Atmos. Chem. Phys. Discuss., 7, S9921–S9928, 2008
www.atmos-chem-phys-discuss.net/7/S9921/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Organic composition of carbonaceous aerosols in an aged prescribed fire plume" *by* B. Yan et al.

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Received and published: 5 September 2008

Responses to Reviewers’ Comments

Manuscript Title: “Organic Composition of Carbonaceous Aerosols in an Aged Prescribed Fire Plume” by B. Yan, M. Zheng, Y.T. Hu, S. Lee, H. K. Kim and A. G. Russell Ms. Ref. No.: acpd-2007-0516-ms

The authors appreciate all reviewers’ valuable comments and suggestions on this manuscript. Please see our responses attached below.

Reviewer 1 Comments: P7, L17; what was the basis for using a multiplication factor of 1.4 to convert measured carbon to OC? Shouldn’t the high content of biogenic and other polar organics drive this factor considerably higher than 1.4, as shown by other similar studies cited by the authors? Response: The OM (organic matter)/OC

factor of 1.4 has been changed to 1.5, and the ratio (OM/PM2.5) of 60% has been recalculated as 70%. The value of 1.5 was estimated as (OC + unidentified mass)/OC in the previous study based on source emissions from prescribed burning in Georgia. The reference (Lee et al., 2008) has been added in the revised manuscript.

Comments: P8, L4-7; are CO2 data available in that area? CO2 concentrations are excellent tracers of the prevailing atmospheric dilution conditions and would put several statements made in this section into the right perspective and eliminate any of the vagueness. Response: Unfortunately, there are no CO2 data available at these routine monitoring sites.

Comments: P11, L5-7; there is increased secondary organic formation in Atlanta in February? Can this argument be supported by either meteorological or atmospheric (O3 etc.) data? Response: Secondary organic formation was enhanced in Atlanta, GA by this plume of wood combustion even in February. The statement is supported not only by elevated ambient concentrations of secondary organic tracers (2-methyltetrols, pinonic acid, pinic acid and dicarboxylic acids) but also by the ozone concentration increasing by about 50% (from around 60 to 90 ppb, which is quite elevated for this time of year) at the same time as when the plume hit Atlanta in the early evening. These data and the related discussions are given in Section 3.3 (page 10-11).

Comments: P13, L19; “factions” or “fractions” Response: It has been revised as “fractions”.

Reviewer 2 Comments: A central assumption in the study is that the airmasses sampled before, during and after the fire originated from the same area. To support this, the authors should show back-trajectories for the 3 day period to show that the "before" and "after" air masses are from the biomass burning area (and if not, it should be discussed whether it is of concern for the analysis). A map of the sampling location and its relation to the prescribed burns would also be very useful. Response: Actually, we do not make an assumption that the air masses originate in the same area, but

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that primary sources other than prescribed fire have similar influences on Atlanta, GA before, during and after the event day. This is clearly established by the data obtained in this work and has further foundation from prior studies. Prior studies have shown that there is a significant increase in OC within Atlanta compared with a more rural site (e.g., Yorkville, GA), and this is further found to be true by analyzing regional EC and OC levels from the Southeastern Aerosol Research and Characterization (SEARCH) network (Edgerton et al., 2005; Hansen et al., 2003; Zheng et al., 2006). More convincing, however, is the analysis of the monitor locations and specific tracers of major, non-biomass burning primary sources. Each sample (‘before_fire’, ‘event’ and ‘after_fire’) in this study was composed of several 24-hr filters from three ASACA sites (South Dekalb, Fort McPherson and Fire station #8). These monitors are spread out around metro Atlanta and capture the urban mix of primary sources. By compositing the filters, the results are relatively insensitive to changes in wind direction. Furthermore, during the period (before, during and after the event day), organic tracers of mobile sources and meat cooking, such as hopanes, steranes and cholesterol, varied slightly in concentration, strongly supporting that those major source impacts varied little (Figures 4-5). On the other hand, the continuous data show very large increases in both PM2.5 and OC when the plume, which originated in a rural area, hit these urban monitors, and these increases overwhelm the background in this area (Lee et al., 2008). Previous studies have also identified the unique biomass burning plume using direct data analysis, wind trajectory and chemical modeling (Hu et al., 2008; Lee et al., 2008). A more detailed explanation has been added in the revised manuscript (Section 3.2, page 10) as follow:

“Vehicular engine exhaust and meat cooking are the two other major sources of primary organic carbon in the Atlanta urban area (Lee et al., 2007; Liu et al., 2005; Marmur et al., 2005; Zheng et al., 2007). The before, during and after concentrations of hopanes and steranes, both organic markers for internal combustion engine emissions, did not show significant variation, i.e. 1.7, 1.7 and 1.0 ng m-3 (Figure 4). Cholesterol, an organic tracer of meat cooking, did not exhibit major variation as well

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(Figure 5). Both suggest that primary sources other than prescribed fire have similar influences on Atlanta, GA before, during and after the event day. This point has further foundation from prior studies, which have shown that there is a significant increase in OC within Atlanta compared with a more rural site (e.g., Yorkville, GA) by analyzing regional EC and OC levels from the Southeastern Aerosol Research and Characterization (SEARCH) network (Edgerton et al., 2005; Hansen et al., 2003; Zheng et al., 2006). Furthermore, the sampling sites used here are spread out around metro Atlanta and capture the urban mix of primary sources. By compositing the filters, the results are relatively insensitive to changes in wind direction. On the other hand, very large increases in continuous PM2.5 and OC values were observed when the plume, which originated in a rural area, hit the urban monitors in Atlanta (Lee et al., 2008). These increases are consistent with those increases measured on the filters, and overwhelmed the background in this area. On the event day, PM2.5 increased by 23.9 μg m-3 (over 60%) while OC increased by 14.2 μg m-3 (over 80%). Biomass burning-related organic tracers (levoglucosan, resin acids and retene) increased by 7-23 times. In addition, Community Multi-scale Air Quality (CMAQ) modeling showed that the timing of the increase in OC is consistent with when the prescribed fire plume impacted Atlanta (Hu et al., 2008). These points strongly support that vehicular sources and meat cooking emissions are not responsible for the large increase in PM2.5 and OC on the event day, and that the greatly increased carbonaceous aerosol concentrations during the smoke episode are from prescribed fires. Source apportionment results calculated using organic molecular marker-based chemical mass balance (CMB-MM) model also indicate significant increases of prescribed burning emissions on the event day, but not for other major primary sources (i.e. vehicular source and meat cooking) (Lee et al., 2008).”

Comments: How "aged" is the biomass burning aerosol sampled in this study? Which aspects of the source profile is expected to remain when the aerosol is additionally aged and diluted for a couple of days? Response: Trajectory analysis gives an age of 3-4 hours for the biomass burning aerosols sampled and investigated in this study

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(Hu et al., 2008; Lee et al., 2008). The aged source profiles developed in this research were compared with fresh emission source profiles tested previously. The results indicate that among wood combustion-related tracers, levoglucosan, n-alkanes, and water-soluble potassium are relatively stable during the short transport time, but PAHs, resin acids, and n-alkanoic acids are more subject to aging.

Comments: The authors state that relatively little information on speciation from aged biomass burning aerosol is available (in part because of the HULIS-like nature of the carbonaceous fraction). However, there is well-established literature on using other techniques, such as the Aerosol Mass Spectrometer (AMS), to identify and attribute biomass burning contributions to aerosol levels. It would be good to include a short discussion on such alternatives in the introduction. Response: A short discussion as below has been added into the introduction (page 4) in the revised manuscript as suggested. “In this study, detailed GC/MS speciation of carbonaceous aerosols, along with receptor modeling, is used to quantify impacts from the biomass burning plume, although some other techniques can provide information about aerosol composition and source impacts as well. For example, Aerosol Mass Spectrometer (AMS) is increasingly used to determine real-time size distribution and chemical composition of non-refractory submicron inorganic and organic aerosols (Allan et al., 2003; Canagaratna et al., 2007; Jimenez et al., 2003). Recently, this method has been used to estimate source contributions from biomass burning through quantitatively characterizing hydrocarbon-like and oxygenated organic aerosols (Cottrell et al., 2008; DeCarlo et al., 2008; Zhang et al., 2005). GC/MS allows identification and quantification of hundreds of organic compounds from ambient PM2.5, including n-alkanes, hopanes, steranes, alkanoic acids, alkanedioic acids, PAHs, resin acids, and others (syringols, levoglucosan, cholesterol, 2-methyltetrols, etc.).”

Comments: A reference is required for the C/OC ratio used. Is 1.4 characteristic of biomass burning aerosol? Does the available speciation support this ratio? How does it compare to the C/OC from other techniques, like the AMS? Response: The OM (or-

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ganic matter)/OC factor of 1.4 has been changed to 1.5, and the ratio (OM/PM2.5) of 60% has been recalculated as 70%. The value of 1.5 was estimated as (OC + unidentified mass)/OC in the previous study based on source emissions from prescribed burning in Georgia. The reference (Lee et al., 2008) has been added in the revised manuscript. To our best knowledge, AMS has not been used to characterize carbonaceous aerosols impacted by biomass burning in this area before.

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Interactive comment on Atmos. Chem. Phys. Discuss., 7, 18015, 2007.