Evaluation of optimization methods for solving the receptor model for chemical mass balance

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Abstract: The chemical mass balance (CMB 8.2) model has been extensively used in order to determine source contribution for particulate matters (size diameters less than 10 and 2.5 µm) in air quality analysis. A comparison of the source contribution estimated from three CMB models was realized through optimization techniques, such as ‘fmincon’ (CMB–fmincon) and genetic algorithm (CMB–GA) using MATLAB. The proposed approach was validated using a San Joaquin Valley Air Quality Study (SJVAQS) California Fresno and Bakersfield PM$_{10}$ and PM$_{2.5}$ followed with Oregon PM$_{10}$ data. The source contribution estimated from CMB–GA was better in source interpretation in comparison with CMB 8.2 and CMB–fmincon. The performance accuracies of three CMB approaches were validated using $R^2$, reduced $\chi^2$ and percentage mass tests. The $R^2$ (0.90, 0.67 and 0.81, 0.83), $\chi^2$ (0.36, 0.66 and 0.65, 0.43) and percentage mass (67.36, 55.03 and 94.24 %, 74.85 %) of CMB–GA showed high correlation for PM$_{10}$, PM$_{2.5}$, Fresno and Bakersfield data, respectively. To make a complete decision, the proposed methodology was bench marked with Portland, Oregon PM$_{10}$ data with the best fit with $R^2$ (0.99), $\chi^2$ (1.6) and percentage mass (94.4 %) from CMB–GA. Therefore, the study revealed that CMB with genetic algorithm optimization method exhibiting better stability in determining the source contributions.

Keywords: receptor model; chemical mass balance; source contribution; source profiles; genetic algorithm.

INTRODUCTION

Air pollution is a major concern in the current century due to population exposure, urbanization and industrialization. The concentration level of particulate matter (particles with aerodynamic diameters less than 10 and 2.5 µm) in the urban environment remains a serious problem.\(^1\)\(^-\)\(^3\) The term particulate matter (PM$_{10}$ and PM$_{2.5}$) is used to describe solid or liquid particles that are airborne...
and dispersed. Particles vary in number, size, shape, surface area, chemical composition, solubility and origin across both space and time. Particulate matter originates from a variety of natural and anthropogenic sources and possesses a range of morphological, physical, chemical and thermodynamic properties. Emissions of mineral particulate matter adversely impact on environmental quality in mining regions, transport regions, and even on a global scale. Various anthropogenic (traffic, power plants, biomass burning, etc.) and natural sources (forest fires, soil re-suspension, etc.) emit primary particulate matters (PM$_{10}$ and PM$_{2.5}$) and gaseous pollutants such as SO$_2$, NO$_x$, NH$_3$ and VOC directly into the atmosphere. Secondary particles, formed by transformation of these primary emissions, contribute to the concentrations of ambient particulate matter, which cause adverse effects on human health. Industrialization patterns changed due to stringent air quality standards with many heavily polluting industries moving from developed countries.

Source identification of particulate matter is one of the key components in air quality management planning. Apportionment studies were attempted to develop and implement air pollution control strategies in many urban areas across the world. Receptor models are widely used to estimate the source contribution of construction activities, fossil fuel combustion, traffic re-suspension, geologic, motor vehicle exhaust, vegetative burning to ambient air pollution. The CMB model combines the chemical and physical characteristics of particles or gases measured at the sources and the receptors to quantify the source contribution to the receptor. The CMB enables the source contributions of ambient PM$_{10}$ and PM$_{2.5}$ to be determined through effective-variance least squares regression, weighted least square regression and the method of moments. Source apportionment (SA) of PM using robotic chemical mass balance (RCMB) reduces the uncertainty due to the human judgment through the best-fit combination of source profiles used as input data. Quantification of uncertainty in RCMB using the traditional Monte Carlo approach and polynomial chaos method were also proposed. Uncertainties in the input variable used to solve the chemical mass balance are the receptor concentration uncertainty and source profile uncertainty. The United States Environmental Protection Agency (USEPA) developed the tool CMB8.2 that resolves using both the uncertainties to obtain the source contribution at the receptor locations. The combined CMB and multivariate source apportionment methods, such as positive matrix factorization (PMF) and the Unmix model, has been widely used for the refined source contribution and source profile estimation in air quality research.

The current research article compares the source contribution results of CMB8.2, CMB–fmincon and CMB–GA. The difference in the estimation of the source contributions by the three approaches were illustrated using San Joaquin Valley Air Quality Study (SJVAQS) of Fresno and Bakersfield, the PM$_{10}$ and
PM$_{2.5}$ data were taken from Chow et al. (1992 and 1993). These data were collected every six days between June 1988 and 1989. A total of 35 and 49 observations of PM$_{2.5}$ and PM$_{10}$, respectively, from the Fresno site and 48 and 33 observations of PM$_{2.5}$ and PM$_{10}$ from Bakersfield sites were respectively obtained. The profile data of ten different sources, such as paved road dust, vegetative burning, crude oil combustion, motor vehicles, lime stone (construction), marine, ammonium sulfate, ammonium nitrate, secondary organic carbon (SOC) and sodium nitrate are available in the literature. The proposed methodology were validated through Portland, Oregon PM$_{10}$ data with marine, urban dust, auto exhaust and residual oil combustion as possible sources of emission. Source contribution estimates from CMB8.2, CMB–fmincon and CMB–GA models were used to predict the receptor concentration (C$_{pre}$) data. The percentage error between the experimental (c$_{exp}$) and predicted concentrations (c$_{pre}$) were compared using the statistical approach of $R^2$, $\chi^2$ and percentage mass to validate the effect of uncertainty and optimization solvers in the three CMB models.

EXPERIMENTAL

CMB receptor model

The CMB receptor model expresses the concentrations of different chemical species ($c_{i\times1}$ / µg m$^{-3}$) measured at a monitoring site (or receptor) as a linear sum of products of the source profile ($F_{i\times j}$ / µg µg$^{-1}$) and source contribution ($S_{j\times1}$ / µg m$^{-3}$):

$$c_{i(\delta i)} = \sum F_{i(\delta j)}S_{j(\delta i)}$$

where $i$ is the number of species measured; $j$ is the number of source categories for one receptor sample. The mass fraction of the emissions from each source type is known as the source profile, µg µg$^{-1}$. Profiles are measured on samples from these sources at times and locations to represent emission compositions, µg m$^{-3}$ while receptor measurements are made. The basic assumptions of CMB model are: 1) compositions of source emissions are constant over the period of ambient and source sampling; 2) no reaction between the chemical species (i.e., they add linearly); 3) all sources with a potential for contributing to the receptor have are identified and have had their emissions characterized; 4) the number of sources or source categories is less than or equal to the number of species; 5) the source profiles are linearly independent of each other; 6) measurement uncertainties are random, uncorrelated, and normally distributed. CMB quantifies contributions from chemically distinct source-types rather than contributions from individual emitters. Sources with similar chemical and physical properties cannot be distinguished from each other by CMB. The CMB model calculates source contribution estimates for each individual ambient sample. The combination of source profiles that best explains the ambient measurements may differ from one sample to the next owing to differences in emission rates.

CMB 8.2

CMB software version 8.2 was developed by the United States Environmental Protection Agency (USEPA). The input to the software contains one day or average receptor concentration, µg m$^{-3}$, data and measured source profile of the possible sources of air pollution at the locality and their corresponding uncertainties. The output of the model is source contribution
to air pollution, µg m$^{-3}$. Performance measures for the least squares calculation in CMB 8.2 are $R^2$, reduced $\chi^2$ and percent mass. The $\chi^2$ is the weighted sum of the squares of the differences between the measured ($c_{\text{exp}}$) and calculated ($c_{\text{pre}}$) fitting species concentrations:

$$\chi^2 = \frac{1}{I-J} \sum_{i=1}^{I} \left( \frac{c_i - \sum_{j=1}^{J} F_{ij} S_j}{V_{ii}} \right)^2$$  \hspace{1cm} (2)

The weighting, $V_{ii}$, is inversely proportional to the squares of the uncertainty in the source profiles and ambient data for each species.

Ideally, there should be no difference between the calculated and measured concentrations of the species and $\chi^2$ would equal zero. A value less than 1 indicates a very good fit to the data, while values between 1 and 2 are acceptable. $\chi^2$ values greater than 4 indicate that concentrations of one or more species are not well explained by the source contribution estimates.

The percent mass can be expressed by Eq. (3), the percent ratio of the sum of the source contribution estimated by the model to the measured mass concentration:

$$\text{Percent mass} = \frac{100 \left( \sum_{j=1}^{J} S_j \right)}{C_t}$$  \hspace{1cm} (3)

where $C_t$ is the total measured mass.

Percentage mass should equal 100 %, although values ranging from 80 to 120 % are acceptable. If the measured mass is very low (<5 to 10 µg m$^{-3}$), the percent mass may be outside this range because the uncertainty in the mass measurement is of the order of 1 to 2 µg m$^{-3}$.

$R^2$ is the fraction of the variance in the measured concentrations that is explained by the variance in the calculated concentrations of the species:

$$R^2 = 1 - \frac{(I-J) \chi^2}{\sum_{j=1}^{J} c_j^2 / V_{jj}}$$  \hspace{1cm} (4)

$R^2$ is determined by linear regression of the measured vs. model-calculated values for the fitted species. The value of $R^2$ ranges from 0 to 1.0. The closer the value is to 1.0, the better is the source contribution estimates explaining the measured concentrations. When $R^2$ is less than 0.8, the source contribution estimates do not explain the observations with the fitting source profiles and/or species very well. The effective variance solution is derived by minimizing the weighted sums of the squares of the differences between the measured and calculated values of $c_i$ and the measured values of $F_{ij}$. The ambient data for 26 species from PM$_{10}$ and PM$_{2.5}$ as well as the profiles and uncertainties for ten sources are available. The constrained optimization routine in MATLAB R2008a ‘fmincon’ were used to optimize the source contributions and thus the difference between $c_{\text{pre}}$ and $c_{\text{exp}}$ of the species involved in this problem. The objective was to minimize the sum of the squares of differences between the experimental ($c_{\text{exp}}$) and model...
predictions \( c_{\text{pre}} \) of the receptor concentrations. Both the uncertainties in the receptor concentrations and in the source profiles were taken into consideration in the model to calculate the optimized source contribution from CMB–fmincon. The source profile \( (10 \times 26) \), receptor concentration \( (26 \times 1) \) and receptor concentration uncertainty \( (26 \times 1) \) were arranged in a Microsoft Excel sheet and used while executing the program in MATLAB R2008a. The calculated source contribution from the model was used to predict the concentration \( c_{\text{pre}} \) of the species in PM\(_{10}\) and PM\(_{2.5}\) of the SJVAQS Fresno and Bakersfield data. The execution of CMB by ‘fmincon’ solver in MATLAB R2008a is shown in Fig. S-1 of the Supplementary material to this paper. In comparison with CMB8.2, CMB–fmincon makes use of an objective function, which significantly reduces the run time between \( c_{\exp} \) and \( c_{\text{pre}} \) species concentrations. CMB–fmincon accounts both the uncertainties and hence reduces the error in the source contribution estimation with constrained optimization of chemical mass balance function.

**CMB with genetic algorithms (CMB–GA)**

A genetic algorithm (GA) is a stochastic global search method that works in the same manner as natural biological evolution. GA operates on a population of potential solutions applying the principle of survival of the fittest to produce better approximations to the solution. At each generation, a new set of approximations is created by the process of selecting individuals according to their level of fitness in the problem domain and breeding them together using operators borrowed from natural genetics. This process leads to the evolution of populations of individuals that are better suited to their environment than the individuals from which they were created, just as in natural adaptation.\(^{31}\) The algorithm consists of a main routine containing the optimization code and a subroutine containing the objective function of the code. In MATLAB R2008a, this is represented as shown in Fig. S-2 of the Supplementary material. Some details of the method are also given in the Supplementary material.

**Analysis of Fresno and Bakersfield PM\(_{10}\) and PM\(_{2.5}\) data\(^{28,29}\)**

California San Joaquin Valley (SJV) encompasses nearly 64,000 km\(^2\) and a population in excess of three million people. The majority of this population is centered in the large urban areas of Bakersfield and Fresno, although nearly 100 smaller communities are situated in the region. This population base, combined with oil and gas production and refining, waste incineration, electrical cogeneration, agriculture, transportation, commerce, and light manufacturing activities, leads to air pollution emissions and concentrations that approach those of the metropolitan area of Los Angeles (South Coast Air Basin-SOCAB) in southern California.\(^{28}\)

The average concentrations of PM\(_{10}\) and PM\(_{2.5}\) containing 26 species observed at Fresno and Bakersfield, California, in 1988–1989 were used to model in the CMB from Chow et al., (1992 and 1993).\(^{28,29}\) Hence, the receptor concentration matrix had a size of \( 26 \times 1 \). The contents of NO\(_3^−\), SO\(_4^{2−}\), NH\(_4^+\), elemental carbon (EC), organic carbon (OC), Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Se, Br, Sr, Zr, Ba and Pb in PM\(_{10}\) and PM\(_{2.5}\) were analyzed to estimate the source contributions. The 26 species present in PM\(_{10}\) and PM\(_{2.5}\) were identified as originating from ten sources according to the emission inventory, which is known as the source profile (percentage of mass emitted) for central California.\(^{29}\) Hence, the source profile matrix had the size of \( 26 \times 10 \). Since the elemental compositions of several sources were identified in the data base of USEPA (SPECIATE) and various studies,\(^{5,30,32}\) it was recommended that the particulate matter (PM\(_{2.5}\) & PM\(_{10}\)) sources be characterized locally for source apportionment studies.\(^{33}\)
Analysis of Portland, OR, PM$_{10}$ data$^{30}$

The average concentrations of 23 species in PM$_{10}$ analyzed at Portland, OR, were used to model the proposed CMB from Watson et al. (1984)$^{30}$. The source composition data and 24 h average concentration of OC, EC, NO$_3^-$, SO$_4^{2-}$, F, Na, Mg, Al, Si, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br and Pb were used as the input species in the three CMB models. The sources compositions of marine, urban dust, auto exhaust and residual oil combustion were used as source profile data in the CMB. The corresponding uncertainty values were also determined as per the literature followed.$^{30}$

Source contribution and predicted concentration

Source contribution values of each source were obtained from CMB8.2, CMB–fmincon and CMB–GA for the Fresno, Bakersfield and Oregon data. The measured source profile and obtained source contribution were used to calculate $c_{pre}$ of the species measured through backward trajectory. The errors between the experimental and predicted concentrations for the three approaches were also estimated.

RESULTS AND DISCUSSIONS

The source contributions for the analyzed species in PM10 and PM2.5 obtained using CMB8.2, CMB–fmincon and CMB–GA for the Fresno, Bakersfield and Oregon data are presented, respectively, in Figs. S-3–S-5 of the Supplementary material. It was observed that the source contributions obtained using CMB–fmincon and CMB–GA fitted better than those obtained using CMB8.2. The statistical parameters obtained for the PM$_{10}$ and PM$_{2.5}$ samples from the Fresno, Bakersfield and Oregon sites using CMB8.2, CMB–fmincon and CMB–GA software are presented in Tables I–III.

**TABLE I. Statistical validation of the results obtained for the analysis of the SJVAQ, California, Fresno data using the three chemical mass balance models**

<table>
<thead>
<tr>
<th>Statistical parameter</th>
<th>CMB8.2 PM$_{10}$</th>
<th>CMB8.2 PM$_{2.5}$</th>
<th>CMB–fmincon PM$_{10}$</th>
<th>CMB–fmincon PM$_{2.5}$</th>
<th>CMB–GA PM$_{10}$</th>
<th>CMB–GA PM$_{2.5}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R^2$</td>
<td>0.80</td>
<td>0.56</td>
<td>0.87</td>
<td>0.59</td>
<td>0.90</td>
<td>0.67</td>
</tr>
<tr>
<td>$\chi^2$</td>
<td>0.57</td>
<td>1.03</td>
<td>0.60</td>
<td>0.89</td>
<td>0.36</td>
<td>0.66</td>
</tr>
<tr>
<td>Mass, %</td>
<td>63.23</td>
<td>53.81</td>
<td>65.00</td>
<td>54.12</td>
<td>67.36</td>
<td>55.03</td>
</tr>
</tbody>
</table>

**TABLE II. Statistical validation of the results obtained for the analysis of the SJVAQ, California, Bakersfield data using the three chemical mass balance models**

<table>
<thead>
<tr>
<th>Statistical parameter</th>
<th>CMB8.2 PM$_{10}$</th>
<th>CMB8.2 PM$_{2.5}$</th>
<th>CMB–fmincon PM$_{10}$</th>
<th>CMB–fmincon PM$_{2.5}$</th>
<th>CMB–GA PM$_{10}$</th>
<th>CMB–GA PM$_{2.5}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R^2$</td>
<td>0.71</td>
<td>0.62</td>
<td>0.78</td>
<td>0.75</td>
<td>0.81</td>
<td>0.83</td>
</tr>
<tr>
<td>$\chi^2$</td>
<td>1.13</td>
<td>0.85</td>
<td>0.80</td>
<td>0.83</td>
<td>0.65</td>
<td>0.43</td>
</tr>
<tr>
<td>Mass, %</td>
<td>88.4</td>
<td>65.64</td>
<td>91.37</td>
<td>71.23</td>
<td>94.24</td>
<td>74.85</td>
</tr>
</tbody>
</table>

The two stations Fresno and Bakersfield were significant receiving various types of atmospheric pollution from large urban locations in California. The significant quantities of sulfate and nitrate in the ambient aerosol may be of second-
ary origin. Hence ammonium sulfate, ammonium nitrate, sodium nitrate and organic carbon (OC) were considered as a “pure” secondary source profile. Ammonium nitrate, paved road dust, secondary organic carbon, motor vehicle, ammonium sulfate, limestone were observed as the major contributors to PM$_{10}$ at the Fresno station. Primary crude oil, marine and vegetation burning were the successive emission sources of PM$_{10}$ to the ambient air (Fig. S-3a–c). Secondary aerosols (NH$_4$NO$_3$, secondary OC and (NH$_4$)$_2$SO$_4$) were observed as the major sources of PM$_{2.5}$ with contributions from paved road dust, primary crude oil and marine sources as not neglectable sources of PM$_{2.5}$ at the Fresno station (Fig. S-3d–f). Moreover, vegetative burning, limestone (construction activities) and motor vehicle emissions were other possible sources of PM$_{2.5}$ at the Fresno station.

### TABLE III. Statistical validation of the results obtained for the analysis of the Portland, Oregon data using the three chemical mass balance models

<table>
<thead>
<tr>
<th>Statistical parameter</th>
<th>CMB8.2</th>
<th>CMB–fmincon</th>
<th>CMB–GA</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R^2$</td>
<td>0.97</td>
<td>0.98</td>
<td>0.99</td>
</tr>
<tr>
<td>$\chi^2$</td>
<td>1.89</td>
<td>1.63</td>
<td>1.6</td>
</tr>
<tr>
<td>Mass, %</td>
<td>93.1</td>
<td>93.5</td>
<td>94.4</td>
</tr>
</tbody>
</table>

Paved road dust, motor vehicle emission, secondary nitrate, secondary sulfate, limestone (construction activities), primary crude oil and marine sources were observed as the major PM$_{10}$ emissions at Bakersfield (Fig. S-4a–c). Secondary aerosols, paved road dust, primary crude oil, marine, limestone were observed as the major PM$_{2.5}$ emission sources (Fig. S-4d–f). Vegetative burning seems to have contributed less to both PM$_{10}$ and PM$_{2.5}$ emissions at the Bakersfield station.

The source contribution optimized using the genetic algorithm (CMB–GA) showed the least percentage error between $c_{\text{exp}}$ and $c_{\text{pre}}$ concentrations of the species at Fresno and Bakersfield based on statistical parameters, as can be seen in Tables I and II. The Percentage mass value obtained from CMB–GA for PM$_{10}$ and PM$_{2.5}$ were acceptable for both Fresno (67.36 and 55.03 %) and Bakersfield (94.24 and 74.85 %) sites than CMB8.2 and CMB–fmincon optimization. The percentage mass values obtained for PM$_{10}$ via all three approaches were acceptable (63.23, 65 and 63.58 % for Fresno and 88.4, 91.37 and 94.24 % for Bakersfield), which indicates the estimates of the source contribution were well fitted in all three models. Hence, it could be predicted that optimization with the genetic algorithm yields better source contribution compared to CMB8.2 and CMB–fmincon optimizations. The performance measure by the $\chi^2$ values from the respective three approaches were acceptable and the values obtained from CMB–GA for PM$_{10}$ and PM$_{2.5}$ were 0.36 and 0.66 for the Fresno site and 0.65 and 0.43 for the Bakersfield site, the data of which proved the good results. Hence, all the concentrations of the species were best explained by the source contribution...
estimate from the genetic algorithm optimization, then from the fmincon and then from the CMB8.2 models. It was found that the error percentages between the experimental and predicted receptor concentrations were the lowest from the genetic algorithm approach as compared to the CMB8.2 and CMB–fmincon approaches. The successive $R^2$ values for the PM$_{10}$ data were best fit in the order of CMB–GA (0.81), then CMB–fmincon (0.78) and then CMB8.2 (0.71) and for PM$_{2.5}$ CMB–GA (0.83), then CMB–fmincon (0.75) and then CMB8.2 (0.62).

The source contributions obtained from the three respective CMB models for Portland, Oregon PM$_{10}$ data are shown in Fig. S-5a–c. The best source estimation was observed from CMB–GA with the best $R^2$ (0.99), $\chi^2$ (1.6) and percentage mass (94.4 %) then CMB–fmincon and then CMB8.2, as can be seen in Table III. The $\chi^2$ value between 1 and 2 is acceptable and the large percentage mass indicates the better estimation of the source contribution through CMB–GA.

Comparison between the experimental and calculated concentrations of 26 species in PM$_{10}$ and PM$_{2.5}$ analyzed through estimates of the source contribution from the three respective CMB approaches in the Fresno (Fig. 3) and Bakersfield

![Graphs](https://example.com/graphs)

**Fig. 3.** Experimental concentrations ($c_{\text{exp}}$) and the concentrations of species predicted by CMB8.2, CMB–fmincon and CMB–GA ($c_{\text{pre-CMB8.2}}$, $c_{\text{pre-CMB-fmincon}}$ and $c_{\text{pre-CMB-GA}}$, respectively) in: a) PM$_{10}$ and b) PM$_{2.5}$ from Fresno data.
(Fig. 4) data revealed possible deviations of the concentrations species from the real data. Since the genetic algorithm approach revealed large percentage mass (both Fresno and Bakersfield) with low $\chi^2$ and a better $R^2$ value, accurate concentrations of the species are predicted than the respective values obtained from CMB–fmincon and CMB8.2. The experimental and predicted species concentration data obtained for Portland, Oregon PM$_{10}$ is shown in Fig. 5. The least errors between $C_{\text{exp}}$ and $C_{\text{pre}}$ were observed for the data analyzed by CMB–GA than the data analyzed by CMB–fmincon or CMB8.2.

![Graph](image)

**Fig. 4.** Experimental concentrations ($C_{\text{exp}}$) and concentrations of species predicted by CMB8.2, CMB–fmincon and CMB–GA ($C_{\text{pre-CMB8.2}}$, $C_{\text{pre-CMB-fmincon}}$ and $C_{\text{pre-CMB-GA}}$, respectively) in: a) PM$_{10}$ and b) PM$_{2.5}$ from Bakersfield data.
CONCLUSIONS

The output of chemical mass balance model gives the contribution of each source type represented by a composition to the total mass, as well as to each chemical species in the receptor concentration. A comparison of CMB receptor models was performed to understand the efficiency in source contribution through various optimization techniques. The source profile uncertainty and receptor concentration uncertainties were used in the CMB8.2 software tool developed by USEPA. Optimized source contributions were obtained by CMB–GA and CMB–fmincon. The best estimate of the source contributions from the converged solution through CMB–GA was possible by a large number of generations. The source contributions obtained from the CMB8.2 deviated more in comparison with those obtained from the CMB–fmincon and CMB–GA because of the constrained optimization by ‘fmin con’ and GA solvers. The model accuracy was validated by various performance measures such as $R^2$, $\chi^2$ and percentage mass for the three respective CMB approaches. Very high correlations between $c_{\text{pre}}$ and $c_{\text{exp}}$ were obtained from CMB–GA for the Bakersfield data (0.81 and 0.83) than from CMB–fmincon and CMB8.2. $\chi^2$ (0.36, 0.66 and 0.65, 0.43) and percentage mass (67.36, 55.03 and 94.24 %, 74.85 %) from the CMB–GA model illustrated more accurate data for PM$_{10}$ and PM$_{2.5}$ from Fresno and Bakersfield. The methodology was followed with Portland, OR, PM$_{10}$ data that also resulted in the best fit from CMB–GA with $R^2$, $\chi^2$ and percentage mass values of 0.99, 1.6 and 94.4 %, respectively. The study revealed that CMB method with genetic algorithm optimization has better stability and more accuracy in determining the contributions of sources than CMB–fmincon and CMB8.2.
SUPPLEMENTARY MATERIAL

The execution of CMB by ‘fmincon’ solver in MATLAB R2008a, Fig. S-1, execution of the genetic algorithm code, Fig. S-2, details of the CMB–GA method and source contributions for the analyzed species in PM10 and PM2.5 obtained using CMB8.2, CMB–fmincon and CMB–GA for the Fresno, Bakersfield and Oregon stations, Figs. S-3–S-5, are available electronically from http://www.shd.org.rs/JSCS/, or from the corresponding author on request.

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