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Source Apportionment of Fine Particulate Matter in the Southeastern United States

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ABSTRACT

Particulate matter (PM) less than 2.5 μm in size ($PM_{2.5}$) source apportionment by chemical mass balance receptor modeling was performed to enhance regional characterization of source impacts in the southeastern United States. Secondary particles, such as NH_4HSO_4, $(NH_4)_2SO_4$, NH_4NO_3, and secondary organic carbon (OC) (SOC), formed by atmospheric photochemical reactions, contribute the majority (>50%) of ambient $PM_{2.5}$ with strong seasonality. Source apportionment results indicate that motor vehicle and biomass burning are the two main primary sources in the southeast, showing relatively more motor vehicle source impacts rather than biomass burning source impacts in populated urban areas and vice versa in less urbanized areas. Spatial distributions of primary source impacts show that each primary source has distinctively different spatial source impacts. Results also find impacts from shipping activities along the coast. Spatiotemporal correlations indicate that secondary particles are more regionally distributed, as are biomass burning and dust, whereas impacts of other primary sources are more local.

INTRODUCTION

Epidemiological studies suggest that ambient particulate matter (PM) has significant associations with adverse respiratory and cardiovascular health effects,[1–5] and prompted the U.S. Environmental Protection Agency (EPA) to promulgate National Ambient Air Quality Standards (NAAQS) for PM less than 2.5 μm in size ($PM_{2.5}$) (15 $\mu g/m^3$ as annual standard and 65 $\mu g/m^3$ as 24-hr standard).[6] The majority of the past epidemiological studies focused on linking human exposures to PM mass and its chemical components.[1–6] More recent studies have been conducted to understand associations between PM emission sources and human exposure.[7–10]

Associated with the new NAAQS, the EPA established the $PM_{2.5}$ chemical Speciation Trend Network (STN) program to provide nationally consistent data for the assessment of trends.[11] Twenty-four-hour integrated filter-based samples are collected every 3 or 6 days at each monitoring site. The samples are analyzed to determine gravimetric mass and chemical composition, including ions, trace elements, and carbonaceous compounds (i.e., organic and elemental carbons: organic carbon [OC] and elemental carbon [EC]).

Prior $PM_{2.5}$ source apportionment studies conducted in the Southeast[12–20] have assessed the applicability of different source apportionment methods. However, little information about regional characterization of $PM_{2.5}$ source impacts in the region is provided in the studies. The main goal of this study was to conduct source apportionment of $PM_{2.5}$ and develop a regional perspective of source impacts in the southeastern United States 23 STN sites from six southeastern states were selected (Figure 1). A chemical mass balance receptor model (CMB) was applied to identify primary source contribution for ambient measurement data collected at 23 STN sites between January 2002 and November 2003. Seasonal variation and spatiotemporal correlations of the identified PM sources were examined. This study provides useful information for possible future epidemiological studies that aim to improve our understanding of the association between fine PM sources and human health exposure and ultimately help develop effective PM control strategies.

IMPLICATION

Regional characterization of $PM_{2.5}$ source impacts was estimated in the southeastern United States The source apportionment results show that secondary $PM_{2.5}$ is regionally distributed, as are biomass burning and dust. Other primary sources are more local. Much of the total regional $PM_{2.5}$ can be attributed to sulfur dioxide (SO_2) and biomass burning emissions, whereas mobile sources are significant in cities. Regional haze programs as well as $PM_{2.5}$ control strategy development in the Southeast can use the quantitative source impacts for planning.

METHODS

Primary OC Estimation

The CMB receptor model is used to estimate primary source contributions by using ambient measurement and source profile data, applying a linear equation for conservation of species.

$$C_i = \sum_{j=1}^{J} f_{i,j} S_j + \varepsilon_i,\ i = 1, \ldots . n \qquad (1)$$

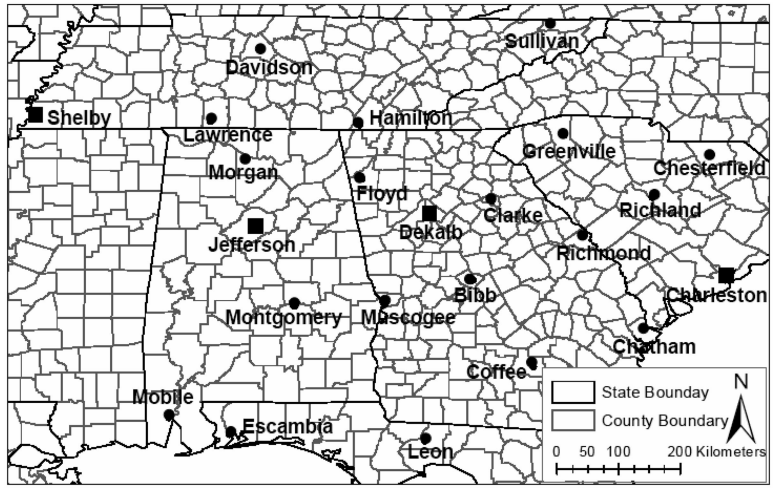


Figure 1. STN ambient monitoring sites in the southeastern United States, measurements every 3 days (■) or 6 days (●).

where C_i is the ambient concentration of species i, f_i is the fraction of species i in source j, S_j is the source contribution of source j, and ε_i is an error term. Chemical species used in CMB are assumed to be nonreactive.[21] However, OC measured at a receptor site includes primary OC emitted from emission sources and secondary OC (SOC) from photochemical formation. To apply CMB using OC information one must either add a SOC source, or, as done here, estimate primary OC. Simply adding a pure SOC source could lead to collinearity problems with OC dominant sources. Therefore, it is desirable to estimate primary OC before the source apportionment. Although many studies have been conducted to understand SOC,[22–33] it is problematic separating primary from SOC via direct chemical analysis.

Table 1. Primary OC/EC ratio estimations by combining two different EC tracer methods.

State	County	Primary OC/EC Ratio from Ambient Data (95% Confidence Interval)	Median Primary OC/EC Ratio from Emission Inventory[a] (Min-Max)	Min-Max Primary OC/EC Ratio from a Combined Method
Georgia	Bibb	5.84 (5.13–6.56)	3.31 (2.90–4.70)	5.13–8.30
	Coffee	6.99 (6.14–7.85)	4.02 (2.10–7.11)	3.66–12.38
	Clarke	4.08 (2.95–5.20)	2.84 (2.84–4.00)	3.01–5.75
	Chatham	4.06 (3.04–5.08)	3.38 (2.87–5.19)	3.45–6.24
	Dekalb	3.02 (2.70–3.34)	2.10 (1.51- 2.44)	2.18–3.51
	Floyd	7.33 (4.56–10.09)	5.86 (5.41–5.49)	6.76–8.12
	Muscogee	7.63 (5.95–9.30)	3.13 (2.10–4.95)	5.12–12.05
	Richmond	6.42 (5.05–7.08)	3.53 (2.65–4.54)	4.82–8.27
Alabama	Jefferson	2.74 (2.47–3.01)	4.05 (3.93–4.42)	2.66–2.99
	Mobile	4.35 (3.00–5.69)	3.67 (3.26- 4.74)	3.86–5.60
	Montgomery	4.13 (3.13–5.14)	2.48 (1.93–3.77)	3.21–6.30
	Morgan	6.99 (5.35–8.63)	2.57 (2.15–3.35)	5.85–9.14
Florida	Escambia	5.03 (4.15–5.90)	4.46 (3.71–10.55)	4.18–11.89
	Leon	3.53 (3.22–3.85)	5.23 (2.78–12.64)	1.88–8.53
South Carolina	Charleston	3.83 (3.42–4.23)	3.40 (2.63–5.43)	2.96–6.11
	Chesterfield	7.88 (5.53–10.23)	3.36 (2.63–4.27)	6.20–10.02
	Greenville	5.86 (4.43–7.28)	2.55 (1.99–3.02)	4.57–6.94
	Richland	4.65 (3.74–5.56)	3.72 (3.37–4.06)	4.21–5.07
Tennessee	Davidson	3.11 (2.48–3.75)	2.24 (1.84–2.64)	2.56–3.66
	Hamilton	5.06 (4.12–5.99)	2.30 (1.64–3.14)	3.60–6.89
	Lawrence	4.89 (3.92–5.86)	2.78 (2.32–4.75)	4.08–8.34
	Shelby	3.45 (3.02–3.89)	1.72 (1.31–2.08)	2.63–4.16
	Sullivan	3.81 (2.82–4.80)	3.63 (3.59–4.39)	3.76–4.61

Notes: [a]Primary $PM_{2.5}$ source categories and references for OC and EC weight fraction of $PM_{2.5}$ were used to obtain monthly primary OC/EC ratios based on emission inventory. On-road: light/heavy duty gasoline and diesel vehicles;[42] Non-road: off-highway gasoline and diesel vehicles;[42] Point non-electricity generation: fuel combustion (coal,[43] distilled oil,[44] natural gas[45]), mineral production,[46] pulp and paper production,[46] metal production;[46] Point electricity generation: power plant (coal,[43] distilled oil,[44] natural gas[45]); Area: wild fires, [47] prescribed burning, [42] agricultural burning,[44] yard waste burning, [43] land clearing,[43] fuel combustion (coal,[43] distilled oil,[44] natural gas[45]), residential wood burning, [42] waste incineration,[44] meat cooking;[42] Dust: agricultural production,[48] construction,[49] paved road dust,[43] unpaved road dust.[43]

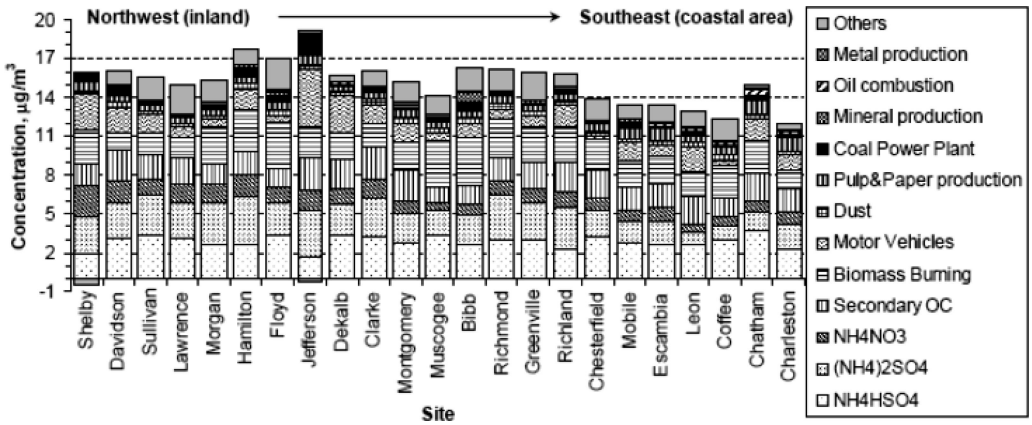


Figure 2. Source apportionment results averaged from November 2002 to November 2003.

The EC tracer method, an indirect method, has been used to estimate primary and SOC, because EC is a good tracer for carbonaceous particles from primary combustion sources.[34–40] In general, there are two approaches in the EC tracer method to separate primary and SOC. One is using ambient measurement data and the other is based on primary emission inventory data. In this study, a combined method was used to estimate monthly primary OC/EC ratios. First, using ambient OC and EC data at each site, a primary OC/EC ratio was derived. A recent study shows that Deming linear least-squares regression is the superior method among several ambient EC tracer methods.[41] Deming regression was applied to daily OC and EC data in the lowest 10% by OC/EC ratio. Second, monthly primary OC/elemental carbon ratios were obtained by compiling primary OC and EC emission data (eq 2). On the basis of the 2001 National Emission Inventory, annual PM emissions were calculated for various categories from counties within 25 km of each monitoring site. Monthly temporal profiles of PM emissions were applied to get monthly PM emissions at each site and then a source-specific OC and EC weight fraction from source emission experiments was multiplied to obtain monthly OC and EC emissions (Table 1):

$$\left[\frac{OC}{EC}\right]_p = \frac{\sum_{i=1}^{n} PM2.5_i \times OC_{f,i}}{\sum_{i=1}^{n} PM2.5_i \times EC_{f,ip}} \tag{2}$$

where $[OC/EC]_p$ is the monthly primary OC to EC ratio, $PM2.5_i$ is the monthly primary $PM_{2.5}$ emission (tons/month) from a source *i*, and $OC_{f,i}$ is the weight fraction of $PM_{2.5}$ from source *i*, and $EC_{f,i}$ is the weight fraction of $PM_{2.5}$ from a source *i*. The OC and EC weight fractions of $PM_{2.5}$ from the Interagency Monitoring of Protected Visual Environments (IMPROVE) method[50] were used because more comprehensive emissions data were available. Ambient STN OC/EC data were obtained using the National Institute of Occupational Safety and Health (NIOSH) method.[51] Deming regression provides one primary OC/EC ratio for the entire ambient dataset at each site. However, the primary OC/EC ratio varies as primary emissions change seasonally. The variability of monthly primary OC/EC ratios from a median ratio on the basis of EPA National Emission Inventory 2001 was weighted to the primary OC/EC ratio from the Deming regression (ambient data) to obtain monthly primary OC/EC ratios (eq 3).

$$\left[\frac{OC}{EC}\right]_{m,p} = \left[\frac{OC}{EC}\right]_{dr} + \left[\frac{OC}{EC}\right]_{dr} \times \left(\frac{\left[\frac{OC}{EC}\right]_p - \left[\frac{OC}{EC}\right]_{median,p}}{\left[\frac{OC}{EC}\right]_{median,p}}\right) \tag{3}$$

where $[OC/EC]_{m,p}$ is the final monthly primary OC to EC ratio, $[OC/EC]_{dr}$ is the primary OC to EC ratio from Deming regression, $[OC/EC]_p$ is the monthly primary OC to EC ratio from eq 2, and $[OC/EC]_{median,\ p}$ is the median value of $[OC/EC]_p$.

Although this combined method gives monthly primary OC to EC ratios at each site, there may be some limitations in the method. For example, biomass burning (e.g., wild fires, prescribed burning) is a major OC source. Emission inventory data may not correctly represent actual $PM_{2.5}$ emissions from wild fires because of their sporadic nature. Estimated monthly primary OC/EC ratios may not capture daily variability of the primary OC/EC ratio. The aforementioned factors can introduce uncertainties in the primary/secondary OC estimates.

Source Apportionment

Major primary sources used in the source apportionment include motor vehicles, biomass burning, dust, coal combustion, oil combustion, mineral, metal, and pulp and paper production. Theoretical profiles based on molecular weight fraction for ammonium bisulfate, ammonium sulfate, and ammonium nitrates were also included to identify inorganic secondary particle formation. The source profile for motor vehicles was generated by weighted average of catalyst-equipped gasoline power vehicles and diesel vehicles on the basis of their estimated emissions.[52,53] The biomass burning source profile was generated by averaging six source tests of southern woods.[54] The source profile used for dust was from

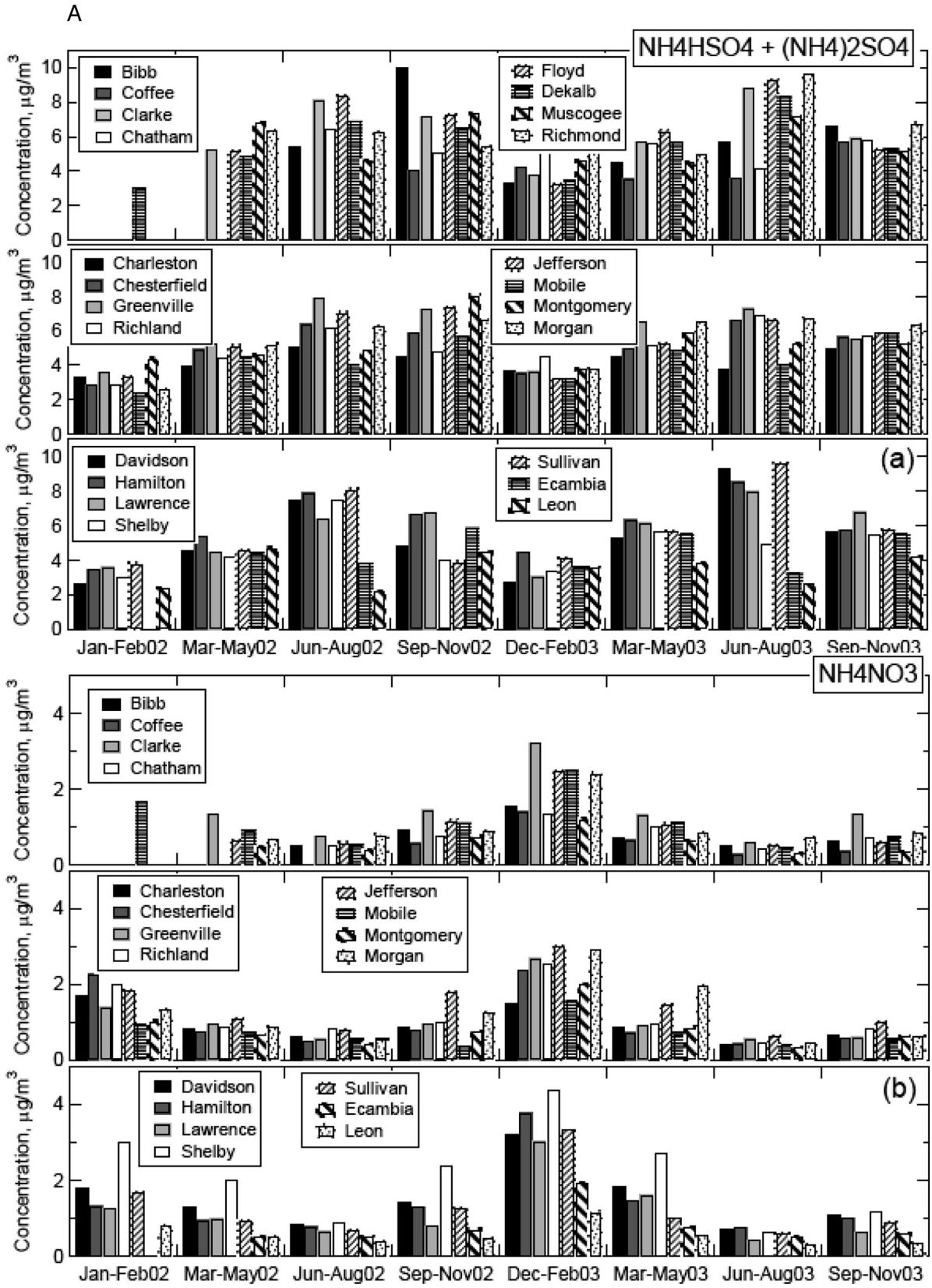
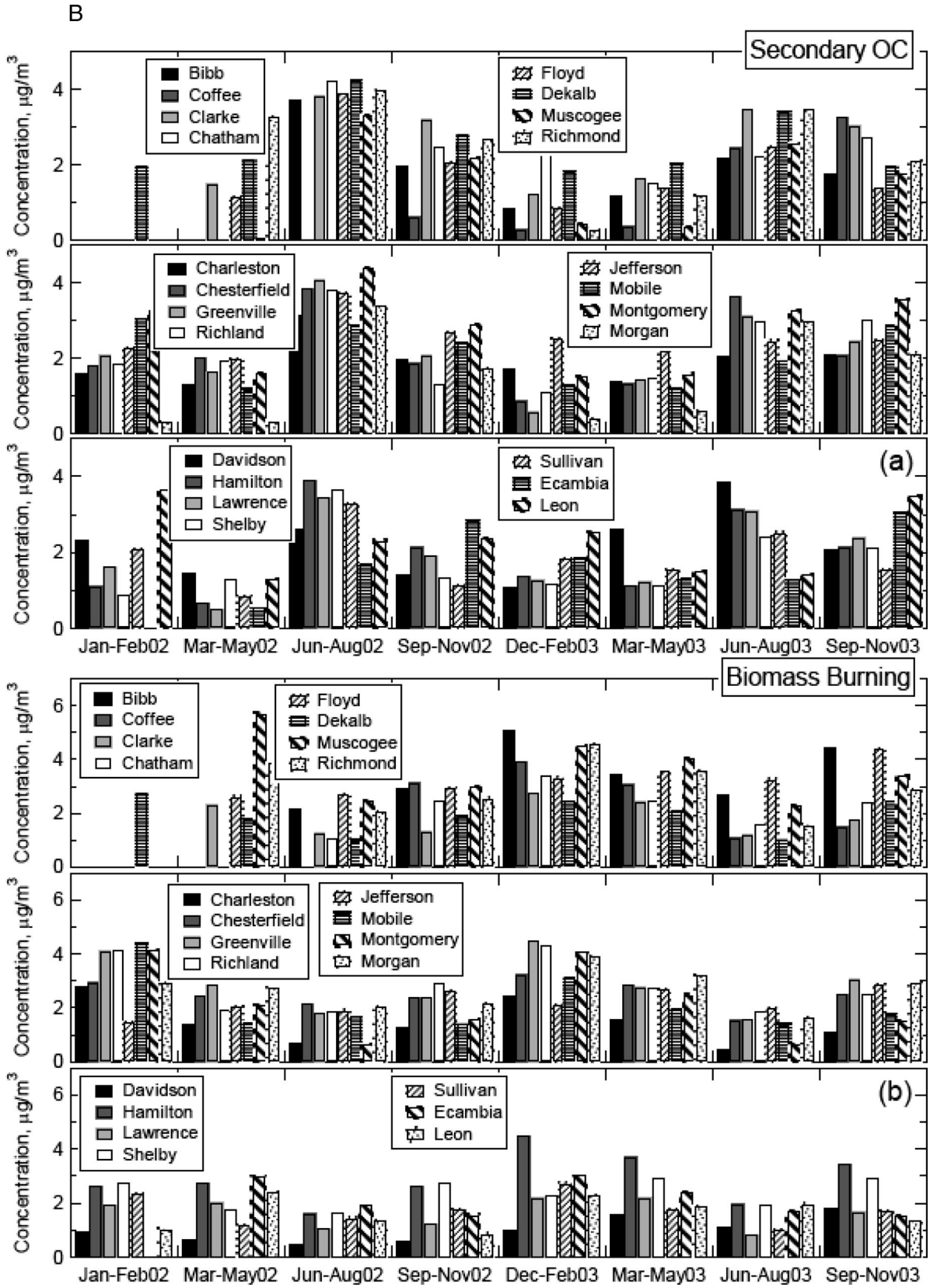


Figure 3. Panel A shows seasonal source contributions: (a) NH_4HSO_4 + $(NH_4)_2SO_4$ and (b) NH_4NO_3. Panel B shows seasonal source contributions: (a) SOC and (b) biomass burning. Panel C shows seasonal source contributions: (a) dust and (b) motor vehicles. Panel D shows seasonal source contributions: (a) pulp/paper production and (b) coal combustion. Panel E shows seasonal source contributions: (a) oil combustion and (b) metal production.



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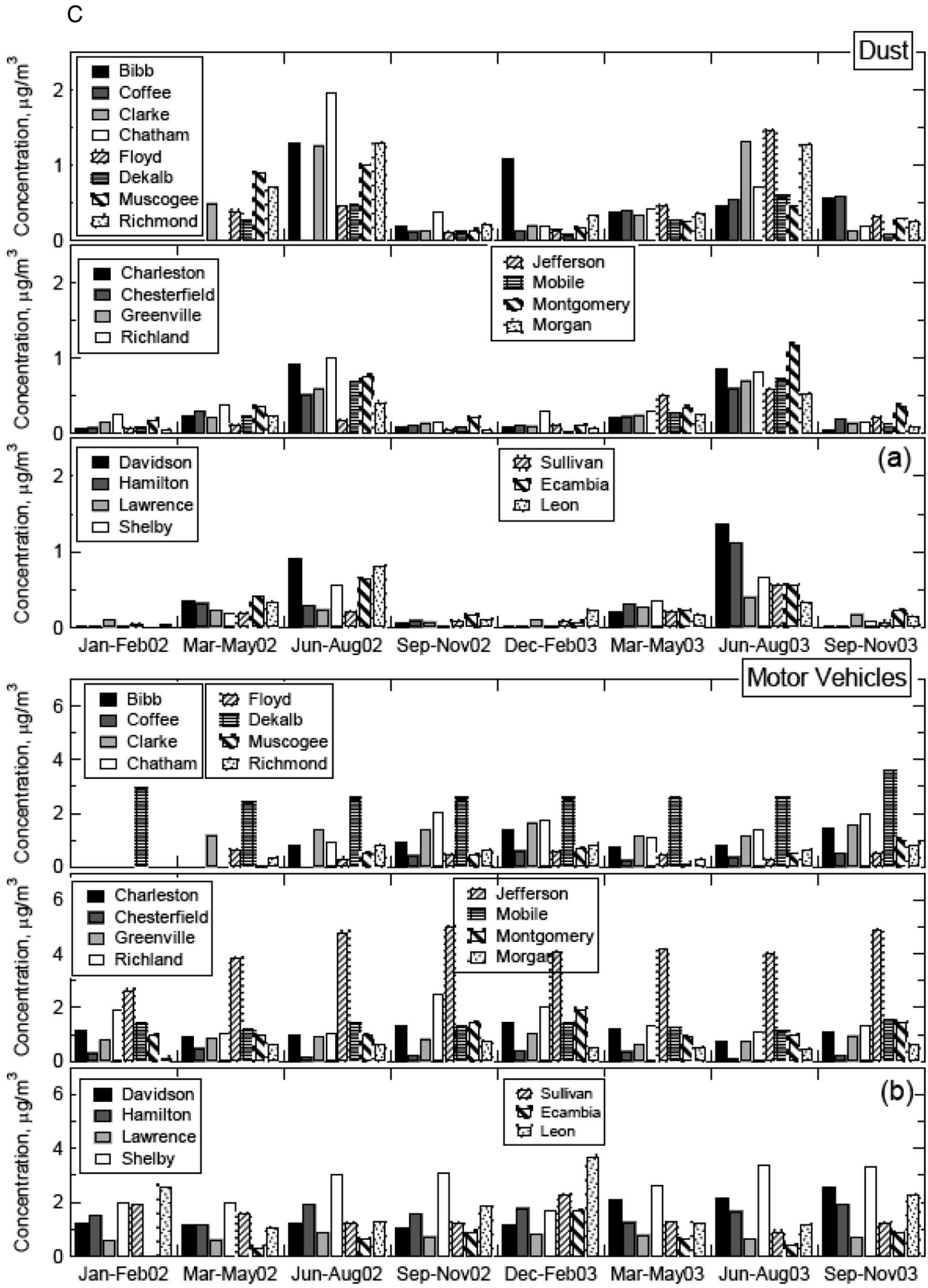

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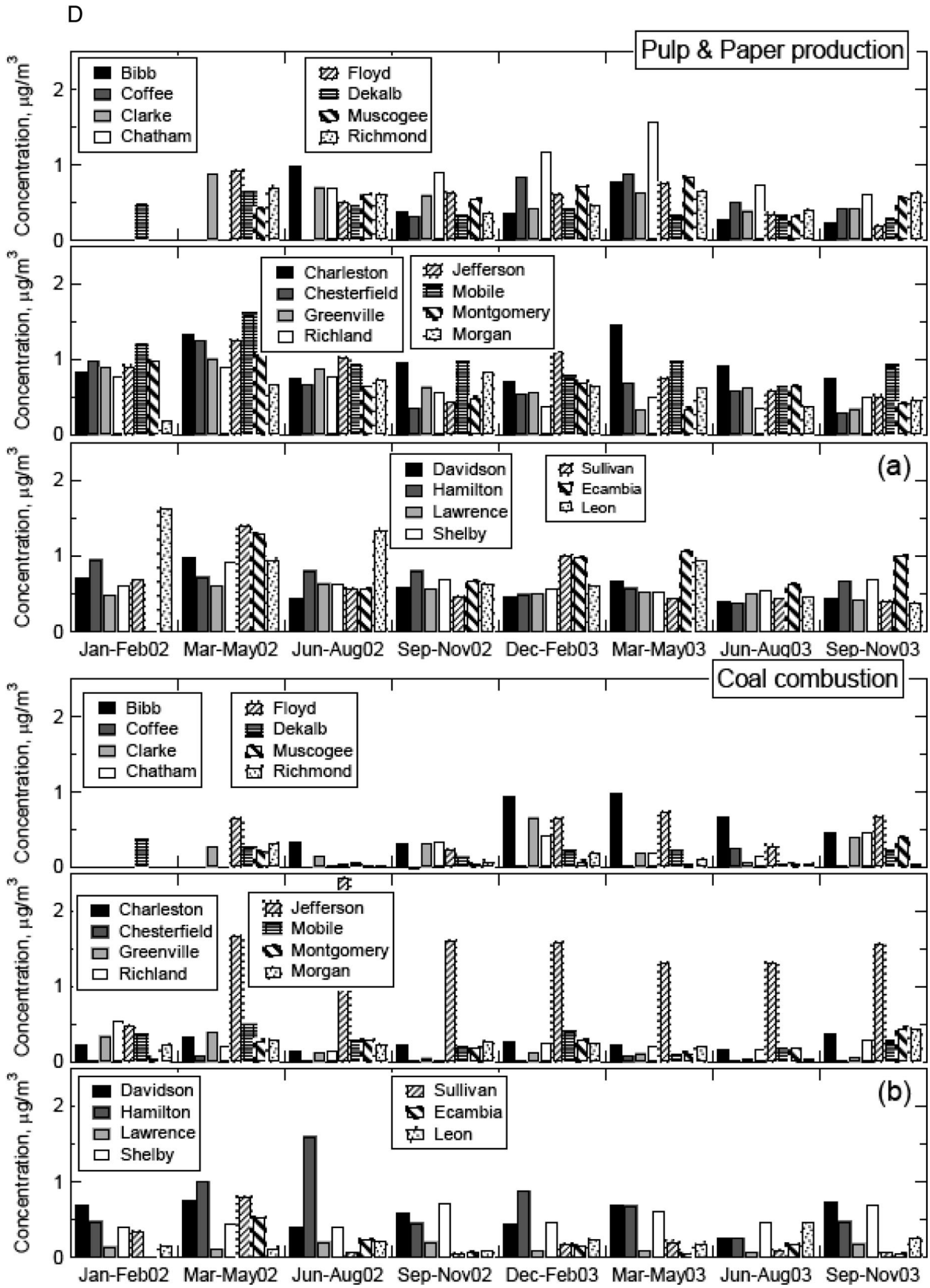

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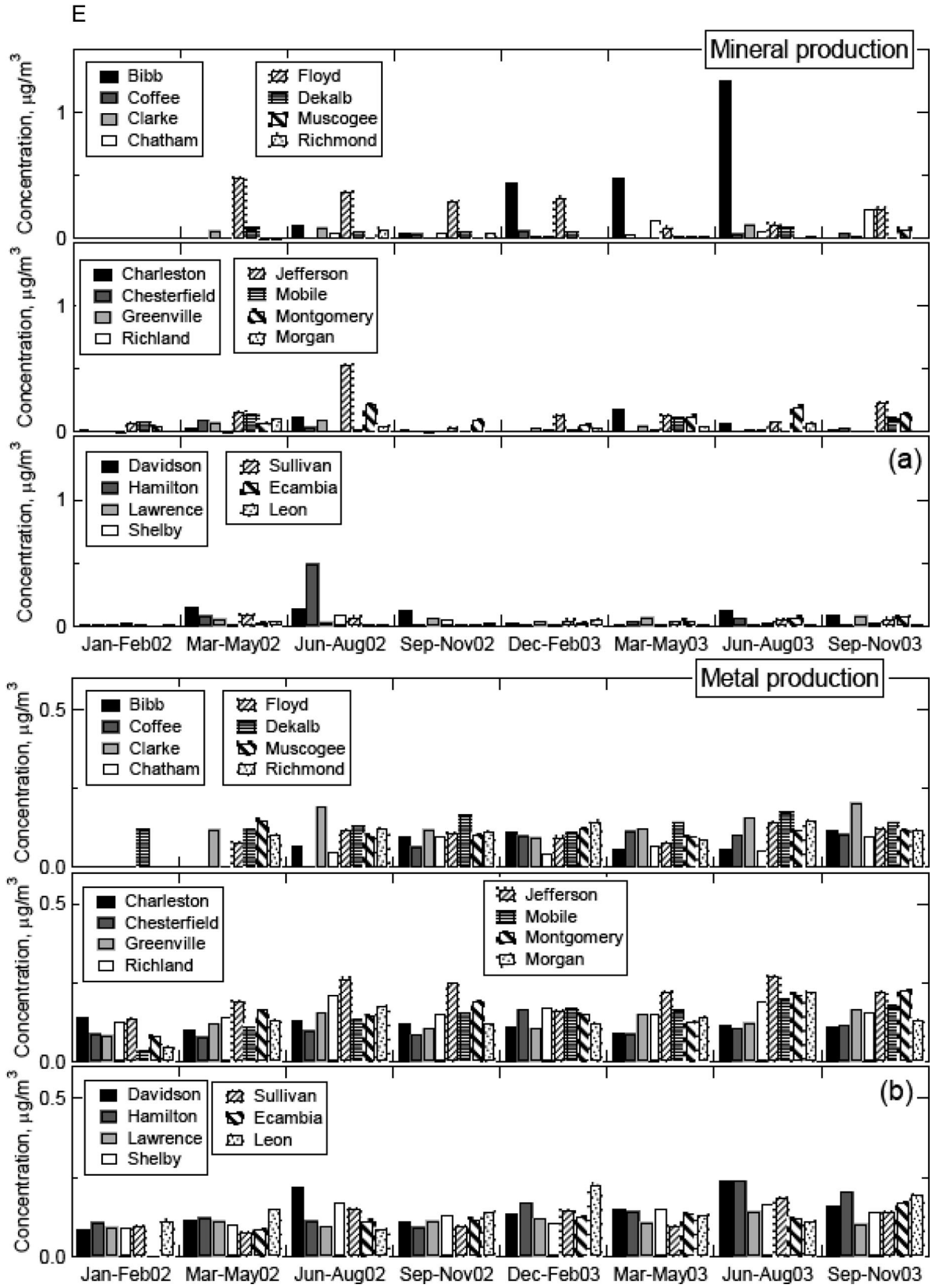

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measurements in Alabama.[48] The source profile for coal combustion was from Chow et al. (2004).[43] Other industrial source profiles were from Sareef (1987).[46] When the reported ambient concentration of trace elements was below the detection limit, it was replaced with a value of half of its detection limit. Uncertainty for each species in ambient data was calculated as 5% of its concentration plus one-third of its detection limit.[55]

RESULTS AND DISCUSSION

CMB source apportionment was done by targeting χ^2 values less than 4 and *r*-square values larger than 0.8 over a period covering January 2002 to November 2003. In general, annual averaged $PM_{2.5}$ concentrations are relatively higher inland than in the coastal areas (Figure 2). Results show that most (>50%) of ambient $PM_{2.5}$ are secondary from photochemical reactions. Motor vehicles and biomass burning are major primary sources in the study area. Dust and industrial sources typically follow in importance.

Source contribution results averaged for three months representing each of the four seasons, winter (December–February), spring (March–May), summer (June–August), and fall (September–November) (Figure 3) show strong seasonality for secondary particles. Sulfate particles (NH_4HSO4 and $(NH_4)_2SO_4$) are higher in the warmer seasons whereas NH_4NO_3 is higher in the colder seasons. Like sulfate particles, SOC is also higher in the warmer seasons when the atmosphere is photochemically more active. Two primary sources, biomass burning and dust, have a strong seasonality. Biomass burning contributes more in the colder seasons when residential wood burning, prescribed burning, and agricultural burning are increased. In contrast, dust is higher in the drier summer season. There is no distinct seasonality for other primary sources.

To address spatial distributions in the impacts from each source category, surface maps were created by using one year averaged source apportionment results (Figure 4) and the inverse distance squared weighted method in ESRI ArcGIS 9.[56] However, the interpolated values depend highly on the availability of source apportionment results at receptor sites, and such source impacts are increasingly uncertain as one is more distant from the observation site. Although less quantitative in between receptors, they are meant to help identify spatial patterns in the Southeast. Furthermore, the impacts are not directly proportional to emissions, but also include the impacts of dispersion. Impacts of very local emissions can be over-emphasized by this approach, but given the density of the monitors, this can still provide a good depiction of the spatial distribution of source impacts. In the northwestern part of the study area, NH_4HSO_4 is relatively lower than $(NH_4)_2SO_4$, but NH_4NO_3 is relatively larger than that in the southeastern area. This indicates that particles in the northwest are more neutralized by NH_3 forming $(NH_4)_2SO_4$, with excess NH_3 producing NH_4NO_3. In contrast, the southeastern area experiences relatively higher NH_4HSO_4 but lower $(NH_4)_2SO_4$ and NH_4NO_3 than the northwestern part, which is indicative of more acidic particles due to less NH_3 neutralization. This is consistent with NH_3 emissions showing relatively higher emissions in the Northwest than in the southeastern area (Figure 5). Motor vehicle source contributions are relatively larger in the more populated areas such as Dekalb (Atlanta), Jefferson

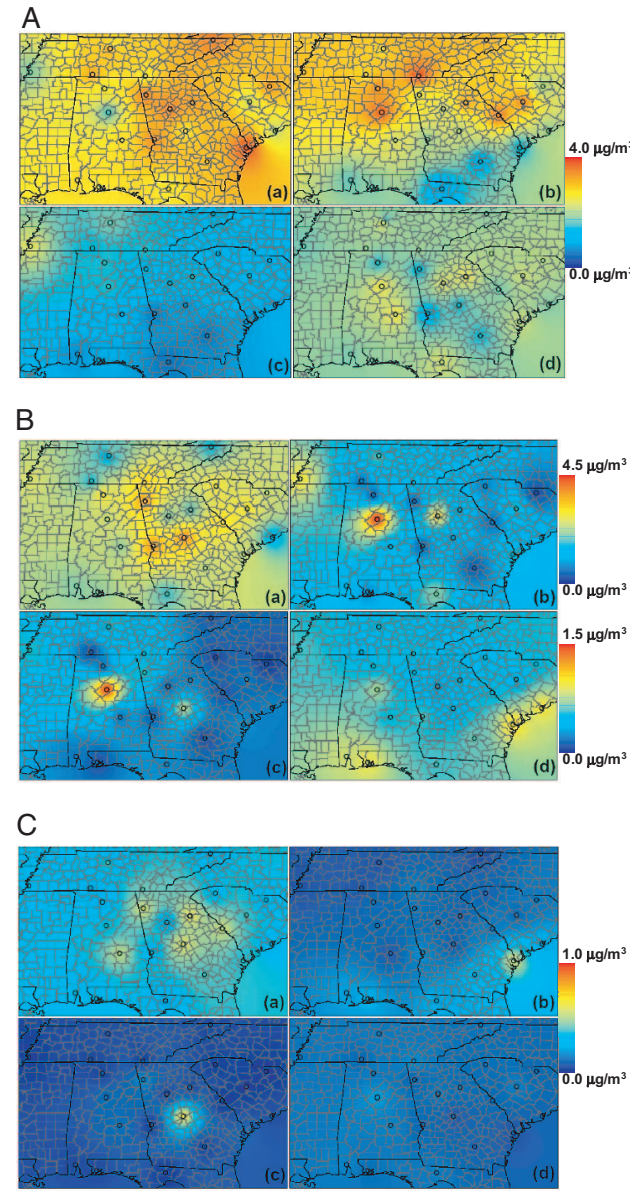

Figure 4. Panel a shows interpolated, spatial distribution of source contributions: (a) NH_4HSO_4, (b) $(NH_4)_2SO_4$, (c) NH_4NO_3, and (d) SOC. No monitors in NC or LA were used in the analysis, so the extrapolations to those states should be regarded as particularly uncertain. Panel b shows spatial distribution of source contributions: (a) biomass burning, (b) motor vehicles, (c) coal combustion, and (d) pulp and paper production. Panel c shows spatial distribution of source contributions: (a) dust, (b) oil combustion, (c) mineral production, and (d) metal production.

(Birmingham), and Shelby (Memphis) whereas biomass burning contributions are larger in the less urbanized areas where burning is more prevalent and actively used in controlled applications for land management purposes.[57] For coal combustion, higher contributions occur in the areas close to source locations and are highest at Jefferson (Birmingham), AL where industrial facilities use coal for fuel. Relatively higher pulp and paper source contribution occurs along the coastal line where pulp and paper mills are located. Oil combustion contribution is also relatively higher along the coast especially at Chatham (Savannah), GA. This corresponds to $PM_{2.5}$ emissions from shipping

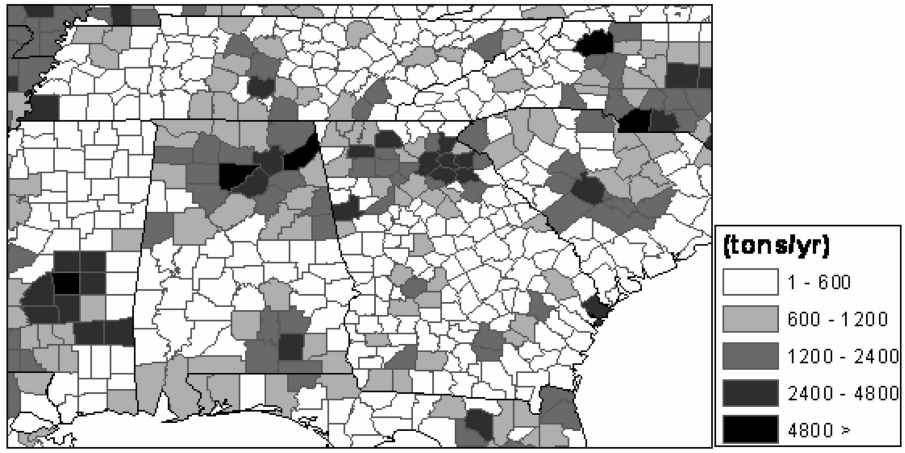

Figure 5. Annual NH_3 county emissions based on EPA 2001 National Emission Inventories.

activities in the coastal port areas because residual oil is used for ships.[58] Mineral production is higher at Bibb, Floyd, and Jefferson. Jefferson County has relatively higher metal production impacts than other sites, but its source contribution is the lowest among the included industrial sources.

Spatiotemporal correlations between all possible pairs of sites were calculated for each source category to understand which sources have more local or regional impact in the study area. In general, the correlations decrease as the distance between sites increases. This trend is distinct for the secondary origin particles (Figure 6), for which the

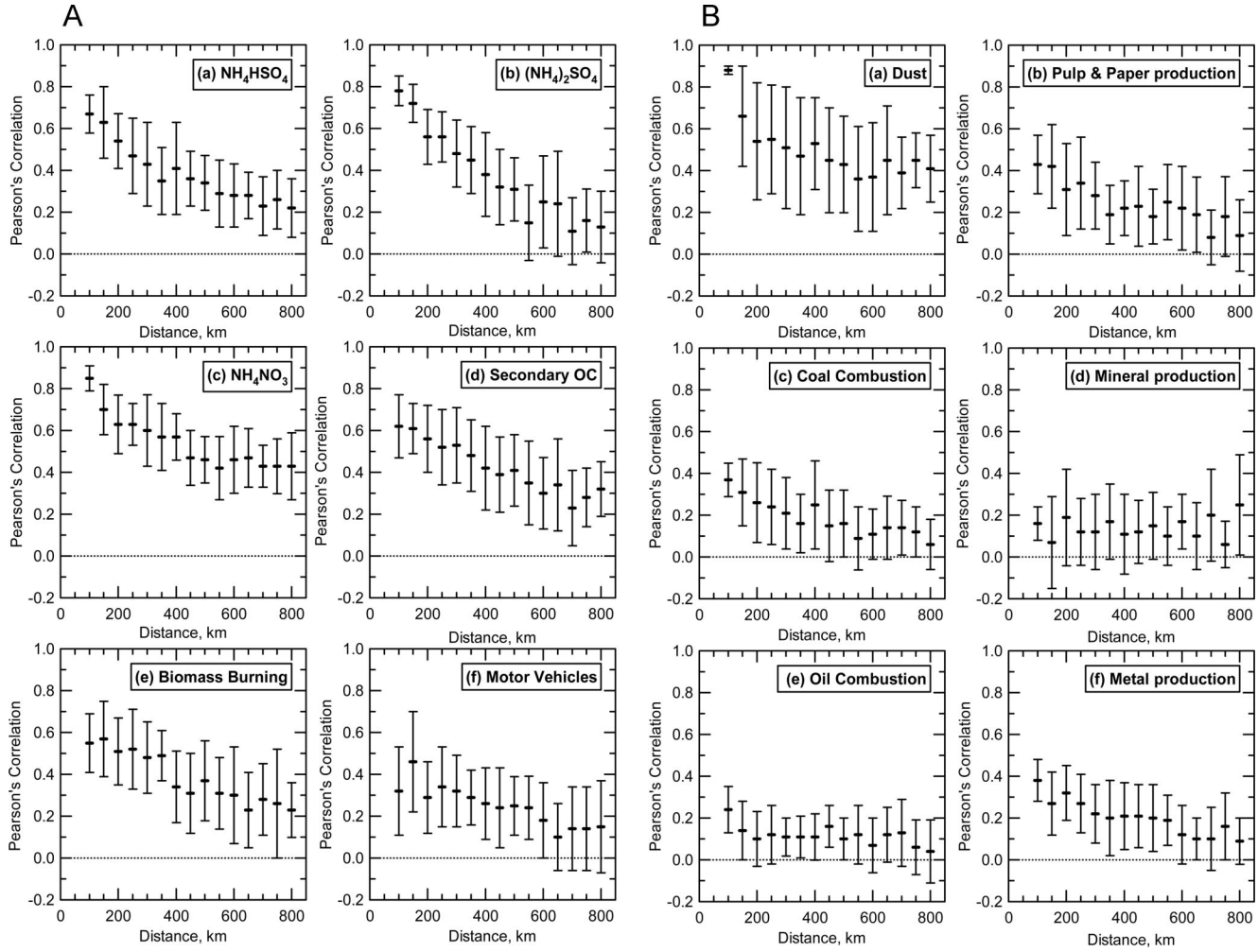

Figure 6. Panel A shows spatial-temporal correlations of source contributions. (a) NH_4HSO_4, (b) $(NH_4)_2SO_4$, (c) NH_4NO_3, (d) SOC, (e) biomass burning, and (f) motor vehicle. Panel B shows spatial-temporal correlations of source contributions. (a) dust, (b) pulp/paper production, (c) coal combustion, (d) mineral production, (e) oil combustion, and (f) metal production.

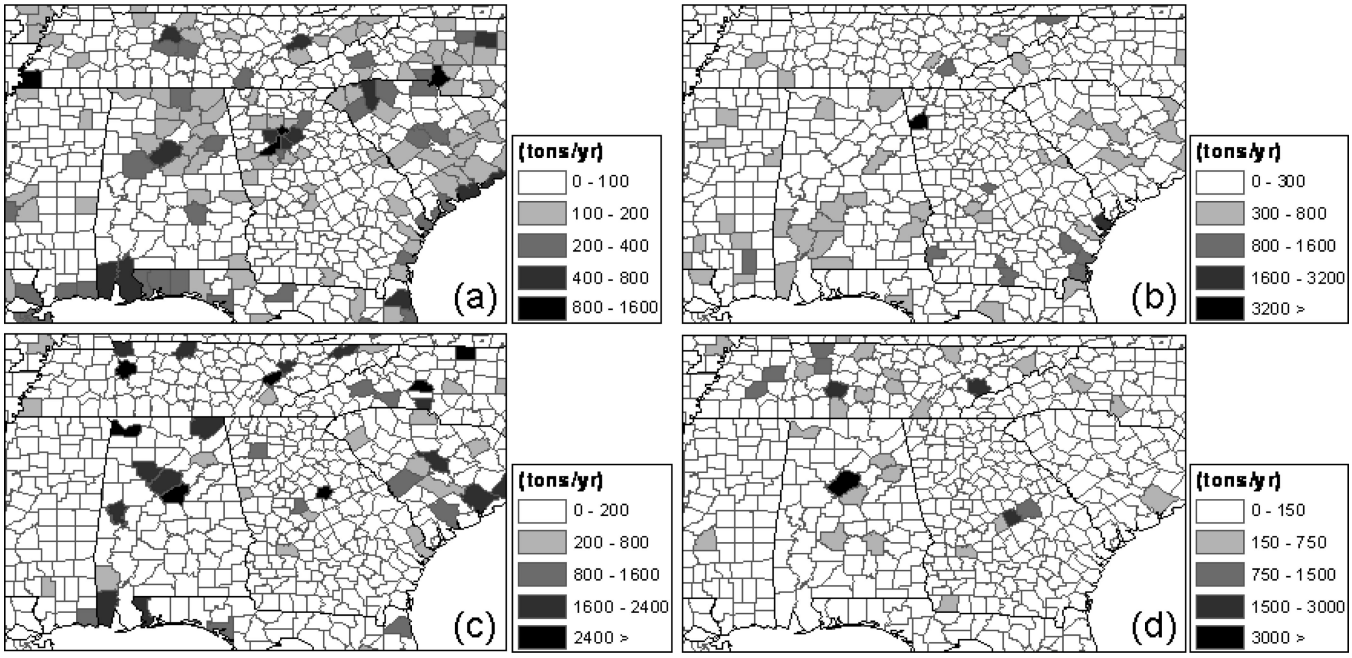

Figure 7. Annual primary $PM_{2.5}$ county emissions based on EPA 2001 National Emission Inventories: (a) motor vehicle, (b) pulp/paper production, (c) coal combustion, and (d) metal production.

correlation coefficients become less than 0.5 once the pair is more than approximately 200–400 km apart, thus showing the regional nature of secondary species. For the primary sources, correlation coefficients are generally small, even for short distances, indicating a local nature of their impacts. Hence, the secondary particles formed from chemical reactions during atmospheric transport and dispersion have better correlation across larger distances than the primary source categories. It suggests that secondary particles underlying atmospheric formation have a more regional character whereas the primary sources are more local.

Source apportionment results consistently show more motor vehicle impacts relative to biomass burning in the urbanized areas but *vice versa* in the less urbanized areas. However, motor vehicle source impacts in Jefferson (Birmingham) are higher than in Dekalb (Atlanta), although $PM_{2.5}$ emission inventories show relatively less motor vehicle emissions in Jefferson than Dekalb (Figure 7a). Jefferson is located in a valley which is surrounded by long parallel mountain ridges, whereas Dekalb is located in more flat terrain.[59] The geographical environment of Jefferson results in less dispersion of pollutants such that the source impact is greater. Source apportionment results suggest that pulp/paper production impacts relatively higher along the coastal area. Pulp/paper production emission inventories also show a similar pattern suggesting relatively higher emissions along the coast (Figure 7b). On the basis of emission inventories, the highest pulp/paper emission occurs in Floyd, GA. However, this is not found from source apportionment. For coal combustion, both source apportionment results and emission inventories show relatively higher impacts on the Tennessee and North Alabama areas (Figure 7c). Metal production impact is the highest at Jefferson, AL, where inventories also suggest the highest emissions (Figure 7d). Source impacts of oil combustion are relatively higher along the coast, especially at Chatham (Savannah). Most commercial ships (~70–80%) use residual oil which contain more contaminants[58] and approximately 80% of ship emissions are concentrated mainly near the shore where the ship traffic density is the highest.[60]

CONCLUSIONS

Source apportionment using CMB receptor model was performed for 24-hr ambient $PM_{2.5}$ measurement data from EPA STN sites in the southeastern United States. Secondary particles formed by atmospheric photochemical reactions make up the majority (>50%) of ambient $PM_{2.5}$ and have strong seasonality. Motor vehicles and biomass burning are the two main primary sources. Motor vehicles are the highest primary source contributor in urban areas whereas biomass burning dominates more in less urbanized areas. Spatiotemporal correlations show that secondary particles are more regionally distributed, and primary particles are more locally. It implies that targeted control strategies can be developed for specific regions on the basis of the most important sources identified and the relative costs of emission reductions. The comparisons with primary $PM_{2.5}$ emission inventories suggest that the source apportionment results support the general characteristics of the inventories. However, there are notable differences to resolve for some sources (e.g., pulp/paper production).

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