Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-832
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 12 October 2018

Atmospheric Chemistry and Physics Discussions

Sources of Airborne Ultrafine Particle Number and Mass Concentrations in California

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Abstract

Regional concentrations and source contributions are calculated for airborne particle number concentration (PNC) and ultrafine particle mass concentration ($PM_{0.1}$) in the San Francisco Bay Area (SFBA) and the South Coast Air Basin (SoCAB) surrounding Los Angeles with 4 km spatial resolution and daily time resolution for selected months in the years 2012, 2015, and 2016. Performance statistics for daily predictions of PNC concentrations meet the threshold normally required for regulatory modeling of $PM_{2.5}$ (MFB< ± 0.5 and MFE < 0.75). Predicted source contributions to $PM_{0.1}$ are in good agreement with results from receptor-based studies that use molecular markers for source apportionment at four locations in California. Different sources dominated regional concentrations of PNC and $PM_{0.1}$ because of the different emitted particle size distributions and different choices for heating fuels. Non-residential natural gas combustion (38-74%) made the largest single contribution to PNC concentrations at the ten regional monitoring locations, followed by nucleation (6-14%), wood smoke (1-8%), food cooking (1-9%), and mobile sources (4-8%). In contrast, wood smoke (25-49%) was the largest source of $PM_{0.1}$ in the SFBA followed by mobile sources (15-33%), non-residential natural gas combustion (13-28%), and food cooking (4%-14%). Non-residential natural gas combustion (42-57%) was the largest $PM_{0.1}$ source at the SoCAB sites, followed by traffic sources (16-35%) and food cooking (6-14%). The study region

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encompassed in this project is home to more than 25M residents, which should provide sufficient power for future epidemiological studies on the health effects of airborne ultrafine particles. Correlations between $PM_{2.5}$ and PNC are low (R^2=0.35) suggesting that the health effects of these metrics may be assessed independently. All of the $PM_{0.1}$ and PNC outdoor exposure fields produced in the current study are available free of charge at http://webwolf.engr.ucdavis.edu/data/soa_v2/monthly_avg2.

1. Introduction

Numerous epidemiological studies have identified positive correlations between exposure to ambient particulate matter (PM) and increased risk of respiratory and cardiovascular diseases, premature mortality and hospitalization (Pope et al., 2002;Pope et al., 2004;Pope et al., 2009;Dockery and Stone, 2007;Ostro et al., 2015;Ostro et al., 2006;Ostro et al., 2010;Brunekreef and Forsberg, 2005;Fann et al., 2012;Gauderman et al., 2015;Miller et al., 2007). Most of these studies have not fully addressed ultrafine particles (UFPs; Dp<0.1µm) because these particles make a very small contribution to total ambient PM mass (Ogulei et al., 2007). Toxicity studies suggest that UFPs may be especially dangerous to human health since they have higher toxicity per unit mass (Li et al., 2003;Nel et al., 2006;Oberdorster et al., 2002) and can penatrate the lungs and enter the bloodstream and secondary organs (Sioutas et al., 2005). These toxicology results are suggestive but more epidemiological evidence is required before the threat to public health from UFPs can be fully assessed.

Most previous UFP epidemiology studies are based on particle number concentration (PNC) measured at fixed sites using commercially-available instruments. These devices are expensive and they require regular maintence which limits the number of measurement sites that can be deployed. Translating measured PNC into population exposure estimates is also difficult because UFP concentrations change more rapidly over shorter distances than $PM_{2.5}$ (Hu et al., 2014b;Hu et al., 2015;Hu et al., 2014a). Land use regression (LUR) models could potentially be used to interpolate UFP concentrations between sparse measurement locations, but the atmospheric processes governing PNC

concentrations are highly non-linear and (so far) sufficient training data is not generally available for LUR models to estimate PNC exposure over a large enough population to support a definitive epidemiology study (Montagne et al., 2015). Previous attempts to use regional reactive chemical transport models to predict PNC in highly populated regions have focused on nucleation, yieldeding a wide range of predicted concentrations and only modest agreement with measurements when nucleation algorithms were not standardized (Elleman and Covert, 2009b;Zhang et al., 2010;Elleman and Covert, 2009a). Obtaining accurate exposure estimates to PNC in highly populated regions therefore remains a major challenge in UFP epidemiological studies.

Recent work has examined UFP mass ($PM_{0.1}$) as an alternative metric for UFP exposure, and demonstrated that $PM_{0.1}$ can be predicted with reasonable accuracy over large populations using regional reactive chemical transport models (Hu et al., 2014b;Hu et al., 2014a). The $PM_{0.1}$ exposure fields developed using this technique have been used in multiple epidemiological studies that revealed associations with mortality and pre-term birth (Ostro et al., 2015;Laurent et al., 2016). Despite the success of studies using $PM_{0.1}$, techniques that estimate PNC exposure are still needed because a large number of ongoing UFP studies are based on PNC and it is possible that $PM_{0.1}$ and PNC are associated with different types of health effects.

Here we extend the previous work using regional reactive chemical transport models for UFPs to include PNC in the San Francisco Bay Area (SFBA) and the South Coast Air Basin (SoCAB) region around Los Angels which are the two most densely populated major metropolitan location in California. Source contributions to $PM_{0.1}$ and PNC are tracked using the University of California, Davis / California Institute of Technology (UCD/CIT) regional reactive chemical transport model with 4 km spatial resolution. Predicted concentrations during the year 2012 are compared to measurements available at 10 regional monitoring sites. The spatial distribution fields of different particle metrics (PNC, $PM_{0.1}$, $PM_{2.5}$) are combined with population distributions to estimate exposure. To the best of our knowledge, this is the first integrated study of both UFP number and mass using a regional reactive chemical transport model in California.

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2. Model Description

The UCD/CIT chemical transport model used in the current study has been succesfully applied in sevaral previous studies in the San Joaquin Valley (SJV) and the SoCAB (Ying et al., 2008b;Ying et al., 2008a;Hu et al., 2015;Hu et al., 2017;Chen et al., 2010;Held et al., 2004;Held et al., 2005;Hixson et al., 2010;Hixson et al., 2012;Hu et al., 2012;Kleeman and Cass, 2001;Kleeman et al., 2007;Kleeman et al., 1997;Mahmud, 2010;Mysliwiec and Kleeman, 2002;Rasmussen et al., 2013;Ying and Kleeman, 2006;Zhang and Ying, 2010). It includes algorithms for emissions, transport, dry deposition, wet deposition, gas phase chemistry, gas-to-particle conversion, coagulation, and some condensed phase chemical reactions. Nucleation was added to the model for the first time in the current study using the ternary nucleation (TN) mechanism involving H_2SO_4-H_2O-ammonia (NH_3) (Napari et al., 2002). As was the case in previous studies using this algorithm, the resulting nucleation rate was adjusted using a tunable nucleation parameter set to 10^{-5} for new particle nucleation (Jung et al., 2010). The Kerminen and Kulmala (2002) parameterization was added in order to bridge the gap between the 1 nm particle nuclei and their appearance into the smallest size bin of the UCD/CIT model (~10 nm). Several previous modeling studies have been conducted to evaluate the performance of the ternary nucleation mechanism on predicted particle number concentration (PNC) using global and regional models. Jung et al. (2010) found that a scaled version of the ternary H_2SO_4-NH_3-H_2O nucleation theory (Napari et al.,2002 with a supplemental 10^{-5} nucleation tuning factor) added to the PMCAMx-UF model produced PNC predictions in reasonable agreement with observations. The study of Westervelt et al. (2013) also showed that the ternary nucleation parameterization (with a supplemental 10^{-5} nucleation tuning factor) added to the Goddard Earth Observing System global chemical transport model (GEOS-Chem) produced reasonable PNC predictions on average when compared with measurements at five locations spanning various environments. Jung et al. (2008) considered multiple nucleation parameterizations in the Dynamic Model for Aerosol Nucleation (DMAN) to predict the nucleation events and non-events observed during the Pittsburgh Pittsburgh Air Quality Study (PAQS) conducted between July 2001 and September 2002. Their results showed that the ternary nucleation mechanism ((Napari et al., 2002) with a supplemental 10^{-5} nucleation tuning

factor) was a suitable nucleation scheme for 3-D chemical transport models. Although there have been numerous significant efforts to incorporate nucleation algorithms into three-dimensional regional and global models (Jung et al., 2008;Jung et al., 2010;Westervelt et al., 2013;Zhang et al., 2010), nucleation modeling studies are still in the early stages of development and further efforts are needed to reduce the uncertainty in both the nucleation rate and growth mechanisms. In the current study, emission, transport, deposition, and coagulation of UFPs were simulated using operators developed for the model framework, leading to modification of the particle size distribution and the subsequent PNC concentrations. Dynamic condensation / evaporation is not considered for nucleation mode particles because these processes act slowly on the regional scale relative to the other operators and they do not strongly alter the ground-level PNC outside the near-roadway environment (within 300 m or major highways). This 0.3km resolution is beyond the scope of the current modeling exercidse which uses 4 km spatial resolution in the horizontal direction. Furthermore, the dynamic condensation / evaporation calculations are too expensive to track for nucleation mode particles in regional model applications. The model configuration in the current study reflects the focus on regional UFP concentrations, not near-roadway UFP concentrations.

The model domains used in the study are shown in Figure 1. The parent domain with 24 km horizontal resolution covered the entire state of California (referred to as CA_24 km) and the two nested domains with 4 km horizontal resolution covered the SFBA + SJV + South Sacramento Valley air basins (referred as SJV_4 km) and the SoCAB surrounding Los Angeles (referred as SoCAB_4 km). The UCD/CIT model was configured with 16 vertical layers up to a height of 5 km above ground level, with 10 layers in the first 1 km. Previous studies have shown that this vertical configuration captures the air pollution system above California (Hu et al., 2014a;Hu et al., 2014b;Hu et al., 2015). Particulate number, mass, and composition are represented in 15 size bins, with particle diameters being centered within equally spaced logarithmic size interval spanning the diameter range from 0.01 to 10μm.

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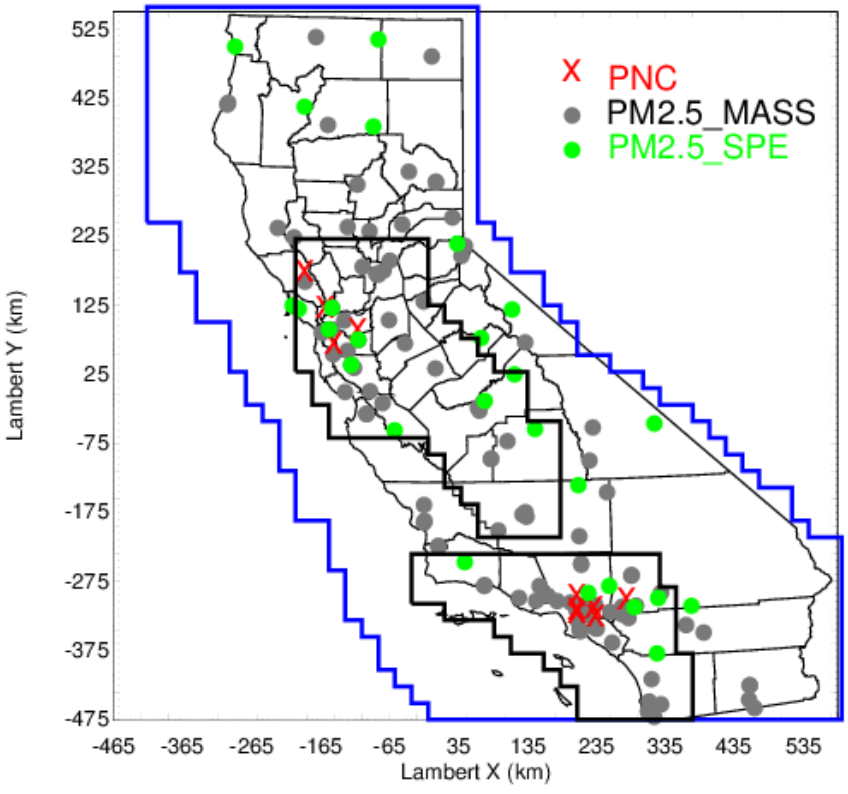

Figure 1: Modeling domains. Blue lines outline the CA_24 km domain, black lines outline the SoCAB_4 km (bottom) and SJV_4 km domains (top). Red crosses represent ten particle number concentration (PNC) sites (fours sites operated by staff at the Bay Area Air Quality Management District (BAAQMD) and six sites from the Multiple Air Toxics Exposure Study IV (MATES IV)). Detailed location information for the PNC sites is listed in Table S3. Green dots represent BAAQMD $PM_{2.5}$ speciation network sites and the Interagency Monitoring of Protected Visual Environments (IMPROVE) sites; gray dots represent the $PM_{2.5}$ federal reference method (FRM) sites.

2.1 Meteorological Fields

Hourly meteorological fields during the modeling period were generated by the Weather Research and Forecasting (WRF) model version 3.4 with three nested domains that had horizontal resolutions of 36 km, 12 km and 4 km, respectively. In the present simulations, the WRF model was configured with 50 vertical layers (up to 100 hpa) and four-

dimensional data assimilation (FDDA) nudging was utilized to improve the agreement between model predictions and observed meteorological patterns (Otte, 2008b, a). WRF predictions for wind speed, temperature, and relative humidity were compared to measurements for seven counties in the SFBA and two counties in SoCAB (see Table S2). Temperature has mean bias (MB) within ~0.2 ˚C and root- mean-square errors (RMSE) between 4-5 ˚C. Wind speed has mean fraction bias (MFB) within ±0.20 and RMSE generally <2.0 m/s. This level of performance is consistent with performance of WRF in previous studies conducted in California (Zhao et al., 2011;Hu et al., 2015).

2.2 Emissions

The emission inventories used in the SFBA were developed by the BAAQMD for the year 2012 based on the regulatory inventory provided by the California Air Resources Board for that same year. The SFBA inventory was processed using the Sparse Matrix Operator Kernel Emissions (SMOKE) v3.7 software package provided by US EPA. SMOKE was configured to separately tag emissions from on-road gasoline vehicles, off-road gasoline vehicles, on-road diesel vehicles, off-road diesel vehicles, food cooking, biomass burning, non-residential natural gas, and all other sources. The emission inventories used in South Sacramento Valley, SJV and SoCAB were provided by the California Air Resources Board.

Measurements conducted in parallel with the current study found that particles emitted from natural gas combustion in home appliances were semi-volatile when diluted by a factor of 25 in clean air, but particles emitted from industrial sources did not evaporate under the same conditions (Xue et al., 2018a). Near-field emissions from residential natural gas sources were therefore set to zero in the current study while emissions from other natural gas combustion sources were retained at their nominal levels. SMOKE results were transformed into size-resolved emissions of particle number, mass, and composition using measured source profiles through an updated version of the emissions model described by Kleeman and Cass (1998). The PM profiles used for each source type were specified as weighted averages from each of the detailed sources within each broad category as summarized in Table S1. Detailed PM source profiles for major sources of

ultrafine particulate matter are based on measurements conducted during source tests (Li and Hopke, 1993;Kleeman et al., 1999, 2000;Robert et al., 2007a;Robert et al., 2007b;Mazaheri et al., 2009). In most cases, these emissions size distributions strongly influence the size distributions of particles in the ambient atmosphere (see Figures S1 and S3). A more detailed discussion of the emissions processing has been presented in a previous study (Hu et al., 2015).

3. Results

3.1 Statistical Evaluation

According to Taylor's Hypothesis (Shet et al., 2017), it is expected that the spatial distribution of model results is more important than the temporal distribution when evaluating performance. In the current study model performance evaluations are limited to the locations where measurements were made. Therefore, the temporal distribution is also considered by comparing predicted vs. measured daily average PNC, $PM_{2.5}$ and individual $PM_{2.5}$ species mass concentrations. The evaluation data set was compiled from several measurement networks including the sites operated by staff at the Bay Area Air Quality Management District (BAAQMD), the IMPROVE sites, the MATES IV sites and FRM sites. Model performance for $PM_{2.5}$ at routine monitoring sites (Figure 1) generally meets the performance criteria suggest by Boylan and Russell (Boylan and Russell, 2006) (mean fractional error (MFE) ≤ +0.75 and mean fractional bias (MFB) ≤ ±0.5) (Table S4). Table S5 shows the MFB and MFE values of gaseous species of O_3, NO, NO_2, CO and SO_2 using daily averages across all measurement sites during the entire simulated period. Gaseous species of O_3, CO, NO, NO_2 and SO_2 have MFBs within ± 0.3 and MFE less than 0.5, indicating general agreement between predictions and measurement for these species. The ability of UCD/CIT predictions for key gas species, mass and chemical component concentrations in the $PM_{0.1}$ and $PM_{2.5}$ size fractions was also evaluated in previous studies (Ying and Kleeman, 2006;Ying et al., 2008a;Ying et al., 2008b;Hu et al., 2012;Chen et al., 2010;Held et al., 2005;Hu et al., 2015;Hu et al., 2017;Venecek et al., 2018). The performance of the UCD/CIT air quality model in these studies generally meets standard model performance criteria. Of greatest interest in the current study,

predicted PNC values were compared to measured $N_{7\text{-}1000}$ (aerosol number concentrations for particle diameters ranging from 7nm-1000nm) values at four sites in the SFBA (Santa Rosa, San Pablo, Redwood City and Livermore) and six sites in SoCAB (Anaheim, Central Los Angeles, Compton, Huntington, Inland-valley and Rubidoux). $N_{7\text{-}1000}$ measurements in the SFBA were made using an Environmental Particle Counter (EPC) Monitor Model 3783 (TSI Inc) while $N_{7\text{-}1000}$ measurements in the SoCAB were made with EPC Model 3781 (TSI Inc). Both monitors can detect ultrafine particles down to 7 nm which is smaller than the first size bin of 10 nm used in model calculations. Previous studies conducted at Fresno, California, suggest that $N_{7\text{-}10}$ accounts for approximately 8% of $N_{7\text{-}1000}$ (Watson et al., 2011), and so some amount of negative bias is expected when comparing predicted PNC (=$N_{10\text{-}1000}$) to measured $N_{7\text{-}1000}$. The evaluation results for PNC summarized in Table 1 follow this expected trend but mean fractional bias (MFB) and mean fractional error (MFE) at each comparison site still meet the performance criteria suggest by Boylan and Russell (2006). This level of performance is comparable to the results from a previous UFP number simulation conducted in Northern California using a modified version of the WRF-Chem model (Lupascu et al., 2015). The good agreement between predicted and measured $PM_{2.5}$, individual $PM_{2.5}$ species, key gas species and PNC builds confidence in the model skill for UFP predictions in the current study.

Table 1 Performance statistics for PNC (=$N_{10\text{-}1000}$) predictions vs. N_7 at individual monitoring sites. Threshold for regulatory PM modeling applications is typically MFB< ± 0.5 and MFE < 0.75.

Location	MFB	MFE	RMSE Particles cm^{-3}
Livermore	0.02	0.25	2831
Redwood city	-0.21	0.37	5141
San Pablo	-0.38	0.51	5392
Santa Rosa	-0.19	0.39	2634
Anaheim	0.39	0.43	7834

Location	MFB	MFE	RMSE Particles cm^{-3}
Central LA	0.11	0.23	5505
Compton	0.25	0.31	6485
Huntington	0.19	0.27	9966
Inland-Valley	0.35	0.38	8494
Rubidoux	0.44	0.47	7106

3.2 $PM_{0.1}$ and PNC Source Apportionment in California

The UCD/CIT model explicitly tracks the mass and the number concentration of particles in each size bin, with tracer species used to quantify source contributions to the primary particle mass (but not number) in that bin. Tracer emissions are empirically set to be 1 % of the total primary particle mass emitted from each source category. This minor addition does not significantly change the particle radius. Tracers are carried through all major aerosol processes including advection, diffusion, coagulation, and deposition. The final tracer concentrations are therefore directly proportional to the primary particle mass from the associated source group. More details describing the source apportionment technique in UCD/CIT model are provided in the previous studies (Ying et al., 2008b;Hu et al., 2017). In the current study, the mass contribution from each source was converted to the number contribution from that source according to equation 1:

$$number_i = mass_i / (\pi/6 * Dp^3 * \rho) \qquad (1)$$

where $number_i$ is the number concentration associated with source *i*, $mass_i$ is the mass concentrations associated with source *i*, Dp is the particle diameter and ρ is the particle density (calculated based on particle composition). This approach assumes that the condensation of new mass does not strongly affect the calculated density of the particles (true when condensation is not dominant in the size bins that contain most of the particle number). The accuracy of this assumption was tested by comparing the sum of the

"reconstructed" particle number (eq 1) across all sources to the actual total particle number tracked by the model yielding error <10% in the current study.

3.2.1 UCD/CIT $PM_{0.1}$ source contributions compared to Chemical Mass Balance (CMB) results

A recently completed study measured the composition of $PM_{0.1}$ at four sites in California and calculated source contributions using molecular markers (Xue et al., 2018b). Figures 2 and 3 compare the source contributions to PM0.1 OC concentrations predicted by the UCD/CIT model and the molecular marker technique at San Pablo, East Oakland, downtown Los Angeles and Fresno during a summer month (August 2015) and a winter month (February 2016). The "others" category in the molecular marker calculation represents unresolved sources, while in the UCD/CIT model "others" represents the sum of non-residential natural gas source combustion, aircraft emissions, and the sources that were not tagged in the current study. In general, predicted source contributions to PM0.1 OC from the molecular marker technique and the UCD/CIT model are in good agreement. Natural gas dominates PM0.1 OC in the summer of 2015 at San Pablo, East Oakland, downtown Los Angeles and Fresno, while wood smoke and aircraft are the major sources of PM0.1 OC in Fresno and East Oakland during the winter of 2016. The importance of ultrafine particles from natural gas combustion has not previously been recognized because these particles lack a unique chemical signature, which causes them to be lumped into the "unresolved" category in receptor-based source apportionment studies. The general agreement in the source contributions from the gasoline, diesel, wood burning, meat cooking and other source categories predicted by the UCD/CIT model and the molecular marker technique builds confidence in the accuracy of the UFP source predictions in the current study.

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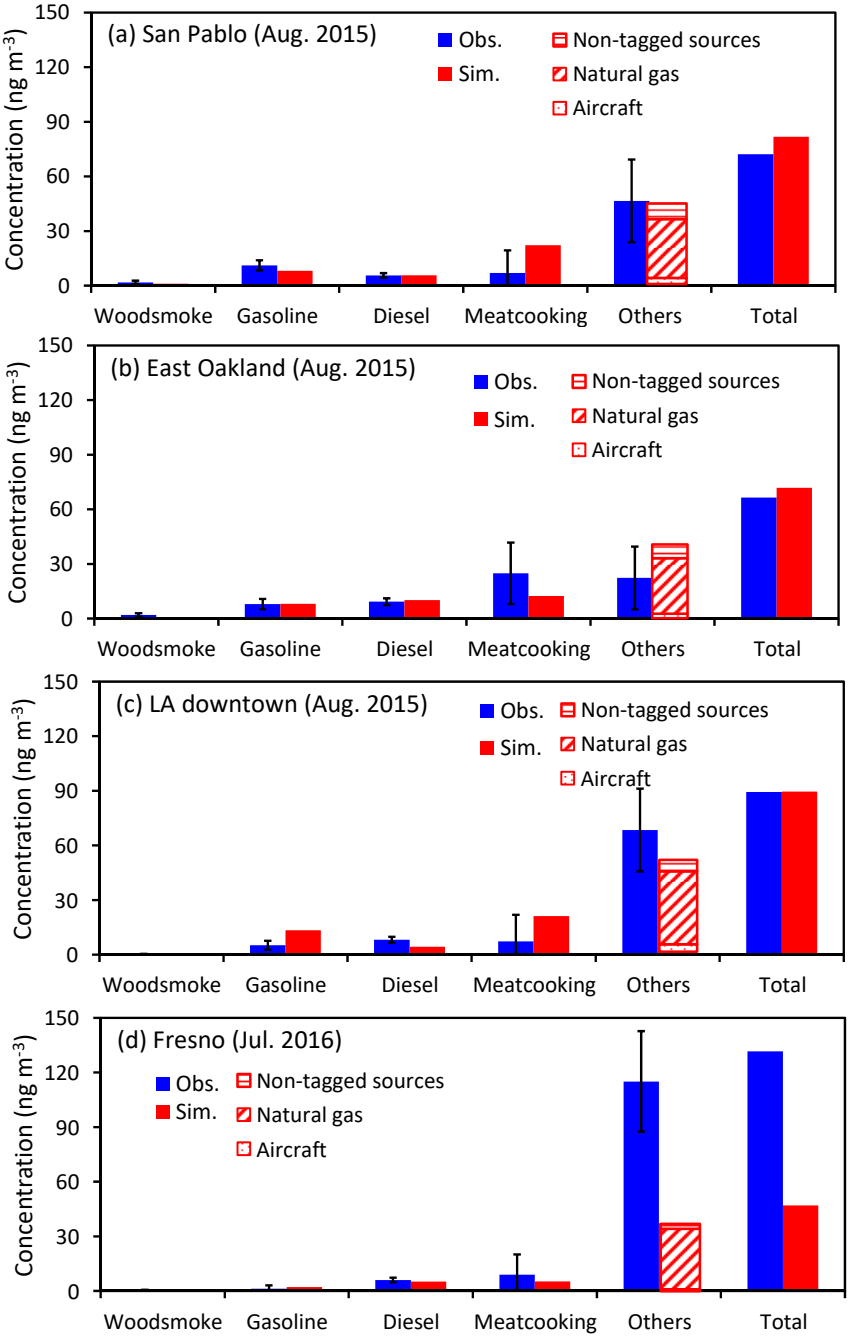

Figure 2: Source contribution to PM0.1 predicted by the CMB receptor model and the UCD/CIT model at four sites in California in August 2015

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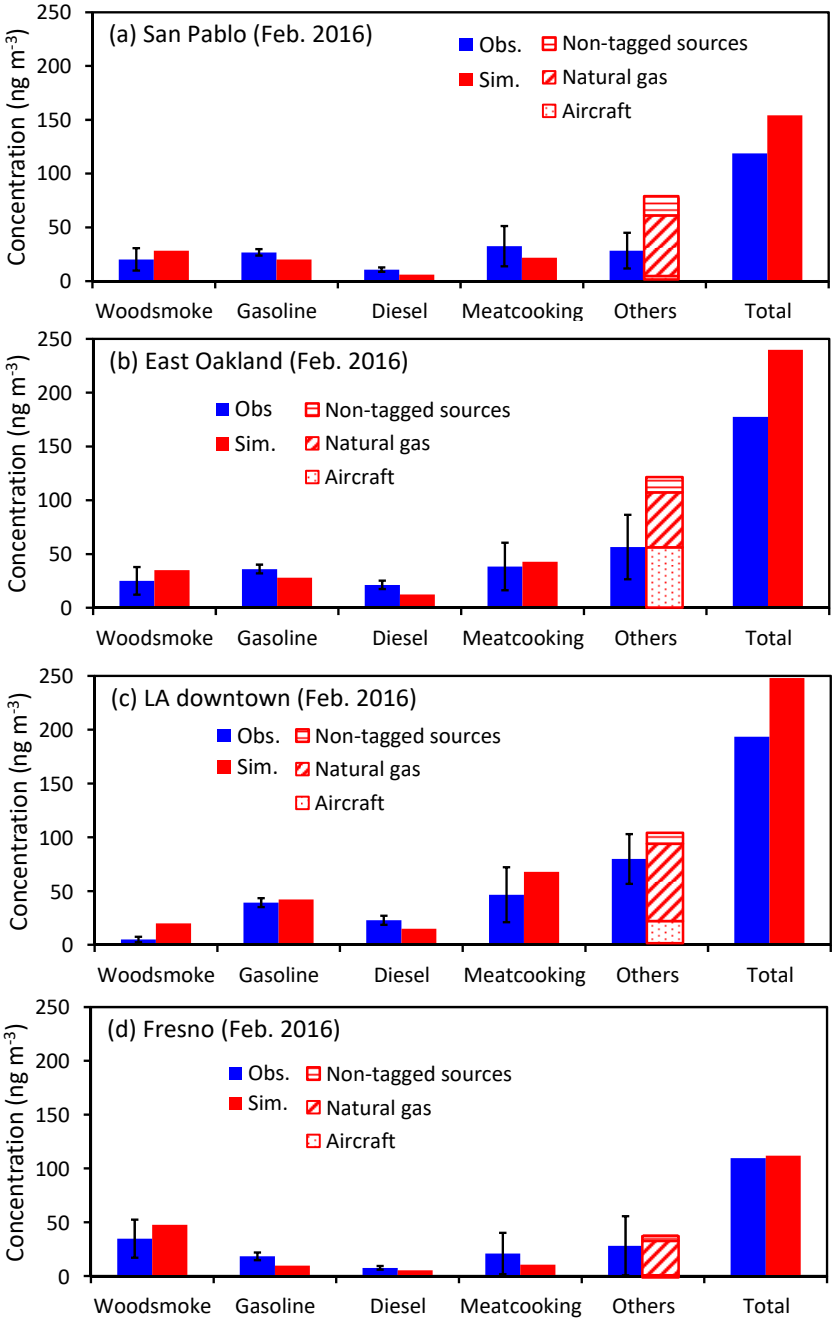

Figure 3: Source contribution to PM0.1 predicted by the CMB receptor model and the UCD/CIT model at four sites in California in February 2016.

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295 **3.2.2 $PM_{0.1}$ and $N_{10\text{-}1000}$ Source contributions in California**

Figures 4-6 and 7-9 show the seasonal variation of major source contributions to primary PNC and $PM_{0.1}$, respectively. The black circles in Figure 4-6 represent the measured $N_{7\text{-}1000}$ at four BAAQMD sites in SFBA and six MATES sites in Los Angeles and Riverside counties. Predicted PNC agrees reasonably well with measured seasonal variations of $N_{7\text{-}1000}$ at San Pablo, Redwood City, Livermore, Anaheim, Central LA, Compton and Huntington. The model under predicts $N_{7\text{-}1000}$ at Santa Rosa and over predicts $N_{7\text{-}1000}$ at Inland-Valley and Rubidoux but overall model performance statistics for PNC are within the guidelines for regulatory $PM_{2.5}$ applications (see Table 1). Nucleation contributes to summer PNC at all sites but makes negligible contributions to $PM_{0.1}$ concentrations. Traffic sources including gasoline- and diesel-powered vehicles make significant contributions to $PM_{0.1}$ concentrations at each measurement site depending on proximity to major freeways. Near-roadway effects on ultrafine particle concentrations are not apparent since these locations were chosen to be regional monitors and so they are more than 300 m from the nearest freeway. Predicted contributions from traffic sources are consistent with the molecular marker results illustrated in Figures 2-3. Traffic contributions to regional PNC concentrations more than 300 m away from roadways are even smaller than $PM_{0.1}$ contributions because the size distribution of particles emitted from motor vehicles peaks at 100 – 200 nm (Robert et al., 2007a;Robert et al., 2007b). Wood smoke makes strong contributions to regional $PM_{0.1}$ concentrations in central California during winter but much smaller contributions in the SoCAB because wood burning is not typically used for home heating in this region. Wood burning contributions PNC are less dominant in central California because the size distribution of particles emitted from wood combustion peaks at 100-300 nm (Kleeman et al., 2008b). The largest source of PNC in central California and PNC+$PM_{0.1}$ in the SoCAB is non-residential natural gas combustion. Industrial processes and power generation that use natural gas do not follow strong seasonal cycles and so the strength of the natural gas source contributions is somewhat constant across seasons subject to variability caused by meteorological conditions.

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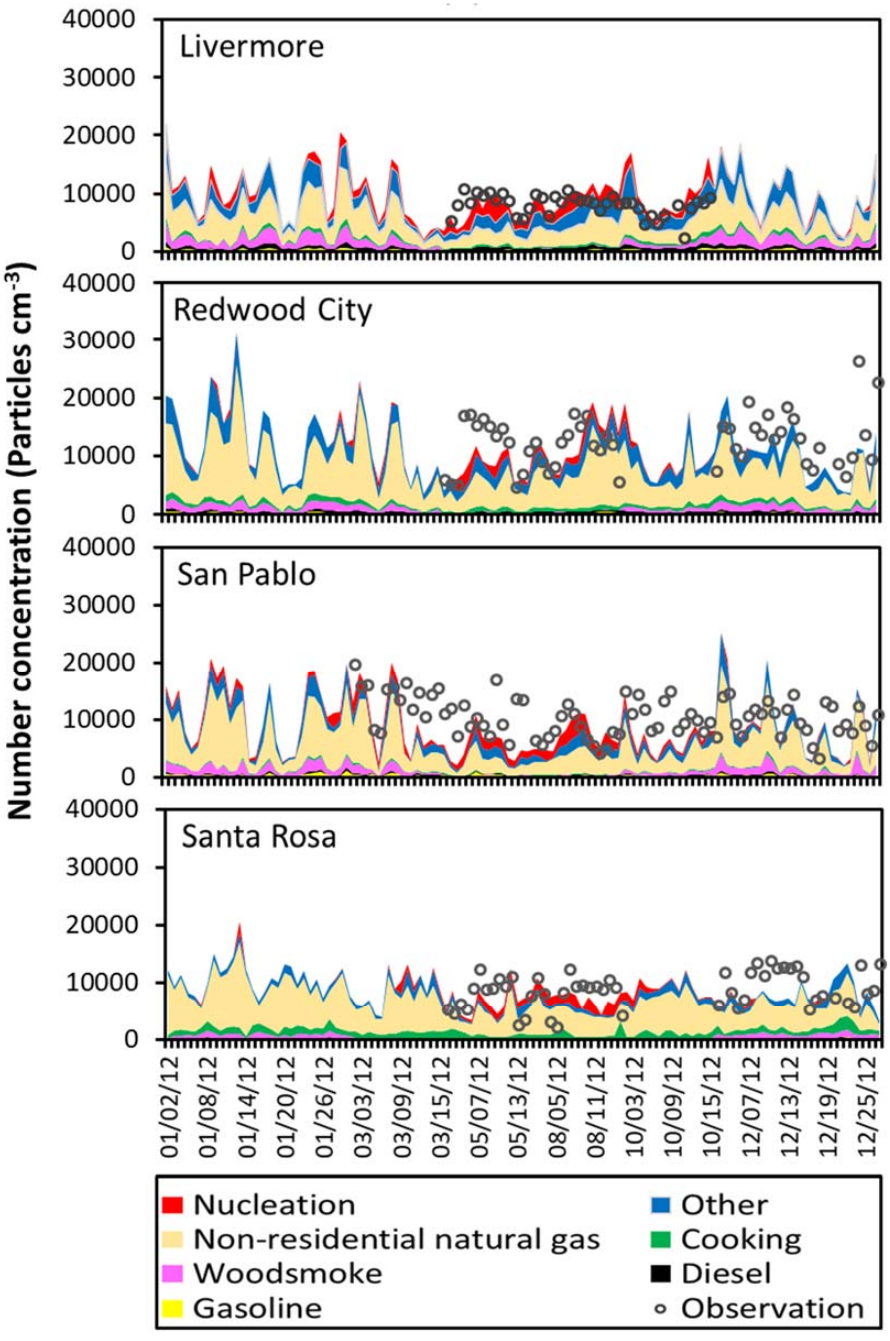

325 Figure 4: Seasonal variation of measured $N_{7\text{-}1000}$ (black circles) and major source contributions to PNC at Livermore, Redwood City, San Pablo and Santa Rosa, respectively.

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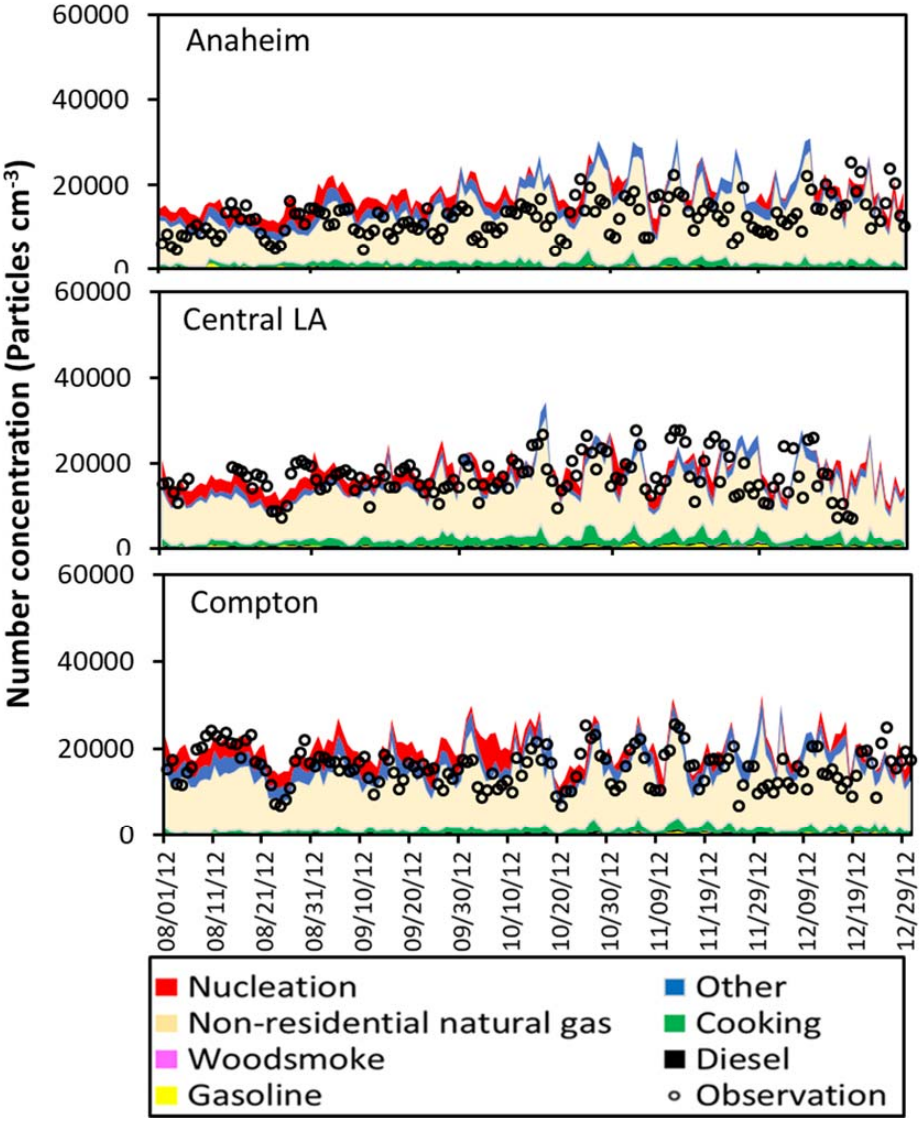

Figure 5: Seasonal variation of measured $N_{7\text{-}1000}$ (black circles) and major source contributions to PNC at Anaheim, Central LA, and Compton, respectively.

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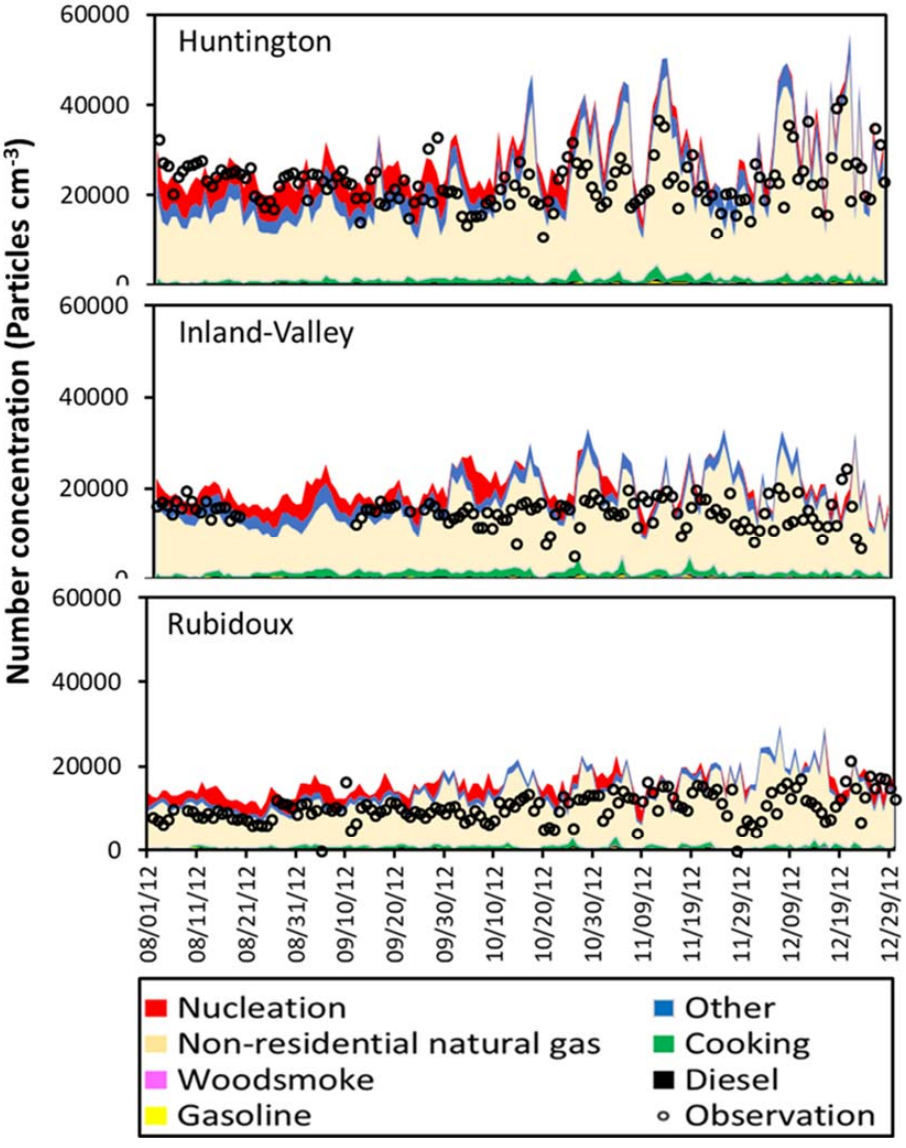

Figure 6: Seasonal variation of measured $N_{7\text{-}1000}$ (black circles) and major source contributions to PNC at Huntington, Inland-Valley, and Rubidoux, respectively.

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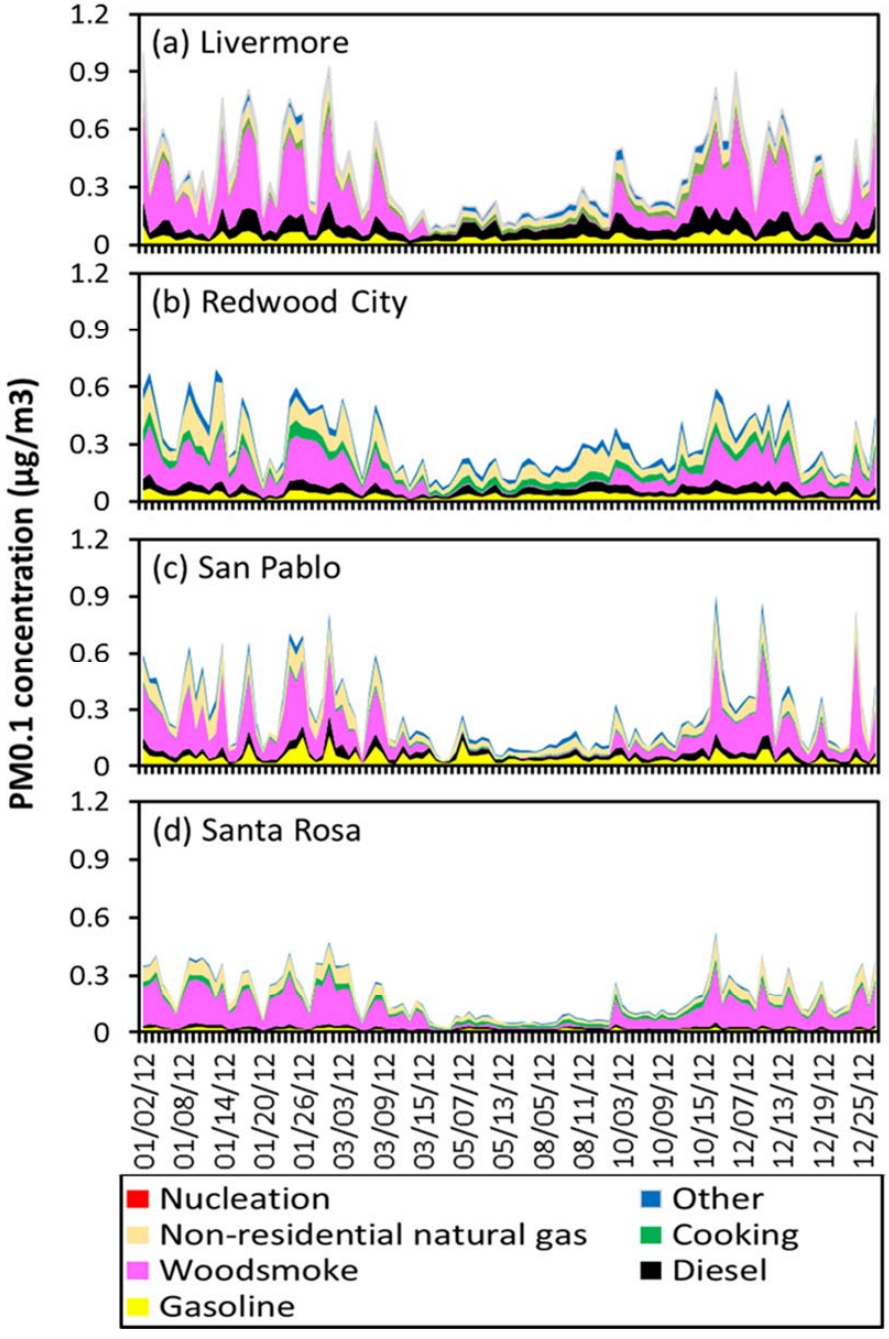

Figure 7: Seasonal variation of major source contributions to $PM_{0.1}$ at Livermore, Redwood City, San Pablo and Santa Rosa, respectively.

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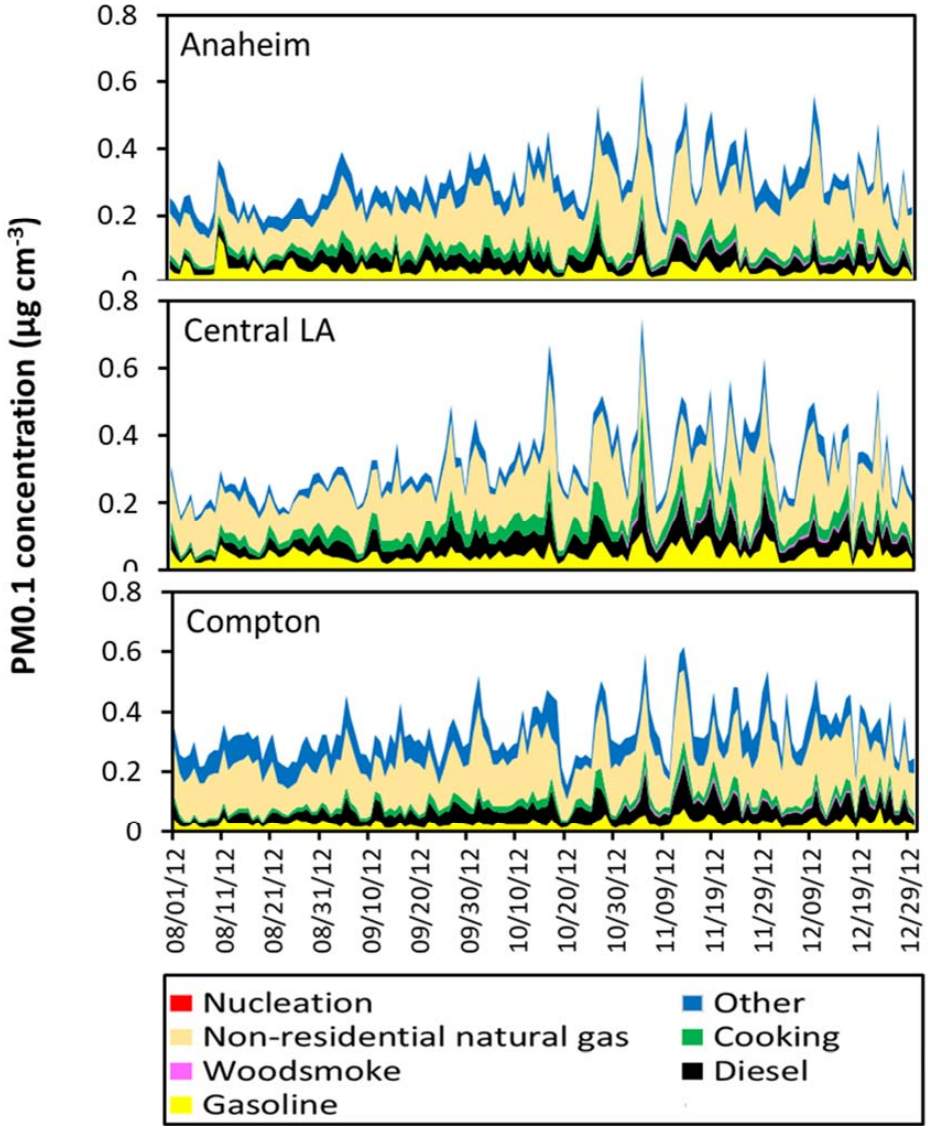

340 Figure 8: Seasonal variation of major source contributions to $PM_{0.1}$ at Anaheim, Central LA, and Compton, respectively.

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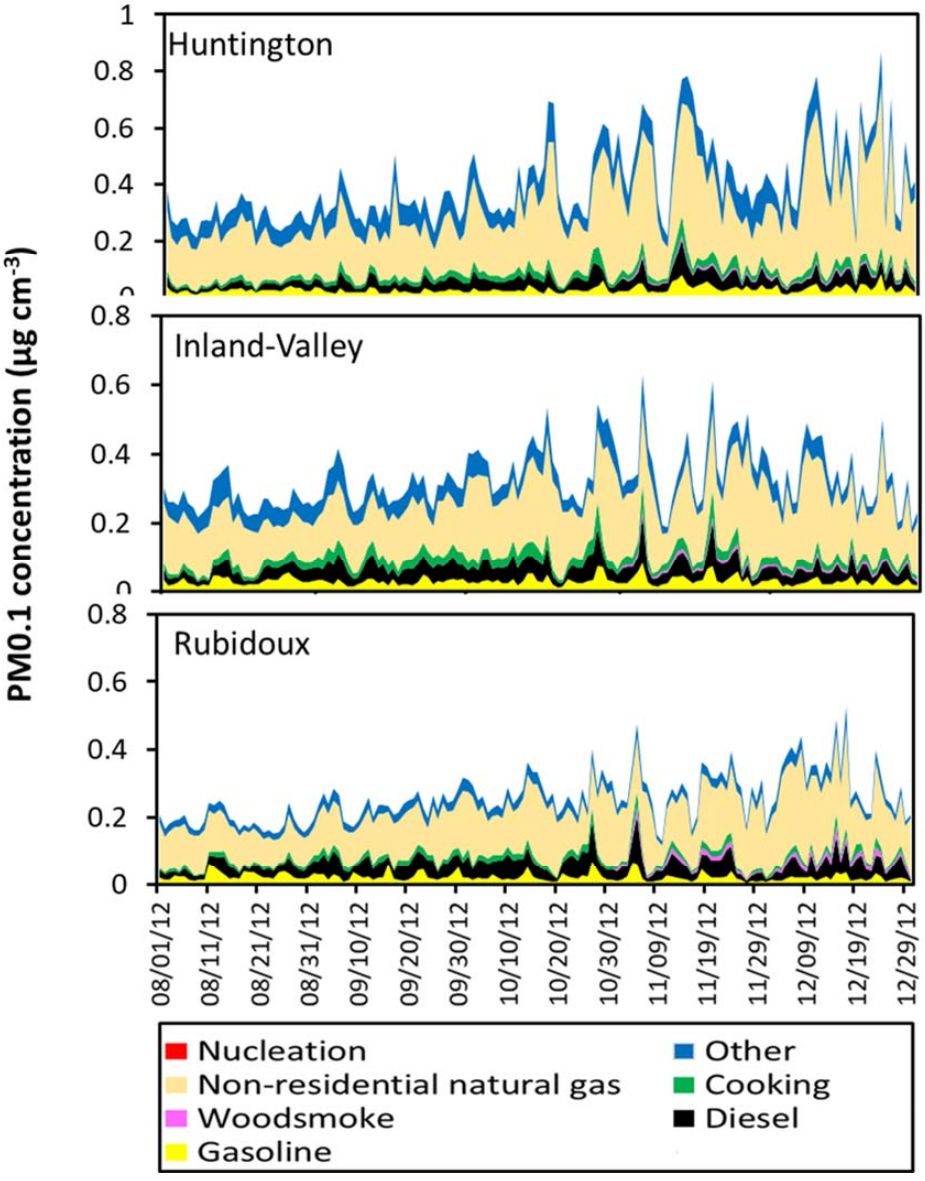

345 Figure 9: Seasonal variation of major source contributions to PM0.1 at Huntington, Inland-Valley, and Rubidoux, respectively.

Figures 10 and Figures 11 show the source contributions to PNC and $PM_{0.1}$, respectively, averaged over the days shown in Figures 4-9. Non-residential natural gas combustion makes the largest predicted contribution to PNC at all the sites that were evaluated. Traditional sources that were tracked including meat cooking, wood smoke, and mobile (gasoline + diesel) accounted for approximately 10-20% of the predicted PNC at the sites selected for study. "Other" sources that were not tagged explicitly in the current study accounted for 8-28% of PNC across these sites. Nucleation is a significant source for of PNC for both BAAQMD sites and MATES sites where sulfur emissions were highest, with contributions ranging from 6-14%.

The dominant PNC contribution from non-residential natural gas combustion reflects the emitted particle size distribution combined with the ubiquitous use of this fuel in the SFBA and SoCAB regions. The chemical composition and size distribution information for non-residential natural gas combustion emissions used in this study was measured by Hildemann (1991) and Li and Hopke (1993), respectively. Size distributions and volatility were further confirmed during on-going field studies conducted by the current authors (Xue et al., 2018a). The estimated non-residential natural gas combustion particle number and mass size distributions are shown in Figure S1 (left column). Clearly, the majority of particles from non-residential natural gas combustion are typically found in diameters <0.05 μm, while particles emitted from other sources such as wood combustion tend to have slightly larger particle diameter (with lower number concentration per unit of emitted mass).

Figures 11 show that wood smoke is the largest $PM_{0.1}$ source at Livermore (36%), San Pablo (35%), while non-residential natural gas combustion still makes the largest contribution to $PM_{0.1}$ at Redwood City (28%), Santa Rosa (41%) and MATES sites (42%-58%) in the SoCAB region. Contributions from cooking and mobile sources are enhanced in $PM_{0.1}$ vs. PNC, with the cooking source accounting for 15% of $PM_{0.1}$ at Santa Rosa and mobile sources (gasoline + diesel) accounting for 34% of $PM_{0.1}$ at the Central LA site, followed by 33% of $PM_{0.1}$ at Livermore site. The different rankings of source contributions to PNC and $PM_{0.1}$ can be explained by the comparison of particle number-size distribution and particle mass-size distribution for the non-residential natural

gas and wood burning sources at four evaluated sites (Figure S1). Particles emitted from non-residential natural gas combustion and wood burning have number distributions that peak at particle diameters of 0.016-0.025 μm and 0.025-0.04 μm, respectively. Non-residential natural gas combustion and wood burning mass distributions, however, peak at particle diameters of 0.025-0.04 μm and 0.10-0.16 μm, respectively.

Figure 12-14 show diurnal variations of measured $N_{7\text{-}1000}$ and predicted PNC averaged over days in August and December 2012. Measured $N_{7\text{-}1000}$ diurnal patterns in August are bimodal with the first peak usually occurring at 6-7 am at four sites in SFBA and 5-6 am at six sites in Los Angeles and Riverside County and the second peak occurring between 12-3 pm. The first summer peak corresponds to morning activities including cooking and traffic "rush hours", while the second peak appears to be related to nucleation events. The predicted PNC diurnal variations in August were in good agreement with measurements at five out of ten sites (Livermore, San Pablo, Anaheim, Compton, and Huntington). The model generally predicts a biomodal diurnal profile with maximum values in reasonable agreement with measurements at these locations. The model failed to capture the mid-day nucleation event at Santa Rosa possibly due to missing SO_2 sources in the emissions inventory upwind from this site. The model overestimated mid-day peak values at Inland-valley and Rubidoux sites. In December, the measured $N_{7\text{-}1000}$ diurnal pattern was also bimodal with the first peak around 7:00-8:00am and the second peak in the evening at around 8pm. This pattern reflects both the emissions activity and the mixing status of the atmosphere throughout the day. The predicted PNC concentration is in good agreement with measurements for the early morning peak but generally underestimated the evening peak possibly due to excess atmospheric mixing after sunset in the model calculations. Nucleation appears to play a small role during winter. Non-residential natural gas combustion is predicted to be the largest source of PNC during morning and evening peaks. The diurnal profiles of non-residential natural gas emissions are included in supplemental information (Figure S2). Industrial natural gas combustion emissions peak during the daytime with lower values at night. Emissions from electricity generation powered by natural gas peak in the morning and evening. Commercial natural gas combustion emissions may either peak in the morning and evening or they may follow a uniform diurnal profile depending on the specific source and location.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-832
Manuscript under review for journal Atmos. Chem. Phys.
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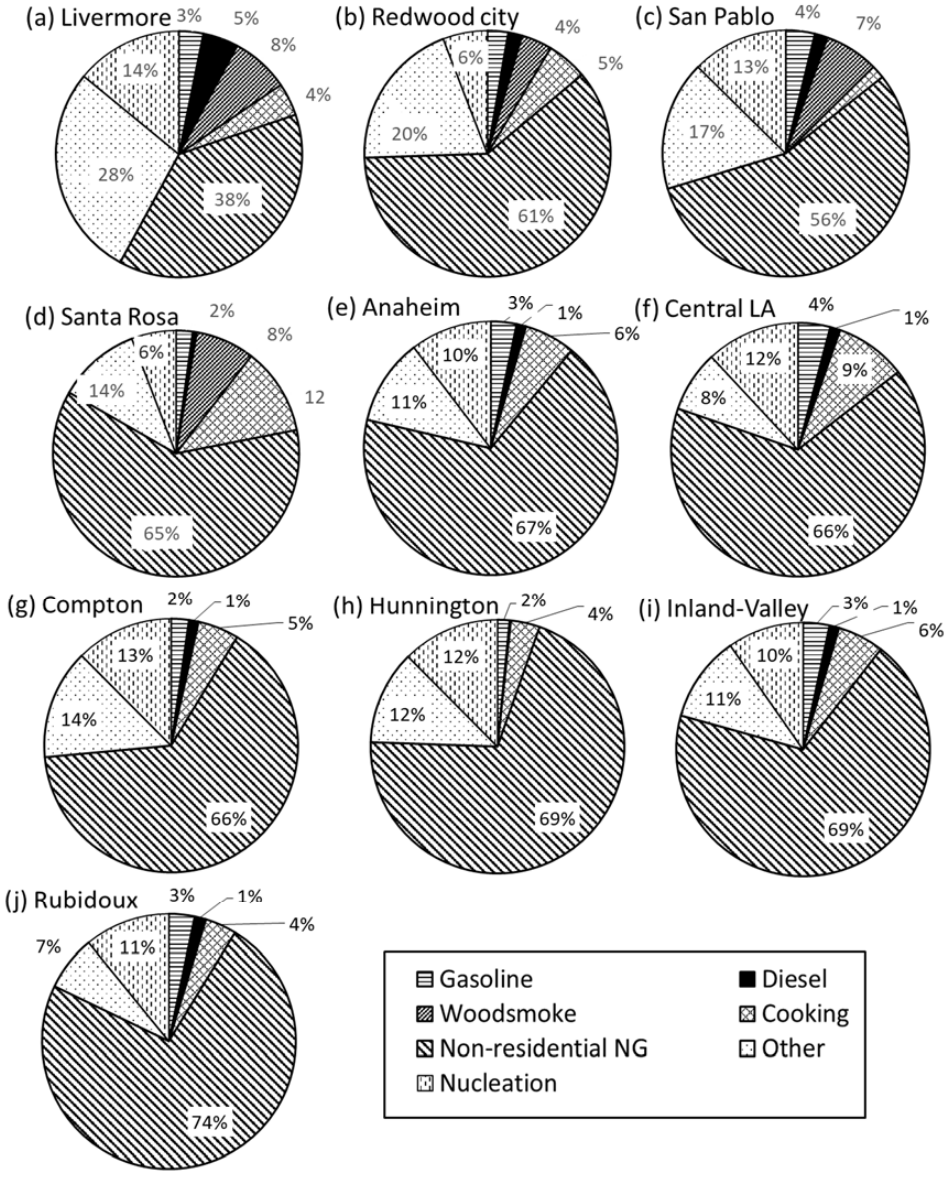

410 Figure 10: The relative source contributions to PNC at Livermore, Redwood City, San Pablo, Santa Rosa, Anaheim, Central LA, Compton, Huntington, Inland-Valley and Rubidoux, respectively. Averaging time included all days shown in Figures 4-6.

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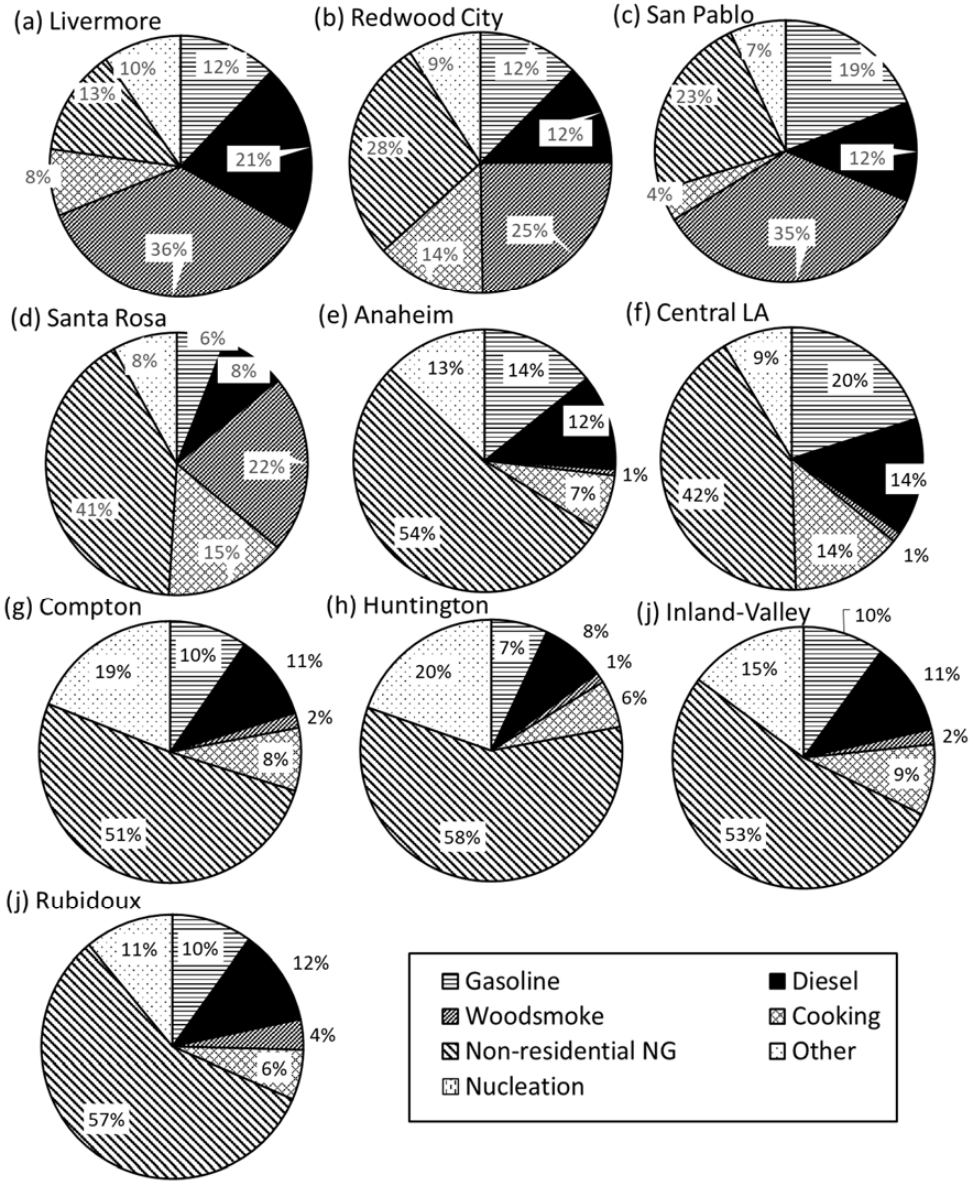

Figure 11: The relative source contributions to $PM_{0.1}$ seasonally averaged at Livermore, Redwood City, San Pablo and Santa Rosa, Anaheim, Central LA, Compton, Huntington, Inland-Valley and Rubidoux, respectively. Averaging time included all days shown in Figures 7-9.

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Manuscript under review for journal Atmos. Chem. Phys.
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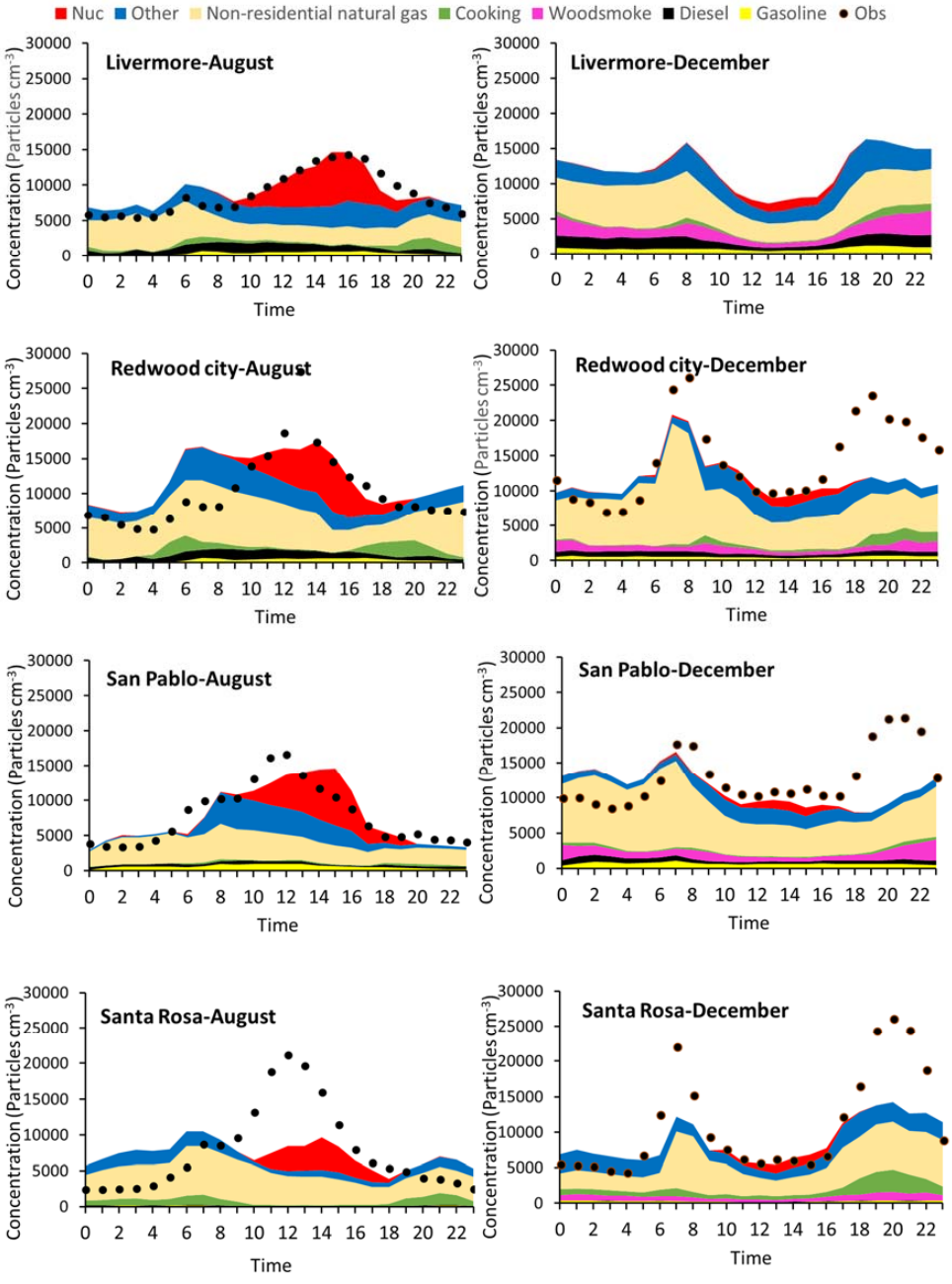

Figure 12: Diurnal variations of measured $N_{7\text{-}1000}$ and predicted PNC averaged for August 2012 (left column) and December 2012 (right column) at Livermore, Redwood city, San Pablo and Santa Rosa.

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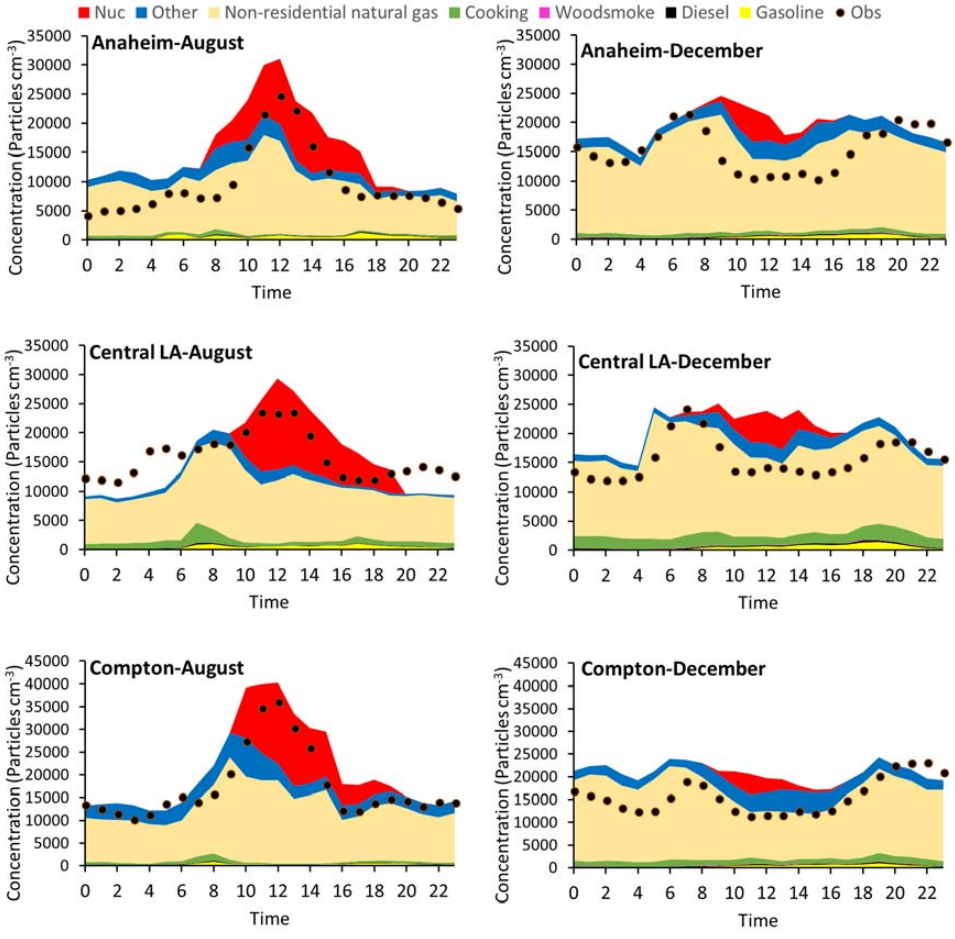

Figure 13: Diurnal variations of measured $N_{7\text{-}1000}$ and predicted PNC averaged for August 2012 (left column) and December 2012 (right column) at Anaheim, Central LA, and Compton.

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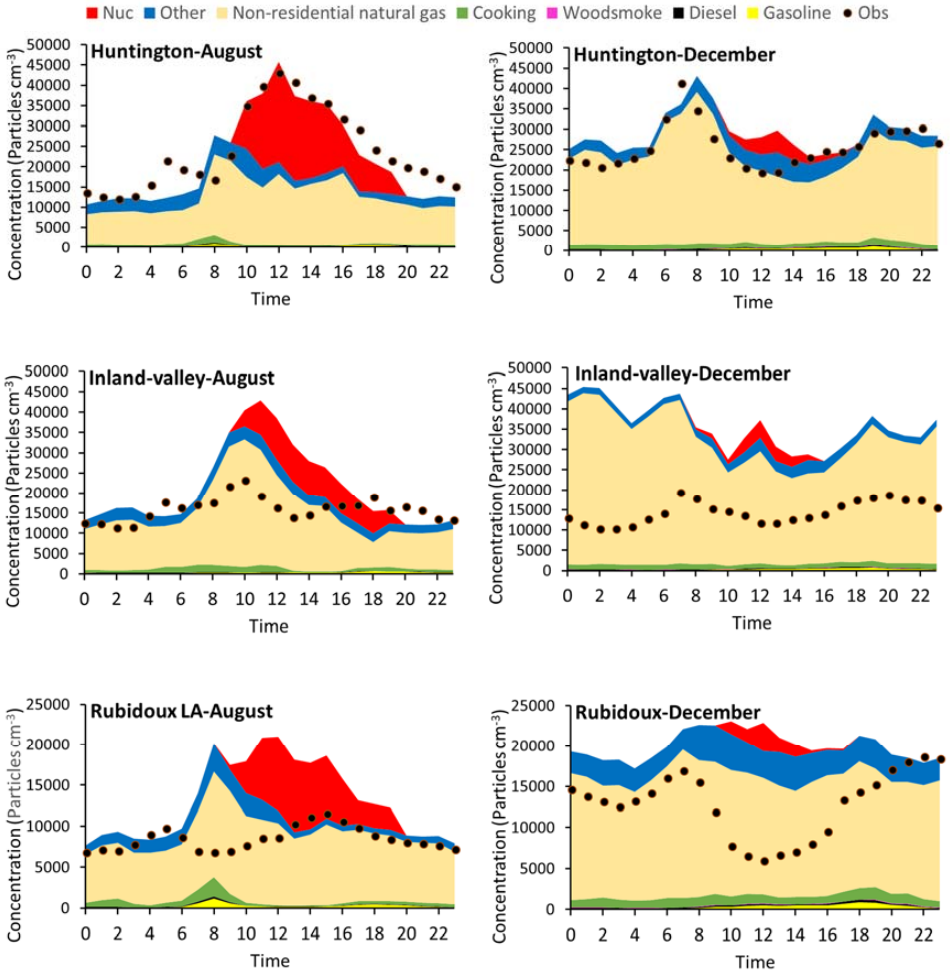

Figure 14: Diurnal variations of measured $N_{7\text{-}1000}$ and predicted PNC averaged for August 2012 (left column) and December 2012 (right column) at Anaheim, Central LA, and Compton.

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Manuscript under review for journal Atmos. Chem. Phys.
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4. Discussion

Previous researchers have used Positive Matrix Factorization (PMF) to calculate source contributions to PNC (Sowlat et al., 2016;Morawska et al., 2008;Gu et al., 2011;Ogulei et al., 2007;Kasumba et al., 2009;Wang et al., 2013;Yue et al., 2008;Friend et al., 2013). The dominant factors resolved by these studies have been traffic, urban background, secondary aerosol, wood burning and nucleation (Sowlat et al., 2016;Morawska et al., 2008;Gu et al., 2011;Ogulei et al., 2007;Kasumba et al., 2009;Wang et al., 2013;Yue et al., 2008;Friend et al., 2013). Particles from natural gas combustion were not separately identified by PMF because they do not contain a unique chemical tracer. It is very likely that natural gas combustion particles are artificially lumped into another source (e.g. traffic) or part of the "urban background" signal identified in previous studies. Natural gas combustion is used extensively in California for electric power, industrial, commercial and residential use (Table S6), and so it seems plausible that this source contributes to ambient UFP concentrations.

The current UFP predictions rely on source profile measurements for wood burning, food cooking, mobile sources, and non-residential natural gas combustion (Cooper, 1989;Harley et al., 1992;Hildemann et al., 1991a;Hildemann et al., 1991b;Houck and L. C., 1989;Kleeman et al., 2008a;Kleeman et al., 2000;Robert et al., 2007b;Robert et al., 2007a;Schauer et al., 1999a, b, 2001, 2002a, b;Taback, 1979). All of these size distributions were measured using appropriate instruments and methods by knowledgeable researchers, but some of these past studies were conducted more than a decade ago. Size distribution information for vehicles, natural gas, etc. have been added to the supplemental information (Figure S3). Changes in fuel composition and emissions control technology in the interim years may have altered the emitted size distributions. New measurements of particle size distributions emitted from natural gas and biomethane combustion were made in parallel with the current project to confirm the source profile measurements from past studies (Xue et al., 2018a). The results of these measurements are consistent with previous size distribution results.

California has tighter air pollution standards than many other regions in the United States due to the severe air quality problems that have historically occurred in the state.

California therefore has a unique mixture of fuels and emissions control technology that may affect the mixture of sources that contribute to atmospheric ultrafine particle concentrations. Venecek et al. (2018) recently used the UCD/CIT air quality model with the 2011 National Emissions inventory to calculate source contributions to $PM_{0.1}$ in 39 major cities across the United States during peak summer photochemical smog episodes in the year 2010. The findings from this study show that natural gas combustion is a major source of ultrafine particles in the regional atmosphere over urban areas across the United States. The public health questions associated with ultrafine particles emitted by natural gas combustion have wide-ranging implications. Similar levels of ultrafine particle concentrations will likely occur in other regions across the world that extensively use natural gas as a fuel source, although other sources of ultrafine particles may also make strong contributions depending on the total mix of fuels in each region.

Recent theories suggest that primary particulate matter composed of semi-volatile organic compounds may evaporate after release to the atmosphere, which may reduce ambient PNC. Measurements conducted in parallel with the current study confirmed that particles emitted from natural gas combustion in home appliances partially evaporated when diluted by a factor of 25 in clean air, but particles emitted from industrial sources did not evaporate under the same conditions (Xue et al., 2018a). Future work should verify the accuracy of the size and composition distributions for all natural gas combustion sources given their apparent importance for predicted PNC.

Evidence from both toxicology and epidemiology will be required to assess the effect of UFPs on public health. It is essential to identify and quantify UFP sources based on both mass ($PM_{0.1}$) and number PNC during this process (Friend et al., 2013). An accurate comparison of both $PM_{0.1}$ and PNC exposure could lay the groundwork for specific assessment of health effects of UFPs and potentially more efficient control strategies for PM emission from major sources (Yue et al., 2008). Ideally, spatial exposure patterns for PNC, $PM_{0.1}$, and $PM_{2.5}$ will be sufficiently unique to separate their individual effects in epidemiological studies. Regression statistics for different metrics were calculated by using all grid cells in the model domain of the current study. The correlations between the various particle metrics were: $R^2(PM_{2.5}$ vs. PNC)=0.35, $R^2(PM_{2.5}$ vs. $PM_{0.1})$=0.63,

R^2($PM_{0.1}$ vs. PNC)=0.75. It seems likely that future epidemiological studies will be able to differentiate between the effects of $PM_{2.5}$ and PNC based on the low R^2 value. The potential for comparisons between $PM_{2.5}$ and $PM_{0.1}$ is less clear cut, but previous work helps understand what may be possible. Ostro et al. (2015) compared the associations between IHD mortality and $PM_{2.5}$ vs. $PM_{0.1}$ in the California Teachers Study (CTS) cohort. Associations between IHD mortality and the sum of $PM_{2.5}$ mass (p-value=0.001) were stronger than associations between IHD mortality and the sum of $PM_{0.1}$ mass (p-value=0.01) but individual components of mass (EC, OC, Cu, etc) all had stronger associations with IHD mortality in the $PM_{0.1}$ size fraction than the $PM_{2.5}$ size fraction.

The current study focuses on outdoor exposure to UFPs that may be useful in future epidemiological studies. Indoor or in-vehicle exposure to UFPs can also be significant (Wallace and Ott, 2011;Rim et al., 2010;Bhangar et al., 2011;Weichenthal et al., 2015;Fruin et al., 2008) but characterizing these micro-environments is beyond the scope of the current manuscript.

5. Conclusions

The UCD/CIT regional chemical transport model has been updated with a nucleation algorithm and combined with the existing size-resolved source profiles of particlualte matter emissions to predict regional source contributions to airborne particle number concentration (PNC) and airborne particulate ultrafine mass ($PM_{0.1}$). Predicted 24-hour average PNC is in good agreement with measured $N_{7\text{-}1000}$ at ten sites across California in summer (Aug) and winter (Dec). Predicted diurnal variation of PNC is in reasonable agreement with measured concentrations but uner-predicts early evening peaks in the winter due to the failure of meteorological calculations to capture the suppressed mixing in the atmosphere at these times. Predicted $PM_{0.1}$ source contributions are in good agreement with $PM_{0.1}$ source contributions measured in a molecular marker study at four sites across California in summer (Aug) and winter (Dec) months. Natural gas combustion is the largest source of regional PNC at all locations outside of the immediate vicinity of other major combustion sources. Nucleation contributed to particle number during the summer months at midday but did not dominate PNC concentrations.

Likewise, traffic sources contributed to PNC but did not dominate over regions more than 300 m away from freeways. Combustion sources such as wood burning, food cooking, and mobile sources made stronger contributions to $PM_{0.1}$ at heavily urbanized locations. Wood burning for home heating had strong seasonal patterns with peak concentrations in winter while other sources contributed more consistently throughout the seasons. Nucleation made a negligible contribution to $PM_{0.1}$ over the urban areas at the focus of the current study.

The current study identifies natural gas combustion as a major source of ultrafine particle number and mass concentrations in urban regions throughout California. The health implications of these natural gas combustion particles should be investigated in future epidemiology studies.

Data Availability: All of the $PM_{0.1}$ and PNC outdoor exposure fields produced in the current study are available free of charge at http://faculty.engineering.ucdavis.edu/kleeman/ which provides a link to the most recent version of the dataset (currently http://webwolf.engr.ucdavis.edu/data/soa_v2/monthly_avg2). Model source code and model input files are available to collaborators via direct email to the corresponding author at mjkleeman@ucdavis.edu.

Acknowledgements: This research was supported by the Bay Area Air Quality Management District under project #2013.218, the Coordinating Research Council under project #A-96, and the California Air Resources Board under project #14-314. None of the project sponsors nor any person acting on their behalf: (1) makes any warranty, express or implied, with respect to the use of any information, apparatus, method, or process disclosed in this report, or (2) assumes any liabilities with respect to use, or damages resulting from the use or inability to use, any information, apparatus, method, or process disclosed in this report.

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