Highlights

- Transference ratio is the ratio of total deposition to the airborne concentration.
- Fifteen years of weekly monitoring data were used to examine transference ratios.
- Transference ratio for oxidized sulfur depends more on site and season than year.
- Findings are generally consistent for monitored oxidized sulfur and nitrogen.
- Annual estimates of total deposition to within ±35% may be expected in the East.

"Transference Ratios" to Predict Total Oxidized Sulfur and Nitrogen Deposition – Part I, Monitoring Results

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Abstract.

The "transference ratio" has been defined as the ratio of total (i.e., dry plus wet) deposition to the airborne concentration of a species of interest and introduced as a metric to help estimate total deposition, given airborne concentration (US EPA, 2011). Use of model-predicted transference ratios is currently under consideration by the US EPA in the formulation of a Secondary National Ambient Air Quality Standard for oxidized nitrogen and oxidized sulfur. Since little is known about transference ratios, their behavior is examined in the current study using monitoring data. This term is defined for oxidized sulfur (T_S) as the ratio of the total oxidized sulfur deposition (from dry plus wet deposition) to the airborne concentration of oxidized sulfur. A 15-year record of weekly measured and data-derived quantities at 12 monitoring sites in the eastern US was examined to determine the variability of T_S that might be expected from field measurements. Weekly T_S displayed considerable variability that depended on site, season, and year, but ANOVA results suggest most heavily on site and season. Using weekly data, the variability of deposition-related quantities and error propagation analysis of T_S both suggest that variation in parameters related to wet deposition processes are generally more important than variation in parameters related to dry deposition processes in determining variation in T_S. Correlations between airborne seasonal concentration of oxidized sulfur and the various components of seasonal deposition (i.e., dry, wet, and total) also underscore the strong influence that the variability in wet deposition processes can have on estimates of total deposition. Analysis of monitoring results suggests that 95% confidence intervals for T_S using weekly results over several years for a specific site and season could be as large as ±235%, but only as large as ±33% using annual aggregates for a specific site. At the annual scale, a regression model of the form that incorporates a site-specific transference ratio, yielded estimates of total oxidized sulfur deposition to within ±25% of the monitored values ≥ 95% of the time. Since all of the major oxidized nitrogen species are not monitored regularly, a parallel analysis was limited to only the monitored oxidized nitrogen species (OxN). Nevertheless, findings for OxN are consistent with those described above for oxidized sulfur. These results suggest that at specific sites in the eastern US, annual estimates of total deposition to within ±25 to ±35% may

be expected using species- and site-specific transference ratios along with annual average monitored airborne concentrations.

Keywords: Transference ratio; oxidized sulfur; oxidized nitrogen; total deposition; CASTNET; NADP

1. Introduction

The "transference ratio" concept was introduced by the US EPA (2011) and was defined as the ratio of total (i.e., dry plus wet) deposition to the airborne concentration of a species of interest. This expresses the notion that the amount of a species deposited to the earth's surface within a given area over a given period of time is proportional to the average airborne concentration of the species over the same area during the same time period, where the proportionality constant is called a transference ratio.

This transference ratio concept has not been well documented in terms of spatial and temporal behavior, variability and associated uncertainty. Other researchers (Barrie, 1991; Galloway et al., 1993; Wojcik and Chang, 1997; Hicks, 2005) have employed a ratio concept in attempts to relate the airborne concentration of a species to its corresponding concentration in precipitation and have found considerable variability in their resulting scavenging and washout ratios. Successful use of the transference ratio concept assumes that conditions prevail to ensure that transference ratios of selected acidic chemical species of interest are sufficiently stationary with respect to space, time, and changing environmental conditions, to permit their use in the estimation of total deposition.

The US Environmental Protection Agency (EPA) is considering the use of transference ratios in the formulation of a Secondary National Ambient Air Quality Standard (NAAQS) for oxides of nitrogen and oxides of sulfur as a means to estimate their total atmospheric deposition (US EPA, 2011). In the development of the Secondary NAAQS, combinations of maximum one-year acceptable (i.e., critical) total acidic deposition loads may be determined for geographical areas of interest. Transference ratios of acidic species of interest (i.e., oxidized nitrogen and oxidized sulfur) may be predicted using air quality models. These model-estimated transference ratios may then be used with monitored atmospheric concentrations to estimate total acidic deposition loads that can be compared with one-year critical total acidic deposition loads to determine compliance of the monitored ambient acidic airborne concentrations.

In view of the possibility that transference ratios determined from air quality model estimates may be employed in a future standard, the goal of the current study is to use actual monitoring data to examine and document the behavior, variability and associated uncertainty of

transference ratios. The goal of a companion article is to use transference ratio estimates from an air quality model to conduct a similar examination of modeled transference ratios and compare modeled and monitored results (Sickles et al., submitted 2012). The objective of the current study is to investigate the transference ratio concept and selected supporting assumptions by examining total oxidized sulfur deposition and airborne concentrations of oxidized sulfur (and to a limited extent, analogous deposition and airborne concentrations of monitored oxidized nitrogen) using monitoring data collected at specific locations (sites) over specific time periods. Since monitoring data are available from the Clean Air Status and Trends Network (CASTNET for dry deposition; http://epa.gov/castnet/javaweb/index.html) and the National Acid Deposition Program (NADP for wet deposition; http://nadp.sws.uiuc.edu/) for all the major oxides of sulfur, the current study focuses primarily on these chemical species (i.e., airborne gaseous sulfur dioxide (SO_2), airborne particulate sulfate (SO_4^{2-}), and sulfate ion (SO_4^{2-}) in precipitation). Oxidized nitrogen is released into the air primarily from anthropogenic sources as nitric oxide (NO) and nitrogen dioxide (NO_2) and together, they are commonly known as NO_X. In the atmosphere these species subsequently undergo chemical transformation to form additional oxidized nitrogen species (i.e., nitric acid (HNO_3), peroxyacetyl nitrate (PAN), nitrous acid (HONO), other organic nitrates, and airborne particulate nitrate (NO_3^-)). Since monitoring data for the total of all major oxidized nitrogen species (commonly known as total reactive oxidized nitrogen, NO_Y) are not currently available, a parallel analysis of NO_Y could not be performed. Nevertheless, using the available monitoring data, analogous analyses of airborne concentration and total deposition were performed for the monitored oxidized nitrogen species, OxN (i.e., airborne monitored oxidized nitrogen (OxN_g) includes gaseous HNO_3 and airborne particulate NO_3^-, and wet monitored oxidized nitrogen (OxN_w) is nitrate ion (NO_3^-) in precipitation).

The following two issues are explored primarily for oxidized sulfur, while only limited results for OxN are presented. First, monitoring data are used to investigate the sources of variability of the transference ratio for oxidized sulfur (T_S). This includes examining: the typical variability of T_S observed in the field using data aggregated at different temporal scales; the relative importance of variables contributing to variation in calculated values of T_S; and spatial and temporal behavior of T_S (see Sections 3.1 to 3.3). Secondly, the end point of applying the concept of transference ratio toward the creation of an air quality standard will likely involve its use in the estimation of total deposition. As a result, the expression using transference ratio and airborne concentration to predict total deposition is cast in the form of a statistical model and evaluated using monitoring data. The resulting uncertainties associated with estimating total oxidized sulfur deposition, D_{Stotal} (and to a limited extent, total OxN deposition, $D_{OxNtotal}$) using the airborne concentration of oxidized sulfur, C_{Sg} (and the corresponding airborne concentration of OxN, C_{OxNg}) are explored in Section 3.4.

2. Approach

Two earlier studies of dry, wet, and total deposition in the eastern US over the 15-years between 1990 and 2004 have been published based on weekly measurements from CASTNET and NADP monitoring sites (Sickles and Shadwick, 2007a&b). The current study initially considered the 22 sites where CASTNET and NADP measurements were collocated for the period 1990-2004 (Sickles and Shadwick, 2007b). The data employed are means of weekly, seasonal (aggregated from weekly means), and annual (aggregated from seasonal means) monitoring results collected at collocated CASTNET/NADP sites where data capture is adequate. Sites, land use, terrain features, elevation, latitude, longitude, and mapped location are shown in Sickles and Shadwick (2007b) and at the CASTNET and NADP web sites. Site location by state may be identified by the first two places of the NADP site designation (e.g., see Table 1). Data capture is acceptable for weekly and seasonal analysis at only the 12 sites identified in Table 1. Six of the 12 sites are in locations that may be representative of ecosystems that are especially vulnerable to acid deposition from oxidized sulfur and oxidized nitrogen, five of these are located in complex (or montane) terrain, and five are at high elevation (see Table 1). Data capture is sufficient (i.e., full years of seasonal data for ≥ 5 years) to permit annual analysis at nine sites for oxidized sulfur and eight sites for OxN (see Tables 2 and 3). Although five of the six acid-vulnerable weekly and seasonal sites and all four of the acid-vulnerable annual sites are located at elevations above 500m, and cloud deposition may be an important contributor to total acidic deposition to vulnerable ecosystems located at high elevations (Sickles and Grimm, 2003); the contribution of cloud deposition to the total is not considered in the current study because it is neither monitored nor modeled extensively on a routine basis. Data described above for the period, 1990-2004, were used in the bulk of the current manuscript; however, corresponding more recent data for the period, 2005-2009, at the same sites were used to a limited extent in the assessment of a statistical model chosen in the current study to predict total deposition from airborne concentration (see Section 3.4).

Measured and data-derived quantities considered in the current study are defined in the Notation Section for oxidized sulfur, and analogous quantities (not shown) are used for OxN. Statistical analyses of selected measured and data-derived quantities have been performed to examine their behavior from different perspectives. A source of variation that is not considered in the statistical summaries is variation due to temporal trend in quantities of interest. Kendall's τ (two-sided tests) (Hollander and Wolfe, 1999) and the conservative Bonferroni approach to simultaneous statistical inference (Miller, 1981) are used to compute significance levels for temporal trends in the seasonal quantities (see Section 3.2). In the current context, this is interpreted as the level of significance for monotone trend, and no specific form of trend (e.g., linear trend) has been assumed.

Three suspected sources of variation contributing to overall variability in T_S are sites, seasons, and years (i.e., location, time of year, and specific year). One method of comparing the

contributions due to these three sources of variation is to partition the variation in an analysis of variance (ANOVA) setting using PC SAS (http://www.SAS.com/). Although the current data do not formally constitute results from a designed experiment, the ANOVA should indicate the relative importance of the sources of variation to the overall week-to-week variation in T_S. Week-to-week variation may also be present in T_S for fixed sites, seasons, and years, but this is treated as 'random noise' in the ANOVA.

The parameter T_S has been computed from several measured and data-derived quantities (see the final expression for T_S in the Notation Section), and variation in these quantities contributes to variation in T_S in a non-linear manner. A propagation of error estimate (Meyer, 1975) has been performed to examine how variation in individual quantities contributes to overall variation in weekly T_S. The propagation of error estimate does not include contributions from mean, covariance, and higher order terms.

The establishment of a secondary NAAQS for oxidized sulfur will likely involve the prediction of D_{Stotal} using C_{Sg}. It has been suggested that this prediction may use the transference ratio for oxidized sulfur in a model equation of the form of $D_{Stotal} = T_S \cdot C_{Sg}$. The variables, site location and season, in addition to airborne concentration, are expected to be major contributors to variability in T_S. In the current study the above expression is cast as a statistical (linear regression) model to predict D_{Stotal} from monitoring site location, season, and C_{Sg}. The general form of statistical model used in the current study for prediction is shown in Equation (1a), where i denotes the monitoring site location, j the season, and k is the year (replication).

$$(D_{Stotal})_{ijk} = \tau_{ij}(C_{Sg})_{ijk} + \varepsilon_{ijk} \qquad (1a)$$

Equation (1a) describes a separate slopes no-intercept weighted regression model with weights $(C_{Sg})^2$ and a slope estimator (τ_{ij}) of the form of T_S, as previously defined. In this case, the slope estimator is the mean of (D_{Stotal} / C_{Sg}). The distribution of ε_{ijk} is normal with mean 0 and variance proportional to $(C_{Sg})^2$.

Since total deposition is the sum of dry and wet deposition, two other models of similar form can be used to predict the two parts that sum to D_{Stotal} (i.e., $D_{Sg} + D_{Sw} = D_{Stotal}$). Here, a model for D_{Sg} may be described in Equation (1b), and a model for D_{Sw} may be described in Equation (1c).

$$(D_{Sg})_{ijk} = \tau_{ij}(C_{Sg})_{ijk} + \varepsilon_{ijk} \qquad (1b)$$

$$(D_{Sw})_{ijk} = \tau_{ij}(C_{Sg})_{ijk} + \varepsilon_{ijk} \qquad (1c)$$

Two approaches for the assessment of Equation (1a) to predict D_{Stotal} from C_{Sg} were used (and also to predict $D_{OxNtotal}$ from C_{OxNg}). In the first approach, an arbitrary nominal confidence level of 99% was chosen for this evaluation, and 99% Prediction Intervals (PI) were calculated for the model for each monitored (or aggregated) concentration according to Draper and Smith (1998). Individual predictions of total deposition were scored for Equation (1a) as falling inside or outside of the resulting calculated PIs and summed to establish a relative frequency. The percentage of predictions of total deposition falling inside the PIs should be close to the nominal confidence level for the model to be considered appropriate. Large departures from the nominal confidence level indicate deficiencies in the model that produced the predictions.

The second approach considered the aggregate behavior of all predictions from the selected model. Here, individual predictions of total deposition defined by Equation (1a) for oxidized sulfur and by its analogue for OxN are expressed relative to the monitored total deposition for oxidized sulfur and OxN at both seasonal and annual temporal resolution. These relative differences (RDs) are defined in Equation (2) and are used to evaluate the probability statement shown in Equation (3) for paired values of predicted D_{Stotal} and monitored D_{Stotal} derived from monitoring data (and similarly for paired values of predicted $D_{OxNtotal}$ and monitored $D_{OxNtotal}$). Equation (3) expresses an a priori statement of acceptable agreement for use in further investigations. The stated probability should be high to ensure confident use of the model. In the current study, an arbitrary RD of ±25% at a probability of ≥ 95% was chosen to define acceptable model performance. For model evaluation, ensembles of RDs were determined and scored as falling inside or outside of ±25% and summed to establish a relative frequency. The relative frequency estimates the probability of meeting the ±25% criterion and should be ≥ 95% for the model to have performed acceptably in the current analysis.

$$RD = 100\% \cdot (\text{predicted } D_{Stotal} - \text{monitored } D_{Stotal}) / \text{monitored } D_{Stotal} \quad (2)$$

$$P\{|RD_{Stotal}| \leq 25\%\} \geq 95\% \quad (3)$$

Each approach of model assessment has two forms, which are subsequently examined, a Level 1 (or weaker) form and a Level 2 (or stronger) form. To provide Level 1 assessment, monitoring data collected between 1990 and 2004 were used to derive site- and (where appropriate) season-specific parameter estimates (i.e., τ_{ij}) for the model described in Equation (1a). To provide more rigorous tests of the model's performance, Level 2 evaluation employed parameter estimates derived from the Level 1 data set. Here, model-estimated deposition was compared with monitored deposition derived from a second, independent set of monitoring data collected at the same sites between 2005 and 2009.

3. Results and Discussion

Analyses of study-wide weekly values, seasonal aggregates of weekly values, and annual aggregates of seasonal values of the following measured and data-derived quantities have been performed by site for oxidized sulfur: $V_{d\ SO2}$; $V_{d\ SO4}$; V_{Sg}; V_{Sw}; PPT; and T_S. For OxN, parallel analyses of weekly values were not performed, and its examination was limited to seasonal aggregates of weekly values and annual aggregates of seasonal values of the following measured and data-derived quantities by site: $V_{d\ HNO3}$; $V_{d\ NO3}$; V_{OxNg}; V_{OxNw}; and T_{OxN} (defined analogously to those for the oxidized sulfur species, above).

For oxidized sulfur, among several summary statistics computed for weekly results, two were selected for display in Table 1 at each of the 12 study sites. These are the seasonal median values of the indicated weekly quantities and their corresponding coefficients of variation (CV = 100 · standard deviation / mean) within seasons, across the 15-years between 1990 and 2004. However, due to space limitations, results for only the summer and winter seasons are displayed in Tables 1 to 4. Corresponding summary statistics for seasonal and annual aggregates of oxidized sulfur are shown in Table 2 at the respective study sites with acceptable data capture. Analogous seasonal and annual summary statistics for OxN are shown in Table 3. In addition, time series illustrations of seasonal and annual T_S are shown by year in Fig. 1 and 2 for the 15-year period 1990-2004 at two selected sites (COW137 and VPI120). Analogous illustrations of T_{OxN} at these same two sites, although not shown, display similar relative behavior. An ANOVA was used to indicate the relative importance of the three main sources of variation (i.e., sites, seasons, and years) to the overall variation in weekly T_S. A rudimentary error propagation analysis of weekly T_S was also performed to identify how variation in individual measured and data-derived quantities contribute to total variability in weekly T_S (Table 4). A statistical model was used to predict D_{Stotal} from monitoring site location, season, and C_{Sg}. An analogous statistical model was also used to predict $D_{OxNtotal}$ from monitoring site location, season, and C_{OxNg}. Finally, deposition estimates from each model were evaluated by comparing PIs, and RDs between modeled and monitored deposition estimates were determined and compared for two sets of monitoring data.

3.1 Analysis of Weekly Data

Site-specific within-season variability of weekly measured and data-derived quantities over all years of the 15-year study period are displayed in Table 1. In addition, the week-to-week variability within seasons was often slightly smaller than the corresponding variability across all weeks in the 15-year data record (not shown). This is likely reflecting additional across-season variability that is absent within seasons. The CVs for quantities associated with wet deposition (PPT and V_{Sw}) are considerably larger than CVs for dry deposition velocities ($V_{d\ SO2}$; $V_{d\ SO4}$; and V_{Sg}). The CVs for V_{Sw} exceed those for V_{Sg} by approximately 600%. Although CVs of the tabulated weekly T_S and the wet deposition quantities are of similar magnitude, the CVs of V_{Sw}

exceed those of T_S by approximately 50%. This suggests that the variability of wet deposition drives the variability of T_S derived from weekly values over several years.

Weekly T_S aggregated from weekly data across all 15 years at each of the 12 eastern study sites exhibit a broad range of variability, with CVs between 31% and 120%, depending on site and season. These results can be used to establish 95% confidence intervals (CIs) (where the half interval width for a 95% CI is 1.96 · CV). This suggests that using weekly results over several years for a specific site and season (this case, although not shown, occurred at Virginia site VPI 120 in fall), could yield 95% CI for weekly T_S as large as (1.96 · 120=) ±235%.

3.1.1 ANOVA

ANOVA was performed with using weekly estimates of T_S, permitting assessment of the relative magnitudes of the sums of squares attributable to the various sources of variation. The resulting comparisons (not shown) suggest that less than 30% ($R^2 = 0.29$) of the variation in weekly estimates of T_S is accounted for by the three main effects (i.e., sites (11.4%), seasons (4.3%), and years (0.5%)) and their interaction terms (12.7%). Alternately, over 70% of the variation remains (in error) after accounting for these three main effects. The contribution of years to variability through its main effect is small and suggests that inter-annual variability in T_S is small relative to other sources of variation. The contribution of seasons to variability has the largest mean square of all sources of variation, while the mean square for site location is somewhat smaller than that for seasons.

3.1.2 Error Propagation

Using weekly data, an error propagation analysis was performed to identify the relative contributions of the variability of each of the pertinent measured and data-derived quantities to the computation of T_S (see the final expression for T_S in the Notation Section). The percentage of total variance in T_S (see Tables 1 and 4) attributed to each quantity at each study site was determined for each season. However, due to space limitations, results are shown in Table 4 for only the summer and winter seasons. The variables determining dry deposition (i.e., C_{SO2}, C_{SO4}, $V_{d\,SO2}$, and $V_{d\,SO4}$) are grouped together in Table 4, as are those determining wet deposition (i.e., C_{Sw} and PPT). The percentage of total variance in T_S contributed by dry deposition variables are summed and appear under the "Dry" heading in Table 4, while the corresponding sum for wet deposition variables appear under the "Wet" heading. The contributions of dry and wet deposition variables sum to 100% of the variation in T_S. In general, variability in $V_{d\,SO2}$ and $V_{d\,SO4}$ as well as C_{SO4} are minor contributors to total variance in T_S. In contrast, the variability of PPT and C_{Sw} are major contributors to total variance in T_S, with C_{SO2} and C_{SO4}, normally showing less importance than C_{Sw}. The right-most two columns of Table 4 (i.e., Dry and Wet) reflect larger relative variability of wet deposition processes and related variables in comparison to those of dry deposition. Wet deposition is an episodic and frequently locally heterogeneous process influenced strongly by meteorological processes. Dry deposition is a continuous process

with deposition velocities influenced strongly by both the chemical species being deposited (e.g., values for gases are generally higher than for corresponding aerosols) and the character of the receiving surface (e.g., values for soluble gases to moist surfaces are generally higher than to dry surfaces) (Cooter and Schwede, 2000). The differences reflected in Table 4 are likely the result of fundamental differences in the nature of dry versus wet deposition at the weekly and site-specific temporal and spatial scales considered.

3.2 Analysis of Seasonal Aggregates

Site-specific, season-to-season variability of seasonal values aggregated by year over the 15-year study period from weekly measured and data-derived quantities is displayed in Tables 2 and 3 for oxidized sulfur and OxN, respectively. Variability of seasonal T_S is reduced in comparison to that derived from weekly values across the 15-year study period and, depending on site and season, yields CVs between 8% and 48% (the latter case, although not shown, occurred at WST109 in spring). Analogous results of the variability of seasonal T_{OxN} yields corresponding CVs between 5% and 31% (the latter case occurred at WST109 in winter). This suggests that using seasonal values aggregated by year over several years for a specific site and season, could yield 95% CI for seasonal T_S as large as ($1.96 \cdot 48$=) ±94%, and 95% CI for seasonal T_{OxN} as large as ±61%. CIs for seasonal transference ratios from seasonal aggregates also appear to be slightly smaller for OxN than oxidized sulfur.

Time series illustrations by season and year of the aggregated measured and data-derived quantities for oxidized sulfur (i.e., $V_{d\,SO2}$; $V_{d\,SO4}$; V_{Sg}; V_{Sw}; PPT; and T_S) were prepared (as well as for the analogous OxN quantities). In the interest of brevity, plots of aggregated seasonal and annual T_S are shown at two sites (COW137 and VPI120) for the period 1990-2004 in Fig. 1 and 2. They illustrate both the broad range of values that T_S can assume across sites as well as the considerable temporal variability both across seasons and to a lesser extent, across years. These results are consistent with earlier findings and emphasize the importance of site-specific factors on the transference ratio at a given location. Similar behavior has also been noted for T_{OxN} (not shown).

Sizeable multi-year trends in oxidized sulfur and nitrogen concentration and corresponding dry and wet deposition have been noted in earlier studies (e.g., Lehmann et al., 2005; Sickles and Shadwick, 2007a). As a result, the aggregated seasonal quantities, listed above, were examined for trends over the 15-year period of record. The collection of results was assessed over all sites and seasons (accounting for simultaneous statistical inference), leading to rejection of the hypothesis that the examined quantities exhibit temporal trends at the 0.05 level. Although not shown, similar behavior was noted for the corresponding quantities for OxN.

3.3 Analysis of Annual Aggregates

Site-specific, year-to-year variability of annual values aggregated by year over the 15-year study period from seasonal aggregates of weekly measured and data-derived quantities for the study sites are displayed for oxidized sulfur in Table 2 (and for OxN in Table 3). The variability of annual T_S using aggregated annual values yields annual CVs between 3% and 17%, depending on site. This variability is reduced in comparison to that found for weekly T_S aggregates based on weekly values across the 15-year study period (Table 1) and for seasonal T_S based on aggregated seasonal values (Table 2). This suggests that using aggregated annual results over several years for a specific site (in this case, COW137), could yield 95% CI for annual T_S as large as ($1.96 \cdot 17$=) ±33%. Analogous examination of the variability of annual T_{OxN} in Table 3 yields annual CVs between 5% and 14%, depending on site. Thus, the variability of annual T_{OxN} is also reduced in comparison to the seasonal T_{OxN} based on aggregated seasonal values. This suggests that using aggregated annual results over several years for a specific site (in this case, COW137), could yield 95% CI for annual T_{OxN} as large as ±27%. Consistent with earlier seasonal results, CIs for annual transference ratios from annual aggregates appear to be slightly tighter for OxN than oxidized sulfur.

3.4 Statistical Models to Predict Total Oxidized Sulfur and Nitrogen Deposition

Using Equations (1a), (1b), and (1c), correlations of D_{Sg}, D_{Sw}, and D_{Stotal} with C_{Sg} were explored using seasonal oxidized sulfur monitoring results. The largest correlations between C_{Sg} and the various components of deposition are with D_{Sg} (Equation (1b)). These correlations (not shown) are always positive and greater than 0.64 (maximum of 0.997). This may be expected, since in actual practice the determination of dry deposition for both airborne SO_2 and airborne sulfate take on similar functional forms to Equation (1b), where deposition velocity replaces transference ratio. In contrast, the smallest correlations occur between C_{Sg} and D_{Sw} (Equation (1c)). There are small negative correlations in nine of 48 cases, and correlations are less than 0.70 in all but two cases (maximum of 0.835). In actual practice the determination of wet deposition, takes on a similar functional form as Equation (1c), where precipitation rate (PPT) replaces transference ratio. However, in this case the airborne concentration of oxidized sulfur (C_{Sg}) is not used, and the sulfate concentration in precipitation (C_{Sw}) is used instead. Previous studies of scavenging ratios based on both monitoring and modeling results (Barrie, 1991; Galloway et al., 1993; Wojcik and Chang, 1997; Hicks, 2005) have shown very large variability in the ratio of precipitation-borne to airborne concentrations for both oxidized sulfur and oxidized nitrogen. Thus, the relatively poor correlations for Equation (1c) may also be expected. As a result, correlations between C_{Sg} and D_{Stotal} (Equation (1a)) are intermediate. These findings for seasonal results are consistent not only with earlier examination of CVs for dry versus wet deposition weekly quantities (Section 3.1) but also with error propagation analysis of weekly results (Section 3.1.2) and suggest that variability in parameter and prediction estimates for the model described by Equation (1a) will be influenced heavily by variation in wet deposition

processes. It should also be noted that while wet deposition processes are not considered directly, they are considered indirectly as the site-specific parameter, τ_{ij}, (i.e., transference ratio) in the form of the model described by Equation (1a).

Individual predictions of total deposition and 99% PIs for the individual predictions evaluated at values of monitored airborne concentration have been generated for a model defined by Equation (1a) for oxidized sulfur and by the analogue for OxN using PC SAS. The 99% PIs and RDs between the model-estimated and monitored values of total deposition at the monitored airborne concentration at each site have been examined to provide two levels of model assessment. Results of these assessments are summarized in Table 5.

In Level 1 assessments, the model defined by Equation (1a) satisfies the 99% PI at both seasonal and annual temporal resolution for both oxidized sulfur and OxN over 98% of the time. For example, considering the case of seasonal oxidized sulfur, 99% PIs were determined for the 514 cases involving monitored D_{Stotal}, and in all cases (or 100% of the time) of paired observed monitored C_{Sg} and D_{Stotal}, the monitored D_{Stotal} value falls within its 99% PI. In Level 2 assessments, the pass rate is reduced at both seasonal and annual temporal resolution, and over 85% of the cases meet the 99% PI criterion. The apparent reduction in pass rate is likely due to the increased rigor associated with using monitoring data from the second, independent data set against PIs established using the first data set; perhaps due to the smaller sample sizes in the Level 2 assessments; and possibly due to model shortcomings. Nevertheless, Level 2 assessments reveal a high percentage of cases meeting the 99% PI criterion, but the pass rate is less than from Level 1 assessment.

The RDs for Level 1 assessment of Equation (1a), shown in Table 5, are within ±25% over 70% of the time in all cases for both oxidized sulfur and OxN. However, the arbitrary acceptability criterion of Equation (3) where RDs must be within ±25% at least 95% of the time is not satisfied at seasonal temporal resolution. At annual temporal resolution, Equation (1a) satisfies the acceptability criterion for both oxidized sulfur and OxN in Equation (3), respectively. Level 2 assessment deteriorates slightly, perhaps a result of the smaller sample sizes. Although the arbitrary acceptability criterion of Equation (3) is not satisfied at seasonal resolution, Level 2 RDs are also within ±25% over 70% of the time for Equation (1a). At annual temporal resolution, Equation (1a) satisfies the acceptability criterion of Equation (3) for both oxidized sulfur and OxN.

Fig. 3 shows monitored airborne seasonal concentration of oxidized sulfur versus RD between the corresponding monitored and modeled total deposition for Level 2 assessments. As indicated in Table 5, 71% (132) of the 185 paired results show RDs falling within the ±25% criterion specified by Equation (3). In general, the proportion of cases meeting the above criteria appears to increase with concentration. Sites located in complex (or montane) terrain are

identified by "C" in Fig. 3. Although C_{Sg} tends to be lower at selected complex terrain sites because their locations may be more remote, other differences in the density or distribution of points that might be associated with monitors located in complex terrain versus other locations are not visually apparent. This suggests that the site-specific considerations included in Equation (1a) for modeling concentration-deposition behavior yields results that are not discernibly different for sites located in complex terrain versus other locations.

4. Conclusions

Use of model-predicted "transference ratios" is currently under consideration by the US EPA in the formulation of a Secondary NAAQS for oxidized nitrogen and oxidized sulfur. This term is an empirical parameter defined for oxidized sulfur (T_S) as the ratio of the total oxidized sulfur deposition (from dry plus wet deposition) to the airborne concentration of oxidized sulfur. A 15-year record of weekly measured and data-derived quantities at 12 monitoring sites in the eastern US was examined to determine the variability of T_S that might be expected from field measurements. Weekly T_S displayed considerable variability that depended on site, season, and year, but ANOVA results suggest most heavily on site and season. Using weekly data, the variability of deposition-related quantities and error propagation analysis of T_S both suggest that variation in parameters related to wet deposition processes are generally more important than variation in parameters related to dry deposition processes in determining variation in T_S. Correlations between airborne seasonal concentration of oxidized sulfur and the various components of seasonal deposition (i.e., dry, wet, and total) also underscore the strong influence that the variability in wet deposition processes can have on estimates of total deposition. Analysis of monitoring results suggests that 95% CI for T_S using weekly results over several years for a specific site and season could be as large as ±235%, and as large as ±94% using seasonal aggregates for a given site, but only as large as ±33% using annual aggregates for a specific site. At the annual scale, a regression model of the form that incorporates a site-specific transference ratio, yielded estimates of D_{Stotal} within ±25% of the monitored values ≥ 95% of the time. Since all of the major oxidized nitrogen species are not monitored regularly, a parallel analysis was limited to only the OxN species. Nevertheless, findings for OxN are consistent with those described above for oxidized sulfur. These results suggest that at specific sites in the eastern US, annual estimates of total deposition to within ±25 to ±35% may be expected using species- and site-specific transference ratios along with annual average monitored airborne concentrations. However, failure to consider the influence of important wet deposition processes directly in the formulation of a model that incorporates a site-specific transference ratio, will likely contribute appreciable uncertainty to estimates of total deposition.

Notation

C_{SO2} – airborne concentration of gaseous SO_2;
C_{SO4} – airborne concentration of aerosol/particulate SO_4^{2-};
C_{Sg} – airborne concentration of oxidized sulfur is the sum of airborne concentrations of gaseous SO_2 and aerosol/particulate SO_4^{2-}, expressed as sulfur, (i.e., $C_{Sg} = C_{SO2} + C_{SO4}$);
C_{Sw} – SO_4^{2-} ion concentration in precipitation, expressed as sulfur;
D_{SO2} – dry deposition of gaseous SO_2, expressed as sulfur, is the product of deposition velocity and airborne concentration of SO_2 (i.e., $D_{SO2} = V_{d\,SO2} \cdot C_{SO2}$);
D_{SO4} – dry deposition of aerosol/particulate SO_4^{2-}, expressed as sulfur, is the product of deposition velocity and airborne concentration of SO_4^{2-} (i.e., $D_{SO4} = V_{d\,SO4} \cdot C_{SO4}$);
D_{Sg} – dry deposition of airborne oxidized sulfur, expressed as sulfur, is the sum of dry deposition of gaseous SO_2 and aerosol/particulate SO_4^{2-} (i.e., $D_{Sg} = D_{SO2} + D_{SO4}$);
D_{Sw} – wet deposition of precipitation-borne oxidized sulfur (SO_4^{2-}), expressed as sulfur, is the product of precipitation rate and SO_4^{2-} ion concentration in precipitation (i.e., $D_{Sw} = PPT \cdot C_{Sw}$);
D_{Stotal} – total oxidized sulfur deposition, expressed as sulfur, is the sum of dry and wet sulfur deposition (i.e., $D_{Stotal} = D_{Sg} + D_{Sw}$);
PPT – precipitation rate;
$V_{d\,SO2}$ – dry deposition velocity for gaseous SO_2;
$V_{d\,SO4}$ – dry deposition velocity for aerosol/particulate SO_4^{2-};
V_{Sg} – dry deposition velocity for airborne oxidized sulfur; this is an *empirical parameter* and can be calculated (i.e., $V_{Sg} = D_{Sg} / C_{Sg}$);
V_{Sw} – wet deposition velocity for precipitation-borne oxidized sulfur (SO_4^{2-}); this is an *empirical parameter* and can be calculated (i.e., $V_{Sw} = D_{Sw} / C_{Sg} = PPT \cdot C_{Sw} / C_{Sg}$);
T_S – transference ratio for oxidized sulfur; this is an *empirical parameter* and can be calculated (i.e., $T_S = [D_{Stotal} / C_{Sg}] = [(D_{Sg} + D_{Sw}) / C_{Sg}] = [V_{Sg} + V_{Sw}] = [(V_{d\,SO2} \cdot C_{SO2} + V_{d\,SO4} \cdot C_{SO4} + PPT \cdot C_{Sw}) / (C_{SO2} + C_{SO4})]$).

Acknowledgments

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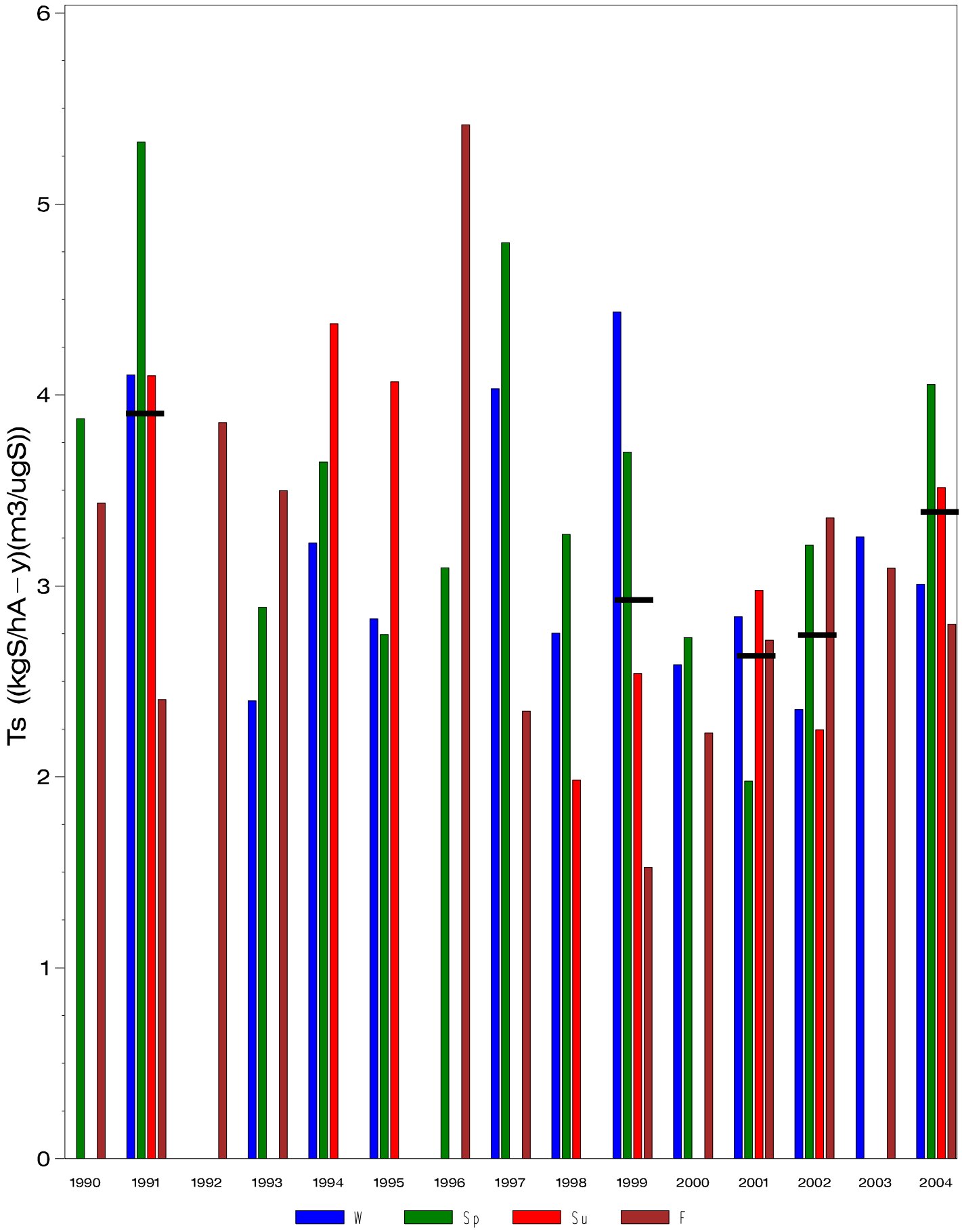
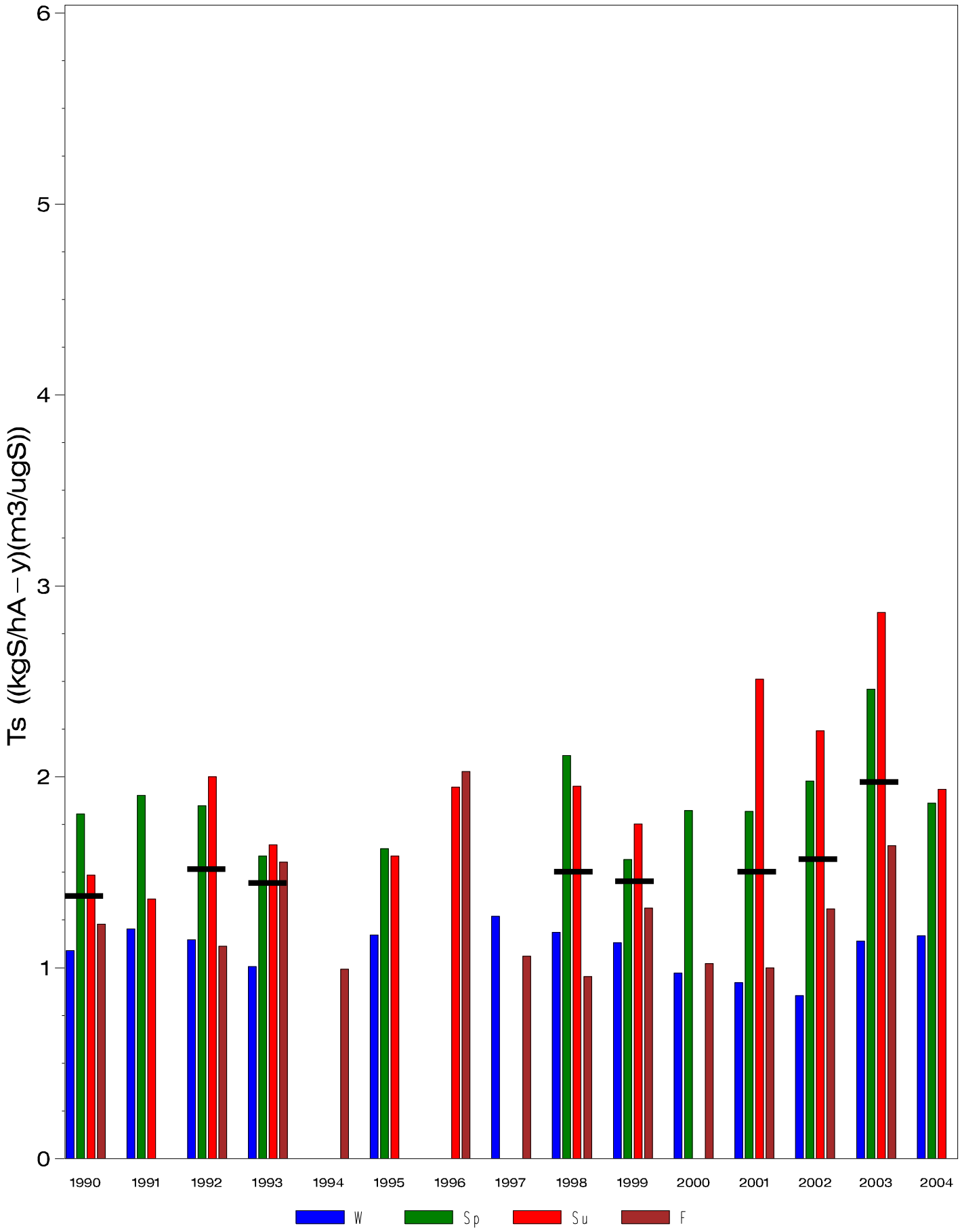
List of Figures

Seasonal and Annual Ts at COW137 1990 – 2004



Seasonal and Annual Ts at VPI120 1990 – 2004



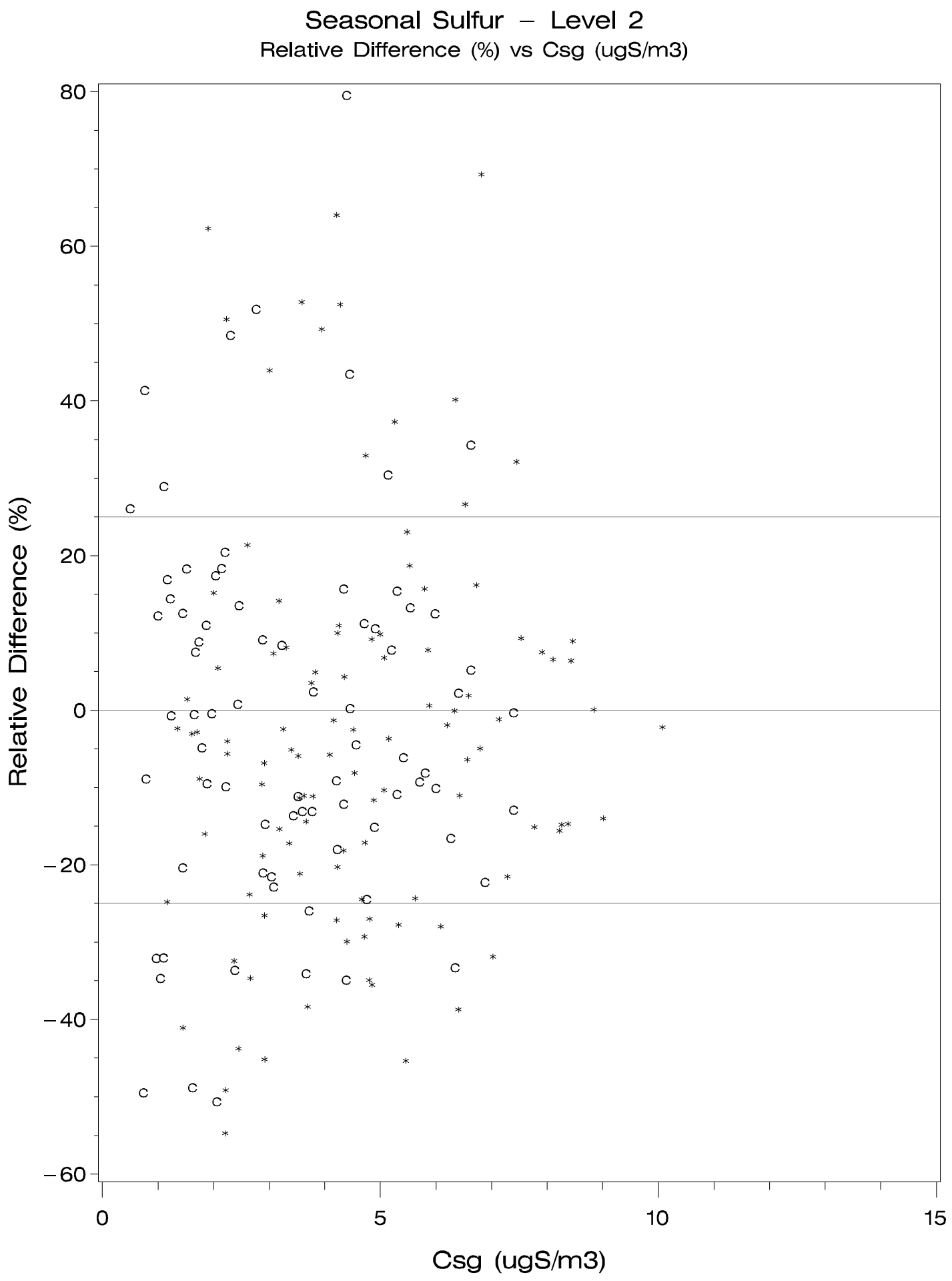

Seasonal Sulfur – Level 2
Relative Difference (%) vs Csg (ugS/m3)
Relative Difference (%)
80
60
40
20
0
−20
−40
−60
0
5
10
15
Csg (ugS/m3)

Table 1. Summer and winter seasonal median values and CVs for dry and wet deposition velocity and related parameters for oxidized sulfur across sites for the period 1990-2004 using aggregated weekly data

CASTNET	NADP	Season	n	$V_{d\ SO2}$[d]		$V_{d\ SO4}$[d]		V_{Sg}[d]		PPT[e]		V_{Sw}[e]		T_S	
				Median	CV	Median	CV	Median	CV	Median	CV	Median	CV	Median	CV
BVL130	IL11	Su	133	0.35	17	0.09	24	0.26	19	21	102	0.54	98	2.54	73
CAD150	AR03	Su	92	0.27	21	0.09	20	0.19	20	23	95	0.56	123	2.38	103
COW137[a,b,c]	NC25	Su	132	0.29	20	0.09	20	0.14	20	22	98	0.91	92	3.33	81
KEF112[a,c]	PA29	Su	138	0.33	20	0.09	22	0.24	21	18	97	0.61	93	2.71	71
MCK131	KY03	Su	110	0.49	21	0.12	18	0.30	22	17	92	0.34	97	2.03	62
OXF122	OH09	Su	125	0.40	23	0.13	17	0.31	23	21	93	0.42	97	2.24	65
PAR107[a,b,c]	WV18	Su	145	0.52	20	0.13	18	0.32	23	23	92	0.62	100	2.90	73
SHN418[a,b,c]	VA28	Su	126	0.38	22	0.14	18	0.24	20	19	110	0.33	136	1.91	97
VIN140	IN22	Su	122	0.44	19	0.13	16	0.39	18	24	88	0.37	88	2.39	53
VPI120[a,b,c]	VA13	Su	135	0.38	17	0.13	16	0.24	19	18	100	0.31	93	1.74	60
WSP144	NJ99	Su	109	0.38	17	0.12	21	0.29	17	18	109	0.51	121	2.59	88
WST109[a,b]	NH02	Su	122	0.24	21	0.06	26	0.13	26	20	101	1.34	107	4.59	101
BVL130	IL11	W	97	0.33	17	0.04	26	0.29	17	7	107	0.13	161	1.37	72
CAD150	AR03	W	108	0.22	13	0.06	31	0.18	17	24	94	0.69	94	2.86	79
COW137	NC25	W	143	0.19	12	0.08	33	0.17	15	35	76	0.81	85	3.18	72
KEF112	PA29	W	92	0.20	11	0.06	34	0.19	11	16	78	0.13	98	1.06	48
MCK131	KY03	W	102	0.51	10	0.09	28	0.44	10	17	115	0.17	133	1.91	51
OXF122	OH09	W	142	0.40	10	0.08	31	0.38	11	14	105	0.11	114	1.54	35
PAR107	WV18	W	127	0.37	13	0.08	31	0.37	12	16	88	0.17	97	1.69	36
SHN418	VA28	W	115	0.32	7	0.11	25	0.30	8	17	107	0.13	97	1.35	39
VIN140	IN22	W	120	0.46	13	0.06	31	0.43	12	13	95	0.12	102	1.73	33
VPI120	VA13	W	130	0.25	9	0.08	21	0.22	9	16	85	0.13	206	1.12	101
WSP144	NJ99	W	138	0.40	14	0.09	30	0.40	13	15	95	0.10	111	1.57	31
WST109	NH02	W	90	0.14	17	0.04	50	0.12	19	17	90	0.26	126	1.19	101

[a]Site location vulnerable to acidic deposition

[b]Site located in complex (or montane) terrain

[c]High elevation site (>500m)

[d]Dry deposition variables;

[e]Wet deposition variables;

Units: deposition velocity - cm/s; precipitation rate - cm/season; CV - %; transference ratio - [(kg ha^{-1} y^{-1})(m^{3} μg^{-1})]; to convert transference ratio from the previous units into cm/s, divide by 3.1536.

Table 2. Summer, winter, and annual median values and CVs for dry and wet deposition velocity and related parameters for oxidized sulfur across sites for the period 1990-2004 using aggregated seasonal and annual data

CASTNET	NADP	Season	n	$V_{d\ SO2}$[a]		$V_{d\ SO4}$[a]		V_{Sg}[a]		PPT[b]		V_{Sw}[b]		T_S	
				Median	CV	Median	CV	Median	CV	Median	CV	Median	CV	Median	CV
BVL130	IL11	Su	13	0.35	11	0.09	14	0.25	10	28	34	0.48	43	2.41	31
CAD150	AR03	Su	9	0.27	8	0.09	12	0.19	8	24	33	0.44	38	2.00	29
COW137	NC25	Su	8	0.28	15	0.10	10	0.14	5	34	33	0.89	33	3.24	29
KEF112	PA29	Su	11	0.32	12	0.09	9	0.24	8	36	28	0.66	37	2.90	29
MCK131	KY03	Su	10	0.47	16	0.12	5	0.29	13	29	30	0.36	41	2.11	26
OXF122	OH09	Su	12	0.38	17	0.13	11	0.31	16	26	33	0.37	36	2.23	25
PAR107	WV18	Su	11	0.52	18	0.13	10	0.33	13	39	33	0.84	34	3.70	27
SHN418	VA28	Su	9	0.39	19	0.14	9	0.26	13	38	28	0.44	23	2.24	18
VIN140	IN22	Su	12	0.43	11	0.13	8	0.38	10	36	26	0.38	30	2.38	19
VPI120	VA13	Su	12	0.38	11	0.12	8	0.25	11	28	33	0.34	34	1.94	22
WSP144	NJ99	Su	8	0.41	10	0.12	10	0.31	9	30	28	0.61	29	2.82	20
WST109	NH02	Su	11	0.24	14	0.06	12	0.13	15	31	21	1.74	26	5.93	24
BVL130	IL11	W	10	0.33	9	0.04	9	0.29	8	14	34	0.15	50	1.39	20
CAD150	AR03	W	10	0.23	4	0.06	9	0.19	7	40	17	0.79	28	3.10	22
COW137	NC25	W	12	0.19	4	0.08	10	0.18	4	51	23	0.75	27	2.92	22
KEF112	PA29	W	5	0.20	4	0.06	9	0.19	5	25	9	0.14	27	1.07	11
MCK131	KY03	W	9	0.50	4	0.09	11	0.44	3	26	29	0.18	42	1.99	14
OXF122	OH09	W	14	0.39	4	0.08	11	0.37	4	20	24	0.13	28	1.59	9
PAR107	WV18	W	7	0.37	4	0.08	13	0.36	4	25	18	0.18	26	1.73	8
SHN418	VA28	W	11	0.32	2	0.11	8	0.29	2	28	29	0.12	29	1.33	9
VIN140	IN22	W	8	0.44	7	0.07	14	0.42	6	18	29	0.12	29	1.70	10
VPI120	VA13	W	13	0.25	7	0.09	8	0.23	7	21	26	0.13	25	1.14	11
WSP144	NJ99	W	12	0.41	6	0.09	12	0.40	6	22	29	0.11	33	1.62	8
WST109	NH02	W	5	0.14	7	0.04	14	0.12	5	25	12	0.43	27	1.77	22
BVL130	IL11	Annual	6	0.36	4	0.07	13	0.28	3	99	12	0.37	19	2.07	11
COW137	NC25	Annual	5	0.22	10	0.10	6	0.16	4	160	12	0.76	20	2.93	17
MCK131	KY03	Annual	5	0.49	3	0.11	4	0.37	2	115	13	0.31	9	2.18	3
OXF122	OH09	Annual	8	0.40	9	0.12	9	0.36	8	109	18	0.26	22	1.94	13
PAR107	WV18	Annual	5	0.37	10	0.12	11	0.34	6	115	12	0.38	16	2.30	10
SHN418	VA28	Annual	5	0.32	11	0.13	7	0.27	7	140	22	0.30	23	1.80	14
VIN140	IN22	Annual	6	0.45	4	0.10	7	0.41	4	122	15	0.25	22	2.13	10
VPI120	VA13	Annual	8	0.30	5	0.11	7	0.24	5	98	21	0.24	21	1.50	12
WSP144	NJ99	Annual	6	0.40	4	0.11	7	0.37	4	104	14	0.31	19	2.14	8

[a]Dry deposition variables;

[b]Wet deposition variables;

Units: for most entries see Table 1; annual precipitation rate - cm/y

Table 3. Summer, winter, and annual median values and CVs for dry and wet deposition velocity and related parameters for monitored oxidized nitrogen across sites for the period 1990-2004 using aggregated seasonal and annual data

CASTNET	NADP	Season	n	$V_{d\ HNO3}$[a]		$V_{d\ NO3}$[a]		V_{OxNg}[a]		PPT[b]		V_{OxNw}[b]		T_{OxN}	
				Median	CV	Median	CV	Median	CV	Median	CV	Median	CV	Median	CV
BVL130	IL11	Su	13	1.03	11	0.09	14	0.91	11	28	34	1.28	33	7.03	19
CAD150	AR03	Su	7	1.19	7	0.10	12	1.38	5	24	37	1.64	42	9.89	24
COW137	NC25	Su	8	1.26	7	0.10	10	1.23	8	34	33	4.04	28	16.41	20
KEF112	PA29	Su	11	1.32	7	0.09	9	1.28	7	36	28	3.17	26	13.74	18
MCK131	KY03	Su	10	1.40	6	0.12	5	1.19	6	29	30	1.61	36	8.75	21
OXF122	OH09	Su	12	1.68	9	0.13	11	1.55	8	26	33	1.40	38	9.42	17
PAR107	WV18	Su	11	1.66	11	0.13	10	1.67	9	39	33	3.68	27	17.21	19
SHN418	VA28	Su	10	1.78	10	0.14	8	1.61	9	36	28	1.52	28	9.84	17
VIN140	IN22	Su	12	1.58	6	0.13	8	1.31	8	36	26	1.41	30	8.57	16
VPI120	VA13	Su	12	1.75	6	0.12	8	1.51	7	28	33	1.43	36	9.37	17
WSP144	NJ99	Su	8	1.40	9	0.12	10	1.46	7	30	28	1.65	27	9.99	14
WST109	NH02	Su	11	0.96	13	0.06	12	0.75	12	31	21	5.28	21	19.16	17
BVL130	IL11	W	9	0.54	6	0.04	7	0.15	18	15	31	0.40	34	1.83	26
CAD150	AR03	W	10	0.79	5	0.06	9	0.63	11	40	17	1.80	20	7.54	15
COW137	NC25	W	12	0.84	6	0.08	10	0.84	6	51	23	3.29	27	13.13	22
KEF112	PA29	W	5	0.72	5	0.06	9	0.55	13	25	9	1.86	5	7.74	5
MCK131	KY03	W	9	1.33	4	0.09	11	0.85	11	26	29	0.69	42	4.94	25
OXF122	OH09	W	13	1.01	5	0.08	9	0.44	13	20	25	0.65	28	3.46	19
PAR107	WV18	W	7	1.06	7	0.08	13	0.74	6	25	18	2.28	21	9.57	15
SHN418	VA28	W	11	1.24	5	0.11	8	0.98	4	28	29	0.72	26	5.33	12
VIN140	IN22	W	8	1.00	7	0.07	14	0.37	14	18	29	0.55	23	2.93	18
VPI120	VA13	W	13	0.92	5	0.09	8	0.76	7	21	26	0.76	27	4.78	14
WSP144	NJ99	W	12	1.05	5	0.09	12	0.57	13	22	29	0.83	32	4.30	22
WST109	NH02	W	5	0.48	7	0.04	14	0.37	9	25	12	4.92	33	16.51	31
COW137	NC25	Annual	5	1.07	4	0.10	6	1.12	5	160	12	3.09	21	13.57	14
MCK131	KY03	Annual	5	1.40	3	0.11	4	1.06	7	116	13	1.26	16	7.39	12
OXF122	OH09	Annual	7	1.35	8	0.12	10	0.95	8	96	17	0.93	16	6.07	8
PAR107	WV18	Annual	6	1.39	9	0.11	10	1.28	8	112	12	2.54	12	12.00	8
SHN418	VA28	Annual	5	1.47	5	0.13	7	1.28	2	140	22	1.06	13	7.38	6
VIN140	IN22	Annual	6	1.32	5	0.10	7	0.81	5	124	15	1.04	15	5.81	10
VPI120	VA13	Annual	7	1.38	4	0.11	6	1.22	6	96	10	1.00	11	7.09	5
WSP144	NJ99	Annual	6	1.28	6	0.11	7	1.14	4	104	14	1.27	19	7.57	10

[a]Dry deposition variables;

[b]Wet deposition variables;

Units: see Tables 1 and 2

Table 4. Error propagation analysis for transference ratio of oxidized sulfur by site and season for the period 1990-2004 using weekly data

CASTNET	NADP	n	Season	% of Total Summed Variance in T_S							
				C_{SO2}[a]	C_{SO4}[a]	$V_{d\ SO2}$[a]	$V_{d\ SO4}$[a]	C_{Sw}[b]	PPT[b]	Dry[c]	Wet[d]
BVL130	IL11	133	Su	4.6	1.8	0.1	0.01	30.7	62.8	7	93
CAD150	AR03	92	Su	1.9	4.3	0.0	0.01	33.4	60.4	6	94
COW137	NC25	132	Su	0.4	4.5	0.0	0.01	40.0	55.2	5	95
KEF112	PA29	138	Su	5.1	1.6	0.1	0.01	26.0	67.2	7	93
MCK131	KY03	110	Su	7.0	1.3	0.5	0.02	33.2	57.9	9	91
OXF122	OH09	125	Su	7.1	1.4	0.6	0.01	22.5	68.5	9	91
PAR107	WV18	145	Su	4.8	1.2	0.1	0.01	45.6	48.2	6	94
SHN418	VA28	126	Su	4.7	1.4	0.2	0.02	32.6	61.1	6	94
VIN140	IN22	122	Su	8.2	0.8	0.6	0.01	35.9	54.5	10	90
VPI120	VA13	135	Su	3.5	1.2	0.2	0.02	29.7	65.4	5	95
WSP144	NJ99	109	Su	2.8	1.2	0.1	0.01	23.5	72.4	4	96
WST109	NH02	122	Su	0.8	11.9	0.0	0.00	22.2	65.1	13	87
BVL130	IL11	97	W	33.9	0.1	2.7	0.00	11.6	51.7	37	63
CAD150	AR03	108	W	8.6	1.1	0.0	0.00	42.8	47.5	10	90
COW137	NC25	143	W	12.3	1.0	0.0	0.01	27.2	59.6	13	87
KEF112	PA29	92	W	32.6	0.1	1.1	0.01	18.0	48.2	34	66
MCK131	KY03	102	W	28.2	0.1	0.8	0.01	16.2	54.8	29	71
OXF122	OH09	142	W	36.7	0.0	2.4	0.01	12.6	48.3	39	61
PAR107	WV18	127	W	45.5	0.0	2.1	0.01	15.1	37.3	48	52
SHN418	VA28	115	W	27.7	0.1	0.3	0.01	19.2	52.7	28	72
VIN140	IN22	120	W	51.5	0.0	3.4	0.01	8.9	36.2	55	45
VPI120	VA13	130	W	24.5	0.1	0.6	0.01	29.4	45.3	25	75
WSP144	NJ99	138	W	34.6	0.0	3.6	0.01	24.0	37.8	38	62
WST109	NH02	90	W	12.0	0.4	0.1	0.01	36.2	51.3	13	87

[a]Dry deposition variables;
[b]Wet deposition variables;
[c]Summed percentage of total variance in T_S contributed by dry deposition variables;
[d]Summed percentage of total variance in T_S contributed by wet deposition variables.

Table 5. Level 1 and 2 assessment summaries for Equation (1a) to estimate total deposition from airborne concentration

Model Assessment	Temporal Resolution	Species	n	% Passing Following Tests 99% PIs	RD ≤ ± 25%
Level 1[a]					
	Seasonal	SOx	514	100	79
	Seasonal	OxN	513	98	83
	Annual	SOx	54	100	100
	Annual	OxN	47	100	100
Level 2[b]					
	Seasonal	SOx	185	96	71
	Seasonal	OxN	185	96	76
	Annual	SOx	26	88	96
	Annual	OxN	23	100	100

[a]Uses 1990-2004 monitoring data to derive parameters and assess model (Equation 1a)

[b]Uses Level 1 parameters and 2005-2009 monitoring data to assess model (Equation 1a)