Contributions of Biomass Burning and Other Sources to Fine Particulate Carbon at Rural Locations Throughout the United States

Extended Abstract # 77

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INTRODUCTION

Carbonaceous compounds are a significant component of fine particulate matter (PM2.5) and contribute to haze in class I areas. The Regional Haze Rule sets the goal of returning visibility on the worst haze days to natural conditions by 2064. To achieve this goal, it is necessary to understand the contributions of natural and anthropogenic sources to the particulate carbon in class I areas.

A receptor-oriented, Lagrangian particle dispersion model with highly simplified physical/chemical processes was developed to simulate primary and secondary PM2.5 total carbon (TC) concentrations in rural areas[1]. The model uses readily available meteorological and emission inputs and simulates the contributions from various source types, including wildfires and area sources. Modeling evaluation revealed potential spatial and temporal biases in the modeled TC concentrations and source attributions. In this work, the biases are reduced by incorporating the modeled source attribution results into a hybrid model in the receptor framework.

MODELED TC SOURCE ATTRIBUTIONS

The receptor-oriented, Lagrangian particle dispersion model is based on the CAPITA (Center for Air Pollution Impact and Trend Analysis) Monte Carlo model (CMC)[2] and simulates the contributions from eight source categories, including biomass burning and secondary organic carbon (SOC) from vegetation. The model is based on 6-day-back airmass histories generated using 40-km meteorological data from the Eta Data Assimilation System (EDAS) and the Western Regional Air Partnership (WRAP) 36-km emission inventory, aggregated to 24-h emission rates. The WRAP biomass burning emissions were replaced by the National Center for Atmospheric Research (NCAR) regional fire emissions model version 2.0[3]. TC removal and formation mechanisms are simulated using a simplified parameterization of atmospheric processes based on pseudo-first-order rate equations. The rate coefficients are empirical functions of meteorological parameters derived from measured, modeled, and literature data. These functions were optimized such that the simulated TC concentrations reproduce the average spatial and temporal patterns in measured 2008 TC concentrations from the IMPROVE monitoring network, as well as measured SOC fractions at two eastern U.S. sites.

The optimized model was used to simulate 2006–2008 TC at 148 rural and 14 urban IMPROVE monitoring sites. The contributions from the modeled source types average over each month and the three years of simulations are presented in Figure 1 for three U.S. subregions. As illustrated, the CMC model was able to reproduce the seasonality well throughout the rural U.S. In regions, such as the Northeast, it also reproduced the average TC concentrations well. However, systematic biases were also evident. For example, in much of the eastern U.S., e.g., Appalachia, the summer TC was underestimated indicating that the contributions from one or more source types were underestimated. Also, contributions from biomass burning could be significantly over- or underestimated due to errors in the biomass burning emissions and airmass transport. This is clearly evident in Figure 1, where in March at Appalachia and June–October along the California Coast the simulated contributions from fires were larger than the measured TC. While biases in the simulated TC are evident in Figure 1, potential compensating errors between contributions from different source types are not. For example, the good correspondence between the measured and simulated TC in the Northeast could hide a systematic underestimation from vegetation balanced by an overestimation in contributions from area sources.

HYBRID RECEPTOR MODEL

The conservation of mass for a single species such as TC can be defined as the sum of the contributions from all sources. For the simulation of measured TC concentrations by modeled source contributions, the conservation of mass can be written as

Equation 1. $C_i = \sum_j g_{ij} + e_i$

where:

C_i = concentration of the i^{th} measurement at the receptor site, units are µg/m^3
g_{ij} = modeled contribution of the k^{th} source to the receptor on the i^{th} measurement, units are µg/m^3
e_i = residual for the i^{th} measurement, units are µg/m^3

The distribution of residuals is the error of the system due to errors in the measurements, mismatch between the model and measurement spatial and temporal scales, i.e., representativeness error, and errors in the modeling system. The measurement error is thought to be nonbiased, but systematic errors could occur in both the representativeness and modeling errors. The modeling biases are due to errors in the model formulations and model inputs, such as emissions, and could vary for each source category and by space and time. In this application, we modeled the errors as a multiplicative bias for each source type j and group of observations l and an additive random error with zero mean and standard deviation σ. Equation 1 can then be written as

Equation 2. $C_i^l = \sum_j g_{ij}^l s_j^l + \sigma^l$

where:

$s_j^l = \left(1 + a_j^l\right)$ are unitless scaling coefficients and a_j^l are the unitless multiplicative biases

This equation is identical in form to the synthesis inversion equation for emission estimation[4,5], but no physical interpretation is given to the regression coefficients. Provided the measured concentrations and modeled source attributions are known, the scaling coefficient $\boldsymbol{s}$ can be found through a variety of inversion methods. In this work we used the same Bayesian least square regression method as Schichtel[5] to solve equation 2 for $\boldsymbol{s}$. Subsequently, these scaling coefficients were used to refine the source attribution results to reduce the systematic biases.

The Bayesian regression method incorporates prior estimates of the source attribution scaling coefficients and their variances into a weighted least square regression analysis such that

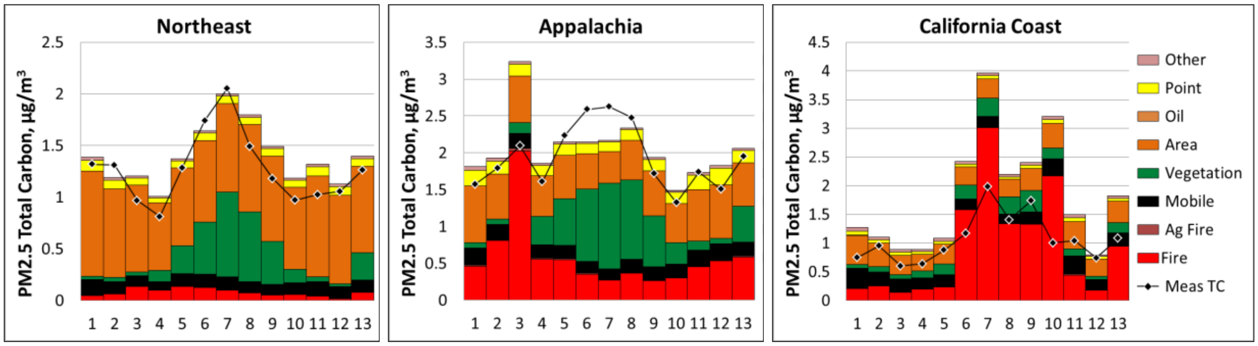
Equation 3. $\boldsymbol{s} = [\boldsymbol{G}^T\boldsymbol{X}\boldsymbol{G} + \boldsymbol{W}]^{-1}[\boldsymbol{G}^T\boldsymbol{X}\boldsymbol{C} + \boldsymbol{W}\boldsymbol{z}]$

Equation 4. The covariance matrix of $\boldsymbol{s}$ is $[\boldsymbol{G}^T\boldsymbol{X}\boldsymbol{G} + \boldsymbol{W}]^{-1}$

where $\boldsymbol{z}$ is the vector of the prior source attribution scaling coefficients and is unitless. $\boldsymbol{W}$ is the inverses of the error covariance matrices for the measured concentrations and has units (µg/m^3)^2. $\boldsymbol{X}$ is the inverse of the error covariance matrices for the prior estimates and is unitless.

This set of equations was solved for each group of observations *l* using an extended data-weighted least squares technique[6,7].

Modeled Source Attributions



Refined Source Attributions Using the Hybrid Model

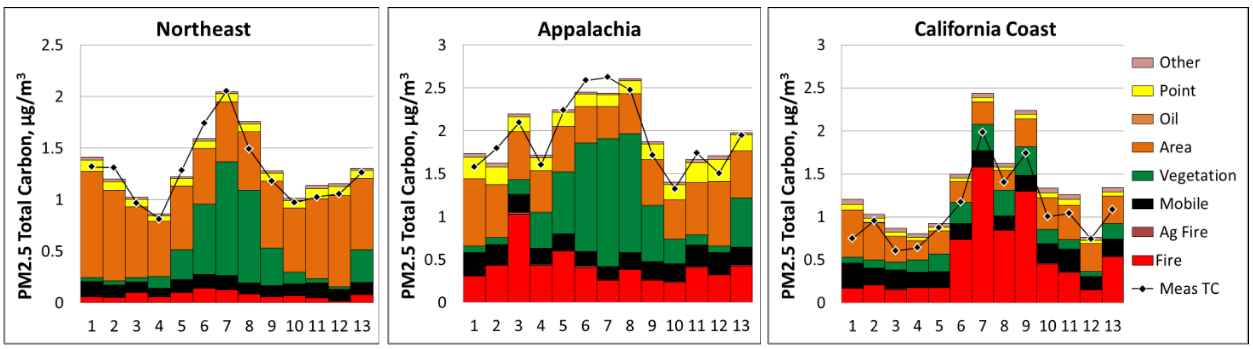


Figure 1. The average contributions of modeled and refined source types to TC for three United States regions. The average measured TC concentrations are also shown. The average values are for each month of the year from 2006 through 2008. Month 1 is January and month 13 is the annual average.

SOURCE ATTRIBUTION REFINEMENT

The CMC model simulates hourly TC concentrations and source contributions. These hourly values were aggregated to the same 24-h periods used in the IMPROVE network. Only data corresponding with IMPROVE's 1 in 3 day sampling schedule were used in the analysis. The CMC model had very poor performance at the fourteen urban, four southern California, and two Washington state sites.

To account for spatial and temporal variation in the model biases, Equations 3 and 4 were solved using data from each monitoring site and its 20 closest neighboring sites for each quarter of the year, with quarter 1 starting in January. This provided a balance between spatial and temporal resolution and enough data for stable inversions. ***W*** was estimated from the uncertainties of the measured TC data which were estimated as $\sigma_c = \sqrt{(0.17 * C_i)^2 + (mdl^2)}$ where *mdl* is the minimum detection limit which was set to[8] 0.35 µg/m^3. Evaluation of the monthly mean modeled and measured TC values found a modeling error of about 50%. Since the errors in the individual source attribution estimates could not be evaluated, it was assumed that all source attribution estimates had a 50% error and ***X*** was derived accordingly. The a priori scaling estimates were all set to 1, i.e., it was assumed the model was unbiased.

The CMC model often had poor performance when impacted by nearby wild or prescribed fires, resulting in large over- or underestimated TC concentrations. These high concentration outliers could bias the regression analysis used in the hybrid model. To reduce this, all measured and simulated TC concentrations greater than 8 µg/m^3 were excluded from the hybrid model runs. Eight µg/m^3 is the 99th TC percentile at the Washington DC site. This is an urban site with little contribution from fires and represents an upper bound on nonfire contributions to TC. This is a conservative estimate for when fires are the dominant contributor to rural TC and affected 1.8% of the data. Although these concentrations were removed from the hybrid model, they were incorporated into the refined source attribution results. This was done by assuming that the difference between the measured TC and the sum of the modeled nonfire source contributions was due to fire. Then the modified source contributions were scaled by the hybrid model scaling coefficients.

RESULTS

The quarterly and annual average source contributions from each source type from the initial model and hybrid model runs were examined. Some compensating biases in the initial model results were found, with average contributions from vegetation underestimated by about 10% over the year and 15% during summer months. This was compensated for by apparent overestimations of contributions from area sources and fire by ~10%. This is evident in the Northeast results (Figure 1), where the refined attributions maintain the excellent agreement with the measured TC concentrations, but the summer contributions from vegetation were increased while the contributions from the area sources were decreased. The low contributing source types, such as oil and gas and point sources, had scale coefficients near 1. This is likely due to the fact that the contributions from these source types were small compared to the error in the system as opposed to these source types being unbiased. The hybrid model also improved the fit between the simulated and measured TC in many regions. As shown in Figure 1, the underestimated TC during the summer months at Appalachian sites by the CMC model was removed by increased TC contributions from vegetation. The large overestimation from biomass burning along the California coast was also removed.

On average over the United States, fires and vegetation accounted for about 60% of the TC. These contributions are seasonal, accounting for about 75% of the TC in quarter 3 and about 33% in quarter 1. In fact, most of the seasonality in the TC is due to these two sources. Vegetation and fire are primarily natural sources, and their contributions represent limits on the reduction of carbonaceous aerosol for improving visibility in these rural areas.

Of anthropogenic sources, area sources were the largest contributors, accounting for about 40% of the TC in quarter 1 and 15% in quarter 3. This is followed by mobile sources, which accounted for 5–15% of the TC. In contrast to vegetation and fire, the anthropogenic sources have the largest contributions during the winter months.

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