Impacts of prescribed fires on air quality over the southeastern United States in spring based on modeling and ground/satellite measurements

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Prescribed burning is a large aerosol source in the southeastern United States. Its air quality impact is investigated using 3-d model simulations and analysis of ground and satellite observations. Fire emissions for 2002 are calculated based on a recently developed VISTAS emission inventory. March was selected for the investigation because it is the month with most active prescribed fires. Inclusion of fire emissions significantly improved model performance. Model results show that prescribed fire emissions lead to ~30% enhancements of mean OC and EC concentrations in the Southeast and a daily increase of PM2.5 up to 25 μg m^{-3}, indicating that fire emissions can lead to PM2.5 nonattainment in affected regions. Surface enhancements of CO up to 200 ppbv are found. Fire count measurements from the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the NASA Terra satellite show large springtime burning in most states, which is consistent with the emission inventory. These measurements also indicate that the inventory may underestimate fire emissions in the summer.

Introduction

About 5 million acres of forest, crop, and range land are burned annually in the southeastern United States. Prescribed fires comprise 60% of the burned acreage (*1*) and are the most common because of the use for forest resource management, such as reducing hazardous fuels and improving wildlife habitats (*2*). While effective and economical, the emissions of aerosols and gases can adversely affect local and regional air quality.

Prescribed burning is one of the most important sources of carbonaceous aerosols in the Southeast (*3*). It contributes ~15% of total particulate matter (PM) emissions over this region (*1*). Since all southeastern states except Florida have PM2.5 nonattainment areas (http://www.epa.gov/pmdesignations/statedesig.htm), this additional contribution of PM2.5 from prescribed burning is of great concern for air quality management. A better understanding of air quality impacts from fire emissions is necessary (*4-6*).

More attention has been drawn to wildfires than prescribed fires since the intensity per event is much larger in wildfires. While less intense, prescribed fires are very frequent in the Southeast in winter and spring, leading to a potentially large impact on regional air quality. The chemical composition of prescribed fire emissions differs from wildfires due to the nature of under-story burning of prescribed fires (*7*). We use the fire emission inventories for 2002 developed by the Visibility Improvement - State and Tribal Association of the Southeast (VISTAS) program. It is the most comprehensive compilation to date on the basis of fire activities reports from county, state, and federal agencies of the southeastern states (*1*).

The fire impacts on the air pollutants, CO, organic (OC) and elemental carbon (EC), can be better evaluated using chemical transport model simulations with the improved emission inventory. The model simulations are evaluated with surface measurements of these species. We choose March 2002 as the studying period, which is the month of the most burned acres in the VISTAS inventory.

The VISTAS inventory can be evaluated more directly using fire count measurements from the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the NASA Terra satellite. Its large spatial coverage has many advantages in monitoring wildfire activities (*8,9*). However, most prescribed fires are under-story and the effectiveness of the MODIS fire detection algorithm (*10*) for under-story fires is unknown. The temperature threshold of 310 K in the MODIS fire detection algorithm can be too high for identifying the relatively small and cool prescribed fires (*11*). To minimize uncertainties, we evaluate if spatially-averaged (state-level) seasonal variations of estimated burned areas and MODIS fire counts are in agreement. March is the month with the most active burning in the VISTAS inventory. We expect to find a large signal in MODIS fire count measurements.

Method

EPA Models-3 Community Multiscale Air Quality (CMAQ) modeling system (*12*) is used to simulate the distributions of gas and particulates emitted from fires. Surface observations of O_3, CO, and aerosols are employed for model evaluations. Satellite measurements of lower tropospheric CO are also used to study its fire enhancements. The uncertainty of fire emissions in the VISTAS inventory is discussed in its comparison with satellite fire count products.

VISTAS Emission Inventory. The VISTAS emission inventory was developed from the 1999 National Emissions Inventory (NEI) version 2 and used 2002 as the base year (*1*). Its anthropogenic emissions are projected from NEI 99 emission inventory. Fire emissions were recalculated for the southeastern states based on the updated fire records collected from state and federal fire agencies. Complete updates for prescribed fire emissions are available in 9 of the 10 Southeastern states (except VA), with 5 states (AL, FL, GA, MS, and SC) having the most complete data. Among the other 4 southeastern states (KY, NC, TN, WV), partial updates have been done whenever the government reports were available. For model evaluation, we therefore focus more on the states with the more complete data.

Among the four fire types, wildfire, prescribed burning, agricultural fires, and land clearing fires, prescribed burning plays a dominant role in all months except May when wildfire is the largest. Figure 1 compares the monthly mean emissions of three major fire emitted pollutants (OC, EC, and CO). Prescribed fire emissions cover large regions of the southeastern US, although the geographic variability is large. Most of the high emission spots appear in 4 states, Georgia, Florida, Alabama, and South Carolina. Burning is the most active in March when prescribed fire compromises >70 % of the total burned area.

Air Quality Modeling System. CMAQ version 4.4 with the SPARC99 chemical mechanism (*13*) and AERO3 aerosol module (*14*) is used. The meteorological fields were assimilated using Penn State/NCAR MM5 with the NCEP reanalysis data (*15*,*16*). The 148×112 model-grid domain covers the contiguous United States and part of southern Canada and northern Mexico with a grid distance of 36 km. We specified 19 vertical layers, of which 12 are below 1 km. The study period is March 2002. Nested simulation using 12-km resolution does not improve the model results due in part to the relatively coarse temporal and spatial resolution of the fire emission inventory (Supporting information).

CMAQ emission inputs were prepared from the VISTAS inventory using the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System (http://www.smoke-model.org/index.cfm) version 2.2. Annual county-level fire emissions were allocated to each month based on VISTAS reported burned areas in each state. These emissions are distributed in SMOKE using forest distributions as surrogates. The monthly temporal resolution of fire emissions contributes to the discrepancies between model results and measurements. The default EPA daily emission profile in SMOKE is applied to distribute prescribed fire emissions (mostly from 10 am to 8 pm). Biogenic emissions are calculated with the BEIS3 model. The detailed model configurations are described in supporting information.

Considering active boundary layer mixing in daytime, the effect from plume rise mainly comes from the fraction injected above the top of the boundary layer. Since controlled prescribed burning is smaller in scale and cooler in temperature than wildfires (*17*), the injected fraction from prescribed fires is also smaller. There is currently not enough information from the VISTAS inventory to properly treat plume rise in the model and hence plume rise above the boundary layer is not included. We consider this uncertainty in model evaluations with measurements and find some support for plume rise in analyzing MOPITT data although it is qualitative (Supporting information).

Default CMAQ initial and boundary conditions are used except for CO. For CO with a relatively long chemical lifetime, the default CO concentration at 80 ppbv in the surface layer leads to underestimations with the observations at surface sites (*18*) and in the free troposphere (*19*). We therefore specify CO initial and boundary conditions with GEOS-CHEM simulations (*20*), which have been evaluated extensively (*19, 21*).

We performed two CMAQ runs, one base run with all emissions and one sensitivity run without prescribed fire emissions over 10 VISTAS states. A comparison is then conducted between the two simulations to reveal the enhancements of certain air pollutants from fire emissions, such as CO and carbonaceous particles. Modeled distributions of fire pollutants are spatially consistent with the spatial distributions of the fire emissions (Supporting information).

Surface CO and Aerosol Observations. Model simulations are evaluated with surface observations from two networks, the Southeastern Aerosol Research and Characterization project (SEARCH) (*18*) and Interagency Monitoring of Protected Visual Environments (IMPROVE) (*22*) (Figure 2). Both gases and aerosols are measured at the 4 pairs of urban-rural SEARCH stations. Only aerosols are measured at the IMPROVE stations, located mainly in rural areas. OC and EC measurements from SEARCH and IMPROVE are comparable since the thermo-optical reflectance (TOR) method was used in both networks. More extensive CO measurements from the EPA AIRNow network cannot be used since only values > 0.5 ppmv were reported and CO concentrations at rural sites were lower than the reporting limit.

MODIS Fire Counts. MODIS fire detection products from the NASA Terra satellite (*10*) are used to evaluate the seasonal variations of the VISTAS fire inventory. Cloud pixels were removed before the fire detection algorithm was applied (*10*). Level 3 daily Terra MODIS global products (MOD14A1) include daytime and nighttime observations (~10:30 am and pm). Most fire counts are observed during daytime, consistent with the diurnal profile in SMOKE. The horizontal resolution of this dataset is 1×1 km^2. Given the difference in temporal resolutions of MODIS measurements and the VISTAS inventory, we focus on the monthly statistics.

Air quality impacts at surface sites

We first examined model simulations with available surface observations using published statistical metrics (Supporting information). Model results capture the general characteristics of observed O_3, CO, EC, OC, and PM2.5.

Ozone. Prescribed burning has very little influence on simulated O_3 during this period. At SEARCH sites, it only leads to a 1.5% ozone increase, which is much smaller than the impacts by biomass burning in the tropics (*e.g.*, *23*, *24*). More extensive evaluation using the measurements from the EPA AIRNow network in the Southeast shows similar results. One of the reasons is that emissions of NO_x from prescribed fire are much smaller than those from fossil fuel sources over North America. NOx concentrations only increase by 1% at SEARCH sites. Another reason is that photochemistry is relatively slow in March.

Carbon Monoxide. We use the relatively long-lived biomass burning tracer CO to examine the influence of fire emissions in the region. On average, fire emissions increase surface CO concentrations by 6%. Since the EPA AIRNow measurement sites report CO only when its mixing ratios are CO > 0.5 ppmv, we are limited to SEARCH sites. Among these, we only find two sites (CTR and OLF) with significant signals (Figure 3). These peaks are identified as simultaneous increases of CO, EC, and OC and the model captures better high CO concentrations in the measurements with fire emissions. During the period of March 5 to 10, both sites show high concentrations of CO. Without prescribed fire emissions, the model would underestimate CO by up to 200 ppbv (or 130%).

The observations also show another high-CO episode between March 12 and 16. However, the model is only able to simulate high CO at the OLF site. A few reasons may contribute to this discrepancy. Chief among them is the quality of the emission inventory.

There are uncertainties in the magnitudes of emissions and the temporal and spatial resolutions are not high enough to capture all impact peaks driven by fire emissions at a specific site. Nonetheless, the evaluation presented here indicates that the CMAQ model with the currently available emission inventory has some predicting skills and provides a useful way to assess the typical impacts of fire emissions.

OC and EC. Observations of OC and EC come from SEARCH and IMPROVE networks. We compare the model results with the observations at 4 selected sites, CTR (SEARCH), GFP (SEARCH), EVER (IMPROVE), and MACA (IMPROVE) (Figure 4 and Supporting information: Figure S4). The effects on OC and EC are similar. The base run with full emissions agrees better with observations than the sensitivity run without prescribed fire emissions. The differences between the two simulations are due to fire emissions, which can be significant. The increases of OC and EC from fire emissions are up to 5 times at the CTR site, where local burning is large. Here we define local burning as that in the county where the site is located. What is perhaps unexpected are that the increases of OC and EC (by 100% on some occasions) at the MACA site, where there is almost no local burning. Simulated enhancements are entirely due to regional transport. It is evident from this comparison that the measurement frequencies of once per 3 days (as in most sites used here) can miss the effects of fire emissions.

The dispersion of episodic fire emitted pollutants highly depends on the meteorological conditions. Although the fire emissions are continuously distributed in the model due to the lack of burning timing information, the model did not simulate continuous aerosol enhancement at surface. The episodic aerosol enhancements are evident in fire induced high OC and EC concentrations (Figure 4). It indicates a combined influence of fire emissions and meteorological conditions.

We compared all OC and EC at 24 stations available in the Southeast (Figure 5). The scattering around the 1:1 line reflects the uncertainties in model formulation and inputs, which includes emission uncertainties. Without fire emissions, the model results have clear low biases. Table 1 shows the observed and simulated mean concentrations of EC and OC. Prescribed fire emissions lead to a simulated mean OC increase from 1.33 to 2.24 μgC m^{-3} (or 68%) compared with the observed mean of 1.92 μgC m^{-3} and a simulated mean EC increase from 0.29 to 0.49 μgC m^{-3} (or 69%) compared with the observed mean of 0.51 μgC m^{-3}. The inclusion of fire emissions does not significantly reduce scattering of simulated data against the measurements (Supporting information), which reflects the uncertainties in simulated impacts of individual fire events. Averaged over the 10 southeastern states, the monthly simulated mean contributions of prescribed fires to OC and EC are 28% (0.8 μgC m^{-3}) and 31% (0.2 μgC m^{-3}), respectively. However, the simulated maximum increases of daily OC and EC from prescribed emissions are much larger, 13.4 and 3.2 μgC m^{-3}, respectively. Prescribed fire can lead to an enhancement of daily carbonaceous matter (CM = OM + EC, organic matter (OM) = 1.4 × OC) up to 22 μg m^{-3}, or 63% of the EPA standard of 24-hour PM2.5 concentration of 35 μg m^{-3}.

A positive correlation is found between the local (county) burned areas and OC and EC enhancements at 24 sites. The correlation coefficients for OC and EC are 0.65 and 0.63 respectively. Therefore, local burning only explains about 40% of fire induced variance. Regional transport is an important factor. To further study the transport effects, we divided the 10 VISTAS states into two groups. In the states with high fire emissions (AL, GA, FL, and SC), prescribed fires contribute to 39% and 42% of OC and EC, respectively. In the states with low fire emissions (KY, MS, NC, TN, VA, and WV), its mean contributions to OC and EC are smaller but still significant at 14% and 17%, respectively.

To further estimate the effects of prescribed fire emissions, we compute the concentrations of organic matter and carbonaceous matter. The simulated mean concentrations of OM, CM, and PM2.5 at 24 sites are 2.9, 3.3, and 9.3 $\mu g\ m^{-3}$, respectively. CM accounts for 37% and 25% of PM2.5 in states with high and low fire emissions, respectively. The simulated mean contributions of prescribed fires to PM2.5 are 12% and 3%, respectively, over states with high and low local burning emissions. The average OC/EC ratio in high burning areas is smaller than in low burning area (*25*), consistent with previous studies (*26*, *27*).

Fire emission evidence from satellite measurements

MOPITT CO at 850 hPa. Based on model comparison with surface measurements (Figure 3), we selected two episodes to compare the model results to the integrated lower tropospheric CO column measurements (reported as 850 hPa) by the Measurements Of Pollution In The Troposphere (MOPITT) satellite (*28*) in the Southeast (Supporting information). Generally, MOPITT shows CO enhancements over regions where the model simulates impacts from fire emissions, although the enhancement levels are lower in the model. The simulated CO vertical distribution may be problematic due to inadequate simulations of plume rise or vertical transport.

Terra MODIS fire counts. Direct detection of burned areas from satellite instruments is ineffective for prescribed fires, since the burning is understory by design. Fire count measurements based on surface temperature changes are more useful. The spatial resolution of MODIS fire counts is 1×1 km^2. The detection efficiency of prescribed fires with scales <1 km is unknown. Quantitative comparison is therefore difficult between burned areas in the VISTAS inventory and MODIS fire counts. The correlation coefficient is 0.57 between the monthly MODIS fire counts and the burned areas in VISTAS inventory at the state level in 2002. We focus on the qualitative aspect in the comparison by examining if March is the month with the largest amount of fires as found in the VISTAS inventory.

We show the comparisons for the 4 states (AL, FL, GA, and SC) with large fire emissions in Figure 6. The secondary peak in May in the VISTAS inventory for Georgia is due to wildfires. Generally, the VISTAS inventory shows consistent spring maximum and summer minimum in the Southeast. MODIS fire counts show clear spring maximum in Florida and South Carolina, in good agreement with the VISTAS inventory. In Georgia, the May fire counts are larger than in March, likely because of wildfires. In the VISTAS inventory, wildfires account for ~80% of the total burned areas in May in GA. It has higher burning temperature than prescribed fires due to less control (*13*). These large wildfires are much easier for satellite to detect than prescribed fires.

The largest difference for spring is found over Alabama, where MODIS fire counts are fairly low. Previous model evaluations with surface measurements of CO at the CTR and OLF sites (Figure 3) and OC and EC at the CTR site (Figure 4) clearly demonstrate the large impacts of fire emissions in Alabama. Chemical mass balance

(CMB) (*29*) and positive matrix factorization analyses (*30*) showed a spring peak of wood burning in the Southeast. Therefore, we believe that MODIS fire counts failed to detect fire activities in Alabama in spring 2002. The mean temperature for prescribed fires is 440 K (*13*); the detection sensitivity of MODIS begins to decrease in this temperature range (*10*). Moreover, prescribed fires may still in the early burning stage when Terra satellite passes over at 10:30 am local time. Some underestimation is expected. The reason for much lower detection in Alabama than Georgia or South Carolina, however, is unclear.

Three out of four states (AL, GA, and SC) show peaks of fire activities (fire counts) in the summer, which is not present in the VISTAS inventory. First, the fully-grown forest canopies in the summer shield the upwelling radiation from fires and reduce the thermal signal and detection efficiency of the MODIS instrument than spring. The second potential interference factor is the presence of cloud. We examined MODIS cloud fractions in 2002 over the Southeast. There are 10-30% monthly variations through this year. But no large decrease of cloud fraction is found from spring to summer. Another possible factor is hot and dry surfaces in summer, such as the urban and industrial regions, which could trigger false positives (*10*). It is however not supported by spatial analysis of MODIS fire spots in Alabama in August. It shows locations mostly over forest areas with a few on cropland or grassland. Indications are that the VISTAS inventory may have underestimated fire emissions in summer. More detailed analysis is currently under way.

CMAQ analyses of surface pollutant concentrations and satellite measurements suggest large enhancements of OC, EC, and CO due to fire emissions in March. CO is a good tracer to understand the transport of fire emissions. However, surface observational evidence is limited. More CO measurements are available from the EPA AIRNow network, but the current lower limit of 0.5 ppmv in reported values is too high and renders the data unusable. To explore in greater detail MOPITT measurements, measurements of CO vertical profiles are needed. The monthly mean enhancement of PM2.5 is 8% (0.8 μg m^{-3}) and daily enhancement is up to 25 μg m^{-3}, posing a problem for the attainment of the daily national PM2.5 standard of 35 μg m^{-3}. For more detailed analysis, fire emission inventories with better spatial and temporal resolutions than VISTAS are needed. A higher temporal resolution is critical to simulate better episodic PM2.5 enhancements. A higher measurement frequency of OC and EC aerosols at the surface networks is needed to more adequately characterize the fire impacts.

Acknowledgement

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Supporting Information Available

Supporting Information includes simulated regional enhancements from fire emissions, model performance evaluation, model comparisons with 36 and 12 km grids, and MOPITT observed CO enhancement at 850 hpa. This information is available free of charge via the Internet at http://pubs.acs.org.

Literature Cited

(1) Barnard, W.; Sabo, E. Documentation of the base G2 and best & final 2002 base year, 2009 and 2018 emission inventories for VISTAS, MACTEC Inc, **2008**. Available at http://www.vistas-sesarm.org/documents/VISTABF2003-20-2008.pdf.

(2) Hardy, C. C.; Hermann, S. M.; Mutch, R. E. Overview. In *Smoke Management Guide for Prescribed and Wildland Fire*; Hardy, C. C., et al., Eds.; NFES 1279, National Wildfire Coordination Group: Boise, ID, **2001**. Available at http://www.nwcg.gov/pms/pubs/SMG/SMG-72.pdf.

(3) Liu, Y. Variability of wildland fire emissions across the contiguous United States. *Atmos. Environ*., **2004**, 38, 3489-3499.

(4) Evans, L. F.; Weeks, I. A.; Eccleston, A. J.; Packham, D. R. Photochemical ozone in smoke from prescribed burning of forest. *Environ. Sci. Technol*., **1977**, 11, 896-900.

(5) Wotawa, G.; and Trainer, M. The influence of Canadian forest fires on pollutant concentrations in the United States. *Science*, **2000**, 288, No5464, 324-327.

(6) McKeen, S. A.; Wotawa, G.; Parrish, D. D.; et al. Ozone production from Canadian wildfires during June and July of 1995. *J. Geophy. Res*., 2002, 107, 4192, 10.1029/2001JD000697.

(7) Lee, S.; Baumann, K.; Schauer, J. J.; et al. Gaseous and particulate emissions from prescribed burning in Georgia. *Environ. Sci. Technol*. **2005**, 39, 9049-9056.

(8) Kaufman, Y.; Ichoku, C.; Giglio, L.; et al. Fires and smoke observed from the Earth Observing System MODIS instrument: products, validation, and operational use. *International J. of Remote Sensing*, *2003*, 24, 1765-1781.

(9) Csiszar, I.; Denis, L.; Giglio, L.; et al. Global fire activity from two years of MODIS data. *International J. of Wildland Fire*, **2005**, 14, 117-130.

(10) Giglio, L.; Descloitres, J.; Justice, C.O.; Kaufman, Y. An enhanced contextual fire detection algorithm for MODIS. *Remote Sensing of Environment*, **2003**, 87, 273-282.

(11) Wang, W.; Qu, J.J.; Hao, X.; Liu, Y.; and Sommers, W.T. An improved algorithm for small and cool fire detection using MODIS data: A preliminary study in the southeastern United States. *Remote Sensing of Environment*, **2007**, 108, 163-170.

(12) Byun, D.W.; Ching, J.K.S. *Science Algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) Modeling System*; U.S. Environmental Protection Agency: Research Triangle Park, NC, **1999**; EPA/600/R-99/030, US EPA.

(13) Carter, W.P.L. *Implementation of the SAPRC99 chemical mechanism into the Models-3 Framework*. **2000**. Available at ftp://ftp.cert.ucr.edu/pub/carter/pubs/s99mod3.pdf.

(14) Binkowski and Roselle, 2003 F.S. Binkowski and S.J. Roselle, Models-3 community multiscale air quality (CMAQ) model aerosol component 1. Model description, *Journal of Geophysical Research*, **2003,** 108, 2001JD001409.

(15) Olerud, D. T.; Sims, A. MM5 sensitivity Modeling in Support of VISTAS (Visibility Improvement – State and Tribal Association), VISTAS task 2e deliverable; **2003.**

(16) Grell, G.; J., D.; Stauffer, D. A description Penn State/NCAR Mesoscale Model (MM5), NCAR/TN-398+STR.; **1994**.

(17) Kennard, D. K.; Outcalt, K. W.; Jones, D.; O'Brien, J. J. Comparing techniques for estimating flame temperature of prescribed fires. *Fire Ecology,* **2005**, 1, 75 – 84.

(18) Hansen, D.A.; Edgerton, E.S.; Hartsell, B.E.; et al. The Southeastern aerosol research and characterization study: Part 1 -- Overview. *J. Air Waste Manage. Assoc.* **2003**, 53, 1460–1471.

(19) Wang, Y.; et al. Late-spring increase of trans-Pacific pollution transport in the upper troposphere. *Geophys. Res. Lett*. **2006**, 33, 10.1029/2005GL024975.

(20) Bey, I.; Jacob, D. J.; Yantosca, R. M.; et al. Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation. *J. Geophys. Res*., **2001**, 106, 23,073 – 23,096.

(21) Choi, Y.; Wang, Y.; Zeng, T; et al. Evidence of lightning NOx and convective transport of pollutants in satellite observations over North America. *Geophys. Res. Lett.* **2005**, 32, 10.1029/2004GL021436.

(22) Malm, W. C.; et al. Spatial and seasonal pattern and temporal variability of haze and its constituents in the United States, Report III. **2000**, ISSN: 0737-5352-47. Available at http://vista.cira.colostate.edu/improve/Default.htm.

(23) Jacob, D. J.; Heikes, B. G.; Fan, S. -M.; et al. Origin of ozone and NO_x in the tropical troposphere: A photochemical analysis of aircraft observations over the South Atlantic basin. *J. Geophys. Res.*, **1996**, 101, 24,235-24,250.

(24) Thompson, A.; Pickering, K. E.; McNamara, D. P.; et al. Where did tropospheric ozone over southern Africa and the tropical Atlantic come from in October 1992? Insights from TOMS, GTE TRACE A, and SAFARI 1992, *J. Geophys. Res*., **1996**, 101, 24251-24278.

(25) National Air Quality and Emissions Trends Report, 2003 special studies edition, EPA 454/R-03-005, **2003**. Available at http://www.epa.gov/airtrends/aqtrnd03/.

(26) Lim, H.-J.; Turpin, B. J. Origins of primary and secondary organic aerosol in Atlanta: Results of time-resolved measurements during the Atlanta supersite experiment. *Environ. Sci. Technol*. **2002**, 36, 4489-4496.

(27) Yu, S; Dennis, R. L.; Bhave, P. V.; Eder, B. K. Primary and secondary organic aerosols over the United States: estimates on the basis of observed organic carbon (OC) and elemental carbon (EC), and air quality modeled primary OC/EC ratios. *Atmos. Environ*. *2004*, 38, 5257–5268.

(28) Deeter, M. N.; Emmons, L.K.; D.P. Edwards; and J.C. Gille. Vertical resolution and information content of CO profiles retrieved by MOPITT. *Geophy. Res*. Lett. **2004**, 31, 10.1029/2004GL020235

(29) Zheng, M.; Cass, G. R.; Schauer, J. J.; Edgerton, E. S. Source apportionment of PM2.5 in the southeastern United States using solvent-extractable organic compounds as tracers. *Environ. Sci. Technol*. **2002**, 36, 2361-2371.

(30) Liu, W.; Wang, Y.; Russell, A.; Edgerton, E. Atmospheric aerosols over two urban-rural pairs in southeastern United States: Chemical composition and sources. *Atmos. Environ*. **2005**, 39, 4453-4470,.

Table 1. Mean and median OC and EC concentrations from observations and model simulations with and without prescribed fire emission at 24 stations in the Southeast in March 2002.

		Observation ($\mu gC\ m^{-3}$)	Model w/ fire ($\mu gC\ m^{-3}$)	Model w/o fire ($\mu gC\ m^{-3}$)
OC	Mean	1.92	2.24	1.33
	Median	1.49	1.54	1.00
EC	Mean	0.51	0.49	0.29
	Median	0.37	0.30	0.18

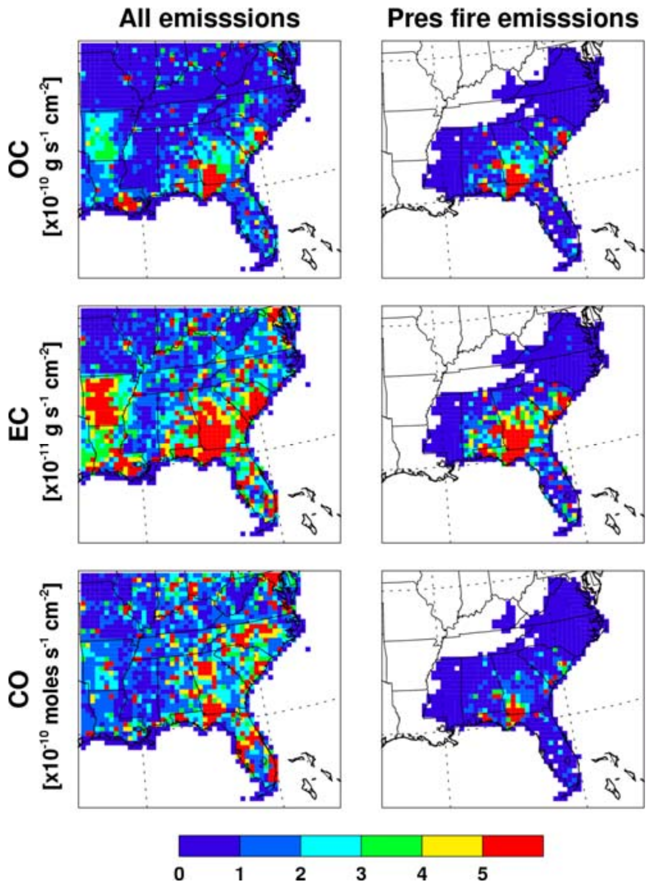

Figure 1. SMOKE processed OC, EC, and CO emissions from standard VISTAS emission inventory and prescribed fires over the southeastern United States in March 2002.

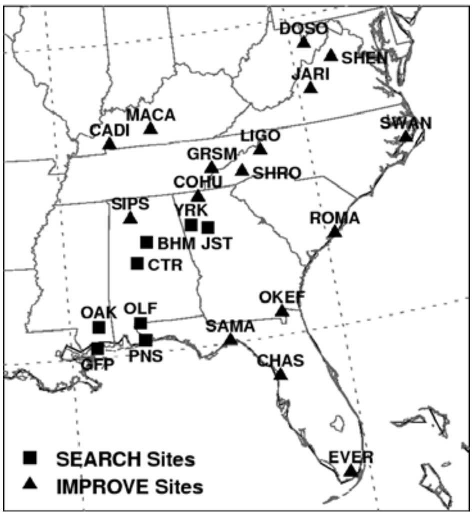

Figure 2. 8 SEARCH (squares) and 16 IMPROVE (triangles) sites over the southeastern United States.

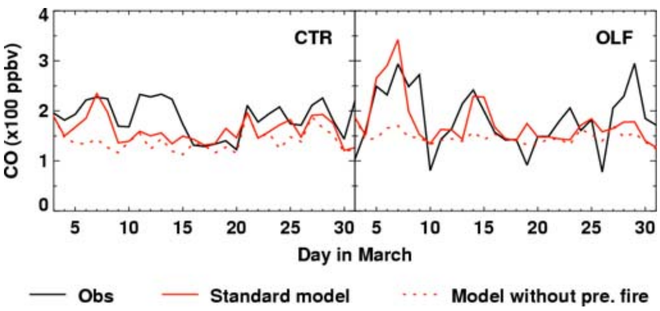

Figure 3. Daily mean CO concentrations (ppbv) at 2 SEARCH sites in March 2002 from observations (black lines), model simulations with (red solid lines) and without (red dotted lines) prescribed fire emissions, respectively. The site locations are shown in Figure 2.

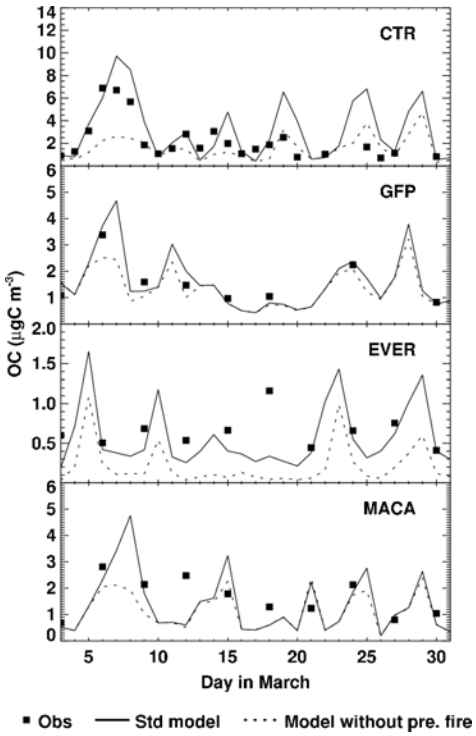

Figure 4. Daily mean OC concentrations (μgC m^{-3}) at 4 stations in March 2002 from observations (squares), model simulations with (solid lines) and without (dotted lines) prescribed fire emissions, respectively. The site locations are shown in Figure 2.

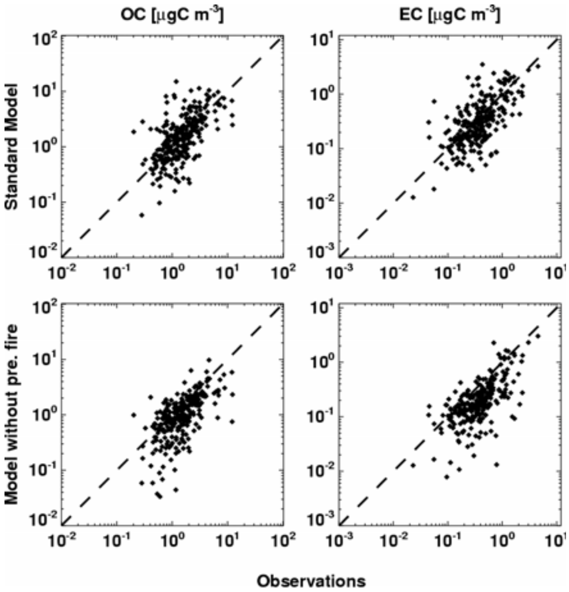

Figure 5. Comparisons between daily OC and EC observations and two simulations with and without prescribed fire emissions at 24 SEARCH and IMPROVE sites.

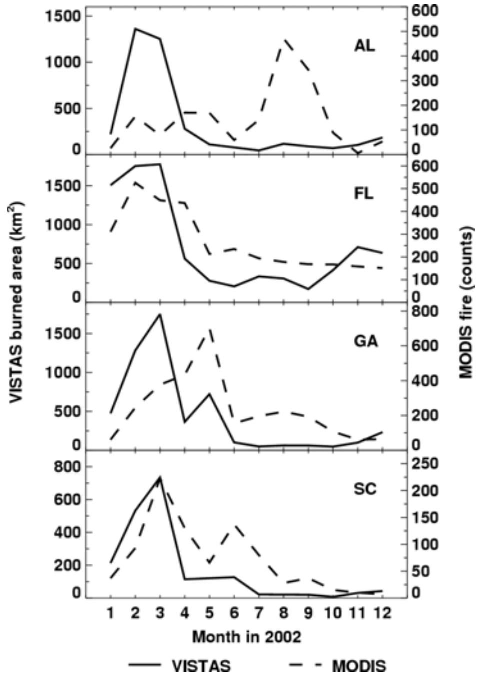

Figure 6. Monthly variations of total burned areas for all fire types from VISTAS inventory (solid lines) and the corresponding Terra MODIS fire counts (dashed lines) over Alabama, Florida, Georgia, and South Carolina in 2002.

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Modeling analyses of surface and satellite observations show that prescribed fire emissions significantly affect air quality in the Southeast

Supporting information

Impacts of prescribed fires on air quality over the southeastern United States in spring based on modeling and ground/satellite measurements

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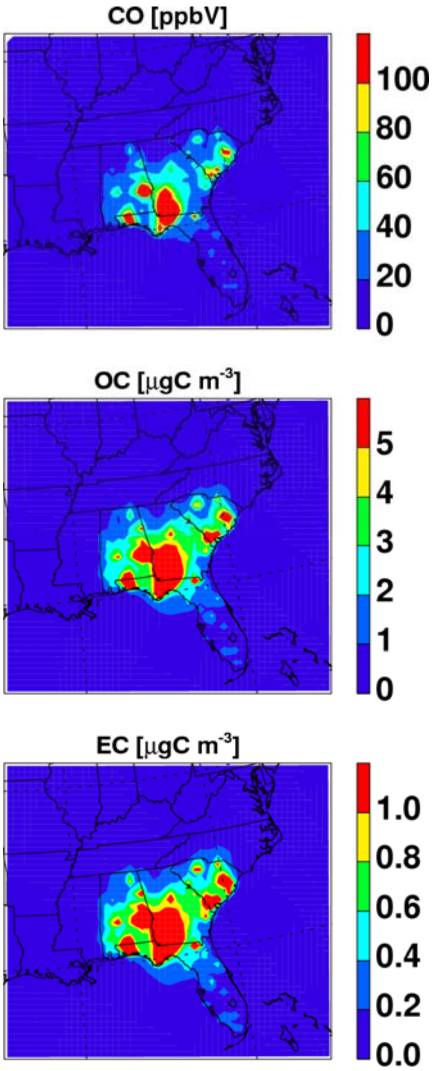
I. Simulated regional enhancements of fire emitted pollutants

The simulated monthly mean enhancements of CO, OC, and EC due to prescribed fire emissions are shown in Figure S1. It is obtained by subtracting the sensitivity results without fire emissions from the base run results. Large increases are mostly found in AL, FL, GA, and SC. The distribution of enhancements correlates well with prescribed fire emissions (Figure 1).



Supporting Information. Figure S1. Monthly enhancements of CO, OC, and EC due to prescribed fire emissions in the southeastern United States in March 2002.

II. CMAQ model performance evaluations

1. Ozone and CO

Three statistical measures recommended by US EPA (*1*) are used here, Mean Normalized Bias (MNB), Mean Normalized Gross Error (MNGE) of all pairs > 60 ppb of ozone and 200 ppbv of CO, and Average Peak Prediction Accuracy (APPA). Hourly ozone observations from the EPA AIRNow and SEARCH networks in March 2002 are compared to the CMAQ ozone simulation results. CO observations are only available from the SEARCH network.

Table S1. Statistics of model performance evaluation for ozone and CO[1].

Species	N_OBS[2]	Obs_Mean[2] (ppbv)	Sim_Mean[2] (ppbv)	Mean Bias (ppbv)	Mean Error (ppbv)	MNB (%)	MNGE[3] (%)	APPA (%)
1-hr O_3	6176	47.9	49.9	2.0	8.2	6.1	23.8	23.6
8-hr O_3	11454	47.4	49.0	1.7	7.0	5.0	20.9	7.7
CO	2423	279.0	250.1	-28.8	134.7	15.9	1.7	-15.4

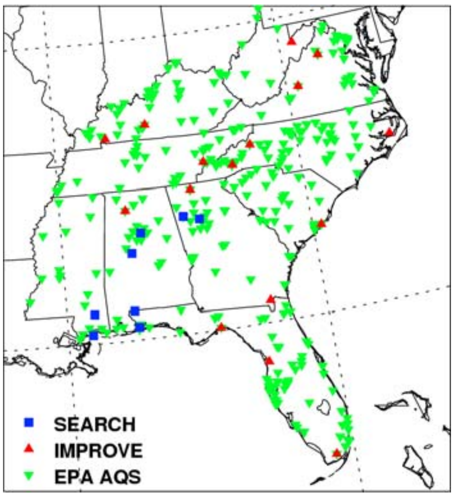
[1] The recommended US EPA (2005) criteria for MNB, MNGE, and APPA for ozone are ±15 %, ±30 %, and ±20 %, respectively.

[2] N_OBS denotes the number of observations. Obs_Mean denotes the observed mean. Sim_Mean denotes the simulated mean.

[3] MNGE is calculated for data with ozone >60 ppbv and CO >200 ppbv.

The CMAQ model errors for 1-hr and 8-hr ozone and CO are within the EPA recommended range. Model simulated CO values tend to have small low biases. However, the CO statistics are based on the measurements from only 4 urban-rural pair sites (SEARCH) compared to many more ozone observation sites (AIRNow and SEARCH) (Figure S2). Increasing model resolution to 12 km does not solve the problem. The underestimate may be partially due to the CO lateral boundary conditions taken from the global GEOS-CHEM model in our simulation. The lifetime of CO in spring is >1 month. The errors of boundary conditions would propagate into the model domain.

However, there are clearly not enough measurement sites to determine if such a case can be made.



Supporting Information. Figure S2. SEARCH (blue), IMPROVE (red), and AIRNow (green) network sites over the southeastern United States.

2. OM, EC, and PM2.5

We use the recently suggested statistical measures (*2*) to evaluate the model performance for particulates. Daily aerosol observations from the EPA IMPROVE and SEARCH networks are employed in the evaluation (Figure S2). The TOR method is used for OC and EC measurements in both networks.

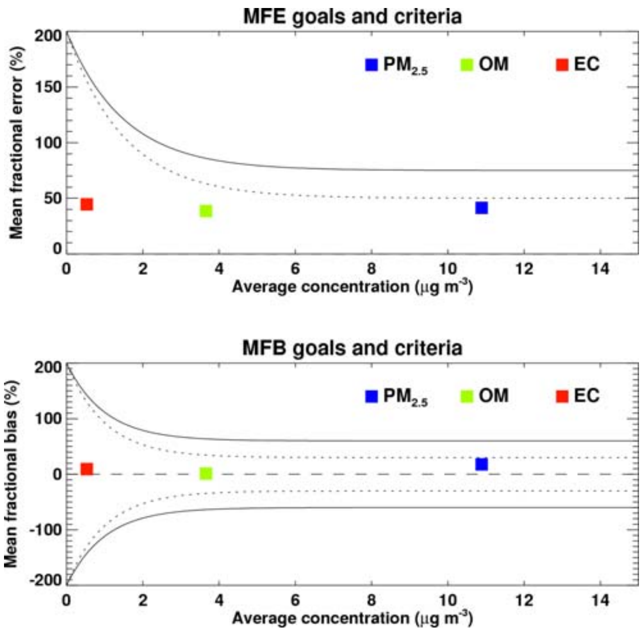
Table S2. Statistics of model performance evaluation for OM, EC, and PM2.5

Species	N_OBS[1]	Obs_Mean[1] ($\mu g/m^3$)	Sim_Mean[1] ($\mu g/m^3$)	Mean Bias ($\mu g/m^3$)	Mean Error ($\mu g/m^3$)	MFB[2] (%)	MFE[2] (%)
OM	261	2.69	3.13	0.44	1.61	5.3	35.0
EC	261	0.51	0.49	-0.02	0.28	-5.7	35.7
PM2.5	377	9.71	9.83	0.12	4.16	-2.4	30.0

[1] N_OBS denotes the number of observations. Obs_Mean denotes the observed mean. Sim_Mean denotes the simulated mean.

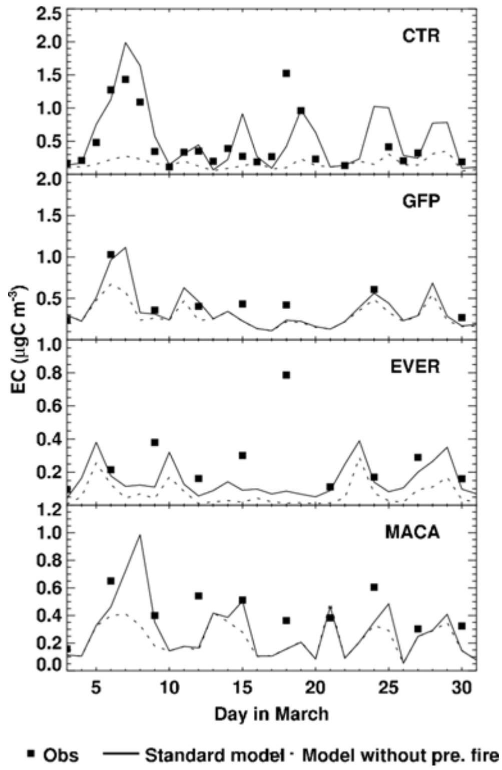
[2] MFB - Mean fractional bias; MFE - Mean fractional error.

Figure S3 illustrates our model results along with the suggested goals (good performance) and criteria (acceptable performance) of MFB and MFE (*2*). The model results are within recommended ranges, indicating that aerosol species are simulated reasonably well.



Supporting Information. Figure S3. MFB and MFE values for simulated PM2.5, OM, and EC. Solid and dotted lines represent the MFE and MFB goals and criteria (*2*), respectively.

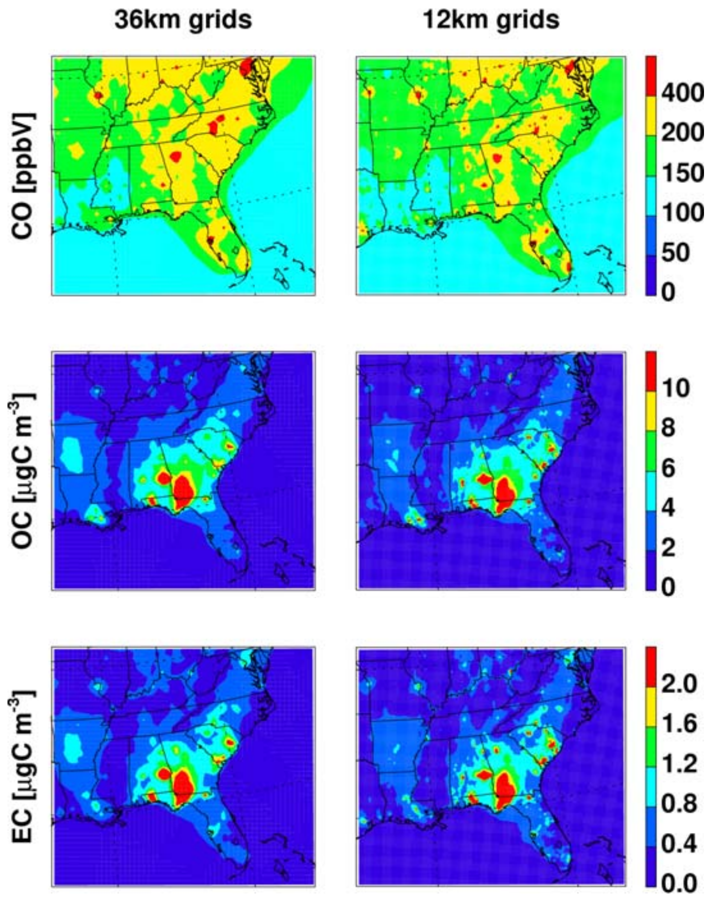
EC simulations were compared to the observations at 4 selected sites in the southeastern United States (Figure S4). The standard model run with prescribed fire emissions generally captures better the observed EC enhancements as in the case of OC (Figure 4).



Supporting Information. Figure S4. Daily mean EC concentrations (µgC m^{-3}) at 4 stations in March 2002 from observations (squares), model simulations with (solid lines) and without (dotted lines) prescribed fire emissions, respectively. The site locations are shown in Figure 2.

III. CMAQ simulation results with 36 km vs 12 km grids

Figure S5 shows that the distributions of CO, OC, and EC are quite similar in the model results. A higher spatial resolution does not improve the simulation results in this season. Another CMAQ model study (*3*) shows similar results between model resolutions of 32 and 8 km.



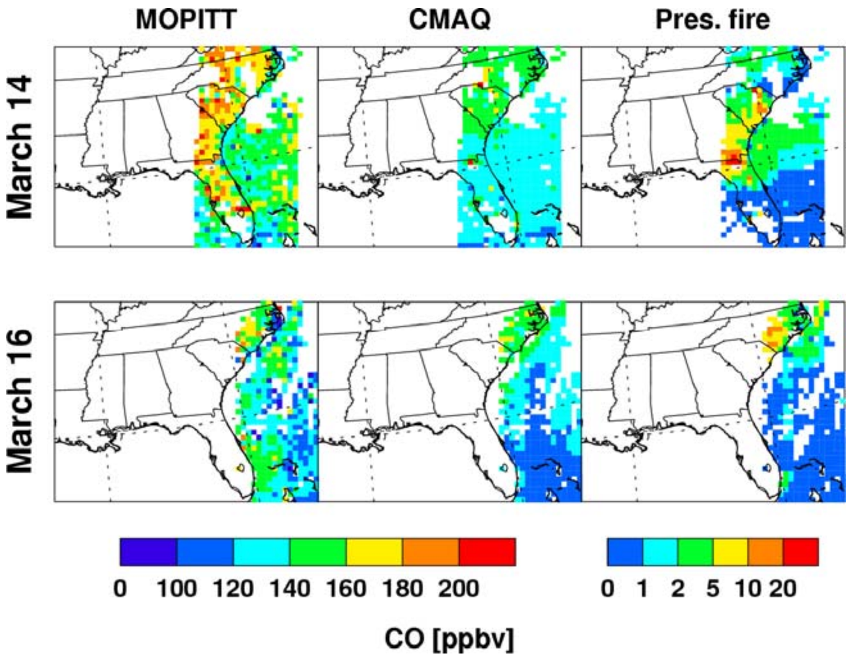
Supporting Information. Figure S5. Distributions of simulated surface CO, OC, and EC with horizontal resolutions of 36 and 12 km, respectively.

IV. MOPITT CO at 850 hPa

Tropospheric column CO is measured using correlation spectroscopy by the Measurement of Pollution in the Troposphere (MOPITT) instrument onboard the NASA Terra satellite (*4*). MOPITT version 3 level 2 products of free tropospheric CO are used to evaluate the fire plume dispersion. Daytime measurements are available once per day at 10:30 AM local time. The spatial resolution is 22×22 km^2. We choose to only analyze daytime data to avoid the uncertainties due to low sensitivity in the night with lower surface temperature (*5*).

MOPITT measurements of lower tropospheric CO are characterized by the retrievals at 850 hPa (*5*). Since the degree of freedom for signal is normally <2 (*6*), MOPITT CO at this level also includes signals from boundary layer, which is influenced by fire emissions. Comparisons between model results and MOPITT measurements indicate that only qualitative agreement can be found.

Surface measurements at SEARCH sites (CTR and OLF) show high CO enhancements on March 14 (Figure 3). MOPITT observations in the eastern part of the Southeast also showed high CO concentrations. We show the comparison results for March 14 and 16 in Figure S6. Model simulated CO concentrations are lower. Sensitivity analysis indicates large amounts of CO enhancements up to 108 ppbv (or 49%) in the coastal regions of Georgia and South Carolina, which is consistent with MOPITT measurements. However, the fire impacts are underestimated. There are two potential reasons. The first is that CO emissions from fire sources are underestimated. Figure 3 suggests that model agreement with surface measurements is reasonable, although the spatial coverage is too limited to draw a general conclusion. The second is that MOPITT observed CO enhancements are biased towards fires that inject CO into the free troposphere since MOPITT is more sensitive to CO in the free troposphere than the boundary layer (*4*). The uplifting of fire CO plumes in these cases may not be adequately simulated by the model in part because plume rise is not included in the model. Measurements of vertical CO profiles are needed to resolve this issue. On March 16, the impacts by fire emissions are much lower. MOPITT CO concentrations are also lower, in better agreement with the measurements. The simulated fire enhancements up to 12 ppbv (or 8%) are over the coastal regions of South Carolina, where MOPITT measurements also show higher concentrations.



Supporting Information. Figure S6. CO concentrations at 850 mb from MOPITT, standard CMAQ simulations, and prescribed fire emissions (standard – sensitivity CMAQ simulations) on 14 and 16 March, respectively. Only the areas with MOPITT coverage are shown for CMAQ simulations. Model simulations are processed with averaging kernels (*5*) in the comparison.

V. Model configurations

The modeling system consists of 3 components, MM5, SMOKE, and CMAQ. Their configurations are described in more detail in this section.

MM5 version 3.6.2 was used for the meteorological simulation with four dimensional data assimilation (FDDA) (*7*). The modeling domain has a horizontal resolution of 36 km. There are 34 vertical layers in MM5 simulations. They are reduced to 19 layers in the Meteorology-Chemistry Interface Processor (MCIP) (version 2) for

SMOKE and CMAQ simulations. The main physical options include Kain-Fritsch2 (KF2) convective parameterization, Pleim-Xiu boundary layer and land surface model, and Rapid Radiative Transfer Model (RRTM) radiation.

Model emissions are prepared by SMOKE version 2.2 with the input of the VISTAS emission inventory. Several source categories are addressed including biogenic sources by BEIS3, on-road mobile sources by MOBILE6, non-road mobile sources, area source, and stationary point sources. Actual electrical generating units (EGU) and fire emissions in the base year 2002 are used within VISTAS domain. Fire emissions are split into area and point sources. Point sources are fewer in number and are allocated at by time and locations of fires. In point source calculation, hourly-specific plume rise was computed to distribute the emissions vertically. Area fire emissions are evenly distributed each day in the month, and spatially surrogated by the forest area and type in each county.

CMAQ version 4.4 was used to simulate the impacts of prescribed fire emissions. SAPRC99 module is chosen for gas phase chemistry simulation. AERO3 module with aqueous-phase reactions is used for aerosol simulations. The vertical diffusion in the boundary layer is computed by the EDDY module. Cloud convection is computed by CLOUD_RADM.

Literature Cited

(31) US EPA, Analyses in attainment demonstrations for guidance on the use of models and other the 8-hour ozone NAAQS. Draft Final Report, EPA-454/R-99-004, February 17, 2005. The US Environmental Protection Agency, Office of Air and Radiation/Office of Air Quality Planning and Standards, Research Triangle Park,

NC, **2005**.

(32) Boylan, J. W.; Russell, A. G., PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models. *Atmospheric Environment*, **2006**, 40, 4946-4959.

(33) Zhang, Y.; Liu, P.; Queen, A.; Misenis, C.; Pun B.; Seigneur, C.; Wu, S.-Y. A comprehensive performance evaluation of MM5-CMAQ for the Summer 1999 Southern Oxidants Study episode—Part II: Gas and aerosol predictions. *Atmos. Environ.*, **2006**, 40, 4839-4855.

(34) Drummond, J. R.; Mand, G. S. The Measurements of Pollution in the Troposphere (MOPITT) instrument: overall performance and calibration requirements. *J. Atmos. Oceanic Tech.*, **1996**, 13, 314-320.

(35) Deeter, M.N.; et al. Operational carbon monoxide retrieval algorithm and selected results for the MOPITT instrument. *J. Geophys. Res.*, **2003**, 108, 4399, 10.1029/2002JD003186.

(36) Deeter, M. N.; Emmons, L.K.; D.P. Edwards; and J.C. Gille. Vertical resolution and information content of CO profiles retrieved by MOPITT. *Geophy. Res*. Lett. **2004**, 31, 10.1029/2004GL020235

(37) Olerud, D. T.; Sims, A. MM5 sensitivity Modeling in Support of VISTAS (Visibility Improvement – State and Tribal Association), VISTAS task 2e deliverable; **2003.**