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Atmospheric Chemistry and Physics Discussions

Ultrafine Particulate Matter Source Contributions across the Continental United States

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Atmospheric Chemistry and Physics Discussions

Abstract

The regional concentration of airborne ultrafine particulate matter mass (Dp < 0.1 µm; $PM_{0.1}$) was predicted with 4km resolution in 39 cities across the United States during summer time air pollution episodes. Calculations were performed using a regional chemical transport model with 4 km spatial resolution operating on the National Emissions Inventory created by the US EPA. Measured source profiles for particle size and composition between 0.01 – 10 µm were used to translate PM total mass to $PM_{0.1}$. $PM_{0.1}$ concentrations exceeded 2 µg m^{-3} during summer pollution episodes in major urban regions across the US including Los Angeles, the San Francisco Bay Area, Houston, Miami, and New York. $PM_{0.1}$ spatial gradients were sharper than $PM_{2.5}$ spatial gradients due to the dominance of primary aerosol in $PM_{0.1}$. Artificial source tags were used to track contributions to primary $PM_{0.1}$ and $PM_{2.5}$ from fifteen source categories. As expected, on-road gasoline and diesel vehicles made significant contributions to regional $PM_{0.1}$ in all 39 cities even though peak contributions within 0.3 km of the roadway were not resolved by the 4 km grid cells. Food cooking also made significant contributions to $PM_{0.1}$ in all cities but biomass combustion was only important in locations impacted by summer wildfires. Aviation was a significant source of $PM_{0.1}$ in cities that had airports within their urban footprints. Industrial sources including cement manufacturing, process heating, steel foundries, and paper & pulp processing impacted their immediate vicinity but did not significantly contribute to $PM_{0.1}$ concentrations in any of the target 39 cities. Natural gas combustion made significant contributions to $PM_{0.1}$ concentrations due to the widespread use of this fuel for electricity generation, industrial applications, residential, and commercial use. The major sources of primary $PM_{0.1}$ and $PM_{2.5}$ were notably different in many cities. Future epidemiological studies may be able to differentiate $PM_{0.1}$ and $PM_{2.5}$ health effects by contrasting cities with different ratios of $PM_{0.1}$ / $PM_{2.5}$. In the current study, cities with higher $PM_{0.1}$ / $PM_{2.5}$ ratios include Houston TX, Los Angeles CA, Birmingham AL, Charlotte NC, and Bakersfield CA. Cities with lower $PM_{0.1}$ to $PM_{2.5}$ ratios include Lake Charles LA, Baton Rouge LA, St. Louis MO, Baltimore MD, and Washington DC.

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Atmospheric Chemistry and Physics Discussions

1. Introduction

Airborne particulate matter (PM) has been linked with premature mortality and numerous other health risks in cities across the world (see for example references (Laden, Neas et al. 2000, Pope, Burnett et al. 2002, Dominici, Peng et al. 2006, Ostro, Broadwin et al. 2006, Franklin, Zeka et al. 2007, Pope, Ezzati et al. 2009, Kheirbek, Wheeler et al. 2013, Aneja, Pillai et al. 2017)). Despite years of progress (EPA 2017), PM concentrations in many urban regions in the United States still exceed health-based standards resulting in an increase of non-accidental mortality (Franklin, Zeka et al. 2007, Baxter, Duvall et al. 2013). Toxicology testing suggests that ultrafine particles with diameter < 0.1 μm may be the most harmful size fraction within $PM_{2.5}$ (Oberdorseter, Gelein et al. 1995, Pekkanen, Timonen et al. 1997, Oberdurster 2000, Li, Siotas et al. 2003, Ostro, Hu et al. 2015). Initial attempts to analyze ultrafine particles in epidemiology studies have used particle number concentration as a surrogate for ultrafine particle exposure, but this approach has not found consistent relationships with health effects (Ostro, Hu et al. 2015). In contrast, a recent epidemiology study based on ultrafine particle mass ($PM_{0.1}$) found significant associations with premature mortality (Ostro, Hu et al. 2015). Follow-up studies have also found significant associations between $PM_{0.1}$ and reproductive outcomes including birth weight and preterm birth (Bergin, Russell et al. 1996, Laurent, Hu et al. 2016). These findings have biological plausibility, since ultrafine particles may cross cell membranes and interfere with the internal cell function (Sioutas, Delfino et al. 2005). The toxic material found in ultrafine particles has greater surface area due to the small particle diameter making the material more available for chemical reaction. Ultrafine particles can therefore have a larger impact when deposited deep into the lung cavity where they are not easily removed (Li, Siotas et al. 2003, Nel, Xia et al. 2006).

A national monitoring network for PM10 and $PM_{2.5}$ has been operating throughout the continental US for almost 20 years. Multiple studies have performed source apportionment calculations for coarse and fine PM using these measurements (Zheng, Cass et al. 2002, Reff, Bhave et al. 2009, Ham and Kleeman 2011, Zhang, Hu et al. 2014). In contrast, measurements of $PM_{0.1}$ are limited to focused field campaigns lasting for short time periods with even fewer studies attempting source apportionment calculations (Kleeman, Riddle et al. 2009). Multiple barriers have prevented the widespread deployment of $PM_{0.1}$ monitoring networks including (i) the low concentration of $PM_{0.1}$ mass, which challenges the detection limits of analytical methods,

(ii) the artifacts associated with collecting $PM_{0.1}$ samples, (iii) the additional workload involved in operating the collection devices, and (iv) the sharp spatial gradients of $PM_{0.1}$ fields. Expensive investments in $PM_{0.1}$ monitoring are unlikely to occur without compelling evidence linking $PM_{0.1}$ to public health. Early epidemiological studies for $PM_{0.1}$ must therefore use some other technique besides direct measurements to calculate population exposure.

Various methods such as the source-resolved PMCAMx chemical transport model, the chemical mass balance (CMB) model, photochemical box models and land use regression (LUR) models have been used to track source contributions to primary organic matter, elemental carbon and in some cases particle number concentration (PNC) over areas in the Eastern U.S. and parts of Europe and Asia (Gaydos, Stanier et al. 2005, Lane, Pinder et al. 2007, Wang, Hopke et al. 2011, Posner and Pandis 2015, Cattani, Gaeta et al. 2017, Wolf, Cyrys et al. 2017, Simon, Patton et al. 2018, Zhong, Nikolova et al. 2018). However, these methods are limited in one or more aspects of their ability to predict population exposure ultrafine particles over large analysis domains. Source resolved models, such as PMCAMx, have been demonstrated for PNC but not for $PM_{0.1}$ (Posner and Pandis 2015). CMB models need measurements of specific molecular markers at numerous sites to resolve the sharp spatial gradients of ultrafine particle source contributions. LUR models need comprehensive measurements that act as training data sets in order to extend throughout a modeling domain (Lane, Pinder et al. 2007).

Hu et al. (Hu, Zhang et al. 2014) calculated population exposure to $PM_{0.1}$ in California using a regional source-oriented chemical transport model supported by measured profiles for particle size and composition emitted by dominant sources. Predictions were compared to all available fine and ultrafine particle measurements over the period 2000-2010 with good agreement observed for the dominant chemical components of $PM_{0.1}$ mass including organic aerosol, elemental carbon, and numerous trace metals (Hu, Zhang et al. 2014). The 4km spatial resolution used in these calculations supported multiple epidemiological studies based on spatial gradients of exposure (Ostro, Hu et al. 2015, Laurent, Hu et al. 2016). These encouraging results motivate the expansion of the $PM_{0.1}$ exposure technique to other locations.

Here we use the Eularian source-oriented UCD/CIT chemical transport model to predict the concentration of $PM_{0.1}$ in thirty-nine urban regions throughout the US during summer pollution events in 2010. The calculation tracks contributions from fifteen (15) primary particle sources through a simulation of all major atmospheric processes while retaining information

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Discussion started: 3 September 2018

about particle size, composition and source origin (Hu, Zhang et al. 2014). The results of this calculation reveal US national trends in $PM_{0.1}$ concentrations for the first time and suggest locations where the differential health effects of $PM_{0.1}$ and $PM_{2.5}$ can best be studied.

2. Methods

2.1 Simulation Dates

Simulations within each target city were carried out during peak summer air pollution events in 2010. Peak air pollution events typically had measured 1-hr maximum ozone (O_3) concentrations greater than 70 ppb. Regional pollution events caused by atmospheric stagnation were selected whenever possible as opposed to special events caused by unusual occurrences such as wildfires. Measured $PM_{2.5}$ 24-hr concentrations during peak summer pollution events ranged between 3.2-30 $\mu g/m^3$ depending on the location. The simulation dates in each city are listed in Table 1 and a map of the city locations is shown in the supplemental information Figure S1. The aggregation of these events across the US enables a comparison of typical air pollution episodes within different cities.

Table 1. City, Simulation Date, 2010 Population and Geographical Region

City	2010 Date	2010 Population	US Geographical Region
Atlanta	March 29 - April 1	422765	South East
Austin	August 23 - August 26	815260	South
Bakersfield	August 23 - August 26	348938	West
Baltimore	August 7 - August 10	621210	East Coast
Baton Rouge	October 6 - October 9	229584	South
Birmingham	October 6 - October 9	212107	South East
Boston	August 29 - September 1	620451	East Coast
Charlotte	March 30 - April 2	738710	South East
Cincinnati	August 25 - August 28	296904	Midwest
Cleveland	August 25 - August 28	396009	Midwest
Dallas	August 23 - August 26	1201000	South
Denver	July 13 - July 16	603421	West
Detroit	August 25 - August 28	711299	Midwest
El Paso	July 11 - July 14	651665	West
Fresno	August 23 - August 26	497090	West

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
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Discussion started: 3 September 2018

Atmospheric Chemistry and Physics Discussions

City	2010 Date	2010 Population	US Geographical Region
Hartford	August 29 - September 1	125312	East Coast
Houston	October 6 - October 9	2103000	South
Indianapolis	August 25 - August 28	830952	Midwest
Jacksonville	March 29 - April 1	823291	South East
Kansas City	August 25 - August 28	460639	Midwest
Lake Charles	October 6 - October 9	72268	South
Los Angeles	September 23 - September 26	3796000	West
Louisville	August 7 - August 10	300000	Midwest
Memphis	October 6 - October 9	647609	Midwest
Miami	March 30 - April 2	400769	South East
Nashville	October 7 - October 10	1800000	Midwest
New York City	August 29 - September 1	8190000	East Coast
Philadelphia	August 27 - August 30	1529000	East Coast
Phoenix	June 19 - Jun3 22	1449000	West
Portland	August 23 - August 26	585286	West
Richmond	August 7 - August 10	204351	East Coast
Sacramento	August 22 - August 25	466488	West
Salt Lake City	August 18 - August 21	186505	West
San Antonio	August 23 - August 26	1334000	South
San Diego	September 23 - September 26	1306000	West
San Francisco	August 22 - August 25	805704	West
St. Louis	August 25 - August 28	319257	Midwest
Tulsa	August 25 - August 28	392443	Midwest
Washington DC	August 7 - August 10	604453	East Coast

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2.2 Model Description

The UCD/CIT model predicts the evolution of gas and particle phase pollutants in the atmosphere in the presence of emissions, transport, deposition, chemical reaction and phase
125 change (Held, Ying et al. 2005) as represented by Eq. (1)

$$\frac{\partial C_i}{\partial t} + \nabla \cdot uC_i = \nabla K \nabla C_i + E_i - S_i + R_i^{gas}(C) + R_i^{part}(C) + R_i^{phase}(C) \quad (1)$$

where C_i is the concentration of gas or particle phase species *i* at a particular location as a function of time *t*, *u* is the wind vector, *K* is the turbulent eddy diffusivity, E_i is the emissions rate, S_i is the loss rate, R_i^{gas} is the change in concentration due to gas-phase reactions, R_i^{part} is the change in concentration due to particle-phase reactions and R_i^{phase} is the change in concentration due to phase change (Held, Ying et al. 2005). Loss rates include both dry and wet deposition. Phase change for inorganic species occurs using a kinetic treatment for gas-particle conversion (Hu, Zhang et al. 2008) driven towards the point of thermodynamic equilibrium (Nenes, Pilinis et al. 1998). Phase change for organic species is also treated as a kinetic process with vapor pressures of semi-volatile organics calculated using the 2-product model (Carlton, Bhave et al. 2010). More sophisticated approaches for secondary organic aerosol (SOA) formation (Cappa, Jathar et al. 2016) were also tested in the current study but these required a larger number of assumptions and they did not produce higher SOA concentrations in the $PM_{0.1}$ size fraction. Nucleation was not included in the current study and so particle number concentrations will not be discussed. Likewise, model spatial resolution was 4km over the 4.2 million km^2 of simulated urban areas and so near-roadway concentrations of ultrafine particles on spatial scales of ~0.1 km will not be presented.

A total of 50 particle-phase chemical species are included in each of 15 discrete particle size bins that range from 0.01-10 µm particle diameter (Held, Ying et al. 2005). Artificial source tags are used to quantify source contributions to the primary particle mass for a specific bin size, therefore allowing for the direct contribution of each source of $PM_{2.5}$ and $PM_{0.1}$ mass to be determined. Gas-phase concentrations of oxides of nitrogen (NOx), volatile organic compounds (VOCs), oxidants, ozone, and semi-volatile reaction products were predicted using the SAPRC-11 chemical mechanism (Carter and Heo 2013).

2.3 Model Inputs

Anthropogenic emissions were generated using the Sparse Matrix Operator Kernel Emissions (SMOKEv3.7) modeling system applied to the 2011 National Emissions Inventory. Emissions from each of the four major source sectors (area, mobile, non-road and point were tagged to create fifteen (15) different emissions groups: on road diesel, on road gasoline, off road diesel, off road gasoline, biomass, food cooking, natural gas, process heaters, distillate oil, aviation, cement, coal, steel foundries, paper products and all other emissions. Size and

160 composition-resolved source profiles were then assigned to the PM emissions within each of these groups using the UCD/CIT emissions processor based on the most recent measurements available in the literature (Robert, VanBergen et al. 2007a, Robert, Kleeman et al. 2007b, Kleeman, Robert et al. 2008). Some of the fifteen (15) source categories were represented using weighted average source profiles from multiple sources as described in Table S1.

165 Daily values for 2010 wildfire emissions were generated using the Global Fire Emissions Database (GFED) (Giglio, Randerson et al. 2013). Biogenic emission rates were generated using the Model of Emissions of Gases and Aerosols from Nature (MEGANv2.1). The gridded geo-referenced emission factors and land cover variables required for MEGAN calculations were created using the MEGANv2.1 pre-processor tool and the ESRI_GRID leaf area index and plant 170 functional type files available at the Community Data Portal (Guenther, Jiang et al. 2012).

Meteorology parameters used to drive the UCD/CIT CTM were generated using the Weather Research and Forecasting model (WRFv3.6) and WRF preprocessing system (WPSv3.6). Meteorological fields were created within 3 nested domains with horizontal resolutions of 36km, 12km, and 4km, respectively. Each domain had 31 telescoping vertical 175 levels up to a top height of 12km. Four-dimensional data assimilation (FDDA) or "FDDA nudging" was used to anchor meteorological predictions to measured values (Hu et al., 2010). Meteorological data and gridded map projections needed for 2010 emissions modeling were taken from the corresponding WRF simulations using the meteorology-chemistry interface processor (MCIP).

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2.3 Supporting Measurements

Ambient hourly ozone measurements and daily $PM_{2.5}$ measurements were obtained from the EPA AQS API / Query AirData (EPA). Model predictions are compared to these measurements to build confidence in the accuracy of the overall modeling system since $PM_{0.1}$ 185 measurements are not available during any of the peak summer pollution events studied here.

3. Results

Predicted 1-hr ozone concentrations were compared to measurements averaged within each city to indirectly evaluate the accuracy of the emissions inventories and meteorology fields. 190 Many of the sources that emit ozone precursors also emit ultrafine particles. Likewise,

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 3 September 2018

Atmospheric Chemistry and Physics Discussions

meteorological parameters including wind speed and mixing depth influence the concentrations of all pollutants including ultrafine particles. Successful prediction of ozone is therefore a necessary step in the accurate prediction of ultrafine particle concentrations during summer photochemical smog episodes. Figure 1 illustrates the time series of predicted vs measured ozone concentration for four (4) representative cities spanning the South, East, Midwest and West US regions. The full set of comparisons for all 39 cities are shown in the supplemental information Figure S2. In general, model simulations capture the peak ozone concentration and diurnal pattern during the pollution events. Mean fractional bias (MFB) and mean fractional error (MFE) summary statistics meet EPA criteria in 37 out of 39 cities (Table S2 in the supplemental information).

Predicted 24-hr $PM_{2.5}$ concentrations were compared to measurements as a second check on the accuracy of model features needed to predict ultrafine particle concentrations. Many of the combustion sources that emit primary particles within the $PM_{2.5}$ size fraction also emit $PM_{0.1}$. The Chemical Speciation Monitoring Network (CSN) operated by the U.S. Environmental Protection Agency (EPA) measures $PM_{2.5}$ mass and chemical composition at more than 260 sites throughout the U.S. including many of the 39 cities studied in the current analysis (Solomon, Crumpler et al. 2014). Elemental carbon (EC) and organic compounds (OC) are the chemical components most relevant for both the $PM_{2.5}$ and the $PM_{0.1}$ size fractions. Figure 2 illustrates predicted vs measured 24-hr $PM_{2.5}$ EC and OC concentrations for all 39 cities while Figure S3 illustrates predicted vs. measured 24-hr $PM_{2.5}$ total mass comparisons. In general, the model slightly under predicts $PM_{2.5}$ EC, OC, and mass with regression slopes ranging from 0.62 for EC to 0.97 for OC. The negative bias in model predictions may stem from the 4km spatial averaging inherent in the calculations vs. the influence of sources closer than 4 km to the measurement site in the urban environment such as highways, restaurants, etc. This trend is reflected in the performance of ozone predictions during the evening hours for Los Angeles and New York City (Figure 1), where measured ozone concentrations fall to zero due to titration from nearby NOx emissions while predicted ozone concentrations remain greater than zero due to dilution of NOx emissions within 4 km grid cells. The MFB and MFE for $PM_{2.5}$ predictions are summarized in the supplemental information Table S2.

As was the case for ozone predictions, $PM_{2.5}$ model performance meets EPA criteria (MFE<0.75) in 37 out of 39 cities, building confidence in the accuracy of the model results for

PM concentrations. MFB values lower than 0.15 and MFE values lower than 0.35 are considered the goal or "excellent" in model performance. In the current study, the average MFB and MFE across all 39 cities was 0.126 and 0.379 for O_3, and -0.27 and 0.38 for $PM_{2.5}$ respectively.

$PM_{0.1}$ measurements are not available for model evaluation in the 39 cities across the US in 2010 at the core of the current study, but measurements are available in California in the years 2015 and 2016 that can serve to evaluate similar modeling procedures. Yu et al. (Yu, Venecek et al. 2018) compared $PM_{0.1}$ concentrations in Los Angeles, Fresno, East Oakland, and San Pablo, California predicted using the UCD/CIT air quality model to receptor-based source apportionment calculations based on measured concentrations of molecular markers in the ultrafine particle size fraction (Xue, Xue et al. 2018). Good agreement was found between predictions from these two independent techniques for $PM_{0.1}$ concentrations associated with gasoline engines, diesel engines, food cooking, wood burning, and "other sources". Further details of this comparison are provided by (Yu, Venecek et al. 2018). This evaluation of the modeling procedures builds confidence in the $PM_{0.1}$ source predictions across the US in the current study.

Figure 3 illustrates a composite representation of $PM_{2.5}$ and $PM_{0.1}$ mass across the US during the summer pollution episodes listed in Table 1. The spatial plot in Figure 3 is constructed using the intermediate 12km simulation results from multiple simulations stitched together to cover a broader geographical area. Regional $PM_{0.1}$ concentrations reach a maximum value of 5 μg m^{-3} in a few isolated grid cells with wildfires but concentrations generally exceed 2 μg m^{-3} in major urban regions across the US including Los Angeles, the San Francisco Bay Area, Houston, Miami, and New York. The comparison between $PM_{2.5}$ mass (Figure 3a) and $PM_{0.1}$ mass (Figure 3b) shows that $PM_{0.1}$ spatial gradients are sharper with less regional contributions between "hot spots". Locations in the Midwestern and Eastern US outside of cities with high $PM_{2.5}$ concentrations due to secondary formation (sulfate and secondary organic aerosol) did not have corresponding high concentrations of $PM_{0.1}$. Most major urban centers had noticeable peaks of both $PM_{2.5}$ and $PM_{0.1}$. This pattern presents a challenge for epidemiological studies seeking to differentiate the effects of $PM_{2.5}$ and $PM_{0.1}$ because the locations with differential exposure (high $PM_{2.5}$ but low $PM_{0.1}$) have low population density which will reduce the power of the analysis.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 3 September 2018

Atmospheric Chemistry and Physics Discussions

The UCD/CIT model explicitly tracks source contributions to particle mass in each size bin using artificial source tags. Pie charts of $PM_{2.5}$ and $PM_{0.1}$ source contributions are illustrated in Figure 3 for selected major cities. Pie charts for $PM_{0.1}$ source contributions in all 39 US cities are shown in Figure 4. The detailed source profiles within each city are based on the nested 4km simulation results during the pollution events listed in Table 1. Source contribution spatial plots for the entire US are shown in the supplemental information Figures S4-S7 and pie charts for $PM_{2.5}$ source contributions in all 39 US cities are shown in the supplemental information Figure S8. As expected, on-road gasoline and diesel vehicles made significant contributions to regional $PM_{0.1}$ in all 39 cities even though peak contributions within 0.3 km of the roadway were not resolved by the 4 km grid cells. Food cooking also made significant contributions to $PM_{0.1}$ in all cities but biomass combustion was only important in locations impacted by summer wildfires. Aviation was a significant source of $PM_{0.1}$ in cities that had airports within their urban footprints. Industrial sources including cement manufacturing, process heating, steel foundries, and paper & pulp processing impacted their immediate vicinity but did not significantly contribute to $PM_{0.1}$ concentrations in any of the target 39 cities. Natural gas combustion made significant contributions to $PM_{0.1}$ concentrations due to the widespread use of this fuel for residential, commercial, and industrial applications. Natural gas contributions were especially significant in locations with high levels of industrial use such as chemical refineries or in locations with significant levels of natural gas fired power plants.

The major sources of primary $PM_{0.1}$ and $PM_{2.5}$ were notably different in many cities (compare Figure 3a and 3b). The sources that contribute most strongly to $PM_{2.5}$ are on road diesel, gasoline, food cooking, coal and “other” which includes break and tire wear from mobile sources and dust. Natural gas combustion makes minor contributions to primary $PM_{2.5}$ mass since particles from this source have a mass distribution peaking at ~0.05 µm particle diameter (Chang, Chow et al. 2004) with all of the emitted mass in the $PM_{0.1}$ size fraction. In contrast, other combustion sources using more complex fuels such as on-road vehicles have a mass distribution peaking at ~0.1 µm with at least half the emitted mass outside the $PM_{0.1}$ size fraction (Robert, VanBergen et al. 2007a, Robert, Kleeman et al. 2007b). Likewise, food cooking contributes strongly to $PM_{2.5}$ concentrations but the emitted particle mass distribution peaks at 0.2 µm with the majority of the mass outside the $PM_{0.1}$ size fraction.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 3 September 2018

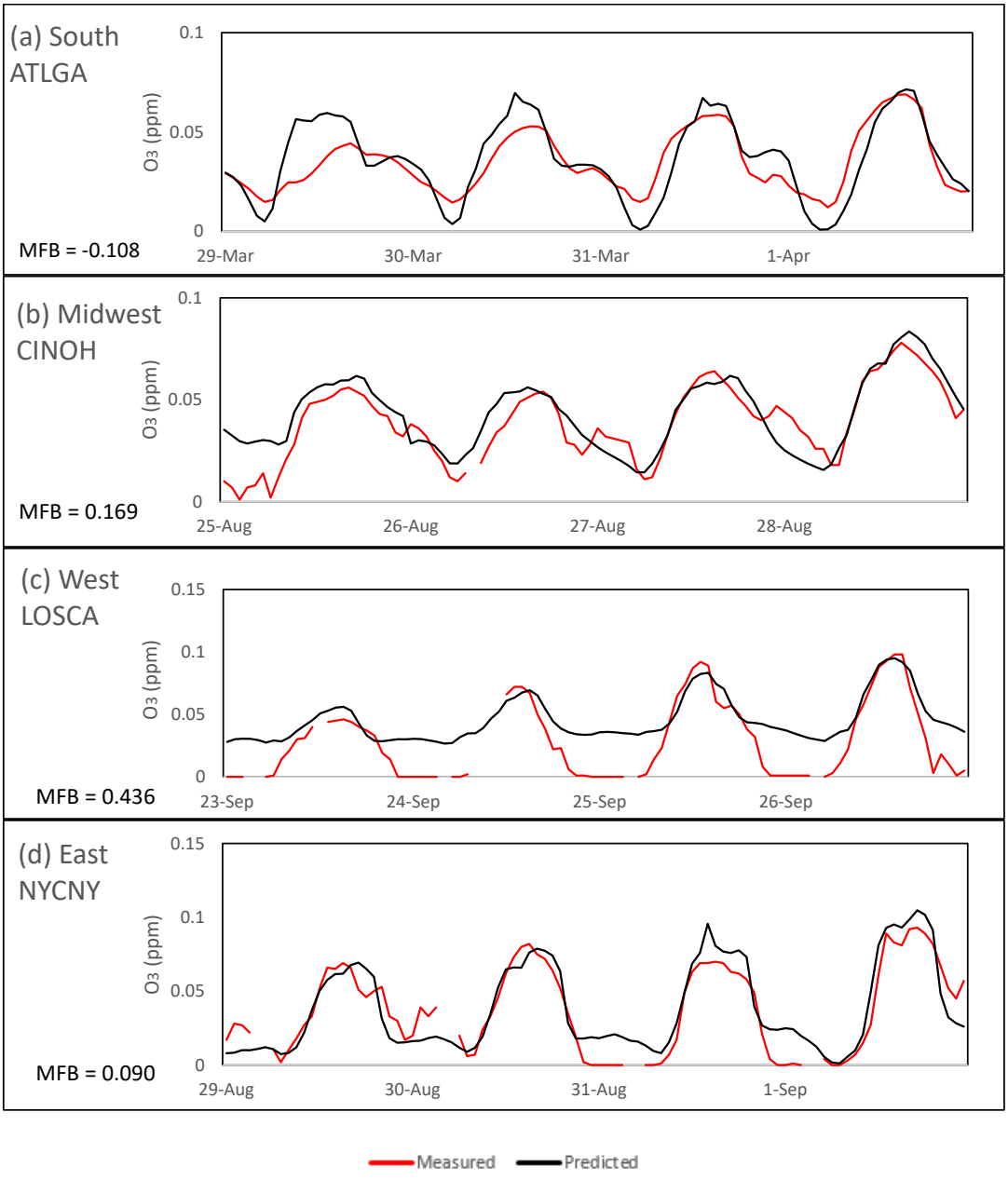

Figure 1. Time series of 1-hr measured vs predicted ozone concentration (ppm) for 4 selected city scenarios representative of the major geographical regions across the Continental United States

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
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Discussion started: 3 September 2018

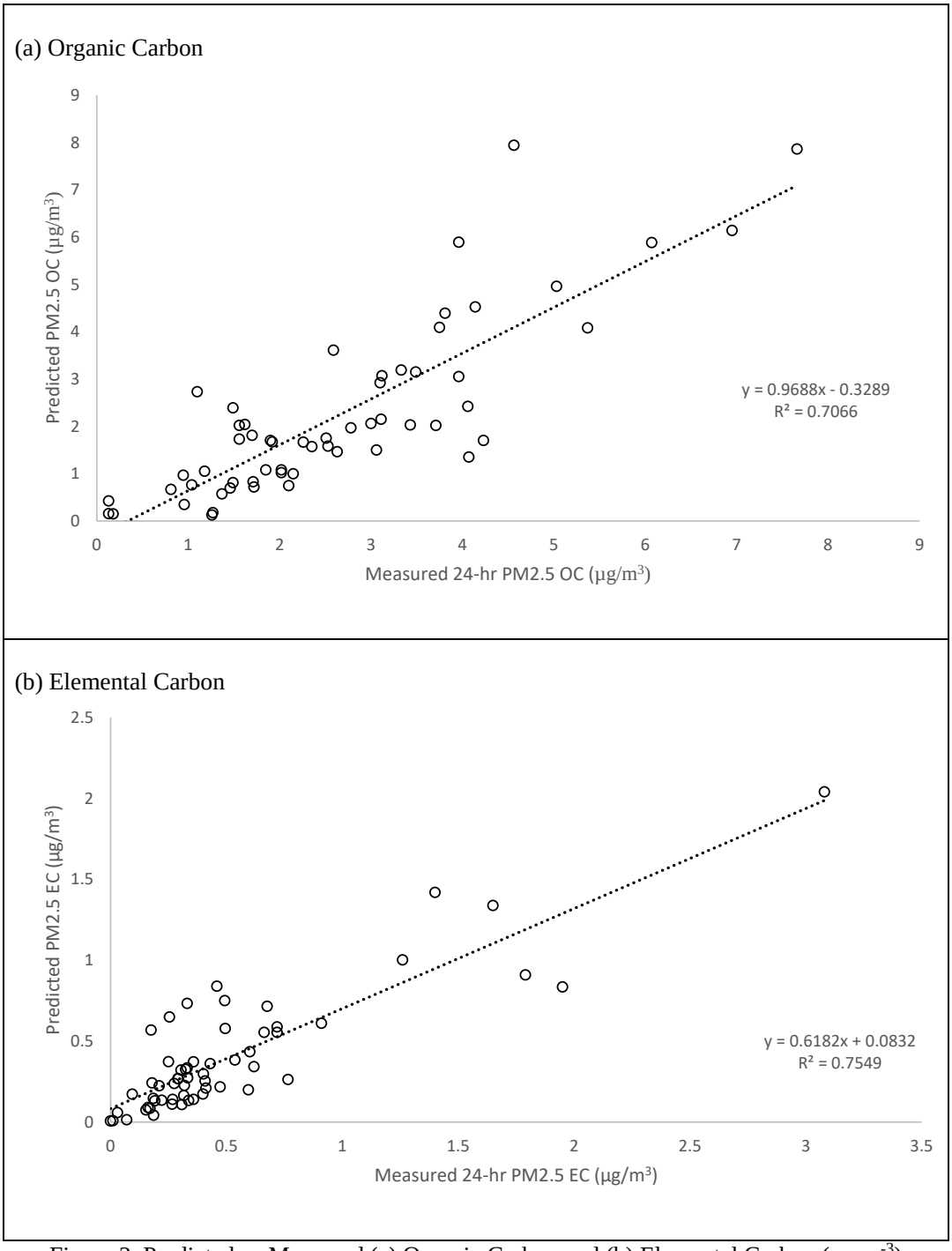

Figure 2. Predicted vs Measured (a) Organic Carbon and (b) Elemental Carbon (µg m^{-3})

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
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Discussion started: 3 September 2018

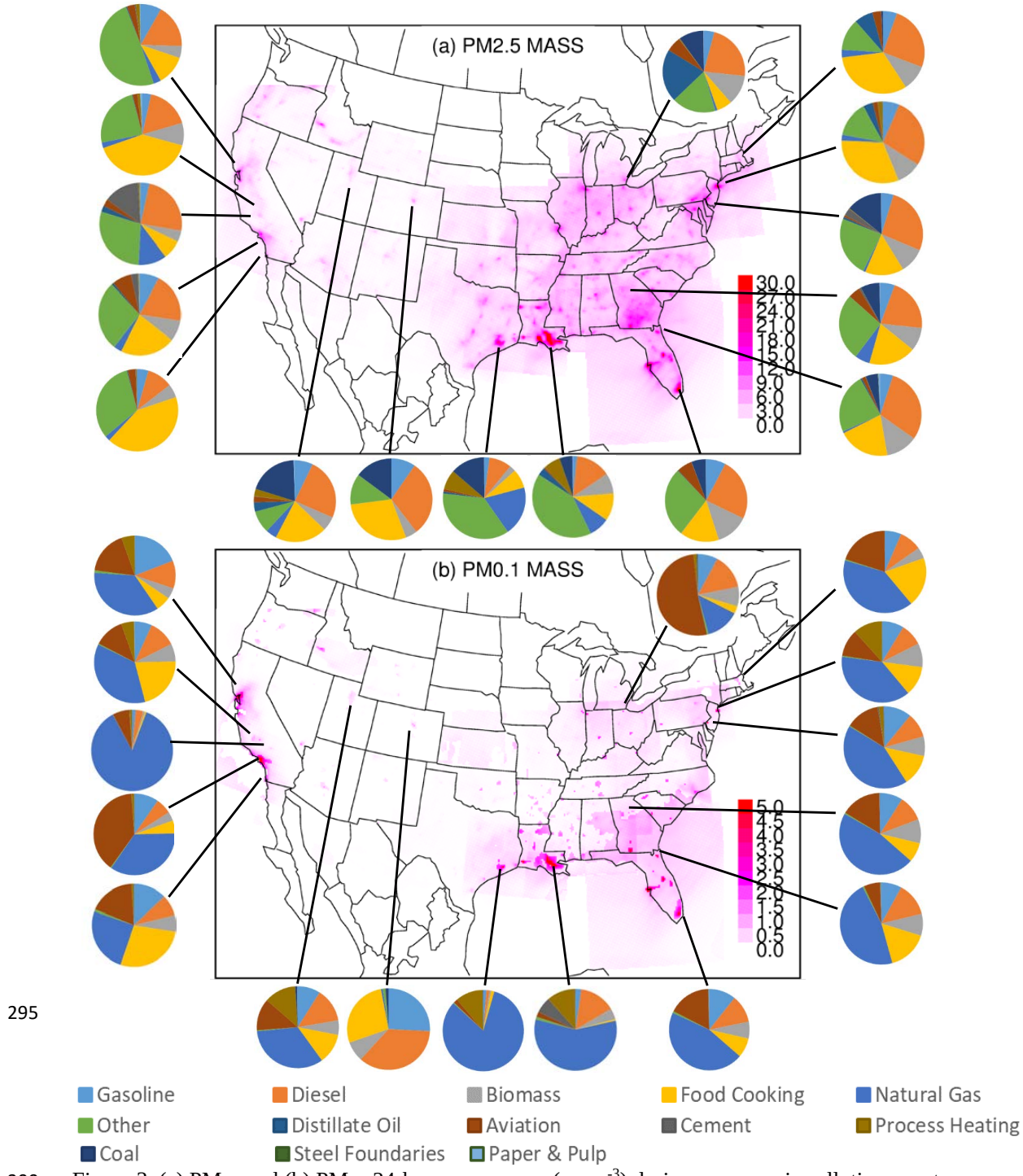

Figure 3. (a) $PM_{2.5}$ and (b) $PM_{0.1}$ 24-hr average mass (μg m^{-3}) during summer air pollution event. Scale drawn to highlight all areas of US. Actual Max for (a) = 109.28 μg/m^3 (b) = 7.71 μg/m^3.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
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Discussion started: 3 September 2018

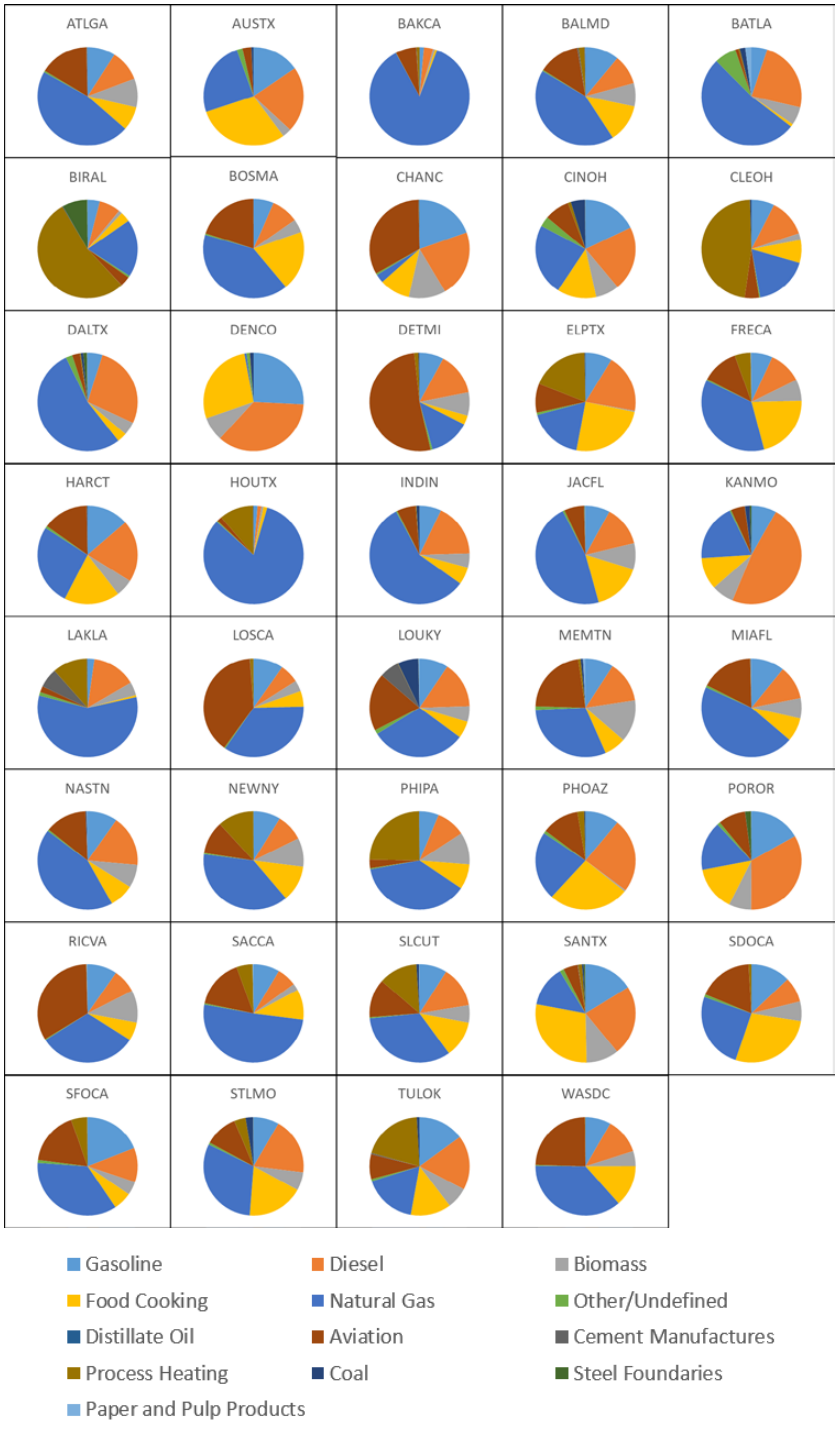


Figure 4. $PM_{0.1}$ source contribution for 39 cities across the continental US

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 3 September 2018

Atmospheric Chemistry and Physics Discussions

305

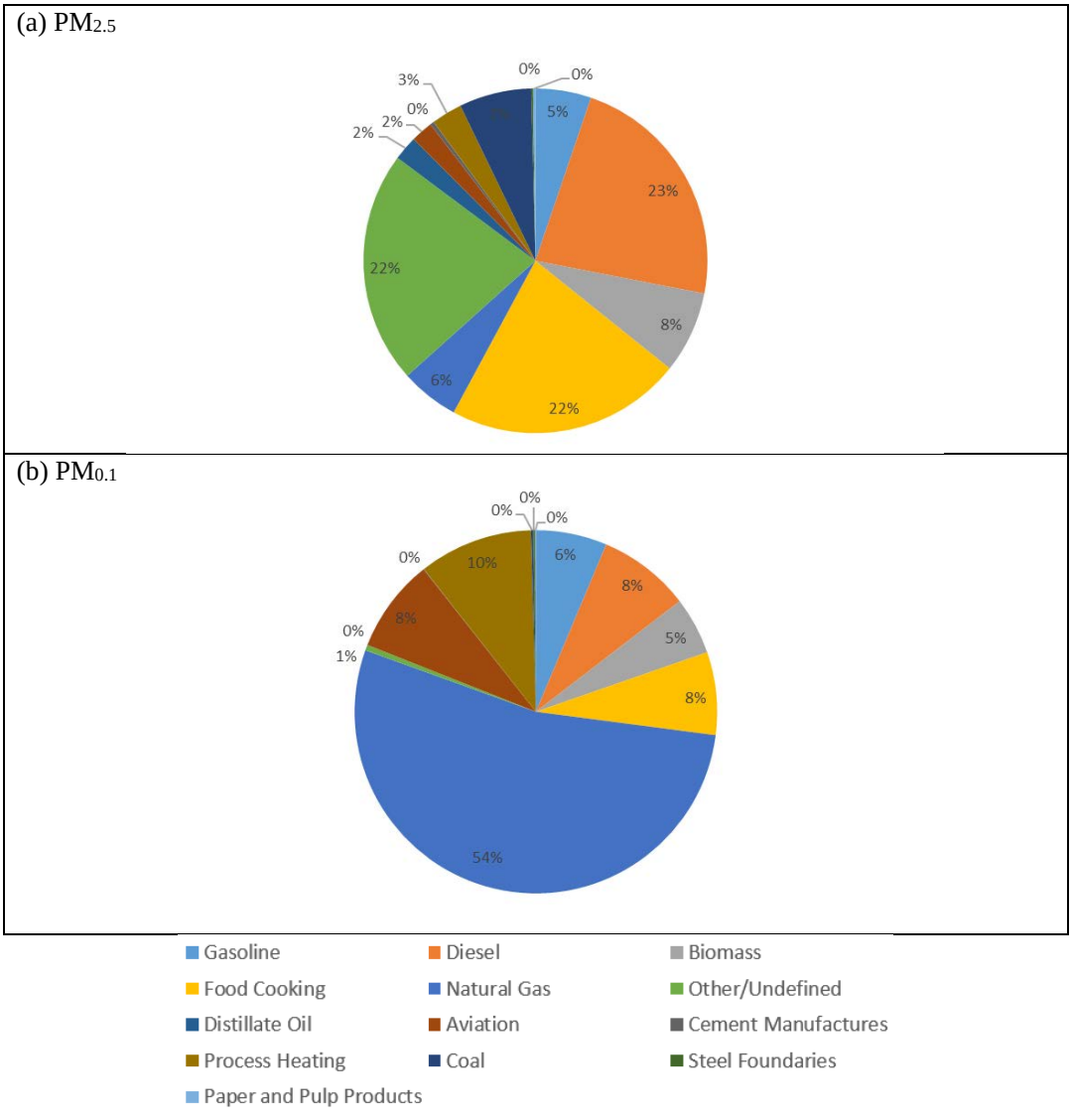


Figure 5. Population weighted average source contribution across the 39 major cities in the continental US for (a) $PM_{2.5}$ and (b) $PM_{0.1}$

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 3 September 2018

Atmospheric Chemistry and Physics Discussions

4. Discussion

Figure 5 illustrates the population-weighted average $PM_{0.1}$ source contributions across all 39 study cities shown in Table 1. This calculation highlights the importance of natural gas combustion particles in the $PM_{0.1}$ size fraction and the minor role that these natural gas combustion particles play in the $PM_{2.5}$ size fraction. Natural gas typically consists of +93% methane with the balance of the fuel made up by higher molecular weight alkanes and trace impurities. In addition to background sulfur compounds in the natural gas, sulfur-containing odorants such as mercaptans are commonly added to aid in leak detection.

Natural gas combustion does not emit high amounts of particulate matter per J of energy in the fuel, but the widespread use of natural gas suggests that it could still contribute significantly to ambient $PM_{0.1}$ concentrations. Natural gas combustion accounted for 29% of total US energy consumption in 2016 (Energy 2017). In contrast, gasoline combustion accounted for 17% of US energy consumption and diesel fuel combustion accounted for approximately 6% of US energy consumption in 2016. Gasoline and diesel fuel combustion in motor vehicles also emit most particles in the size fraction larger than $PM_{0.1}$ (Robert, VanBergen et al. 2007a, Robert, Kleeman et al. 2007b) whereas natural gas combustion emits particles entirely within the $PM_{0.1}$ size fraction (Chang, Chow et al. 2004). Taken together, these facts support the potential importance of natural gas combustion for ambient $PM_{0.1}$ concentrations.

The five (5) states with the highest consumption of natural gas in 2016 were Texas (14.7%), California (7.9%), Louisiana (5.7%), New York (5%), and Florida (4.8%). These consumption patterns are reflected in the natural gas distribution system (Figure 6a) and the predicted $PM_{0.1}$ concentration field associated with natural gas combustion (Figure 6b). Natural gas end-use included electric power generation (36%), industrial applications (34%), residential use (16%), commercial use (11%), and transportation (3%).

Lane et al. (2007) used a source-resolved version of PMCAMx and individual emission inventories to determine source contributions of primary organic material ($POM_{2.5}$) (Lane, Pinder et al. 2007). Lane et al. note that $POM_{2.5}$ associated with natural gas sources ranged from 0.1 to 0.8 $\mu g/m^3$. Chang et al in 2004 measured emitted particle size distributions for gas-fired stationary combustion that fell between 10-100nm (Chang, Chow et al. 2004). The combination of these two results indicates that the natural gas mass component of $POM_{2.5}$ predicted by Lane et al. is consistent with the magnitude of the PM0.1 mass associated with natural gas combustion

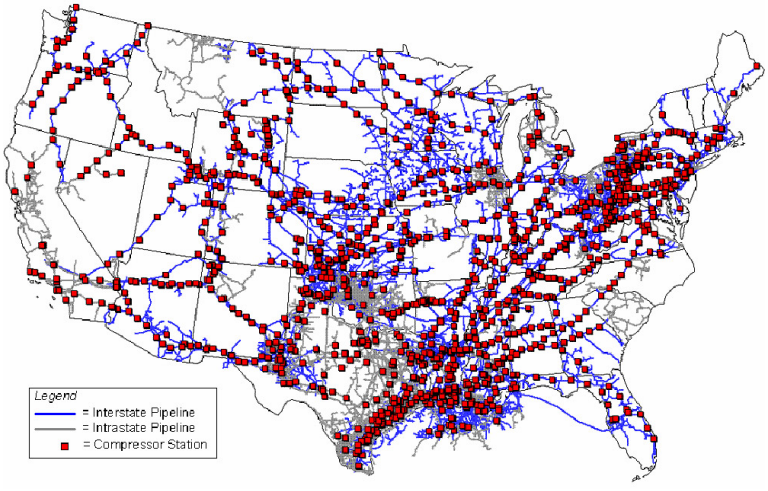
found in the current study. Lane et al. were not studying $PM_{0.1}$ and so the major role of natural gas in this size fraction was not identified.

Posner and Pandis (2015) utilized PMCAMx with the LADCO 2001 BaseE source-resolved mass emissions inventory for a July 2001 prediction of PNC over the Eastern United States with 36 km resolution (Posner and Pandis 2015). Posner and Pandis used a "zero-out" method in combination with source-specific size distribution to study the percent contribution of six major sources (on road gasoline, industrial, non-road diesel, on road diesel, biomass and dust) of PNC. They found that PNC was made up of 36% on-road gasoline, 31% industrial, 18% non-road diesel, 10% on-road diesel, 1% biomass burning and 4% long-range transport (Posner and Pandis 2015). The emissions particle number inventory was normalized based on PM10 mass from each source and particle emissions from natural gas sources were assumed negligible, which effectively removed natural gas sources from the simulation. This has minor effects on $PM_{2.5}$ and PM_{10} predictions, but the results of the current study suggest that natural gas combustion contributions significantly to ultrafine particle concentrations.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 3 September 2018

Atmospheric Chemistry and Physics Discussions

(a) Natural Gas Compressor Stations and Pipelines throughout the U.S. (Map courtesy of U.S. Energy Information Administration)



(b) UCD/CIT CTM Field Plot of $PM_{0.1}$ from Natural Gas sources

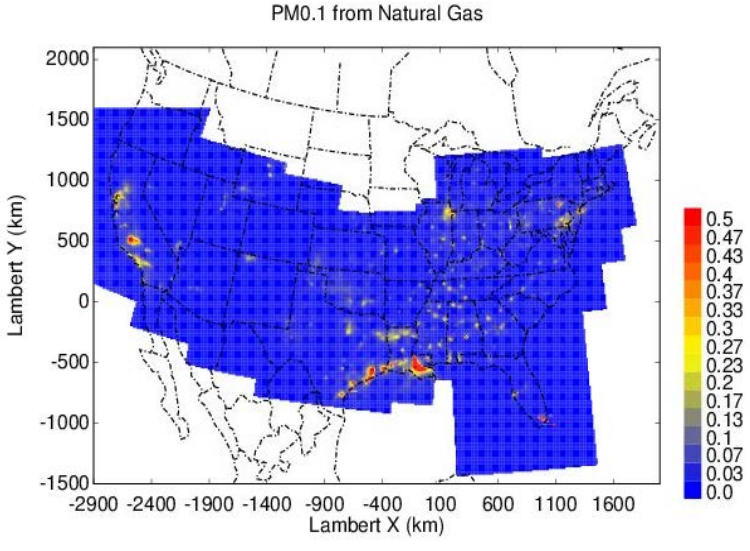


Figure 6. (a) Natural Gas compressor stations and pipelines across the US and (b) $PM_{0.1}$ Natural Gas combustion concentrations (μg m^{-3}).

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 3 September 2018

Atmospheric Chemistry and Physics Discussions

Future epidemiological studies may be able to differentiate $PM_{0.1}$ and $PM_{2.5}$ health effects by contrasting cities with different ratios of $PM_{0.1}$ / $PM_{2.5}$. Figure 7 illustrates the correlation between $PM_{2.5}$ and $PM_{0.1}$ concentrations in the 39 cities considered in the current analysis. Cities with higher $PM_{0.1}$ / $PM_{2.5}$ ratios include Houston TX, Los Angeles CA, Birmingham AL, Charlotte NC, and Bakersfield CA. Cities with lower $PM_{0.1}$ to $PM_{2.5}$ ratios include Lake Charles LA, Baton Rouge LA, St. Louis MO, Baltimore MD, and Washington DC. Measurements should be conducted in these locations to verify the contrast in $PM_{0.1}$ / $PM_{2.5}$ concentrations in preparation for future exposure analysis.

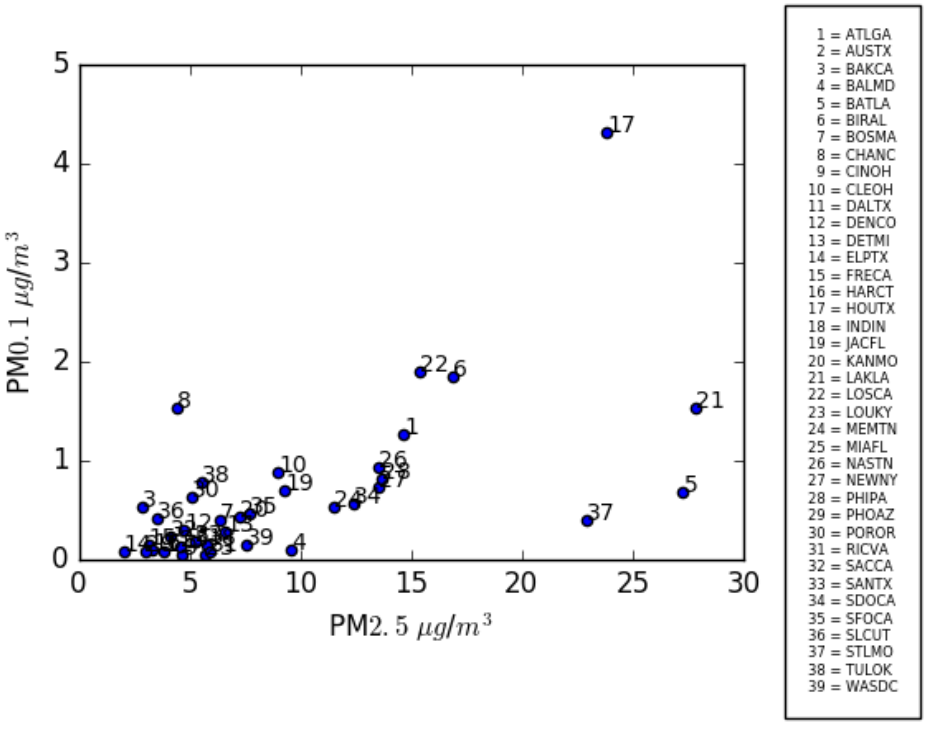


Figure 7. Scatter plot showing correlation between 24-hr average $PM_{2.5}$ and $PM_{0.1}$ for the 39-cities.

Future epidemiological studies may also be able to use the contrast in $PM_{0.1}$ source contributions between different cities to separately identify health effects. In the current study, the similarity in $PM_{0.1}$ source contributions between cities was calculated as a dot product. A

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 3 September 2018

source contribution vector was created for each city with 13 elements set equal to the normalized % contribution from each source. The dot product of each city source-vector with other city source-vectors was then calculated using eq. (2)

$$\vec{a} \cdot \vec{b} = \|\vec{a}\|\|\vec{b}\|\cos(\theta) \quad (2)$$

where $\vec{a}$ is the vector of city *i*, $\vec{b}$ is the vector of source for city *j*, $\|\vec{a}\|$ is the magnitude of city *i*, $\|\vec{b}\|$ is the magnitude of the vector for city *j* and θ is the angle between the two vectors ranging from 0 to 90º. *cos(θ)* quantifies the similarity in source contributions between the two cities. Rearranging Eq. (2) *cos(θ)* can be solved using Eq. (3)

$$\cos(\theta) = \left(\frac{\vec{a} \cdot \vec{b}}{\|\vec{a}\|\|\vec{b}\|}\right) \quad (3)$$

cos(*θ*) ranges between zero (0) for no correlation to one (1) for perfect correlation between the source vectors. Figure 8 illustrates the value of *cos(θ)* calculated for city comparisons for $PM_{0.1}$ (lower left) and $PM_{2.5}$ (upper right) source-vectors. The cities were arranged by region defined in Table 1 and starting from East, South East, South, Midwest and West in order to observe any geographical patterns. $PM_{2.5}$ source-vectors were found to be slightly more homogenous across all U.S. cities than $PM_{0.1}$ source vectors. Regional clusters with similar source contributions are apparent, especially on the East Coast where cities are closer in proximity to one another. Few regional clusters were observed for $PM_{0.1}$ source vectors, suggesting that emissions control programs may need to be tailored to each region. Natural gas combustion is prevalent in many locations, but the remaining sources of ultrafine particles vary strongly with location.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833
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Discussion started: 3 September 2018

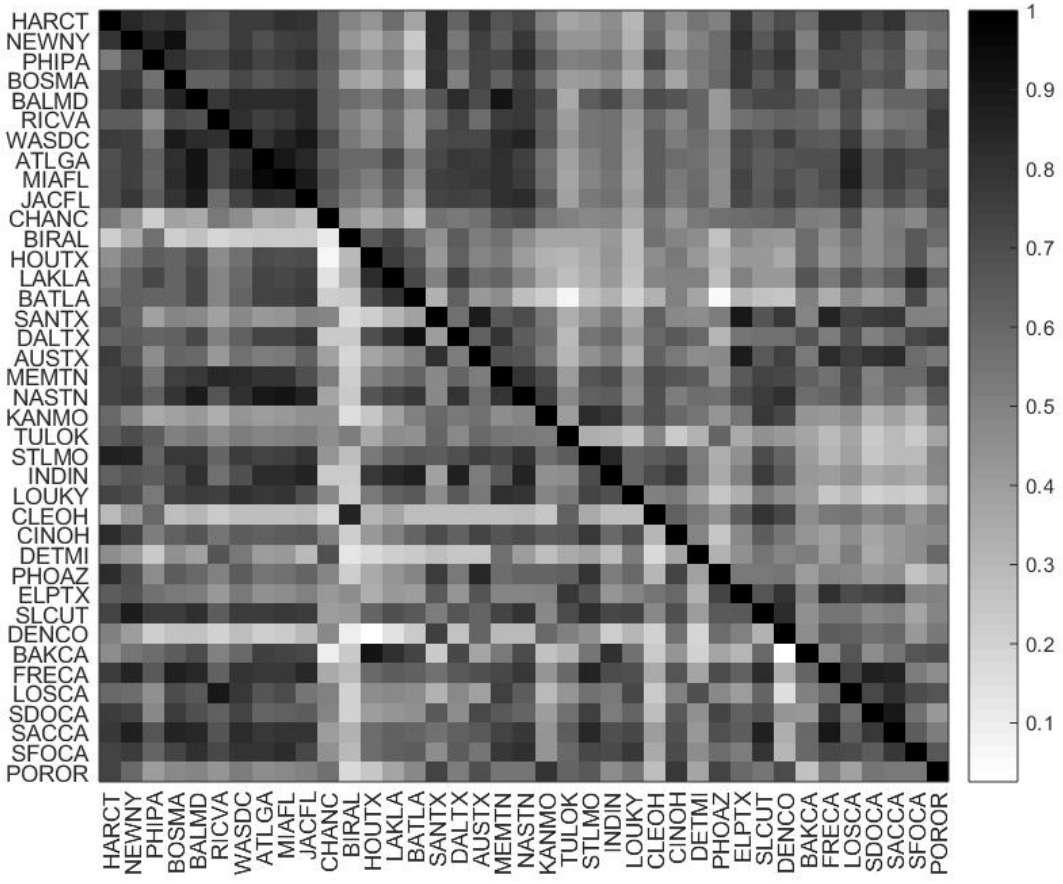

Figure 8. Normalized dot product between the 13 source types and each city for $PM_{2.5}$ (upper right) and $PM_{0.1}$ (lower left). The scale represents 100% (black) to 0% (white) correlation. Cities are organized by region in the following order: East, South East, South, Midwest and West.

5. Conclusion

The UCD/CIT regional chemical transport model was used to predict source contributions to $PM_{0.1}$ across the continental United States during peak photochemical smog periods during the year 2010. Model performance for $PM_{2.5}$ and ozone predictions met the recommendations for regulatory applications building confidence in the emissions inputs and meteorological fields used to drive the calculations. Similar model exercises carried out for episodes in California in 2015 and 2016 find good agreement between predicted $PM_{0.1}$ source

415 contributions and receptor-based $PM_{0.1}$ source contributions calculated using measured concentrations of molecular markers (Yu, Venecek et al. 2018). Regional $PM_{0.1}$ concentrations exceeded 2 μg m^{-3} during summer pollution episodes in major urban regions across the US including Los Angeles, the San Francisco Bay Area, Houston, Miami, and New York. $PM_{0.1}$ spatial gradients were sharper than $PM_{2.5}$ spatial gradients due to the dominance of primary 420 aerosol in $PM_{0.1}$. This finding suggests that $PM_{0.1}$ measurement networks needed to support epidemiology must be denser than comparable $PM_{2.5}$ measurement networks. Non-residential natural gas combustion was identified as a major source of $PM_{0.1}$ across all major cities in the United States. On-road gasoline and diesel vehicles made significant contributions to regional $PM_{0.1}$ in all 39 cities even though peak contributions within 0.3 km of the roadway were not 425 resolved by the 4 km grid cells. This is consistent with other studies that have found an exponential decrease in ultrafine particle concentrations outside of major roadways (Wang, Hopke et al. 2011). Food cooking also made significant contributions to $PM_{0.1}$ in all cities but biomass combustion was only important in locations impacted by summer wildfires. Aviation was a significant source of $PM_{0.1}$ in cities that had airports within their urban footprints. The 430 major sources of primary $PM_{0.1}$ and $PM_{2.5}$ were notably different in many cities. Future epidemiological studies may be able to differentiate $PM_{0.1}$ and $PM_{2.5}$ health effects by contrasting cities with different ratios of $PM_{0.1}$ / $PM_{2.5}$ sources.

Data Availability: All of the $PM_{0.1}$ and PNC outdoor exposure fields produced in the current 435 study are available free of charge at http://faculty.engineering.ucdavis.edu/kleeman/.

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