Spatial pattern in the influence of sulfur dioxide emissions from Arizona and New Mexico copper smelters
by Milo Douglas Adkison

A thesis submitted in partial fulfillment of the requirements for the degree of Master of Science in Biological Sciences
Montana State University

Abstract:
A linear relationship between SO2 emissions from Arizona and New Mexico copper smelters and SO4 deposition in Colorado, ascribed by previous authors to long range transport in the atmosphere, is reexamined. Correlations between SO2 emissions rates and SO4 concentrations in deposition at monitoring stations across the continental U.S. are calculated. The effect on these correlations of correcting for the effects of potential confounding factors (amount of rainfall, seasonal pattern in deposition, and local emissions) is examined. The effect of removing the strongest signal in emissions is examined. The existence of spatial patterns in observed correlations predicted by atmospheric transport of materials from the smelters is examined.

The spatial patterns found are consistent with the hypothesis of long range transport of materials in some respects but not in others. The effect of removing site-specific seasonal pattern in deposition is to increase the mean correlation observed at most stations. The effect of removing the largest fluctuation in emissions is the same, and it is shown that this is because this fluctuation is asynchronous to the nationwide seasonal pattern in deposition.

Removing the largest fluctuation in emissions also increases observed correlations in Colorado, contrary to expectation if long range transport is the cause of the observed correlation. Also contrary to the transport hypothesis are the large number of monitoring locations remote from Arizona and New Mexico smelters which show correlations of larger magnitude, both positive and negative.

It is concluded that the evidence for long range transport from this analysis is ambiguous. Theoretical considerations reveal inherent weaknesses in any analysis of this sort. The conclusions of previous authors using this methodology are not supported.

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Milo Douglas Adkison

This thesis has been read by each member of the thesis committee and has been found to be satisfactory regarding content, English usage, format, citations, bibliographic style, and consistency, and is ready for submission to the College of Graduate Studies.

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To my parents

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ABSTRACT

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It is concluded that the evidence for long range transport from this analysis is ambiguous. Theoretical considerations reveal inherent weaknesses in any analysis of this sort. The conclusions of previous authors using this methodology are not supported.

INTRODUCTION

The extent to which distant sources contribute to the deposition of acidic materials has great implications for air quality regulation (1). Accordingly, an effort has been made to determine the range and magnitude of long range transport of such materials (2,3,4,5,6,7). Non-ferrous metal smelters in the southwestern U.S. are of particular interest, both because they produce more than half of the SO_2 emissions in the region (6), and because these emissions fluctuate wildly (Figure 1). These smelters thus provide an opportunity for detecting the extent of long distance transport.

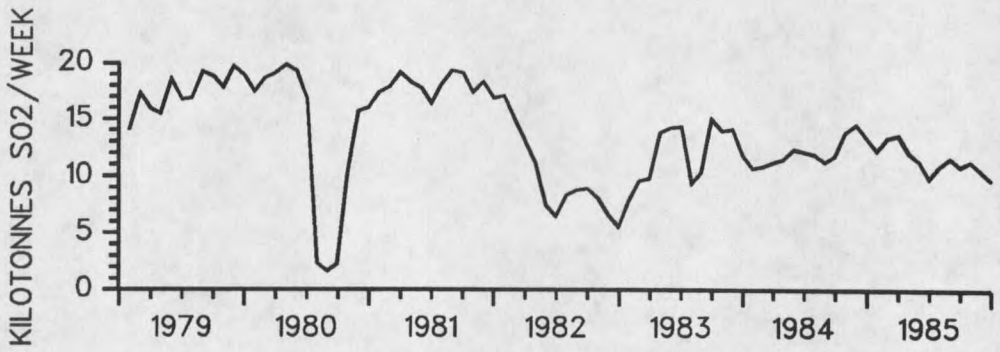

Figure 1. Time series of summed Arizona and New Mexico smelter emissions of SO_2

An apparently significant correlation between southwest smelter emissions and volume-weighted averages of sulfate concentrations in wet deposition in Colorado has been reported (6,7). That correlation was enhanced by removal of the seasonal pattern in deposition, which was attributed to seasonal changes in atmospheric rates of conversion of SO_2 to SO_4. However, correlation between two temporal series cannot of itself prove a causal relationship. A stronger case could be made by showing that other aspects of the data are consistent with a proposed mechanism.

The purpose of this analysis was to test the consistency of the long range transport hypothesis with currently available data. The hypothesis is that appreciable amounts of sulfur dioxide (SO_2) emitted by copper smelters in Arizona and New Mexico are being transported by the atmosphere to distant locations, at least as far away as Colorado. If the hypothesis is true, certain spatial patterns should exist. First, the correlation between smelter SO_2 emissions rates and the concurrent concentrations of SO_4 in wet deposition should tend to decrease with distance from the smelters. This is partly because the odds of the polluted air mass passing over a fixed point decreases the further the point is from the source of the pollution and partly because diffusion

and other meteorological processes will have more opportunity to modify the concentration of the pollutant.

Second, at any given distance, the correlation of deposition with emissions should be greater in the direction of the prevailing winds than in any other direction, because this direction corresponds to the presumed transport mechanism. As winds 600 meters above the surface in southwestern Arizona (the center of the smelting region) are principally from the south and west (8) (persistent surface winds also show these directional tendencies, although easterly winds are important at some locations), the influence of smelters should be strongest northeast of this part of Arizona.

Third, if the correlation between emissions and deposition is due to transport of materials the observed correlation should increase when deposition data are corrected for the effects of phenomena unrelated to distant emissions, such as the amount of precipitation, meteorologically-driven seasonality in deposition, or the influence of local SO_2 emissions.

Finally, if the correlation between emissions and deposition is due to the transport of materials, the observed correlation will be weaker during episodes when the variation in the emissions signal is reduced, since the reduction of emissions variation will increase the

relative importance of other factors unrelated to the proposed mechanism.

A related hypothesis consistent with the observed correlation between Arizona and New Mexico smelters and deposition in Colorado is that the effect of long range transport is enhanced by certain sorts of orographic features. High elevation sites might receive more materials or these materials might be more readily detectable due to the increase in precipitation amount and frequency at such sites.

These predictions were tested using correlation analysis, calculating correlations between the time trajectories of SO_2 emissions and SO_4 concentrations in wet deposition at monitoring stations at various distances from the presumed source. Examined were (1) the relationship between the observed correlation and the distance from the monitoring site to the smelter, (2) the difference in the observed correlations for monitoring sites northeast of smelters versus those in other directions, and (3) the relationship between these correlations and other factors such as the elevation of the monitoring sites. Also examined were the effects on these patterns of (a) removing the relationship between the amount of precipitation and the concentration of sulfate, (b) removing seasonal patterns in sulfate

concentration (9,10,11,12), (c) removing correlations between sulfate concentration and local sources of SO_2 emissions (4), (d) simultaneously removing the effects of precipitation amount, seasonal pattern, and local emissions, and (e) throwing away all data for 1980, the year which contains the largest fluctuation in smelter emissions.

Any analysis relying solely on correlations between emissions and wet deposition is vulnerable to several kinds of interference. The first is that the trajectories of air masses vary in time. The second is that air masses which have followed the same trajectory will have experienced different meteorological histories, which will have differentially affected the concentration of the materials they contain. Thus, the amount of sulfur aerosols reaching a fixed monitoring location is a function not only of the fluctuation in emissions, but also of the fluctuation in meteorology (4,8,10,13,14). The larger such fluctuations, the lower the correlation between emissions and deposition.

Another possible interference arises from using materials deposited in precipitation as the measure of the amount of material transported. Such measurements might contain biases, as only air masses conducive to precipitation are sampled. Certainly there will exist

large periods of time for which no measurements are available. Sampling ambient air would be a much more direct measurement. However, no network of aerometric samplers exists with an equivalent monitoring site density and location remoteness which match those of the National Atmospheric Deposition Program wet deposition monitoring network.

MATERIALS AND METHODS

Data Used in the Analysis

Weekly wet deposition data were obtained from the Acid Deposition System database for all National Atmospheric Deposition Program (NADP) monitoring sites (10).

Emissions estimates for calendar months, by state, were obtained from the Argonne National Laboratory, covering the period January 1979 to December 1985 (15,16). Because of the crude method used to calculate smelter emissions, smelter emissions data for the states of Arizona, New Mexico, Utah, and Nevada were replaced by estimates based on more detailed data, used previously by Epstein and Oppenheimer in their analyses (7). These latter estimates were compared with estimates from the relevant state air quality bureaus before use. As both sets of estimates of summed Arizona and New Mexico emissions rates show a strong linear relationship (Figure 2), the choice of which to use in a correlation analysis is relatively innocuous. This study chose to use the same

estimates as Epstein and Oppenheimer so as to facilitate comparison.

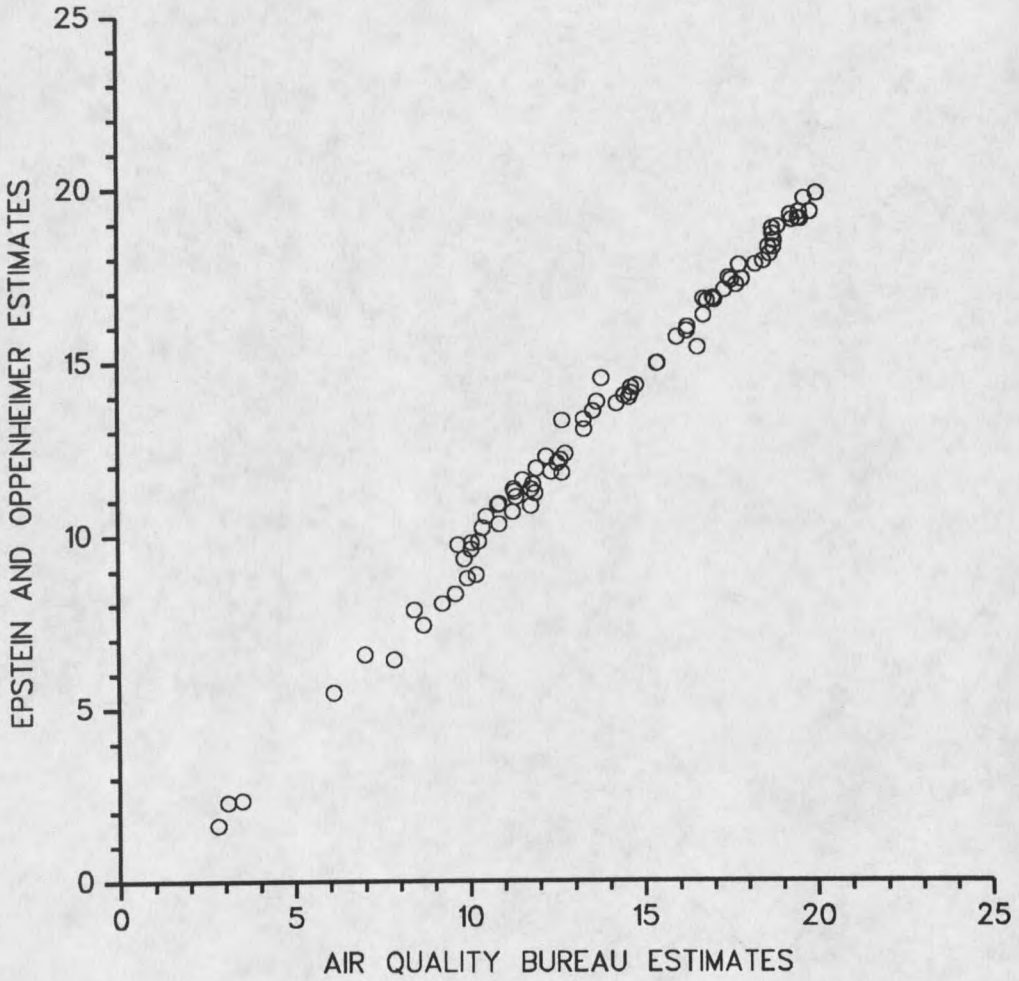

Figure 2. Comparison of estimates of summed Arizona and New Mexico smelter emissions of SO_2 (kilotons SO_2/week)

Data Manipulation

Any deposition record in which the value of gauge or of any of the nine principal measured ions (H^+, SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}) was not recorded was discarded. Due to the presence of occasional extreme outliers in the remainder of the data, observations unusual in any respect other than their sulfate concentration were discarded using the following methodology: each observation was assigned a value equal to its Mahalanobis distance from the centroid of all other observations from the same site. The covariance matrix used in the Mahalanobis distance calculation was the matrix of covariances of gauge and all nine principal ions except sulfate, calculated from all observations except the one for which the distance was being calculated. The ten percent of the observations from each site possessing the largest values were discarded (Figure 3).

Deposition and emissions data were adjusted so that time periods were equivalent. Weekly sulfate concentrations were lumped by volume-weighted averaging of records into four or five week periods chosen to correspond as closely as possible to calendar months. Emissions estimates for these four and five week periods were calculated as a weighted sum of calendar month

emissions estimates, where the weights were the number of days of the calendar month falling into the period. These totals were then divided by the number of weeks in the time period so as to express emissions as a weekly rate (Table 1).

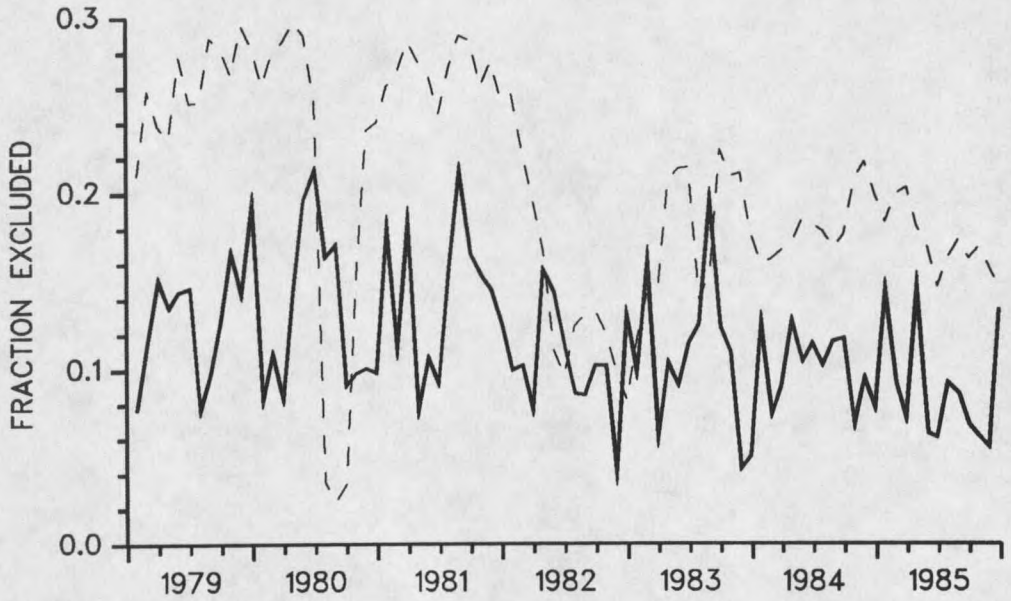

Figure 3. Temporal sequence of the fraction of deposition records excluded. Dashed line is emissions rate for AZ and NM smelters.

Table 1. Temporal series of summed Arizona and New Mexico smelter emissions rates

Period beginning	Period ending	Tons SO_2/day	Period beginning	Period ending	Tons SO_2/day
790103	790130	14029	820630	820803	8327
790131	790227	17231	820804	820831	8783
790228	790403	15903	820901	820928	8880
790404	790501	15427	820929	821102	8048
790502	790529	18505	821103	821130	6579
790530	790703	16772	821201	821228	5484
790704	790731	16825	821229	830201	7865
790801	790828	19245	830202	830301	9611
790829	791002	18848	830302	830329	9793
791003	791030	17799	830330	830503	13797
791031	791127	19717	830504	830531	14268
791128	800101	18908	830601	830628	14332
800102	800129	17435	830629	830802	9335
800130	800226	18692	830803	830830	10334
800227	800401	19102	830831	830927	14971
800402	800429	19868	830928	831101	13981
800430	800603	19327	831102	831129	14108
800604	800701	16802	831130	840103	11831
800702	800729	2367	840104	840131	10679
800730	800902	1634	840201	840228	10852
800903	800930	2308	840229	840403	11217
801001	801028	10225	840404	840501	11475
801029	801202	15720	840502	840529	12349
801203	801230	16015	840530	840703	12173
801231	810203	17399	840704	840731	11923
810204	810303	17914	840801	840828	11260
810304	810331	19127	840829	841002	11799
810401	810428	18317	841003	841030	13855
810429	810602	17822	841031	841127	14533
810603	810630	16357	841128	850101	13312
810701	810728	18116	850102	850129	12265
810729	810901	19339	850130	850226	13345
810902	810929	19142	850227	850402	13584
810930	811103	17368	850403	850430	12077
811104	811201	18318	850501	850528	11333
811202	811229	16854	850529	850702	9839
811230	820202	17088	850703	850730	10919
820203	820302	14960	850731	850903	11596
820303	820330	13055	850904	851001	10885
820331	820427	11111	851002	851029	11339
820428	820601	7424	851030	851203	10553
820602	820629	6433	851204	851231	9738

As a strong relationship existed between gauge and both the mean and the variance of sulfate concentration, some analyses were performed in which the sulfate concentration of each deposition record, while still in weekly form, was adjusted to a weighted departure from that expected for that gauge by subtracting the expected sulfate concentration for the recorded gauge, and then dividing by the standard deviation in concentration for that gauge. These expected values were derived by fitting fourth order polynomials to empirical relationships with gauge calculated using all NADP weekly records (Figures 4 and 5).

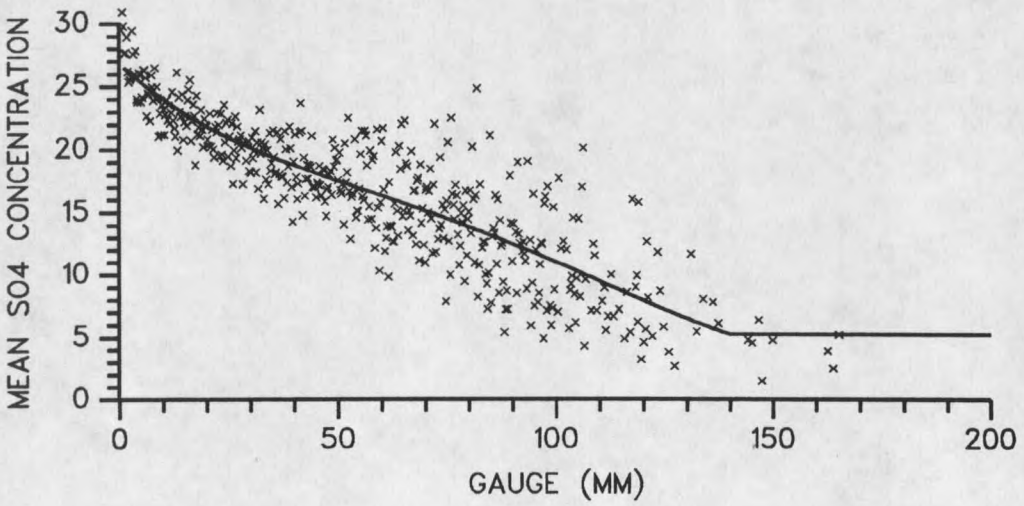

Figure 4. Polynomial fit (truncated) of the relationship between SO_4 concentration and gauge.

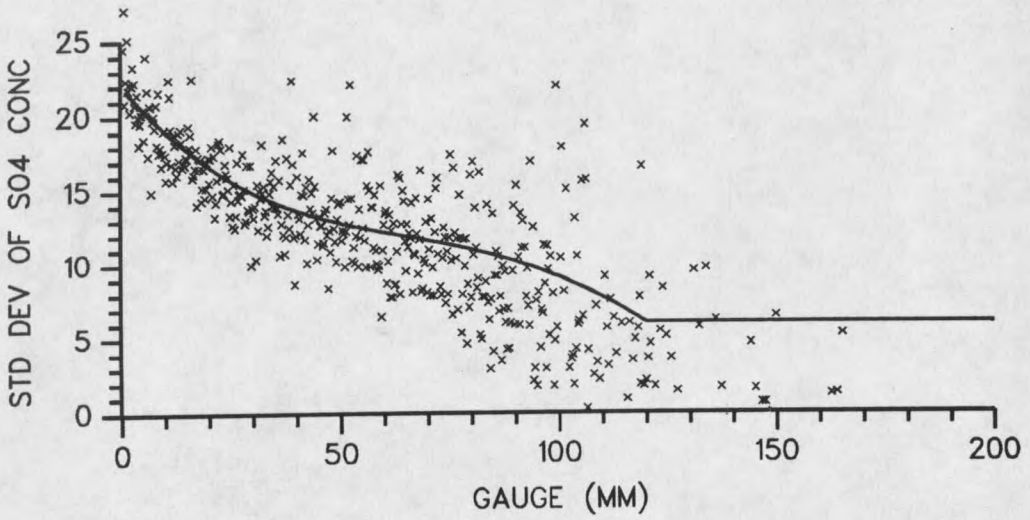

Figure 5. Polynomial fit (truncated) of the relationship between the standard deviation in SO_4 concentration and gauge.

In order to eliminate spurious correlation of deposition with smelter emissions which could arise from local emissions with a temporal pattern similar to that of the smelters, some analyses were performed in which sulfate concentrations were replaced by the residuals of a linear regression of SO_4 concentration on the emissions of all sulfur dioxide sources in the state in which the deposition monitoring site was located (Arizona and New Mexico smelter emissions were not included in their respective state totals).

In order to eliminate a potential masking of the correlation of deposition with smelter emissions which could arise from seasonal meteorological effects driving sulfate concentration, some analyses were performed in which seasonal pattern was subtracted from deposition. The quantity subtracted was the average concentration of all periods in the temporal series separated by integer multiples of lag twelve from the observation. As this was only meaningful where at least two such observations existed, data for which a seasonal average could not be calculated were discarded in this particular calculation.

Additionally, some analyses were performed in which the effect on deposition of gauge, seasonal pattern, and local emissions were simultaneously removed as described above.

Finally, the effect on the observed correlations of throwing away all data from 1980, the year containing the largest fluctuation in smelter SO_2 emissions rates, was examined.

Figure 6. Location of smelters and monitoring stations. Diamonds signify smelters. 'NE' signifies NADP monitoring stations northeast of smelters. 'O' signifies NADP monitoring stations at other orientations.

Calculations and Statistics Used

Correlations were calculated between summed Arizona and New Mexico copper smelter emission rates and volume-weighted average sulfate concentrations in wet deposition, for each site possessing data for at least twelve of the 84 nominal months in the series. The great circle distance from the source smelters (Tucson, AZ was taken as the center of the Arizona and New Mexico smelter region) to each of the deposition monitoring stations was also calculated.

The hypothesis of decreasing correlation with increasing distance was tested using a nonparametric rank concordance test (17). The hypothesis of increased correlations downwind of smelters was tested by first regressing correlations on distance, and then performing a two-sample t-test (17) on the residuals. One of the two samples was sites within 22.5 degrees of due northeast of the smelters; the other was all other sites (Figure 6).

A nonparametric rank concordance test was also used to explore the possibility that stations at higher elevations should have deposition which is more strongly influenced by smelter emissions, since these are more exposed to long range transport. The concordance between elevation and the residuals from a linear regression of correlation on

distance from the smelters was calculated, using either the residuals for all sites or only the residuals for sites northeast of the smelters.

In addition, the change in the observed correlation for each site as a result of the previously described data manipulations was plotted. These manipulations were a) correction of sulfate concentration for gauge, b) correction of sulfate concentration for seasonal pattern, c) correction of sulfate concentration for local emissions, d) correction of sulfate for gauge, seasonal pattern, and local emissions, and e) throwing away all 1980 records.

RESULTS

Test of a negative concordance between the distance separating a monitoring station from the smelters and the correlation between the temporal series of emissions and deposition: If all monitoring stations are considered, the result is generally significant at the .05 level (Table 2), even if p values are corrected for testing multiple hypotheses (We are testing five distinct hypotheses). The one exception arises when using deposition data treated to simultaneously remove the influence of gauge, seasonal pattern, and local emissions. Even the nonsignificant concordance is of the expected sign. The concordances range from -.2164 to -.0579.

When only stations northeast of the smelters are considered, concordances again have the expected sign, although none are significant at the .05 level (Table 3). Concordances range from -.1494 to -.0061.

A roughly linear decline in the average correlation with distance is observed (Figures 12-16). Correlations

for stations within 2500 kilometers of the smelters are mostly positive, with frequent exceptions (e.g. CUBA site, New Mexico - see figures 1-5). Stations further than 2750 kilometers distant frequently show strong negative correlations.

Test of higher correlations for monitoring sites northeast of the smelters than for those lying in other directions: The results of this test are generally significant at the .05 level, even if testing multiple hypotheses is taken into account. However, if deposition data treated to simultaneously remove the effects of gauge, seasonal pattern, and local emissions are used, the result is significant if considered alone and not significant if the Scheffe multiple testing criterion is used (Table 4). A visual examination of the results shows that higher correlations to the northeast are evident at all distances (Figures 17-21).

Table 2. Nonparametric rank concordance test of a negative relationship between correlation and distance from smelters. All monitoring stations.

Deposition data manipulation	Kendall's Tau	Z	p value
a	-0.188	-3.597	.001 **
b	-0.216	-4.151	.00001 **
c	-0.113	-2.107	.018 *
d	-0.159	-3.029	.0012 **
e	-0.058	-1.072	.15

Table 3. Nonparametric rank concordance test of a negative relationship between correlation and distance from smelters. Stations northeast of smelters only.

Deposition data manipulation	Kendall's Tau	Z	p value
a	-0.149	-1.448	.065
b	-0.168	-1.624	.055
c	-0.006	-0.059	.480
d	-0.117	-1.134	.130
e	-0.044	-0.430	.340

* = significant at .05 level
** = significant at .01 level, and significant at .05 level given five hypotheses tested
a = unadjusted data
b = data adjusted to remove gauge effect
c = data adjusted to remove seasonal pattern
d = data adjusted to remove effect of local emissions
e = data adjusted to remove effects of gauge, seasonal pattern, and local emissions

Table 4. T test for difference in mean residual of linear regression of correlation on distance.

Dep. data	mean NE	#	Mean other	#	t	p value
a	0.072	45	-0.027	122	3.39	.001 **
b	0.059	45	-0.022	122	2.49	.006 **
c	0.058	45	-0.023	114	2.51	.006 **
d	0.066	45	-0.026	119	3.19	.002 **
e	0.051	45	-0.021	111	1.93	.035 *

* = significant at .05 level
** = significant at .01 level, and significant at .05 level given five hypotheses tested
a = unadjusted data
b = data adjusted to remove gauge effect
c = data adjusted to remove seasonal pattern
d = data adjusted to remove effect of local emissions
e = data adjusted to remove effects of gauge, seasonal pattern, and local emissions

Test of increasing correlations at any given distance for monitoring sites at higher elevations: Although the sign of all results was correct, the tests were nonsignificant with one exception. All tests using only stations northeast of the smelters were insignificant (Table 5). The test using all monitoring stations and deposition data treated to simultaneously remove the effects of gauge, seasonal pattern, and local emissions was significant, even using a multiple testing criterion (Table 6). The concordance observed was 0.135.

Table 5. Nonparametric rank correlation of elevation with residual of linear regression of correlation on distance. Stations northeast of smelters only.

Deposition data type	Kendall's Tau	Z	p value
a	0.032	0.313	.39
b	0.079	0.763	.23
c	0.101	0.978	.17
d	0.075	0.724	.24
e	0.162	1.565	.06

Table 6. Nonparametric rank correlation of elevation with residual of linear regression of correlation on distance. All monitoring stations.

Deposition data type	Kendall's Tau	Z	p value
a	0.036	0.678	.25
b	0.029	0.554	.29
c	0.072	1.330	.09
d	0.059	1.115	.13
e	0.135	2.495	.007 **

* = significant at .05 level
** = significant at .01 level, and significant at .05 level given five hypotheses tested
a = unadjusted data
b = data adjusted to remove gauge effect
c = data adjusted to remove seasonal pattern
d = data adjusted to remove effect of local emissions
e = data adjusted to remove effects of gauge, seasonal pattern, and local emissions

Change in observed correlations as a result of manipulation of the data: The removal of the effects of gauge or of local emissions caused no significant increase or decrease in observed correlations (Table 7, Figures 22,24). The removal of seasonal pattern, seasonal pattern in conjunction with local emissions and gauge effects, or the removal of 1980 data all resulted in highly significant increases in the mean correlation observed (Table 7, Figures 23,25,26).

Table 7. Test for a difference between mean correlation of smelters with monitoring stations before and after data manipulation.

Deposition data type	difference in means	variance of differences	t
a	0.007	0.005	1.3
b	0.082	0.013	9.2**
c	0.004	0.045	0.8
d	0.081	0.028	6.0**
e	0.048	0.010	7.1**

* = significant at .05 level
** = significant at .01 level, and significant at .05 level given five hypotheses tested
a = data adjusted to remove gauge effect
b = data adjusted to remove seasonal pattern
c = data adjusted to remove effect of local emissions
d = data adjusted to remove effects of gauge, seasonal pattern, and local emissions
e = 1980 data excluded,includes only stations in operation in 1980

DISCUSSION

In accordance with the hypothesis of long range transport, the average correlation between smelter emissions of SO_2 and deposition of SO_4 declines with the distance of the monitoring site from the smelters (Figures 12-16). However, the average correlation near the smelters is very small. Further, the average correlation seems to decline linearly to values which are quite negative at large separations. An asymptotic approach to a correlation of zero would be more consistent with the hypothesis.

Also in accordance with the hypothesis, stations downwind (northeast) of the smelters tend to have correlations which are more positive than stations at the same distance which lie in other directions, apparently at all distances (Figures 17-21). Some puzzling features in the pattern are observed. Among the sites downwind of the smelters, the observed correlation declines only weakly with distance. Some of the largest correlations occur in Wisconsin and Michigan, over 2000 kilometers distant,

while deposition at the CUBA, New Mexico site (500 km distant) is negatively correlated with the smelter signal.

A 'Rocky Mountains' effect, where the high altitude of Colorado sites results in a stronger relationship between these sites and the smelters, is at best weakly supported. The residuals of the relationship between correlation and distance are only slightly related to site elevations. One would also expect a screening effect, so that sites beyond the mountain barrier would not be highly correlated with the smelter signal. The large correlations observed in Wisconsin and Michigan are thus unexpected.

Although these analyses are consistent with previous results showing a correlation between smelter emissions in Arizona and New Mexico and deposition in Colorado (6,7) they cast serious doubt on the attribution of this correlation to long range transport of sulfur aerosols from the smelters to Colorado. Sites in Colorado all show positive correlations, many of which are significant using conventional significance tests. However, comparison of Colorado correlations with correlations in other locations is probably a better test of their significance, both because the data are from a temporal sequence with strong serial autocorrelation and because the null hypothesis is not that correlations are due to chance but rather that they are due to indirect factors.

Stations in unlikely locations show positive correlations of greater magnitude (Figures 30-34). Further, many stations distant from the smelters are highly negatively correlated with emissions (Figures 7-16). These stations are too distant to reasonably be receiving a detectable emissions signal from southwest copper smelters. This distribution of correlations therefore provides a standard for the expectation of the size of correlations due to indirect factors. A comparison of correlations from Colorado with the empirical distribution function of U.S. correlations reveals that Colorado stations range from the upper 90th to upper 30th percentiles (Figures 30-34).

More important is the increase in the correlations observed in Colorado resulting from removal of all 1980 records (Figure 26). This is directly contrary to the predictions of a long range transport mechanism. Why should removing the strongest signal in the emissions data increase the correlation between these emissions and deposition?

The answer seems to come from the answer to a different question. Why, in the majority of cases, for monitoring stations scattered all over the U.S., do the correlations between smelter emissions and deposition increase when the seasonal signal in deposition is

removed? There are two possible explanations; either the Arizona and New Mexico smelter emissions are significantly affecting most of the U.S., or there is a common seasonal pattern to most monitoring stations which is negatively correlated with the seasonal pattern in smelter emissions.

Many authors have observed and explained a common seasonal pattern to sulfate concentration in deposition (26,27,28,29,30,32). Sulfate concentrations in wet precipitation tend towards a summer peak, due to the importance of temperature and sunlight in the rate of conversion of SO_2 to SO_4. An examination of the smelter emissions shows a summer low (Figure 27). This explains why the removal of the seasonal pattern in the deposition of a given site will tend to increase the observed correlation between that site's deposition and the emissions of the smelters.

A closer look at the seasonal pattern in smelter emissions reveals that the seasonality is due to the influence of a single year - 1980, the year in which a strike resulted in near zero summer emissions. If 1980 is removed, no seasonal pattern in smelter emissions remains (Figure 28).

For all sites at which fluctuations in smelter emissions strongly influence fluctuations in deposition, the removal of 1980, the year of greatest fluctuation in

emissions, should result in a decrease in the observed correlation. For those sites for which the observed correlations are owing to coincidental synchrony of seasonality, the removal of 1980 - and consequently of a seasonal pattern in emissions opposite to that in deposition - should result in an increase in the observed correlation.

The mean correlation of those stations which were in operation in 1980 increases significantly when 1980 data are removed (Table 6, Figure 27). Those increasing in correlation by at least 0.1 include the only Arizona station and all five of the Colorado stations used by Epstein and Oppenheimer in their analysis of the impacts of the smelters on Colorado.

CONCLUSIONS

The southwestern smelters probably offer the best presently available opportunity for detection of long-range transport from a point source using this methodology. The southwest smelter emissions present an unmatched situation of a relatively strong emissions signal that is out of synchrony with the mass emissions from the rest of the U.S. Nevertheless, the power of any analysis based on the correlation between emissions and wet deposition is diminished by the indirectness of the estimation of sulfur-containing aerosols, the variation in plume trajectories, site-specific micrometeorology, etc.

Although some features of the spatial patterns in the observed correlations between southwest smelter emissions and deposition at NADP monitoring sites are consistent with the hypothesis of long-range transport of sulfur materials, many others are not. For example, correlations between smelter emissions in the southwest and sulfate deposition in Colorado are not stronger than those observed at locations unlikely to be affected by Arizona

and New Mexico smelter emissions. Those Colorado stations in operation in 1980 did not respond to an almost complete cessation of smelter emissions (Figure 29).

Given the inherent weaknesses of this sort of analysis, the lack of detection of a long-range effect of these smelters cannot be taken to mean that one does not exist. However, one definite conclusion can be drawn; if significant long-range transport of sulfur-containing aerosols occurs here, analyses of this sort are not sensitive enough to detect them unambiguously.

Given these considerations, it is recommended that any future analyses of distant impacts of point emissions sources incorporate some or all of the following features: direct measurements of the concentration of sulfur-containing aerosols in the air, detailed meteorological histories for polluted air masses, and incorporation of natural or experimental tracer materials.

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APPENDIX

Figure 7. Correlations of concentration of SO_4 in wet deposition and SO_2 emissions from smelters. Diamonds signify smelters. Unadjusted deposition.

Figure 8. Correlations of concentration of SO_4 in wet deposition and SO_2 emissions from smelters. Diamonds signify smelters. Deposition adjusted for gauge.

Figure 9. Correlations of concentration of SO_4 in wet deposition and SO_2 emissions from smelters. Diamonds signify smelters. Deposition adjusted for seasonal pattern.

Figure 10. Correlations of concentration of SO_4 in wet deposition and SO_2 emissions from smelters. Diamonds signify smelters. Deposition adjusted for local emissions.

Figure 11. Correlations of concentration of SO_4 in wet deposition and SO_2 emissions from smelters. Diamonds signify smelters. Deposition adjusted for gauge, seasonal pattern, and local emissions.

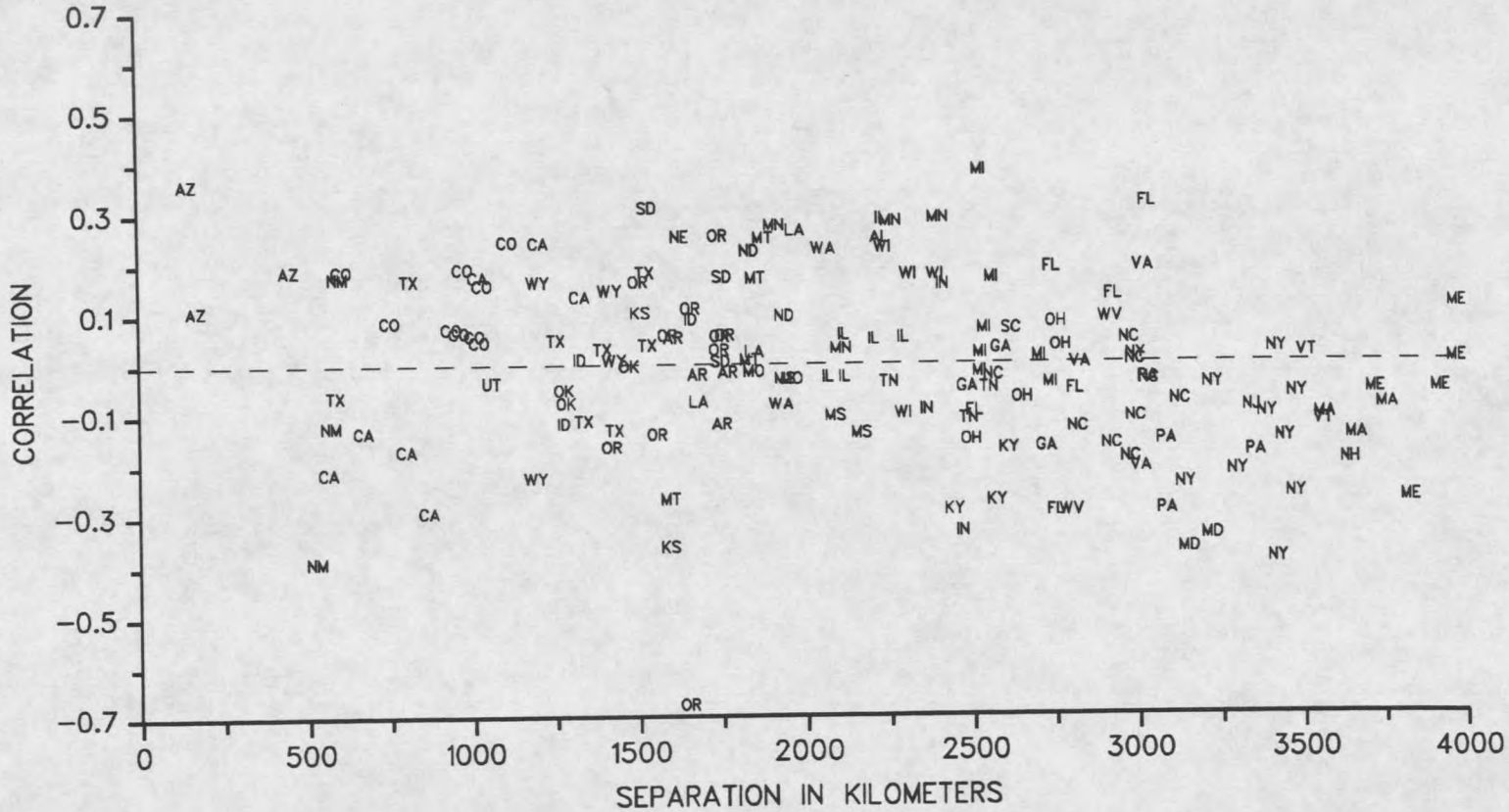

Figure 12. Correlations of concentration of SO_4 in wet deposition and SO_2 emissions from smelters vs distance from smelters to monitor. Stations labelled by state. Dashed line is zero correlation. Unadjusted deposition.

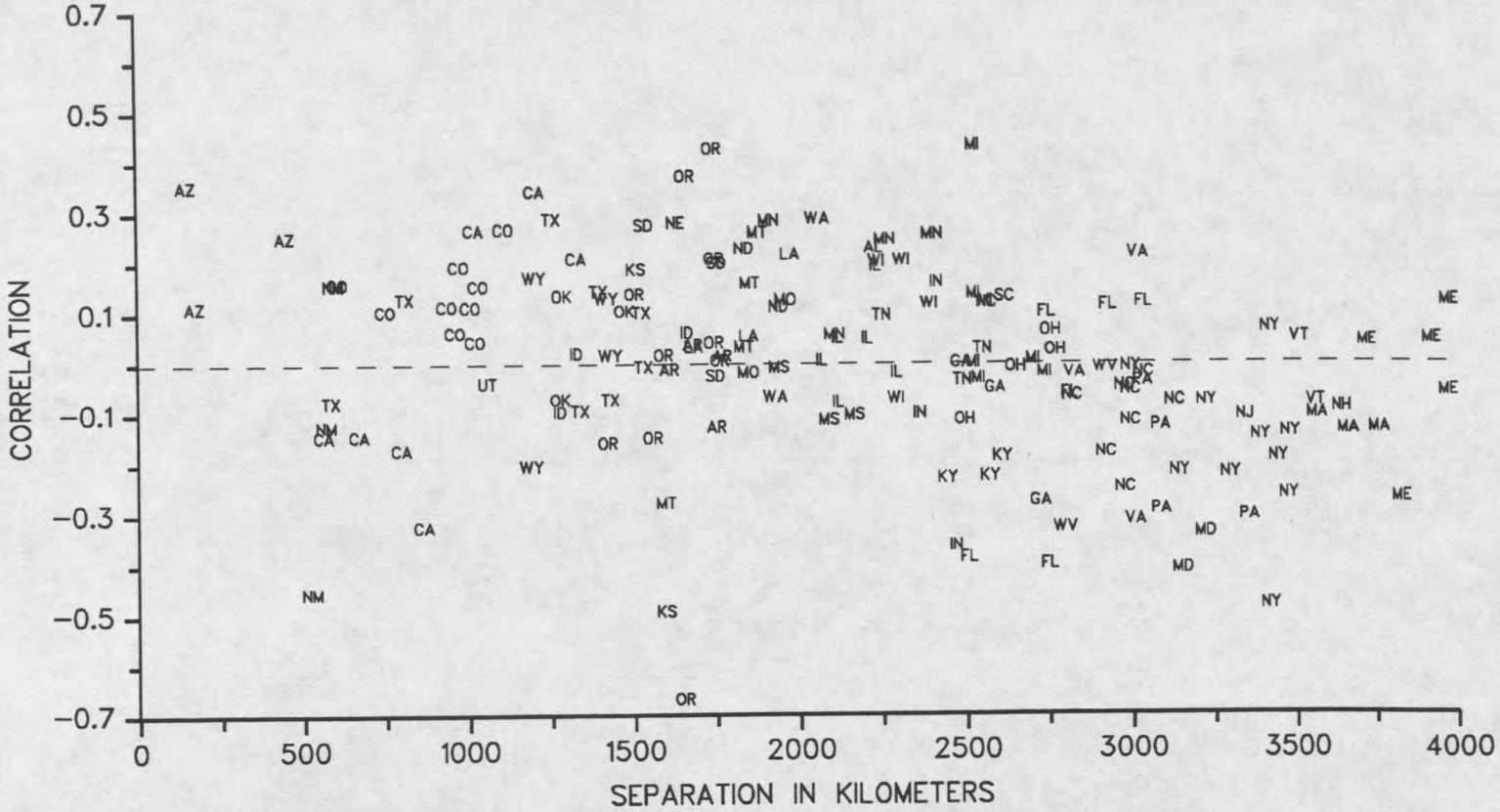

Figure 13. Correlations of concentration of SO_4 in wet deposition and SO_2 emissions from smelters vs distance from smelters to monitor. Stations labelled by state. Dashed line is zero correlation. Deposition adjusted for gauge.

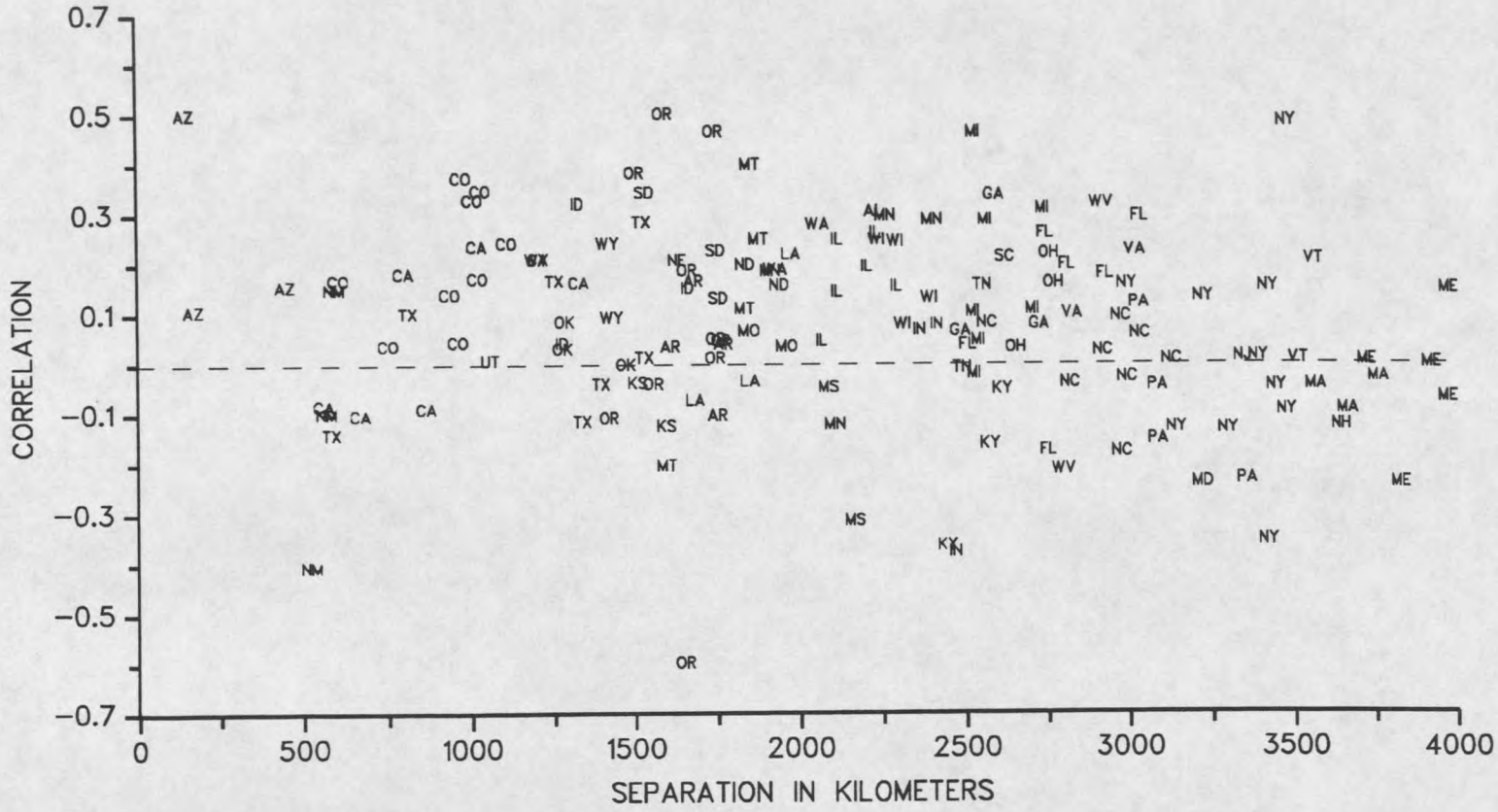

Figure 14. Correlations of concentration of SO_4 in wet deposition and SO_2 emissions from smelters vs distance from smelters to monitor. Stations labelled by state. Dashed line is zero correlation. Deposition adjusted for seasonal pattern.

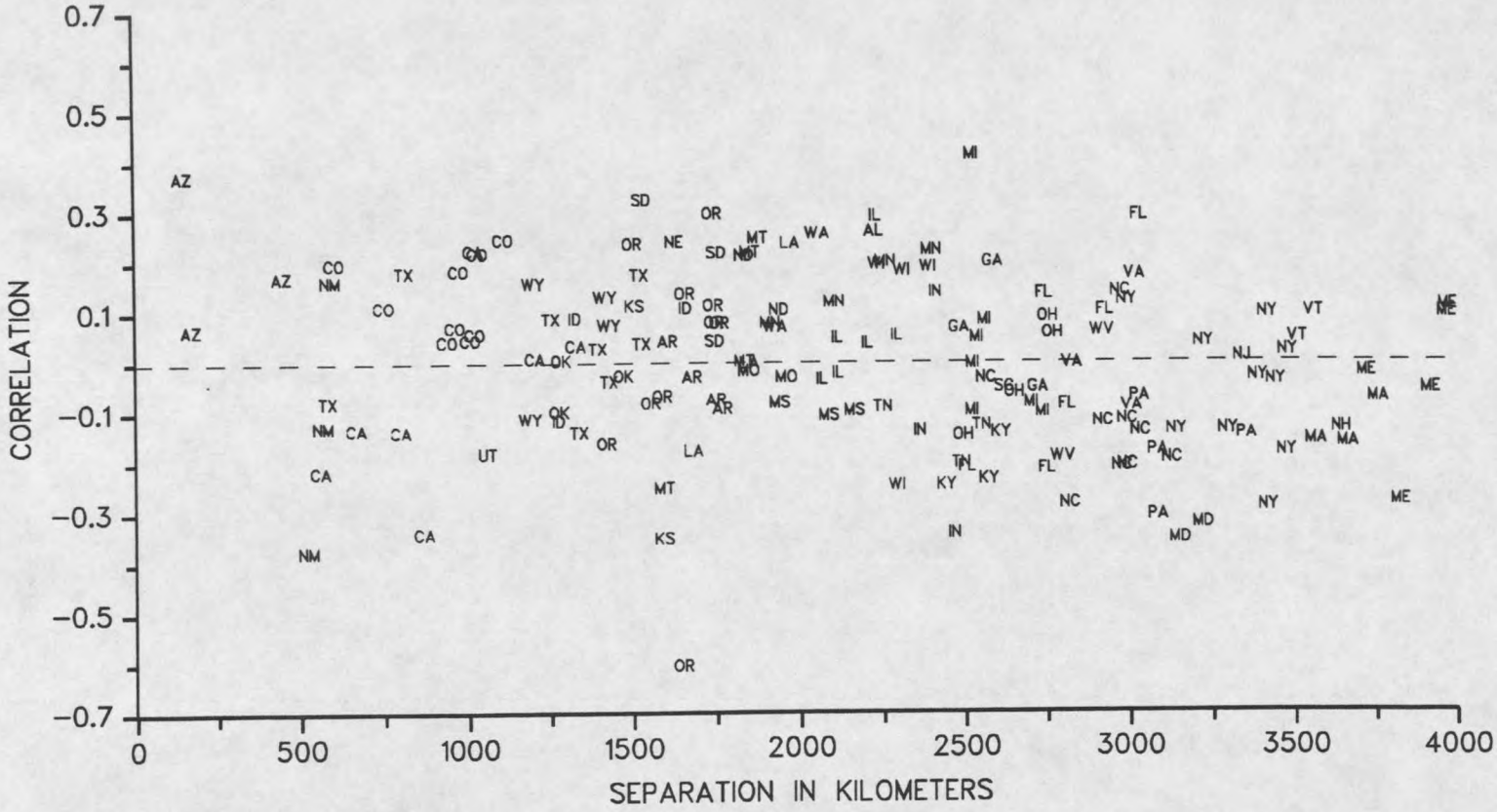

Figure 15. Correlations of concentration of SO_4 in wet deposition and SO_2 emissions from smelters vs distance from smelters to monitor. Stations labelled by state. Dashed line is zero correlation. Deposition adjusted for local emissions.

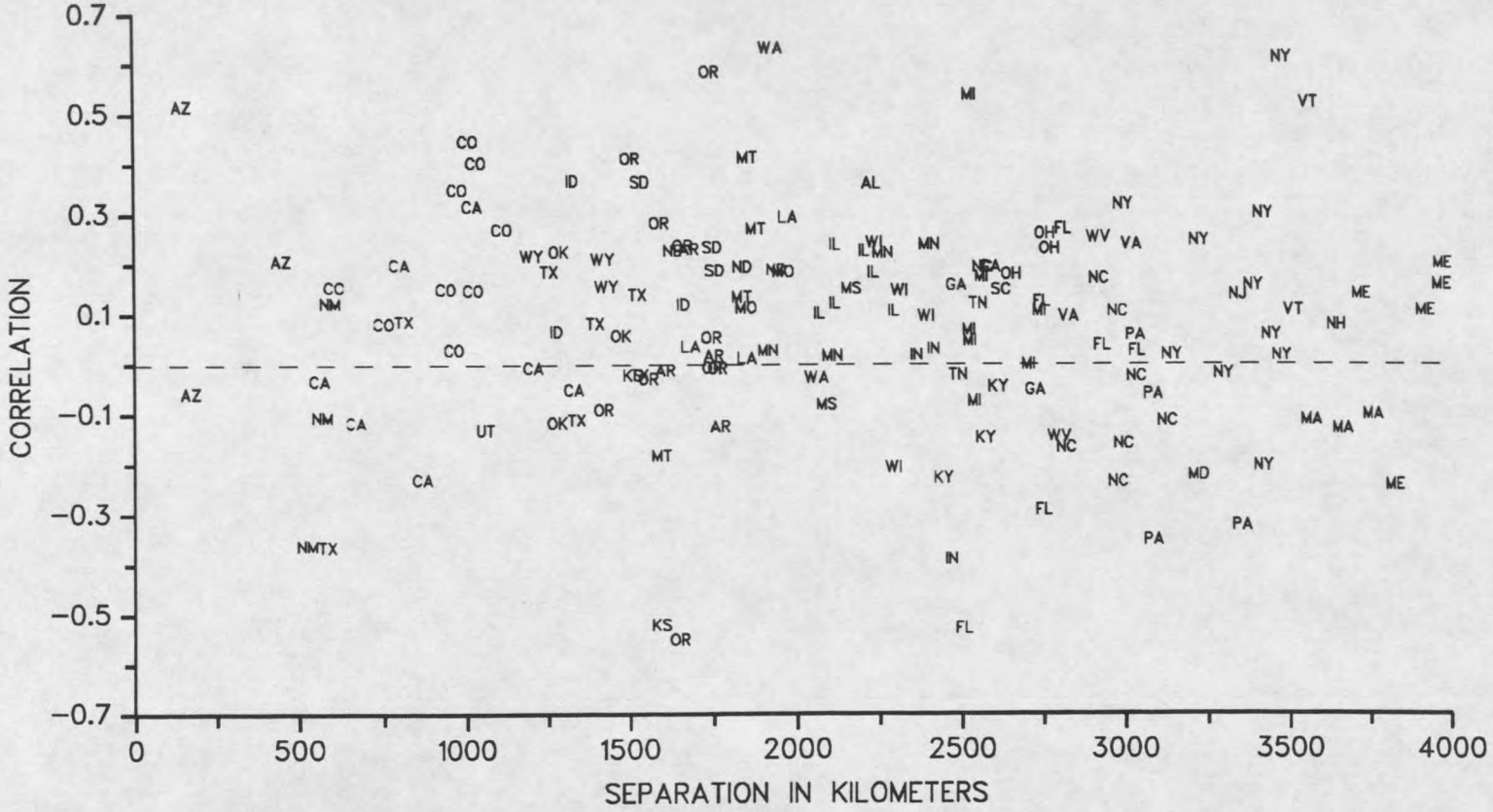

Figure 16. Correlations of concentration of SO_4 in wet deposition and SO_2 emissions from smelters vs distance from smelters to monitor. Stations labelled by state. Dashed line is zero correlation. Deposition adjusted for gauge, seasonal pattern, and local emissions.

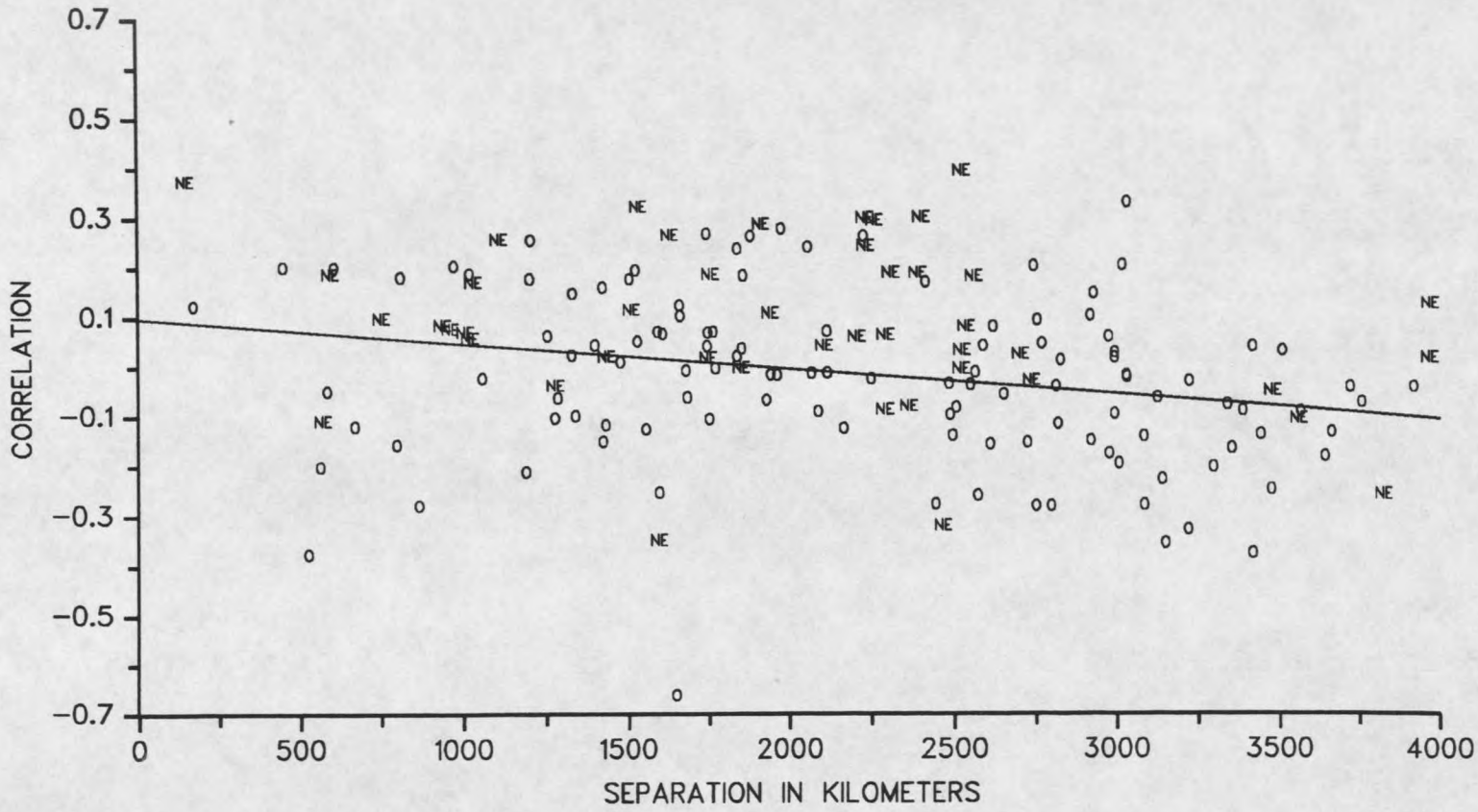

Figure 17. Linear regression of correlation vs distance from smelters to monitor. Stations labelled by orientation. Unadjusted deposition.

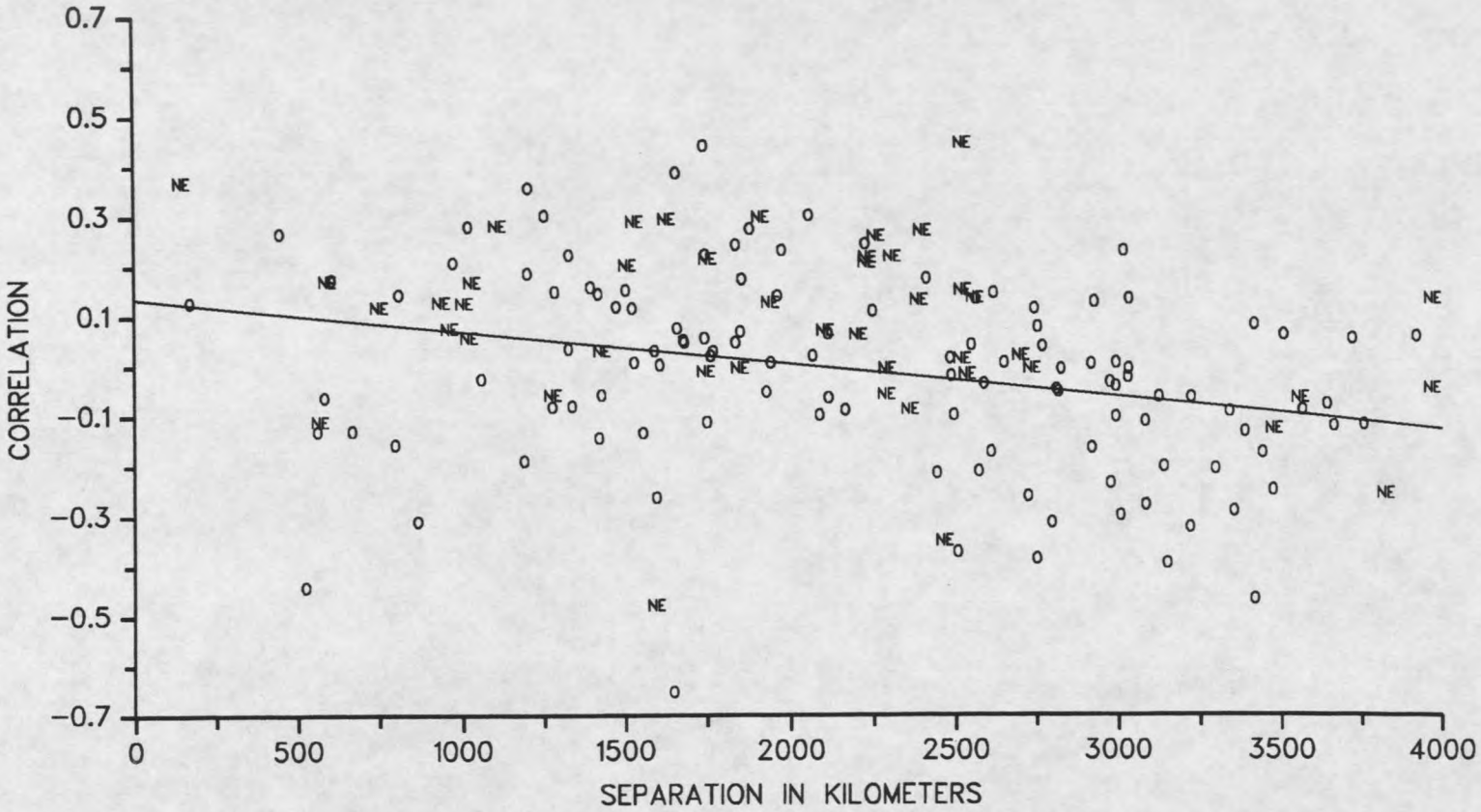

Figure 18. Linear regression of correlation vs distance from smelters to monitor. Stations labelled by orientation. Deposition adjusted for gauge.

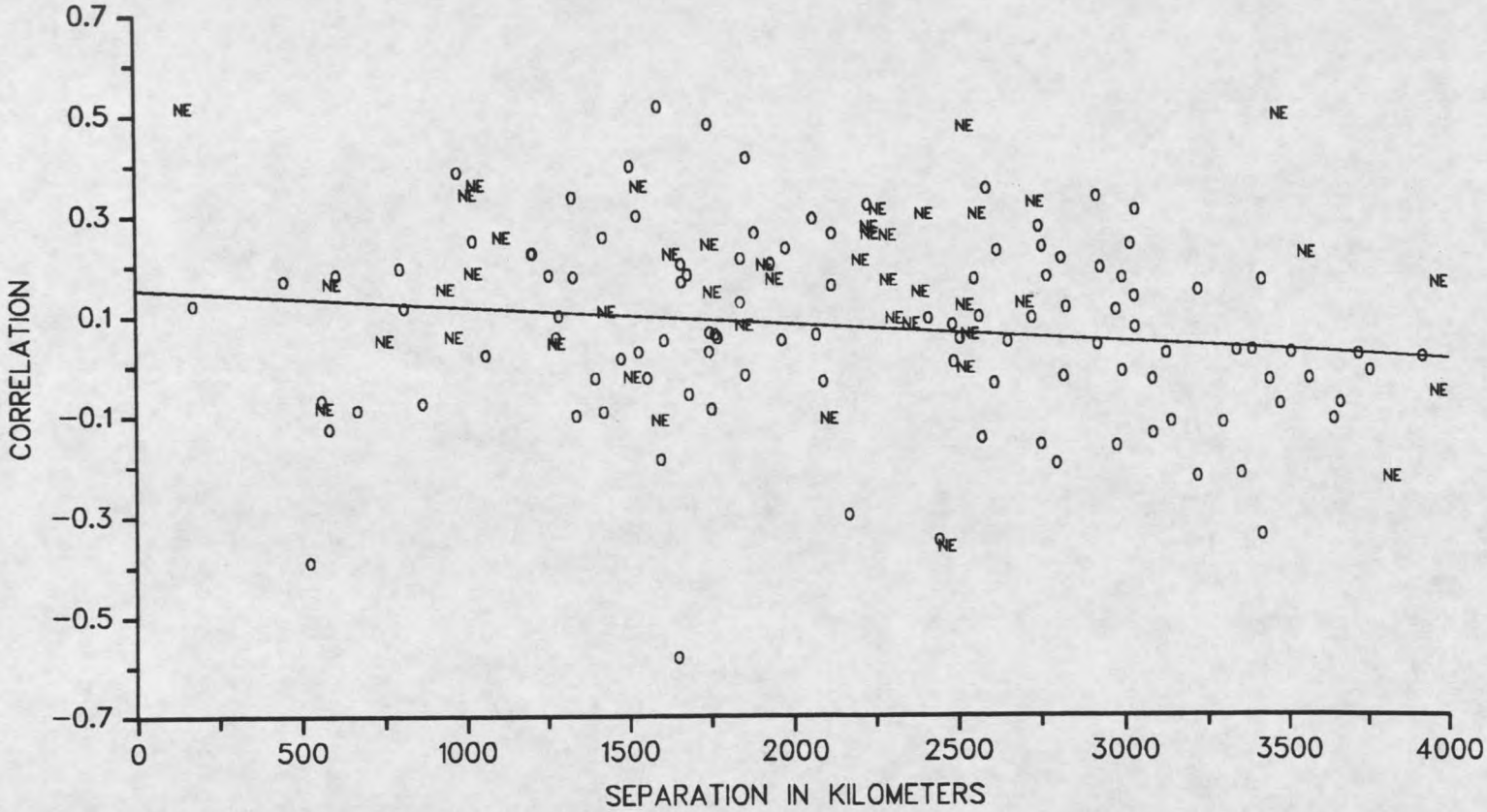

Figure 19. Linear regression of correlation vs distance from smelters to monitor. Stations labelled by orientation. Deposition adjusted for seasonal pattern.

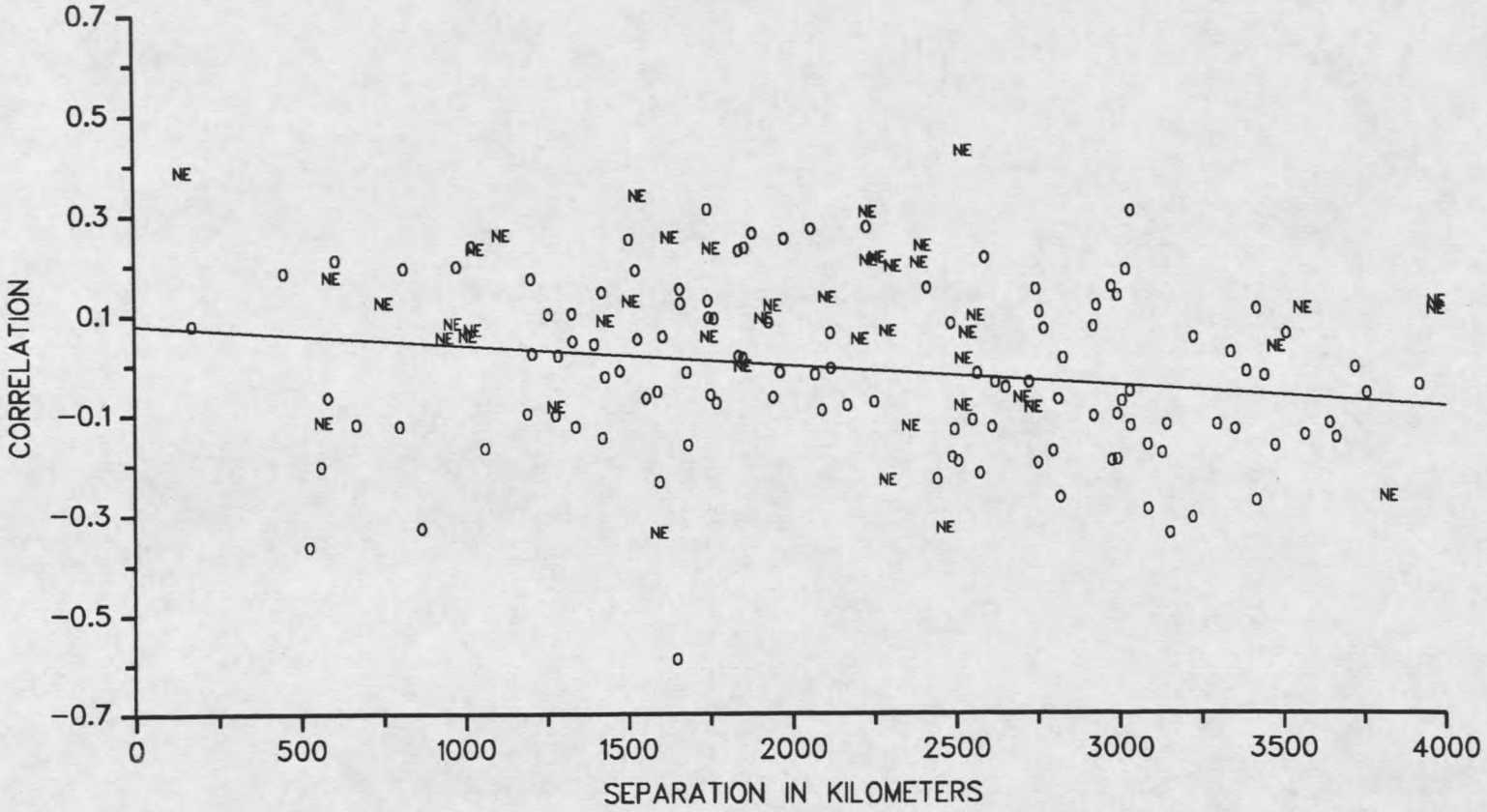

Figure 20. Linear regression of correlation vs distance from smelters to monitor. Stations labelled by orientation. Deposition adjusted for local emissions.

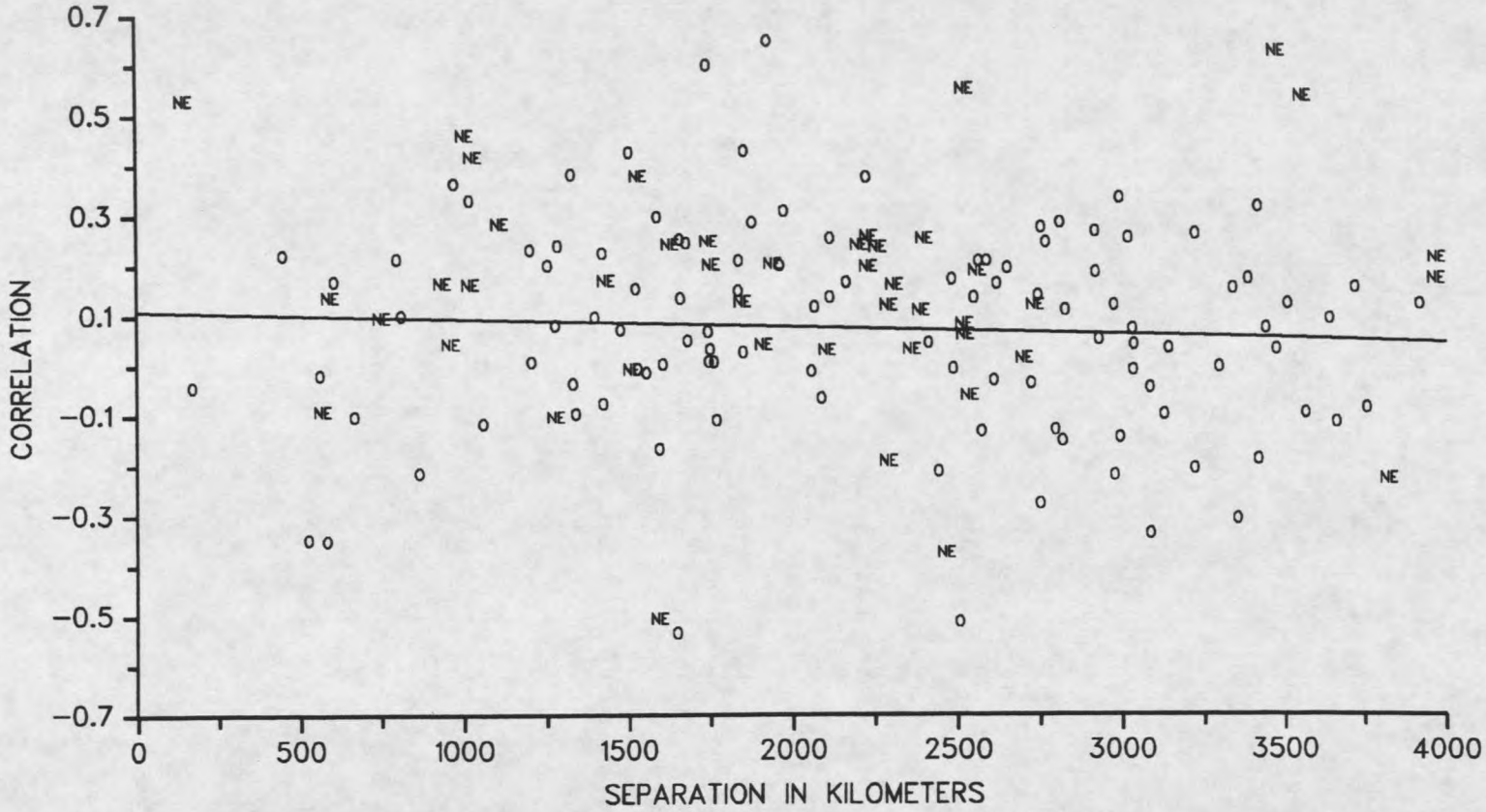

Figure 21. Linear regression of correlation vs distance from smelters to monitor. Stations labelled by orientation. Deposition adjusted for gauge, seasonal pattern, and local emissions.

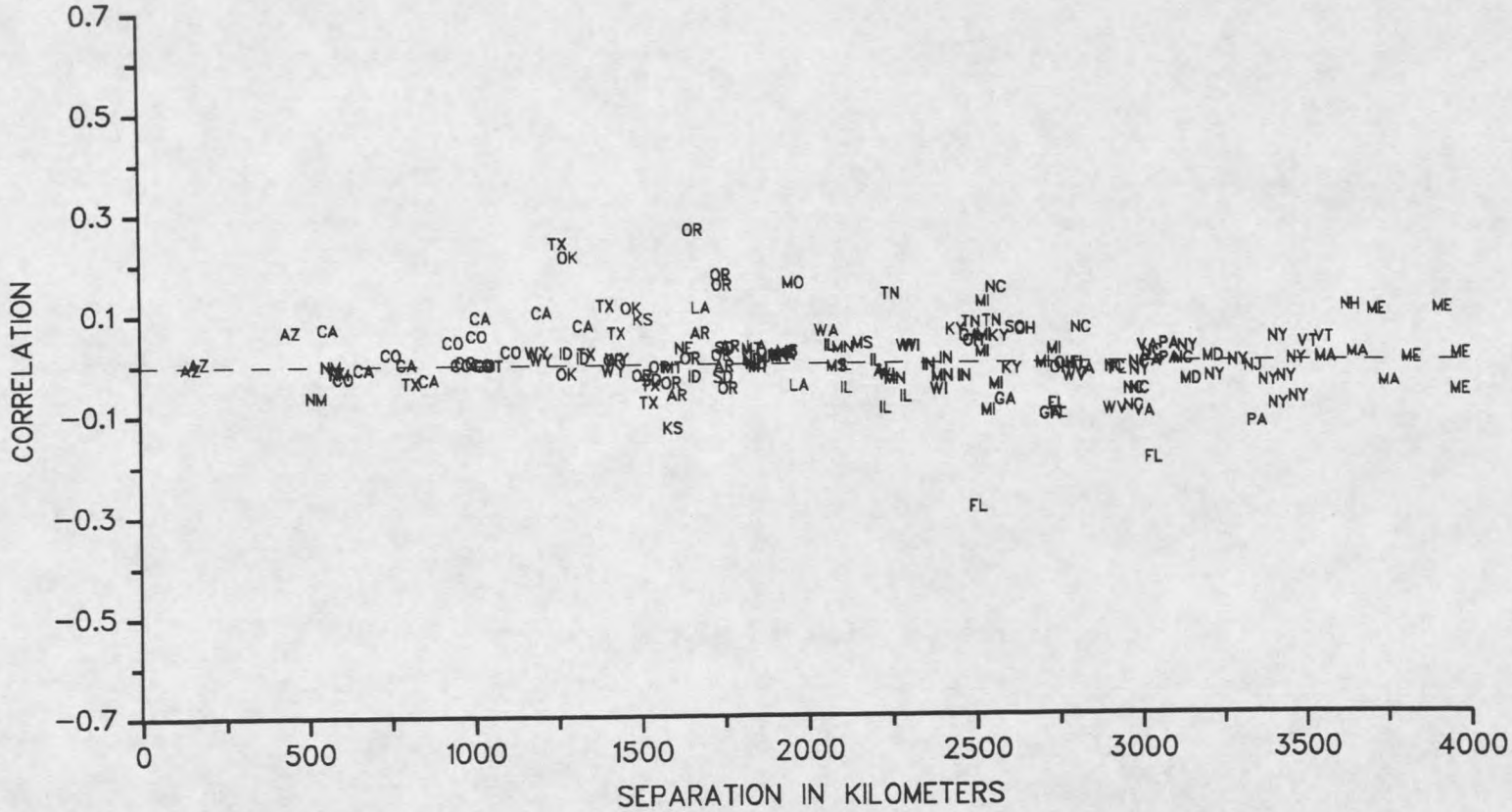

Figure 22. Change in correlation of monitoring station with smelters after deposition corrected for effect of gauge

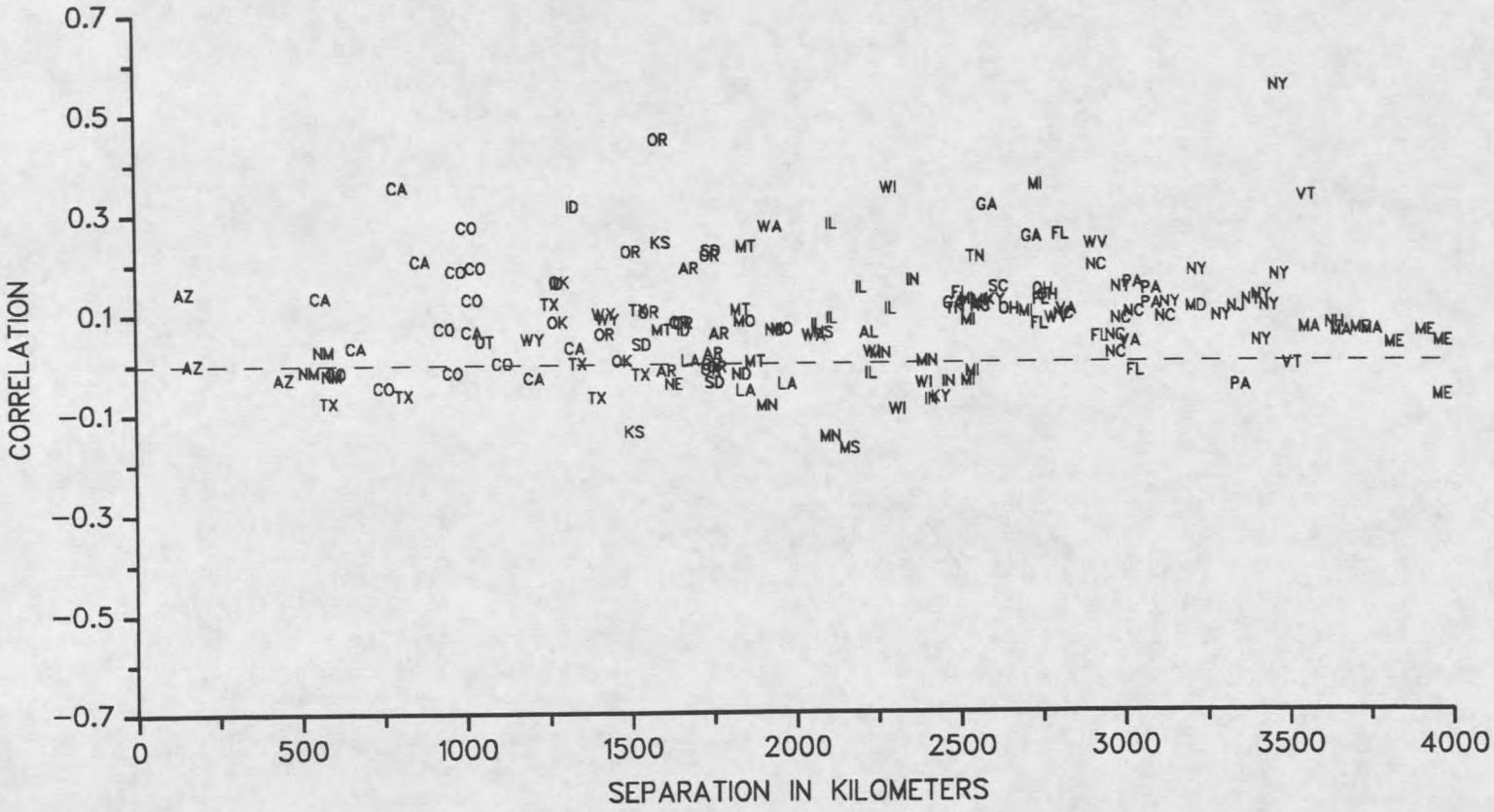

Figure 23. Change in correlation of monitoring station with smelters after deposition corrected for the effect of seasonal pattern

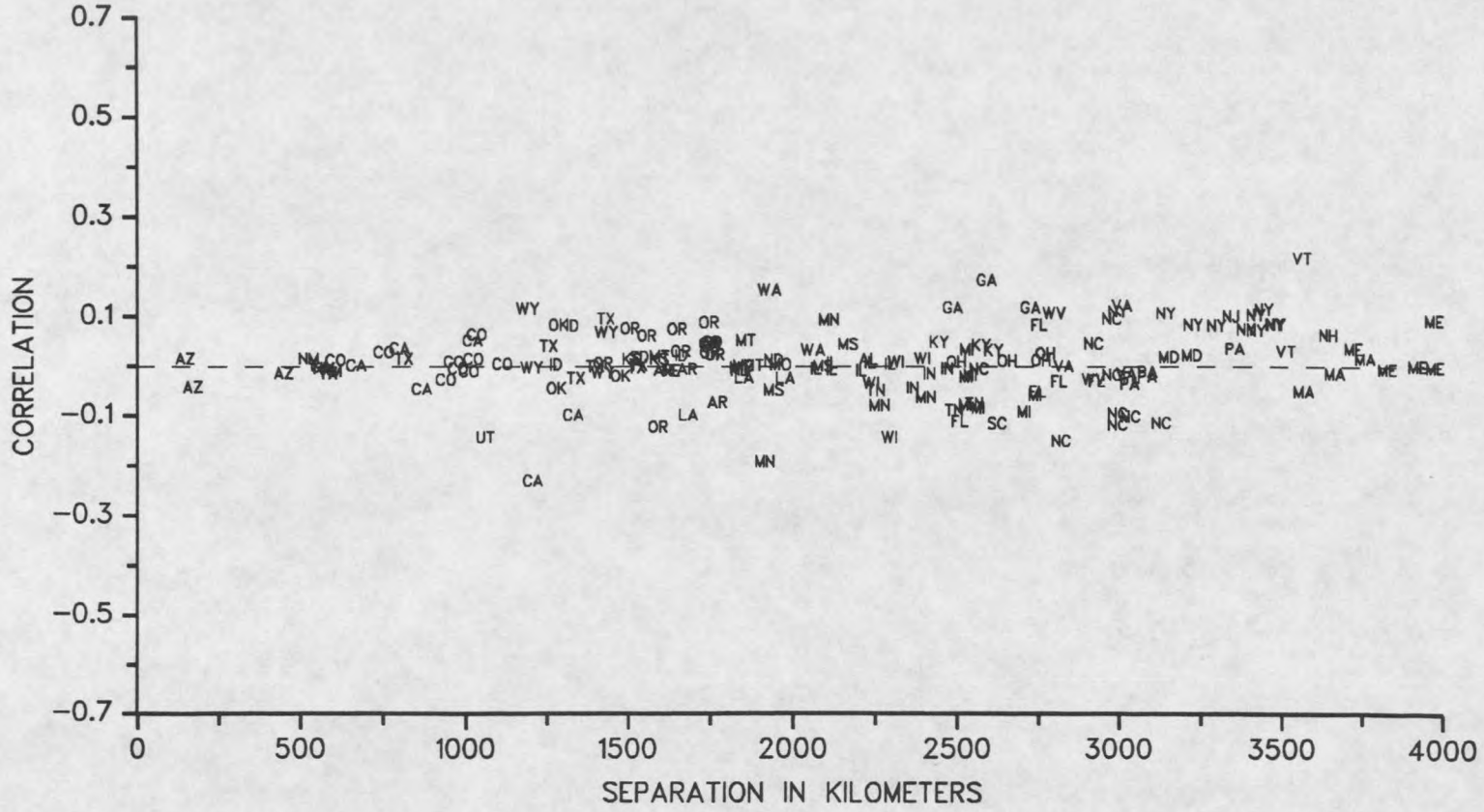

Figure 24. Change in correlation of monitoring station with smelters after deposition corrected for the effect of local emissions

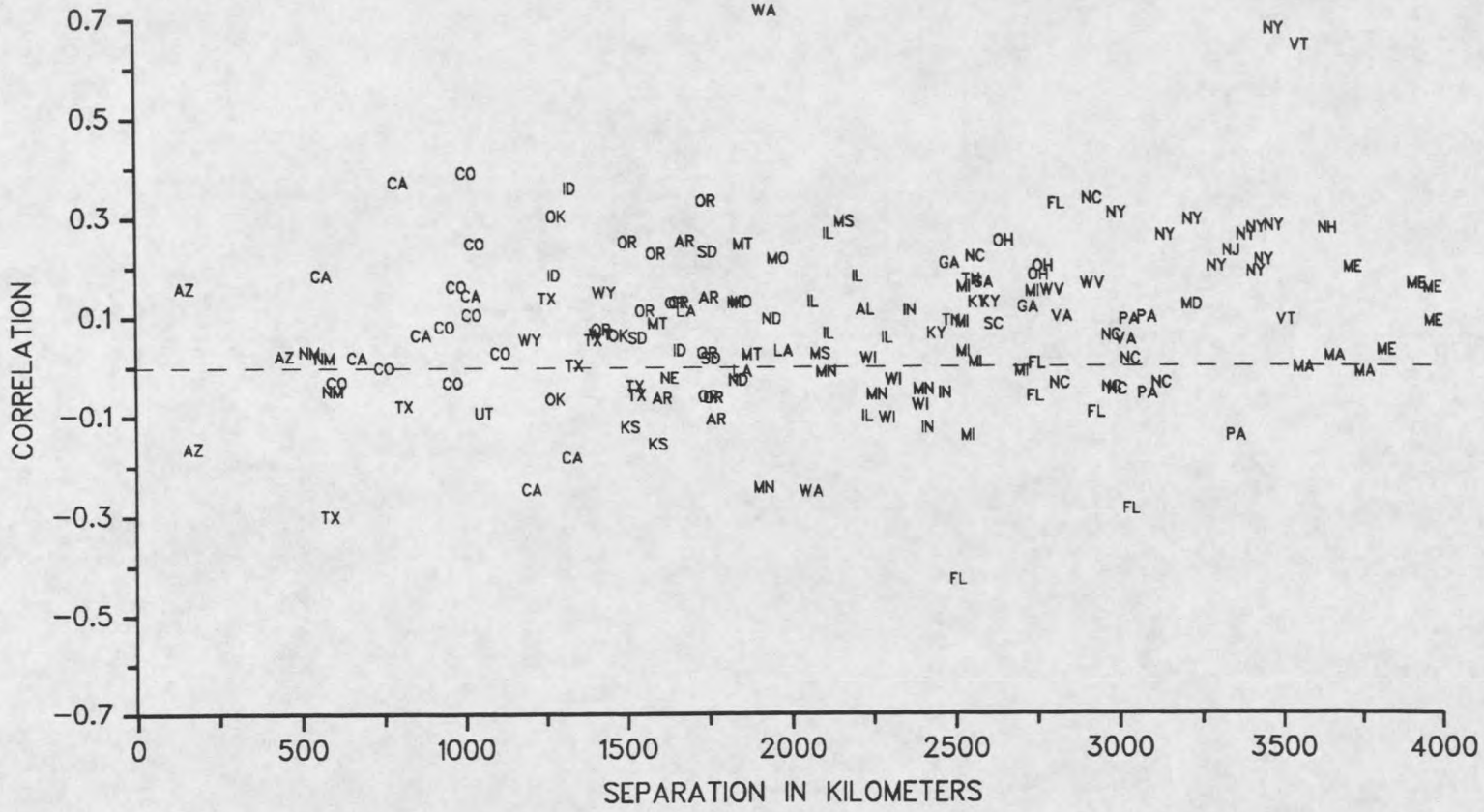

Figure 25. Change in correlation of monitoring station with smelters after deposition corrected for the effect of gauge, seasonal pattern, and local emissions

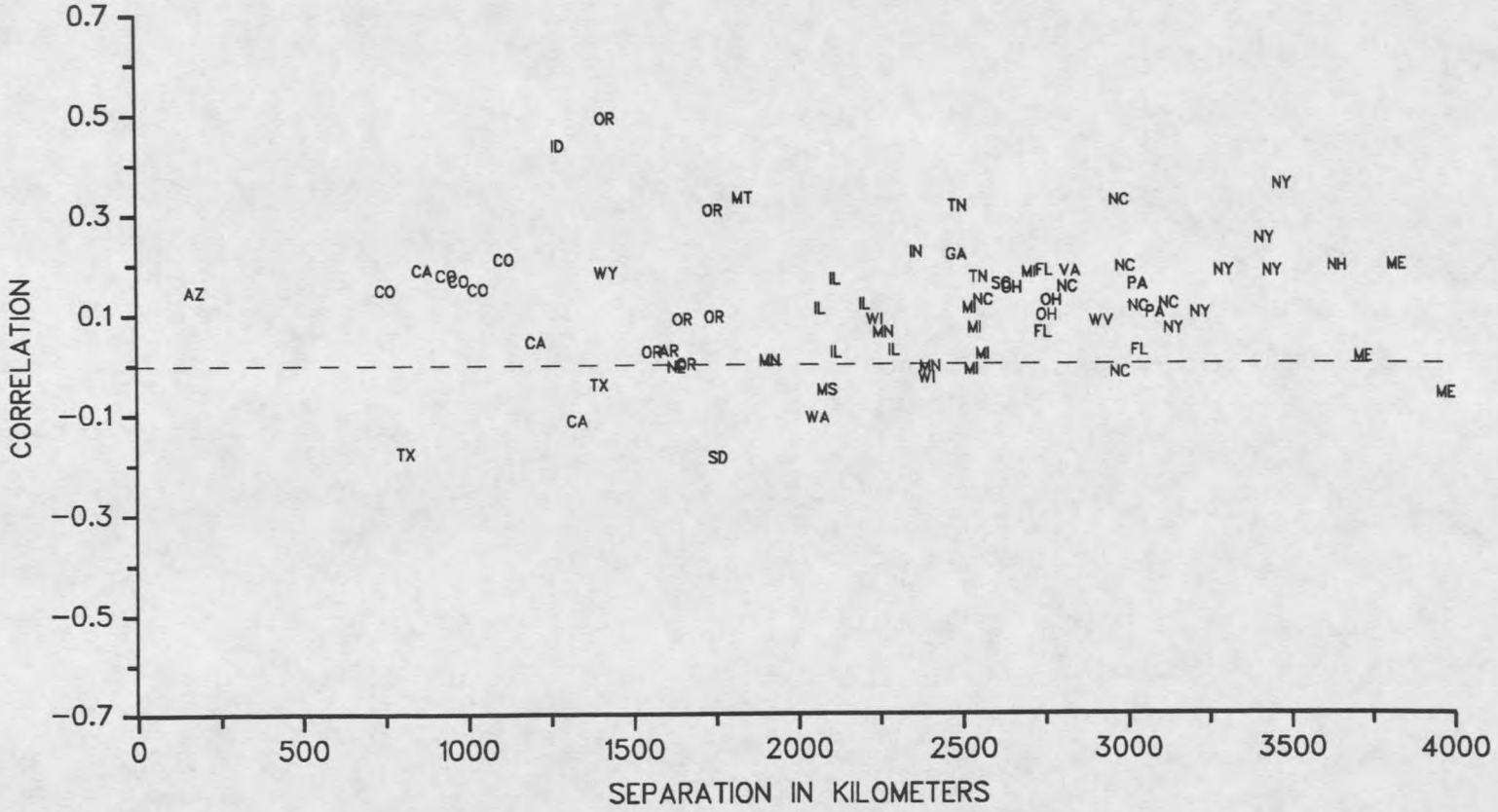

Figure 26. Change in correlation of monitoring station with smelters when 1980 data is excluded. Only those stations in operation in 1980

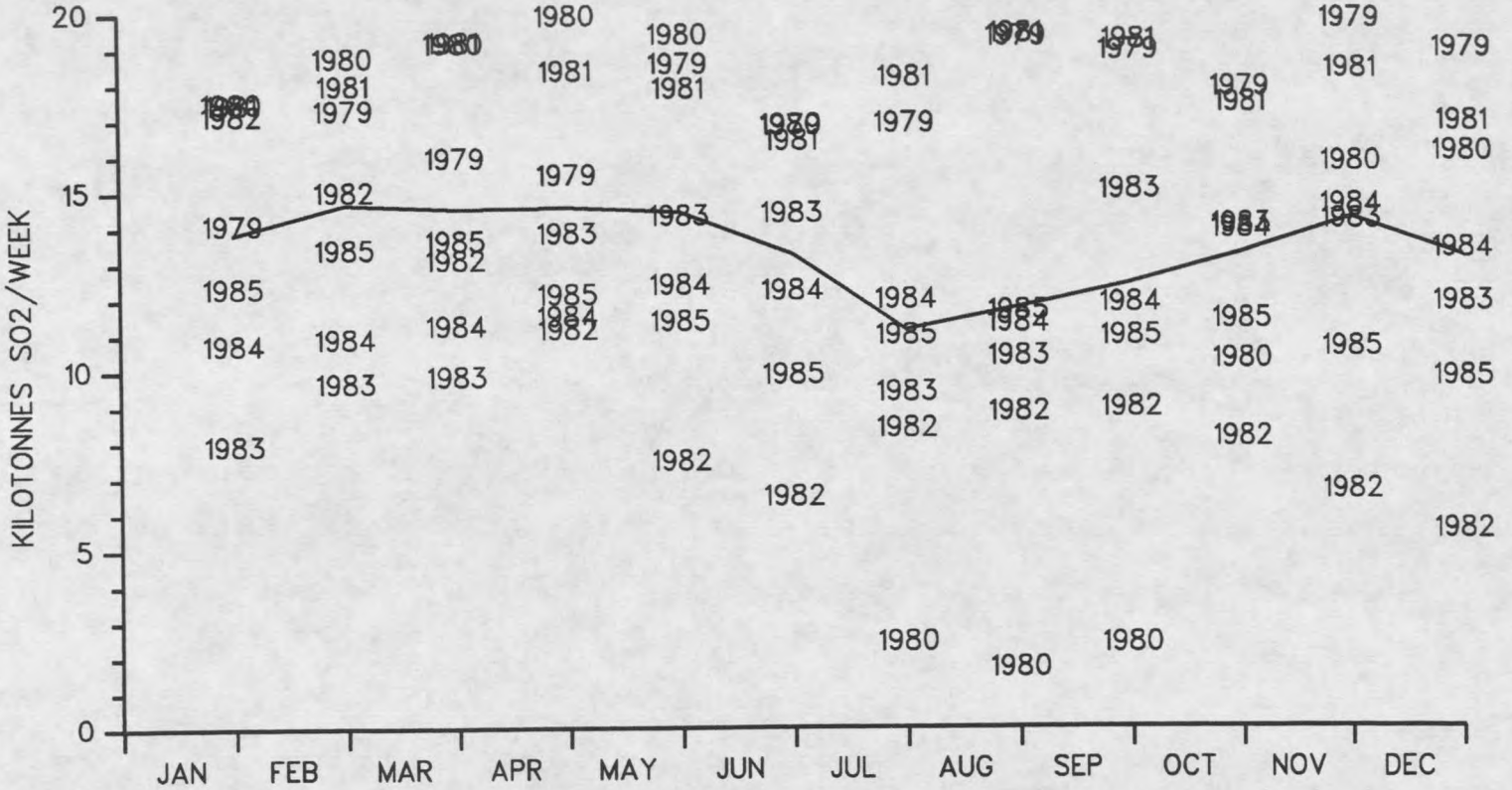

Figure 27. Smelter SO_2 emissions, 1979-1985. Solid line is seasonal average

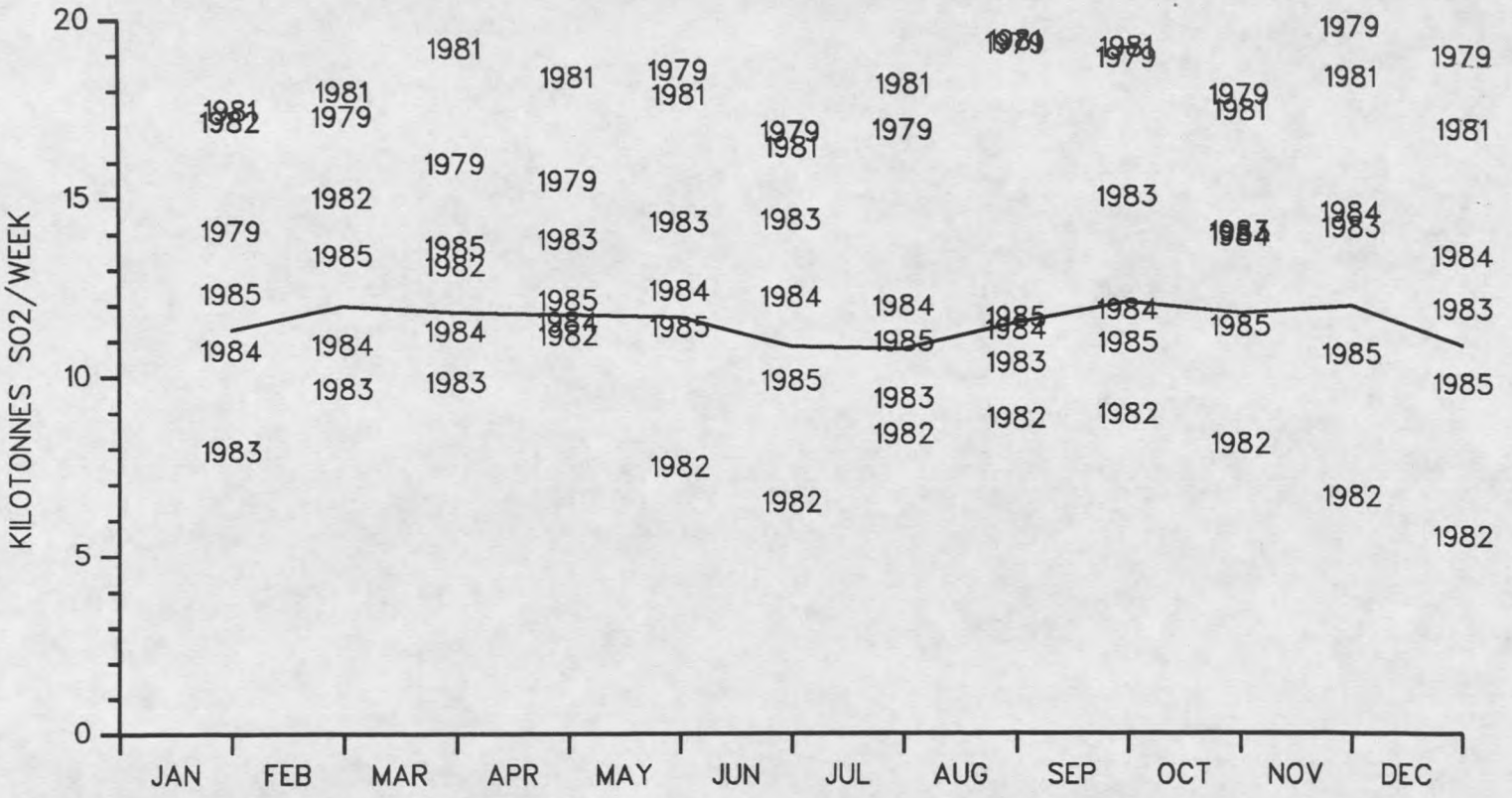

Figure 28. Smelter SO_2 emissions, 1979-1985, excluding 1980. Solid line is seasonal average.

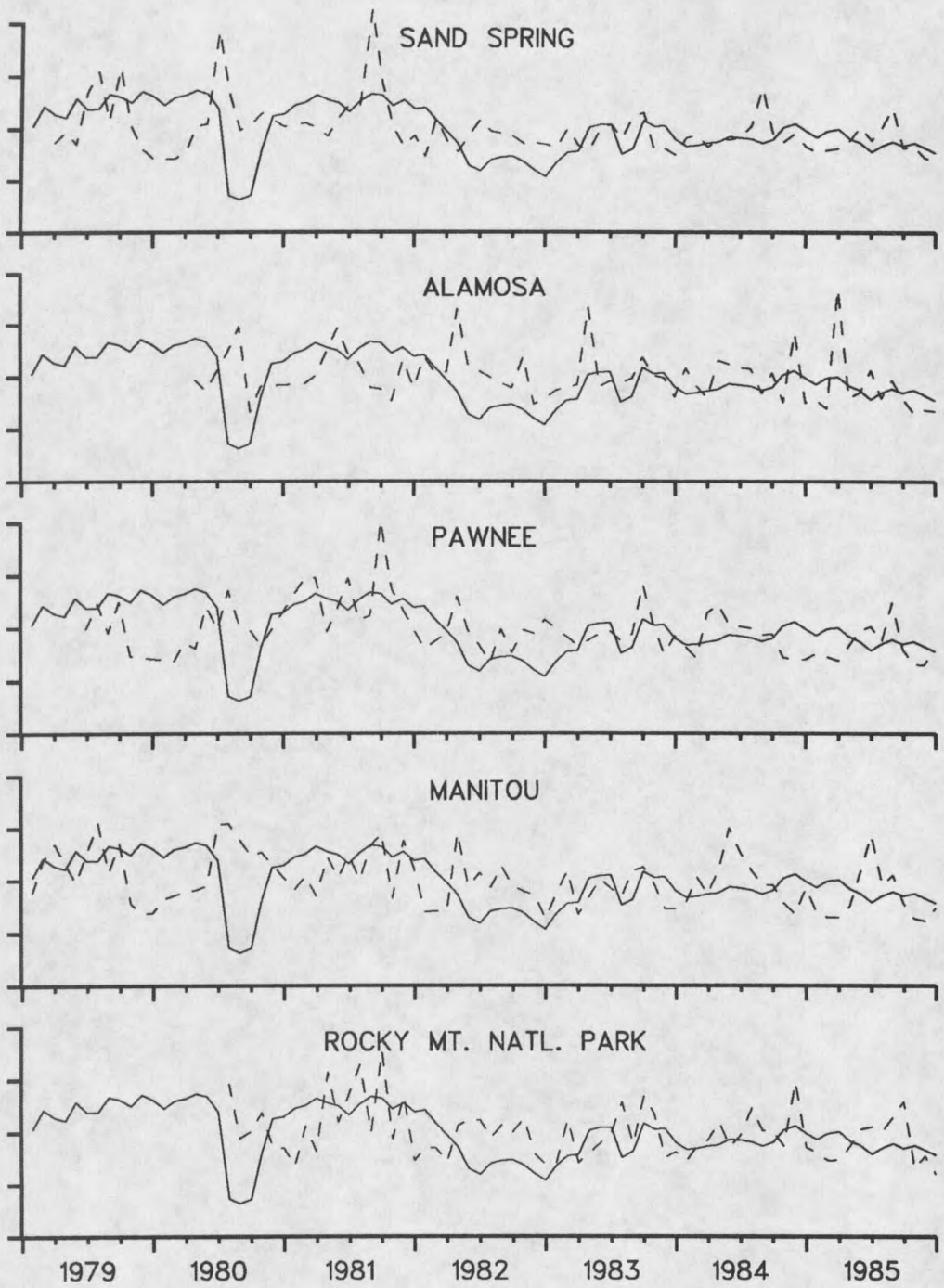

Figure 29. Temporal series of emissions and deposition. Solid line is summed Arizona and New Mexico weekly SO_2 emissions rate from copper smelters. Dashed lines are SO_4 concentrations in wet deposition at Colorado monitoring stations.

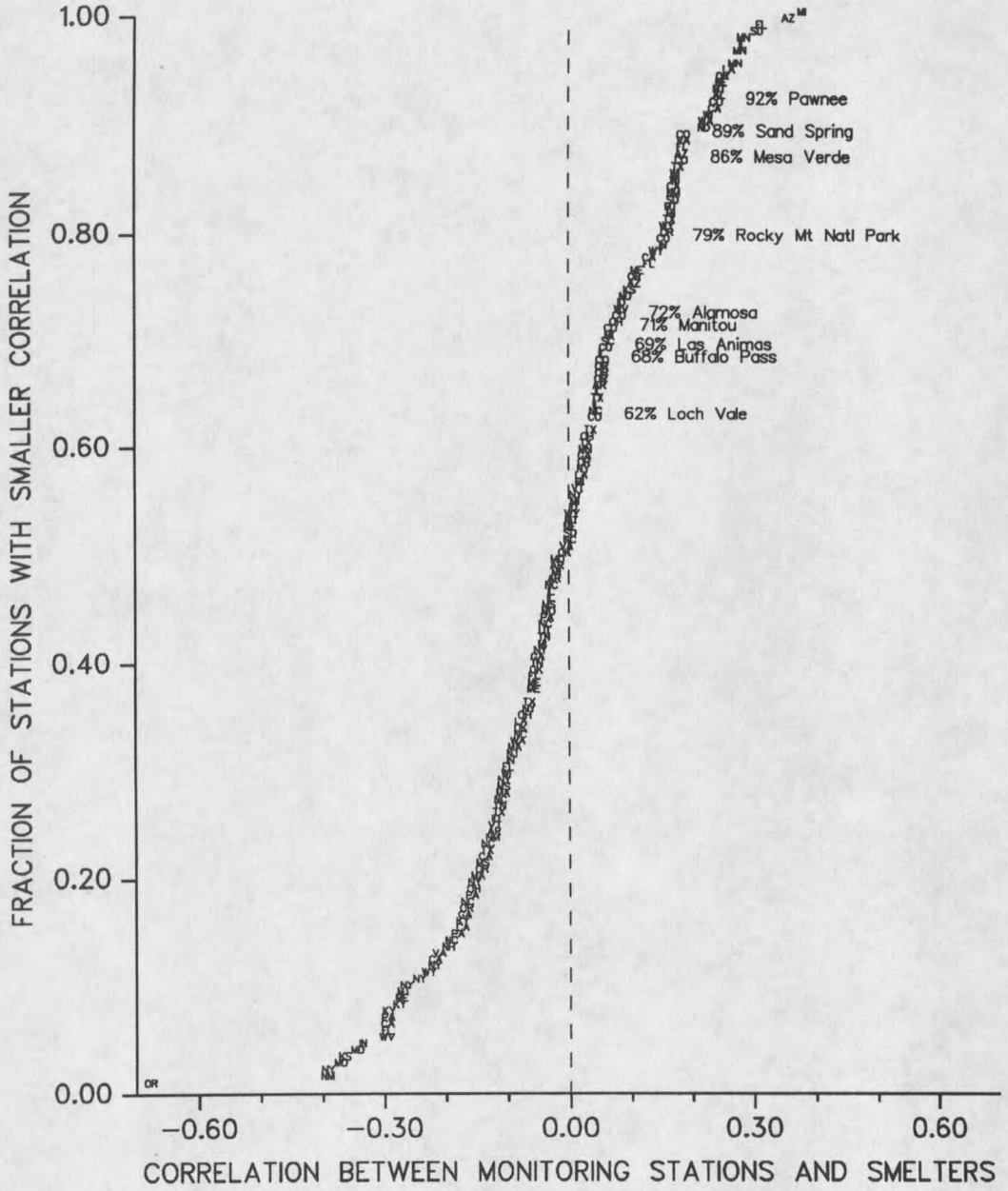

Figure 30. Cumulative distribution function of correlation between monitoring stations and smelters. Unadjusted deposition.

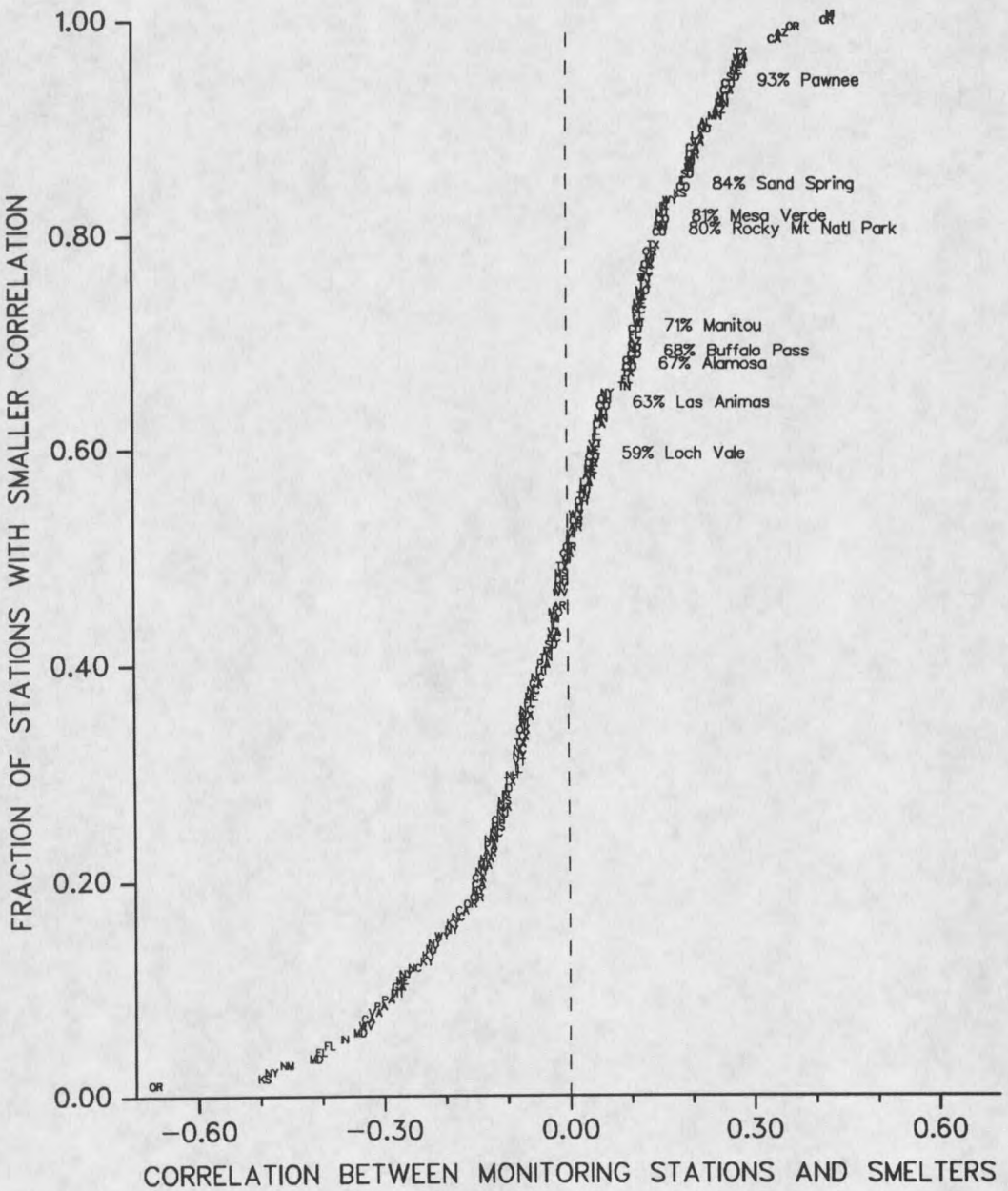

Figure 31. Cumulative distribution function of correlation between monitoring stations and smelters. Deposition adjusted for gauge.

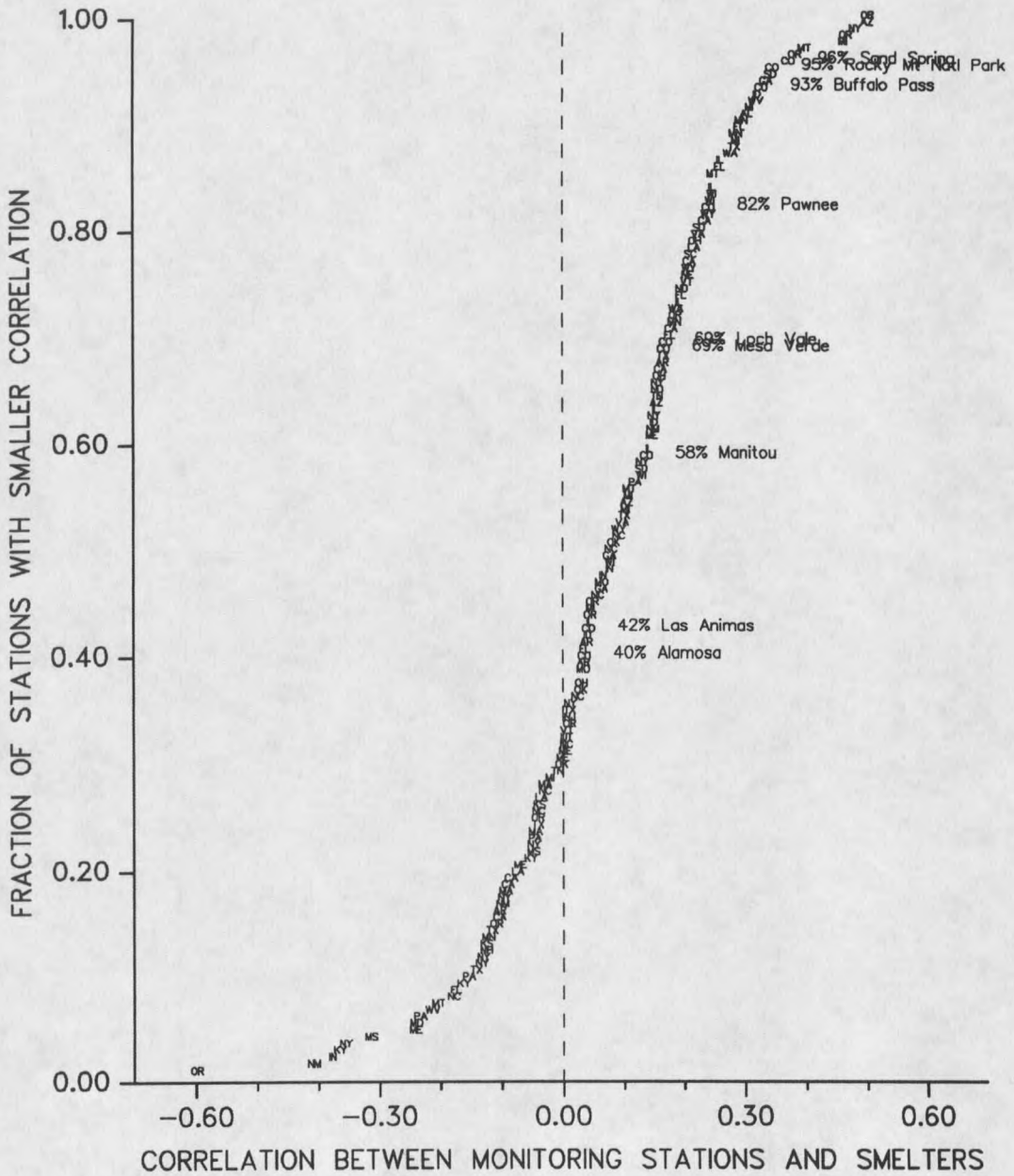

Figure 32. Cumulative distribution function of correlation between monitoring stations and smelters. Deposition adjusted for seasonal pattern.

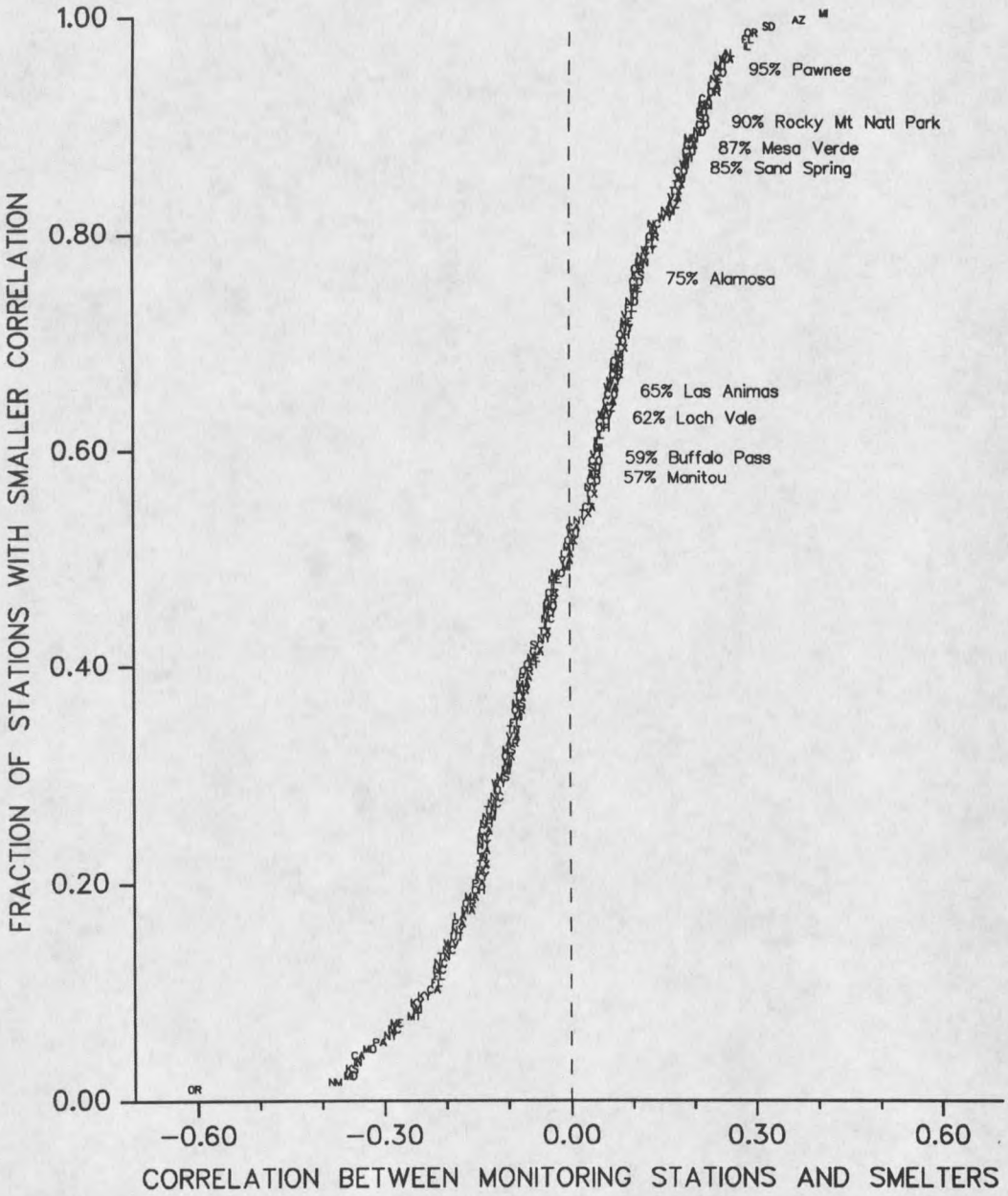

Figure 33. Cumulative distribution function of correlation between monitoring stations and smelters. Deposition adjusted for local emissions.

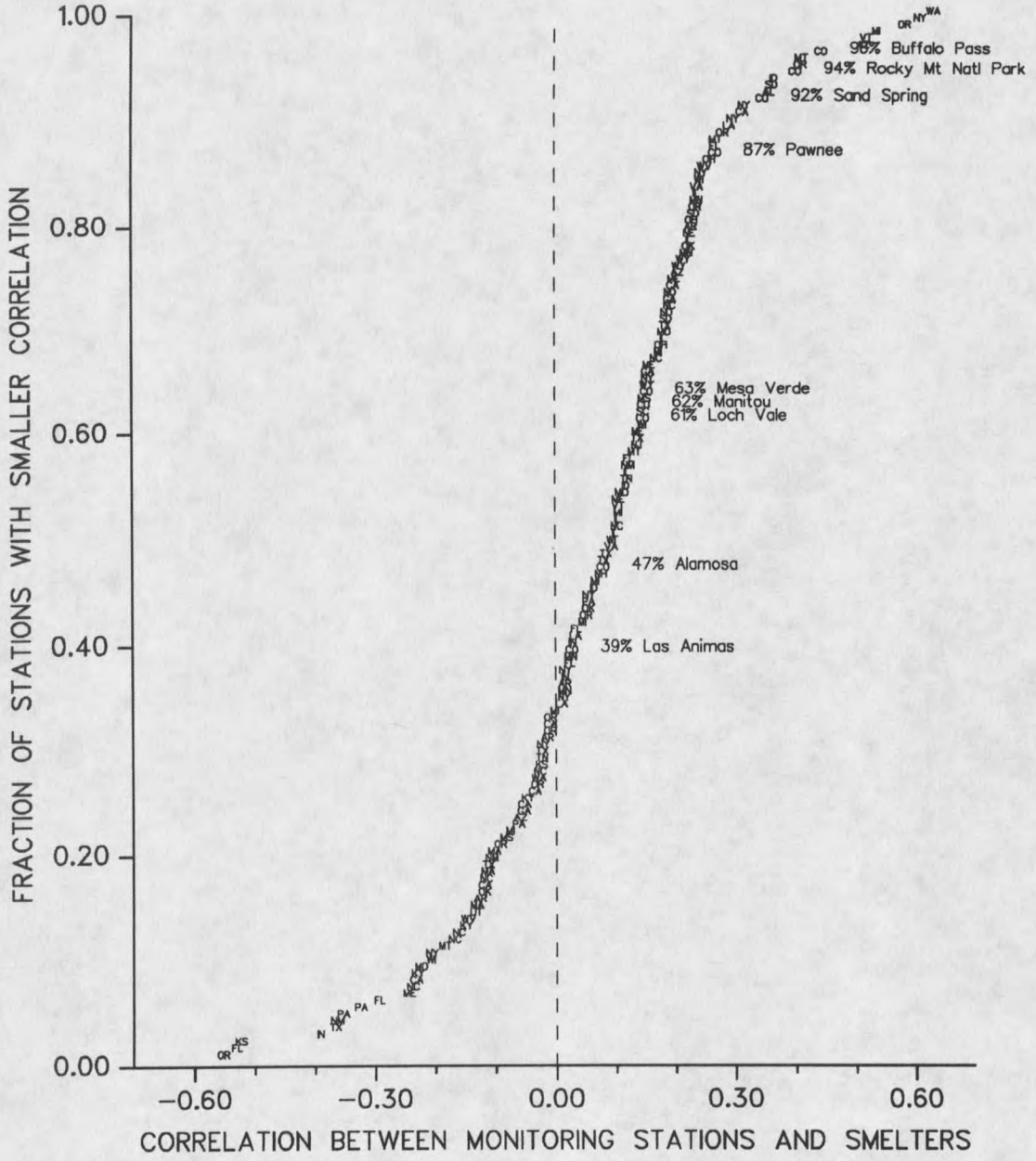

Figure 34. Cumulative distribution function of correlation between monitoring stations and smelters. Deposition adjusted for gauge, seasonal pattern, and local emissions.