

Comparative Evaluation of VOC Source Profiles Developed by PMF and UNMIX Models

R. Ethirajan and S. Mohan

Abstract—Source profiles of hazardous VOCs were developed using PMF and UNMIX models for carrying out source apportionment of ambient VOCs using CMB model. The PMF model fitted the monitored data set very well with five factors. Q values, G-space plots and uncertainty analysis using boot strap technique showed that the performance of the PMF model was good. The source profiles from PMF model resembled most closely the source profiles of petroleum refinery, automobiles, wastewater treatment plants, solvent using industries, and solid waste burning. These source types match well with the prevailing source types in the study area. UNMIX model produced feasible solution with four factors. The source profiles obtained from both the models were compared and observed that the profiles of automobiles, solvent using industries and petroleum refinery developed by PMF model are matching with the profiles developed by UNMIX model. The fourth profile by UNMIX model is partially matching with wastewater treatment plants. The UNMIX model could not distinguish clearly the profile of biomass burning. These observations match very well with the observations of several studies which found that UNMIX model derive less factors compared to PMF model especially when limited data set is used and more contributing sources are available in the study area

Index Terms—PMF model, source profile, UNMIX model, VOC

I. INTRODUCTION

VOCs are often from multiple and heterogeneous sources. VOC emission inventories are difficult to assemble, because significant emissions come from fugitive sources (evaporative). The volatile organic compounds also undergo rapid transformation in the atmosphere [1]. The usefulness of the source orientated models to quantify the source contribution in these circumstances is very limited. Requirement of elaborate input parameters including emission profile, wind profile, vertical temperature profile, etc. makes these models difficult to use. The need for incorporating an appropriate chemical and photochemical transformation mechanism in the source oriented models is the major limitation in using these models for VOC.

On the other hand, a quantitative assessment of the contributions from different sources to the ambient VOC levels may be obtained through inverse modeling approach by applying receptor models without the knowledge of dispersion factors [2]. Receptor models are mathematical models for identifying the sources of air pollution and

quantifying their effects (contribution), primarily on the basis of concentration measurements at the receptor site. Receptor models encompass a variety of techniques that use differences in chemical composition, particle size and concentration patterns in space and time, as well as ratios among specific compounds to identify source types and contribution [3]. The general receptor model equation is as follows.

$$X_{ij} = \sum_{k=1}^P g_{ik} f_{kj} + e_{ij} \quad (1)$$

where x_{ij} is the concentration of j^{th} species in i^{th} sample, g_{ik} is the mass contributed by k^{th} factor in i^{th} sample, f_{kj} is the percentage emission of j^{th} species in the k^{th} factor and e_{ij} is the residual for each sample. The common receptor models include UNMIX model, a kind of factor analysis model, Positive Matrix Factorisation (PMF) model, a modified principal component analysis model and Chemical Mass Balance (CMB) model [4], [5]. PMF and UNMIX models which are capable of estimating both the source profiles and source contribution from the ambient measurement data alone can return only average source contribution estimate for the entire data set. The average source contribution estimate may not be adequate to develop strategies for effective VOC management at all occasions [6]. On the other hand CMB model, a widely used receptor model is capable of estimating source contribution even for a single data set, but only with prior knowledge of prevalent sources and their source profiles [7]. The objective of this study is to develop VOC source profiles of contributing sources at locations where source profiles are not available, using PMF and UNMIX models and comparatively evaluate them for effective VOC management.

II. SOURCE PROFILE

A. General

Source profiles are the mass abundances (fraction of total mass) of a chemical species in source emissions. Source profiles are intended to represent a category of source rather than individual emitters. Source profiles are created by sampling emissions from a variety of single emitters or small groups of emitters. These samples are then subjected to different chemical and physical analyses to determine those properties that will allow contributions from the sources they represent to be distinguished at receptors. Each of these properties must be normalized (scaled) to some common property in the emissions from all sources. The normalization procedure is one in which the measured concentrations are expressed as ratios (fractional abundances) [8].

Developing source profiles for all the possible sources of

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VOC is a very difficult, time consuming and expensive task. A wide variety of normalization factors and measurement units are applied and different conventions for defining VOC are adopted in VOC profile development and hence these profiles vary widely between different studies. Many published VOC profiles are not comparable to each other, or with the ambient measurements, in terms of their normalization. Several studies on source apportionment of ambient VOC, therefore, had used source profiles developed in countries like USA [9], [10]. In petroleum and petrochemical industries, the quantity of VOC emission largely depends on the operating practices, size of operation and meteorological conditions and therefore adopting those profiles may not be appropriate to solve the problem in a judicious way.

B. PMF Model

The PMF model is a multivariate factor analysis tool which uses ambient air measurement data alone to identify the source profiles and the source contributions [11]. PMF converts a matrix of speciated sample data into two matrices - factor contributions matrix and factor profile matrix. It solves the general receptor modeling problem using constrained, weighted least squares. The “constrained” part of the least squares is achieved by assuming that the contributions and mass fractions are all non-negative. The “weighted” part of the least squares is achieved by assigning higher uncertainties to the selected species based on the noise level in the data.

The model minimizes the objective function Q based on the uncertainties.

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left[\frac{x_{ij} - \sum_{k=1}^p g_{jk} f_{kj}}{S_{ij}} \right]^2 \quad (2)$$

where S_{ij} is the uncertainty associated with each data set. The Q values from the model are goodness-of-fit parameters and are an assessment of how well the model fit the input data [11].

C. UNMIX Model

UNMIX model uses the singular value decomposition (SVD) method to estimate the source number by reducing the dimensionality of data space m to p [12]. Assuming that ‘ p ’ number of sources contributing to ‘ m ’ number of species in ‘ n ’ number of samples, the UNMIX model can be expressed as

$$C_{ij} = \sum_{l=1}^p (\sum_{k=1}^p U_{lk} D_{lk}) V_{lj} + \epsilon_{ij} \quad (3)$$

where U , D and V are $n \times p$, $p \times p$ diagonal, and $p \times m$ matrices, respectively; and ϵ_{ij} is the error term consisting of all the variability in C_{ij} not accounted for by the first p principal components. Geometrical concepts of self-modeling curve resolution are used to ensure that the results obey (to within error) the nonnegative constraints on source compositions and contributions. Additional constraints are also used to help in determining the results. UNMIX model also normalizes the data matrix such that all the species are on the same scale with a mean of 1. The data are then projected to a plane perpendicular to the first axis of p -dimensional space. The edges represent the samples that characterize the source. Such edges in point sets are then used to calculate the vertices, which are used with the matrices decomposed by SVD to

obtain the source profiles and contributions [13].

III. AMBIENT VOC MONITORING

As monitoring and assessment of the ambient concentration of various hazardous VOC present in the study area had not been done prior to this study, it was monitored as a part of this study. The sampling and analysis were carried out as per the procedures laid down in USEPA compendium method TO-17 [14]. The study area is dominated by industrial sources, and hence emphasis was given to the assessment of VOC that are more hazardous. The ambient air samples were collected in standard adsorption tubes with 3.67 mm ID and 128.02 mm length and containing tenex and carbopack B in the ratio of 125 mg to 75 mg. A total of 50 samples (23 samples during weekdays and 27 samples during weekends/holidays) were collected between July 2008 and October 2009. The adsorption tubes were thermally desorbed to release the pollutants and the pollutants were analysed in Gas Chromatograph – Mass Spectrometer (GC-MS) [15]. Compounds that are listed under 60 target compounds by USEPA were quantified using appropriate standards. Among these 60 target compounds, about 21 hazardous volatile organic compounds were predominantly present in all the samples. The maximum, minimum and mean concentrations of these 21 species are listed in Table I.

TABLE I: OBSERVED CONCENTRATION OF DIFFERENT VOCs

Sl. No	Compounds	Concentration in $\mu\text{g}/\text{m}^3$		
		Max	Min	Mean
1	Benzene	83.25	13.24	38.23
2	Toluene	352.58	49.87	156.14
3	Ethylbenzene	61.40	5.43	27.37
4	<i>m,p</i> -Xylene	59.52	3.97	26.80
5	<i>o</i> -Xylene	54.52	2.29	22.94
6	1,2-Dichloropropane	59.16	0.00	13.71
7	Trichloroethylene	13.65	0.10	4.59
8	Styrene	22.00	1.00	6.31
9	1,3,5-TMB	27.20	0.41	7.22
10	1,2,4-TMB	12.25	0.42	4.67
11	Naphthalene	23.56	0.43	7.04
12	Isopropylbenzene	5.27	0.00	1.31
13	<i>n</i> -Propylbenzene	9.51	0.10	2.97
14	Carbon Tetrachloride	23.96	0.00	7.75
15	1,4-Dichlorobenzene	18.35	0.00	4.33
16	Secbutylbenzene	3.32	0.00	0.34
17	4-Isopropyltolune	13.17	0.00	1.76
18	Butylbenzene	6.32	0.00	0.42
19	Chloroform	19.01	0.00	20.21
20	1,2-Dichloroethane	84.16	0.00	9.50
21	Trichlorofluoromethane	0.92	0.00	0.04

IV. SOURCE PROFILE DEVELOPMENT USING PMF MODEL

As source profile of all contributing sources in the study area is not available, EPA PMF 3.0 model has been used to develop the source profiles. EPA PMF 3.0 is the latest PMF

version of USEPA minimizes the objective function Q using a program known as the multilinear engine, ME-2 [11]. A set of 50 samples with the concentration of 21 species each has been used as input of the model. Out of these 21 species, 6 species viz. secbutylbenzene, 4-isopropyltoluene, butylbenzene, 1,2-dichloroethane, 1,4-dichlorobenzene, trichlorofluoromethane have been identified as bad species as their ambient concentration was either 0 or not detected in several samples. Among the remaining 15 species 2 species viz. 1,2-dichloropropane and styrene have been identified as weak species as their signal to noise ratio was very low. As the number of data set is relatively smaller, an extra modeling uncertainty of 15% has been used to obtain consistent model output. The model was run with different number of source types ranging from 3 to 9 and it was observed that the model resolved optimum solution when the number of source types was 5. The Q value distribution of the base run of the model is presented in Fig. 1. It can be seen from Fig. 1 that all the 20 base runs of the model had produced converged results in both cases and that the values of Q (Robust) and Q (True) are almost the same. The minimum Q (Robust) estimated in 9th run is about 0.95 times the Q (Theoretical).

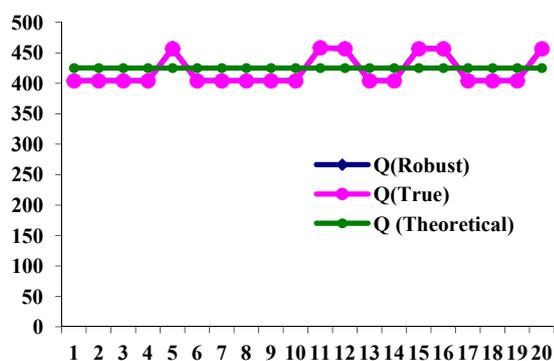


Fig. 1. Distribution of Q value in model base run

TABLE II: SOURCE PROFILES FROM PMF MODEL

Species	Mass Fractions				
	1	2	3	4	5
Benzene	0.02	0.13	0.09	0.08	0.17
Toluene	0.55	0.53	0.45	0.48	0.47
Ethylbenzene	0.01	0.08	0.11	0.05	0.11
<i>m,p</i> -Xylene	0.00	0.10	0.11	0.06	0.09
<i>o</i> -Xylene	0.02	0.10	0.11	0.03	0.06
1,2-Dichloropropane	0.15	0.02	0.01	0.00	0.00
Trichloroethylene	0.05	0.02	0.01	0.01	0.00
Styrene	0.01	0.01	0.01	0.02	0.01
1,3,5,-TMB	0.00	0.00	0.04	0.07	0.01
1,2,4-TMB	0.02	0.00	0.01	0.00	0.04
Naphthalene	0.00	0.00	0.01	0.11	0.01
Isopropylbenzene	0.01	0.00	0.02	0.00	0.00
<i>n</i> -propylbenzene	0.01	0.00	0.02	0.00	0.01
Carbon Tetrachloride	0.04	0.02	0.00	0.05	0.02
1,4-Dichlorobenzene	0.11	0.00	0.00	0.02	0.00

The results indicate that the model had produced optimum results for the monitored data set with five factors. The regression diagnostics of the base run with the minimum Q

value shows that the R^2 value of 13 species except the two week species is about 0.8, which indicates that the model has fit these species well. The model results also indicate that the scaled residuals of all the species are normally distributed as determined by Kolmogorov-Smirnoff test and only two data points exceeded the model output threshold for the scaled residuals [16]. The source profiles developed by PMF model is listed in Table II.

V. SOURCE TYPE IDENTIFICATION

There are several ways by which the actual source types are identified from the source profiles generated by the PMF model. Most of the studies had used tracer elements present in the profile to identify the source types [16], [17]. Reference [18] identified the source types assuming that the profiles obtained from the model will be same as the profiles of various source types available in similar monitoring sites. Some researchers used the prior knowledge on the source types present in the study area and their characteristics [19], [20].

In this study, the source types were identified by comparing the source profiles used/developed by similar studies along with the SPECIATE data base of USEPA (Fig. 2 to Fig. 5). The source profiles obtained in the model resembled most closely the source profiles of PETREF (petroleum refinery), AUTMOB (automobiles), WWTP (wastewater treatment plants), SOLUSE (solvent using industries) and SWBURN (solid waste burning) [21], [22], [23]. These source types match well with the prevailing source types in the study area.

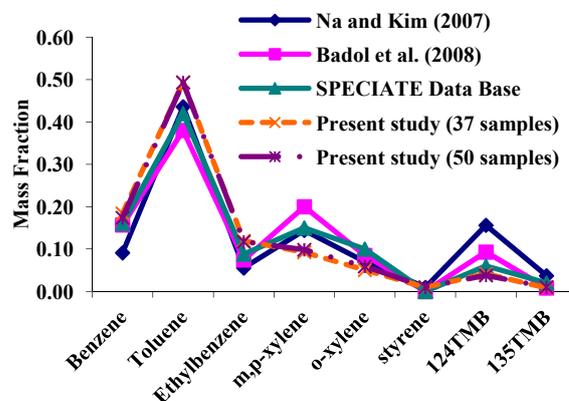


Fig. 2. Source Profiles of Automobiles

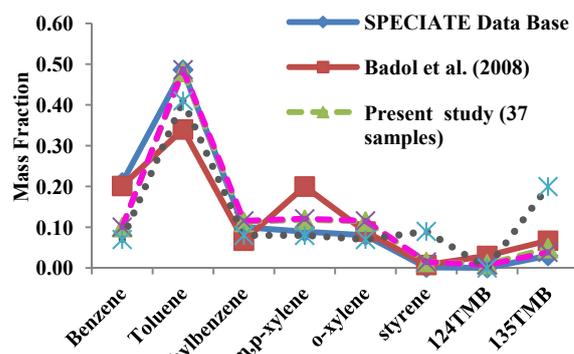


Fig. 3. Source Profiles of Petroleum Refinery

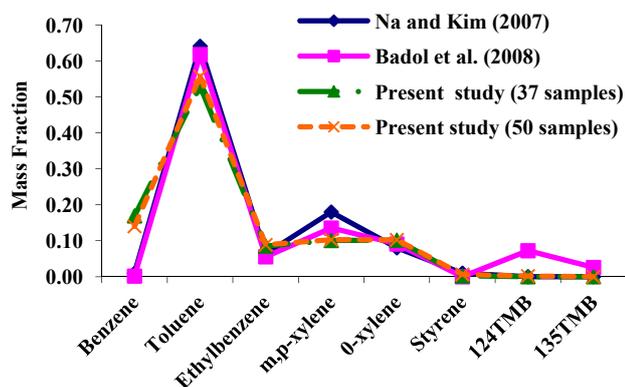


Fig. 4. Source Profiles of Solvent Using Industries

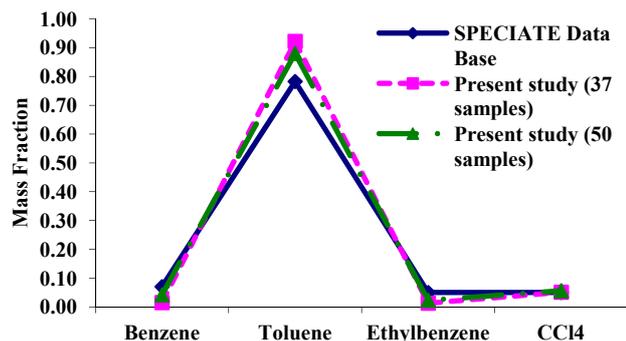


Fig. 5. Source Profiles of Wastewater Treatment Plants

VI. SOURCE PROFILE DEVELOPMENT USING UNMIX MODEL

To comparatively evaluate the source profiles obtained from PMF model and to assess the applicability of UNMIX model for the monitored data set, source profiles have been developed using UNMIX model also. The UNMIX 6.0 version of USEPA has been used in this study for developing source profiles. 50 samples set with 21 species have been used as input to the model. Unlike PMF model, UNMIX model resolves the given data set into different factors on its own. UNMIX determines the edges in the dataset. The number and direction of the edges depend on the number of species chosen for the UNMIX model. UNMIX incorporates the algorithm “NUMFACT” that estimates the number of factors in the data using principal component analysis on randomly sampled subsets of the original data [12]. The source profile developed by UNMIX model is presented in Table III. Though negative composition is physically meaningless, the UNMIX model provides negative composition to ensure that the source contributions are either positive or zero.

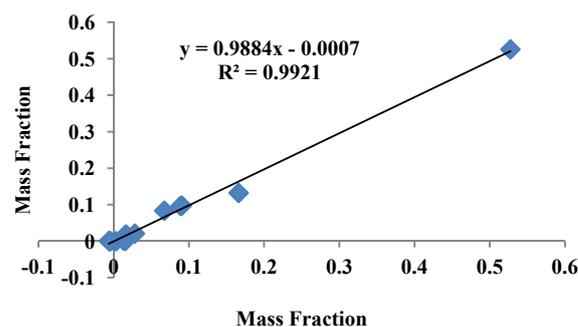
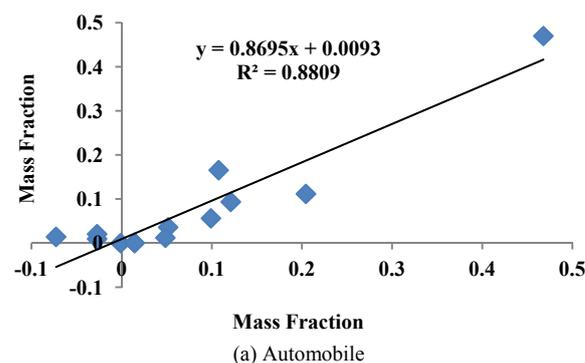
TABLE III: SOURCE PROFILES FROM UNMIX MODEL

Species	Mass Fraction			
	1	2	3	4
Benzene	5.160	-2.210	6.740	28.400
Toluene	20.700	13.500	29.300	90.200
Ethylbenzene	7.580	-4.760	12.800	11.500
m,p-Xylene	7.060	-3.670	7.580	15.500
o-xylene	4.910	-3.260	6.200	15.200
Trichloroethylene	-0.379	2.220	-0.076	2.780

1,3,5-TMB	8.970	-0.501	-1.710	0.529
1,2,4-TMB	-0.197	-0.418	3.230	2.270
Naphthalene	5.580	3.480	-4.560	2.640
Isopropylbenzene	1.680	-0.279	0.889	-0.983
n-Propylbenzene	1.950	-0.957	3.040	-1.090
Carbon Tetrachloride	0.733	3.790	-1.710	4.820
1,4-Dichlorobenzene	-0.719	4.860	0.902	-0.704

VII. COMPARATIVE EVALUATION OF SOURCE PROFILES FROM PMF AND UNMIX MODELS

To comparatively evaluate UNMIX solution and PMF solution for the given data set, a four factor solution was also obtained from PMF model. The Q (Robust) and Q (True) values obtained from PMF model for 4 factor solution was 657.4 and 657.5 respectively as against the Q (Theoretical) value of 490. The high (> 25%) variation between Q (Robust) and Q (Theoretical) indicates that the performance of PMF model when solving the given data set for 4 factors is not good. The source profiles obtained from UNMIX model and PMF model have been compared by plotting scatter plots between the profiles (Fig. 6). It can be seen from the R^2 values of the scatter plots that the profiles of automobiles, solvent using industries and petroleum refinery developed by PMF model are matching well with the profiles developed by UNMIX model. The fourth factor is partially matching with WWTP. The UNMIX model could not distinguish clearly the profile of biomass burning. In general several studies found that UNMIX model derive less factors compared to PMF model especially when limited data set is used and more contributing sources are available in the study area [24], [25], [26]. Though UNMIX model provided a feasible solution to the given data set with four factors, the profiles generated by PMF model is distinct and representative of the study area.



(b) Solvent Using Industries

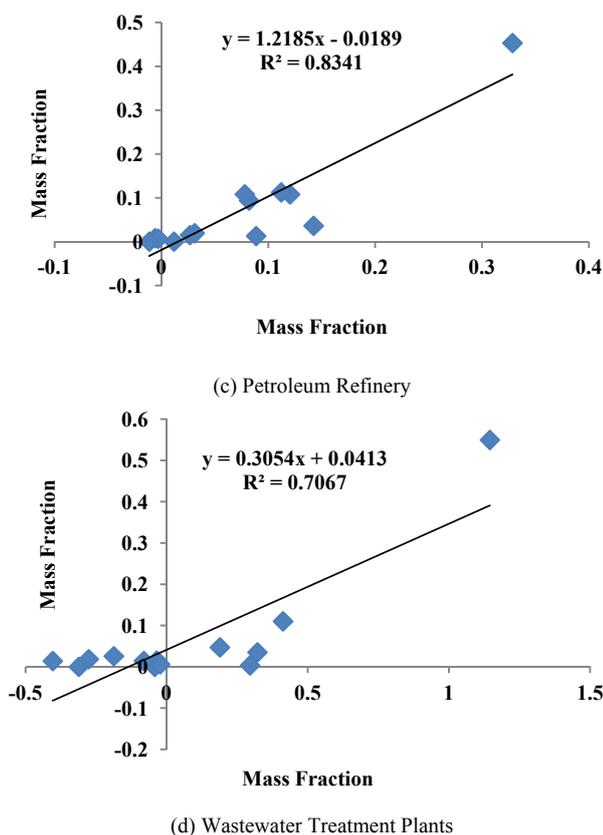


Fig. 6. Scatter plots of source profiles from PMF and UNMIX models

VIII. CONCLUSIONS

The PMF model fitted the monitored data set very well with five factors and generated source profiles. The source profiles obtained from PMF model were compared with the SPECIATE data base of USA and the source profiles used/developed by two other similar studies and found that the profiles resembled most closely the source profiles of petroleum refinery, automobiles, wastewater treatment plants, solvent using industries, and solid waste burning. These source types match well with the prevailing source types in the study area. UNMIX model produced feasible solution with four factors. The source profiles obtained from UNMIX model and from PMF model were compared and observed that the profiles of automobiles, solvent using industries and petroleum refinery developed by PMF model are matching with the profiles developed by UNMIX model. The fourth profile by UNMIX model is partially matching with wastewater treatment plants. The UNMIX model could not distinguish clearly the profile of biomass burning. This observation match very well with the observations of several studies which found that UNMIX model derive less factors compared to PMF model especially when limited data set is used and more contributing sources are available in the study area. Although the PMF model produced stable solution and provided source profiles for five factors, it could not differentiate between the smaller contributing factors like gasoline vehicle, diesel vehicle, fuel filling stations, paints manufacturing and use etc mainly because of limited species in the monitored data. Monitoring more species and more samples may produce source profiles of even small contributors.

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