Letters

Tracing Aerosol Pollution

In their article "Elemental tracers of distant regional pollution aerosols" (13 January 1984, p. 132), Kenneth A. Rahn and Douglas H. Lowenthal use statistical methods to apportion the sulfate component of the acid deposition problem to responsible regions of the United States. We address several problems with their analyses: (i) the aerosol data employed are inadequate for the proposed work; (ii) the use of selenium as the denominator in elemental ratios limits the usefulness of the regional aerosol discrimination approach; and (iii) the authors' final conclusions regarding the sources of U.S. sulfate pollution rest upon the interpretation of regression coefficients that cannot be reliably estimated in the manner proposed. We believe the techniques employed by Rahn and Lowenthal should be carefully examined in order to appreciate the limitations inherent in their analyses.

First, the aerosol data are total aerosol samples (that is, both fine and coarse particles together) and thus may include elemental mass that is unlikely to be transported from region to region. This is especially important for crustal elements, such as manganese. The "noncrustal" adjustment used does not solve this problem, as (for example) a major portion of noncrustal manganese may also occur in the coarse particle fraction (*1*). In addition, these samples were collected over many days, during which transported pollution may be contributed on one day and local elemental tracers on the next. The analysis of such mixed samples might easily give misleading results. To properly study the transport question, one must collect data more appropriate to the task (that is, shorter-term, size-specific aerosol samples).

Most important to the question of data validity, however, is that the proposed regional signatures are based on too few samples and were developed in a poorly documented and seemingly inconsistent manner. For example, tables 1 and 5 in the article contain an elemental signature for Boston derived from only three samples collected south of Providence, Rhode Island (roughly 100 kilometers from Boston). No objective criterion is reported for selecting these samples. There is no evidence presented to suggest that only three samples, no matter how carefully selected, can correctly characterize the Boston urban area's summertime aerosol. In sharp contrast, the Washington, D.C., profile is reportedly based on 2-month averages of a number of sites within (not downwind of) that city. Moreover, the authors' approach does not incorporate the probable effects of aerosol fractionation on such regional signatures during transport to another region. In general, the elemental samples employed were apparently of differing character, and no consistent, defensible, or objective methodology was presented for the development of the regional signatures.

With regard to the authors' analyses of elemental ratios, the division of all elements by selenium in order to develop regional discriminators is not appropriate to many situations of interest. Selenium is not (as the authors state) "found at similar concentrations in diverse source areas." On the contrary, the vast majority of anthropogenic selenium emissions in the United States are due to the combustion of coal (*2*), and selenium has been found in higher concentrations within areas of substantial coal combustion (for example, the Ohio River Valley) (*3*). It has been found that the ratio of two source-related elements can be very unstable in an air mass if there are intervening sources downwind. This problem has been indicated in the literature (*4*) for the manganese/vanadium technique previously proposed by Rahn (*5*). Although a ratio of, for example, vanadium/selenium or manganese/vanadium may look very different between regions or continents, these ratios are not necessarily conserved (in proportion to total pollution) when a polluted air mass passes over downwind tracer sources or is transported into another source region. Thus, elemental ratios may well correctly indicate where a sample was collected (as per the results in the authors' table 4), but not be able to reliably indicate the true source region(s) of a majority of the pollution in a polluted air mass transported to that site. When one considers these applicability limitations, it seems unwise to employ such ratios of source specific elements in most source apportionment analysis situations.

The least-squares analyses summarized in the authors' tables 6 and 7 constitute the most troublesome aspect of their work, raising serious questions about the validity of their aerosol apportionment results. We reran a number of the regressions of trace elements on signatures using the Statistical Analysis System (SAS) (*6*). Since the authors did not retain negative coefficients, we employed non-negative least squares. The regression coefficients were found to be very dependent upon the weightings employed (in direct conflict with the authors' contentions), and many were not statistically significant (that is, when P is greater than 0.05). A representative example is shown in Table 1 for the sample from 3 through 8 August 1979. The two models are identical, except that the first employs the authors' proposed weights, and the second employs weightings equal to the inverse of each element's pooled variance ($1/\sigma^2$), as derived from their table 5. The estimates of the coefficients change between models, most notably the New England coefficient shifts from 0.21 to 0.00. While the merits of each set of weightings might be argued, their importance to the coefficients derived is obvious.

The 95 percent confidence intervals reported by SAS for these coefficients are also included in Table 1. While negative coefficients are prohibited by such bounded regressions, these confidence

Table 1. Regional regression coefficients for data from 3 through 8 August 1979 with alternative weightings. Regressions were solved by the use of SAS with nonnegativity constraints on the estimated coefficients.

Region	Rahn and Lowenthal weightings		$1/\sigma^2$ weightings	
	Coefficient	95-percent confidence interval	Coefficient	95-percent confidence interval
New England	0.21	−1.10 to +1.53	0.00	−2.84 to +2.84
Boston	0.17	−0.01 to +0.35	0.18	−0.41 to +0.78
New York	0.06	−0.31 to +0.43	0.01	−0.62 to +0.64
Washington, D.C.	0.00	−0.44 to +0.44	0.08	−0.99 to +1.15
Interior	0.29	−0.15 to +0.74	0.24	−1.03 to +1.50

intervals clearly show that *none* of the coefficients is statistically different from zero, or from each other for that matter (indicating that comparisons between coefficients are not meaningful). This result is to be expected in view of the large errors in the coefficients arising from (i) the regional profiles' not being properly characterized; (ii) the fact that many of the profiles employed are highly intercorrelated with one another; and (iii) the fact that there are only two degrees of freedom in the proposed regression model. Together, these deficiencies cause the authors' interpretations of the coefficients in their table 7 to be speculative and unsubstantiated by the statistics presented. Furthermore, since the subsequent sulfate regressions employ these coefficients as inputs, the validity of the sulfate apportionments must also be questioned.

In summary, the authors ignore the complexities of the problem they are addressing and do not present the limitations of the techniques they employ. The idea of developing regional profiles is itself not new, but it has been recognized by the scientific community that data and statistical techniques tailored for this approach must be developed before one can move to the application state. The effort required to further develop proper aerosol characterizations and valid statistical techniques will likely be considerable, but the ultimate achievement of more reliable answers regarding the sources of sulfate aerosols is an objective worthy of research.

GEORGE D. THURSTON
Institute of Environmental Medicine, New York University Medical Center, Tuxedo 10987

NAN M. LAIRD
Department of Biostatistics, School of Public Health, Harvard University, Boston, Massachusetts 02115

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We thank Thurston and Laird for commenting on our regional elemental tracer technique. We agree that "data and statistical techniques tailored for this approach must be developed before one can move to the application state." In fact, we have generated an abundance of technical documentation and detailed application to the eastern United States, much of which will be reported shortly (*1–4*). Using the best available statistical techniques, such as those in the Environmental Protection Agency's source-apportionment program (*5*) and modifying them as needed, we have evaluated many aspects of our tracer system (*2–4*), including the points raised by Thurston and Laird. Our results support neither their basic conclusions nor the assertions and reasoning used to reach them, which are stated hypothetically and without supporting data or references to data. We respond briefly below to their points.

- We noted in our article that some of the mass of our tracer elements is supermicron. But Thurston and Laird offer no proof that this coarse mass is not transported between regions or that it affects elemental ratios during transport. We have data to the contrary: during 35 episodes of transport from the Midwest to northern Vermont, none of the elemental ratios to selenium changed (within the detection limit of 20 to 25 percent) beyond what could be explained by slight additions of northeastern aerosol (*2*). Fractionation during transport thus seems to be unimportant during at least the first 1000 kilometers. These data also contradict the assertion that size-specific samples must be used to study transport.
- The duration of receptor samples is not critical to evaluating long-term relations between distant and local sources, because the very nature of apportionment is to resolve mixed sources wherever their location. We have shown that 3-day apportionments are the same as 1-day apportionments over a season (*2, 4*). Detailed understanding of individual transport episodes may require samples even shorter than 1 day, however.
- The number of samples used to derive a signature is hardly an issue. We have shown that subsets of five samples chosen randomly from the sets of 12 and 48 samples comprising two of our newer signatures (*1*) do not differ statistically from the original signatures (*2, 4*). Our Boston-area signature was derived from three samples. Yet Thurston, who has studied hundreds of aerosol samples from metropolitan Boston (*6*), offers no evidence that this signature did not represent Boston adequately.
- The techniques used to derive regional signatures are still evolving and have not yet become standardized or objective. Our method of choice is still to use modal analysis of elemental ratios to identify the aerosol most characteristic of a region and verify its origin by meteorological analysis. Thurston and Laird offer no reasons to the contrary. We are, however, exploring alternative methods, such as cluster and factor analysis. To date, the results have been satisfactory only for clear-cut cases.
- The objection to using selenium as the denominator is groundless: selenium was chosen because its concentrations were similar regionally, especially as compared to those of more variable elements, such as vanadium. Reference elements cannot have *equal* concentrations in all source regions because every element varies from region to region. Also, ratios are used only to construct signatures; regional apportionments are based on individual elemental concentrations. The true problems, if any, in choosing a denominator stem from differences in statistical distributions between numerator and denominator. Distributions of tracer elements are likely to be more similar to one another than to the total mass that Thurston and Laird imply for a denominator.
- The "instability" implied by Thurston and Laird is merely a natural effect of aerosol of a different composition added during transport and is easily dealt with by least-squares apportionment.
- Thurston and Laird state that our signatures were colinear, but do not prove it. Using an objective technique available in the Statistical Analysis System (SAS), we have shown that our five best signatures for eastern North America (*1*) are not more than weakly colinear, and then only within the Northeast or Midwest (*2*). The earlier signatures were somewhat more colinear, but again mostly within regions rather than across them.
- The discussion by Thurston and Laird of the stability and meaning of our regression coefficients is flawed both technically and philosophically. They use two apportionments of one of our samples (their table 1) to show how sensitive regional coefficients are to weighting scheme. In our opinion, that table shows the opposite: the major sources (Boston and Interior) change by 20 percent or less; overall apportionments to Northeast or Midwest were hardly affected. Thus, while regional regression coefficients do depend on the weighting scheme, they do so within important limits. Our original statement about their insensitivity to weighting, which referred only to variations of subjective weights, remains correct even in a more general sense.

But the true dependence on weighting may be still smaller than their table 1 suggests. Although the proper way to estimate uncertainties in apportionments is currently being debated (*7*), it is generally accepted that correct weighting in

Table 1. Regional coefficients for Narragansett, Rhode Island aerosol of 3 through 8 August 1979.

Signature	Type of weighting		
	Subjective*	Sample variance†	Effective variance‡
New England	0.21 ± 0.30	0.16 ± 0.21	
Boston	0.17 ± 0.04	0.17 ± 0.05	0.24 ± 0.05
New York City	0.06 ± 0.09	0.07 ± 0.04	
Washington	0.00 ± 0.10	0.00 ± 0.08	
Interior	0.29 ± 0.10	0.28 ± 0.12	0.41 ± 0.09

*Weights according to our original article. Uncertainties directly from SAS output. †Uncertainties calculated according to (*8*), with variance of sample only. ‡Insignificant sources eliminated according to procedures of (*6*). Uncertainties calculated according to (*8*), with variances in both sample and signatures.

volves either the variance of the receptor sample or the variances of sample and signatures combined with the source strengths (*8*), but not the variances of the signatures alone. This latter weighting is without precedent in the literature. Thurston and Laird thus do not follow the state-of-the-art procedures outlined in a paper of which Thurston was a co-author (*7*). Our Table 1 shows three apportionments of the same sample: our original version based on subjective weightings, one with the proper inverse-variance weightings (from the receptor sample only), and one based on effective-variance weighting (*7, 8*). The stability and meaningfulness of the coefficients are evident in three ways: the results from inverse-variance weighting agree closely with those from subjective weighting, the sense of the results from the more rigorous effective-variance treatment agrees with both our other answers, and the 85 percent confidence intervals around the Boston and Interior coefficients from all three of our treatments exclude zero. We do not see how Thurston and Laird can make such strong assertions about lack of meaning on the basis of the 95 percent confidence level when the opposite answer is available at the 85 percent level. To us, it seems clear that the midwestern coefficient of this sample is nearly double the northeastern coefficient, no matter how they are calculated, and that both are known accurately.

The sweeping rejection by Thurston and Laird of apportionment data based on confidence intervals alone is a classic example of the dangers of depending upon a single statistical argument and its subjective interpretation at the expense of all other knowledge. For a complex system such as the atmosphere and its aerosol, this approach is particularly risky. To be sure, uncertainties of individual regional coefficients are large—we have estimated some to be as great as 30 percent even for the most significant coefficients. But this hardly justifies discarding the entire technique. Abundant data of other types are available with which to counter the conclusion of Thurston and Laird, and they emphasize the reliability of regional coefficients. For example, time-series of regional coefficients correlate tightly with large-scale meteorology (*1*), long-term regional apportionments of sulfate for the Northeast agree well with those derived from transport models and directional studies (*1*), and the validity of our midwestern coefficients has been verified in the 1983 Cross-Appalachian Tracer Experiment (*9*), during which every pulse of perfluorocarbon tracer gas released from Dayton, Ohio, and later sensed near our two sampling sites in New England was preceded by a pronounced maximum of midwestern aerosol (*2, 10*). When all the evidence is considered, it is clear that our tracer system is functioning properly and that regional coefficents are highly meaningful.

KENNETH A. RAHN
DOUGLAS H. LOWENTHAL
Center for Atmospheric Chemistry Studies, University of Rhode Island, Narragansett 02882-1197

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Smoking and Longevity

In response to the communication by J. E. Enstrom (Letters, 31 Aug., p. 878), we would like to make the following comments. We did not question that the survey by Enstrom and Godley (*1, 2*) was representative of the U.S. population. We did question its coding of lifetime nonsmokers. Enstrom asserts his confidence in his procedures and notes that our methods yielded higher smoker-nonsmoker longevity differences than Enstrom and Godley reported (*2*); however, both results are within appropriate limits for such studies.

We draw different conclusions than Enstrom does from the literature he cites. For example, the authors of the three-state Amish study (*3*) reported very similar nonaccidental death rates for Amish men and women age 40 and above, as have the authors of other studies of nonsmoking populations (*4*). These studies show neglible differences in life expectancy between nonsmoking men and women. The Alameda County data of Wingard (*5*) and of Enstrom (*6*) are not directly comparable with our data (*7*) for two reasons: (i) their data is for individuals age 30 to 69, and ours is for individuals age 30 to 105; (ii) classifications that merge continuous and ex-smokers can not be equated with classifications of continuous smokers only. Wingard (*5*) pointed out that her merged classification of continuous and ex-smokers created certain anomalies in the estimates of male-female mortality risk. Our conclusion from the evidence is that smoking explains at least half and perhaps 80 to 90 percent of the male-female mortality difference after age 30.

G. H. MILLER
Studies on Smoking, 125 High Street, Edinboro, Pennsylvania 16412
DEAN R. GERSTEIN
Committee on Basic Research in the Behavioral and Social Sciences, National Research Council, Washington, D.C. 20418

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Erratum: In the listing of recipients of the National Medal of Science (News and Comment, 8 Mar., p. 1183), the affiliation of Helmut E. Landsberg was incorrect. He is emeritus professor at the University of Maryland.