PMF and CMB Source Apportionments of PM$_{2.5}$: A comparison

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ABSTRACT: Two commonly used receptor models, Positive Matrix Factorization (PMF) and Chemical Mass Balance (CMB), are applied to 3-year SEARCH measurements at two urban sites (Atlanta, GA and Birmingham, AL) and two rural sites (Yorkville, GA and Centreville, AL). The measurements include fine particle (PM$_{2.5}$) concentrations of inorganic ionic species, trace metals, elemental and organic carbon from January 2000 through December 2002. Source apportionment results using the two methods are analyzed and compared. Conditional probability functions (CPFs) of wind directions are calculated for source factors identified by PMF and CMB in order to investigate the impact of point sources and elucidate differences between the two methods. Source contributions and profiles of secondary sulfate and nitrate factors derived by the two methods agree well, as do corresponding CPF distributions. Secondary OC factor calculated from PMF correlates well with that found using the EC tracer approach in the CMB analysis, while the source contribution of the former is lower. Derived characteristics of other (mostly primary) source factors can be quite different between the two methods. CMB source profiles for motor vehicles are quite different from the PMF factor, although the corresponding source contributions and CPF results show generally good correlations. The coal combustion factors derived by the two methods are poorly correlated and have very different CPF

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distributions. The wood smoke factor also shows large differences, especially at the two urban sites. PMF resolves two dust factors, one for long-range transport and the other for local/industry sources, while the CMB resolves only one dust source since long-range transport was not considered. PMF was able to resolve an industry source with high zinc concentrations while CMB included a cement source, which was unresolved by PMF. There are several reasons that may explain the disagreements between the results using the two methods: (1) the lack of proper markers for additional sources; (2) errors in estimation of source profiles in CMB due to a lack of local-specific profiles or the incompleteness of profiles; (3) the uncertainties in the estimation of secondary organic carbon using EC tracer method; and (4) the tendency of PMF to mix primary and secondary aerosols due in part to atmospheric mixings.

1. Introduction

Particulate matter has been linked with cardiovascular and respiratory problems, including problems leading to premature mortality (Peters et al., 2001; Peel et al., 2002; Pope et al., 2002; Metzger et al., 2004). PM$_{2.5}$ is also the main anthropogenic cause of pollution-related visibility impairment (Milne et al., 1982), and can contain constituents leading to acid deposition (NADP, 1993). PM$_{2.5}$ is emitted directly from the sources or forms in the atmosphere through reactions of precursor gases, such as nitrogen oxides (NO$_x$), sulfur oxides (SO$_x$), and certain organic vapors. Cost effective air quality management planning relies on identifying how different sources impact pollutant concentrations.

Receptor models, which attribute observed concentrations to sources through statistical and/or meteorological interpretation of data often yield useful insights on the sources of aerosols (Hopke, 2003). Such models are generally based on the observed mass concentrations and appropriate use of mass balance. Various methods have been developed for this purpose.
If the number and nature of the sources in the region are known, the mass contribution of each source to each sample can be estimated using the Chemical Mass Balance (CMB) model (Chow et al., 1991; Schauer et al., 1996). When source information is largely unknown, factor analysis is generally applied. Three factor analysis approaches that have been often used in recent years are UNMIX, Multilinear Engine (ME) and Positive Matrix Factorization (PMF). We use the PMF approach in this work. It has been applied to a number of aerosol data sets (e.g., Polissar et al., 1998, Lee et al., 1999; Liu et al., 2003). Similarities between PMF and CMB include using least-squares fitting to minimize the differences between measured and estimated concentrations. CMB makes use of an effective variance least-squares fitting by using source composition data as independent variables and ambient air quality data as dependent variables. PMF uses an alternating least-square method and only needs ambient measurement data without a priori assumptions of the factor profiles. CMB incorporates uncertainties from both ambient measurement and source profile data while PMF estimates the profile uncertainties based on those from the ambient measurement data.

In this work, both source apportionment methods were applied to four data sets, two urban (Atlanta, GA and Birmingham, AL) and two rural (Yorkville, GA and Centreville, AL). Data used include 3-year measurements from January 2000 to December 2002. Source profiles (as used by CMB) and factors (as developed by PMF), as well as source contributions from the two analyses were compared in order to investigate the usefulness and limitations of each method.

2. Method

2.1 Measurement data
PM$_{2.5}$ composition data analyzed in this study consist of measurements taken at two urban-rural pairs in Alabama (North Birmingham [BHM] and Centreville [CTR]), and Georgia (Atlanta [JST] and Yorkville [YRK]). These sites are operated by the Southeastern Aerosol Research and Characterization Study (SEARCH) (Hansen et al., 2003). Twenty-four hour integrated PM$_{2.5}$ samples were collected daily at the JST site. PM$_{2.5}$ samples were collected every third day at the other sites. Samples were collected using particulate composition monitors (PCM, Atmospheric Research and Analysis, Inc., Durham, NC) that have three sampling lines (air flow rate 16.7 l/min) with inlets 5 m above ground. More detailed descriptions can be found elsewhere (e.g., Hanson et al., 2003; Liu et al., 2005a).

A total of 932 samples for the JST site, 336 samples for the BHM site, 347 samples for the YRK site and 338 samples for the CTR site were obtained and analyzed, covering the time period from January 2000 through December 2002. For each sample, concentrations of the following 19 chemical species were usually available: SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, EC, OC (OC was calculated as OC1+OC2+OC3+OC4+OP and EC as EC1+EC2+EC3-OP), As, Ba, Br, Cu, Mn, Pb, Se, Ti, Zn, Al, Si, K, Ca, and Fe, although there are occasional “missing data” (no reported measurements) for one or more species. Total PM$_{2.5}$ mass concentrations for each day, analytical uncertainty and detection limit for each chemical species were also obtained.

2.2 PMF

PMF (Paatero and Tapper, 1994; Paatero, 1997) was used to analyze PM$_{2.5}$ data at the four sites. In this work, missing data were replaced by the geometric mean of corresponding species and four times of geometric mean as the corresponding error estimates (Polissar et al., 1998). Half of the detection limit was used for the values below the detection limit and 5/6 of the detection limit was used for the corresponding error estimate (Polissar et al., 1998).
With the total PM$_{2.5}$ mass concentration measured for each sample, multiple linear regression (MLR) was performed to regress the mass concentration against the factor scores obtained from PMF. Because of the uncertainties introduced by the measurement matrix, PMF results always have a portion of unexplained variation. Mass concentrations excluding the unexplained variation portion from G factors (factor contributions) were used to regress the factor scores to obtain the quantitative factor contributions for each resolved factor.

2.3 CMB

EPA’s CMB 8.0 model, using the effective variance weighted least-squares fitting, was applied to calculate source contributions to PM$_{2.5}$ on a daily basis (Watson, 2001). All performance diagnostic criteria were met for each calculation. For example, the ratios of the calculated to measured concentrations of fitting species, R-square, Chi-square, percent of attribute mass are 0.5 to 2.0, 0.8 to 1.0, 0 to 4.0, and 100±20% respectively, each time.

The same data treatments of missing and below detection limit data in PMF analysis were used in the CMB analysis. Source profiles for the primary sources selected in the CMB analysis included motor vehicle (Watson et al., 1998), wood burning (Watson et al., 1998), coal combustion from power plants (Chow et al., 2004), Alabama road dust (Cooper et al., 1981), and cement kilns (Chow et al., 2004). The composites of light duty gasoline and heavy duty diesel vehicles are created by weighing emission rates and four different profiles using weights derived from the emission inventories for Georgia and Alabama. The wood burning source profile is also created by weighting emission rates of three wood burning source measurements, soft-wood fireplace, hard-wood fireplace, and woodstove (Zielinska et al., 1998). Ammonium bisulfate (NH$_4$HSO$_4$), ammonium sulfate ((NH$_4$)$_2$SO$_4$), and ammonium nitrate (NH$_4$NO$_3$) were used as inorganic secondary sources.
Primary organic carbon in the CMB analysis was calculated by using the EC tracer approach (Turpin et al., 1991). First, the primary OC to elemental carbon (OC/EC) ratio was estimated and then primary OC was calculated by multiplying OC/EC ratio to EC assuming that all EC is primary. If the estimated primary OC was higher than measured OC, measured OC was used as primary OC.

2.4 Conditional probability function

CPF (Ashbaugh et al., 1985; Kim et al., 2003) estimates the likelihood for a source factor identified by CMB or PMF to originate from a given wind direction. Hourly wind data are used. The CPF is defined as,

\[ CPF_{\Delta \theta} = \frac{m_{\Delta \theta}}{n_{\Delta \theta}} \]  

(1)

where \( m_{\Delta \theta} \) is the number of occurrences of the source contribution exceeding a threshold criterion from wind sector \( \Delta \theta \), and \( n_{\Delta \theta} \) is the total number of data from the same wind sector. In this study, 36 sectors were used (10° in each sector). The threshold criterion of the upper 25th percentile in the source contribution was chosen to define the directionality of a source factor. Sources are likely to originate from the directions that have high conditional probability values. Calm wind (<1m/s) periods were excluded from this analysis.

3. Results

The aforementioned seven source profiles were used in the CMB analysis to derive the source contributions at the four sites. PMF was able to resolve eight and seven factors for the two urban sites and the two rural sites, respectively. PMF factors common to all locations included: (1) secondary sulfate dominated by high concentrations of sulfate and ammonium with a strong seasonal variation peaking in summer; (2) nitrate and the associated ammonium with a seasonal
maximum in winter; (3) “coal combustion/other” factor with the presence of sulfate, EC, OC, and Se; (4) soil, with high levels of Al, Ca, Fe, K, Si and Ti; (5) wood smoke with high concentrations of EC, OC and K; and (6) an industry/dust factor with elevated levels of Ca, Fe, K, Si and Ti. A motor vehicle factor with high concentrations of EC and OC and the presence of some soil dust components was found at the urban sites, but not at the two rural sites. One similar industry factor with high zinc concentrations was found in each site in the PMF analysis.

Secondary sulfate was a dominant source factor in urban and rural areas. CMB had two source profiles to represent secondary sulfate (NH₄HSO₄ and (NH₄)₂SO₄). PMF resolved factor profiles are compatible with the CMB profiles at all four sites (Figure 1) except that OC and small amounts of EC are associated with this factor in the PMF results. In PMF, the OC and EC components reflect the effects of mixing of primary and secondary pollutants while CMB by definition is not affected by mixing. The resolved PMF factor therefore does not represent a single pure source. The OC association implies that secondary organic aerosol formation coincides with the secondary sulfate formation while the small EC content likely reflects an increase of sulfate and EC concentrations during stagnant conditions. In the PMF sulfate factor, molar ratios of ammonium to sulfate were 2.3, 2.0, 2.1 and 1.6 for the JST, YRK, BHM, and CTR site, respectively. The ratios suggest that sulfate is present primarily as ammonium sulfate at these four receptor sites, although sulfate at CTR is probably not fully neutralized. In the CMB results, NH₄HSO₄ had low source contributions at the JST and YRK sites (<10% of secondary sulfate) and relatively high source contributions at the BHM (30%) and CTR (40%) sites. Therefore, the two approaches agreed except at the BHM site. The resolved source contributions from PMF and CMB model are well correlated (Figure 1); the corresponding CPF results are
also in agreement (Figure 2). Average source contributions from both models agree well, although the PMF value is lower at JST (Table 2).

PMF resolved a nitrate factor, which corresponds to the CMB NH$_4$NO$_3$ profile. The source contributions from the two methods are generally in good agreement (Figure 3). Sulfate and some OC are mixed in the PMF profile, likely arising from concurrent oxidations of NO$_2$, SO$_2$, and VOCs. The PMF and CMB source contributions are well correlated and CPF distributions are similar (Figure 4). However, the PMF source contributions tend to be lower except at the BHM site (Table 2).

The CMB source profiles and the PMF factor associated with wood burning are in relatively good agreement, with high concentrations of OC, EC, and K. However, PMF factor profiles are mixed with some sulfate at the two urban sites. For the two rural sites, source contributions agree better between the models (Figure 5). Again, the non-locally measured source profiles (the CMB wood burning source profiles were obtained in Colorado) may, in part, contribute the differences between CMB and PMF. Furthermore, Liu et al. [2005b] found that the PMF resolved wood burning factors at urban sites peaked in winter while those at the rural sites peaked in spring. Winter residential burning appears to be a major contributor at urban sites while agricultural burning in spring appears to dominate at rural sites. The PMF-CPF and CMB-CPF distributions also reflect the urban-rural differences between the two models (Figure 6). At the JST site, the PMF-CPF distribution shows the impact mainly coming from the south while the CMB-CPF distribution shows the impact coming from the south, northeast and northwest. For YRK, source locations are similar. Average source contributions were in good agreement at JST and YRK, while they are very different for the BHM and CTR sites. This “source” has larger contributions in rural areas than in urban areas (Table 2).
The PMF coal combustion factor has strong signals of sulfate, ammonium, EC, OC, and Se at all sites (Figure 7). The CMB source profile has higher levels of elements associated with dust. This difference may be due, in part, to a lack of locally appropriate source profile for use in the CMB, or PMF is “losing” the other elements to a dust factor. Compared to CMB results, PMF has higher source contributions at all four sites. This is due largely to the OC content mixed in this factor. There is increasing evidence that highly acidic particulate matter can catalyze the formation of secondary organic aerosol, and soluble organics may be absorbed in water-laden aerosols (Jang et al., 2002). Thus, the increased OC associated with these particles could arise either by increased condensation of urban OC onto the particles or it could represent acid catalyzed conversion of VOCs including isoprene. The PMF-CPF distributions at YRK and JST sites suggest that this factor has a common source for the two sites, northwest of JST and north of Yorkville. For the CMB-CPF results, this source comes from different locations for the two sites, south-north direction for JST and northwest and south for YRK (Figure 8).

A motor vehicle factor is resolved only at the two urban sites by PMF. Motor vehicle contributions estimated by CMB at the two rural sites are much smaller (~1/10) than the urban sites. We compare the PMF and CMB motor vehicle factors at the two urban sites. This factor has high concentrations of EC and OC. However, the OC/EC ratios in the source profiles assigned in CMB and the factors calculated by PMF are markedly different. The ratios are about 2 in PMF as compared to 0.5 in CMB (Figure 9). The higher OC concentrations in the PMF factor could arise from condensation of secondary OC onto the particles. It is also possible that the EC/OC ratio specified in the CMB profile is in error. Liu et al. [2005b] found that the PMF resolved gasoline and diesel factor profiles agreed with those measured by Cao et al., [2005] but differed from an earlier study by Watson et al., [1994]. Changes in fuel composition could be
part of the reason, though there is considerable variability in emission profile measurements as well (e.g. Cocker et al., 2005). Source contributions from both models are well correlated; the CPF results also agree well. However, PMF estimated higher source contributions for this factor due to the calculated higher OC/EC ratio.

PMF resolved two dust factors. We refer to the first one as “dust” and the second one as “industry/dust”. The dust factor is rich in Si, Al, K, Ca, and Fe, and is associated with some secondary OC and sulfate. There is evidence that this factor comes from long range transport of desert dust. The source contribution peaked in April 2001, July 2001 and July 2002. These are likely intercontinental dust transport events. The April 2001 event is due to transport from Asia (EPA, 2003), while the July episodes in 2001 and 2002 are probably transported from Saharan deserts (Prospero, 2001). The Industry/dust factor has high concentrations of Si, K, Ca, and Fe coupled with secondary OC and sulfate. The EC content in this factor may come from local industrial sources. CMB includes only one dust profile for paved road dust. Compared to the PMF dust factor, the CMB profile has higher concentrations of Si. A cement source is also resolved by CMB at all of the four sites. From the time series comparison (Figures 11 and 12) and the CPF distributions (Figures 13 and 14), the long-range transport dust resolved by PMF is probably distributed to the CMB dust and cement sources.

Major OC sources are wood smoke, motor vehicle and secondary production at urban sites, and wood smoke and secondary production at rural sites. The average OC concentrations apportioned to each source are compared between the two models (Figure 15). At the two urban sites, PMF apportioned more OC to the motor vehicle factor while CMB apportioned more OC to the wood smoke source. At the two rural sites, PMF apportioned more OC to the wood smoke factor. PMF SOC is lower than that found using the EC tracer approach used in CMB analysis.
PMF secondary OC was calculated from the sum of OC fractions mixed in the PMF sulfate, and nitrate factors and unexplained variations. Yuan et al. (2005) previously applied PMF to estimate secondary OC production in Hong Kong and found that PMF estimated secondary OC is lower than obtained using the EC tracer method. The large uncertainty of the EC tracer method arises from the assumption that a single OC/EC ratio can represent a mixture of primary sources varying in time and space, while secondary OC mixed into the primary source factors in the PMF analysis may lead to underestimation. There is also evidence that meat cooking and natural gas contribute significant portion of OC in the Southeast (Zheng et al., 2002, Jaemeen et al., 2005). However, both PMF and CMB can not resolve such sources because of the lack of additional markers and source profiles. These sources must have been distributed among the sources resolved above to an unknown degree. This may also cause some differences between the two models.

4. Conclusions

PMF and CMB methods were applied to 3-year measurements at four monitoring sites in GA and AL to identify major source factors and contributions to PM$_{2.5}$. Primary organic carbon concentrations calculated by the EC tracer approach are used in the CMB analysis. For comparison purposes, corresponding CPF values were calculated using source contributions estimated by PMF and CMB coupled with wind direction measurements at these sites. There is relatively good agreement between the PMF and CMB-derived secondary aerosol source contributions and the resulting CPFs. Both approaches found that secondary sulfate and secondary nitrate account for the majority of measured PM$_{2.5}$ mass at the Southeast, with similar day to day variations. Secondary OC calculated from PMF is well correlated with that from CMB using the EC tracer approach, although the values are typically lower than the latter.
Differences between PMF computed primary source factor contributions and CMB source profiles are much larger than secondary source factors, resulting in the differences in calculated source attributions. The OC/EC ratios in the motor vehicle profiles assigned in CMB and calculated by PMF are markedly different, 2 in PMF as compared to 0.5 in CMB. As a result, higher source contributions are calculated in PMF. PMF and CMB wood burning profiles are in relatively good agreement, with high concentrations of OC, EC, and K. The PMF coal combustion factor has strong signals of sulfate, ammonium, EC, OC, and Se at all sites. The CMB source profile has higher levels of elements associated with dust. PMF was able to resolve two dust factors, one due to long-range transport from Sahara and Asia and the other representing local sources. CMB resolved only one dust source factor since long-range transport is not considered in the CMB source profiles. PMF resolved an industry factor with high zinc concentrations while CMB resolved a cement source.

Several factors may have contributed to the different apportionment resulting using PMF and CMB. First, there are no appropriate markers to apportion the emissions from meat cooking and natural gas. Secondly, the source profiles in CMB may be inaccurate due to (a) a lack of locally available source profiles (the vegetative burning and coal combustion source profiles used in CMB were based on measurements in Colorado and Texas, respectively), (b) the uncertainties in the mobile source profiles, and (c) a lack of long range transport dust profile. Thirdly, there appears to be a high bias in the secondary organic carbon contributions estimated using the EC tracer method in the CMB analysis compared to the PMF results. Lastly, PMF has a tendency to mix primary and secondary aerosols due in part to atmospheric mixing. In addition, PMF and CMB results are affected by measurement uncertainties and missing data. Neither PMF nor CMB accounts for the seasonal variations of source profiles.
Acknowledgement. This study was supported by the Southern Company and US EPA under grant RD-83215901.

References:


Peel, J., Tolbert, P., Klein, M., Metzger, K., Flanders, W. D., Todd, K., Mulholland, J., Ryan, P. B., Frumkin, H. (2002). Ambient air pollution and respiratory emergency department
Table 1. PMF Factor- CMB source contribution correlation coefficients ($R^2$) at the four sites

<table>
<thead>
<tr>
<th>Source</th>
<th>JST</th>
<th>BHM</th>
<th>YRK</th>
<th>CTR</th>
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<tbody>
<tr>
<td>Sulfate</td>
<td>0.99</td>
<td>0.93</td>
<td>0.97</td>
<td>0.97</td>
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<tr>
<td>Nitrate</td>
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<td>0.91</td>
<td>0.99</td>
<td>0.98</td>
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<tr>
<td>Coal</td>
<td>0.02</td>
<td>0.04</td>
<td>0.01</td>
<td>0.0003</td>
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<td>Wood smoke</td>
<td>0.27</td>
<td>0.35</td>
<td>0.73</td>
<td>0.72</td>
</tr>
<tr>
<td>Motor vehicle</td>
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<td>0.89</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>Dust</td>
<td>0.15</td>
<td>0.2</td>
<td>0.42</td>
<td>0.5</td>
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<tr>
<td>SEOC</td>
<td>0.64</td>
<td>0.45</td>
<td>0.49</td>
<td>0.37</td>
</tr>
<tr>
<td>CMB cement vs. PMF industry / dust</td>
<td>0.12</td>
<td>0.42</td>
<td>0.17</td>
<td>0.19</td>
</tr>
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Table 2. The comparison of average factor contributions (%) to PM$_{2.5}$ mass concentrations between CMB and PMF at the four sites

<table>
<thead>
<tr>
<th>PMF</th>
<th>CMB</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>JST</td>
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<tr>
<td>(NH$_4$)$_2$SO$_4$</td>
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<td>Sulfate</td>
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<td>Nitrate</td>
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<tr>
<td>SEOC</td>
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<tr>
<td>undetermined</td>
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</tr>
</tbody>
</table>
Figure captions

Figure 1. Comparison of source profiles and contributions of secondary sulfate resolved from PMF and CMB at the four sites. The top panel shows the source profiles and the bottom panel shows the correlations the source contributions resolved at the four sites.

Figure 2. Comparison of sulfate CPF distributions between PMF and CMB results. The top panel shows the CPF distributions for JST site and the bottom panel shows the CPF distributions for YRK site.

Figure 3. Same as Figure 1 but for secondary nitrate.

Figure 4. Same as Figure 2 but for secondary nitrate.

Figure 5. Same as Figure 1 but for wood smoke.

Figure 6. Same as Figure 2 but for wood smoke.

Figure 7. Same as Figure 1 but for coal combustion.

Figure 8. Same as Figure 2 but for coal combustion.

Figure 9. Comparison of source profiles and contributions of motor vehicle from PMF and CMB at the JST and BHM sites. The top panels show the source profiles, the middle panel shows the correlations of the source contributions, and the bottom panel shows the CPF distributions at JST.

Figure 10. Comparison of source profiles of dust and “Industry /dust” from PMF and dust and cement from CMB at the four sites.

Figure 11. Comparison of source contributions of dust and “industry /dust” from PMF and dust and cement from the CMB at JST and BHM sites.

Figure 12. Comparison of source contributions of dust and “industry /dust” from PMF and dust and cement from CMB at the YRK and CTR sites.
Figure 13. Comparison of the dust factor CPF distributions between PMF and CMB. The top panel shows the CPF distributions for JST and the bottom panel shows the CPF distributions for YRK.

Figure 14. Comparison of industry/dust factor PMF-CPF and cement PMF-CPF distributions. The top panel shows the CPF distributions for JST and the bottom panel shows the CPF distributions for YRK.

Figure 15. Comparison of PMF and CMB averaged OC source contributions for the four sites.
Figure 1
Figure 2
Figure 3
Figure 5
Figure 6
Figure 7
Figure 8
Figure 9
Figure 10
Figure 11
Figure 12
Figure 13
Figure 14
Figure 15