IMPACTS OF BIOMASS BURNING EMISSIONS ON AMBIENT $PM_{2.5}$ IN THE SOUTHEASTERN UNITED STATES USING CMAQ

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1. INTRODUCTION

Biomass burning is combustion processes that consume biomass fuels, either through natural processes (e.g. wildfires) or man-made processes (e.g. prescribed burning, agriculture field burning, land clearing, wood burning in fireplace and woodstove, residential leaf burning). During combustion, large amounts of air pollutants, e.g. particulate matter (PM), volatile organic compounds (VOCs), nitrogen oxides (NO_X), ammonia (NH_3), carbon monoxide (CO), etc, can be emitted. In the United States, biomass burning contributes about 35% of the fine particulate matter ($PM_{2.5}$), i.e. PM with aerodynamic diameter less than 2.5 µm emissions (US-EPA, 2004a). Much of the mass emitted is carbonaceous (70-95%) and carbonaceous material is a substantial component of $PM_{2.5}$ in the US (NARSTO, 2003). Previous $PM_{2.5}$ source apportionment studies using both receptor models and emission-based models suggest significant contributions from biomass burning in the southeastern United States (Kim et al., 2003; Kim, 2003; Liu et al., 2005; Park et al., 2006; Zheng et al., 2002; Zheng et al., 2006).

Here, air quality impacts from biomass burning emissions are estimated using Community Multi-scale Air Quality model (CMAQ). Their accuracy is greatly affected by the quality of emission inputs, which are first addressed in this study. Improved understanding of biomass burning emissions are then applied to study air quality impacts from biomass burning during different seasons, characterized by different levels of biomass burning emissions, as well as different meteorological conditions and physical-chemical processes of pollutants in the atmosphere.

2. METHODS

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2.1 Air quality modeling

Four months in 2002 (January, March, May and July) are selected. Specifically, biomass burning emissions during winter and spring are typically much larger than during summer in Georgia, with emissions from prescribed burning (the largest individual biomass burning source) peaking in March, emissions from fireplaces and woodstoves in January, emissions from wildfires in May, and very low biomass burning emissions in July. Air quality in these months is simulated with the Community Multiscale Air Quality (CMAQ) model v. 4.3 using SAPRC-99 chemical mechanism (Byun and Ching, 1999). Results of the fist two days for each month are discarded as model initialization periods. The modeling domain has 19 vertical layers reaching to about 15 km vertically, with a 36 m bottom layer. It covers the southeastern United States at 12-km resolution. Initial and boundary conditions are supplied by simulations on a 36-km resolution grid covering the United States (Unified RPO modeling domain). Meteorological conditions for the episodes are simulated with the NCAR's 5th generation Mesoscale Model (MM5), described in detail elsewhere (Grell et al., 1994; Olerud and Sims, 2003). Emission inventories are obtained from VISTAS (VISTAS, 2005), and then processed through the Carolina Environmental Program's (CEP) Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System v. 2.1 (Houyoux et al., 2000).

Simulations from the above modeling are evaluated by comparing model results with observations collected as part of the Interagency Monitoring of Protected Visual Environments (IMPROVE), the SouthEastern Aerosol Research and Characterization (SEARCH), the Assessment of Spatial Aerosol Composition in Atlanta (ASACA) and the Speciation Trends' Network (STN) networks. Organic carbon (OC) observations are converted to organic matter (OM) by multiplying using a 1.4 factor, which has been widely used, though recent studies suggest it is low (Turpin and Lim, 2001).

Impacts of uncertainties in biomass burning emissions on air quality modeling are investigated in January 2002, since emissions during this episode from the various biomass burning sources are significant. These impacts are further compared with results from a receptor modeling, Chemical Mass Balance (CMB). The CMB modeling employed both organic compounds and elemental species (e.g. trace metals) (Zheng et al., 2002). Relative distributions of organic compounds in source emissions provide additional means to identify source contributions that cannot be uniquely identified by elemental compositions alone.

Air quality impacts from biomass burning emissions are obtained by conducting CMAQ simulations twice: both with and without biomass burning emissions (brute force method). Both primary and secondary impacts are assessed. Source contributions from individual biomass burning sources are accomplished by tracing primary organic matter (the major component as discussed later) emissions from these sources in CMAQ. These tracer emissions are generated by SMOKE using specific speciations, and then input into CMAQ together with other emissions. These tracers are treated as non-reactive species, and go through similar physical processes as other primary carbonaceous aerosol species (Baek et al., 2005).

2.2 Uncertainties in total amount of emissions

Two different emission inventories for wildfires, prescribed and agriculture field burning and land clearing, all specific to Georgia, are employed, including EPA 2001 modeling platform emission inventory (EPA 2001) (US-EPA, 2004a), and VISTAS 2002 (VISTAS, 2005). They are developed using different estimation methods.

2.3 Uncertainties in temporal and spatial characteristics of emissions

Accounting for when and where fires actually occur are important to detailed studies of the impacts of biomass burning on ambient $PM_{2.5}$. Biomass burning emissions in EPA 2001 and VISTAS 2002 have different temporal and spatial resolution. An updated monthly county-level emission inventory for prescribed and agriculture field burning and land clearing is also developed here (UPDATED), using detailed burned area data (GFC, 2005) and same fuel loading and emissions factors as in VISTAS 2002.

2.4 Uncertainties in $PM_{2.5}$ speciation

$PM_{2.5}$ speciation profiles for biomass burning are obtained from EPA (US-EPA, 2004b), with POA as the major species. Current fractions of POA in $PM_{2.5}$ are based on the OC measurements, by multiplying a factor of 1.2-1.4 to account for the other elements bound to C (Fine et al., 2002; Hays et al., 2005). However, molecular level analyses of POA indicate that the POA/OC ratio for wood burning is about 1.9 (Turpin and Lim, 2001). In addition, these analyses only measure less polar organics and don't account for the water-soluble species which comprise 20-80% of the organic aerosol (Saxena and Hildemann, 1996; Sullivan and Weber, 2006). Since more water-soluble organic compounds have higher molecular weight to carbon weight ratios than less water-soluble organic compounds, the POA/OC ratio for biomass burning may be larger than 1.9. As such, current speciation profiles tend to underestimate the POA fractions. Here, POA fractions for wildfires, prescribed burning and wood burning in fireplaces and woodstoves are recalculated by subtracting fractions of EC (f_{EC}), SO4 (f_{SO4}), NO3 (f_{NO3}) and other unspecified mass (f_{other}) from 1:

$$f_{POA} = 1 - f_{EC} - f_{SO4} - f_{NO3} - f_{other} \qquad (1)$$

The fractions of each species for prescribed burning and wildfires are decided by both field and lab measurements (Lee et al., 2005, Hays et al., 2002, Andreae and Merlet, 2001). Speciation for wood burning in fireplaces and woodstoves combustion is updated by recent lab measurements (Fine et al., 2002, Mcdonald et al., 2000). Significantly different emission characteristics from different crops, e.g. wheat and rice, have been observed in lab measurements (Hays et al., 2005), though lack of agriculture burned area by crops and emission factors for other crops inhibits further improvements. Land clearing appears to emit more visible smoke (GFC, 2005), but specific speciation profiles are not available.

3. RESULTS AND DISCUSSION

3.1 Impacts of biomass burning emission uncertainties on air quality modeling

Emissions from wildfires, prescribed and agriculture field burning, and land clearing in Georgia during 2002 differ significantly in EPA 2001 and VISTAS 2002 emission inventories, due to different estimation methods and input data used (Figure 1). There are around 18,000 tons/year more $PM_{2.5}$ emissions in VISTAS 2002 than EPA

2001. In EPA 2001, emissions from each of the four biomass burning sources are similar. Whereas the difference between emissions from individual biomass burning sources in VISTAS 2002 is fairly large, and prescribed burning contributes about 70% of the total emissions from the four sources combined. These emissions are much larger than emissions from other biomass burning sources (e.g. wood burning in fireplaces and woodstoves).

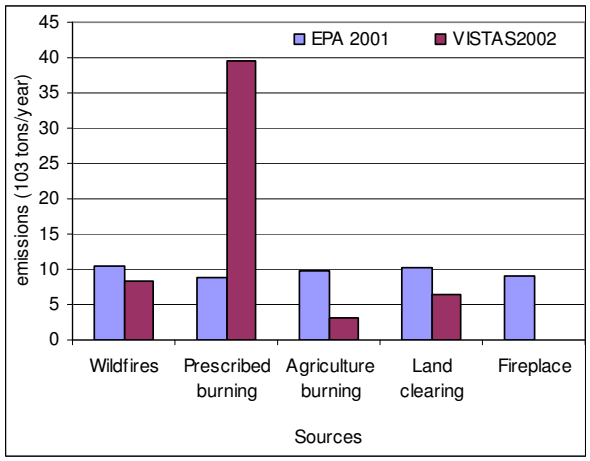

Figure 1 $PM_{2.5}$ emissions from biomass burning within Georgia during 2002 in different emission inventories (10^3 tons/year)

Different temporal and spatial distributions of $PM_{2.5}$ emissions are also found for simulations with the two different emission inventories, as well as corresponding $PM_{2.5}$ concentrations. POA emissions in January with EPA 2001 are more intense in the Atlanta area than with VISTAS 2002, which have denser emissions in the southwestern Georgia (Figure 2). Responsive POA concentrations with EPA 2001 are higher than ones with VISTAS 2002 by 0.7 μg/m^3 for the $PM_{2.5}$ non-attainment area in Atlanta, and less than those with VISTAS 2002 by 0.6 μg/m^3 for the whole Georgia (Figure 2). Sketchy data used in emission estimates by EPA 2001 can partially explain the large difference between inventories, and estimates in VISTAS 2002 agree well with the fact that most forest fires in Georgia are conducted under control, i.e. prescribed (GFC, 2005). Total annual emissions from biomass burning in UPDATED are same as VISTAS 2002. However, significant differences in spatial distributions of POA emissions and concentrations using the two inventories have been found. These differences can be attributed to the application of county-specific monthly temporal profiles.

(a) EPA 2001

(b) VISTAS 2002

(c) UPDATED

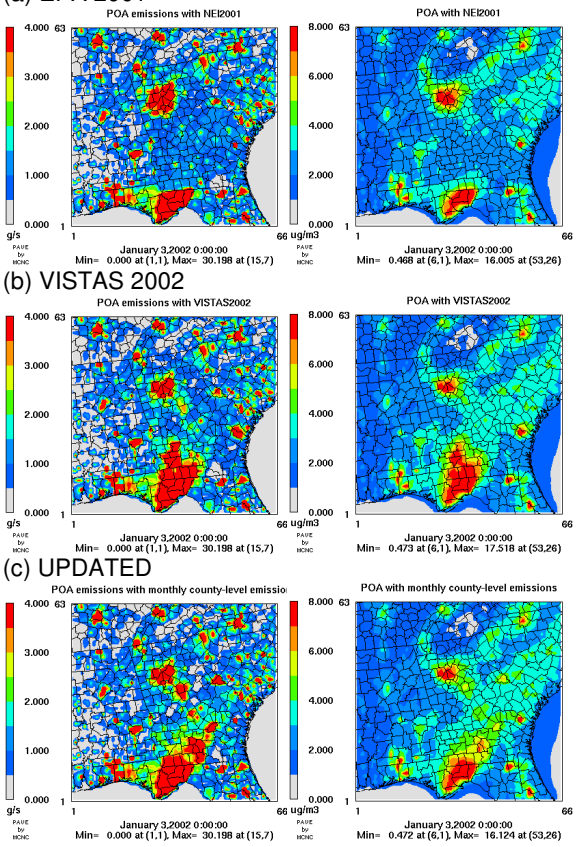

Figure 2 Monthly-average POA emissions (left column) and concentrations (right column) using different emission inventories during January 2002

POA fractions derived using equation 1 increase 0.13 for wildfires and prescribed burning, and 0.30 for wood burning in fireplaces and woodstoves, along with decreased fractions of other $PM_{2.5}$ components (Table 1). Given POA and OC ratios are larger than 1.9 for biomass burning emissions according to molecular level analyses, updated OC fractions are less than 0.5. They are much lower than the measurements (0.602-0.788 for forest fires and 0.530-0.718 for residential wood burning based on the measurements mentioned above).

Table 1 $PM_{2.5}$ speciation profiles for biomass burning sources

		POA	EC
Wildfire & Prescribed burn	EPA	0.770	0.160
	Updated	0.898	0.056
Fireplace and Woodstove	EPA	0.566	0.108
	Updated	0.865	0.108

Model performance statistics with VISTAS 2002 are slightly better than those with EPA 2001,

whereas they are almost same as those with UPDATED due to the fact that most monitors are outside of the affected regions (Table 2). Updated speciation profiles improve EC performance and deteriorate OM performance. $PM_{2.5}$ source contributions from biomass burning during January using the UPDATED emission inventory and improved speciation profiles are 13.2 μg/m^3 at JST and 4.2 μg/m^3 at YRK (as $PM_{2.5}$), which are much higher than the results from CMB analysis (1.59 μg/m^3 at JST and 0.450 μg/m^3 at YRK. Further evaluation of source contributions from individual biomass burning shows that emissions from wood burning in fireplaces and woodstoves have the largest air quality impact at JST and YRK, contributing around 90% of POA from all biomass burning at JST and 70% at YRK during January. It is resulted from its compact emissions in the Atlanta area, despite small annual emissions in Georgia. These emissions have relatively large uncertainties because of estimation methods and data used, and should be improved. Since it is out of the range of this study and 90% emission reduction is simply applied to avoid their large impacts. The reduction improves the model performance for both EC and OM (Table 2), though discrepancies between simulations and observations still remain due to other sources of uncertainties and are not investigated here.

Table 2 EC and OM performance with different emission uncertainties during January 2002 (Means of EC and OM observations are 0.84 μg/m^3 and 4.83 μg/m^3 respectively.)

Species	Uncertainties in emissions	SIM_mean (μg/m^3)	MFB (%)	MFE (%)
EC	EPA 2001	1.13	22.1	58.5
	VISTAS 2002 / UPDATED	1.13	22.0	58.4
	UPDATED & Speciation	1.03	13.3	55.3
	UPDATED & Speciation 90% reduction	0.86	3.9	50.2
OM	EPA 2001	6.02	18.3	54.9
	VISTAS 2002 / UPDATED	5.90	16.3	53.6
	UPDATED & Speciation	6.83	26.3	57.9
	UPDATED & Speciation 90% reduction	5.34	11.9	48.9

3.2 Seasonal source contributions from biomass burning

The UPDATED emission inventory, 90% reduction of emission from wood burning in fireplaces and woodstoves and the improved speciation profiles, are applied in all episodes. Overall performance of simulated $PM_{2.5}$ species during these episodes is fairly good, except for OM during May and July (Figure 3). Underestimation in OM is common in the current CMAQ model, and it is likely due to underestimation of secondary organic aerosol (SOA) formation from biogenic sesquiterpene and isoprene emissions, and polymerization of SOA into nonvolatile particles (Morris et al., 2005).

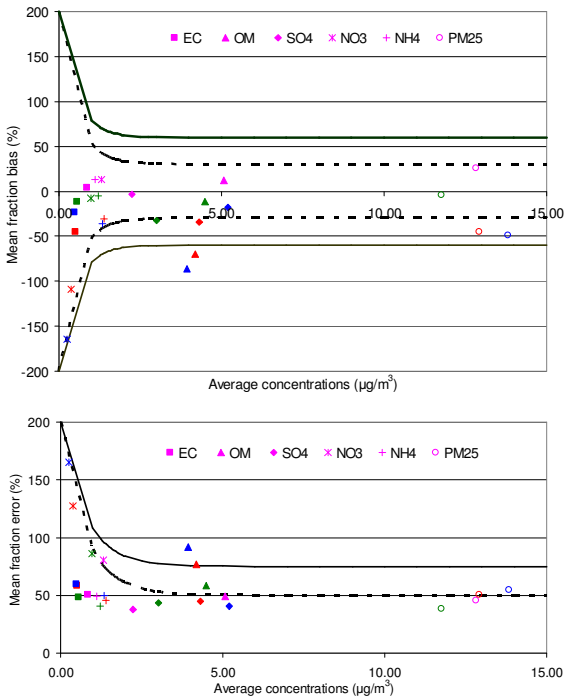

Figure 3 Air quality modeling performance of total and speciated PM2.5 during four months in 2002. (Solid and dashed lines are suggested criteria from Boylan, 2005) January (purple), March (green), May (red), July (blue)

$PM_{2.5}$ source contributions from biomass burning usually coincide with peak $PM_{2.5}$ concentrations and have large temporal and spatial variation, with January and March being impacted the most and negligible impacts during May and July (Figure 4). Biomass burning emissions respectively contribute 3.0, 5.1, 0.8, and 0.3 μg/m^3 of $PM_{2.5}$, constituting 25%, 40%, 9% and 4% of total $PM_{2.5}$ during January, March, May and July for the whole modeling domain. Analyses using receptor models, e.g Positive Matrix Factorization (PMF) and Chemical Mass Balance (CMB), indicated similar seasonal trends (Liu et al., 2005; Zheng et al., 2002). $PM_{2.5}$ source contributions from biomass burning during January and March are concentrated in southwestern Georgia, where large amounts of open burning are conducted. $PM_{2.5}$ concentrations caused by biomass burning in the Georgia $PM_{2.5}$ non-

attainment area are 1.5 and 2.6 μg/m^3 respectively in January and March. About 90% of $PM_{2.5}$ concentrations caused by biomass burning are carbonaceous, including about 85%, 7.5% and 7.5% of POA, SOA and EC respectively. In addition, NH_3 emissions from biomass burning also lead to increased NH_4, contributing about additional 2% of $PM_{2.5}$. Extra NH_3 and NO_X emissions from biomass burning lead to increased NO_3 as well (about 4% of $PM_{2.5}$). No significant increase in SO_4 levels is observed, as indicated in a recent study in Texas (Buzcu et al., 2006).

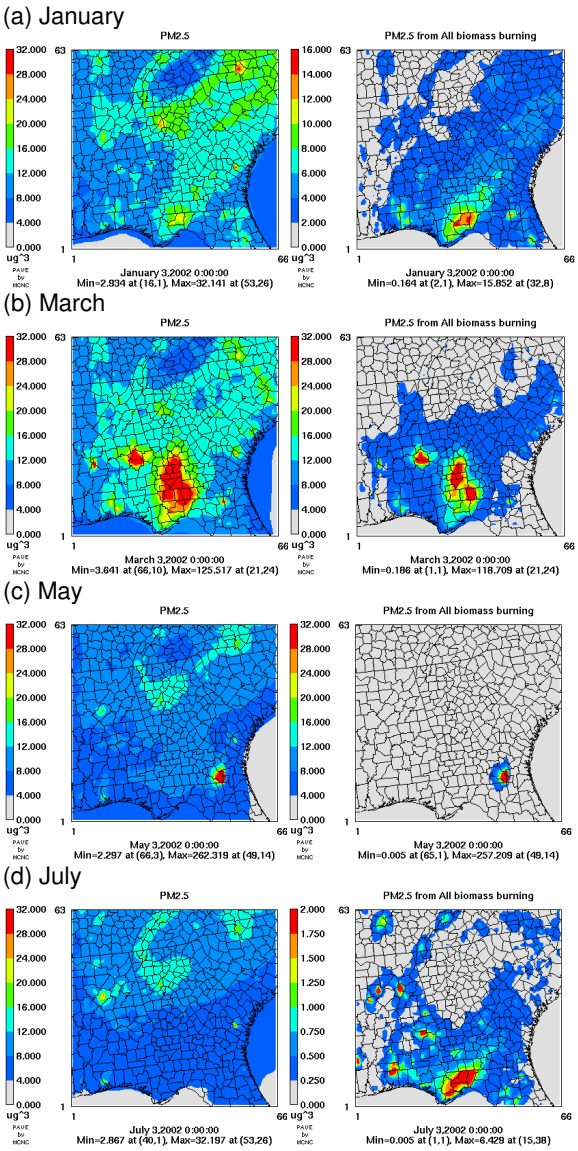

Figure 4 Monthly average $PM_{2.5}$ concentrations (left column) and source contributions from all biomass burning (right column) during January, March, May and July 2002. (Note different scales used.)

Air quality impacts from individual biomass burning sources have similar rank as their emissions, with prescribed burning as the largest individual source. Significant temporal and spatial variation is also observed. Source contributions from prescribed burning peak in March, followed by January, May and July. They concentrate in the southwestern Georgia, influencing much of the region. Source contributions from agriculture burning spatially follow the distribution of agricultural lands and peak in March. Spatial distributions of source contributions from land clearing and wildfires are more sporadic. Specifically, prescribed burning is always the largest single biomass burning source in the Georgia $PM_{2.5}$ non-attainment area, followed by land clearing. Source contributions from wood burning in fireplaces and woodstoves are very uncertain and require further studies.

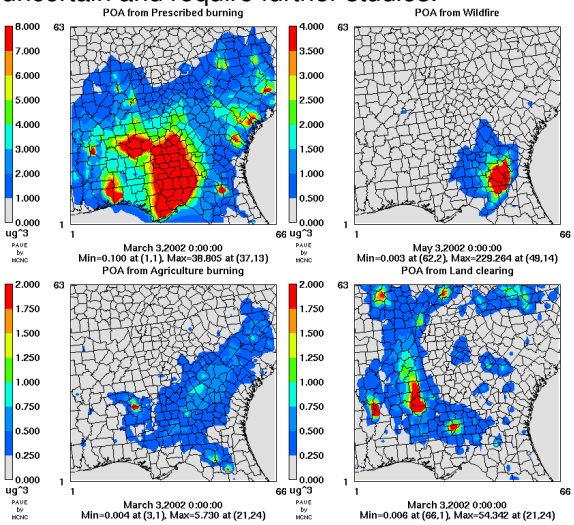

Figure 5 Monthly average POA contributions from prescribed fires (upper left), wildfires (upper right), agriculture burning (lower left) and land cleaning (lower right) during the their peak month

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