

Modeling Air Quality during the California Regional PM₁₀/PM_{2.5} Air Quality Study (CPRAQS) using the UCD/CIT Source Oriented Air Quality Model - Part II. Regional Source Apportionment of Primary Airborne Particulate Matter

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Abstract

A comprehensive air quality modeling project was carried out to simulate regional source contributions to primary airborne particle concentrations in California's central Valley. A three week stagnation episode lasting from December 15, 2000 to January 7, 2001, was chosen for study using the air quality and meteorological data collected during the California Regional PM₁₀/PM_{2.5} Air Quality Study (CRPAQS). The UCD/CIT source oriented air quality model was applied to this episode using both the source-oriented external mixture configuration and an internal mixture with artificial tracers so that source contribution information could be retrieved in less time.

The majority of the predicted and measured primary airborne particulate matter mass was composed of elemental carbon (EC) and organic carbon (OC). Previous work has shown that base case EC and OC predictions made by the UCD/CIT model are in good agreement with observations. Model results from the current study show that the highest EC and OC concentrations occur in urban areas and along transportation corridors where primary emissions are largest. Lower concentrations of primary EC and OC are predicted at rural locations in the SJV.

Source contributions predicted by the UCD/CIT air quality model were compared to receptor-oriented source apportionment results produced by the Chemical Mass Balance (CMB) model at Fresno and Angiola. The relative contributions from major sources predicted by the UCD/CIT model agree with the CMB model results, building confidence in the accuracy of the UCD/CIT model predictions at locations where the CMB results are not available. Wood smoke was identified as the major regional source of primary OC in airborne particles in the winter SJV episode, accounting for approximately 50% of the total PM_{2.5}. Diesel engines were also found to be a significant contributor to primary PM_{2.5} OC and the largest contributor to the predicted PM_{2.5} EC averaged over a typical day. EC contributions from wood smoke increased at night and sometimes reached as high as 40% of the total PM_{2.5} EC. The contribution of fugitive dust to primary PM_{2.5} mass was also predicted to be significant, especially in rural areas, but this result is likely biased high by the lack of an appropriate diurnal profile for dust emissions.

The results of the current study suggest that reductions in wood burning and diesel engine activity would reduce the regional concentration of primary PM_{2.5} during severe winter stagnation events in the SJV.

1 Introduction

Fresno, Bakersfield and Sacramento rank 2nd, 3rd and 9th in the list of top 25 cities most polluted by airborne particles with diameters smaller than 2.5 μm ($\text{PM}_{2.5}$) (American Lung Association, 2005). These statistics place the San Joaquin Valley (SJV) in central California among the most heavily polluted air basins in the United States. Numerous studies have identified strong correlations between $\text{PM}_{2.5}$ and various respiratory (von Klot et al., 2002; Murr et al., 2004) and cardiovascular (Delfino et al., 1996; de Hartog et al., 2003) symptoms. One set of hypotheses about the mechanistic link between airborne particles and health effects focuses on the source-origin and chemical composition of primary particles (emitted directly from sources). A better understanding of regional source contributions to primary particulate matter in the SJV would help to identify threats to public health and provide the basis for future regulations designed to improve air quality.

The most severe SJV $\text{PM}_{2.5}$ episode in recent history occurred during the California Regional $\text{PM}_{10}/\text{PM}_{2.5}$ Air Quality Study (CRPAQS) in December 2000 and January 2001. Elevated fine PM concentrations occurred throughout the entire SJV over a 3 week period (Chow et al., 2006a). The recorded hourly $\text{PM}_{2.5}$ concentrations exceeded 200 $\mu\text{g m}^{-3}$ in Bakersfield (Herner et al., 2005) (which is approximately 5.7 times the National Ambient Air Quality Standard for 24-hour average $\text{PM}_{2.5}$ concentrations). A database containing measured gaseous and particulate matter concentrations, meteorological measurements, and emissions estimates has been constructed as a part of the CRPAQS study to support an evaluation of the underlying cause for this severe air quality problem and to develop cost-effective emission control strategies.

The purpose of this study is to (1) develop a computationally effective source-oriented air quality model for the simultaneous determination of source contributions to the regional distribution of primary PM mass in multiple size fractions, (2) validate the modeled source contribution to primary PM against the CMB source apportionment calculation based on measured molecular markers and (3) apply the model to study regional source contributions to primary $\text{PM}_{2.5}$ concentrations in California's central Valley during CRPAQS.

2 Background

Receptor-oriented statistical models are the traditional tools used in air quality studies to identify source contributions to PM concentrations. These models are based on the principle of mass conservation for non-reactive chemical components in the emitted particles (Watson et al., 2002). The Chemical Mass Balance (CMB) model requires emissions profiles from each major source category to determine the source contributions to primary particle concentrations (Watson et al., 2001). The source resolution of CMB models is usually limited by the co-linearity of the profiles. For example, diesel and gasoline engine exhaust particles are hard to separate because the emission characteristics of the two sources are very similar (Watson et al., 1994). In the past decade, various source-specific organic tracers have been identified from different emission source categories (Schauer et al., 1996; Fraser and Lakshmanan, 2000). The application of the organic molecular markers can greatly improve the resolution and reduce the uncertainties associated with statistical source apportionment methods (Chow et al., 2006b).

Statistical source apportionment techniques have been applied in the past to determine the source contributions to PM at receptor sites in central California for primary PM_{10} (Chow et al.,

1992) and $PM_{2.5}$ (Schauer et al., 1996; Chow et al., 2006b; Chen et al., 2007) concentrations. Residential wood combustion was identified as the major contributor to $PM_{2.5}$ concentrations during wintertime PM episodes in central California, with a valleywide average contribution of 24%. Primary emissions from motor vehicles account for 10-15% of the $PM_{2.5}$ mass (Chen et al., 2007).

Recent research has shown that regional transport of PM precursors and fine PM can also have a significant impact on local air quality at receptor sites (Chow et al., 1996; Solomon and Magliano, 1999; Ying and Kleeman, 2006; MacDonald et al., 2006). Thus, future emission control plans will need to take a regional approach. However, the receptor-oriented statistical tools can only be applied at locations where detailed and accurate PM chemical composition has been measured. The cost of operating a large-scale receptor-oriented study to obtain regional source attribution information is almost prohibitive due to intensive field sampling and laboratory analysis. New tools are needed to efficiently identify the contributions of emission sources to observed PM on a regional scale, so that air quality improvement strategies can consider the impact of surrounding regions on specific non-attainment areas.

3 Model Description

The UCD/CIT source-oriented air quality model tracks particles emitted from different sources separately through the simulated atmosphere in the presence of all major aerosol processes (emissions, transport, deposition, gas-to-particle conversion, coagulation). The UCD/CIT model can also be configured to represent the particles as an internal mixture in which particles emitted from different sources into a single virtual particle class and thus no source information is explicitly retained in the model simulation. In this study, a unique inert artificial tracer, which is empirically set to be 1% of the total mass of the chemical species emitted from each source category, is automatically injected into the model emissions. Variations on this approach have been described in previous studies (Ying and Kleeman, 2004; Bhave et al., 2004; Lane et al., 2007). The 1% injected inert mass does not significantly change the particle radius and the dry deposition rate. The evolution of the tracer species concentrations is explicitly tracked along with other chemical components of the particles. The simulated artificial tracer concentration for a given source directly correlates with the amount of PM mass emitted from that source. After determining the amount of primary mass emitted from each source, an appropriate source-specific emission profile is used to recover the source contribution of each chemical component to the total primary PM mass using the following equation:

$$C_{i,j} = A_{i,j} \times T_i \quad (1)$$

where $C_{i,j}$ represents the concentration of the j^{th} chemical component from the i^{th} particle emission category. A is the source profile matrix so that $A_{i,j}$ represents the mass of the j^{th} chemical species per unit mass of PM emitted from the i^{th} emission source. T_i is the model predicted particle mass concentration for the i^{th} source using artificial tracer mass for that source.

The internal mixture with artificial tracer method greatly reduces the computation time of calculation, and the memory / external storage footprint for the source apportionment of primary particulate matter. The number of model particle species is increased only slightly to track the concentration of the artificial tracers from each source. This allows the efficient source apportionment for a large number of primary particle sources on a regional scale.

The full source-oriented external mixture aerosol representation more accurately simulates the physical/chemical properties of particles (especially their hygroscopicity) compared to the internal mixture approach (Kleeman et al., 1997). The internal mixture particle representation is an approximation made to increase efficiency that may lead to some inaccurate predictions for particle composition and size distributions. It is thus important to verify that the overall concentrations predicted by the internal and external particle representation are similar for each source apportionment exercise using the internal mixture method. Previous studies show little difference in the predicted primary particle concentrations using internal and external mixed particle representations (Ying et al., 2004, 2007). In this study, the source apportionment results for the primary particulate matter using the internal mixture particle representation are compared with the results from a full source-oriented external mixture particle simulation to further validate the internal mixture results. Differences between the internal vs. source-oriented external results reflect the intrinsically different behavior of particles due to compositional changes as well as numerical approximations necessary in the internal mixture simulation.

Both the source-oriented external mixture and internal mixture representations require that the emissions inventory be divided into different source categories with similar source profiles. The source-oriented calculations retain the source separation throughout the entire model simulation providing explicit source apportionment results. The internal mixture approach requires that a representative source profile be specified to transform tracer concentrations into chemical species concentrations at the end of the simulation. This step can introduce approximation error into the calculation when sources that have different source profiles are lumped into the same category tracked by a single artificial tracer. Several approaches can be used to estimate the effective source profiles at a receptor location under these conditions. In this study, the emission profiles for each source category over the entire model domain were averaged to generate the representative category profile. Figure 1 shows the average fraction of elemental carbon (EC) and organic compounds (OC) per unit of primary mass emitted from fugitive dust, road dust, diesel engines, catalyst-equipped gasoline engines, non-catalyst-equipped gasoline engines, wood smoke, meat cooking, combustion of high-sulfur fuel, and other sources using this approach. The domain-average source profile may differ from local conditions since emissions within each category are the sum of many sub-categories that each use slightly different emission profiles (example: idling diesel engines and loaded diesel engines have different emissions profiles but they are both averaged into the diesel category). One possible solution to this problem is to calculate average emissions profiles for sub-domains so that greater heterogeneity can be represented. A future study will be conducted to explore this alternative way of estimating emission profiles for internally mixed source apportionment calculations.

4 Model Application

The internally mixed air quality model with artificial tracers was applied to study the source contributions to primary $PM_{2.5}$ mass and chemical composition in the central Valley of California during December 15, 2000 - January 7, 2001. The simulation was carried out using 4 km horizontal grid resolution with 190 x 190 grid cells in a domain that covers the entire central Valley of California (see Figure 1 of Ying et al. (2008b)). The computation domain covers land areas with surface elevation below 2000 meters and ocean regions 100 km off the coastal line. Details about the model

setup and the preparation of the model meteorology, initial and boundary condition fields are described by Ying et al. (2008b) and are not repeated here. The source contributions to secondary PM and total PM_{2.5} and PM_{0.1} mass concentrations are also predicted and the results are documented in a separate paper (Ying et al., 2008a).

Raw gridded emissions of NO_x, SO_x, VOC, NH₃ and PM and the associated EIC (Emission Identification Code) number for the entire modeling episode were provided by the California Air Resources Board (CARB). Emissions were processed further to apply VOC and PM source profiles and to split emissions into different source categories. In the current study, emissions from fugitive dust, road dust, diesel engines, catalyst-equipped gasoline engines, non-catalyst-equipped gasoline engines, wood combustion, food cooking, high-sulfur fuel combustion, and other sources are separated into into different categories based on their EIC number. Table 1 lists the total gas and PM emissions for all the model emission source categories for a typical weekday (December 19, 2000) during the study episode. Based on the totals from the emission inventory, wood smoke accounts for most of the OC emissions while diesel engines account for the majority of the EC emissions.

5 Results

The base case model results have been verified by comparison with measured gas and particle concentrations and the calculation of model performance statistics. Routine measurements were made throughout the December 15, 2000 - January 7, 2001 period with more detailed measurements of particle size and composition made during three Intensive Operating Periods (IOPs) (Dec 15-18, 2000; Dec 26-28, 2000; Jan 4-7, 2001). A detailed discussion of the base case results can be found in a separate manuscript (Ying et al., 2008b). In summary, good agreement for both gas and particle phase pollutants was found at most measurement sites. The model correctly simulated the regional buildup of nitrate concentrations during IOP2 and the subsequently elevated nitrate concentrations during IOP3. The general agreement between the predicted and observed concentrations by the base case model simulation provides a solid foundation for the source apportionment of primary PM in this study.

5.1 Internal Versus External Mixture Source Apportionment

Figure 2 shows the calculated source contribution to 24 hour average EC, OC, and primary PM_{2.5} mass on December 28, 2000 using the source-oriented externally mixed and internally mixed particle representation. In this comparison study, both models are executed using a horizontal grid resolution of 8 km for computational efficiency. Different symbols on the figure indicate different source categories. For each source category, the predicted concentrations at five stations (Bethel Island, Sacramento, Fresno, Angiola and Bakersfield) are shown on the figure. The source contributions predicted by the internally mixed model with artificial tracers agree very well with the source-oriented externally mixed aerosol approach for EC, OC and PM_{2.5} mass concentrations above 1 $\mu\text{g m}^{-3}$. The slope (k) and R^2 values for PM_{2.5}, OC and EC are 1.03(k)/0.987, 1.01/0.996 and 0.88/0.992, respectively. The agreement diverges slightly when the predicted concentrations are lower than 1 $\mu\text{g m}^{-3}$ with slope and R^2 values for PM_{2.5}, OC and EC values 0.956/0.969, 1.17/0.844 and 0.853/0.645, respectively.

5.2 Comparison with CMB Results

The daily-average primary PM source apportionment predicted by UCD/CIT model was compared with the results from an independent CMB source apportionment calculation that resolved road dust, gasoline combustion, diesel combustion, food cooking, and wood burning contributions to PM_{2.5} (Chow, 2005; Chow et al., 2006b).

Figure 3 shows the averaged relative source contributions to primary PM_{2.5} at Angiola (Panel (a)) and Fresno (Panel (b)) during all 3 IOPs. Fresno is the largest urban area in the SJV while Angiola represents a typical rural area in the Valley. The mobile source category shown in Figure 3 represents the sum of the source contributions from diesel and gasoline engines. Dust sources were not included when the relative source contributions were calculated due to large positive bias in the raw PM emission inventory for fugitive dust (see discussion in section 5.3). The UCD/CIT and CMB models have very similar source apportionment predictions at both sites. At Angiola, wood burning was the major source for primary particles, accounting for 45-59% of primary PM_{2.5} mass (excluding dust particles). The contribution from mobile emission sources ranged from 25-37% (excluding dust particles). At Fresno, wood burning was found to be the dominant source for primary particles, accounting for 63 - 75% of primary PM_{2.5} mass (excluding dust particles).

The meat cooking contributions predicted by the UCD/CIT source-oriented model and the CMB receptor-oriented model are in good agreement at Fresno but differ significantly at Angiola. As shown in Figure 3, the relative contribution of meat cooking to primary PM_{2.5} predicted by the UCD/CIT model is less than 5%, while the CMB model predicts more than 30%. Cholesterol is the major organic marker in the meat cooking profile used in the CMB source apportionment calculation. Due to the short sampling durations in the CRPAQS study (5-8 hours per sample) and the low absolute cholesterol concentrations, the measurement error for cholesterol was large and only limited samples were useful (Chow et al., 2006b). This measurement uncertainty could lead to the overestimation of the meat cooking contributions by the CMB approach at Angiola.

The daily averaged source contributions at Fresno during all 3 IOPs from UCD/CIT and CMB models are shown in Figures 4(a) and 4(b), respectively. Wood burning was predicted as the dominant source of primary PM_{2.5} by the UCD/CIT model at all times, and the contribution of meat cooking is consistently below 5 $\mu\text{g m}^{-3}$. CMB predictions also identify wood burning as the main source of primary PM_{2.5} at Fresno, but the contribution from meat cooking was calculated to be greater than 10 $\mu\text{g m}^{-3}$ on some days. Also, CMB predictions show larger day-to-day variation for primary PM_{2.5} source apportionments than the UCD/CIT model. As shown in Figure 4(b), the contribution of wood burning varies from 3 - 35 $\mu\text{g m}^{-3}$. The average of the daily CMB results across all 3 IOPs differs from the CMB results calculated using aggregate measurements, suggesting that the daily CMB results have significant uncertainty. Given the uncertainty in the daily CMB measurements, the level of agreement between daily UCD/CIT and CMB predictions is considered satisfactory.

The results shown in Figure 3 demonstrate that the source contributions to primary PM_{2.5} predicted using UCD/CIT source-oriented air quality model generally agree with the CMB receptor model predictions at a rural and an urban site during the study period. This builds confidence that the UCD/CIT model properly represents the major sources and atmospheric processes of primary PM in this study.

5.3 Primary Source Apportionment at Receptor Sites

Figure 5 shows the predicted hourly-averaged relative source contributions to $PM_{2.5}$ EC, $PM_{2.5}$ OC and $PM_{2.5}$ mass for Fresno during the study period. The EC and OC mass concentrations from each source category are derived from the inert tracer concentrations using the average EC and OC fractions for each source category shown in Figure 1. The relative source contribution was calculated by dividing the predicted EC and OC mass for each source category by the total EC and OC mass concentrations, which are directly predicted by the model. The sum of the mass concentrations do not always total 100% because domain-average emissions profiles used to calculate source contributions during internally mixed simulations do not always capture emissions variability in each sub-region (see discussion in Section 3). The error introduced by this approximation is less than 20% in all cases.

Panel 5(a) shows the calculated source contributions to EC at Fresno during the entire modeling episode are dominated by emissions from diesel engines with smaller contributions from wood smoke. Clear differences in the diurnal variation can be observed for these two EC sources. The wood smoke EC peaks at approximately 20% during the night and falls to approximately 0% during the day when the mixing height increases and wood burning activity for home heating is reduced. The contribution of EC from diesel engines peaks during daytime hours following the general traffic pattern. Panel 5(b) shows that approximately 70-80% of the OC in Fresno comes from wood smoke while approximately 10-15% of the OC originates from meat cooking. Fresno is the largest population center in the SJV so the contribution from meat cooking is quite significant. This prediction agrees with the CMB calculations, as shown in Figure 4. Contributions from gasoline combustion to primary OC are small. The diurnal variation of wood smoke OC peaks during the night (similar to wood smoke EC). Panel 5(c) shows the relative source contribution to total primary $PM_{2.5}$ mass at Fresno. Contributions to secondary PM are not shown here to better illustrate the diurnal variation of the primary PM contribution from each source category. The detailed source contribution to secondary PM components will be shown in a companion paper. The relative contribution of primary PM to total $PM_{2.5}$ varies from 50-80%, with a daily minimum occurring at noon or early afternoon when the primary particles are significantly diluted due to increased mixing height that allows secondary PM formed in the upper atmosphere to reach the surface. Wood smoke accounts for approximately 50% of the total $PM_{2.5}$ mass. Dust particles contribute approximately 10% of the total $PM_{2.5}$ concentrations. The relative contribution from primary PM decreased significantly during December 21-23, 2000 and January 6-7, 2001. Further analysis indicates that the decrease in primary PM during these periods corresponds to two significant inter-region transport events in the SJV. The details of the inter-region transport analysis will be documented in a separate paper.

Figure 6 shows the source contributions to $PM_{2.5}$ EC, $PM_{2.5}$ OC and $PM_{2.5}$ mass concentrations at Angiola during the entire model episode. Panel 6(a) shows that EC originates mainly from diesel engines but does not have clear diurnal variation. Panel 6(b) shows that OC at Angiola is mainly associated with wood smoke that also does not show a clear diurnal variation. The relatively constant source contributions throughout the day most likely indicate that the influence from local sources is small. Panel 6(c) shows the relative source contribution to total $PM_{2.5}$ mass at Angiola. Primary PM accounts for 20-50% of the predicted $PM_{2.5}$ concentration in December 2000 and approximately 20% in January 2001. Fugitive dust particles account for approximately 40% of the predicted $PM_{2.5}$ mass concentration. The high dust contribution predictions are caused by

the high dust emissions in the raw inventory. Recent analysis of the CARB emission inventory found that the fugitive dust $PM_{2.5}/TSP$ ratios were likely overestimated in the CARB PM profiles (Gaffney, 2006). This is likely the cause of the $PM_{2.5}$ mass over-predictions in the base case model simulation at Angiola.

5.4 Regional Source Apportionment of Primary PM

Figure 7 shows the source contribution to 24-hour average $PM_{2.5}$ EC concentrations on December 28, 2000 over the entire computational domain. The concentration scales on each panel are set to best illustrate the regional distribution of each source. Panel 7(a) shows that high total $PM_{2.5}$ EC concentrations occur in the urban areas with maximum concentrations approaching $9 \mu g m^{-3}$. In rural Valley areas, the 24-hour average EC concentrations are approximately $2 \mu g m^{-3}$. Significant outflow from the San Francisco Bay area to the Pacific Ocean can also be seen. Panel 7(c) shows that the 24-hr average contribution of wood smoke to $PM_{2.5}$ EC varies from 0.1 - $0.4 \mu g m^{-3}$, with highest concentration occurring in the populated urban areas in the Valley. Panel 7(d) shows that diesel engines are the dominant EC source in the modeling domain. The highest predicted diesel EC concentrations occur in the San Francisco Bay area, which is the largest urban area in central California. Diesel EC concentrations are also high in other urban areas in the central Valley. Rural EC concentrations from diesel engines are approximately $2 \mu g m^{-3}$, which is likely due to the regional transport of the emissions from Interstate 5 and CA Highway 99 that transect the SJV. Contributions to $PM_{2.5}$ EC from gasoline engines (Panel 7e and f) and meat cooking (Panel 7g) are predicted to be negligible based on the current emissions inventory. Contributions to $PM_{2.5}$ EC from high sulfur fuel combustion (Panel 7h) are significant around the two air force bases in the modeling domain (Travis Air Force Base and Edwards Air Force Base). All other anthropogenic sources contribute approximately $0.5 \mu g m^{-3}$ of EC in the Valley with some higher concentrations of $2 \mu g m^{-3}$ in the northern end of the Valley.

Figure 8 shows the predicted source contributions to 24-hour average $PM_{2.5}$ OC concentrations on December 28, 2000. Panel 8(a) shows that the high total OC concentrations occur in the urban areas with maximum concentrations approaching $55 \mu g m^{-3}$. In rural areas the 24-hour average $PM_{2.5}$ OC concentrations are less than $10 \mu g m^{-3}$. The difference between urban vs. rural concentrations is greater for OC than EC. This is due to the fact that most of the EC originates from on-road diesel engines and significant EC emissions from the CA Highway 99 and Interstate 5 can be transported to nearby rural locations in a relatively short amount of time. In contrast, OC is mainly emitted from residential wood combustion and meat cooking in urban areas. The $PM_{2.5}$ OC concentrations in the rural areas are thus much lower than the concentrations near the urban centers where most of the OC is emitted. Panel 8(b) shows that the predicted regional $PM_{2.5}$ OC contribution from fugitive dust is rather uniform in the Valley with highest concentrations in regions between Fresno and Angiola. Road dust contributions are largest in the urban areas. Both fugitive and road dust make a small contribution to the total $PM_{2.5}$ OC. Panel 8(d) shows that wood smoke is the single largest source of $PM_{2.5}$ OC in urban areas. Emissions of $PM_{2.5}$ OC from diesel and gasoline engines (Panel 8e,f and g) are lower and account for less than 10% of the OC in the domain. The contribution of meat cooking to total $PM_{2.5}$ OC concentrations is highest in the San Francisco Bay area and in Fresno with reduced contributions at Sacramento. This is likely due to the higher wind speed in Sacramento area that dilutes primary emissions. The high sulfur fuel combustion contribution to total $PM_{2.5}$ OC is approximately $2.5 \mu g m^{-3}$ around the two air force

bases in the modeling region. Other sources contribute 2-9 $\mu g m^{-3}$ of $PM_{2.5}$ OC in the Valley.

6 Conclusions

The internally mixed source-oriented air quality model with artificial tracers developed in this study efficiently determines the source contributions to primary particulate matter during the wintertime California Regional $PM_{10}/PM_{2.5}$ Air Quality Study. The internal mixture with artificial tracer method was validated by comparison to a simulation using the full source-oriented external mixture particle representation. The predicted source attribution to primary $PM_{2.5}$ was validated by comparison to CMB results at Fresno and Angiola. The relative contributions from source-oriented model predictions show good agreement with the CMB results for major source categories.

Wood smoke is the major source of primary $PM_{2.5}$ OC in airborne particles in the SJV during a severe winter stagnation event. Daily-average $PM_{2.5}$ EC concentrations are dominated by diesel engines. Diesel engines were also a significant contributor to primary $PM_{2.5}$ OC. Primary PM contributions from wood smoke increase at night, accounting for a maximum of 40% and 90% of the total $PM_{2.5}$ EC and OC, respectively. Sharp gradients of PM concentrations were predicted around major urban areas.

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References

- American Lung Association. State of the air 2005. Technical report, 2005.
- P. V. Bhave, G. A. Pouliot, and M. Zheng. Source apportionment of primary carbonaceous aerosol using the community multiscale air quality model. In *Air Pollution Modeling and Its Application XVI*. Kluwer Academic/Plenum Publishers, New York, 2004.
- L. W. A. Chen, J. G. Watson, and J. C. Chow. Quantifying $\text{pm}_{2.5}$ source contributions for the san joaquin valley with multivariate receptor models. *Environmental Science & Technology*, 41(8): 2818–2826, 2007.
- J. Chow. California regional PM₁₀/PM_{2.5} air quality study (crpaqs) initial data analysis of field program measurement. project report to arb. Technical report, Desert Research Institute, 2005.
- J. C. Chow, J. G. Watson, D. H. Lowenthal, P. A. Solomon, K. L. Magliano, S. D. Ziman, and L. W. Richards. Pm₁₀ source apportionment in California San-Joaquin valley. *Atmospheric Environment Part a-General Topics*, 26(18):3335–3354, 1992.
- J. C. Chow, J. G. Watson, Z. Q. Lu, D. H. Lowenthal, C. A. Frazier, P. A. Solomon, R. H. Thuilier, and K. Magliano. Descriptive analysis of pm(2.5) and pm(10) at regionally representative locations during sjvaqs/auspex. *Atmospheric Environment*, 30(12):2079–2112, 1996.
- J. C. Chow, L. W. A. Chen, J. G. Watson, D. H. Lowenthal, K. A. Magliano, K. Turkiewicz, and D. E. Lehrman. PM_{2.5} chemical composition and spatiotemporal variability during the California regional PM₁₀/PM_{2.5} air quality study (CRPAQS). *Journal of Geophysical Research-Atmospheres*, 111(D10), 2006a.
- J. C. Chow, J. G. Watson, D. H. Lowenthal, L.-W. A. Chen, B. Zielinska, L. Rinehart, and K. L. Magliano. Evaluation of organic markers for chemical mass balance source apportionment at the fresno supersite. *Atmos. Chem. Phys. Discussion*, 6:10341–10372, 2006b.
- J. J. de Hartog, G. Hoek, A. Peters, K. L. Timonen, A. Ibald-Mulli, B. Brunekreef, J. Heinrich, P. Tiittanen, J. H. van Wijnen, W. Kreyling, M. Kulmala, and J. Pekkanen. Effects of fine and ultrafine particles on cardiorespiratory symptoms in elderly subjects with coronary heart disease - the ultra study. *American Journal of Epidemiology*, 157(7):613–623, 2003.
- R. J. Delfino, B. D. Coate, R. S. Zeiger, J. M. Seltzer, D. H. Street, and P. Koutrakis. Daily asthma severity in relation to personal ozone exposure and outdoor fungal spores. *American Journal of Respiratory and Critical Care Medicine*, 154(3):633–641, 1996.
- M. P. Fraser and K. Lakshmanan. Using levoglucosan as a molecular marker for the long-range transport of biomass combustion aerosols. *Environmental Science & Technology*, 34(21):4560–4564, Nov. 2000.
- P. Gaffney. Updating the ARB PM_{2.5} size speciation profiles for fugitive dust sources. Technical report, 2006.

- J. D. Herner, J. Aw, O. Gao, D. P. Chang, and M. J. Kleeman. Size and composition distribution of airborne particulate matter in northern California: I-particulate mass, carbon, and water-soluble ions. *Journal of the Air & Waste Management Association*, 55(1):30–51, 2005.
- M. J. Kleeman, G. R. Cass, and A. Eldering. Modeling the airborne particle complex as a source-oriented external mixture. *Journal of Geophysical Research-Atmospheres*, 102(D17):21355–21372, 1997.
- T. E. Lane, R. W. Pinder, M. Shrivastava, A. L. Robinson, and S. N. Pandis. Source contribution to primary organic aerosol: Comparison of the results of a source-resolved model and the chemical mass balance approach. *Atmospheric Environment*, 41:3758–3776, 2007.
- C. MacDonald, M. McCarthy, T. Dye, N. Wheeler, H. Hafner, and P. Roberts. Transport and dispersion during wintertime particulate matter episodes in the San Joaquin Valley, California. *Journal of the Air and Waste Management Association*, 56(7):961–976, 2006.
- L. E. Murr, E. V. Esquivel, and J. J. Bang. Characterization of nanostructure phenomena in airborne particulate aggregates and their potential for respiratory health effects. *Journal of Materials Science-Materials in Medicine*, 15(3):237–247, 2004.
- J. J. Schauer, W. F. Rogge, L. M. Hildemann, M. A. Mazurek, and G. R. Cass. Source apportionment of airborne particulate matter using organic compounds as tracers. *Atmospheric Environment*, 30(22):3837–3855, 1996.
- P. A. Solomon and K. L. Magliano. Objectives and design of central California’s 1995 integrated monitoring study of the California regional PM₁₀/PM_{2.5} air quality study. *Journal of the Air & Waste Management Association*, 49:199–215, 1999.
- S. von Klot, G. Wolke, T. Tuch, J. Heinrich, D. W. Dockery, J. Schwartz, W. G. Kreyling, H. E. Wichmann, and A. Peters. Increased asthma medication use in association with ambient fine and ultrafine particles. *European Respiratory Journal*, 20(3):691–702, 2002.
- J. G. Watson, J. C. Chow, Z. Q. Lu, E. M. Fujita, D. H. Lowenthal, D. R. Lawson, and L. L. Ashbaugh. Chemical mass-balance source apportionment of pm(10) during the southern California air-quality study. *Aerosol Science and Technology*, 21(1):1–36, 1994.
- J. G. Watson, J. C. Chow, and J. E. Houck. PM_{2.5} chemical source profiles for vehicle exhaust, vegetative burning, geological material, and coal burning in northwestern colorado during 1995. *Chemosphere*, 43(8):1141–1151, 2001.
- J. G. Watson, T. Zhu, J. C. Chow, J. Engelbrecht, E. M. Fujita, and W. E. Wilson. Receptor modeling application framework for particle source apportionment. *Chemosphere*, 49(9):1093–1136, 2002.
- Q. Ying and M. J. Kleeman. Efficient source apportionment of airborne particulate matter using an internally mixed air quality model with artificial tracers. *Environmental Science & Engineering (China)*, 1(1):91–99, 2004.

- Q. Ying and M. J. Kleeman. Source contributions to the regional distribution of secondary particulate matter in California. *Atmospheric Environment*, 40(4):736–752, 2006.
- Q. Ying, M. Mysliwiec, and M. J. Kleeman. Source apportionment of visibility impairment using a three-dimensional source-oriented air quality model. *Environmental Science & Technology*, 38(4):1089–1101, 2004.
- Q. Ying, M. P. Fraser, R. J. Griffin, J. J. Chen, and M. J. Kleeman. Verification of a source-oriented externally mixed air quality model during a severe photochemical smog episode. *Atmospheric Environment*, 41(7):1521–1538, 2007.
- Q. Ying, J. Lu, and M. J. Kleeman. Modeling air quality during the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) using the CIT/UCD source oriented air quality model - Part III. source apportionment of secondary particulate matter. *Atmospheric Environment*, Submitted, 2008a.
- Q. Ying, J. Lu, P. Livingstone, P. Allen, and M. J. Kleeman. Modeling air quality during the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) using the CIT/UCD source oriented air quality model - Part I. Base case model results. *Atmospheric Environment*, Submitted, 2008b.

List of Tables

1	Total gas and PM emission for a typical weekday	15
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Table 1: Total gas and PM emission for a typical weekday

	EC	OC	N(-III) (kg/day)	S(VI)	N(V)	CO (kmol/day)	NO	NO ₂	SO ₂
Paved Road Dust	0.0	10805.0	47.0	1581.0	77.0	3.0	1.0	0.0	0.0
Fugitive Dust	1698.0	25911.0	85.0	1928.0	400.0	0.0	0.0	0.0	0.0
Wood Smoke	1153.0	149316.0	146.0	549.0	549.0	47619.0	333.0	38.0	39.0
Diesel Engine	16915.0	4777.0	14.0	66.0	61.0	9110.0	17336.0	1983.0	410.0
Non-cat. Engine	126.0	5405.0	2.0	8.0	0.0	70141.0	1567.0	179.0	5.0
Cat.Engine	119.0	2615.0	0.0	92.0	0.0	156137.0	9819.0	1123.0	0.0
Meat Cooking	44.0	9217.0	0.0	15.0	24.0	0.0	0.0	0.0	0.0
High Sulfur Fuel	210.0	1401.0	25.0	3339.0	254.0	4987.0	1433.0	164.0	662.0
Other	8988.0	37111.0	970.0	3714.0	650.0	43876.0	6423.0	735.0	823.0

List of Figures

1	Averaged $PM_{2.5}$ EC and OC fractions for each emission source category used in the EC and OC source apportionment calculations.	17
2	Source contribution to the 24-hour average EC, OC, and primary $PM_{2.5}$ on December 26, 2000 calculated using the externally mixed aerosol model and the internal mixture with artificial tracer model. Different symbols represent different emission source categories. The data points included in the figures are predicted concentrations at Bethel Island, Sacramento, Fresno, Angiola and Bakersfield.	18
3	Relative source contribution to the averaged primary $PM_{2.5}$ mass concentrations predicted by the CMB receptor model and the UCD/CIT source-oriented air quality model at (a) Angiola and (b) Fresno. The contribution from mobile source is the sum of the contribution from gasoline engines and diesel engines. The contribution of dust particles is not included in this calculation. Units are $\mu g m^{-3}$	19
4	Source contribution to 24-hour average primary $PM_{2.5}$ mass concentrations at Fresno (FSF) during the IOPs calculated using (a) CMB and (b) UCD/CIT source oriented air quality model. Units are $\mu g m^{-3}$	20
5	Relative source contribution to EC, OC and $PM_{2.5}$ mass at Fresno from December 15, 2000 to January 7, 2001	21
6	Relative source contribution to EC, OC and $PM_{2.5}$ mass at Angiola from December 15, 2000 to January 7, 2001	22
7	Source contribution to $PM_{2.5}$ EC concentrations on December 28, 2000. The scale on each panel is different. Units are $\mu g m^{-3}$	23
8	Source contribution to $PM_{2.5}$ OC concentrations on December 28, 2000 The scale on each panel is different. Units are $\mu g m^{-3}$	24

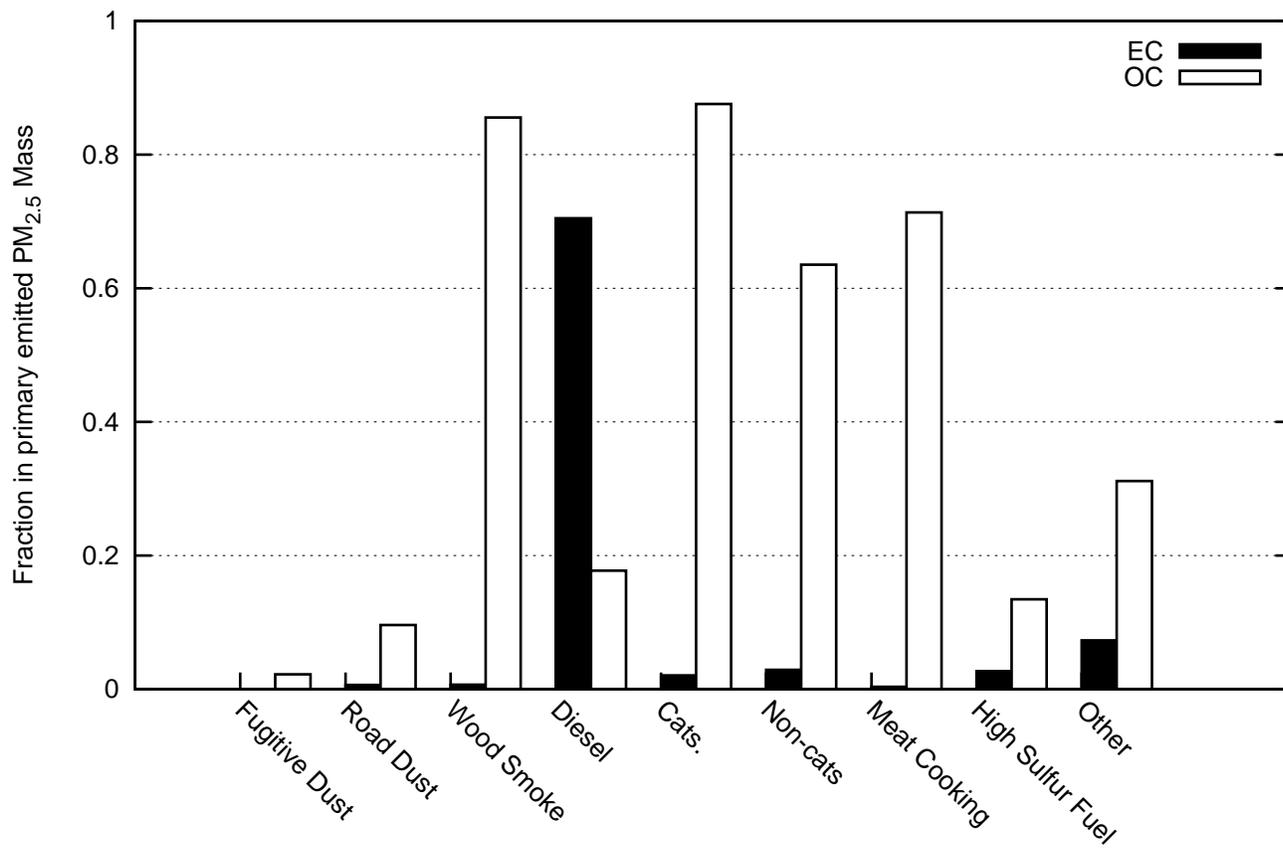


Figure 1: Averaged PM_{2.5} EC and OC fractions for each emission source category used in the EC and OC source apportionment calculations.

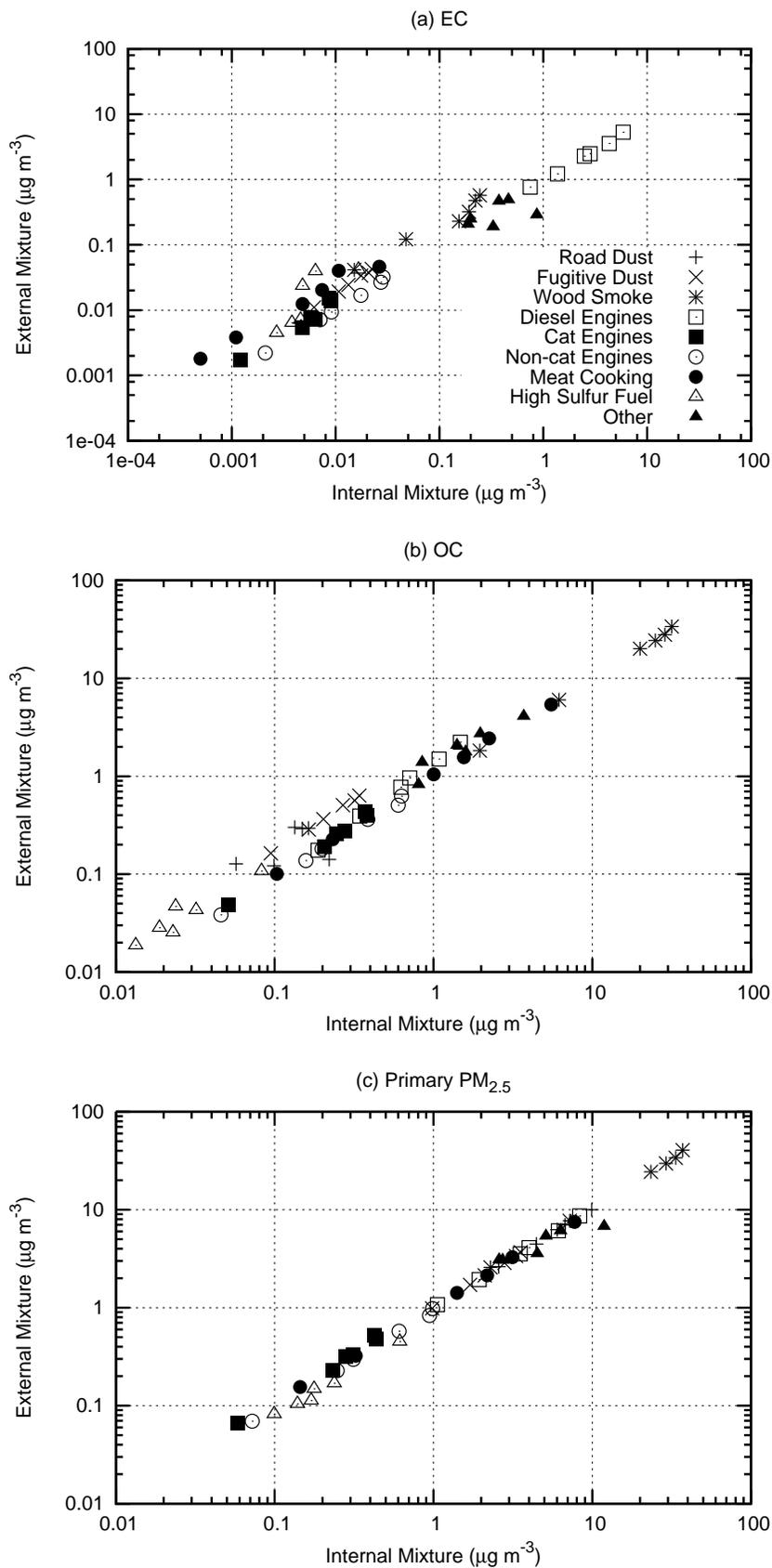


Figure 2: Source contribution to the 24-hour average EC, OC, and primary PM_{2.5} on December 26, 2000 calculated using the externally mixed aerosol model and the internal mixture with artificial tracer model. Different symbols represent different emission source categories. The data points included in the figures are predicted concentrations at Bethel Island, Sacramento, Fresno, Angiola and Bakersfield.

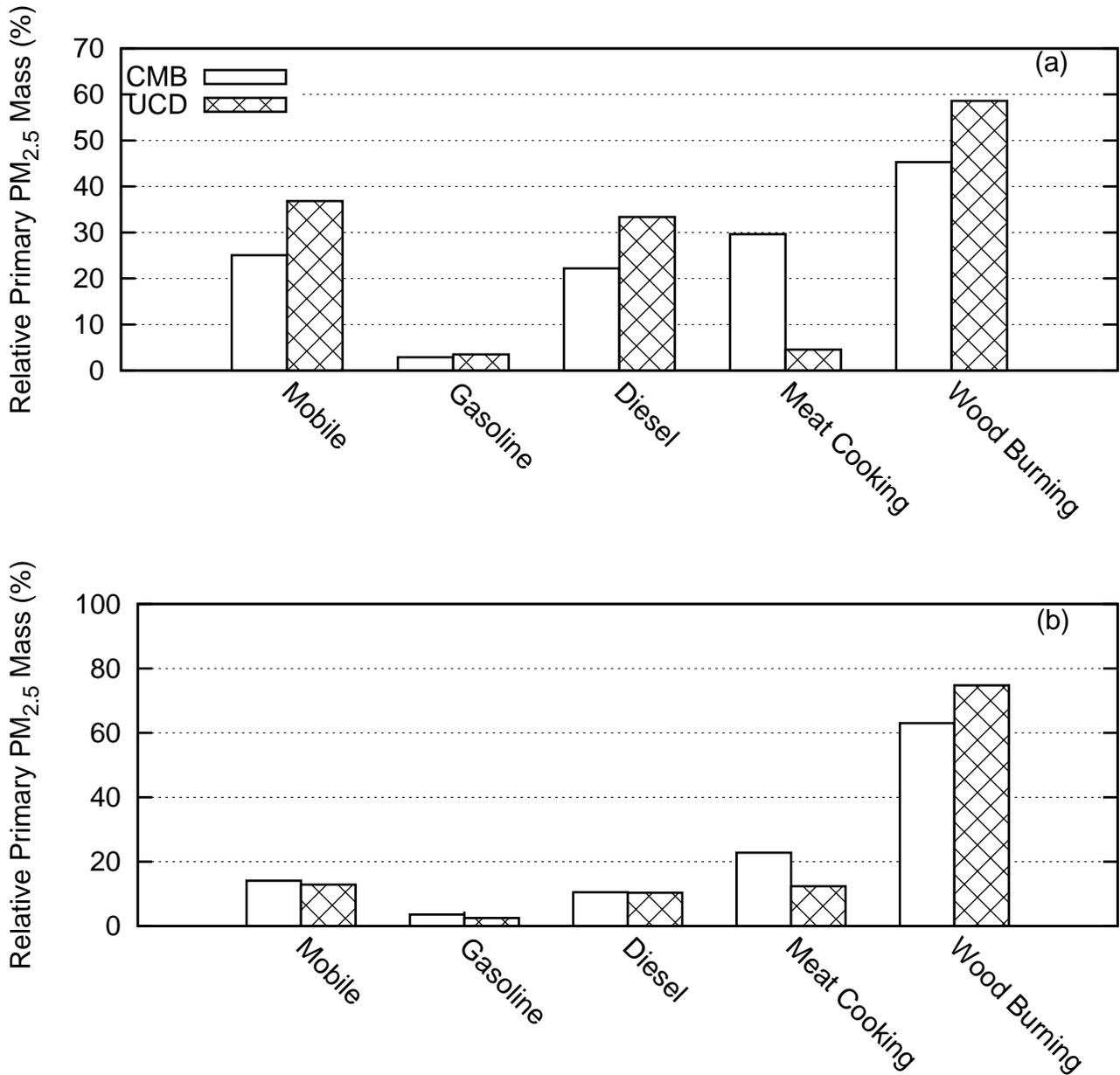


Figure 3: Relative source contribution to the averaged primary PM_{2.5} mass concentrations predicted by the CMB receptor model and the UCD/CIT source-oriented air quality model at (a) Angiola and (b) Fresno. The contribution from mobile source is the sum of the contribution from gasoline engines and diesel engines. The contribution of dust particles is not included in this calculation. Units are $\mu\text{g m}^{-3}$.

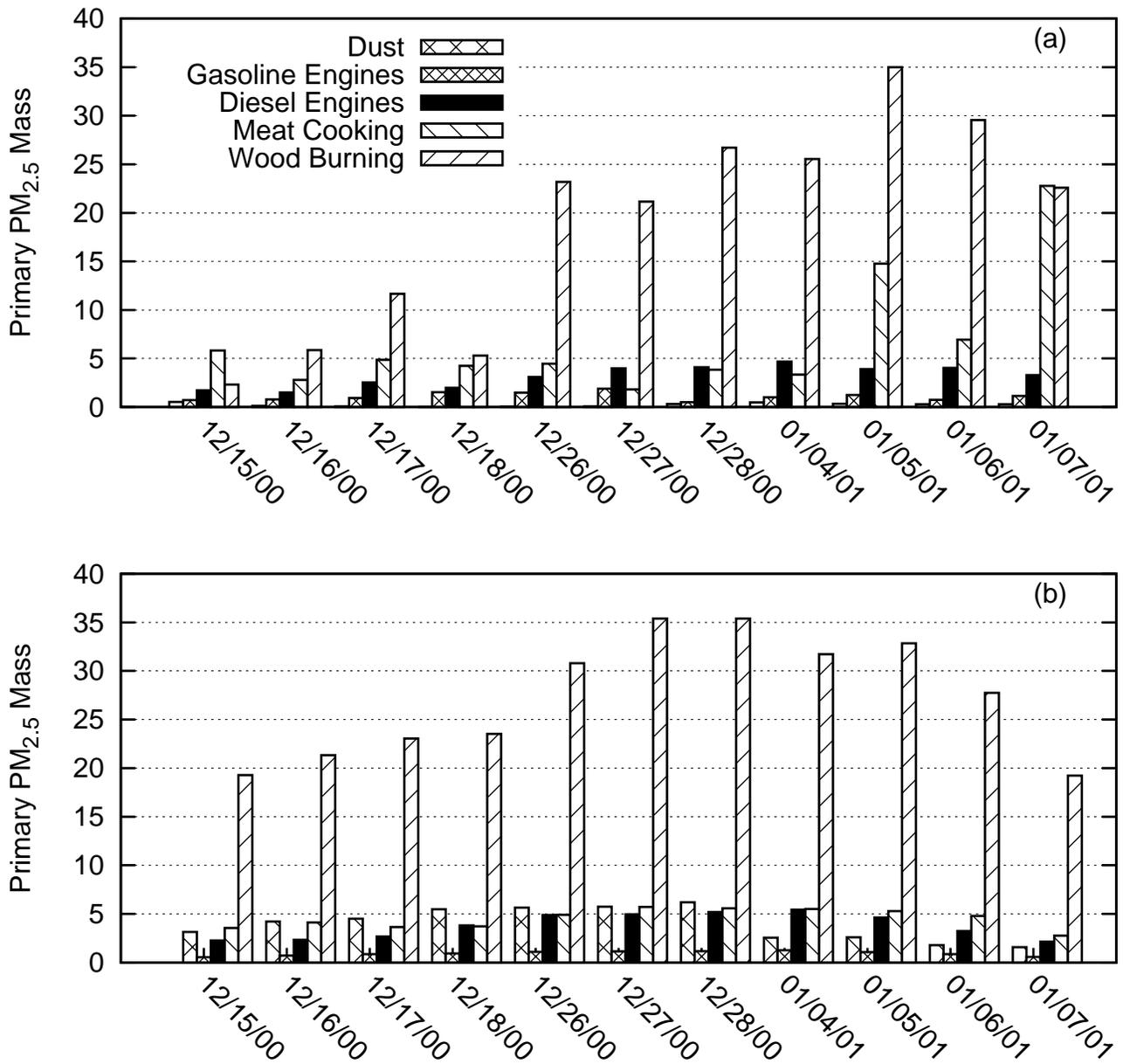


Figure 4: Source contribution to 24-hour average primary $PM_{2.5}$ mass concentrations at Fresno (FSF) during the IOPs calculated using (a) CMB and (b) UCD/CIT source oriented air quality model. Units are $\mu g m^{-3}$.

Figure 5: Relative source contribution to EC, OC and PM_{2.5} mass at Fresno from December 15, 2000 to January 7, 2001

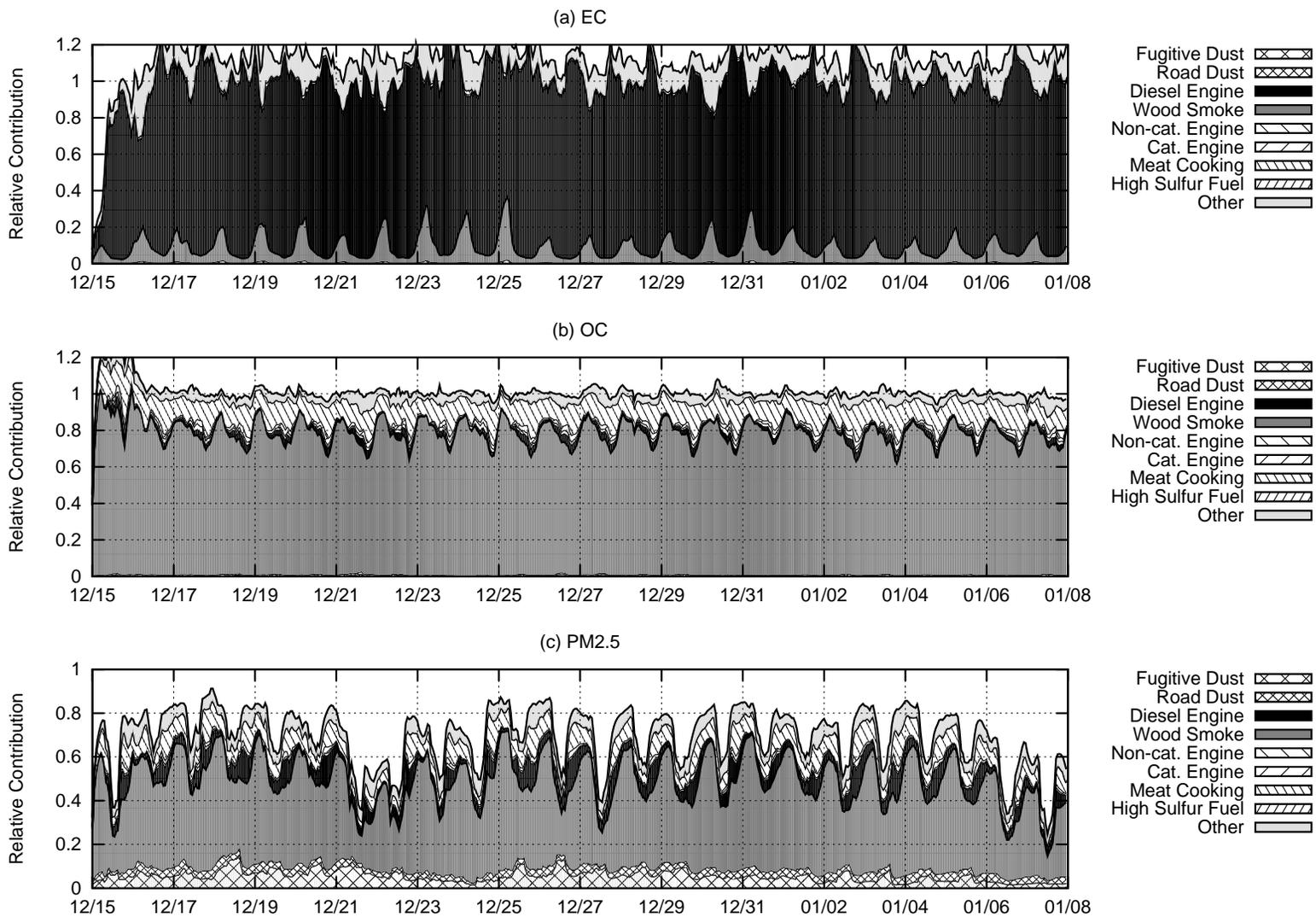
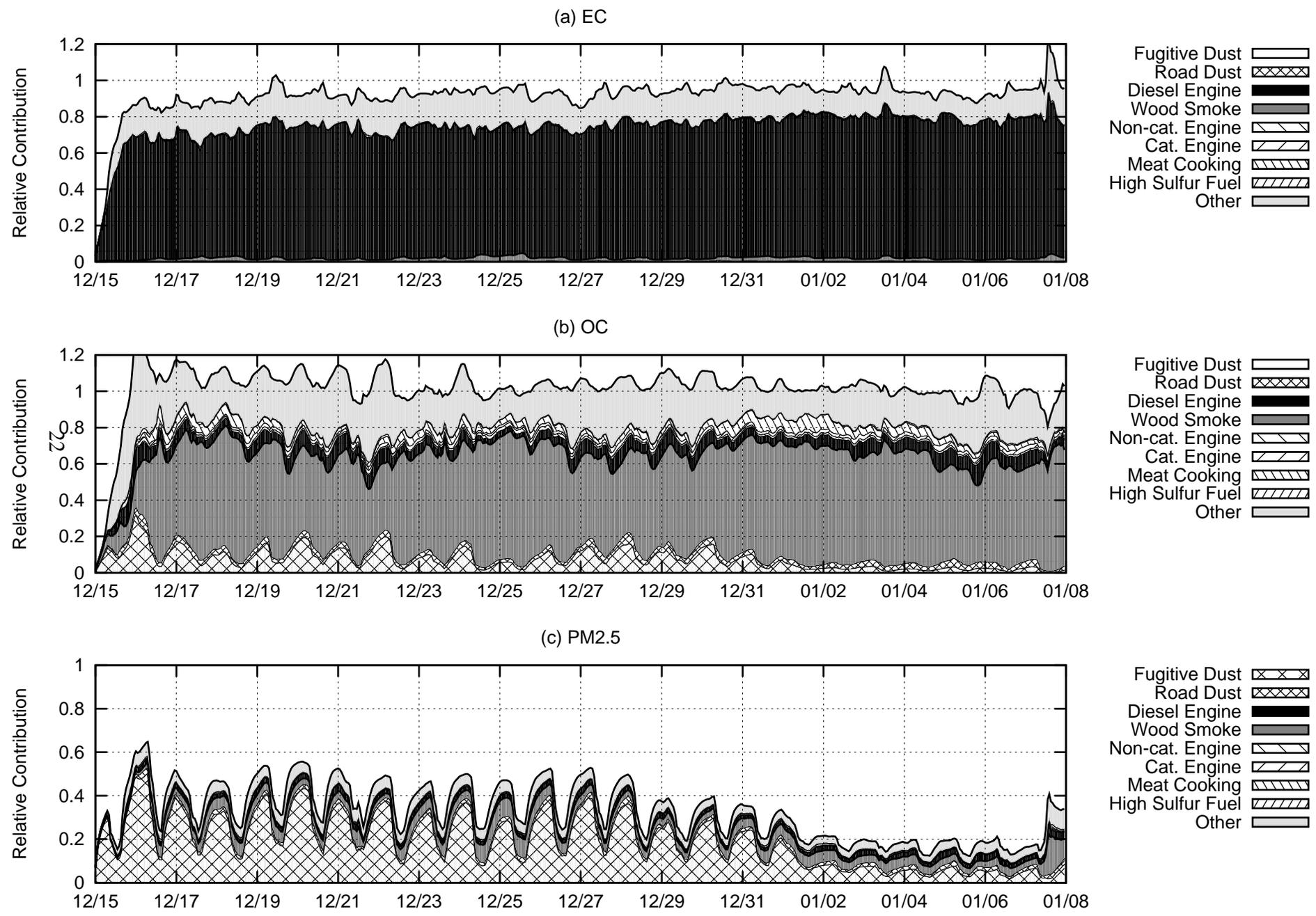


Figure 6: Relative source contribution to EC, OC and PM_{2.5} mass at Angiola from December 15, 2000 to January 7, 2001



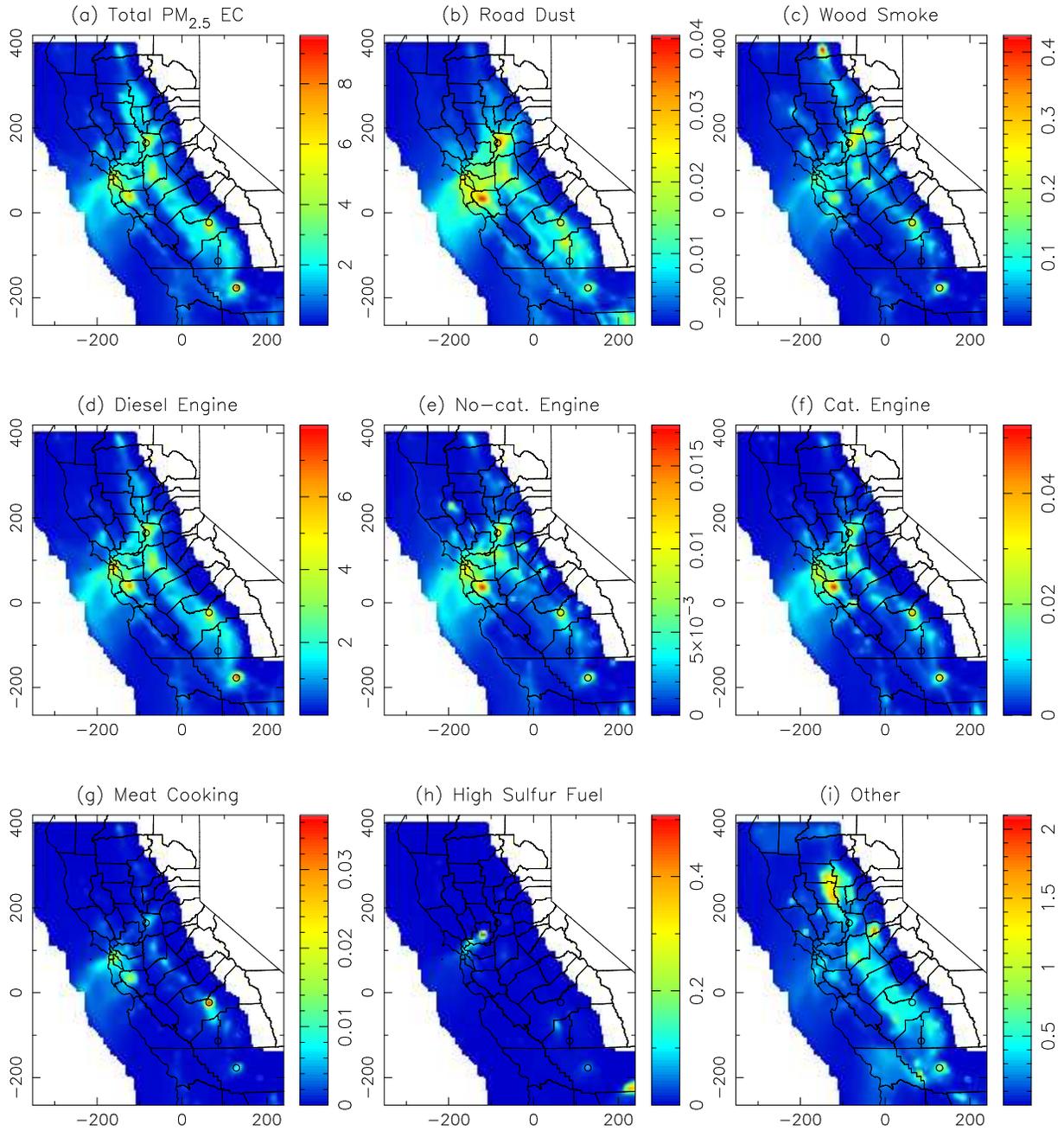


Figure 7: Source contribution to $PM_{2.5}$ EC concentrations on December 28, 2000. The scale on each panel is different. Units are $\mu g m^{-3}$

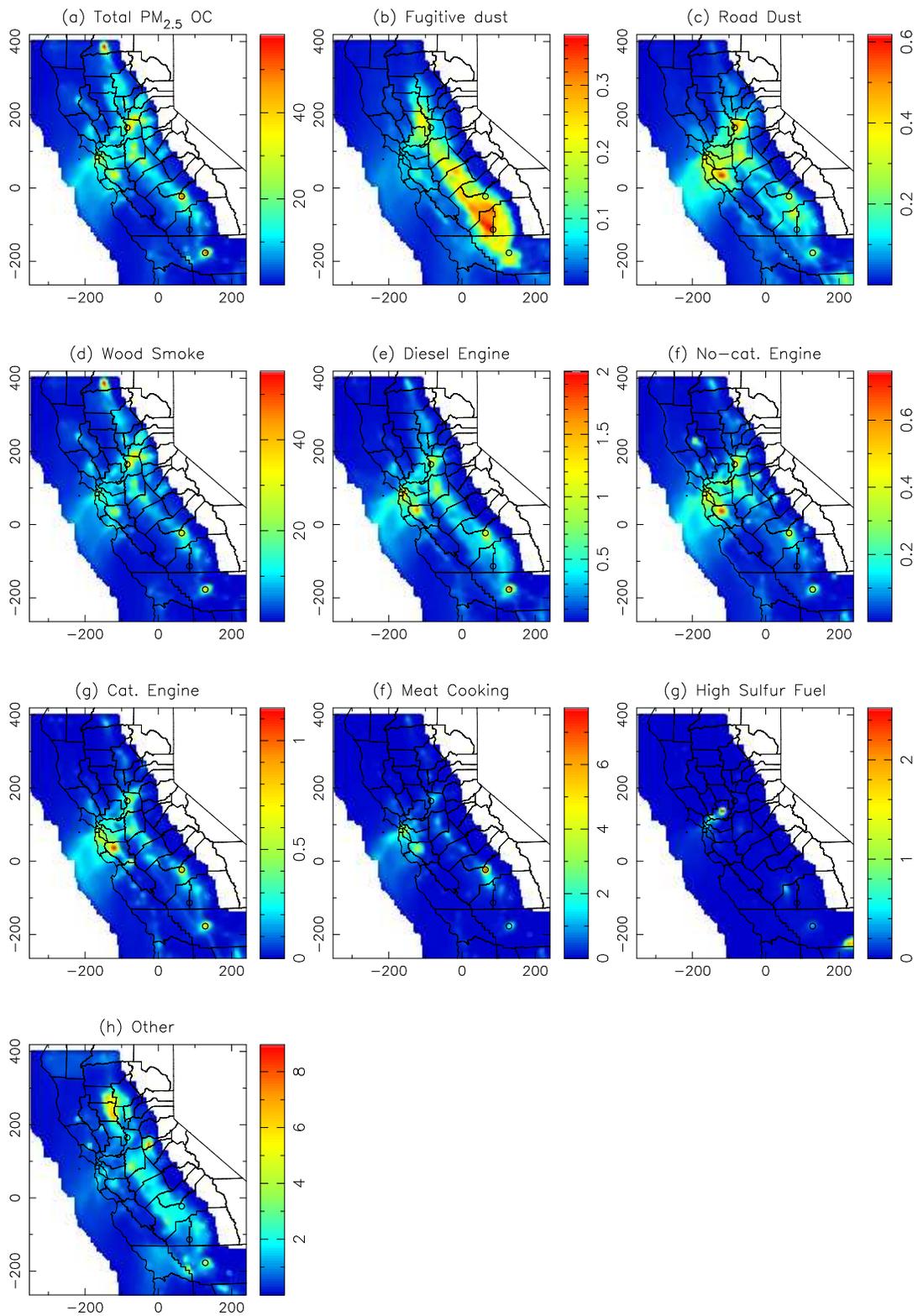


Figure 8: Source contribution to $PM_{2.5}$ OC concentrations on December 28, 2000 The scale on each panel is different. Units are $\mu g m^{-3}$