

**Supplementary Information for:**

**Determining the spatial and seasonal variability in OM/OC ratios across the U.S.  
using multiple regression**

Heather Simon<sup>1</sup>, Prakash V. Bhave<sup>2</sup>, Jenise L. Swall<sup>2</sup>, Neil H. Frank<sup>1</sup>, William C. Malm<sup>3</sup>

<sup>1</sup> US EPA, Office of Air Quality Planning and Standards, Research Triangle Park, NC

<sup>2</sup> US EPA, National Exposure Research Laboratory, Atmospheric Modeling and Analysis  
Division, Research Triangle Park, NC

<sup>3</sup> National Park Service, Colorado State University/Cooperative Institute for Research in  
the Atmosphere, Fort Collins, CO

Table of contents:

Section S1. Methodology

Section S1.1. Calculating the variance of regression coefficients – **p. 2**

Section S1.2. Sample R code – **p. 3**

Section S2. Sulfate coefficient analysis – **p. 7**

Section S3. Sensitivity of  $\beta_{OC}$  to the inclusion of  $\beta_{EC}$  and assumptions about OC artifacts  
– **p. 10**

Section S4. Maps of regression coefficients – **p. 15**

Section S5. Tabulated regression results – **p. 20**

Section S6. References – **p. 49**

## S1. Methodology

### S1.1. Calculating the variance of regression coefficients

To find the estimated variance associated with the regression coefficients in equation (11), we need to make some additional calculations. This discussion, like that in Sec. 2.3, is based entirely on the work of Fuller (1987) (Sec 3.1.2), conforming to his original notation as much as is feasible. We begin by defining the matrix  $\hat{M}_{z\pi z}$  as

$$\hat{M}_{z\pi z} = n^{-1} \sum_{t=1}^n [\tilde{\sigma}_{v\pi t}^{-1} (Z_t' Z_t - \Sigma_{a\pi t})] \quad (S1)$$

We are most interested in the lower right submatrix of  $\hat{M}_{z\pi z}$ ; i.e., the submatrix which remains when the first row and first column of  $\hat{M}_{z\pi z}$  are removed. We call this  $k \times k$  submatrix  $\hat{M}_{x\pi x}$ , where  $k$  is the number of explanatory variables in the regression model.

The estimated covariance matrix associated with our regression coefficients is given by

$$\hat{V}(\hat{\beta}) = n^{-2} \hat{M}_{x\pi x}^{-1} \left\{ \sum_{t=1}^n [\tilde{\sigma}_{v\pi t}^{-1} (X_t' X_t + \tilde{\sigma}_{v\pi t}^{-1} \Sigma_{u\pi t} \tilde{\beta} \tilde{\beta}' \Sigma_{u\pi t})] \right\} \hat{M}_{x\pi x}^{-1} \quad (S2)$$

As mentioned in Sec 2.3, the diagonal elements of the matrix given by  $\hat{V}(\hat{\beta})$  are the estimated variances associated with each of the regression coefficients (each of the elements of  $\hat{\beta}$ ). The square roots of these variances are referred to as the estimated standard errors for the regression coefficients.

## S1.2. Sample R Code

The following R code can be used to calculate regression coefficients

---

```
# load functions necessary for these calculations
source("func_for_beta_est.r")

# calculate response variable
data$response <- data$PM25_Value - (data$EC_Value + 1.2*data$knon_Value +
1.8*data$Cl_Value)

# Set up a data frame with response variable and covariates. Each entry in the data
# frame (measured sample) includes values and reported uncertainties for the PM
# components
regdata <- data.frame(y = data$response, sulfate = data$ammsulfate, nitrate <-
  data$amnitrate_Value, OC = data$OC_Value, soil = data$soil_Value, y_Unc =
  data$response_Unc, sulfate_Unc = data$ammsulfate_Unc, nitrate_Unc =
  data$amnitrate_Unc, OC_Unc = data$OC_Unc, soil_Unc = data$soil_Unc)

# Create a data frame. Each row contains the name of the covariate value in the
# first column and the name of the column containing the uncertainty values for that
# variable in the second column.
names.covariates.columns.df <- data.frame(value=c("sulfate", "nitrate", "OC", "soil"),
sd=c("sulfate_Unc", "nitrate_Unc", "OC_Unc", "soil_Unc"), stringsAsFactors=F)
num.covariates = nrow(names.covariates.columns.df)

# Create a data frame containing just one row. The first column holds the name of
# the column for the response variable value. The second column holds the name
# of the column for the error associated with the response.
names.response.columns.df <- data.frame(value="y", sd="y_Unc", stringsAsFactors=F)

# obtain preliminary estimate for betas
prelim.beta.est <- find.prelim.beta.est(regdata, names.response.columns.df,
  names.covariates.columns.df)

# Calculate var.qq given preliminary estimate
var.qq <- find.var.qq(regdata, names.response.columns.df, names.covariates.columns.df,
  prelim.beta.est)

# calculate new beta est, G, M.zpiz
beta.est.etc <- find.beta.est.etc(regdata, names.response.columns.df,
  names.covariates.columns.df, var.qq, prelim.beta.est)
beta.est <- beta.est.etc$beta.est
M.zpiz <- beta.est.etc$M.zpiz
var.beta.est <- find.beta.est.var(regdata, names.response.columns.df,
  names.covariates.columns.df, var.qq, M.zpiz)
stdev.beta.est <- sqrt(diag(var.beta.est))
var.beta.est <- find.beta.est.var(regdata, names.response.columns.df,
  names.covariates.columns.df, var.qq, M.zpiz)
stdev.beta.est <- sqrt(diag(var.beta.est))

sulfate_coeff <- beta.est[1]
nitrate_coeff <- beta.est[2]
oc_coeff <- beta.est[3]
soil_coeff <- beta.est[4]
sulfate_stdev <- stdev.beta.est[1]
nitrate_stdev <- stdev.beta.est[2]
oc_stdev <- stdev.beta.est[3]
soil_stdev <- stdev.beta.est[4]
```

---

Below is the text from a file that defines the functions needed to estimate the regression coefficients and standard deviations: func\_for\_beta\_est.r.

---

```
##### find.prelim.beta.est function #####
find.prelim.beta.est <- function(data.df, names.response.columns.df,
  names.covariates.columns.df){
```

```

# The number of observations is equal to the number of rows of data.df
num.obs <- nrow(data.df)

# The number of covariates is equal to the number of rows of
# names.covariates.columns.df.
num.covariates <- nrow(names.covariates.columns.df)

# Initialize at 0.
M.xx <- matrix(0.0, nrow=num.covariates, ncol=num.covariates)
M.xy <- rep(0.0, num.covariates)

for (j in 1:num.obs){

  # Calculate beta estimate.
  X.j <- as.vector(as.matrix(data.df[j, names.covariates.columns.df$value]))
  Y.j <- data.df[j, names.response.columns.df$value]

  M.xy <- M.xy + (X.j * Y.j)

  # Covariance matrix of measurement standard deviations among covariates.
  cov.uu <- diag(as.vector(as.matrix(data.df[j, names.covariates.columns.df$sd]^2)),
  ncol=num.covariates, nrow=num.covariates)
  M.xx <- M.xx + ( X.j %*% t(X.j) - cov.uu )
}

M.xy <- M.xy / num.obs
M.xx <- M.xx / num.obs

return(as.vector(solve(M.xx) %*% M.xy))
}

##### find.var.qq function #####
find.var.qq <- function(data.df, names.response.columns.df, names.covariates.columns.df,
prelim.beta.est){

# The number of observations is equal to the number of rows of
# data.df
num.obs <- nrow(data.df)

# The number of covariates is equal to the number of rows of
# names.covariates.columns.df.
num.covariates <- nrow(names.covariates.columns.df)

# Initialize at 0.
sig.qq <- 0.0
A <- matrix(0, num.covariates+1, num.covariates+1)
M <- matrix(0, num.covariates+1, num.covariates+1)

# Loop through the observations, adding a contribution from each to sig.qq.
for (j in 1:num.obs){

  # Identify response, covariates, and combined error matrix for
  # observation j.
  X.j <- as.vector(as.matrix(data.df[j, names.covariates.columns.df$value]))
  Y.j <- data.df[j, names.response.columns.df$value]
  cov.aa <- diag(as.vector(as.matrix(data.df[j, c(names.response.columns.df$sd,
names.covariates.columns.df$sd)]^2)), ncol=num.covariates+1,
nrow=num.covariates+1)

  ## Estimate var.qq.
  first.part <- ( (Y.j - (t(X.j) %*% prelim.beta.est))^2 ) / (num.obs -
num.covariates)
  one.and.neg.beta <- c(1.0, -prelim.beta.est)
  second.part <- ( t(one.and.neg.beta) %*% cov.aa %*% one.and.neg.beta ) / num.obs

  sig.qq <- sig.qq + (first.part - second.part)

  ## Calculate generalized eigenvalues.
  A <- A + ( c(Y.j, X.j) %*% t(c(Y.j, X.j)) )
  M <- M + cov.aa
}

```

```

    }

# Find the minimum of the generalized eigenvalues det(A - lamda M)
# = 0. Since our M is diagonal, we can simplify this to finding
# the eigenvalues (in the standard fashion) of inv(M) %*% A. We
# know that these eigenvalues must be real, so any small imaginary
# parts are numerical artifacts.
lambda <- min ( Re( eigen(solve(M) %*% A)$values ) )

# If lambda is smaller than one, then sig.qq should be 0,
# instead of the value we calculated in the loop.
if (lambda < 1)
  return(0)
else
  return(as.vector(sig.qq))
}

##### find.var.vv.for.indiv.obs function #####
# Assumes no correlation among covariate measurement errors and no
# correlation between covariate response measurement errors.
find.var.vv.for.indiv.obs <- function(var.qq, response.sd, covariates.sd,
prelim.beta.est){

  var.ww <- response.sd^2
  cov.uu <- diag(covariates.sd^2, ncol=num.covariates, nrow=num.covariates)

  return( as.vector(var.qq + var.ww + ( t(prelim.beta.est) %*% cov.uu %*%
    prelim.beta.est ) ) )
}

##### find.beta.est.etc function #####
find.beta.est.etc <- function(data.df, names.response.columns.df,
  names.covariates.columns.df, var.qq, prelim.beta.est){

# The number of observations is equal to the number of rows of data.df
num.obs <- nrow(data.df)

# The number of covariates is equal to the number of rows of
# names.covariates.columns.df.
num.covariates <- nrow(names.covariates.columns.df)

# Initialize to 0.
G <- matrix(0.0, nrow=num.covariates, ncol=num.covariates)
mult1 <- matrix(0.0, nrow=num.covariates, ncol=num.covariates)
mult2 <- rep(0.0, num.covariates)
M.zpiz <- matrix(0.0, nrow=num.covariates+1, ncol=num.covariates+1)

for (j in 1:num.obs){

# Find var.vv for this observation.
var.vv <- find.var.vv.for.indiv.obs(var.qq, response.sd=as.vector(data.df[j,
  names.response.columns.df$sd]), covariates.sd=as.vector(as.matrix(data.df[j,
  names.covariates.columns.df$sd])), prelim.beta.est)

cov.uu <- diag(as.vector(as.matrix(data.df[j, names.covariates.columns.df$sd]^2)),
  ncol=num.covariates, nrow=num.covariates)
cov.uv <- -cov.uu %*% prelim.beta.est

# Now, we have enough info to get G.
X.j <- as.vector(as.matrix(data.df[j, names.covariates.columns.df$value]))
G <- G + ( ( X.j %*% t(X.j) ) * var.vv) + (cov.uv %*% t(cov.uv)) )

# Find final beta estimate.

# Find Y.j
Y.j <- data.df[j, names.response.columns.df$value]

```

```

# Combine with var.vv, X.j, and cov.uu.
# First multiplier.
mult1 <- mult1 + ( ( X.j %*% t(X.j) ) - cov.uu ) / var.vv )
mult2 <- mult2 + ( (X.j * Y.j) / var.vv )

# Find M.zpiz.

Z.j <- c(Y.j, X.j)
cov.aa <- diag(as.vector(as.matrix(data.df[j, c(names.response.columns.df$sd,
names.covariates.columns.df$sd)]^2)), ncol=num.covariates+1,
nrow=num.covariates+1)
M.zpiz <- M.zpiz + ( ( Z.j %*% t(Z.j) ) - cov.aa ) / var.vv )

}

G <- G / num.obs
beta.est <- solve(mult1) %*% mult2
M.zpiz <- M.zpiz / num.obs

return(list(beta.est=as.vector(beta.est), G=G, M.zpiz=M.zpiz))
}

##### find.beta.est.var function #####
find.beta.est.var <- function(data.df, names.response.columns.df,
names.covariates.columns.df, var.qq, M.zpiz){

# The number of observations is equal to the number of rows of
# data.df
num.obs <- nrow(data.df)

# The number of covariates is equal to the number of rows of
# names.covariates.columns.df.
num.covariates <- nrow(names.covariates.columns.df)

# Initialize to 0.
mid.part <- matrix(0.0, nrow=num.covariates, ncol=num.covariates)

for (j in 1:num.obs){

cov.uu <- diag(as.vector(as.matrix(data.df[j, names.covariates.columns.df$sd]^2)),
ncol=num.covariates, nrow=num.covariates)
cov.uv <- -cov.uu %*% prelim.beta.est

X.j <- as.vector(as.matrix(data.df[j, names.covariates.columns.df$value]))

var.vv <- find.var.vv.for.indiv.obs(var.qq, response.sd=as.vector(data.df[j,
names.response.columns.df$sd]), covariates.sd=as.vector(as.matrix(data.df[j,
names.covariates.columns.df$sd])), prelim.beta.est)

mid.part <- mid.part + ( ( X.j %*% t(X.j) ) + ((cov.uv %*% t(cov.uv))/var.vv) ) /
var.vv )
}

M.xpix <- M.zpiz[-1, -1]

return( (1.0/(num.obs^2)) * (solve(M.xpix) %*% mid.part %*% solve(M.xpix)) )
}

```

## S2. Sulfate coefficient analysis

Table S1 shows how  $\beta_{\text{sulf}}$  should change with laboratory RH and degree of sulfate neutralization (DSN). The DSN is calculated assuming that all nitrate is in the form of ammonium nitrate and that any ammonium not bound to nitrate is bound to sulfate (Pinder et al., 2008). A DSN of 2 means that two moles of ammonium are available to bond with every mole of sulfate, indicating fully neutralized ammonium sulfate.

$$DSN = \frac{NH_4(\text{moles}) - NO_3(\text{moles})}{SO_4(\text{moles})} \quad (\text{S3})$$

We used the AIM model (Wexler and Clegg, 2002) to estimate total water mass associated with sulfate aerosols for both the dry hysteresis branch and for supersaturated aerosols. Ammoniated sulfate switches from wet to dry at its efflorescence RH (Colberg et al., 2003).

Table S1. Estimated  $\beta_{\text{sulf}}$  values based on laboratory RH and DSN for dry (red) and wet (blue) particles.

		Degree of sulfate neutralization (DSN)										
		1	1.1	1.2	1.3	1.4	1.5	1.6	1.7	1.8	1.9	2
Laboratory relative humidity (RH)	30%	1.03	1.00	0.99	1.01	1.04	0.94	0.94	0.96	0.97	0.99	1.00
	31%	1.04	1.01	1.00	1.01	1.05	0.94	0.94	0.96	0.97	0.99	1.00
	32%	1.05	1.02	1.01	1.02	1.05	0.94	0.94	0.96	0.97	0.99	1.00
	33%	1.06	1.03	1.02	1.03	1.06	0.94	0.94	0.96	0.97	0.99	1.00
	34%	1.07	1.04	1.03	1.04	1.07	1.12	0.94	0.96	0.97	0.99	1.00
	35%	1.08	1.05	1.04	1.05	1.08	1.13	0.94	0.96	0.97	0.99	1.00
	36%	1.09	1.06	1.05	1.06	1.09	1.13	1.18	0.96	0.97	0.99	1.00
	37%	1.11	1.07	1.06	1.06	1.10	1.14	1.19	0.96	0.97	0.99	1.00
	38%	1.12	1.08	1.07	1.07	1.11	1.15	1.20	0.96	0.97	0.99	1.00
	39%	1.13	1.09	1.08	1.08	1.12	1.16	1.21	0.96	0.97	0.99	1.00
	40%	1.14	1.11	1.09	1.10	1.13	1.18	1.22	1.26	0.97	0.99	1.00
	41%	1.15	1.12	1.10	1.11	1.14	1.19	1.23	1.27	1.30	0.99	1.00
42%	1.17	1.13	1.11	1.12	1.15	1.20	1.24	1.28	1.31	1.34	1.00	

Figure S1 shows seasonal variation in the laboratory RH where the filters were weighed, based on several years of data. Though the variation is modest, laboratory RH values are slightly higher during quarter 3 and lowest during quarter 4. Ninety percent of the samples were weighed between 5 and 29 days after sampling, so samples are generally weighed during the same time of year as they are sampled.

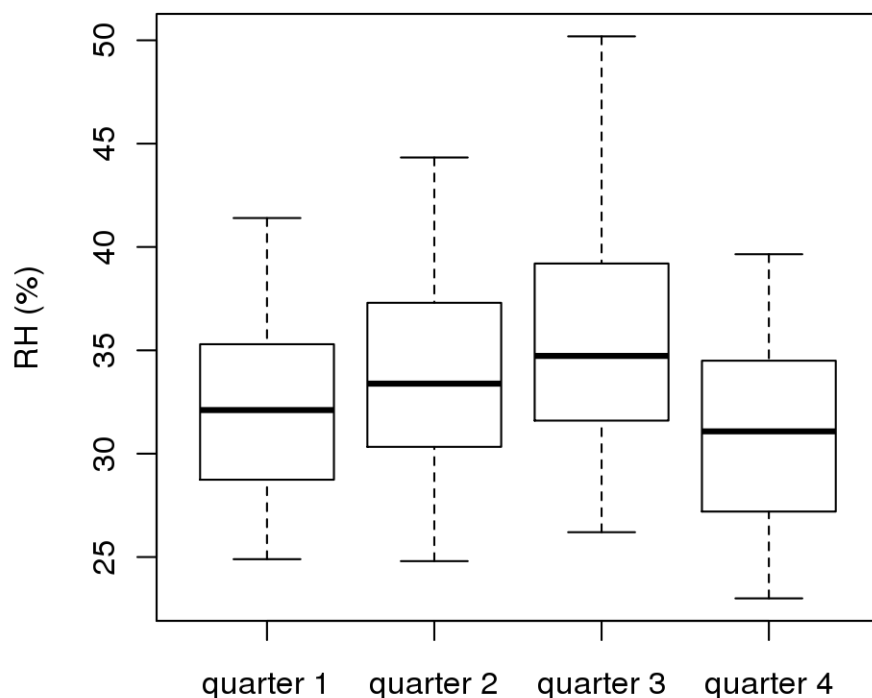


Figure S1. Seasonal RH variation in the IMPROVE gravimetric measurement laboratory

To determine if the seasonal variation in our  $\beta_{\text{sulf}}$  estimates is reasonable, we examined measurements collected between 1999 and 2007 across the CSN where ammonium concentrations are routinely measured along with sulfate and nitrate (downloaded September 24, 2009 from [http://www.epa.gov/cgi-bin/htmSQL/mxplorer/query\\_spe.hsql](http://www.epa.gov/cgi-bin/htmSQL/mxplorer/query_spe.hsql)). These calculations show that DSN does indeed vary seasonally in the southeast, great lakes and northeast regions, with less seasonal variation in the central and western regions. The seasonal variations in DSN are consistent with measurements reported from the Pittsburgh supersite which showed that sulfate was fully neutralized in the winter but not in the summer (Khlystov et al., 2005). The calculated DSN values are used to approximate  $\beta_{\text{sulf}}$  at CSN sites using Table S1 and assuming laboratory RH values of 35% in q1, 37% in q2, 39% in q3, and 35% in q4 (Fig.



S2b) . Except for the western region, our approximations of CSN  $\beta_{\text{sulf}}$  show a seasonal pattern similar to that estimated by our regression analysis of IMPROVE data (copied from Fig. 9 to Fig. S2a to facilitate comparison) with both having higher values in the summer and lower values in the winter. This analysis suggests that the trends predicted by this regression analysis are reasonably explained by known physical phenomena.

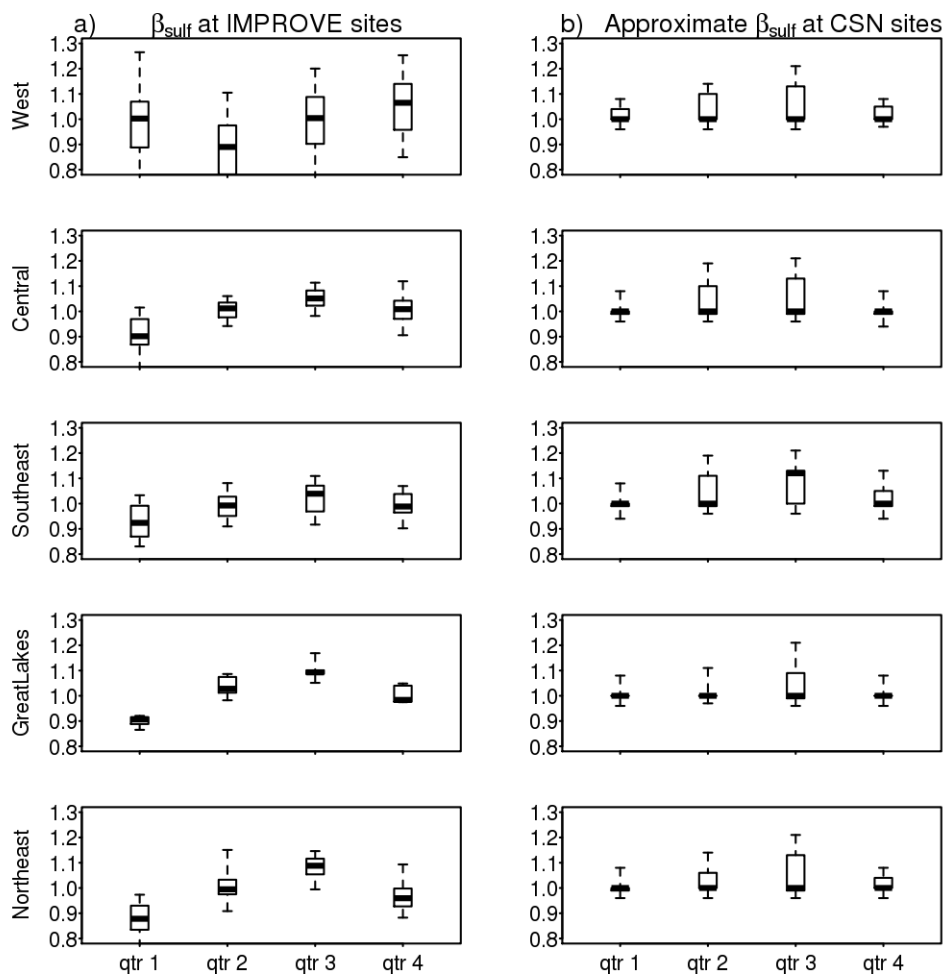


Figure S2. (a) Seasonal and temporal trends in  $\beta_{\text{sulf}}$  from regression of IMPROVE data. (b) Calculated  $\beta_{\text{sulf}}$  values based on CSN measurements of  $\text{NH}_4^+$ ,  $\text{SO}_4^{2+}$ , and  $\text{NO}_3^-$ , and RH in the IMPROVE gravimetric measurement laboratory.

### S3. Sensitivity of $\beta_{OC}$ to the inclusion of $\beta_{EC}$ and assumptions about OC artifacts

The component of  $PM_{2.5}$  reported as EC may not be purely graphitic and therefore may have some non-carbon mass associated with it. In that case, the EC coefficient in Eq. (5) could be greater than one. Also, there is some uncertainty in the measurement method used to split total carbon (TC) into EC and OC which could lead to either a positive or negative EC artifact. Average EC/TC values have been reported to shift by around 15% due to changes in measurement equipment (White, 2007). For these reasons, we investigate the net effect of assuming an EC coefficient of 1. We perform 10 sets of site- and quarter-specific EiV regressions in which we fix the coefficient for EC at various values (0, 0.25, 0.5, 0.75, 1.25, 1.5, 1.75, 2, 2.5, and 3). This analysis shows that when the EC coefficient is fixed between 0.25 and 1.75, most  $\beta_{OC}$  values change by less than 0.2 (see Fig. S3). When the EC coefficient is changed to 0 or 2,  $\beta_{OC}$  is affected substantially.

To explore this further, we repeat all of the site- and quarter-specific regressions using both EC and OC as explanatory variables (Eq. S4).

$$PM_{2.5,i} = \beta_{OC} OC_i + \beta_{sulf} (NH_4)_2 SO_{4,i} + \beta_{nit} NH_4 NO_{3,i} + \beta_{soil} SOIL_i + \beta_{EC} EC_i + 1.8 \times Cl_i^- + 1.2 \times KNON_i + \varepsilon_i \quad (S4)$$

Twenty five percent of the EC coefficients fall below -0.3 and 50% fall below 0.3. Such low coefficients are unrealistic and can cause substantial overestimates of  $\beta_{OC}$ . The results reported by Hand and Malm (2006) show the opposite effect with most EC coefficients exceeding one. About one quarter of their reported EC coefficients are greater than 3 and one is as high as 11. These EC coefficients appear to be unrealistically high and are likely an artifact of co-linear explanatory variables used in their OLS regression. Again, Fig. S3 demonstrates that high EC coefficients like those from Hand and Malm (2006) would cause drastic underestimates of  $\beta_{OC}$ .

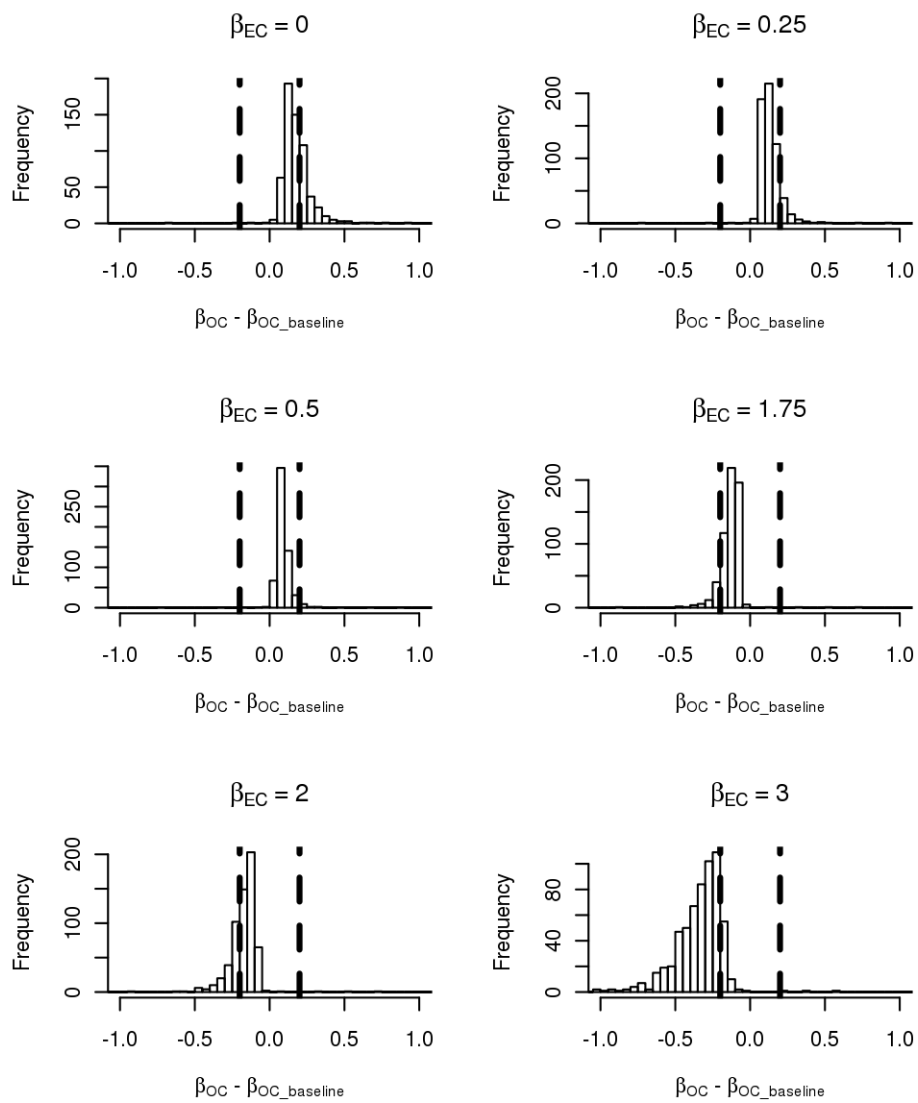


Figure S3. Change in  $\beta_{OC}$  when EC coefficient ( $\beta_{EC}$ ) is altered from the baseline value of 1 to other fixed values: 0, 0.25, 0.5, 1.75, 2, and 3. Vertical dashed lines encompass all site- and quarter-specific regressions in which  $\beta_{OC}$  deviates by less than 0.2 from the baseline results presented in Section 3.3.

We conduct a separate analysis to estimate the actual EC coefficients. To accomplish this, we perform a set of regressions in which we use TC instead of OC as our covariate (Eq. S5).

$$\begin{aligned}
 PM_{2.5,i} = & \beta_{TC} TC_i + \beta_{sulf} (NH_4)_2 SO_{4,i} + \beta_{nit} NH_4 NO_{3,i} + \beta_{soil} SOIL_i \\
 & + 1.8 \times Cl_i^- + 1.2 \times KNON_i + \varepsilon_i
 \end{aligned}
 \tag{S5}$$

We expect the actual coefficient for TC ( $\beta_{TC}$ ) to be an intermediate value between our original  $\beta_{OC}$  results and the actual EC coefficient. By applying Eq. (S5), we find that  $\beta_{TC}$  is very close to our original  $\beta_{OC}$  results for most site- and quarter-specific regressions. On average,  $\beta_{TC}$  is slightly lower than  $\beta_{OC}$  (see Fig. S4). Only 3% of the TC coefficients differ from our original  $\beta_{OC}$  values by more than 0.2. It may seem counter-intuitive that results using Eq. (S5) would be so similar to the original regression results, whereas including EC as a separate covariate (as in Eq. (S4)) has a much larger effect.

Assuming a maximum measurement artifact of 15%, we can set a lower bound for  $\beta_{EC}$  around 0.85. From this we can infer that  $0.85 < \beta_{EC} < \beta_{TC} < \beta_{OC}$ . Over 80% of the estimated TC coefficients from Eq. (S5) fall in the range of 1.2 to 1.9. It follows that the true EC coefficients lie between 0.85 and 1.9. Consequently, EC coefficients in this analysis are much closer to 1 than the EC coefficients estimated by treating EC as a separate explanatory variable. Combining this analysis with the results shown in Fig. S3, we conclude that our assumption of an EC coefficient equal to 1 does not greatly bias our  $\beta_{OC}$  results.

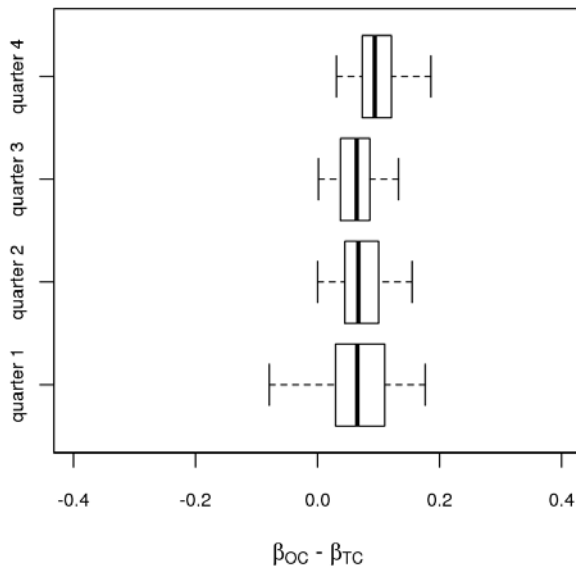


Figure S4: Comparison of our baseline OC coefficients from Eq. (5) to TC coefficients obtained using Eq. (S5).

As mentioned in Sect. 3.3,  $\beta_{OC}$  is influenced by differences in the OC sampling artifacts on quartz versus Teflon filters. Whereas the literature is inconclusive regarding negative artifacts, quartz filters are more prone to positive artifact than Teflon filters. The IMPROVE data include a network-wide and month-specific correction for positive OC artifact on the quartz filter, but no correction for the Teflon filter. Quartz-behind-quartz backup filters are collected at six IMPROVE sites (Chiricahua, Grand Canyon, Mount Rainier, Okefenokee, Shenandoah, and Yosemite). Each month, the median of all quartz-behind-quartz backup filters from these six sites is used as a network-wide average value for positive OC artifact. The reported OC concentrations are calculated by subtracting the median artifact value for that month ( $\mu\text{g}/\text{filter}$ ) from each OC sample at all sites ( $\mu\text{g}/\text{filter}$ ) before converting filter measurements to ambient concentrations of  $\mu\text{g}/\text{m}^3$  (McDade, 2008). Here we evaluate the effect of using a single median artifact at all IMPROVE sites.

Since backup filters are only collected at 6 monitoring sites, it is not possible to determine how much site-to-site variability occurs network-wide. However, we perform a sensitivity study in which we look at site-to-site variability in back-up filter concentrations within the six sites used to create the median OC artifact value. For this analysis, all OC values for these six sites are recalculated using sample-specific backup filter values instead of the network-wide monthly median. We repeat the EiV regression analysis using these new sample-specific-corrected OC values and evaluate changes in  $\beta_{OC}$ . These results are shown in Fig. S5. In all regressions, changes in  $\beta_{OC}$  values are modest, with the average change being 0.05 (3%) and the maximum change being 0.14 (9%). Although it is not known how representative these six sites are of the network as a whole, this analysis suggests that using a single artifact correction network-wide does not substantially affect our estimations of  $\beta_{OC}$ .

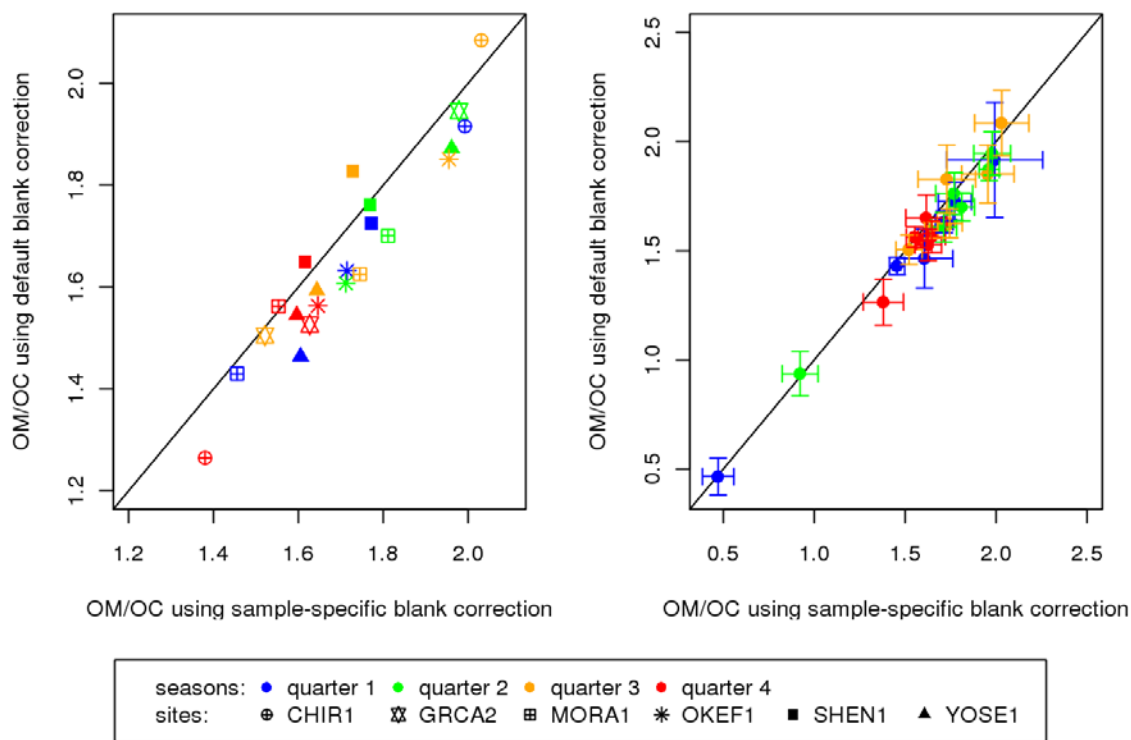
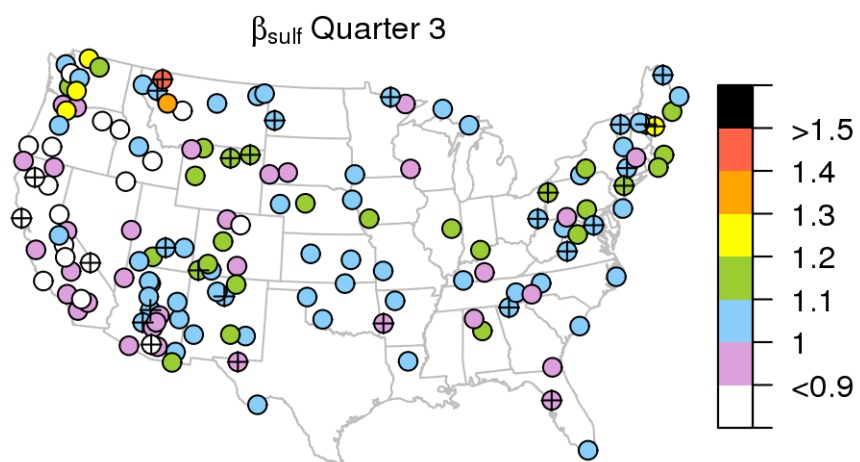
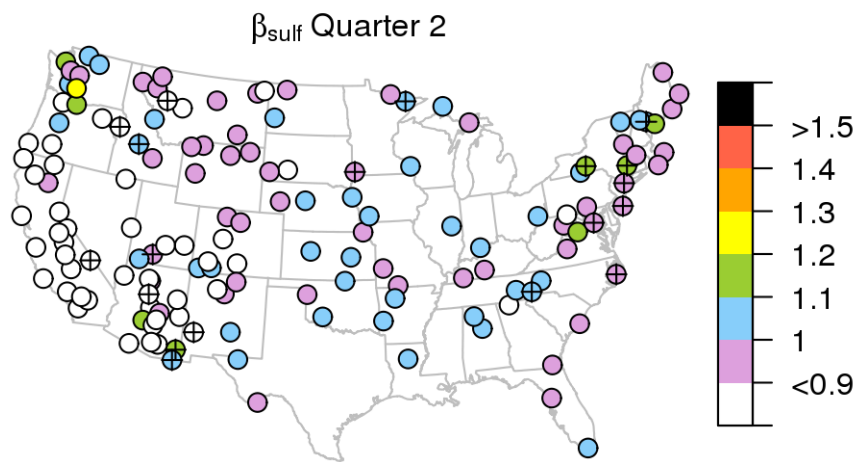
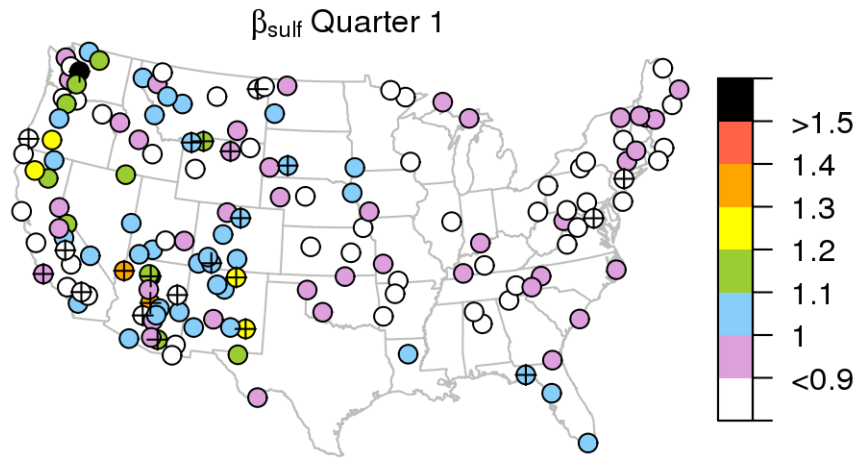
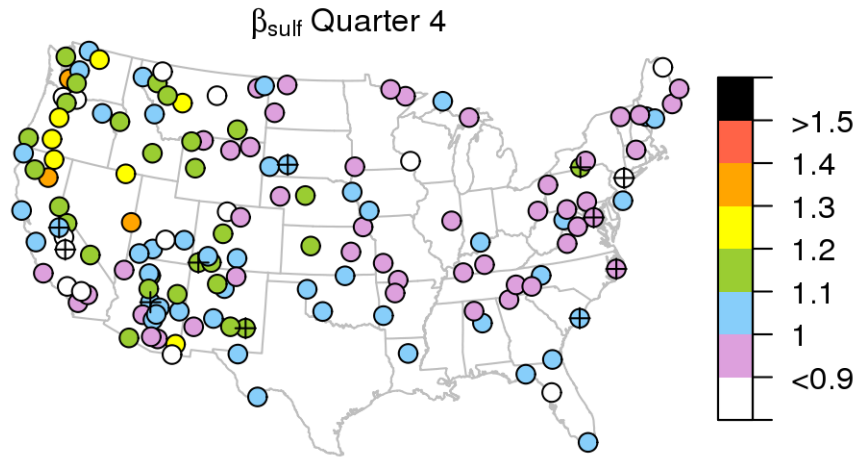


Figure S5. Comparison of  $\beta_{OC}$  values when using default artifact correction versus sample-specific artifact correction for only good regressions (left) and for all quarter-specific regressions (right). Uncertainty bars in the right-hand plot are standard error values for  $\beta_{OC}$  at each site and quarter.

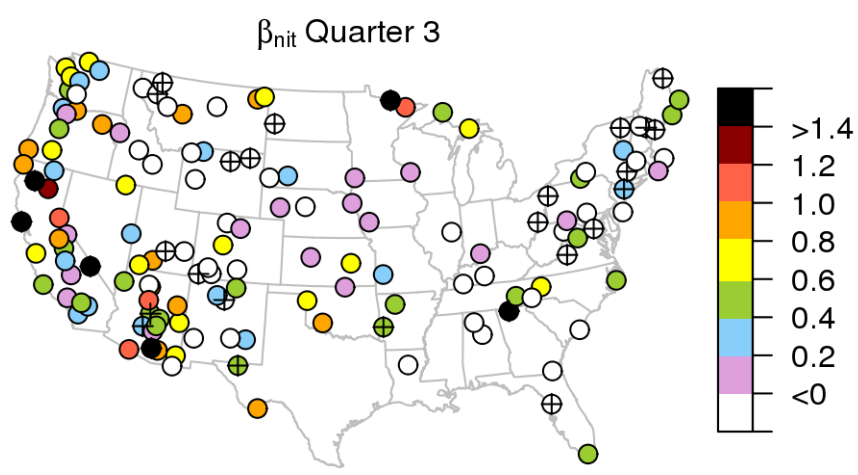
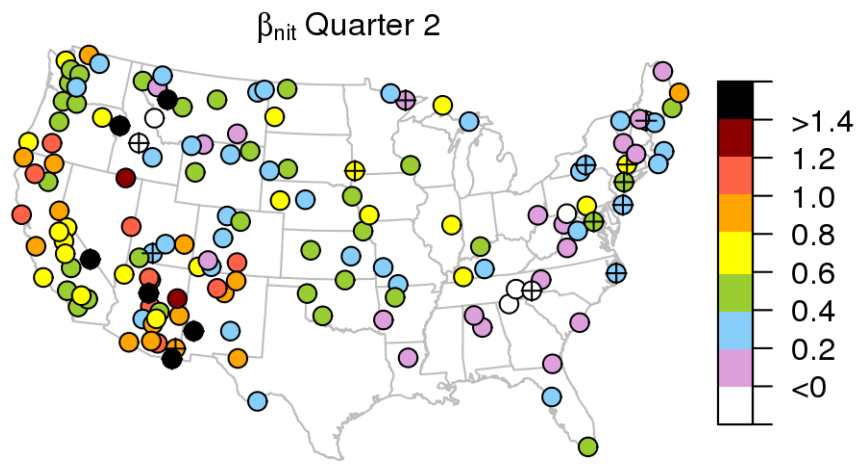
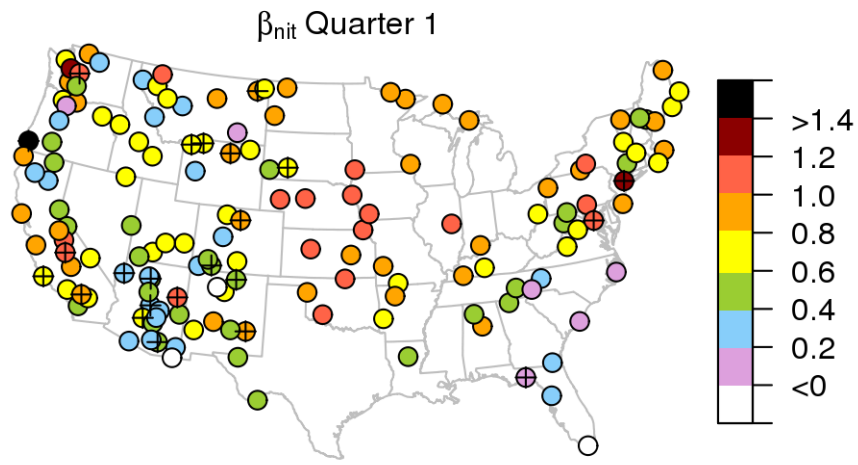
S4. Maps of regression coefficients

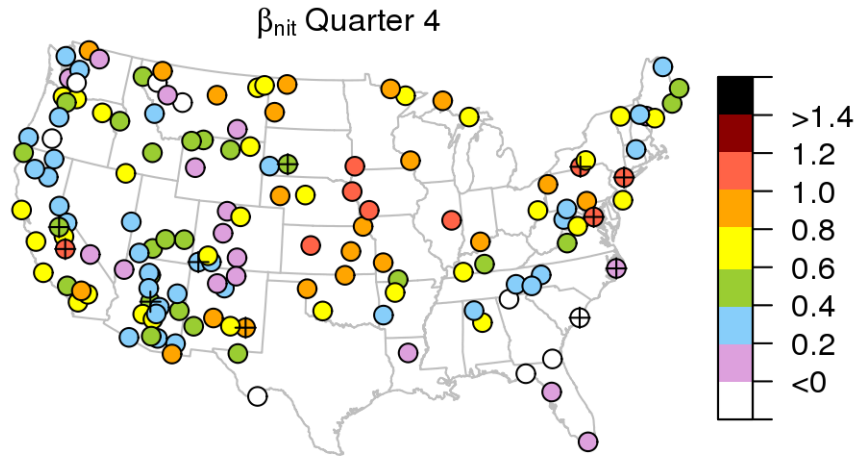




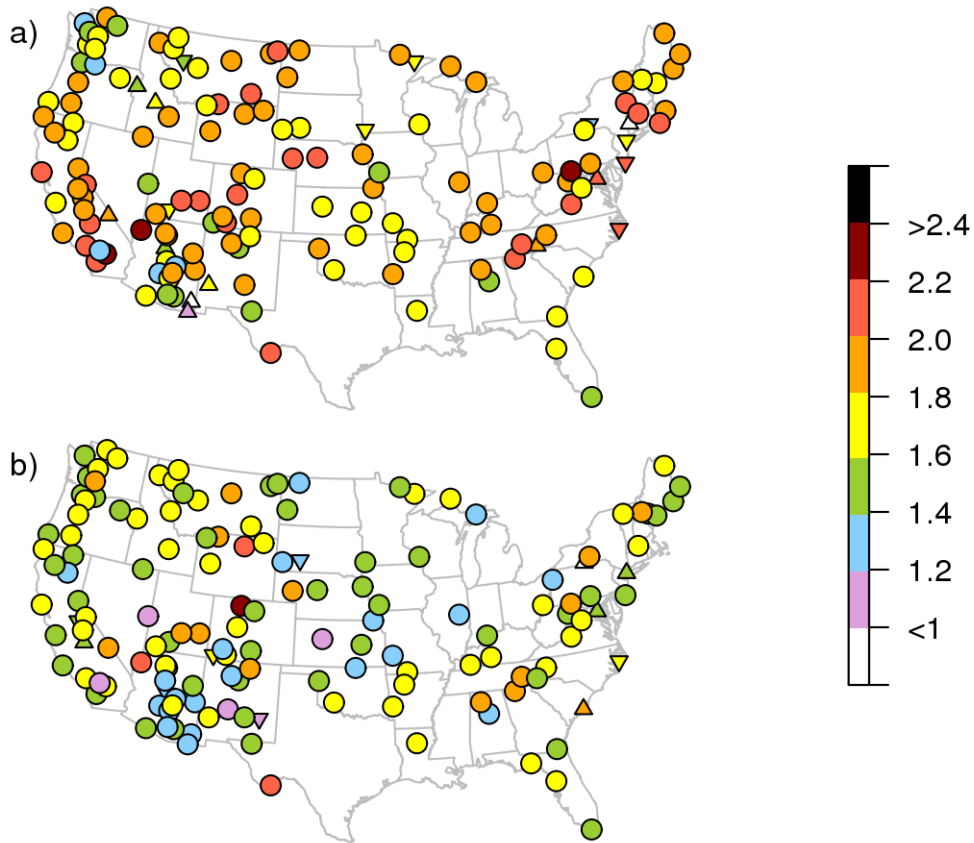
Figures S6-S9: Maps of sulfate coefficients in quarters 1-4. High confidence results are plotted with colored dots. Regressions that were flagged for problematic coefficients or temporal trends in the residual errors are marked with crosses or black dots.







Figures S10-S13: Maps of nitrate coefficients in quarters 1-4. High confidence results are plotted with colored dots. Regressions that were flagged for problematic coefficients or temporal trends in the residuals are marked with crosses or black dots.



Figures S14:  $\beta_{OC}$  values for quarter 2 (top) and quarter 4 (bottom). High confidence results are depicted by circles, regressions with questionable residual trends are depicted by downward facing triangles, and regressions with any physically unreasonable coefficient are depicted by upward facing triangles..

S5. Tabulated regression results

Table S2. Multiyear regression results. Physically unreasonable coefficients are shown in bold.

site	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	$\beta_{soil}$
Acadia NP	1.82 +/- 0.04	1.05 +/- 0.01	0.13 +/- 0.06	0.95 +/- 0.11
Addison Pinnacle	1.44 +/- 0.07	1.10 +/- 0.02	0.58 +/- 0.04	1.24 +/- 0.18
Agua Tibia	1.74 +/- 0.05	0.97 +/- 0.02	0.52 +/- 0.02	0.98 +/- 0.04
Arendtsville	1.63 +/- 0.06	1.06 +/- 0.02	0.77 +/- 0.02	0.49 +/- 0.13
Badlands NP	1.60 +/- 0.04	0.97 +/- 0.03	0.30 +/- 0.05	0.98 +/- 0.05
Bandelier NM	1.48 +/- 0.04	1.05 +/- 0.03	0.47 +/- 0.04	1.11 +/- 0.02
Big Bend NP	1.92 +/- 0.07	1.00 +/- 0.01	0.32 +/- 0.08	1.07 +/- 0.02
Birmingham	1.39 +/- 0.04	1.08 +/- 0.02	0.64 +/- 0.06	1.02 +/- 0.04
Bliss SP (TRPA)	1.76 +/- 0.02	0.95 +/- 0.03	0.38 +/- 0.05	1.01 +/- 0.03
Blue Mounds	1.70 +/- 0.05	0.93 +/- 0.02	1.05 +/- 0.01	0.79 +/- 0.05
Bondville	1.65 +/- 0.07	1.10 +/- 0.02	0.85 +/- 0.02	0.82 +/- 0.09
Bosque del Apache	1.28 +/- 0.05	0.96 +/- 0.02	0.79 +/- 0.04	1.06 +/- 0.02
Boundary Waters Canoe Area	1.80 +/- 0.04	0.95 +/- 0.02	0.77 +/- 0.02	0.82 +/- 0.10
Bridger Wilderness	1.85 +/- 0.03	0.97 +/- 0.03	0.23 +/- 0.09	1.01 +/- 0.03
Bridgton	1.76 +/- 0.04	1.07 +/- 0.02	-0.05 +/- 0.07	1.09 +/- 0.13
Brigantine NWR	1.79 +/- 0.06	1.03 +/- 0.02	0.52 +/- 0.04	0.79 +/- 0.14
Bryce Canyon NP	1.52 +/- 0.04	1.04 +/- 0.04	0.50 +/- 0.05	1.08 +/- 0.03
Cabinet Mountains	1.76 +/- 0.02	1.02 +/- 0.03	0.17 +/- 0.07	1.11 +/- 0.03
Cadiz	1.68 +/- 0.05	1.04 +/- 0.01	0.71 +/- 0.02	0.96 +/- 0.05
Caney Creek	1.75 +/- 0.04	0.99 +/- 0.01	0.44 +/- 0.03	0.98 +/- 0.02
Canyonlands NP	2.03 +/- 0.05	0.91 +/- 0.03	0.40 +/- 0.04	1.18 +/- 0.02
Cape Cod	1.78 +/- 0.05	1.03 +/- 0.02	0.18 +/- 0.06	0.99 +/- 0.13
Cape Romain NWR	1.69 +/- 0.04	1.02 +/- 0.01	<b>-0.27 +/- 0.10</b>	1.00 +/- 0.05
Capitol Reef NP	2.03 +/- 0.05	0.83 +/- 0.04	0.40 +/- 0.04	1.14 +/- 0.02
Casco Bay	1.56 +/- 0.03	1.17 +/- 0.02	0.18 +/- 0.07	1.03 +/- 0.10
Cedar Bluff	1.43 +/- 0.09	1.05 +/- 0.04	0.88 +/- 0.02	1.02 +/- 0.06
Chassahowitzka NWR	1.60 +/- 0.04	1.00 +/- 0.02	0.17 +/- 0.10	1.01 +/- 0.03
Cherokee Nation	1.49 +/- 0.04	1.06 +/- 0.02	0.87 +/- 0.02	0.97 +/- 0.02
Chiricahua NM	1.45 +/- 0.07	1.15 +/- 0.02	0.25 +/- 0.10	1.11 +/- 0.02
Cloud Peak	1.94 +/- 0.03	0.97 +/- 0.03	0.31 +/- 0.08	1.05 +/- 0.03
Cohutta	1.88 +/- 0.06	0.96 +/- 0.01	0.12 +/- 0.05	0.82 +/- 0.05
Columbia Gorge #1	1.57 +/- 0.03	0.85 +/- 0.03	0.57 +/- 0.03	1.13 +/- 0.06
Columbia River Gorge	1.47 +/- 0.03	0.96 +/- 0.04	0.62 +/- 0.02	1.05 +/- 0.02
Connecticut Hill	1.53 +/- 0.08	1.07 +/- 0.02	0.61 +/- 0.04	0.69 +/- 0.15
Crater Lake NP	1.71 +/- 0.02	1.08 +/- 0.03	0.15 +/- 0.10	0.95 +/- 0.03
Craters of the Moon NM	1.85 +/- 0.03	0.88 +/- 0.04	0.50 +/- 0.02	1.04 +/- 0.02

site	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	$\beta_{soil}$
Crescent Lake	1.97 +/- 0.05	0.92 +/- 0.03	0.92 +/- 0.02	0.86 +/- 0.05
Death Valley NP	1.83 +/- 0.05	0.87 +/- 0.03	0.70 +/- 0.09	1.16 +/- 0.02
Dolly Sods Wilderness	1.51 +/- 0.05	1.06 +/- 0.01	0.28 +/- 0.05	1.03 +/- 0.08
Dome Lands Wilderness	1.79 +/- 0.06	0.77 +/- 0.05	0.70 +/- 0.02	1.12 +/- 0.05
Douglas	1.31 +/- 0.08	1.04 +/- 0.04	0.60 +/- 0.15	1.02 +/- 0.01
El Dorado Springs	1.44 +/- 0.04	1.04 +/- 0.01	0.77 +/- 0.02	1.07 +/- 0.03
Ellis	1.59 +/- 0.05	1.06 +/- 0.02	0.86 +/- 0.02	0.93 +/- 0.03
Everglades NP	1.58 +/- 0.04	1.05 +/- 0.02	0.31 +/- 0.10	1.02 +/- 0.02
Flathead	1.74 +/- 0.02	0.97 +/- 0.03	0.17 +/- 0.06	1.01 +/- 0.04
Fort Peck	1.86 +/- 0.04	0.88 +/- 0.02	0.68 +/- 0.02	0.96 +/- 0.04
Frostberg Reservoir (Big Piney Run)	1.95 +/- 0.06	0.94 +/- 0.01	0.24 +/- 0.04	0.96 +/- 0.08
Gates of the Mountains	1.73 +/- 0.02	1.02 +/- 0.03	0.15 +/- 0.07	1.02 +/- 0.04
Gila Wilderness	1.49 +/- 0.03	0.96 +/- 0.03	0.77 +/- 0.18	1.10 +/- 0.02
Glacier NP	1.72 +/- 0.02	0.88 +/- 0.03	0.79 +/- 0.06	0.91 +/- 0.03
Great Basin NP	1.66 +/- 0.04	1.01 +/- 0.04	0.13 +/- 0.10	1.11 +/- 0.02
Great Gulf Wilderness	1.83 +/- 0.04	1.02 +/- 0.02	0.06 +/- 0.07	0.82 +/- 0.12
Great River Bluffs	1.67 +/- 0.06	0.94 +/- 0.02	0.86 +/- 0.01	0.87 +/- 0.13
Great Sand Dunes NM	1.83 +/- 0.05	0.96 +/- 0.04	0.18 +/- 0.08	1.02 +/- 0.01
Great Smoky Mountains NP	1.86 +/- 0.05	1.05 +/- 0.01	0.18 +/- 0.05	1.06 +/- 0.08
Guadalupe Mountains NP	1.60 +/- 0.09	1.02 +/- 0.03	0.53 +/- 0.05	1.04 +/- 0.01
Hance Camp at Grand Canyon NP	1.54 +/- 0.04	1.15 +/- 0.03	0.57 +/- 0.06	1.11 +/- 0.02
Hells Canyon	1.67 +/- 0.02	0.89 +/- 0.04	0.64 +/- 0.02	1.01 +/- 0.04
Hercules-Glades	1.64 +/- 0.04	0.99 +/- 0.01	0.58 +/- 0.02	1.00 +/- 0.03
Hoover	1.80 +/- 0.03	0.94 +/- 0.04	0.37 +/- 0.08	1.20 +/- 0.03
Ikes Backbone	1.30 +/- 0.05	1.03 +/- 0.04	0.40 +/- 0.05	1.19 +/- 0.02
Indian Gardens	1.71 +/- 0.05	0.99 +/- 0.03	0.36 +/- 0.08	1.11 +/- 0.02
Isle Royale NP	1.95 +/- 0.04	1.03 +/- 0.02	0.81 +/- 0.02	0.61 +/- 0.13
James River Face Wilderness	1.72 +/- 0.04	1.04 +/- 0.01	0.29 +/- 0.05	0.92 +/- 0.08
Jarbidge Wilderness	1.84 +/- 0.04	0.94 +/- 0.03	0.72 +/- 0.05	0.99 +/- 0.02
Joshua Tree NP	1.92 +/- 0.07	0.87 +/- 0.03	0.62 +/- 0.02	0.93 +/- 0.03
Kaiser	1.83 +/- 0.03	0.76 +/- 0.04	0.72 +/- 0.03	0.99 +/- 0.03
Kalmiopsis	1.53 +/- 0.02	0.93 +/- 0.04	0.93 +/- 0.15	1.06 +/- 0.08
Lassen Volcanic NP	1.67 +/- 0.03	1.06 +/- 0.04	0.31 +/- 0.06	1.06 +/- 0.04
Lava Beds NM	1.68 +/- 0.03	1.04 +/- 0.07	0.25 +/- 0.11	1.04 +/- 0.07
Linville Gorge	1.78 +/- 0.04	1.10 +/- 0.01	-0.07 +/- 0.07	0.83 +/- 0.08
Livonia	1.66 +/- 0.07	1.08 +/- 0.02	0.71 +/- 0.02	1.06 +/- 0.07
Lostwood	1.81 +/- 0.04	0.91 +/- 0.02	0.80 +/- 0.02	0.99 +/- 0.05
Lye Brook Wilderness	1.94 +/- 0.06	1.00 +/- 0.02	0.31 +/- 0.04	0.79 +/- 0.12
M.K. Goddard	1.50 +/- 0.05	1.08 +/- 0.02	0.69 +/- 0.03	0.93 +/- 0.15
Mammoth Cave NP	1.88 +/- 0.05	0.93 +/- 0.01	0.42 +/- 0.02	0.91 +/- 0.05
Marthas Vineyard	1.87 +/- 0.06	1.04 +/- 0.01	0.18 +/- 0.06	0.93 +/- 0.12
Meadview	1.84 +/- 0.07	0.99 +/- 0.03	0.32 +/- 0.06	1.10 +/- 0.02

site	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	$\beta_{soil}$
Medicine Lake	1.84 +/- 0.04	0.91 +/- 0.02	0.69 +/- 0.02	0.99 +/- 0.04
Mesa Verde NP	1.79 +/- 0.06	1.08 +/- 0.05	0.18 +/- 0.09	1.19 +/- 0.02
Mohawk Mt.	1.53 +/- 0.07	1.02 +/- 0.02	0.28 +/- 0.06	1.11 +/- 0.19
Monture	1.67 +/- 0.02	1.01 +/- 0.03	0.22 +/- 0.13	1.08 +/- 0.03
Moosehorn NWR	1.70 +/- 0.04	1.02 +/- 0.02	0.23 +/- 0.07	0.70 +/- 0.14
Mount Baldy	1.44 +/- 0.03	1.05 +/- 0.02	0.55 +/- 0.06	1.09 +/- 0.02
Mount Hood	1.79 +/- 0.03	1.17 +/- 0.03	0.21 +/- 0.06	1.00 +/- 0.06
Mount Rainier NP	1.59 +/- 0.03	1.20 +/- 0.04	0.38 +/- 0.13	1.16 +/- 0.09
Mount Zirkel Wilderness	2.02 +/- 0.04	0.82 +/- 0.03	0.33 +/- 0.06	1.04 +/- 0.03
Nebraska NF	1.91 +/- 0.07	0.98 +/- 0.03	0.77 +/- 0.02	0.84 +/- 0.07
New York City	1.62 +/- 0.09	1.01 +/- 0.03	0.91 +/- 0.04	<b>0.27 +/- 0.19</b>
North Absaroka	1.91 +/- 0.03	0.97 +/- 0.03	0.32 +/- 0.06	1.11 +/- 0.03
North Cascades	1.79 +/- 0.03	1.10 +/- 0.03	0.75 +/- 0.14	0.89 +/- 0.07
Northern Cheyenne	1.91 +/- 0.03	1.03 +/- 0.03	0.10 +/- 0.05	0.96 +/- 0.04
Okefenokee NWR	1.65 +/- 0.03	0.98 +/- 0.01	0.04 +/- 0.10	0.97 +/- 0.03
Olympic	1.53 +/- 0.03	1.11 +/- 0.03	0.47 +/- 0.06	0.99 +/- 0.10
Omaha	1.75 +/- 0.06	1.00 +/- 0.02	0.97 +/- 0.02	0.67 +/- 0.07
Organ Pipe	1.45 +/- 0.08	0.99 +/- 0.02	0.44 +/- 0.07	1.09 +/- 0.02
Pasayten	1.69 +/- 0.02	1.08 +/- 0.03	0.19 +/- 0.06	1.09 +/- 0.05
Petrified Forest NP	1.66 +/- 0.05	1.05 +/- 0.03	0.44 +/- 0.09	1.04 +/- 0.02
Phoenix	1.25 +/- 0.02	0.95 +/- 0.03	0.64 +/- 0.03	1.08 +/- 0.02
Pinnacles NM	1.69 +/- 0.06	0.93 +/- 0.05	0.65 +/- 0.05	1.05 +/- 0.13
Point Reyes National Seashore	1.58 +/- 0.07	0.95 +/- 0.03	0.78 +/- 0.03	1.03 +/- 0.17
Presque Isle	1.79 +/- 0.03	0.94 +/- 0.01	-0.01 +/- 0.05	0.94 +/- 0.03
Proctor Maple R. F.	1.86 +/- 0.04	1.01 +/- 0.01	0.46 +/- 0.04	0.69 +/- 0.13
Puget Sound	1.39 +/- 0.03	0.85 +/- 0.03	1.00 +/- 0.04	0.73 +/- 0.09
Quabbin Summit	1.76 +/- 0.04	0.96 +/- 0.01	0.30 +/- 0.04	0.85 +/- 0.11
Quaker City	1.64 +/- 0.06	1.07 +/- 0.01	0.55 +/- 0.03	0.86 +/- 0.09
Queen Valley	1.51 +/- 0.07	1.01 +/- 0.03	0.57 +/- 0.02	1.08 +/- 0.02
Redwood NP	1.73 +/- 0.03	0.95 +/- 0.03	0.80 +/- 0.09	0.88 +/- 0.09
Rocky Mountain NP	1.84 +/- 0.05	0.81 +/- 0.05	0.56 +/- 0.04	1.05 +/- 0.03
Sac and Fox	1.56 +/- 0.05	0.98 +/- 0.02	0.90 +/- 0.01	0.93 +/- 0.05
Saguaro NM	1.35 +/- 0.06	1.00 +/- 0.03	0.38 +/- 0.04	1.14 +/- 0.01
Saguaro West	1.35 +/- 0.11	0.99 +/- 0.04	0.37 +/- 0.05	1.12 +/- 0.02
Salt Creek	1.42 +/- 0.10	1.02 +/- 0.03	0.94 +/- 0.04	1.10 +/- 0.02
San Gabriel	1.86 +/- 0.05	0.81 +/- 0.03	0.50 +/- 0.02	1.02 +/- 0.04
San Geronio Wilderness	1.46 +/- 0.06	0.83 +/- 0.04	0.77 +/- 0.01	0.96 +/- 0.04
San Pedro Parks	1.62 +/- 0.05	1.02 +/- 0.03	0.18 +/- 0.11	1.16 +/- 0.02
San Rafael	1.70 +/- 0.05	0.95 +/- 0.03	0.53 +/- 0.03	1.06 +/- 0.05
Sawtooth NF	1.65 +/- 0.03	1.01 +/- 0.07	<b>-1.79 +/- 0.50</b>	1.21 +/- 0.05
Seney	1.66 +/- 0.04	0.97 +/- 0.02	0.70 +/- 0.02	0.78 +/- 0.14
Sequoia NP	1.68 +/- 0.04	0.79 +/- 0.05	0.90 +/- 0.01	1.09 +/- 0.06

site	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	$\beta_{soil}$
Shamrock Mine	1.92 +/- 0.04	1.05 +/- 0.03	0.11 +/- 0.06	0.93 +/- 0.01
Shenandoah NP	1.73 +/- 0.06	1.10 +/- 0.01	0.36 +/- 0.04	1.02 +/- 0.08
Shining Rock Wilderness	1.80 +/- 0.08	1.02 +/- 0.02	<b>-0.27 +/- 0.12</b>	0.95 +/- 0.07
Sierra Ancha	1.30 +/- 0.04	1.04 +/- 0.03	0.29 +/- 0.06	1.13 +/- 0.02
Sikes	1.71 +/- 0.03	1.06 +/- 0.01	0.14 +/- 0.05	1.03 +/- 0.02
Sipsy Wilderness	1.85 +/- 0.04	0.98 +/- 0.01	0.23 +/- 0.03	0.97 +/- 0.04
Snoqualmie Pass	1.64 +/- 0.05	1.09 +/- 0.05	0.43 +/- 0.06	0.98 +/- 0.15
St. Marks	1.65 +/- 0.04	1.04 +/- 0.01	0.15 +/- 0.12	1.02 +/- 0.03
Starkey	1.63 +/- 0.02	0.90 +/- 0.04	0.70 +/- 0.02	1.15 +/- 0.03
Sula Peak	1.73 +/- 0.02	0.99 +/- 0.04	-0.07 +/- 0.09	0.99 +/- 0.03
Swanquarter	1.80 +/- 0.05	1.01 +/- 0.01	0.01 +/- 0.06	1.06 +/- 0.05
Sycamore Canyon	1.29 +/- 0.04	1.13 +/- 0.04	0.47 +/- 0.06	1.04 +/- 0.01
Tallgrass	1.44 +/- 0.04	1.04 +/- 0.02	0.80 +/- 0.02	1.00 +/- 0.04
Theodore Roosevelt	1.83 +/- 0.04	0.96 +/- 0.03	0.80 +/- 0.03	0.91 +/- 0.03
Three Sisters Wilderness	1.75 +/- 0.02	1.07 +/- 0.03	0.35 +/- 0.10	0.99 +/- 0.04
Thunder Basin	1.86 +/- 0.03	0.94 +/- 0.02	0.54 +/- 0.03	0.92 +/- 0.02
Tonto NM	1.71 +/- 0.05	1.00 +/- 0.03	0.27 +/- 0.04	1.08 +/- 0.01
Trinity	1.63 +/- 0.03	1.08 +/- 0.05	0.40 +/- 0.06	0.94 +/- 0.06
UL Bend	1.91 +/- 0.03	0.86 +/- 0.02	0.81 +/- 0.03	1.04 +/- 0.03
Upper Buffalo Wilderness	1.63 +/- 0.04	1.04 +/- 0.01	0.68 +/- 0.02	1.01 +/- 0.03
Viking Lake	1.54 +/- 0.05	1.05 +/- 0.02	0.99 +/- 0.01	0.91 +/- 0.06
Voyageurs NP #2	1.70 +/- 0.04	0.94 +/- 0.02	0.87 +/- 0.02	0.97 +/- 0.13
Washington D.C.	1.67 +/- 0.06	1.07 +/- 0.02	0.78 +/- 0.03	<b>0.21 +/- 0.14</b>
Weminuche Wilderness	1.78 +/- 0.04	1.00 +/- 0.04	<b>-0.19 +/- 0.13</b>	1.09 +/- 0.02
Wheeler Peak	1.68 +/- 0.06	1.08 +/- 0.04	0.17 +/- 0.13	1.24 +/- 0.03
White Mountain	1.63 +/- 0.06	1.08 +/- 0.03	0.55 +/- 0.04	1.13 +/- 0.02
White Pass	1.82 +/- 0.04	1.18 +/- 0.04	0.14 +/- 0.09	0.87 +/- 0.06
White River NF	1.96 +/- 0.05	1.08 +/- 0.04	<b>-0.18 +/- 0.11</b>	1.17 +/- 0.02
Wichita Mountains	1.53 +/- 0.05	1.10 +/- 0.02	0.81 +/- 0.02	0.95 +/- 0.03
Wind Cave	1.72 +/- 0.03	0.93 +/- 0.03	0.41 +/- 0.03	1.10 +/- 0.03
Yellowstone NP 2	1.75 +/- 0.02	0.87 +/- 0.03	0.52 +/- 0.04	1.08 +/- 0.03
Yosemite NP	1.64 +/- 0.02	1.01 +/- 0.03	0.77 +/- 0.03	1.09 +/- 0.05
Zion Canyon	1.76 +/- 0.05	1.11 +/- 0.03	0.28 +/- 0.05	1.15 +/- 0.02

Table S3. Quarterly regressions eliminated because of high colinearity among covariates

site	quarter	max $ r_p $ among covariate pairs	correlated covariates
Northern Cheyenne	3	0.86	sulfate:nitrate
Sula Peak	3	0.87	oc:nitrate
Cape Cod	4	0.90	sulfate:soil
Lye Brook Wilderness	4	0.86 : 0.86	oc:sulfate ; soil:sulfate
Marthas Vineyard	4	0.85	soil:sulfate
Mohawk Mt.	4	0.88	soil:nitrate
Puget Sound	4	0.86	sulfate:nitrate

Table S4. Quarterly regressions eliminated because of high correlation between residual error ( $\epsilon_i$ ) and a  $PM_{2.5}$  component ( $|r_s| > 0.4$ ).

site	quarter	Max $ r_s $ between $\epsilon_i$ and $PM_{2.5}$ constituents	$PM_{2.5}$ constituents correlated to $\epsilon_i$
Bosque del Apache	2	0.45	chloride
M.K. Goddard	2	0.41	chloride
Mount Hood	2	0.44 ; 0.46	OC ; EC
Salt Creek	2	0.49	chloride
St. Marks	2	0.53	chloride
Bosque del Apache	3	0.48	chloride
Hercules-Glades	3	0.41	chloride
Lostwood	3	0.45	chloride
Sac and Fox	3	0.44	chloride
St. Marks	3	0.50	chloride
Dome Lands Wilderness	4	0.44	soil
Sawtooth NF	4	0.42	KNON



Table S5. High-confidence quarter-specific regression results. This table includes 10 regressions flagged for an outlier year, but excluding that year did not change the regression coefficients (see Table S6).

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Acadia NP	1	1.66 +/- 0.14	0.89 +/- 0.04	0.62 +/- 0.11	7.38	-0.21
Addison Pinnacle	1	1.46 +/- 0.11	0.82 +/- 0.03	0.98 +/- 0.05	5.93	-0.12
Agua Tibia	1	1.38 +/- 0.12	1.08 +/- 0.08	0.56 +/- 0.04	9.52	0.07
Arendtsville	1	1.21 +/- 0.09	0.88 +/- 0.03	1.18 +/- 0.04	6.15	-0.38
Bandelier NM	1	1.15 +/- 0.08	1.05 +/- 0.06	0.71 +/- 0.06	10.48	-0.30
Big Bend NP	1	1.83 +/- 0.12	0.97 +/- 0.03	0.40 +/- 0.08	6.88	0.04
Birmingham	1	1.50 +/- 0.05	0.87 +/- 0.04	0.82 +/- 0.07	5.88	-0.14
Bliss SP (TRPA)	1	1.65 +/- 0.09	0.97 +/- 0.08	0.40 +/- 0.08	14.37	-1.83
Blue Mounds	1	1.53 +/- 0.14	1.00 +/- 0.04	1.08 +/- 0.02	5.24	0.12
Bondville	1	1.44 +/- 0.14	0.89 +/- 0.04	1.03 +/- 0.03	6.41	-0.19
Bosque del Apache	1	1.05 +/- 0.09	0.99 +/- 0.06	0.91 +/- 0.06	9.64	-0.08
Boundary Waters Canoe Area	1	1.62 +/- 0.25	0.87 +/- 0.06	0.88 +/- 0.04	9.16	0.59
Bridger Wilderness	1	1.81 +/- 0.19	0.89 +/- 0.06	0.36 +/- 0.12	13.66	-0.46
Bridgton	1	1.48 +/- 0.09	0.98 +/- 0.03	0.59 +/- 0.11	7.59	-0.04
Brigantine NWR	1	1.44 +/- 0.10	0.88 +/- 0.03	0.94 +/- 0.06	5.90	-0.10
Bryce Canyon NP	1	1.11 +/- 0.17	1.05 +/- 0.09	0.65 +/- 0.06	16.44	-1.81
Cabinet Mountains	1	1.48 +/- 0.08	1.03 +/- 0.05	0.38 +/- 0.12	14.49	1.49
Cadiz	1	1.47 +/- 0.09	0.91 +/- 0.03	0.93 +/- 0.03	7.14	-0.33
Caney Creek	1	1.64 +/- 0.07	0.89 +/- 0.03	0.62 +/- 0.04	7.95	-0.80
Canyonlands NP	1	1.31 +/- 0.25	0.95 +/- 0.07	0.65 +/- 0.08	11.49	-0.13
Cape Cod	1	1.62 +/- 0.14	0.85 +/- 0.03	0.80 +/- 0.10	6.38	-0.08
Cape Romain NWR	1	1.60 +/- 0.05	0.99 +/- 0.03	0.04 +/- 0.14	6.68	-0.30
Capitol Reef NP	1	1.37 +/- 0.28	0.82 +/- 0.12	0.62 +/- 0.10	15.98	-1.60
Casco Bay	1	1.38 +/- 0.06	0.95 +/- 0.04	0.95 +/- 0.12	7.48	-0.63
Cedar Bluff	1	1.18 +/- 0.13	0.87 +/- 0.07	1.01 +/- 0.03	9.92	-1.06

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Chassahowitzka NWR	1	1.46 +/- 0.06	1.07 +/- 0.03	0.25 +/- 0.18	6.49	0.12
Cherokee Nation	1	1.32 +/- 0.08	0.94 +/- 0.05	1.00 +/- 0.03	7.72	-0.45
Chiricahua NM	1	1.99 +/- 0.25	0.88 +/- 0.09	0.29 +/- 0.16	10.11	-0.49
Cohutta	1	1.84 +/- 0.06	0.86 +/- 0.02	0.41 +/- 0.05	6.50	-0.10
Columbia Gorge #1	1	1.41 +/- 0.06	0.79 +/- 0.06	0.62 +/- 0.04	9.11	-0.89
Columbia River Gorge	1	1.00 +/- 0.08	0.79 +/- 0.09	0.82 +/- 0.04	12.37	0.66
Connecticut Hill	1	1.59 +/- 0.18	0.75 +/- 0.04	1.04 +/- 0.05	6.13	-0.25
Crater Lake NP	1	1.18 +/- 0.11	1.23 +/- 0.08	0.45 +/- 0.17	17.40	-2.30
Craters of the Moon NM	1	1.24 +/- 0.24	0.82 +/- 0.10	0.67 +/- 0.04	17.40	-0.57
Crescent Lake	1	1.31 +/- 0.20	0.95 +/- 0.07	1.04 +/- 0.03	9.64	-0.61
Death Valley NP	1	1.42 +/- 0.23	1.02 +/- 0.11	0.76 +/- 0.14	11.50	1.20
Dolly Sods Wilderness	1	1.38 +/- 0.07	0.99 +/- 0.03	0.57 +/- 0.06	7.56	-0.05
Dome Lands Wilderness	1	1.21 +/- 0.12	0.69 +/- 0.10	0.86 +/- 0.03	10.14	-0.11
Douglas	1	1.86 +/- 0.14	0.62 +/- 0.14	-0.06 +/- 0.25	6.33	0.27
El Dorado Springs	1	1.30 +/- 0.06	0.90 +/- 0.04	0.91 +/- 0.03	7.32	-0.20
Ellis	1	1.24 +/- 0.09	0.97 +/- 0.05	0.99 +/- 0.03	8.14	-1.08
Everglades NP	1	1.97 +/- 0.13	1.02 +/- 0.03	-0.10 +/- 0.20	7.86	0.23
Flathead	1	1.48 +/- 0.09	0.91 +/- 0.05	0.62 +/- 0.10	11.99	0.36
Frostberg Reservoir (Big Piney Run)	1	2.03 +/- 0.12	0.83 +/- 0.03	0.46 +/- 0.06	5.77	-0.12
Gates of the Mountains	1	1.26 +/- 0.11	1.08 +/- 0.05	0.32 +/- 0.10	15.05	0.20
Gila Wilderness	1	1.18 +/- 0.09	1.06 +/- 0.06	0.75 +/- 0.21	9.18	-0.02
Glacier NP	1	1.52 +/- 0.03	0.79 +/- 0.04	1.05 +/- 0.06	7.46	-0.68
Great Basin NP	1	1.05 +/- 0.10	1.06 +/- 0.13	0.49 +/- 0.15	13.34	-1.00
Great Gulf Wilderness	1	1.62 +/- 0.18	0.97 +/- 0.04	0.46 +/- 0.12	8.38	-0.31
Great River Bluffs	1	1.42 +/- 0.20	0.86 +/- 0.07	1.00 +/- 0.03	8.40	-0.28
Great Sand Dunes NM	1	1.15 +/- 0.12	1.01 +/- 0.07	0.74 +/- 0.09	10.27	-0.41
Great Smoky Mountains NP	1	1.85 +/- 0.06	0.87 +/- 0.02	0.54 +/- 0.05	6.33	0.02

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Guadalupe Mountains NP	1	1.29 +/- 0.28	1.14 +/- 0.13	0.53 +/- 0.10	11.30	1.00
Hells Canyon	1	1.33 +/- 0.10	0.97 +/- 0.09	0.72 +/- 0.03	13.60	-0.64
Hercules-Glades	1	1.61 +/- 0.07	0.87 +/- 0.04	0.74 +/- 0.04	8.07	-0.23
Hoover	1	1.42 +/- 0.17	1.17 +/- 0.08	0.43 +/- 0.09	15.29	0.01
Indian Gardens	1	1.20 +/- 0.16	1.15 +/- 0.10	0.32 +/- 0.08	10.96	1.37
Isle Royale NP	1	2.02 +/- 0.28	0.92 +/- 0.06	0.87 +/- 0.03	9.10	0.79
James River Face Wilderness	1	1.64 +/- 0.06	0.90 +/- 0.02	0.69 +/- 0.05	6.06	-0.10
Jarbidge Wilderness	1	1.12 +/- 0.18	1.17 +/- 0.09	0.69 +/- 0.08	16.68	0.89
Joshua Tree NP	1	1.26 +/- 0.16	0.79 +/- 0.10	0.78 +/- 0.03	10.58	0.46
Kaiser	1	1.06 +/- 0.13	1.02 +/- 0.09	1.00 +/- 0.05	12.89	-1.33
Lassen Volcanic NP	1	1.39 +/- 0.09	1.16 +/- 0.07	0.35 +/- 0.08	13.82	-1.05
Lava Beds NM	1	1.43 +/- 0.06	1.07 +/- 0.07	0.41 +/- 0.15	14.09	-1.13
Linville Gorge	1	1.80 +/- 0.05	0.92 +/- 0.02	0.33 +/- 0.08	6.82	-0.23
Livonia	1	1.58 +/- 0.09	0.92 +/- 0.03	0.92 +/- 0.02	6.01	-0.17
Lostwood	1	1.11 +/- 0.16	0.95 +/- 0.04	0.96 +/- 0.04	9.27	-0.92
Lye Brook Wilderness	1	1.60 +/- 0.23	0.87 +/- 0.05	0.77 +/- 0.07	9.20	-0.33
M.K. Goddard	1	1.57 +/- 0.09	0.79 +/- 0.03	0.98 +/- 0.04	6.55	-0.26
Mammoth Cave NP	1	1.79 +/- 0.07	0.77 +/- 0.02	0.67 +/- 0.03	6.77	-0.22
Marthas Vineyard	1	1.77 +/- 0.16	0.84 +/- 0.04	0.73 +/- 0.12	5.80	-0.29
Medicine Lake	1	1.59 +/- 0.19	0.89 +/- 0.04	0.76 +/- 0.04	10.64	-0.44
Mesa Verde NP	1	1.39 +/- 0.17	1.10 +/- 0.09	0.26 +/- 0.14	12.42	0.85
Mohawk Mt.	1	1.47 +/- 0.15	0.94 +/- 0.04	0.58 +/- 0.09	7.32	0.10
Monture	1	1.29 +/- 0.06	1.05 +/- 0.05	0.73 +/- 0.18	14.05	0.68
Moosehorn NWR	1	1.29 +/- 0.10	0.97 +/- 0.03	0.76 +/- 0.11	7.11	-0.07
Mount Baldy	1	1.31 +/- 0.05	1.02 +/- 0.04	0.60 +/- 0.07	9.14	0.06
Mount Hood	1	1.80 +/- 0.14	1.17 +/- 0.06	0.02 +/- 0.13	16.56	0.39
Mount Rainier NP	1	1.46 +/- 0.04	0.97 +/- 0.06	0.90 +/- 0.17	9.86	0.44
Mount Zirkel Wilderness	1	1.17 +/- 0.26	0.91 +/- 0.09	0.68 +/- 0.09	13.37	0.06

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Nebraska NF	1	1.45 +/- 0.22	0.70 +/- 0.08	1.11 +/- 0.05	10.90	-0.15
North Cascades	1	1.62 +/- 0.12	1.04 +/- 0.07	0.93 +/- 0.27	15.51	0.36
Northern Cheyenne	1	1.86 +/- 0.11	0.95 +/- 0.04	0.19 +/- 0.06	10.85	-0.11
Okefenokee NWR	1	1.64 +/- 0.05	0.94 +/- 0.02	0.25 +/- 0.14	6.33	-0.25
Olympic	1	1.36 +/- 0.06	1.00 +/- 0.07	0.67 +/- 0.09	8.86	0.02
Omaha	1	1.23 +/- 0.12	1.02 +/- 0.03	1.09 +/- 0.03	4.82	0.01
Organ Pipe	1	1.40 +/- 0.13	1.01 +/- 0.06	0.35 +/- 0.10	8.48	0.83
Pasayten	1	1.47 +/- 0.11	1.11 +/- 0.06	0.27 +/- 0.08	16.62	0.64
Pinnacles NM	1	1.39 +/- 0.23	0.82 +/- 0.24	0.84 +/- 0.13	13.03	1.07
Point Reyes National Seashore	1	1.02 +/- 0.13	0.88 +/- 0.05	0.99 +/- 0.04	7.92	-0.93
Presque Isle	1	1.36 +/- 0.07	0.87 +/- 0.03	0.91 +/- 0.11	5.49	0.00
Proctor Maple R. F.	1	1.32 +/- 0.09	0.91 +/- 0.02	0.96 +/- 0.05	6.22	-0.04
Puget Sound	1	1.25 +/- 0.05	0.66 +/- 0.09	1.23 +/- 0.08	6.18	-0.05
Quabbin Summit	1	1.47 +/- 0.11	0.93 +/- 0.03	0.65 +/- 0.07	6.14	-0.07
Quaker City	1	1.81 +/- 0.10	0.86 +/- 0.02	0.79 +/- 0.03	6.66	-0.09
Queen Valley	1	1.31 +/- 0.16	0.94 +/- 0.09	0.54 +/- 0.03	7.88	0.34
Redwood NP	1	1.65 +/- 0.07	0.76 +/- 0.05	0.97 +/- 0.13	7.65	-0.85
Sac and Fox	1	1.27 +/- 0.08	0.84 +/- 0.05	1.07 +/- 0.03	7.23	-0.34
Saguaro West	1	1.33 +/- 0.20	0.96 +/- 0.13	0.28 +/- 0.08	8.45	0.73
San Gabriel	1	1.10 +/- 0.18	0.74 +/- 0.10	0.76 +/- 0.04	10.47	-0.40
San Pedro Parks	1	1.70 +/- 0.28	1.04 +/- 0.09	-0.11 +/- 0.18	12.96	0.03
Sawtooth NF	1	1.10 +/- 0.04	0.94 +/- 0.12	0.68 +/- 0.47	14.36	-1.80
Seney	1	1.42 +/- 0.16	0.91 +/- 0.03	0.86 +/- 0.03	7.24	0.87
Shenandoah NP	1	1.72 +/- 0.08	0.85 +/- 0.02	0.78 +/- 0.04	7.21	-0.43
Shining Rock Wilderness	1	1.58 +/- 0.08	0.97 +/- 0.03	0.13 +/- 0.11	8.78	-0.33
Sierra Ancha	1	1.15 +/- 0.09	1.07 +/- 0.09	0.29 +/- 0.10	11.57	-0.45
Sikes	1	1.52 +/- 0.05	1.01 +/- 0.02	0.44 +/- 0.05	5.92	-0.07
Sipsy Wilderness	1	1.81 +/- 0.05	0.84 +/- 0.02	0.44 +/- 0.04	6.30	-0.04

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Starkey	1	1.40 +/- 0.05	0.77 +/- 0.05	0.78 +/- 0.03	10.02	-0.53
Sula Peak	1	1.30 +/- 0.10	1.05 +/- 0.07	0.36 +/- 0.16	15.56	-2.26
Swanquarter	1	1.71 +/- 0.09	0.99 +/- 0.02	0.19 +/- 0.09	6.60	0.02
Sycamore Canyon	1	1.21 +/- 0.05	0.95 +/- 0.06	0.52 +/- 0.05	8.21	0.25
Tallgrass	1	1.44 +/- 0.08	0.77 +/- 0.05	0.97 +/- 0.03	8.75	-0.50
Theodore Roosevelt	1	1.01 +/- 0.14	1.06 +/- 0.04	0.90 +/- 0.04	7.70	-0.31
Three Sisters Wilderness	1	1.65 +/- 0.09	1.05 +/- 0.06	0.33 +/- 0.20	14.39	-1.15
Thunder Basin	1	1.48 +/- 0.10	0.88 +/- 0.05	0.74 +/- 0.05	7.62	0.08
Tonto NM	1	1.52 +/- 0.14	1.05 +/- 0.08	0.33 +/- 0.06	8.99	0.16
Trinity	1	1.42 +/- 0.07	1.26 +/- 0.08	0.37 +/- 0.11	13.19	-0.85
UL Bend	1	1.65 +/- 0.12	0.81 +/- 0.04	0.90 +/- 0.04	9.92	0.05
Upper Buffalo Wilderness	1	1.46 +/- 0.06	0.87 +/- 0.03	0.89 +/- 0.03	7.83	-0.55
Viking Lake	1	1.27 +/- 0.09	0.92 +/- 0.04	1.12 +/- 0.02	5.27	0.01
Voyageurs NP #2	1	1.12 +/- 0.12	0.89 +/- 0.04	0.98 +/- 0.02	7.15	0.14
Weminuche Wilderness	1	1.14 +/- 0.10	1.01 +/- 0.07	0.45 +/- 0.14	10.27	-1.08
White Mountain	1	1.51 +/- 0.17	1.05 +/- 0.07	0.60 +/- 0.06	9.25	-0.14
White Pass	1	1.30 +/- 0.20	1.15 +/- 0.08	0.59 +/- 0.16	22.56	-1.95
White River NF	1	1.29 +/- 0.27	1.02 +/- 0.11	0.20 +/- 0.15	14.38	-0.04
Wichita Mountains	1	1.27 +/- 0.07	0.92 +/- 0.04	1.01 +/- 0.03	8.22	-0.84
Wind Cave	1	1.35 +/- 0.08	0.95 +/- 0.04	0.55 +/- 0.04	9.80	0.15
Yosemite NP	1	1.47 +/- 0.13	1.00 +/- 0.10	0.88 +/- 0.04	13.25	-2.28
Zion Canyon	1	1.17 +/- 0.12	1.07 +/- 0.08	0.55 +/- 0.07	11.12	-0.95
Acadia NP	2	1.93 +/- 0.06	0.97 +/- 0.02	0.45 +/- 0.11	6.54	0.38
Addison Pinnacle	2	1.63 +/- 0.13	1.08 +/- 0.03	0.35 +/- 0.10	7.37	-1.00
Agua Tibia	2	2.10 +/- 0.09	0.79 +/- 0.05	0.58 +/- 0.05	6.55	0.04
Arendtsville	2	1.84 +/- 0.12	0.99 +/- 0.03	0.69 +/- 0.05	7.31	-0.45
Badlands NP	2	1.69 +/- 0.09	0.89 +/- 0.06	0.45 +/- 0.14	11.19	0.13
Bandelier NM	2	1.43 +/- 0.09	0.98 +/- 0.07	0.82 +/- 0.27	7.86	0.07

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Big Bend NP	2	2.16 +/- 0.12	0.96 +/- 0.03	0.24 +/- 0.28	6.72	0.19
Birmingham	2	1.41 +/- 0.08	1.07 +/- 0.04	0.18 +/- 0.34	5.54	0.21
Bliss SP (TRPA)	2	1.94 +/- 0.05	0.67 +/- 0.05	0.91 +/- 0.13	7.31	-0.48
Bondville	2	1.95 +/- 0.12	1.03 +/- 0.04	0.63 +/- 0.05	8.55	-0.67
Bridger Wilderness	2	1.83 +/- 0.07	0.93 +/- 0.05	0.45 +/- 0.18	7.90	-0.13
Cabinet Mountains	2	1.82 +/- 0.05	0.98 +/- 0.05	0.45 +/- 0.18	6.96	0.09
Cadiz	2	1.90 +/- 0.07	0.96 +/- 0.02	0.67 +/- 0.06	6.17	-0.07
Caney Creek	2	1.87 +/- 0.10	1.03 +/- 0.03	0.01 +/- 0.16	6.51	-0.15
Canyonlands NP	2	2.14 +/- 0.12	0.76 +/- 0.06	0.89 +/- 0.24	7.51	0.21
Cape Cod	2	1.98 +/- 0.12	0.98 +/- 0.03	0.35 +/- 0.14	7.03	-0.99
Cape Romain NWR	2	1.76 +/- 0.08	0.98 +/- 0.02	0.04 +/- 0.21	6.76	-0.01
Capitol Reef NP	2	2.20 +/- 0.12	0.75 +/- 0.08	0.33 +/- 0.24	7.39	-0.29
Casco Bay	2	1.72 +/- 0.07	1.10 +/- 0.03	0.25 +/- 0.14	7.36	-0.67
Cedar Bluff	2	1.69 +/- 0.10	1.03 +/- 0.04	0.40 +/- 0.05	6.96	-0.06
Chassahowitzka NWR	2	1.79 +/- 0.10	0.96 +/- 0.03	0.24 +/- 0.21	6.90	-0.02
Cherokee Nation	2	1.72 +/- 0.05	1.00 +/- 0.02	0.45 +/- 0.05	5.38	-0.07
Cloud Peak	2	1.92 +/- 0.07	0.99 +/- 0.06	0.30 +/- 0.16	7.81	-0.07
Cohutta	2	2.15 +/- 0.10	0.90 +/- 0.03	-0.26 +/- 0.19	5.76	-0.18
Columbia Gorge #1	2	1.57 +/- 0.06	0.85 +/- 0.06	0.52 +/- 0.09	7.03	-0.08
Columbia River Gorge	2	1.34 +/- 0.08	1.14 +/- 0.08	0.40 +/- 0.18	8.28	-0.61
Crater Lake NP	2	1.81 +/- 0.06	0.90 +/- 0.05	1.08 +/- 0.26	7.85	-0.14
Craters of the Moon NM	2	1.97 +/- 0.06	0.95 +/- 0.05	0.24 +/- 0.12	8.30	-0.26
Crescent Lake	2	2.02 +/- 0.13	0.94 +/- 0.06	0.73 +/- 0.06	9.17	0.14
Dolly Sods Wilderness	2	1.94 +/- 0.14	0.99 +/- 0.03	0.07 +/- 0.14	6.90	-0.26
Dome Lands Wilderness	2	2.05 +/- 0.14	0.64 +/- 0.11	0.56 +/- 0.06	7.08	-0.23
El Dorado Springs	2	1.78 +/- 0.07	0.98 +/- 0.03	0.27 +/- 0.06	6.24	-0.24
Ellis	2	1.88 +/- 0.08	0.98 +/- 0.03	0.51 +/- 0.06	6.52	-0.39
Everglades NP	2	1.53 +/- 0.04	1.04 +/- 0.03	0.44 +/- 0.21	7.28	0.10

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Flathead	2	1.76 +/- 0.05	0.97 +/- 0.06	0.02 +/- 0.22	8.40	-0.52
Fort Peck	2	1.95 +/- 0.08	0.90 +/- 0.04	0.28 +/- 0.06	7.60	0.41
Frostberg Reservoir (Big Piney Run)	2	2.25 +/- 0.14	0.89 +/- 0.03	-0.07 +/- 0.10	5.34	0.34
Gates of the Mountains	2	1.74 +/- 0.08	0.89 +/- 0.08	0.45 +/- 0.20	9.98	0.30
Glacier NP	2	1.73 +/- 0.05	0.98 +/- 0.08	0.27 +/- 0.55	7.39	-0.73
Great Basin NP	2	1.56 +/- 0.14	0.88 +/- 0.10	1.10 +/- 0.37	9.56	0.10
Great Gulf Wilderness	2	1.70 +/- 0.07	1.06 +/- 0.03	0.04 +/- 0.33	7.36	-0.15
Great River Bluffs	2	1.69 +/- 0.07	1.01 +/- 0.03	0.41 +/- 0.05	8.14	0.01
Great Sand Dunes NM	2	1.89 +/- 0.11	0.75 +/- 0.09	1.07 +/- 0.27	7.76	-0.20
Great Smoky Mountains NP	2	2.15 +/- 0.10	1.01 +/- 0.03	-0.15 +/- 0.18	6.27	-0.39
Guadalupe Mountains NP	2	1.52 +/- 0.15	1.01 +/- 0.05	0.81 +/- 0.24	6.62	0.27
Hance Camp at Grand Canyon NP	2	1.89 +/- 0.10	0.93 +/- 0.06	0.81 +/- 0.15	7.05	-0.22
Hercules-Glades	2	1.72 +/- 0.07	0.98 +/- 0.03	0.39 +/- 0.10	6.94	-0.27
Hoover	2	2.04 +/- 0.07	0.70 +/- 0.06	0.71 +/- 0.20	8.25	0.38
Ikes Backbone	2	1.71 +/- 0.14	0.63 +/- 0.09	1.05 +/- 0.24	8.69	0.28
Indian Gardens	2	1.90 +/- 0.11	0.74 +/- 0.07	1.01 +/- 0.22	7.07	0.17
Isle Royale NP	2	1.94 +/- 0.07	1.09 +/- 0.03	0.62 +/- 0.08	8.44	-0.01
James River Face Wilderness	2	2.19 +/- 0.09	0.91 +/- 0.03	0.02 +/- 0.12	6.36	-0.17
Jarbidge Wilderness	2	1.83 +/- 0.08	0.86 +/- 0.07	1.21 +/- 0.36	8.64	-0.56
Joshua Tree NP	2	2.39 +/- 0.14	0.62 +/- 0.08	0.56 +/- 0.04	7.39	-0.18
Kaiser	2	1.87 +/- 0.08	0.78 +/- 0.07	0.61 +/- 0.08	7.88	0.22
Kalmiopsis	2	1.70 +/- 0.07	0.87 +/- 0.06	0.76 +/- 0.22	8.14	-0.33
Lassen Volcanic NP	2	1.72 +/- 0.05	0.96 +/- 0.05	0.47 +/- 0.09	8.70	1.16
Lava Beds NM	2	1.73 +/- 0.06	0.88 +/- 0.06	0.85 +/- 0.19	9.34	0.22
Linville Gorge	2	1.99 +/- 0.08	1.02 +/- 0.02	0.01 +/- 0.16	5.82	0.08
Livonia	2	1.97 +/- 0.13	1.01 +/- 0.03	0.44 +/- 0.05	7.43	-0.38

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Lostwood	2	1.96 +/- 0.07	0.92 +/- 0.04	0.41 +/- 0.04	8.26	-0.07
Lye Brook Wilderness	2	2.00 +/- 0.10	0.99 +/- 0.03	0.10 +/- 0.15	6.81	-0.20
Mammoth Cave NP	2	1.99 +/- 0.08	0.93 +/- 0.02	0.27 +/- 0.08	6.04	-0.10
Marthas Vineyard	2	2.04 +/- 0.12	1.00 +/- 0.03	0.27 +/- 0.13	6.57	-0.18
Meadview	2	2.23 +/- 0.17	0.74 +/- 0.07	0.76 +/- 0.22	7.15	-0.03
Medicine Lake	2	2.17 +/- 0.07	0.83 +/- 0.04	0.39 +/- 0.07	8.83	-0.10
Mesa Verde NP	2	1.42 +/- 0.15	1.09 +/- 0.12	0.66 +/- 0.37	10.12	0.89
Moosehorn NWR	2	1.87 +/- 0.07	0.91 +/- 0.03	0.89 +/- 0.15	8.04	-0.44
Mount Baldy	2	1.83 +/- 0.10	0.79 +/- 0.07	0.94 +/- 0.17	6.55	-0.05
Mount Rainier NP	2	1.70 +/- 0.06	1.07 +/- 0.07	0.45 +/- 0.21	8.46	0.34
Mount Zirkel Wilderness	2	1.89 +/- 0.12	0.94 +/- 0.08	0.30 +/- 0.18	9.21	-0.46
Nebraska NF	2	2.10 +/- 0.19	1.03 +/- 0.08	0.37 +/- 0.07	10.49	0.01
North Absaroka	2	2.05 +/- 0.08	0.94 +/- 0.07	0.18 +/- 0.14	8.55	-0.14
North Cascades	2	1.90 +/- 0.07	1.02 +/- 0.06	0.81 +/- 0.24	8.42	0.06
Northern Cheyenne	2	2.06 +/- 0.07	0.96 +/- 0.04	0.13 +/- 0.09	7.45	-0.09
Okefenokee NWR	2	1.61 +/- 0.07	1.00 +/- 0.03	0.13 +/- 0.22	6.98	0.01
Olympic	2	1.39 +/- 0.08	1.13 +/- 0.07	0.74 +/- 0.17	7.30	-0.24
Omaha	2	1.91 +/- 0.09	1.03 +/- 0.04	0.53 +/- 0.05	8.00	-0.12
Organ Pipe	2	1.74 +/- 0.16	0.77 +/- 0.07	0.86 +/- 0.23	6.20	0.01
Pasayten	2	1.58 +/- 0.07	1.09 +/- 0.07	0.38 +/- 0.24	10.47	-0.24
Petrified Forest NP	2	1.94 +/- 0.11	0.72 +/- 0.07	1.24 +/- 0.18	6.54	0.20
Phoenix	2	1.23 +/- 0.08	1.11 +/- 0.08	0.21 +/- 0.20	5.95	-0.12
Pinnacles NM	2	1.65 +/- 0.07	0.78 +/- 0.04	1.00 +/- 0.07	6.64	0.36
Point Reyes National Seashore	2	2.06 +/- 0.24	0.69 +/- 0.08	1.12 +/- 0.25	8.31	0.01
Presque Isle	2	1.86 +/- 0.05	0.91 +/- 0.03	0.03 +/- 0.20	5.69	0.03
Proctor Maple R. F.	2	1.95 +/- 0.09	1.01 +/- 0.03	0.25 +/- 0.13	6.81	-0.46
Puget Sound	2	1.48 +/- 0.07	0.98 +/- 0.06	0.44 +/- 0.09	6.25	-0.18
Quabbin Summit	2	2.02 +/- 0.10	0.91 +/- 0.03	0.01 +/- 0.14	6.58	-0.29



Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Quaker City	2	1.83 +/- 0.14	1.07 +/- 0.03	0.17 +/- 0.11	7.39	-0.37
Queen Valley	2	1.73 +/- 0.16	0.85 +/- 0.07	0.88 +/- 0.18	6.73	-0.33
Redwood NP	2	1.81 +/- 0.12	0.88 +/- 0.06	0.95 +/- 0.23	7.81	-0.70
Rocky Mountain NP	2	1.67 +/- 0.10	0.94 +/- 0.08	0.47 +/- 0.07	8.43	-0.43
Sac and Fox	2	1.86 +/- 0.07	0.92 +/- 0.03	0.50 +/- 0.04	6.65	-0.43
Saguaro NM	2	1.44 +/- 0.13	0.80 +/- 0.09	1.02 +/- 0.30	7.39	-0.20
Saguaro West	2	1.59 +/- 0.19	0.76 +/- 0.11	0.85 +/- 0.29	6.93	-0.15
San Gabriel	2	2.10 +/- 0.10	0.71 +/- 0.05	0.47 +/- 0.03	7.47	0.05
San Geronio Wilderness	2	1.40 +/- 0.12	0.81 +/- 0.09	0.79 +/- 0.03	8.35	0.54
San Pedro Parks	2	1.85 +/- 0.09	0.66 +/- 0.08	1.12 +/- 0.24	8.41	0.03
San Rafael	2	1.81 +/- 0.12	0.81 +/- 0.07	0.71 +/- 0.09	8.42	0.13
Seney	2	1.83 +/- 0.07	0.97 +/- 0.02	0.31 +/- 0.07	6.86	-0.25
Sequoia NP	2	1.84 +/- 0.08	0.75 +/- 0.07	0.69 +/- 0.06	6.59	0.04
Shamrock Mine	2	2.01 +/- 0.11	1.01 +/- 0.07	0.27 +/- 0.23	5.14	-0.22
Shenandoah NP	2	1.76 +/- 0.10	1.13 +/- 0.03	0.21 +/- 0.09	6.69	-0.30
Sierra Ancha	2	1.36 +/- 0.07	0.98 +/- 0.06	0.59 +/- 0.16	6.43	-0.42
Sikes	2	1.79 +/- 0.06	1.06 +/- 0.03	0.02 +/- 0.20	5.60	-0.11
Sipsy Wilderness	2	1.87 +/- 0.07	1.01 +/- 0.02	0.02 +/- 0.11	5.39	0.05
Snoqualmie Pass	2	1.73 +/- 0.09	0.95 +/- 0.09	0.44 +/- 0.17	10.95	-0.88
Starkey	2	1.74 +/- 0.07	0.88 +/- 0.07	0.62 +/- 0.23	9.55	0.22
Sula Peak	2	1.71 +/- 0.07	1.04 +/- 0.11	-0.06 +/- 0.53	10.45	-0.26
Tallgrass	2	1.63 +/- 0.05	1.06 +/- 0.02	0.22 +/- 0.05	5.98	-0.69
Theodore Roosevelt	2	1.83 +/- 0.10	1.03 +/- 0.06	0.60 +/- 0.08	8.73	0.44
Three Sisters Wilderness	2	1.81 +/- 0.06	1.01 +/- 0.06	0.46 +/- 0.20	8.79	-0.06
Thunder Basin	2	1.86 +/- 0.08	0.96 +/- 0.05	0.59 +/- 0.10	6.27	-0.15
Tonto NM	2	1.94 +/- 0.12	0.75 +/- 0.07	0.78 +/- 0.16	5.73	-0.28
Trinity	2	1.80 +/- 0.07	0.81 +/- 0.06	1.08 +/- 0.21	8.68	-0.19
UL Bend	2	1.88 +/- 0.06	0.94 +/- 0.04	0.59 +/- 0.15	7.37	0.03

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Upper Buffalo Wilderness	2	1.80 +/- 0.08	1.01 +/- 0.03	0.54 +/- 0.08	6.49	-0.22
Viking Lake	2	1.59 +/- 0.07	1.05 +/- 0.03	0.77 +/- 0.04	7.92	-0.63
Voyageurs NP #2	2	1.84 +/- 0.05	0.97 +/- 0.03	0.33 +/- 0.09	7.54	-0.34
Weminuche Wilderness	2	1.94 +/- 0.10	0.88 +/- 0.08	0.06 +/- 0.25	7.88	-0.21
Wheeler Peak	2	1.67 +/- 0.13	0.93 +/- 0.11	0.86 +/- 0.34	9.25	0.21
White Mountain	2	1.85 +/- 0.13	1.07 +/- 0.07	0.23 +/- 0.28	7.22	-0.02
White Pass	2	1.78 +/- 0.08	1.21 +/- 0.06	0.33 +/- 0.20	9.65	0.38
White River NF	2	2.14 +/- 0.10	0.87 +/- 0.07	0.28 +/- 0.19	8.32	0.12
Wichita Mountains	2	1.74 +/- 0.10	1.09 +/- 0.04	0.47 +/- 0.09	6.69	-0.54
Wind Cave	2	1.72 +/- 0.06	0.97 +/- 0.03	0.27 +/- 0.05	6.39	-1.02
Yellowstone NP 2	2	1.78 +/- 0.06	0.94 +/- 0.05	0.32 +/- 0.11	7.11	-0.70
Yosemite NP	2	1.87 +/- 0.05	0.86 +/- 0.04	0.72 +/- 0.07	5.53	-0.18
Zion Canyon	2	1.83 +/- 0.11	1.10 +/- 0.07	0.40 +/- 0.31	6.67	-0.31
Acadia NP	3	1.80 +/- 0.06	1.12 +/- 0.02	0.59 +/- 0.31	6.50	-0.15
Addison Pinnacle	3	1.82 +/- 0.11	1.08 +/- 0.02	0.49 +/- 0.46	6.85	-0.30
Agua Tibia	3	1.96 +/- 0.08	0.96 +/- 0.03	0.38 +/- 0.06	5.41	-0.30
Arendtsville	3	1.89 +/- 0.11	1.12 +/- 0.02	-0.06 +/- 0.11	6.22	-0.16
Badlands NP	3	1.80 +/- 0.05	0.94 +/- 0.05	0.22 +/- 0.17	9.80	-0.52
Big Bend NP	3	1.57 +/- 0.17	1.06 +/- 0.03	0.80 +/- 0.25	6.67	0.16
Birmingham	3	1.59 +/- 0.13	1.11 +/- 0.04	-0.48 +/- 0.55	6.10	-0.13
Bliss SP (TRPA)	3	1.79 +/- 0.04	0.88 +/- 0.06	1.18 +/- 0.26	6.44	-0.37
Blue Mounds	3	2.07 +/- 0.05	1.06 +/- 0.03	0.05 +/- 0.09	5.83	-0.42
Bondville	3	1.95 +/- 0.14	1.19 +/- 0.03	-0.05 +/- 0.17	8.03	-0.10
Boundary Waters Canoe Area	3	1.81 +/- 0.04	0.99 +/- 0.04	1.08 +/- 0.61	7.84	-0.64
Bridger Wilderness	3	1.83 +/- 0.05	1.10 +/- 0.09	-0.15 +/- 0.53	9.07	-0.86
Brigantine NWR	3	1.92 +/- 0.12	1.06 +/- 0.03	-0.06 +/- 0.29	8.06	-0.25
Bryce Canyon NP	3	1.44 +/- 0.07	1.13 +/- 0.08	0.88 +/- 0.53	10.89	-0.42
Cabinet Mountains	3	1.84 +/- 0.04	1.02 +/- 0.10	-0.27 +/- 0.41	6.38	-0.45

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Cadiz	3	1.91 +/- 0.17	1.07 +/- 0.03	-0.05 +/- 0.55	7.81	-0.50
Canyonlands NP	3	2.04 +/- 0.08	1.02 +/- 0.07	-0.28 +/- 0.61	8.07	-0.16
Cape Cod	3	1.76 +/- 0.09	1.11 +/- 0.03	-0.09 +/- 0.33	8.50	-0.45
Cape Romain NWR	3	1.74 +/- 0.15	1.03 +/- 0.03	-0.33 +/- 0.40	8.17	-0.16
Cedar Bluff	3	1.78 +/- 0.08	1.05 +/- 0.03	0.14 +/- 0.14	6.06	-0.12
Cherokee Nation	3	1.82 +/- 0.10	1.05 +/- 0.03	0.07 +/- 0.24	5.13	-0.01
Chiricahua NM	3	1.93 +/- 0.13	1.02 +/- 0.04	0.78 +/- 0.43	7.69	-0.09
Columbia Gorge #1	3	1.61 +/- 0.05	0.98 +/- 0.07	0.35 +/- 0.15	7.36	-0.07
Columbia River Gorge	3	1.63 +/- 0.06	0.92 +/- 0.11	0.86 +/- 0.29	7.57	-0.09
Connecticut Hill	3	1.75 +/- 0.11	1.11 +/- 0.02	-0.33 +/- 0.36	6.04	-0.07
Crater Lake NP	3	1.78 +/- 0.04	0.90 +/- 0.11	0.61 +/- 0.77	8.54	-2.03
Craters of the Moon NM	3	1.93 +/- 0.05	0.79 +/- 0.09	-0.13 +/- 0.24	7.42	-0.60
Crescent Lake	3	2.02 +/- 0.05	1.09 +/- 0.04	0.09 +/- 0.08	6.17	-0.87
Dolly Sods Wilderness	3	1.77 +/- 0.12	1.05 +/- 0.02	-0.14 +/- 0.46	6.77	-0.25
Dome Lands Wilderness	3	1.92 +/- 0.07	0.93 +/- 0.07	0.13 +/- 0.10	5.97	-0.46
Douglas	3	1.45 +/- 0.12	1.10 +/- 0.05	-0.42 +/- 0.39	6.26	0.16
El Dorado Springs	3	1.48 +/- 0.09	1.10 +/- 0.03	0.34 +/- 0.23	5.99	-0.03
Ellis	3	1.85 +/- 0.11	1.04 +/- 0.04	0.71 +/- 0.24	6.34	-0.49
Everglades NP	3	1.52 +/- 0.10	1.05 +/- 0.03	0.58 +/- 0.25	7.12	0.05
Fort Peck	3	1.82 +/- 0.04	1.01 +/- 0.05	0.98 +/- 0.34	7.19	-0.13
Frostberg Reservoir (Big Piney Run)	3	1.90 +/- 0.12	0.98 +/- 0.02	0.02 +/- 0.56	4.83	0.10
Gates of the Mountains	3	1.80 +/- 0.04	0.75 +/- 0.10	0.95 +/- 0.34	7.23	-0.68
Gila Wilderness	3	1.50 +/- 0.06	1.06 +/- 0.05	-0.22 +/- 0.84	9.11	-0.28
Great Basin NP	3	1.88 +/- 0.05	0.90 +/- 0.05	0.32 +/- 0.36	7.17	-0.36
Great Gulf Wilderness	3	1.91 +/- 0.08	1.09 +/- 0.02	-1.69 +/- 1.66	6.85	-0.12
Great River Bluffs	3	1.96 +/- 0.05	0.98 +/- 0.02	0.09 +/- 0.09	5.56	-0.50
Great Sand Dunes NM	3	2.12 +/- 0.13	0.90 +/- 0.11	-0.35 +/- 1.30	8.90	-0.40

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Great Smoky Mountains NP	3	1.84 +/- 0.15	1.07 +/- 0.03	0.43 +/- 0.82	6.94	-0.32
Hance Camp at Grand Canyon NP	3	1.52 +/- 0.06	1.13 +/- 0.06	0.93 +/- 0.40	8.28	-0.47
Hells Canyon	3	1.74 +/- 0.03	0.84 +/- 0.11	0.10 +/- 0.53	6.94	-0.25
Hoover	3	1.83 +/- 0.06	0.97 +/- 0.07	0.02 +/- 0.39	8.10	-0.42
Indian Gardens	3	1.65 +/- 0.10	1.09 +/- 0.08	-0.35 +/- 0.51	8.41	-0.49
Isle Royale NP	3	1.93 +/- 0.06	1.09 +/- 0.03	0.55 +/- 0.85	7.46	-0.63
Jarbidge Wilderness	3	1.93 +/- 0.06	0.81 +/- 0.09	0.77 +/- 0.69	6.22	-0.59
Joshua Tree NP	3	2.19 +/- 0.13	0.93 +/- 0.06	0.30 +/- 0.13	7.37	-0.17
Kaiser	3	2.00 +/- 0.05	0.73 +/- 0.06	0.46 +/- 0.07	6.30	-0.56
Kalmiopsis	3	1.65 +/- 0.05	0.83 +/- 0.08	0.96 +/- 0.28	8.59	-0.42
Lassen Volcanic NP	3	1.76 +/- 0.04	0.85 +/- 0.07	1.32 +/- 0.25	7.76	0.88
Lava Beds NM	3	1.75 +/- 0.07	0.98 +/- 0.21	0.35 +/- 1.20	8.65	-0.62
Linville Gorge	3	2.01 +/- 0.15	1.09 +/- 0.03	0.80 +/- 1.24	6.27	-0.32
Livonia	3	2.02 +/- 0.19	1.10 +/- 0.03	0.16 +/- 0.22	8.11	-0.41
Lye Brook Wilderness	3	1.98 +/- 0.08	1.03 +/- 0.02	0.28 +/- 0.38	6.59	0.09
Mammoth Cave NP	3	2.24 +/- 0.15	0.91 +/- 0.03	-0.46 +/- 0.40	5.78	-0.18
Marthas Vineyard	3	1.78 +/- 0.08	1.10 +/- 0.02	0.17 +/- 0.22	6.35	-0.07
Meadview	3	1.80 +/- 0.10	0.98 +/- 0.05	0.57 +/- 0.26	7.45	-0.01
Medicine Lake	3	1.73 +/- 0.04	1.03 +/- 0.05	0.63 +/- 0.29	7.58	0.28
Monture	3	1.88 +/- 0.12	1.37 +/- 0.55	-6.81 +/- 8.29	7.53	-0.05
Moosehorn NWR	3	1.65 +/- 0.06	1.09 +/- 0.03	0.48 +/- 0.46	7.90	-0.44
Mount Baldy	3	1.51 +/- 0.05	1.05 +/- 0.04	0.79 +/- 0.37	6.89	-0.81
Mount Hood	3	1.76 +/- 0.04	1.29 +/- 0.08	0.08 +/- 0.16	8.69	0.63
Mount Rainier NP	3	1.65 +/- 0.06	1.19 +/- 0.08	0.52 +/- 0.30	8.52	0.37
Mount Zirkel Wilderness	3	2.02 +/- 0.07	0.97 +/- 0.07	-0.62 +/- 0.44	6.67	-1.14
Nebraska NF	3	2.03 +/- 0.06	1.11 +/- 0.04	-0.19 +/- 0.14	7.31	-0.51
North Absaroka	3	1.86 +/- 0.05	1.11 +/- 0.09	0.31 +/- 0.42	7.02	-0.96

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
North Cascades	3	1.73 +/- 0.05	1.20 +/- 0.07	0.67 +/- 0.31	7.16	-0.16
Okefenokee NWR	3	1.90 +/- 0.12	0.92 +/- 0.03	-0.28 +/- 0.29	7.30	0.00
Olympic	3	1.68 +/- 0.06	1.00 +/- 0.05	0.71 +/- 0.14	7.76	-0.51
Omaha	3	2.12 +/- 0.08	1.04 +/- 0.03	0.09 +/- 0.14	6.09	-0.46
Organ Pipe	3	1.14 +/- 0.17	0.93 +/- 0.05	1.13 +/- 0.23	7.51	0.25
Pasayten	3	1.66 +/- 0.03	1.17 +/- 0.08	0.20 +/- 0.55	7.68	0.45
Petrified Forest NP	3	1.79 +/- 0.07	1.01 +/- 0.04	0.87 +/- 0.29	7.28	-0.66
Pinnacles NM	3	1.84 +/- 0.04	0.90 +/- 0.03	0.68 +/- 0.08	6.86	0.19
Puget Sound	3	1.53 +/- 0.05	0.87 +/- 0.04	0.71 +/- 0.10	6.04	-0.15
Quabbin Summit	3	1.78 +/- 0.05	1.00 +/- 0.02	-0.15 +/- 0.17	5.62	0.41
Queen Valley	3	1.90 +/- 0.13	0.98 +/- 0.05	0.09 +/- 0.16	7.39	-0.23
Redwood NP	3	1.83 +/- 0.05	0.94 +/- 0.05	0.95 +/- 0.18	7.94	-0.54
Rocky Mountain NP	3	1.97 +/- 0.08	0.84 +/- 0.11	0.18 +/- 0.18	8.26	-0.51
Saguaro NM	3	1.32 +/- 0.12	0.97 +/- 0.06	0.97 +/- 0.28	8.01	0.07
Salt Creek	3	1.78 +/- 0.14	1.00 +/- 0.05	0.35 +/- 0.24	8.47	0.30
San Gabriel	3	2.06 +/- 0.08	0.90 +/- 0.04	0.10 +/- 0.07	7.00	-0.32
San Geronio Wilderness	3	1.84 +/- 0.08	0.85 +/- 0.06	0.44 +/- 0.04	6.76	-0.20
San Pedro Parks	3	1.54 +/- 0.08	1.10 +/- 0.06	0.23 +/- 0.49	8.28	-0.08
San Rafael	3	2.00 +/- 0.06	0.85 +/- 0.04	0.57 +/- 0.09	7.33	-0.43
Sawtooth NF	3	1.88 +/- 0.07	1.02 +/- 0.24	-4.18 +/- 2.96	7.19	-0.15
Seney	3	1.61 +/- 0.06	1.04 +/- 0.03	0.77 +/- 0.45	7.71	-0.67
Sequoia NP	3	1.87 +/- 0.06	0.87 +/- 0.07	0.28 +/- 0.12	5.92	0.11
Shamrock Mine	3	2.02 +/- 0.07	1.06 +/- 0.07	-0.25 +/- 0.56	6.01	-0.59
Shenandoah NP	3	1.76 +/- 0.13	1.11 +/- 0.02	0.53 +/- 0.40	7.50	-0.17
Shining Rock Wilderness	3	2.08 +/- 0.20	0.98 +/- 0.03	-0.82 +/- 0.80	6.68	-0.30
Sierra Ancha	3	1.32 +/- 0.07	0.99 +/- 0.06	0.41 +/- 0.38	8.03	-0.21
Sikes	3	1.88 +/- 0.09	1.03 +/- 0.03	-0.37 +/- 0.36	5.79	-0.14
Sipsy Wilderness	3	2.27 +/- 0.11	0.92 +/- 0.03	-0.53 +/- 0.41	5.54	-0.20

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Snoqualmie Pass	3	1.79 +/- 0.06	1.04 +/- 0.08	0.39 +/- 0.20	8.20	-0.60
Starkey	3	1.75 +/- 0.04	0.75 +/- 0.11	0.97 +/- 0.43	8.04	0.15
Swanquarter	3	1.83 +/- 0.09	1.02 +/- 0.02	0.45 +/- 0.27	5.46	0.01
Sycamore Canyon	3	1.32 +/- 0.08	1.03 +/- 0.11	1.05 +/- 0.58	7.34	0.26
Tallgrass	3	1.51 +/- 0.09	1.06 +/- 0.03	0.69 +/- 0.34	6.37	-0.10
Three Sisters Wilderness	3	1.80 +/- 0.04	1.05 +/- 0.08	0.44 +/- 0.33	5.43	-0.64
Tonto NM	3	1.75 +/- 0.10	0.98 +/- 0.05	0.45 +/- 0.28	7.33	-0.54
UL Bend	3	1.89 +/- 0.05	1.09 +/- 0.07	-1.04 +/- 0.94	7.52	-0.58
Upper Buffalo Wilderness	3	1.88 +/- 0.08	1.01 +/- 0.02	0.45 +/- 0.27	6.04	-0.35
Viking Lake	3	1.81 +/- 0.07	1.12 +/- 0.03	0.13 +/- 0.12	5.95	-0.10
Weminuche Wilderness	3	1.80 +/- 0.08	1.15 +/- 0.09	-0.92 +/- 0.94	9.46	-0.82
Wheeler Peak	3	1.63 +/- 0.11	1.15 +/- 0.08	0.49 +/- 0.45	9.63	0.18
White Mountain	3	1.71 +/- 0.11	1.10 +/- 0.04	-0.04 +/- 0.32	7.19	-0.47
White Pass	3	1.81 +/- 0.06	1.27 +/- 0.10	-0.22 +/- 0.35	10.27	0.11
White River NF	3	1.80 +/- 0.08	1.17 +/- 0.08	0.74 +/- 0.64	8.89	-0.42
Wichita Mountains	3	1.69 +/- 0.09	1.08 +/- 0.03	0.83 +/- 0.18	5.32	-0.06
Wind Cave	3	1.83 +/- 0.04	0.98 +/- 0.05	-0.48 +/- 0.42	6.44	-0.66
Yellowstone NP 2	3	1.80 +/- 0.04	0.98 +/- 0.11	-0.46 +/- 0.51	8.58	-0.81
Yosemite NP	3	1.67 +/- 0.04	1.01 +/- 0.07	0.96 +/- 0.28	7.49	0.11
Zion Canyon	3	1.83 +/- 0.07	1.08 +/- 0.04	0.73 +/- 0.34	6.28	-0.42
Acadia NP	4	1.56 +/- 0.09	0.99 +/- 0.04	0.55 +/- 0.11	8.50	-0.37
Agua Tibia	4	1.54 +/- 0.09	0.95 +/- 0.06	0.63 +/- 0.04	9.37	-0.70
Arendtsville	4	1.49 +/- 0.11	0.96 +/- 0.03	0.89 +/- 0.05	7.98	-0.74
Bandelier NM	4	1.43 +/- 0.05	1.06 +/- 0.04	0.39 +/- 0.08	9.10	-0.46
Big Bend NP	4	2.00 +/- 0.14	1.01 +/- 0.03	-0.22 +/- 0.21	8.13	-0.26
Birmingham	4	1.38 +/- 0.05	1.07 +/- 0.04	0.75 +/- 0.08	5.11	-0.10
Bliss SP (TRPA)	4	1.59 +/- 0.04	1.11 +/- 0.06	0.38 +/- 0.14	9.77	-0.70
Blue Mounds	4	1.57 +/- 0.09	0.96 +/- 0.05	1.11 +/- 0.03	6.56	0.13

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Bondville	4	1.31 +/- 0.10	0.98 +/- 0.04	1.05 +/- 0.03	7.49	-0.29
Bosque del Apache	4	1.15 +/- 0.08	1.01 +/- 0.04	0.83 +/- 0.08	8.70	0.11
Boundary Waters Canoe Area	4	1.65 +/- 0.11	0.97 +/- 0.05	0.80 +/- 0.04	8.87	0.26
Bridger Wilderness	4	1.71 +/- 0.12	1.12 +/- 0.08	0.18 +/- 0.21	14.92	-0.88
Bridgton	4	1.60 +/- 0.08	1.06 +/- 0.04	0.19 +/- 0.14	8.38	-0.42
Brigantine NWR	4	1.50 +/- 0.10	1.01 +/- 0.03	0.71 +/- 0.06	6.71	-0.33
Bryce Canyon NP	4	1.49 +/- 0.10	1.07 +/- 0.07	0.51 +/- 0.08	13.47	-2.55
Cabinet Mountains	4	1.66 +/- 0.04	1.01 +/- 0.09	0.41 +/- 0.18	9.42	-0.31
Cadiz	4	1.75 +/- 0.06	0.90 +/- 0.03	0.75 +/- 0.03	6.50	-0.04
Caney Creek	4	1.74 +/- 0.06	1.01 +/- 0.03	0.32 +/- 0.04	7.02	-0.22
Canyonlands NP	4	1.84 +/- 0.15	1.03 +/- 0.07	0.45 +/- 0.08	11.88	0.45
Capitol Reef NP	4	1.96 +/- 0.13	0.86 +/- 0.08	0.53 +/- 0.06	11.25	-0.63
Casco Bay	4	1.50 +/- 0.05	1.00 +/- 0.03	0.66 +/- 0.10	7.34	-0.59
Cedar Bluff	4	1.03 +/- 0.39	1.12 +/- 0.23	1.01 +/- 0.09	14.58	-0.05
Chassahowitzka NWR	4	1.67 +/- 0.05	0.89 +/- 0.02	0.12 +/- 0.10	4.79	0.01
Cherokee Nation	4	1.39 +/- 0.08	1.03 +/- 0.04	0.91 +/- 0.03	7.89	-0.59
Chiricahua NM	4	1.24 +/- 0.10	1.23 +/- 0.04	0.21 +/- 0.14	9.28	1.55
Cloud Peak	4	2.11 +/- 0.13	0.94 +/- 0.07	0.50 +/- 0.25	17.70	-0.45
Cohutta	4	1.83 +/- 0.08	1.00 +/- 0.03	-0.01 +/- 0.06	6.22	-0.44
Columbia Gorge #1	4	1.60 +/- 0.04	0.68 +/- 0.07	0.72 +/- 0.05	8.73	-0.94
Columbia River Gorge	4	1.46 +/- 0.05	0.84 +/- 0.07	0.73 +/- 0.04	9.09	-0.65
Connecticut Hill	4	1.83 +/- 0.18	0.91 +/- 0.04	0.70 +/- 0.06	7.38	-0.89
Crater Lake NP	4	1.68 +/- 0.06	1.22 +/- 0.08	-0.32 +/- 0.25	12.88	-1.87
Craters of the Moon NM	4	1.68 +/- 0.10	1.10 +/- 0.10	0.47 +/- 0.04	12.35	-0.68
Crescent Lake	4	1.90 +/- 0.19	0.98 +/- 0.11	0.95 +/- 0.04	10.63	0.76
Death Valley NP	4	1.84 +/- 0.15	1.12 +/- 0.07	0.09 +/- 0.11	9.79	-0.52
Dolly Sods Wilderness	4	1.43 +/- 0.07	1.02 +/- 0.02	0.39 +/- 0.08	7.23	-0.24
Douglas	4	1.29 +/- 0.14	0.87 +/- 0.13	0.87 +/- 0.28	6.03	-0.09

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
El Dorado Springs	4	1.37 +/- 0.08	0.97 +/- 0.04	0.85 +/- 0.03	7.91	-0.29
Ellis	4	1.53 +/- 0.10	1.04 +/- 0.05	0.86 +/- 0.03	8.74	-1.10
Everglades NP	4	1.52 +/- 0.11	1.09 +/- 0.03	0.17 +/- 0.19	7.48	-0.24
Flathead	4	1.66 +/- 0.04	1.12 +/- 0.06	-0.05 +/- 0.11	8.03	-0.57
Fort Peck	4	1.44 +/- 0.09	0.96 +/- 0.05	0.76 +/- 0.04	10.16	-0.36
Frostberg Reservoir (Big Piney Run)	4	1.85 +/- 0.09	0.93 +/- 0.02	0.33 +/- 0.06	5.15	-0.27
Gates of the Mountains	4	1.67 +/- 0.06	1.25 +/- 0.07	-0.18 +/- 0.16	11.47	0.06
Gila Wilderness	4	1.61 +/- 0.09	0.97 +/- 0.05	0.43 +/- 0.31	9.70	-0.16
Glacier NP	4	1.65 +/- 0.03	0.87 +/- 0.06	0.94 +/- 0.07	6.97	-0.47
Great Basin NP	4	1.01 +/- 0.09	1.33 +/- 0.07	0.24 +/- 0.13	14.95	-1.22
Great Gulf Wilderness	4	1.88 +/- 0.13	0.91 +/- 0.04	0.26 +/- 0.10	8.58	0.22
Great River Bluffs	4	1.45 +/- 0.15	0.86 +/- 0.07	0.91 +/- 0.03	10.20	-0.11
Great Sand Dunes NM	4	1.43 +/- 0.09	1.06 +/- 0.06	0.09 +/- 0.18	11.34	-1.24
Great Smoky Mountains NP	4	1.83 +/- 0.08	0.96 +/- 0.03	0.36 +/- 0.07	6.90	-0.32
Guadalupe Mountains NP	4	1.55 +/- 0.18	1.04 +/- 0.07	0.49 +/- 0.08	9.50	-0.70
Hance Camp at Grand Canyon NP	4	1.54 +/- 0.07	1.16 +/- 0.05	0.59 +/- 0.09	11.29	0.25
Hells Canyon	4	1.64 +/- 0.05	1.17 +/- 0.10	0.51 +/- 0.04	9.27	-0.97
Hercules-Glades	4	1.62 +/- 0.07	0.96 +/- 0.03	0.55 +/- 0.03	7.54	-0.70
Hoover	4	1.63 +/- 0.06	1.13 +/- 0.07	0.24 +/- 0.15	12.62	0.88
Indian Gardens	4	1.69 +/- 0.06	1.09 +/- 0.04	0.36 +/- 0.11	7.55	0.29
Isle Royale NP	4	1.62 +/- 0.11	1.05 +/- 0.04	0.88 +/- 0.03	8.24	0.70
James River Face Wilderness	4	1.61 +/- 0.05	0.99 +/- 0.02	0.42 +/- 0.07	6.20	-0.18
Jarbidge Wilderness	4	1.55 +/- 0.09	1.26 +/- 0.09	0.77 +/- 0.07	13.67	-0.40
Joshua Tree NP	4	1.64 +/- 0.12	0.95 +/- 0.07	0.64 +/- 0.03	10.63	-0.56
Kaiser	4	1.74 +/- 0.07	0.86 +/- 0.09	0.68 +/- 0.06	13.01	-1.63
Kalmiopsis	4	1.55 +/- 0.03	1.14 +/- 0.11	0.25 +/- 0.40	8.92	-0.61



Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Lassen Volcanic NP	4	1.39 +/- 0.05	1.31 +/- 0.08	0.28 +/- 0.09	11.99	-0.61
Lava Beds NM	4	1.53 +/- 0.04	1.25 +/- 0.08	0.25 +/- 0.08	9.43	-1.11
Linville Gorge	4	1.66 +/- 0.06	1.04 +/- 0.02	0.38 +/- 0.10	6.41	-0.01
Livonia	4	1.48 +/- 0.12	1.04 +/- 0.04	0.82 +/- 0.04	8.19	-0.69
Lostwood	4	1.25 +/- 0.08	0.96 +/- 0.05	0.99 +/- 0.04	9.37	-1.06
M.K. Goddard	4	1.34 +/- 0.07	1.00 +/- 0.03	0.84 +/- 0.03	6.18	-0.16
Mammoth Cave NP	4	1.69 +/- 0.10	0.95 +/- 0.04	0.45 +/- 0.04	7.97	-0.34
Meadview	4	2.14 +/- 0.17	0.91 +/- 0.07	0.17 +/- 0.11	8.99	0.14
Medicine Lake	4	1.55 +/- 0.11	1.05 +/- 0.05	0.72 +/- 0.05	11.09	0.03
Monture	4	1.59 +/- 0.04	1.19 +/- 0.08	0.11 +/- 0.22	9.89	-0.92
Moosehorn NWR	4	1.57 +/- 0.08	0.95 +/- 0.03	0.52 +/- 0.12	7.84	0.01
Mount Baldy	4	1.39 +/- 0.04	1.05 +/- 0.03	0.44 +/- 0.19	8.88	-0.29
Mount Hood	4	1.66 +/- 0.04	1.20 +/- 0.06	0.54 +/- 0.11	11.27	0.23
Mount Rainier NP	4	1.56 +/- 0.04	1.38 +/- 0.10	0.04 +/- 0.28	10.28	0.75
Mount Zirkel Wilderness	4	2.32 +/- 0.15	0.72 +/- 0.07	0.09 +/- 0.18	14.94	-2.20
Nebraska NF	4	1.51 +/- 0.11	1.12 +/- 0.07	0.72 +/- 0.04	10.13	-0.15
North Absaroka	4	1.83 +/- 0.12	0.99 +/- 0.09	0.41 +/- 0.16	14.77	-0.55
North Cascades	4	1.75 +/- 0.05	1.07 +/- 0.07	0.83 +/- 0.29	10.76	0.23
Northern Cheyenne	4	1.71 +/- 0.07	1.16 +/- 0.06	0.16 +/- 0.11	11.39	0.31
Okefenokee NWR	4	1.60 +/- 0.07	1.04 +/- 0.03	-0.23 +/- 0.19	7.07	-0.02
Olympic	4	1.60 +/- 0.06	1.14 +/- 0.09	0.40 +/- 0.12	9.79	-0.28
Omaha	4	1.47 +/- 0.11	1.02 +/- 0.05	1.06 +/- 0.03	7.15	0.07
Organ Pipe	4	1.46 +/- 0.12	1.15 +/- 0.04	0.29 +/- 0.08	6.92	0.22
Pasayten	4	1.70 +/- 0.05	1.23 +/- 0.06	0.11 +/- 0.12	11.26	0.60
Petrified Forest NP	4	1.57 +/- 0.08	1.12 +/- 0.05	0.37 +/- 0.14	8.12	-0.09
Phoenix	4	1.24 +/- 0.04	0.97 +/- 0.07	0.69 +/- 0.05	5.41	0.17
Pinnacles NM	4	1.55 +/- 0.06	1.02 +/- 0.07	0.60 +/- 0.04	9.07	-0.53
Point Reyes National Seashore	4	1.63 +/- 0.11	1.03 +/- 0.07	0.65 +/- 0.04	8.85	-0.70

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Presque Isle	4	1.71 +/- 0.06	0.89 +/- 0.03	0.31 +/- 0.10	6.85	-0.06
Proctor Maple R. F.	4	1.65 +/- 0.09	0.99 +/- 0.03	0.60 +/- 0.06	8.08	-0.37
Quabbin Summit	4	1.65 +/- 0.12	0.96 +/- 0.04	0.38 +/- 0.09	8.01	-0.10
Quaker City	4	1.61 +/- 0.09	0.98 +/- 0.03	0.67 +/- 0.04	6.48	-0.20
Queen Valley	4	1.34 +/- 0.09	1.07 +/- 0.04	0.69 +/- 0.03	8.04	0.18
Redwood NP	4	1.70 +/- 0.05	1.09 +/- 0.10	0.45 +/- 0.24	7.57	-1.08
Rocky Mountain NP	4	1.44 +/- 0.11	0.96 +/- 0.07	0.64 +/- 0.07	14.04	-2.04
Sac and Fox	4	1.38 +/- 0.10	0.94 +/- 0.05	0.95 +/- 0.03	9.43	-0.47
Saguaro NM	4	1.42 +/- 0.09	0.91 +/- 0.04	0.37 +/- 0.04	7.24	0.13
Saguaro West	4	1.27 +/- 0.09	0.94 +/- 0.06	0.41 +/- 0.04	6.06	-0.21
San Gabriel	4	1.64 +/- 0.10	0.83 +/- 0.08	0.53 +/- 0.04	11.87	-0.40
San Gorgonio Wilderness	4	1.20 +/- 0.11	0.87 +/- 0.08	0.87 +/- 0.03	11.27	-1.21
San Pedro Parks	4	1.38 +/- 0.09	1.13 +/- 0.06	0.20 +/- 0.17	12.38	0.76
San Rafael	4	1.50 +/- 0.10	0.94 +/- 0.07	0.60 +/- 0.04	10.81	-1.19
Seney	4	1.31 +/- 0.11	0.97 +/- 0.04	0.73 +/- 0.03	8.60	0.61
Shamrock Mine	4	1.72 +/- 0.08	1.11 +/- 0.05	0.23 +/- 0.12	8.15	-0.74
Shenandoah NP	4	1.66 +/- 0.10	0.97 +/- 0.03	0.60 +/- 0.05	7.42	0.03
Shining Rock Wilderness	4	1.59 +/- 0.14	0.98 +/- 0.04	0.35 +/- 0.17	9.40	-0.92
Sierra Ancha	4	1.36 +/- 0.06	1.00 +/- 0.05	0.25 +/- 0.09	8.78	-0.43
Sikes	4	1.66 +/- 0.06	1.09 +/- 0.03	0.03 +/- 0.12	6.96	-0.35
Sipsy Wilderness	4	1.80 +/- 0.06	0.94 +/- 0.03	0.36 +/- 0.04	5.81	-0.15
Snoqualmie Pass	4	1.71 +/- 0.05	1.05 +/- 0.07	0.28 +/- 0.07	10.01	-0.72
St. Marks	4	1.69 +/- 0.08	1.05 +/- 0.03	-0.23 +/- 0.23	7.05	-0.12
Starkey	4	1.50 +/- 0.03	1.09 +/- 0.07	0.76 +/- 0.04	8.75	-0.05
Sula Peak	4	1.62 +/- 0.05	1.08 +/- 0.07	0.22 +/- 0.14	11.05	-1.08
Sycamore Canyon	4	1.26 +/- 0.05	1.12 +/- 0.06	0.31 +/- 0.12	7.35	-0.30
Tallgrass	4	1.47 +/- 0.09	0.98 +/- 0.04	0.81 +/- 0.03	8.73	-0.70
Theodore Roosevelt	4	1.54 +/- 0.08	0.93 +/- 0.05	0.99 +/- 0.04	8.36	0.15

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Three Sisters Wilderness	4	1.64 +/- 0.04	1.24 +/- 0.08	0.38 +/- 0.15	9.69	-0.99
Thunder Basin	4	1.78 +/- 0.08	0.92 +/- 0.06	0.61 +/- 0.07	7.24	-0.27
Tonto NM	4	1.62 +/- 0.07	1.06 +/- 0.04	0.23 +/- 0.05	6.71	-0.32
Trinity	4	1.56 +/- 0.04	1.19 +/- 0.10	0.37 +/- 0.08	9.89	-0.92
UL Bend	4	1.85 +/- 0.07	0.89 +/- 0.05	0.91 +/- 0.05	9.37	-0.25
Upper Buffalo Wilderness	4	1.64 +/- 0.08	0.99 +/- 0.04	0.65 +/- 0.04	7.83	-0.49
Viking Lake	4	1.44 +/- 0.09	1.03 +/- 0.04	1.03 +/- 0.02	6.44	-0.11
Voyageurs NP #2	4	1.50 +/- 0.08	0.91 +/- 0.04	1.00 +/- 0.03	8.54	-0.51
Weminuche Wilderness	4	1.35 +/- 0.12	1.05 +/- 0.07	0.69 +/- 0.27	13.73	-0.55
Wheeler Peak	4	1.81 +/- 0.08	0.98 +/- 0.05	0.14 +/- 0.16	11.14	-1.58
White Mountain	4	1.40 +/- 0.10	1.12 +/- 0.05	0.64 +/- 0.07	9.57	-0.01
White Pass	4	1.90 +/- 0.08	1.20 +/- 0.07	-0.19 +/- 0.21	14.93	-1.02
White River NF	4	1.62 +/- 0.17	1.16 +/- 0.08	0.20 +/- 0.33	16.34	-0.39
Wichita Mountains	4	1.62 +/- 0.09	1.04 +/- 0.04	0.70 +/- 0.03	8.41	-1.04
Wind Cave	4	1.32 +/- 0.08	1.08 +/- 0.07	0.37 +/- 0.08	11.75	0.23
Yellowstone NP 2	4	1.51 +/- 0.08	1.11 +/- 0.08	0.52 +/- 0.07	12.26	-1.93
Zion Canyon	4	1.78 +/- 0.07	1.09 +/- 0.05	0.29 +/- 0.07	8.62	-0.58

Table S6. Quarter-specific regression results flagged for single outlier year (n = 28) or temporal trend (n = 7) in residual errors. Values in parentheses represent regression results when the outlier year was removed from the dataset. Regressions for which all coefficients changed by less than 0.1 when the outlier year was removed are highlighted in gray. These 10 cases are regarded as high confidence results so they also appear in Table S5.

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)	outlier year
Agua Tibia	1	1.38 +/- 0.12 (1.30 +/- 0.16)	1.08 +/- 0.08 (1.15 +/- 0.09)	0.56 +/- 0.04 (0.52 +/- 0.05)	9.52	0.07	2002
Salt Creek	1	0.74 +/- 0.29 (0.71 +/- 0.17)	1.22 +/- 0.12 (1.03 +/- 0.07)	0.98 +/- 0.07 (1.08 +/- 0.05)	10.70	-0.26	2004
Shamrock Mine	1	1.52 +/- 0.16 (1.64 +/- 0.05)	1.06 +/- 0.07 (0.94 +/- 0.03)	0.43 +/- 0.11 (0.22 +/- 0.14)	10.59	0.08	2002
St. Marks	1	1.63 +/- 0.06	1.02 +/- 0.03	0.06 +/- 0.19	7.53	-0.47	N/A
Big Bend NP	2	2.16 +/- 0.12 (2.18 +/- 0.13)	0.96 +/- 0.03 (0.93 +/- 0.03)	0.24 +/- 0.28 (0.34 +/- 0.26)	6.72	0.19	2002
Blue Mounds	2	1.67 +/- 0.09 (1.67 +/- 0.10)	0.95 +/- 0.04 (1.00 +/- 0.04)	0.74 +/- 0.04 (0.63 +/- 0.05)	9.62	-0.23	2003
Boundary Waters Canoe Area	2	1.69 +/- 0.06 (1.98 +/- 0.06)	1.06 +/- 0.03 (0.94 +/- 0.03)	0.20 +/- 0.10 (0.28 +/- 0.09)	8.25	-0.01	2003
Brigantine NWR	2	2.07 +/- 0.11 (2.17 +/- 0.13)	0.99 +/- 0.03 (0.97 +/- 0.03)	0.35 +/- 0.13 (0.31 +/- 0.13)	6.98	-0.26	2008
Bryce Canyon NP	2	1.62 +/- 0.06 (1.60 +/- 0.06)	0.97 +/- 0.05 (1.08 +/- 0.05)	0.27 +/- 0.15 (0.19 +/- 0.16)	7.56	-0.21	2005
Caney Creek	2	1.87 +/- 0.10 (1.87 +/- 0.09)	1.03 +/- 0.03 (1.00 +/- 0.03)	0.01 +/- 0.16 (-0.08 +/- 0.14)	6.51	-0.15	2002
Connecticut Hill	2	1.28 +/- 0.14	1.15 +/- 0.03	0.40 +/- 0.10	7.92	-0.64	N/A
El Dorado Springs	2	1.78 +/- 0.07 (1.78 +/- 0.06)	0.98 +/- 0.03 (0.96 +/- 0.02)	0.27 +/- 0.06 (0.29 +/- 0.05)	6.24	-0.24	2002
Fort Peck	2	1.95 +/- 0.08 (1.91 +/- 0.08)	0.90 +/- 0.04 (0.92 +/- 0.04)	0.28 +/- 0.06 (0.28 +/- 0.06)	7.60	0.41	2002
Great Gulf	2	1.70 +/- 0.07 (1.63 +/- 0.07)	1.06 +/- 0.03 (1.06 +/- 0.03)	0.04 +/- 0.33 (0.10 +/- 0.34)	7.36	-0.15	2007

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)	outlier year
Wilderness							
Monture	2	1.52 +/- 0.05 (1.65 +/- 0.06)	0.90 +/- 0.08 (0.96 +/- 0.08)	2.23 +/- 0.79 (1.80 +/- 0.72)	10.09	0.43	2004
New York City	2	1.75 +/- 0.14	0.99 +/- 0.05	0.58 +/- 0.08	6.91	-0.62	N/A
Northern Cheyenne	2	2.06 +/- 0.07 (2.07 +/- 0.07)	0.96 +/- 0.04 (0.93 +/- 0.03)	0.13 +/- 0.09 (0.14 +/- 0.07)	7.45	-0.09	2002
Okefenokee NWR	2	1.61 +/- 0.07 (1.65 +/- 0.06)	1.00 +/- 0.03 (1.02 +/- 0.03)	0.13 +/- 0.22 (0.12 +/- 0.19)	6.98	0.01	2005
Swanquarter	2	2.03 +/- 0.08	0.93 +/- 0.02	0.28 +/- 0.14	6.02	-0.61	N/A
Boundary Waters Canoe Area	3	1.81 +/- 0.04 (1.89 +/- 0.05)	0.99 +/- 0.04 (0.96 +/- 0.03)	1.08 +/- 0.61 (1.03 +/- 0.53)	7.84	-0.64	2003
Bridgton	3	1.84 +/- 0.06 (1.89 +/- 0.07)	1.14 +/- 0.02 (1.11 +/- 0.03)	-0.48 +/- 0.45 (-0.73 +/- 0.45)	6.90	-0.22	2002
Cadiz	3	1.91 +/- 0.17 (1.83 +/- 0.16)	1.07 +/- 0.03 (1.07 +/- 0.03)	-0.05 +/- 0.55 (0.02 +/- 0.59)	7.81	-0.50	2002
Caney Creek	3	1.81 +/- 0.08 (1.82 +/- 0.07)	0.97 +/- 0.03 (0.96 +/- 0.02)	0.45 +/- 0.28 (0.31 +/- 0.24)	6.07	-0.18	2002
Chassahowitzka NWR	3	2.11 +/- 0.17	0.93 +/- 0.04	-0.56 +/- 0.35	6.98	-0.21	N/A
Guadalupe Mountains NP	3	1.76 +/- 0.13 (1.93 +/- 0.13)	0.98 +/- 0.04 (0.97 +/- 0.04)	0.57 +/- 0.29 (0.67 +/- 0.31)	7.13	-0.19	2005
Ikes Backbone	3	1.33 +/- 0.10 (1.61 +/- 0.12)	1.01 +/- 0.08 (1.03 +/- 0.08)	0.56 +/- 0.34 (0.28 +/- 0.33)	10.26	-0.24	2005
Mesa Verde NP	3	1.90 +/- 0.09 (1.97 +/- 0.08)	1.11 +/- 0.08 (1.07 +/- 0.08)	-0.29 +/- 0.52 (0.05 +/- 0.64)	9.03	0.14	2002
Phoenix	3	1.19 +/- 0.07 (1.45 +/- 0.07)	1.08 +/- 0.06 (0.99 +/- 0.05)	0.29 +/- 0.18 (0.22 +/- 0.15)	6.20	0.03	2004
Thunder Basin	3	1.88 +/- 0.04	1.14 +/- 0.06	-0.74 +/- 0.50	5.87	-0.78	N/A
Badlands NP	4	1.22 +/- 0.08 (1.35 +/- 0.08)	1.02 +/- 0.06 (0.93 +/- 0.05)	0.49 +/- 0.08 (0.60 +/- 0.07)	11.60	-0.34	2007
Ikes Backbone	4	1.28 +/- 0.06 (1.62 +/- 0.08)	1.06 +/- 0.05 (1.04 +/- 0.05)	0.44 +/- 0.05 (0.32 +/- 0.05)	10.74	0.90	2005
Mesa Verde NP	4	1.79 +/- 0.13 (1.65 +/- 0.11)	1.14 +/- 0.08 (1.21 +/- 0.06)	0.32 +/- 0.12 (0.23 +/- 0.13)	11.63	0.32	2002

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)	outlier year
Salt Creek	4	1.17 +/- 0.20 (1.26 +/- 0.15)	1.16 +/- 0.09 (0.99 +/- 0.07)	0.93 +/- 0.08 (1.03 +/- 0.06)	10.45	-0.17	2003
Swanquarter	4	1.67 +/- 0.12 (1.62 +/- 0.12)	0.98 +/- 0.03 (0.99 +/- 0.03)	0.02 +/- 0.15 (-0.08 +/- 0.15)	6.28	-0.13	2008
Yosemite NP	4	1.56 +/- 0.03	1.05 +/- 0.06	0.59 +/- 0.05	8.93	-1.00	N/A

Table S7. Quarter-specific regression results flagged because of physically unrealistic coefficients. Four cases with a \* are already flagged as low confidence due to an influential outlier year or temporal trend in  $\varepsilon_i$  (see Table S6). New York City and Washington D.C. regressions from all quarters are included here because their multiyear  $\beta_{soil}$  values are physically unrealistic (See Table S2).

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
Badlands NP	1	<b>0.54 +/- 0.07</b>	1.03 +/- 0.04	0.72 +/- 0.08	12.00	1.42
Cloud Peak	1	<b>0.93 +/- 0.30</b>	0.98 +/- 0.09	0.83 +/- 0.13	16.48	0.56
Fort Peck	1	<b>0.99 +/- 0.17</b>	0.87 +/- 0.05	0.90 +/- 0.04	11.01	-0.93
Hance Camp at Grand Canyon NP	1	<b>0.45 +/- 0.08</b>	1.51 +/- 0.05	0.59 +/- 0.06	11.21	1.56
Ikes Backbone	1	<b>0.76 +/- 0.08</b>	1.34 +/- 0.07	0.34 +/- 0.05	11.66	1.11
Kalmiopsis	1	1.37 +/- 0.03	0.75 +/- 0.08	<b>1.66 +/- 0.30</b>	9.25	-0.75
Meadview	1	<b>0.87 +/- 0.13</b>	1.33 +/- 0.07	0.39 +/- 0.07	10.61	1.72
New York City	1	1.59 +/- 0.14	0.76 +/- 0.05	1.34 +/- 0.06	4.82	-0.02
North Absaroka	1	<b>0.90 +/- 0.19</b>	1.17 +/- 0.07	0.61 +/- 0.09	14.93	-0.26
Petrified Forest NP	1	<b>0.96 +/- 0.18</b>	0.90 +/- 0.12	1.05 +/- 0.20	14.88	2.40
Phoenix	1	1.31 +/- 0.04	<b>0.57 +/- 0.10</b>	0.67 +/- 0.04	6.01	0.20
Rocky Mountain NP	1	<b>0.64 +/- 0.17</b>	1.00 +/- 0.09	0.89 +/- 0.06	13.72	-0.24
Saguaro NM	1	<b>0.93 +/- 0.13</b>	1.11 +/- 0.08	0.50 +/- 0.07	8.59	1.52
Salt Creek*	1	<b>0.74 +/- 0.29</b>	1.22 +/- 0.12	0.98 +/- 0.07	10.70	-0.26
San Geronio Wilderness	1	<b>0.81 +/- 0.15</b>	0.84 +/- 0.09	0.93 +/- 0.03	9.02	0.40

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
San Rafael	1	<b>0.92 +/- 0.08</b>	0.94 +/- 0.07	0.77 +/- 0.06	11.48	-0.40
Sequoia NP	1	<b>0.72 +/- 0.14</b>	0.84 +/- 0.13	1.10 +/- 0.03	9.20	-0.22
Snoqualmie Pass	1	<b>0.23 +/- 0.06</b>	<b>1.55 +/- 0.09</b>	1.02 +/- 0.07	19.60	0.86
Washington D.C.	1	1.55 +/- 0.06	0.88 +/- 0.03	1.08 +/- 0.04	5.59	-0.23
Wheeler Peak	1	<b>0.58 +/- 0.26</b>	1.27 +/- 0.17	0.54 +/- 0.23	15.48	0.02
Yellowstone NP 2	1	<b>0.92 +/- 0.11</b>	1.00 +/- 0.06	0.74 +/- 0.05	12.55	-1.07
Bridgton	2	1.85 +/- 0.08	1.01 +/- 0.03	<b>-0.43 +/- 0.23</b>	7.50	-0.23
Chiricahua NM	2	<b>1.00 +/- 0.10</b>	1.15 +/- 0.06	0.88 +/- 0.26	7.17	0.01
Death Valley NP	2	1.82 +/- 0.16	0.71 +/- 0.08	<b>1.41 +/- 0.28</b>	6.26	-0.26
Douglas	2	1.04 +/- 0.17	1.10 +/- 0.14	<b>1.49 +/- 0.56</b>	5.27	0.36
Gila Wilderness	2	1.64 +/- 0.06	<b>0.52 +/- 0.07</b>	<b>2.43 +/- 0.29</b>	7.25	-0.37
Hells Canyon	2	1.56 +/- 0.07	0.81 +/- 0.11	<b>1.87 +/- 0.65</b>	9.47	-0.96
Mohawk Mt.	2	<b>0.76 +/- 0.12</b>	1.18 +/- 0.04	0.72 +/- 0.30	11.47	0.95
Monture*	2	1.52 +/- 0.05	0.90 +/- 0.08	<b>2.23 +/- 0.79</b>	10.09	0.43
New York City*	2	1.75 +/- 0.14	0.99 +/- 0.05	0.58 +/- 0.08	6.91	-0.62
Sawtooth NF	2	1.65 +/- 0.07	1.03 +/- 0.13	<b>-2.00 +/- 1.03</b>	9.74	-1.19
Shining Rock Wilderness	2	1.88 +/- 0.17	1.05 +/- 0.04	<b>-0.64 +/- 0.33</b>	7.89	-0.68
Sycamore Canyon	2	1.59 +/- 0.10	0.73 +/- 0.09	<b>1.52 +/- 0.22</b>	6.20	-0.15
Washington D.C.	2	2.05 +/- 0.10	1.00 +/- 0.03	0.52 +/- 0.07	6.91	-0.52
Bandelier NM	3	1.83 +/- 0.08	1.04 +/- 0.05	<b>-0.96 +/- 0.56</b>	7.76	-0.58
Capitol Reef NP	3	2.04 +/- 0.07	1.04 +/- 0.05	<b>-0.81 +/- 0.46</b>	7.22	-0.86
Casco Bay	3	1.69 +/- 0.05	1.29 +/- 0.03	<b>-0.76 +/- 0.25</b>	7.21	-0.65
Chassahowitzka NWR*	3	2.11 +/- 0.17	0.93 +/- 0.04	<b>-0.56 +/- 0.35</b>	6.98	-0.21
Cloud Peak	3	1.96 +/- 0.04	1.14 +/- 0.07	<b>-0.89 +/- 0.30</b>	6.36	-0.78
Cohutta	3	1.30 +/- 0.17	1.04 +/- 0.04	<b>2.92 +/- 0.74</b>	6.43	-0.10
Death Valley NP	3	1.75 +/- 0.06	0.69 +/- 0.05	<b>1.88 +/- 0.30</b>	7.24	-0.02
Flathead	3	1.82 +/- 0.03	1.09 +/- 0.10	<b>-1.26 +/- 0.57</b>	7.27	-1.13
Glacier NP	3	1.90 +/- 0.06	1.48 +/- 0.19	<b>-4.94 +/- 1.74</b>	8.00	-0.14

Site	quarter	$\beta_{oc}$	$\beta_{sulf}$	$\beta_{nit}$	NME (%)	NMB (%)
James River Face Wilderness	3	2.09 +/- 0.11	1.04 +/- 0.02	<b>-0.41 +/- 0.25</b>	6.22	-0.24
M.K. Goddard	3	1.77 +/- 0.08	1.13 +/- 0.02	<b>-0.46 +/- 0.29</b>	6.84	-0.26
Mohawk Mt.	3	1.75 +/- 0.09	1.03 +/- 0.03	<b>-0.55 +/- 0.21</b>	8.31	0.14
New York City	3	1.75 +/- 0.14	1.13 +/- 0.04	0.38 +/- 0.17	7.61	-0.36
Point Reyes National Seashore	3	1.51 +/- 0.11	0.86 +/- 0.05	<b>1.49 +/- 0.16</b>	9.38	-0.44
Presque Isle	3	1.85 +/- 0.04	1.05 +/- 0.02	<b>-1.28 +/- 0.52</b>	6.12	-0.39
Proctor Maple R. F.	3	2.03 +/- 0.06	1.05 +/- 0.02	<b>-1.05 +/- 0.52</b>	6.09	0.24
Quaker City	3	1.90 +/- 0.13	1.09 +/- 0.02	<b>-0.70 +/- 0.40</b>	6.59	0.01
Saguaro West	3	1.68 +/- 0.35	0.70 +/- 0.14	<b>2.11 +/- 0.63</b>	10.17	-1.59
Theodore Roosevelt	3	1.95 +/- 0.05	1.07 +/- 0.05	<b>-0.46 +/- 0.18</b>	6.58	-0.90
Trinity	3	1.66 +/- 0.04	0.83 +/- 0.08	<b>1.75 +/- 0.36</b>	9.51	1.35
Voyageurs NP #2	3	1.54 +/- 0.06	1.08 +/- 0.06	<b>1.90 +/- 1.20</b>	10.12	-0.18
Washington D.C.	3	2.05 +/- 0.14	1.08 +/- 0.03	-0.09 +/- 0.27	7.36	-0.35
Addison Pinnacle	4	<b>0.26 +/- 0.07</b>	1.19 +/- 0.03	1.11 +/- 0.06	9.05	0.14
Cape Romain NWR	4	1.81 +/- 0.09	1.03 +/- 0.04	<b>-0.63 +/- 0.19</b>	7.98	-0.56
New York City	4	1.42 +/- 0.11	0.87 +/- 0.05	1.17 +/- 0.06	6.02	0.07
Sequoia NP	4	1.43 +/- 0.07	<b>0.45 +/- 0.13</b>	1.04 +/- 0.02	8.85	-0.19
Washington D.C.	4	1.48 +/- 0.07	0.95 +/- 0.04	1.06 +/- 0.06	6.95	-0.42



## S6. References

- Colberg, C. A., Luo, B. P., Wernli, H., Koop, T., and Peter, T.: A novel model to predict the physical state of atmospheric  $\text{H}_2\text{SO}_4/\text{NH}_3/\text{H}_2\text{O}$  aerosol particles, *Atmospheric Chemistry and Physics*, 3, 909-924, 2003.
- Fuller, W. A.: *Measurement error models*, John Wiley & Sons, New York, 1987.
- Hand, J. L., and Malm, W. C.: Review of the IMPROVE equation for estimating ambient light extinction coefficients - final report, Colorado State University, CIRA, 146, 2006.
- Khlystov, A., Stanier, C. O., Takahama, S., and Pandis, S. N.: Water content of ambient aerosol during the Pittsburgh air quality study, *Journal of Geophysical Research-Atmospheres*, 110, 10.1029/2004jd004651, 2005.
- McDade, C. E.: IMPROVE standard operating procedure, Crocker Nuclear Laboratory, University of California, Davis, CASOP 351-2, 258, 2008.
- Pinder, R. W., Dennis, R. L., and Bhavsar, P. V.: Observable indicators of the sensitivity of  $\text{PM}_{2.5}$  nitrate to emission reductions—Part I: Derivation of the adjusted gas ratio and applicability at regulatory-relevant time scales, *Atmospheric Environment*, 42, 1275-1286, doi:10.1016/j.atmosenv.2007.10.039, 2008.
- Wexler, A. S., and Clegg, S. L.: Atmospheric aerosol models for systems including the ions  $\text{H}^+$ ,  $\text{NH}_4^+$ ,  $\text{Na}^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{Br}^-$ , and  $\text{H}_2\text{O}$ , *Journal of Geophysical Research-Atmospheres*, 107, 14, 4207 10.1029/2001jd000451, 2002.
- White, W. H.: IMPROVE data advisory: Shift in EC/OC split with 1 January 2005 TOR hardware upgrade, 2007.