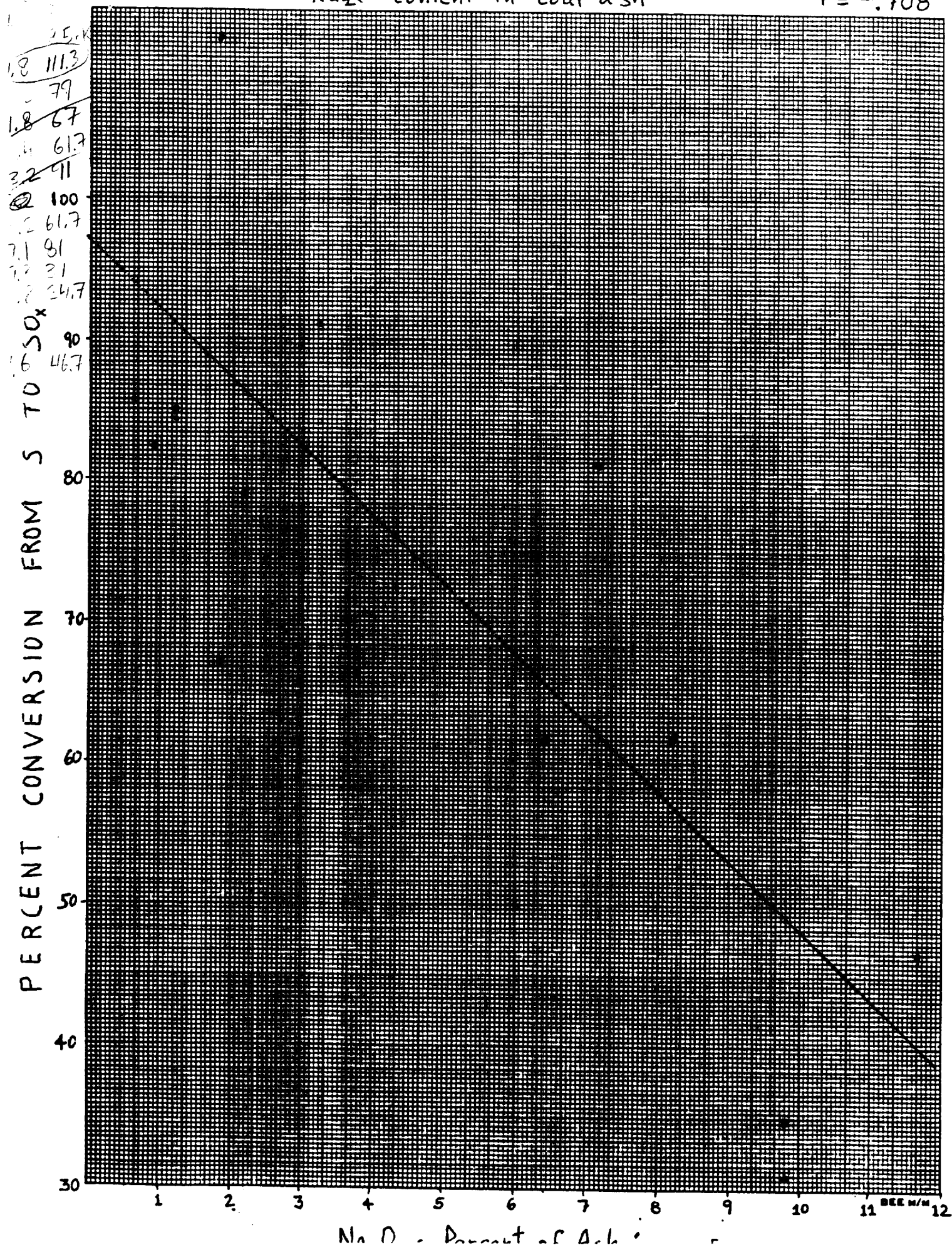
$$\frac{S \text{ lbs sulfur}}{100 \text{ lbs coal}} \times \frac{2 \text{ lbs SO}_2}{1 \text{ lb Sulfur}} \times \frac{2000 \text{ lb}}{\text{ton}} = \frac{40 S \text{ lbs SO}_2}{\text{ton coal}}$$

$$S \left[\frac{\% \text{ Sulfur}}{100 \text{ coal}} \right] \times \left[\frac{2}{1} \right] \rightarrow \left[S_x \right] \frac{155 \text{ SO}_2}{\text{ton}}$$

FIGURE 1: SO_2 emissions as a function of Na_2O content in coal ash

$$y = 96.76 - 4.89x$$

$$r = -0.708$$



% sodium oxide E.F. lb SOx/ton = Value x S
in ash S = % sulfur wet basis

% sulfur
to SOx.

0.6	34.6	86.5
0.86	32.8	82
1.2	34.3	85.75
1.2	34	85
1.6	46.8	117
2.2	33.7	84.25
6.3	26	65
7.1	32.5	81.25
8.2	25	62.5
9.8	12.4	31
9.8	14	35
11.6	20	50

Regression Output:

Constant	96.77185
Std Err of Y Est	14.24715
R Squared	0.692548
No. of Observations	12
Degrees of Freedom	10

X Coefficient(s)	-4.89600
Std Err of Coef.	1.031585