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APPLICATION OF TRACER MASS BALANCE REGRESSION TO WHITEX DATA

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Abstract

The Winter Haze Intensive Tracer EXperiment (WHITEX) was designed to evaluate the feasibility of using a variety of receptor models to attribute emissions from a single point source, the Navajo Electric Generating Station (NGS) to visibility impairment in a number of national park areas. One of the receptor models, Tracer Mass Balance Regression (TMBR), is a linearized version of a deterministic statement of how gases are transported and transformed as they move from source to receptor. It essentially uses variations of unique trace gases or particulates over time to link source emissions to secondary aerosol concentrations. TMBR analysis of data gathered at Hopi Point, Grand Canyon were used to estimate NGS contribution to secondary sulfate and nitrate at that park unit. NGS was estimated to contribute between 70–80% of the sulfate concentration on the rim of Grand Canyon National Park while copper smelters may contribute between 10–30%. Background sulfate is estimated to be between 0.0 and 10%.

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Introduction

There are a number of receptor oriented models that have been used to attribute primary aerosols to their sources, while on the other hand very few attempts have been made to apportion secondary aerosols using receptor oriented techniques. The chemical mass balance (CMB) model has historically been the most popular receptor oriented model. It essentially uses the relative ratios of tracers of opportunity or man-made tracers at the sources and receptor locations to apportion primary species concentrations at the receptor site. Other common types of models include: 1) principal component analysis (PCA); 2) multiple linear regression (MLR); 3) tracer mass balance regression (TMBR); and 4) differential mass balance (DMB). Explanations of these models are given by Watson[1,2,3] Chow,[4] Hopke,[5] and Malm *et al.*[6] Furthermore, Dzubay et al.,[7] Lewis and Stevens,[8] and Stevens and Lewis[9] have integrated a number of these approaches into a hybrid receptor model that can be used to apportion secondary aerosols.

The <u>W</u>inter <u>H</u>aze <u>I</u>ntensive <u>T</u>racer <u>Ex</u>periment (WHITEX) was designed to evaluate the feasibility of using these models to attribute single point source emissions to visibility impairment in selected geographic regions.[10] WHITEX was conducted during January and February 1987 in the vicinity of the Navajo Generating Station (NGS), a large (2250 MWe) coal-fired power plant located relatively close to several national parks including the Grand Canyon. The primary receptor sites were Grand Canyon and Canyonlands National Parks and Glen Canyon National Recreation Area (Page, Arizona). Figure 1, a three-dimensional perspective of the WHITEX study region, shows key monitoring sites, topographic features, and emission sources. This paper explores the use of TMBR to apportion SO_4 concentrations at Hopi Point, Grand Canyon.

The TMBR Model

The TMBR model, a linearized version of the deterministic general mass balance (GMB) formalism, will be presented here without derivation. The interested reader is referred to a companion paper by Malm *et al.*[6] for specific details and assumptions for deriving the TMBR equations.

Notation

The subscript i will be used for indexing the species, j for sources and k for sampling periods. The subscript i_u will be used to index unique tracer species that can be associated with groups of sources all having that tracer in common. A unique tracer species could be replaced with a fraction of some reference species that has been calculated using CMB or some other appropriate model.

C_{ik} = concentration of aerosol species i to be apportioned at the receptor for sampling period k.

C_{i_uk} = ambient concentrations of tracer species i_u at the receptor corresponding to sampling period k.

γ_{i_u} = fractionation coefficient associated with tracer species i_u when TMBR equations have been linearized using some independent physical variable such as relative humidity. In a regression analysis γ_{i_u}'s are the regression coefficients.

ϕ_{i_uk} = a function of known physical variables that can be used to linearize the GMB equations. In the simplest version of the TMBR model $\phi_{i_uk} = 1.0$.

RH_k = relative humidity at the receptor site during sampling period k.

Model Equations

The TMBR equation is given by

$$C_{ik} = \gamma_0 + \sum_{u=1}^{h} \gamma_{i_u} C_{i_uk} \phi_{i_uk} \tag{1}$$

for each sampling period $k = 1, 2, \ldots, s$. h is the number of contributing source or source types to aerosol species C_{ik}. The quantities C_{i_uk}, C_{ik}, and ϕ_{i_uk} are assumed known. We thus have a set of s linear equations in $h+1$ unknowns γ_0, γ_{i_1}, γ_{i_2}, …, γ_{i_h}. If the system of equations has rank $h+1$, the gamma coefficients may be obtained by solving the above system of linear equations. The apportionment of the species of interest to the various groups of sources is then carried out by calculating the individual terms of the equations above.

In the simplest version of the TMBR model $\phi_{i_uk} = 1.0$. Malm *et al.*[6] have shown that with a number of assumptions $\phi_{i_uk} = RH_k$ for the case where SO_2 is oxidized to SO_4. Then Equation 1 becomes

$$C_{SO_4 k} = \gamma_0 + \sum_{u=1}^{h} \gamma_{i_u} C_{i_u k} RH_k. \qquad (2)$$

The use of RH as a linearization parameter suggests that the rate of SO_2 to SO_4 oxidation is proportional to relative humidity. This assumption does not necessarily imply that the RH dependence of oxidation is based on basic physio-chemical processes. The mechanism for RH dependent oxidation may be associated with the amount of time chemical processes are associated with in–cloud mechanisms. As RH climbs, cloud formation increases and the probability of SO_2 oxidation being associated with in–cloud chemistry increases.

Model Assumptions

The TMBR model assumptions are:

- The chemical species used as tracers in the model are assumed to be uniquely emitted by non-overlapping groups of sources. In particular none of the species other than the tracer associated with the source of interest can be emitted by another source unless there is an independent method such as CMB to partition the ambient species concentrations into components attributable to the various groups of sources.
- The composition of source emissions are constant over the period of ambient sampling.
- Deposition and conversion are constant from one sampling period to the next for each subgroup u.
- Measurement errors are random, uncorrelated, and normally distributed.

For the case where $\phi_{i_u k} = RH_k$ additional assumptions are:

- Exponential forms of deposition and conversion equations can be represented by first order approximations.
- The RH at the receptor site is indicative of the amount of time that air parcels spend in contact with clouds and therefore sulfur dioxide oxidation is assumed proportional to RH.

Measurements

Fine particles (smaller than 2.5 μm) were collected on Teflon, nylon, and quartz filters by IMPROVE[11] samplers. These filters were analyzed by a variety of methods: gravimetric analysis, particle induced x-ray emission (PIXE), ion chromatography, and thermal optical reflectance. Concentrations were determined for mass, major and trace elements, sulfate, nitrate plus nitric acid vapor, and organic and elemental carbon. Samples were gathered on a six or twelve hour basis. For purposes of analysis, all data were averaged to twelve hours. A second fine particle sampler, referred to as SCISAS,[12] was run on a 24 hour basis. Trace elements from SCISAS samples were obtained using X-ray fluorescence (XRF). Since in a number of cases Se from IMPROVE samples was below detectable limit, Se derived from SCISAS samples was used in data analysis. Finally, since SCISAS data were taken on a 24 hour sampling schedule and IMPROVE data on a 12 hour schedule, the SCISAS data were disaggregated to 12 hour values.[13]

Deuterated methane (CD_4) was released from one of NGS's three stacks during the WHITEX study.[14] CD_4 is a stable non-reactive species (even at elevated stack temperatures) and is presumed to have a deposition velocity close to zero.[15] Though chemically similar to normal methane, heavy methane has a higher molecular weight which allows it to be distinguished by mass spectrometry at very low concentrations. CD_4 is not emitted by other sources and has an average background concentration of approximately 1 part in 10^{16}.

Data

The NGS power plant was operating continuously, if not uniformly, throughout the WHITEX study period. The three-unit plant was run at times at near its maximum capacity (2250 MW_e); however, much of the time one unit was not operating, and during two days (JD=38-40) two units were not operating.

An attempt was made to vary the tracer emission rate in proportion to the power plant load, i.e., emissions were to maintain a constant CD_4 to SO_2 ratio at NGS. However, the attempt to control the release rate was not particularly successful. The ratio of CD_4 tracer to power plant load (and hence to emissions) was not constant but varied significantly around the average.

Scaling of CD_4 Data

Because the TMBR analysis relies on a constant CD_4/total emission ratio, the CD_4 data must be scaled or 'standardized' to a constant CD_4 emission rate.

Scaling is valid if the CD_4 release rate is constant over a time period which is long compared to travel time from the source to the receptor, or if the CD_4 is scaled as a function of NGS air mass age. The rate of change of CD_4/megawatt ratio was on the order of days, and since travel time of NGS emissions from Page, Arizona to a receptor site is variable and can also be on the order of days, the CD_4 ambient concentrations cannot be directly scaled to the NGS CD_4/megawatt ratio. However, CD_4 data can be scaled using the approximate age of the air mass that carried CD_4 into Grand Canyon. CD_4 scaled to emissions and plume age will be referred to as fully scaled CD_4. The temporal history of the CD_4 release rate, unscaled CD_4 data, and CD_4 data scaled to CD_4 release rate and air mass age for Hopi Point are shown in Figure 2. The standard CD_4 emission rate used was 2.5 mg/MWe-h. Procedures that were used to calculate air mass age for various receptor sites are described by Latimer *et al.*[16] By comparing the scaled and unscaled CD_4 time lines the action of the standardization can be seen; whenever the release rate increases above 2.5 the scaled CD_4 values are reduced, and vice versa.

It should be pointed out that even though CD_4 samples were gathered on all days of the experiment, analysis of CD_4 data was limited to those days shown in Figure 2 because of high cost of the sample analysis procedure. The time periods chosen correspond to days before, during and after elevated sulfate episodes. Shown in Figure 3 are temporal plots of sulfate and sulfur dioxide sulfur, fully scaled CD_4, selenium, arsenic, and relative humidity for Hopi Point, Grand Canyon.

TMBR Calculations

If a single unique tracer such as SCD_4 (scaled CD_4) is associated with a source, a regression analysis using the aerosol that is to be attributed as the dependent variable and tracer as the independent variable has a fairly simple interpretation. The intercept is the average background concentration, while the slope of the regression line multiplied by the tracer is the concentration associated with the source having the unique tracer. On the other hand, if more than one unique tracer is available, then the TMBR analysis can be extended to include as many sources as there are unique tracers, provided the number of sources does not exceed the number of data points.

For purposes of attributing total sulfur or sulfate sulfur to NGS, SCD_4 is considered to be a unique NGS tracer. Furthermore, As was found to be below the detectable limit in samples gathered from within the NGS plume. Therefore, As is considered to be a unique tracer for emissions other than NGS, and most probably is associated with copper smelter emissions. Finally, copper smelter trace element emission profiles usually exhibit relatively small amounts of Se.[17] Therefore, Se is assumed to be a tracer that is primarily associated with coal fired power plant emissions with possible small contributions associated with other sources. For purposes of the following sulfur attribution analysis, Se emissions will be associated with coal fired power plant sulfur.

All regressions were carried out using orthogonal distance regression (ODR).[18] ODR explicitly takes into account the uncertainties in both the dependent and independent variables. Usually the ODR regression coefficients will be greater than the corresponding coefficients obtained with ordinary least square (OLS) and the intercept will be smaller. The intercepts were forced to be positive in all ODR regressions.

Uncertainties

Uncertainty formulas can be derived using propagation of error methods and assuming the covariances between various terms occurring in the derivation are negligible. For details of uncertainty calculations the interested reader is referred to Malm *et al.*[6]

Results for Hopi Point, Grand Canyon

Least squares (ODR) estimates of fractionation coefficients obtained from the TMBR analyses using data from Hopi Point are listed in Tables I and II. Estimates were carried out using total sulfur and sulfate sulfur, particulate nitrate, organic, and light absorbing carbon as the dependent variable and SCD_4, Se, and As as the independent variables. The R^2 values for the regressions using total sulfur and just SCD_4 are low. R^2 can be somewhat improved by including other independent variables such as As. The R^2 with just SCD_4 is 0.36 while the addition of As to the regression model improves the R^2 to 0.56. The low R^2 could be due to varying deposition and conversion rates, or the existence of sources of SO_2 not associated with SCD_4 or trace elements.

Furthermore, no significant relationships were found between just SCD_4 and carbon species. Inclusion of other trace elements in the regression model did not significantly improve the R^2. A relationship with carbon species is not expected; however, since NGS is a large source of NO_x, a relationship between SCD_4 and nitrates is expected. While the regression coefficient of nitrate on SCD_4 is statistically significant, the R^2 is low, which could be due to variation in deposition, oxidation rates, or the existence of other nitrate sources.

Regressions with Se, SCD_4, and As as independent variables and SO_4 as the dependent variable were carried out. Using any one of these variables by themselves yielded R^2 values that were low, suggesting that neither coal fired power plants nor copper smelters contribute, by themselves, all of the sulfates found at Grand Canyon, or that deposition and conversion are highly variable.

The model suggested by Equation 2 was tested using $Se \times RH$, and As or $As \times$ RH as independent variables. Results of this analysis are shown as the first two regression models in Table II. Inclusion of the RH term and As in the regression model increased the R^2 to approximately 0.70. Multiplication of As by RH does not make a large difference in either the goodness of fit or the average amount of sulfur attributed to As (copper smelters), suggesting that SO_2 from smelters may have been mostly converted to SO_4 prior to reaching the monitoring site. If it is assumed that Se is associated with all coal fired power plants and As with copper smelters, an attribution of sulfate to these sources can be achieved. It is estimated that between 83 and 88 percent of the sulfate is associated with coal fired power plant SO_2 emissions, 3 to 7 percent to copper smelters, and approximately 10 percent of the sulfate is unaccounted for or associated with other sources. The mean attributions which result from each individual regression along with uncertainties are shown in Table II.

The next two sets of regressions, presented in Table II, were again done with the same variables, but for time periods for which there are CD_4 data. R^2's are approximately 0.80. For the time periods for which there is CD_4, it is estimated that 80 to 82 percent of the sulfate is associated with coal fired power plants, while 18 to 20 percent is attributed to copper smelters. 0.0 ± 1 percent is attributed to background or other sources than coal fired power plants or copper smelters.

The final four regressions were all done using SCD_4 or SCD_4 multiplied by RH. Based on the R^2 values, it can be seen that the sulfate sulfur is better explained by using $SCD_4 \times RH$ than by SCD_4 alone. It is also evident that the addition of the As term to the regression greatly improves the fit. However, as before, the multiplication of As by RH does not greatly improve the R^2 values. If the regressions without an As term are disregarded due to their poorer R^2's, then the sulfur attribution which results from all of the analyses using SCD_4 are quite similar. The sulfate sulfur for all days with CD_4 data was found to be on average 72 to 82 percent due to NGS, 18 to 28 percent from copper smelters, and 0.0 ± 1 percent from other sources. Interestingly, the differences between the mean sulfur attributed to all power plants and that attributed to NGS are not statistically different from each other.

TMBR best explained the measured sulfate sulfur when the independent variables used were $SCD_4 \times RH$, and $As \times RH$. The results for each time period are shown in Figure 4. Scatter plots of predicted *vs* measured sulfate sulfur are shown in Figure 5.

Conclusions

The TMBR formulation is a linearized version of a deterministic statement of how gases and particles are transformed as they are transported from source to receptor. The assumptions required for linearization have been stated, but the validity of these assumptions has not been explicitly tested. On the other hand, the least square models linking NGS and copper smelter emissions to sulfate concentrations at Grand Canyon had R^2 values between 0.72 and 0.81, which suggests model assumptions were not adversely violated.

Application of the TMBR model to the WHITEX data set yielded interesting insight into physio-chemical processes of transport and transformation of SO_2 between NGS and Grand Canyon, Arizona. Contrary to popular opinion, in–cloud or wet chemical processes appear to be responsible for SO_2–to–SO_4 oxidation at Grand Canyon during periods of high relative humidity. The concentration of sulfate and presumably the formation of sulfate is highly correlated with relative humidity.

Apportionment estimates using the TMBR model and Se as a tracer for coal fired power plants suggest that approximately 80 percent of sulfate found at Grand Canyon can be attributed to coal fired power plants, 10 percent to copper smelters and 10 percent to other sulfate sources. When the analysis is repeated with CD_4, the tracer released from NGS, the TMBR model suggests that NGS is responsible for between 70 and 80 percent of the sulfate while copper smelters contribute between 20 and 30 percent. Although there isn't a statistical difference between all coal fired power plant and NGS contribution to sulfate at Grand Canyon, the analysis suggests upper and lower limits. All coal fired power plants other than NGS may contribute between 0 and 10 percent, NGS may contribute between 70 and 80 percent, copper smelters between 10 and 30 percent and other sources between 0 and 10 percent of the sulfate found at Grand Canyon.

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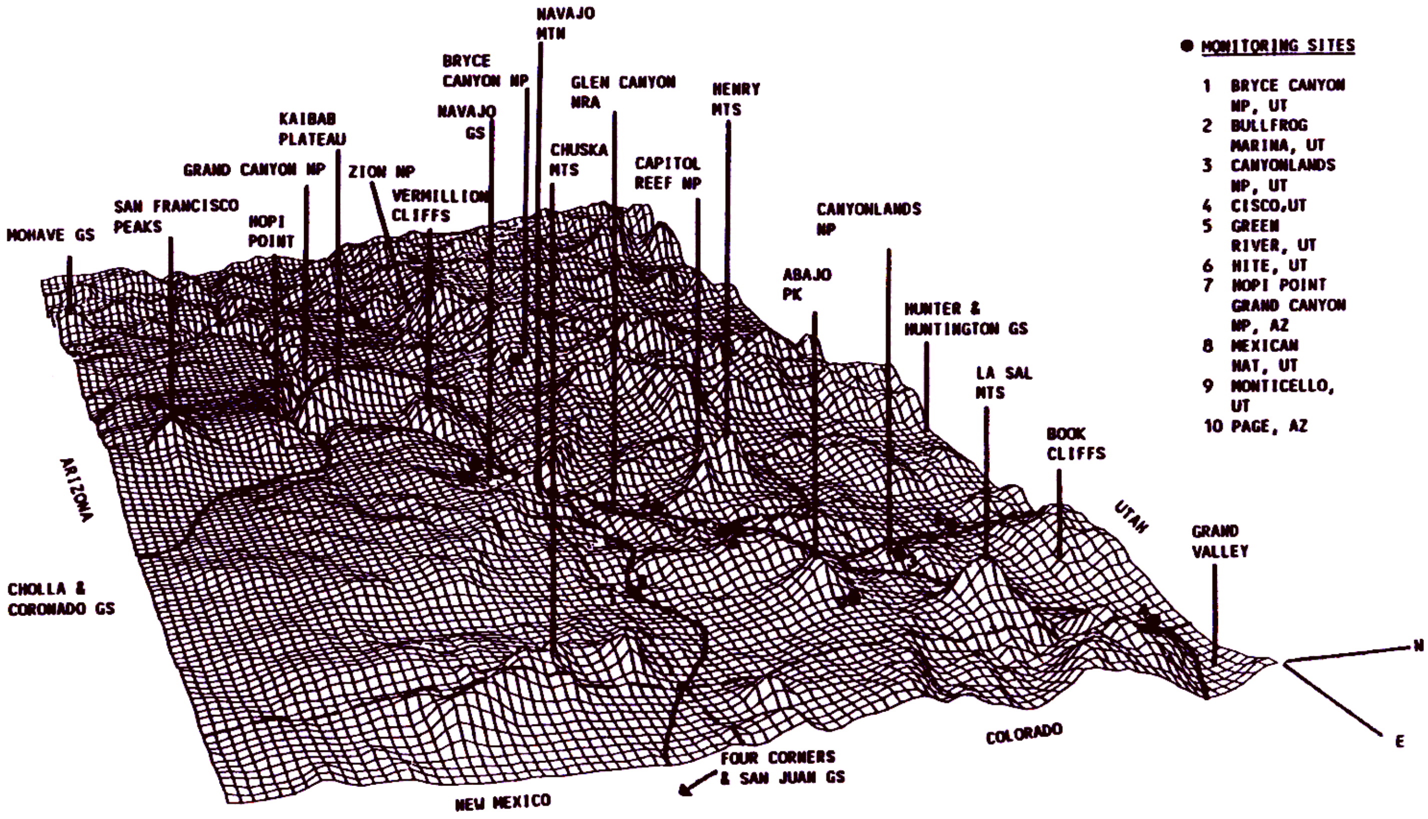

Figure 1. Three dimensional perspective of the WHITEX study region.

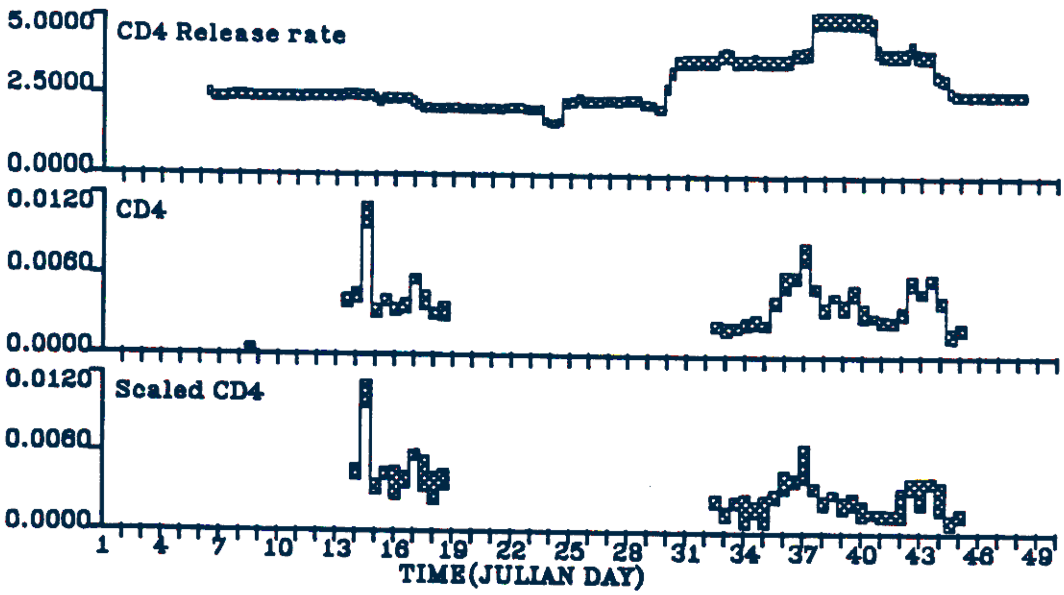

Figure 2. CD_4 release rate, unscaled CD_4, and CD_4 concentrations scaled to release rate and average air mass age for Hopi, Point, Grand Canyon. The hatched area represents one standard error above and below the mean.

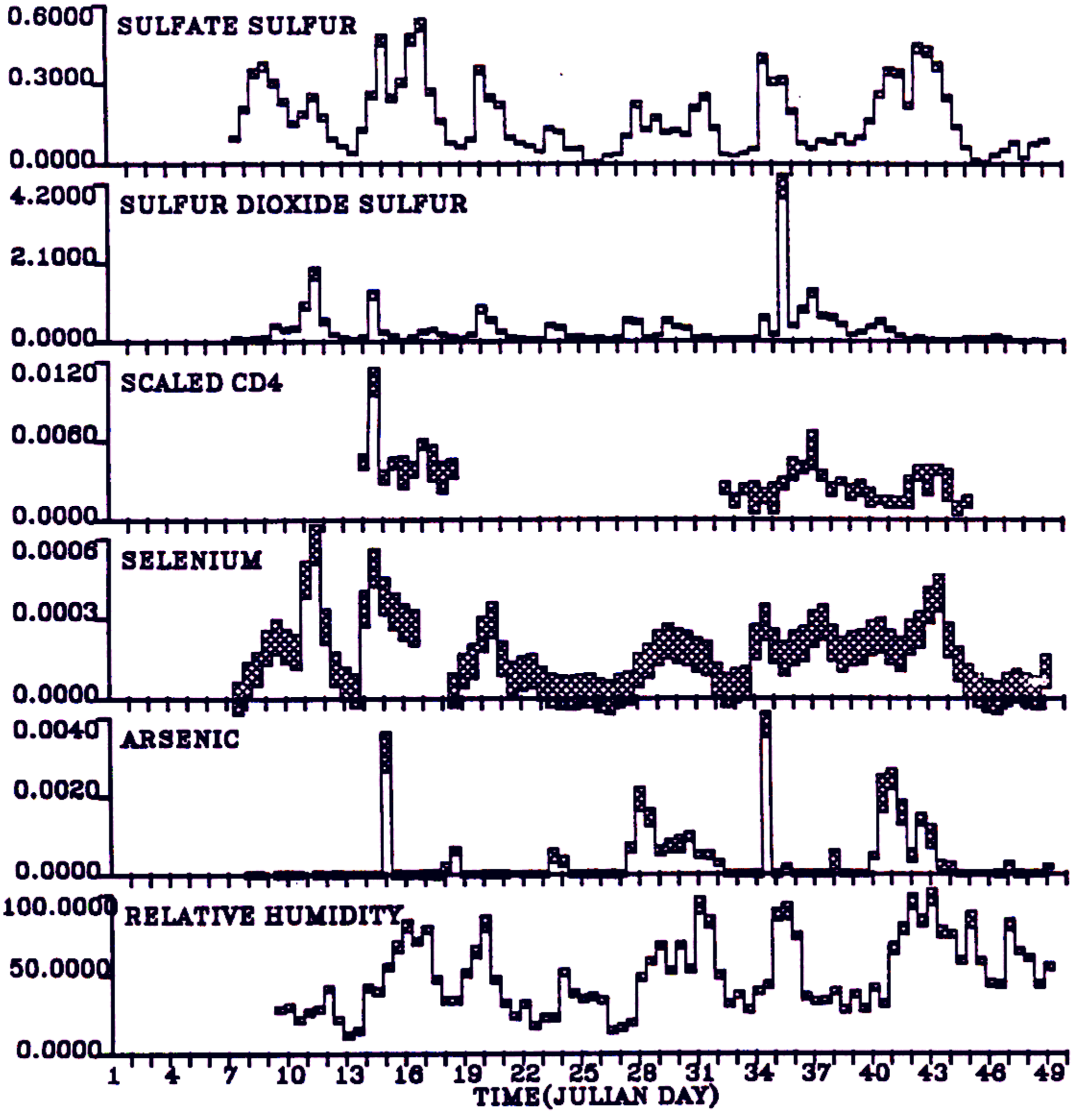

Figure 3. Temporal plot of sulfate sulfur, sulfur dioxide sulfur, fully scaled CD_4, selenium, arsenic, and relative humidity for Hopi Point, Grand Canyon. The hatched area represents one standard error above and below the mean.

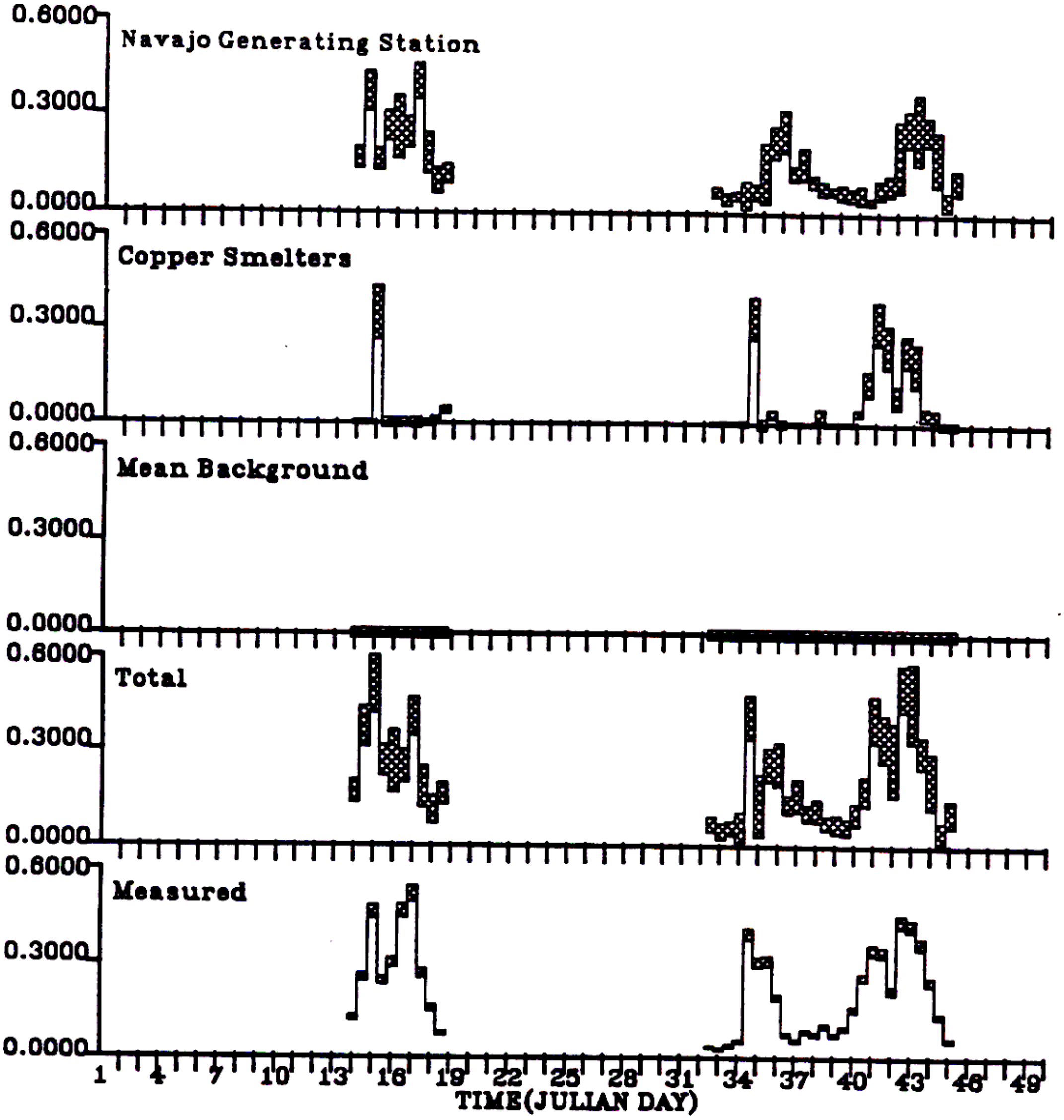

Figure 4. Temporal plot of TMBR source apportionment for sulfate sulfur ($\mu g/m^3$) at Hopi Point using ODR regression with independent variables $SCD_4 \times RH$ and $As \times RH$ for days with CD_4. The hatched area represents one standard error above and below the mean.

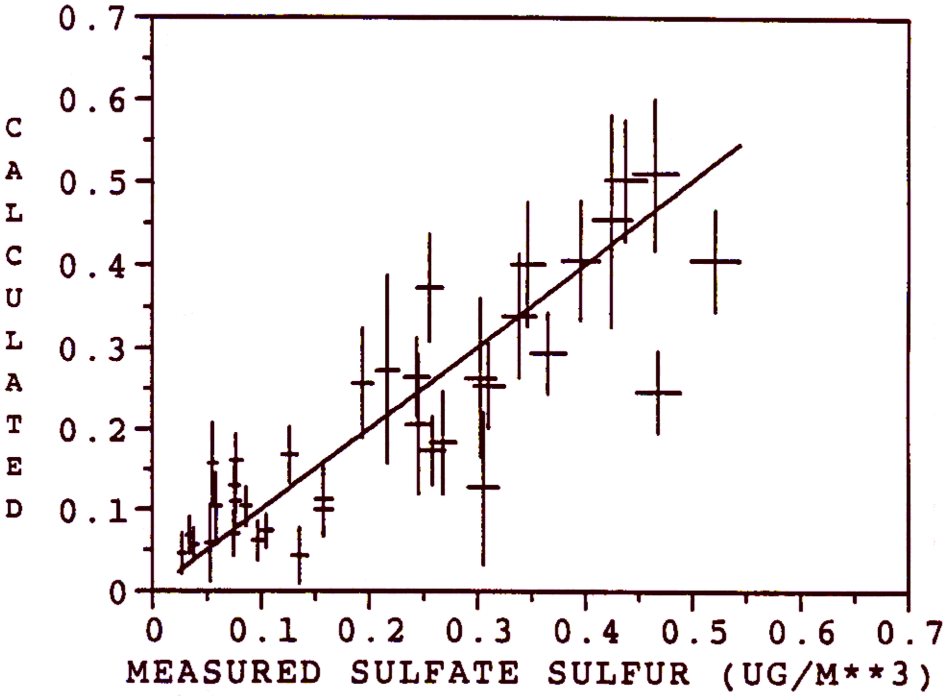

Figure 5. Scatter plot of predicted *vs* measured sulfate sulfur ($\mu g/m^3$) at Hopi Point. Results from ODR regression with independent variables $SCD_4 \times RH$ and $As \times RH$ for days with CD_4. Uncertainty estimates for the calculated results are one standard error on either side of the mean.

Table I. Results of TMBR regressions for total sulfur, nitrate, organic carbon, and elemental carbon at Hopi Point.

Dependent Variable	Regression Model	Days Used	No. of Obs.	R^2	Independent Variable	Regression Coefficient
total sulfur	ODR	All	36	0.06	SCD_4	171.38 ± 42.80
					Intercept	0.00 ± 0.13
total sulfur	ODR	All	35	0.36	SCD_4	167.08 ± 34.73
		ex 35.3 *			Intercept	0.00 ± 0.10
total sulfur	ODR	all	35	0.56	SCD_4	124.77 ± 21.13
		ex 35.3 *			As	180.30 ± 40.76
					Intercept	0.00 ± 0.07
nitrate	ODR	All	36	0.20	SCD_4	31.87 ± 12.08
					Intercept	0.00 ± 0.04
organic carbon	ODR	All	36	0.00	SCD_4	-2.54 ± 21.78
					Intercept	0.09 ± 0.08
light absorbing carbon	ODR	All	36	0.01	SCD_4	5.06 ± 11.77
					Intercept	0.17 ± 0.04

* Julian Day 35.3 was an extreme outlying data point (high SO_2 value).

Table II. Results of TMBR regressions for sulfate sulfur at Hopi Point.

Regression Model	Days Used	No. of Obs.	R^2	Independent Variable	Regression Coefficient	% of sulfur Attributed
ODR	All	76	0.72	$Se \times RH$	19.21 ± 1.97	83±4
				As	35.77 ± 14.90	7±1
				Intercept	0.02 ± 0.01	10±1
ODR	All	76	0.69	$Se \times RH$	20.78 ± 2.40	88±5
				$As \times RH$	0.32 ± 0.37	3±1
				Intercept	0.02 ± 0.02	9±1
ODR	All	33	0.81	$Se \times RH$	13.74 ± 1.38	82±5
	CD_4			As	70.93 ± 11.30	18±2
				Intercept	0.00 ± 0.01	0±1
ODR	All	33	0.80	$Se \times RH$	13.81 ± 1.64	80±5
	CD_4			$As \times RH$	1.39 ± 0.25	20±2
				Intercept	0.0 ± 0.01	0±1
ODR	All	36	0.04	SCD_4	69.71 ± 14.42	100±6
	CD_4			Intercept	0.00 ± 0.05	0±4
ODR	All	36	0.38	$SCD_4 \times RH$	1.20 ± 0.21	100±6
	CD_4			Intercept	0.00 ± 0.03	0±2
ODR	All	36	0.72	$SCD_4 \times RH$	0.99 ± 0.11	82±5
	CD_4			As	71.29 ± 13.96	18±2
				Intercept	0.00 ± 0.02	0±1
ODR	All	36	0.75	$SCD_4 \times RH$	0.90 ± 0.11	72± 4
*	CD_4			$As \times RH$	2.04 ± 0.35	28±2
				Intercept	0.00 ± 0.02	0±1

* Results of this regression were used for source apportionment.