The Relationship Between In-Lake Sulfate Concentration and Estimates of Atmospheric Sulfur Deposition for Subregions of the Eastern Lake Survey*

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SUMMARY

Estimated amounts of the total sulfur deposition at each of the 1798 lakes in the U.S. Environmental Protection Agency's Eastern Lake Survey (Linthurst et al. 1986), obtained by using the ASTRAP model, were compared with the survey measurements of in-lake sulfate concentration on a subregional basis. In general, the sample median in-lake sulfate concentration was qualitatively correlated with the sample median estimated total sulfur deposition, with in-lake concentration increasing with increased estimated deposition. Two subregions, 3A (southern Blue Ridge) and 3B (Florida), however, did not fit this relationship. In-lake sulfate concentrations were higher than expected in Florida and lower than expected in the southern Blue Ridge.

Comparison of our modeled total sulfur deposition with estimated amounts of wet-only sulfate deposition determined by Sullivan et al. (1988) were in good agreement in terms of subregional rank order. More detailed comparison of the magnitudes of the estimates was not done because Sullivan et al. reported deposition in terms of the estimated population medians, obtained by using a weighting procedure based on alkalinity map class, instead of as sample medians. Sullivan et al. also used the weighting procedure to estimate the subregional median in-lake sulfate concentration. Although this weighting does not seem relevant to atmospheric inputs, the effect is small in most cases. The major consequence of applying the weighting is that the estimated population median sulfate concentration for subregion 3B (Florida) is much closer to the general trend between sulfate concentration and sulfur deposition than is the sample median sulfate concentration.

INTRODUCTION

Correlations between the concentration of sulfate in the surface waters of the United States and the estimated flux of sulfur from the atmosphere have been reported by Sullivan et al. (1988) and NAPAP (1989). These correlations were based on a subregional-scale analysis of data collected as part of the Eastern Lake Survey (Linthurst et al. 1986) in which the wet-only sulfur deposition at each lake was estimated from a spatially interpolated combination of rain chemistry data and long-term precipitation records. Because dry deposition of sulfur compounds is also expected to be a significant component of sulfur loading to lakes and watersheds, it is of interest to examine the relationship between lake sulfate concentrations and estimated amounts of the total (including both wet and dry) sulfur deposition. However, observations of total deposition appropriate for this purpose are not yet available. In this study we make use of the ASTRAP model to estimate the annual total sulfur deposition at each lake in the Eastern Lake Survey and compare the subregional distribution of these estimates to the subregional distribution of lake sulfate concentration.

EASTERN LAKE SURVEY SAMPLING

The Eastern Lake Survey (ELS) was conducted by the U.S. Environmental Protection Agency in the fall of 1984. The primary purpose of the survey was descriptive: it was intended to determine the percentage of lakes that are acidic or have low acid neutralizing capacity in potentially sensitive regions of the eastern U.S. (Linthurst et al. 1986). In order to make these determinations, three potentially sensitive regions were identified in the U.S. and a statistical protocol was established so that lakes to be sampled were selected by a systematic random process from the population of lakes in each region. Data from the sampled lakes could then be used to extrapolate to the larger population in each potentially sensitive region. In addition to stratification by region, the sampling design included two other levels of stratification. Each region was subdivided into subregions and each subregion was further subdivided by alkalinity map class. In this report, the term sample refers to data actually obtained in the field and the term population refers to estimates based on extrapolation of the sample data to the larger population.

ESTIMATES OF ATMOSPHERIC DEPOSITION

The ASTRAP model is well documented elsewhere (Shannon 1981; 1985) and the details of its workings need not be repeated here. In the present application, simulation of atmospheric transport and deposition was based on a twenty-four-year meteorological record (1960-1983), and the SO_x emissions for the United States and Canada were obtained from a 1980 inventory. The estimates of the total sulfur deposition obtained by using ASTRAP were added to our on-line version of the ELS database for further analysis. Deposition estimates were made for all of the 1798 lakes sampled, including those 186 "special" lakes which were sampled in addition to the "regular" lakes selected by the statistical protocol established for the survey.

While the meteorological and emissions records do not correspond precisely with the time of lake sampling, it is generally accepted that in-lake concentrations reflect watershed and atmospheric input over some poorly defined period which may range from months to years depending on the lake and watershed characteristics. Because they are based on a long meterological record, our estimates are intended to represent typical deposition. Previous investigations have taken a similar approach to estimate the wet-only loading by calculating deposition as the product of a several-year precipitation-weighted average rain sample sulfate concentration and a longer term (up to thirty year) average of precipitation amount. However, because the data available for these calculations varied from region to region, the wet-only deposition estimates for the Northeast are based on an entirely different source of data than are the estimates for the upper Midwest and Southeast (Sullivan et al. 1988). By using ASTRAP simulations we avoid this lack of uniformity between subregional deposition estimates.

RESULTS AND DISCUSSION

The median lake sulfate concentration for the 1798 sampled lakes is plotted against the median total sulfur deposition estimate for the 11 ELS subregions in Fig. 1. With the exception of two subregions (3A - the southern Blue Ridge and 3B - Florida), the lake sulfate concentrations seem to be directly related to the estimated total sulfur deposition. These two regions have previously been identified as anomalies (Sullivan et al. 1988) though not, in the case of Florida, to the extent shown here. As would be expected, subregions generally are grouped together. Within regions, the gradient of sulfur deposition also is reflected in the lake sulfate concentrations. Although one subregion (2A - northeastern Minnesota) shows a higher sulfate concentration than might be expected, this subregion encompasses the Superior Natural Forest, an area known to have geologic sources of sulfur (Nichols and McRoberts 1986).

Because the Eastern Lake Survey was based on a stratified probability sampling design, users of the database are cautioned

> "...against estimating population parameters or examining relationships among variables with the expectation that these relationships are representative of the population, from sample data without accounting for ...(the appropriate stratum weights)." (Linthurst et al., 1986)

The stratum weights are factors that are to be used when making estimates for the population of lakes by combining data collected from different strata. It should be noted that, within subregions, the strata are based on alkalinity map class.

Alkalinity map classes were subjectively determined on the basis of data from a variety of sources predating the Eastern Lake Survey. Post-analysis of the Eastern Lake Survey data indicates notable discrepancies between the alkalinity of individual lakes and their alkalinity map class, particularly in Florida.

Sullivan et al. (1988) used the weighted combination of data collected in each subregional alkalinity class in their study which showed an almost linear relationship between median lake sulfate concentration and median estimated wet sulfate deposition. Although it may be argued that this weighting is required by the sampling protocol, the weighting is not necessary when the relationships of interest are independent of alkalinity map class (Linthurst et al. 1986). Clearly, this is the case for the flux of sulfur from the atmosphere because there is no reason to expect that the sulfur deposition for a particular lake will depend on the alkalinity map class for that lake. In other words, although it would be necessary to use the alkalinity class weighting to estimate the population median lake sulfate concentration alone, it may be misleading to use the same type of weighting to estimate the population median sulfur deposition. Furthermore, unless there is evidence that the alkalinity class weighting results in an estimate of the representative sulfur deposition for the target population, the weighting should not be used when it is of interest to examine the relationship between lake sulfate concentration and atmospheric sulfur deposition.

In order to avoid this potential problem, our analysis is restricted to the relationship between the sulfate concentration of the sampled lakes and our estimates of the total sulfur deposition for those lakes. Because, as pointed out above, deposition is independent of a lake's location within an alkalinity map class, it would be impossible to determine a representative deposition flux estimate for the target population of lakes without having detailed information about the location of those lakes. Our estimates, however, do provide an estimate of the representative sulfur deposition for the sampled lakes.

In general, the effect of the weighting on the estimated median value of lake sulfate concentration is small. The sample median values for both sulfate concentration and total sulfur deposition flux are listed along with the comparable population estimates in Table 1. This table and Figure 2, a scatter plot of the estimated subregional population median sulfate concentration against the subregional sample median sulfate concentration, show that except for subregion 3B (Florida), the estimated population medians are very close to the sample medians. The population median sulfate for Florida is dominated by the contribution from those lakes in alkalinity class 3 (> 200 μeq/L) and is much lower than the sample median. Although the alkalinity class 2 lakes in this subregion have an extremely high median sulfate concentration (391.4 μeq/L), the weighting for this stratum is so low that their effect is minimal when the combined subregional population median is determined.

Our estimates of the subregional sample median total sulfur deposition are fairly well correlated with the estimated population median wet-only sulfur flux (Table 1, Figure 3). The relative magnitudes of the wet-only and total deposition estimates, illustrated by rank ordering, is very similar for the eleven subregions. Interestingly, although we would expect that our estimates would be higher than the wet-only estimates by about a factor of two, this is apparently not true in the upper Midwest, where the total and wet-only estimates are about equal. Our deposition estimates for the upper Midwest agree very well with an independent set of estimates of total sulfur deposition in 1983 presented by Neary and Dillon (1988). Determination of the causes of the discrepancies between our results and those given by Sullivan et al. (1988) are beyond the scope of this report.

CONCLUSIONS

Modeled estimates of the deposition of total sulfur (wet and dry) at lakes sampled as part of the Eastern Lake Survey show a similar relationship to measurements of lake sulfate concentration as do estimates of wet-only sulfate deposition flux. With the exception of two subregions (3A - the southern Blue Ridge and 3B - Florida), lake sulfate concentration increases with increasing deposition. The sample median sulfate concentration of lakes in Florida is dominated by the contribution from lakes in alkalinity class 2, which is much higher than the median concentration in either of the two other alkalinity classes. Expressing the results in terms of estimated population medians by weighting the contribution from different strata considerably reduces the effect of lakes in alkalinity class 2.

Comparison of the magnitude of the modeled deposition of total sulfur with previously published estimates of the wet-only deposition are in general agreement, with the total deposition estimates being at least a factor of two higher in most subregions. In the four subregions of the upper Midwest, however, the estimates are very similar in magnitude, suggesting problems in one or both methods of estimation.

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Table 1. Sample medians and estimated population medians of in-lake sulfate concentration and estimated sulfur deposition for Eastern Lake Survey subregions. N = the number of lakes sampled in each subregion. Population medians were taken from Linthurst et al. (1986) and wet sulfur deposition from Sullivan et al. (1988). Deposition estimate ranking among subregions (from highest to lowest) is shown in parentheses.

		In-Lake Sulfate Concentration		Sulfur Deposition	
Subregion	N	Sample Median (μeq/L)	Population Median (μeq/L)	Sample Median[a] (g/m^2/yr)	Population Median[b] (g/m^2/yr)
1A	203	120.5	118.7	1.87 (3)	0.77 (4)
1B	156	146.2	159.3	2.56 (1)	0.95 (1)
1C	213	97.0	101.2	1.64 (5)	0.54 (5)
1D	127	132.5	141.1	1.87 (2)	0.63 (2)
1E	184	73.0	74.6	1.11 (6)	0.53 (8)
2A	159	62.7	62.5	0.35 (11)	0.34 (11)
2B	156	69.6	77.7	0.71 (8)	0.50 (9)
2C	187	60.2	56.9	0.69 (9)	0.64 (6)
2D	142	47.0	50.1	0.48 (10)	0.48 (10)
3A	112	29.6	31.8	1.78 (4)	0.79 (3)
3B	159	145.2	93.7	0.82 (7)	0.58 (7)

[a] Total sulfur (wet and dry) deposition estimated by using ASTRAP.

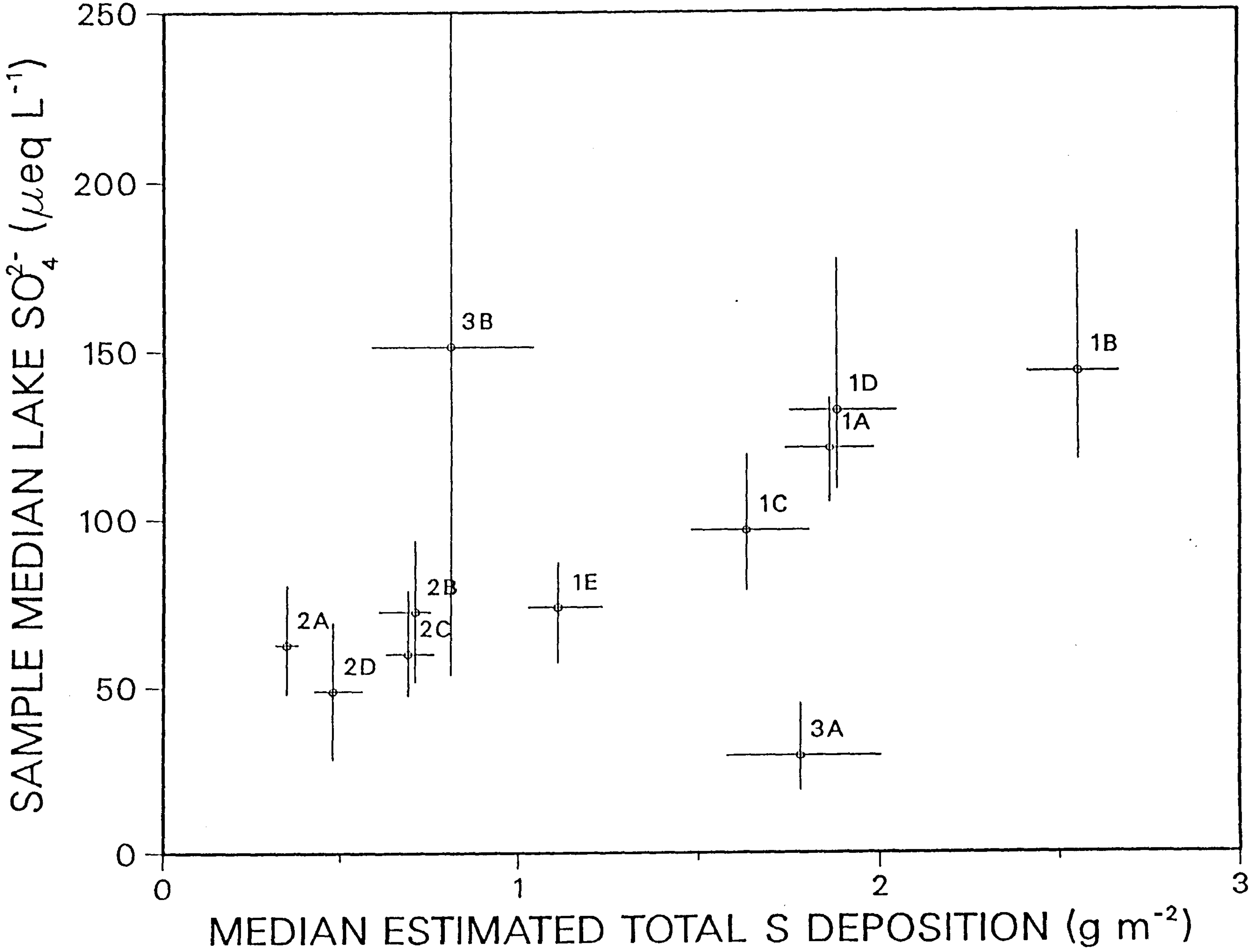
[b] Sulfur (wet only) deposition based on observations reported in Sullivan et al. (1988).

FIGURE CAPTIONS

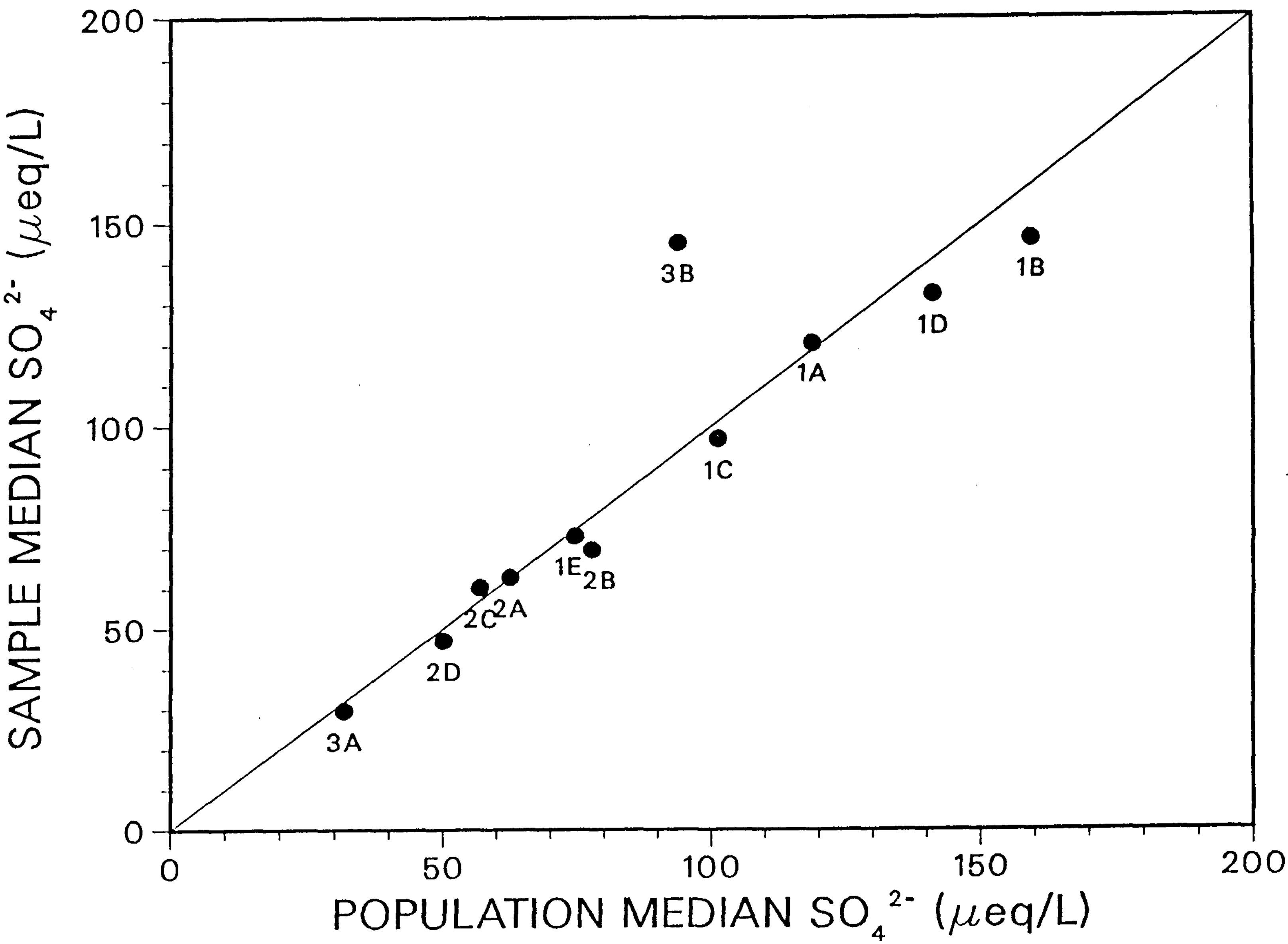
Figure 1. Sample median in-lake sulfate concentration versus sample median total sulfur deposition for subregions of the Eastern Lake Survey. Error bars show the 25th and 75th percentiles of the subregional distributions. Total sulfur deposition estimates were calculated by using ASTRAP.

Figure 2. Comparison of sample median in-lake sulfate concentrations and estimated population in-lake sulfate concentrations (Linthurst et al., 1986) for subregions of the Eastern Lake Survey.

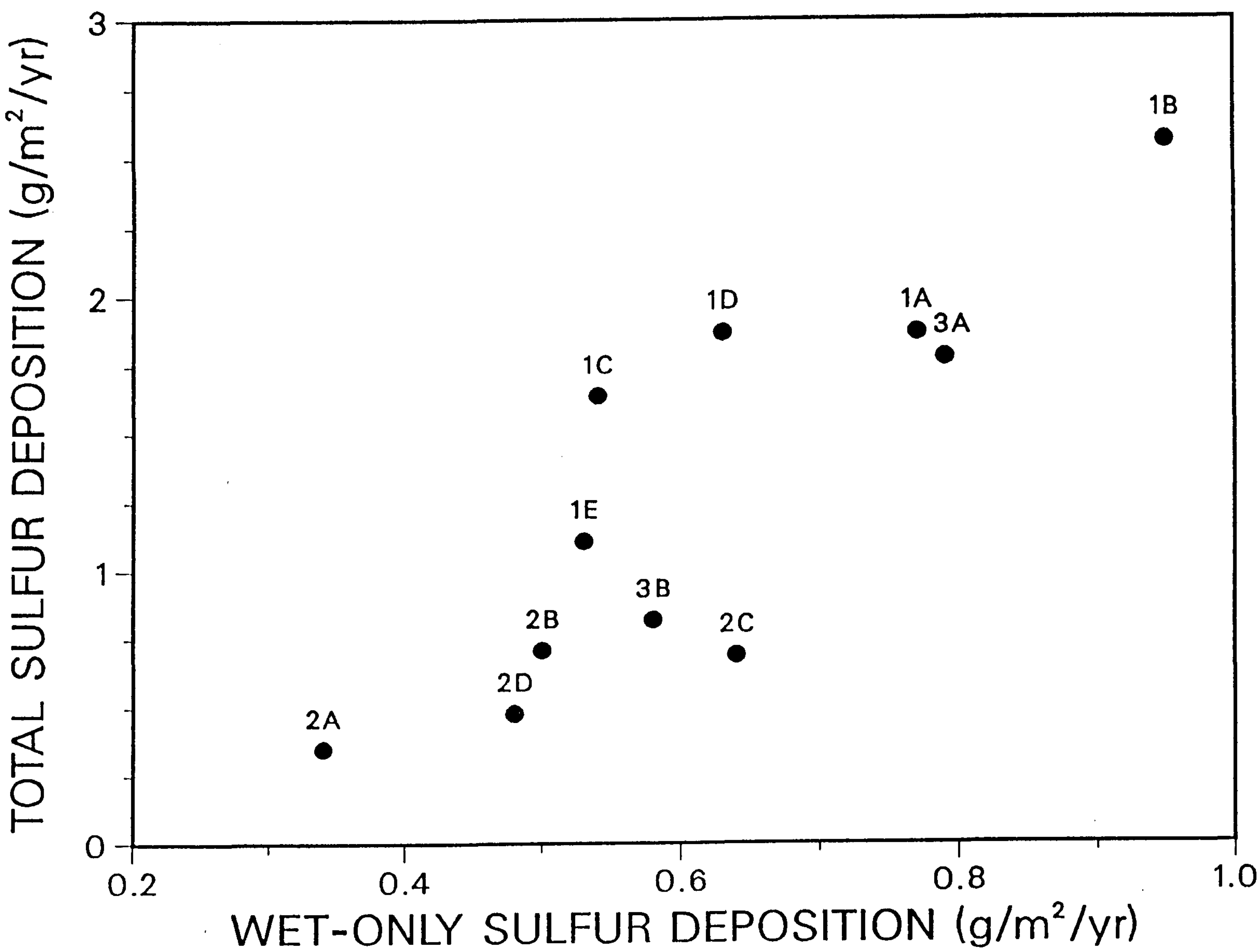
Figure 3. Comparison of sample median estimated total sulfur deposition and estimated population wet-only sulfur deposition (Sullivan et al., 1988) for subregions of the Eastern Lake Survey.



SAMPLE MEDIAN LAKE SO_4^{2-} (μeq L^{-1})
250
200
150
100
50
0
0
1
2
3
MEDIAN ESTIMATED TOTAL S DEPOSITION (g m^{-2})
3B
1B
1D
1A
1C
1E
2B
2A
2C
2D
3A



SAMPLE MEDIAN SO_4^{2-} (μeq/L)
POPULATION MEDIAN SO_4^{2-} (μeq/L)
0
50
100
150
200
3A
2D
2C
2A
1E
2B
1C
1A
3B
1D
1B


TOTAL SULFUR DEPOSITION (g/m²/yr)
WET-ONLY SULFUR DEPOSITION (g/m²/yr)
0
1
2
3
0.2
0.4
0.6
0.8
1.0
1B
1D
1A
3A
1C
1E
3B
2B
2C
2D
2A