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Application of CMB₈ Model for Source Apportionment in Bangkok Metropolitan Area

A. Wangkiat*, H. Garivait**, N. W. Harvey***, and S. Okamoto****.

Abstract

Samples of 24-hours total suspended particulate matter were collected by high volume air samplers during December 1996 to January 1997 at a station in Bangkok, Thailand, and analyzed for 14 elements and 2 carbons. Seven source profiles and the average mass concentration of 42 data ambient data were used to run the Chemical Mass Balance (CMBs) model. The source apportionment by CMBs gave similar results comparing with the former results by Factor Analysis – Multiple Regression (FA-MR) model. The results revealed that major sources of particulate matter in Bangkok were: soil, road dust and automobile (80 %). The minor source contributions were: sea salt (4.22%), refuse incineration and biomass burning (0.82%), steel mill (0.60%) and fuel oil combustion (0.33). The lack of source profile for biomass or open burning in Bangkok resulted in much lower contribution of this source when compared to that from FA-MR. This source profile is needed to improve the CMB calculation of source contribution of particulate matter in Bangkok area.

Key wards: PM10, receptor model, CMB8, factor analysis-multiple regression

Introduction

Bangkok, the capital of Thailand, is facing a serious and deterioration problem of air pollution from suspended particulate matter (PM). At many monitoring stations in this area, the levels of fine particulate matter under 10 micron in aerodynamic diameter (PM₁₀) usually exceed the national 24-hour ambient standard (120 μ g/m³). The annual average of PM₁₀ at roadsides typically range from 70-80 μ g/m³, which was approximately 40% - 60% above its standard (50 μ g/m³). The major sources contributing of these high PM levels are observed to be motor vehicles, dust from roads and construction sites, and industrial PM emissions (Radian, 1998; Garivait, 1999).

In order to develop a comprehensive control strategy for PM in Bangkok, it is very important to identify all of its significant sources contributing to the level of PM in the area. Development of emission inventories is among many attempts to quantify the mass emissions coming from possible major sources of PM to the region. Although this approach allows us to estimate possible sources of PM to a receptor, it does not give information on their loading contributions at the site. Another approach is chemical mass balance method (CMB). This method, on the other hand, provides information on the source

loading to the receptor that cannot be achieved by source inventory data alone.

A few work has attempted to use mass balance model approach to solve the air pollution problem in Bangkok. The purposes of this study were to estimate the source contributions of suspended particulate matter (SPM) in the Bangkok metropolitan area using CMB₈ model, and to compare this result with former work at the same site using Factor Analysis - Multiple Regression Model (FA-MR) by Garivait, 1998.

Source Apportionment Model

The source apportionment model (or receptor model) applied in this study was the Chemical Mass Balance Model Version 8 (CMBs) developed by Watson *et al.*, 1997.

As oppose to a source-based dispersion model, the CMB model has some advantages in its applications. These include: a) it does not depend on emission inventory data, which can be unreliable, and b) the CMB technique has ability to account for pollution, both point and area sources, without computer intensive mathematical techniques, and numerous model inputs for meteorology, topography, deposition rate, etc. (Scheff *et al.*, 1984).

The CMB model can determine which source types are the major contribution of the ambient pollution to the receptor being investigated. The model is receiving much interest by many regulatory agencies. The results form the CMB calculations are very useful for regulators to make decisions concerning suitable control measures, and assigning priority for abatement control, particularly when budget is limited.

Source profiles (i.e., the fractional amount of chemical species contained in an emission from each source type) and their concentrations at the receptor, with appropriate uncertainty estimates, serve as input data to the CMB model. The output consists of the amount contributed by each source-type of each chemical species. The model calculates the values for the contributions from each source and the uncertainties of those values. The uncertainties of the input data are used both to weigh the importance of the values of output data in the solution, and to calculate the uncertainties of the source contributions (Watson *et al.*, 1997).

The CMB model consists of a least squares solution to a set of linear equations that expresses each concentration at receptor of a chemical species as a linear sum of the products of source profile species and source contributions (Watson *et al.*, 1989).

The basic assumptions of the model are as follows:

-Compositions of source emissions are constant over the period of ambient and source sampling, -Chemical species do not react with other,

-All the sources with a potential for significantly contributing to the receptor have been identified and have had their emission characterised.

-The source compositions are linearly independent of each other.

-The number of sources or source categories is less than or equal to the number of chemical species.

-Measurement uncertainties are random, uncorrelated and normally distributed.

CMBs Model Equation

The CMB₈ uses the least squares estimation method to solve the CMB equations. It bears resemblance to linear regression analysis, and consists of the following set of equations:

	Ci	=	$Fi1S1+Fi2S2+\ldots+FijSj\ldots+FIJSJ$	i =1I, j =1J
Where	Ci	=	Concentration of species <i>i</i> measured at a rec	ceptor site
	Fij	=	Fraction of species <i>i</i> in the emission from so	urce j
	Sj	=	Estimate of the contribution of source j	
	Ι	=	Number of chemical species	
	J	=	Number of source types	

These equations will yield a unique solution only when the number of species is equal to or greater than the number of sources. Studies on the model evaluation showed that the greater the number of species, the more precise the apportionment calculated. These simultaneous equations can be solved by an effective variance weighted least squares estimation method (Watson, *et al.*, 1984).

The CMBs software is the new version of CMB currently in use. It employs an effective variance solution, developed and tested by Watson *et al.* (1984). The effective variance solution is derived by minimizing the weight of the squares of the differences between the measured and calculated values of C*i* and F*ij* (Britt and Luecke, 1973). The solution algorithm is an iterative procedure, which calculates a new set of S*j*, based upon the estimated S*j* from the previous iteration. Its solution provides more realistic estimates of the uncertainties of the source contributions, owing to its incorporation of both source profiles and receptor data uncertainties.

Source Profiles in CMB Modelling

For the CMB model, sources that effect ambient air quality with regard to PM must be identified, and their compositions need to be chemically characterized. In Thailand, there are a few inventories of source chemical compositions and concentrations. In 1991, the Japan International Cooperation Agency (JICA) investiged the air quality around Samut Prakarn industrial districts for the air quality planing in that area. Its results are presented in Table 1. In 1998, the Radian International LIC presented the Pollution Control Department (PCD) with a report on the PM abatement strategies. As a result, additional inventories of point and area source profiles in Bangkok area were produced (see Table 2).

The PM source apportionment by CMB⁸ has also been estimated (Radian, 1998). For this study both point and area sources were considered. Consequently, eight potential source profiles were chosen. The percentage mass source profiles inputted into the CMB⁸ model is listed in Table 3.

In Bangkok, for exhaust emissions, those from diesel engines was the greatest, approximately 63%, while emissions from gasoline engines was about 30% (Wibulswas, 1999). Because there are some

similarities between this two source profiles, in this study we did not use source profile from gasoline in the CMB_8 calculations.

In suburb of Bangkok, people often burn their waste, such as paper, plastic, plant leaves and garbage. In CMB model, this open burning has multicollinearlity with refuse incineration. Therefore, in this study these two source profiles were grouped together by arithmetic mean. Finally, only seven profiles were used to run the model.

(% mass)								
	Soil	Road dust	Diesel	Gasoline	Sea salt	Iron steel industry	Fuel oil	Refuse
EC	0.32500	1.69000	62.00000	30.10000	0.00000	3.30000	37.10000	9.40000
OC	0.10000	0.06000	10.00000	26.30000	0.00000	3.30000	2.40000	1.50000
NA	0.65000	0.72000	0.01200	0.02000	30.42000	0.71500	3.78000	8.83000
AL	5.55000	5.00000	0.02400	0.13000	0.00003	1.80000	1.42500	0.97700
Κ	3.15000	2.10000	0.01200	0.20000	1.10000	1.15000	0.13000	10.00000
CA	0.17500	2.60000	0.32500	0.00000	1.20000	4.70000	1.45000	1.36000
SC	0.00045	0.00045	0.00000	0.00001	0.00000	0.00029	0.00003	0.00007
TI	0.27000	0.23000	0.00000	0.00000	0.00000	0.10000	0.04300	0.13800
V	0.00355	0.00460	0.00029	0.00021	0.00001	0.03650	3.77000	0.00270
CR	0.00275	0.00330	0.00450	0.00210	0.00000	0.42300	0.27600	0.05700
MN	0.03650	0.06100	0.00770	0.00510	0.00001	2.77000	0.05000	0.05400
FE	1.20000	1.80000	3.07000	0.49000	0.00003	30.10000	2.83000	0.59000
NI	0.00300	0.00250	0.00000	0.00380	0.00000	0.17000	2.10000	0.01350
ZN	0.00175	0.01300	0.11600	0.14000	0.00000	2.60000	0.17000	0.07940
SE	0.00092	0.00061	0.00003	0.00000	0.00001	0.00510	0.00670	0.00330
BR	0.00175	0.01500	0.00000	2.00000	0.19000	0.01400	0.00600	0.64200
SB	0.00016	0.00034	0.00005	0.00019	0.00000	0.00900	0.00100	0.11200
PB	0.00700	0.01300	0.04900	12.00000	0.00001	1.05000	0.09000	5.34000

Table 1: The compositions of emission sources in Samut Prakarn district.

Source: JICA(1991)

(%mass ± uncertainty)							
	Heavy duty truck	Light duty truck	Motorcycle	Soil	Road dust	Steel mill	Power plant
EC	18.181 ± 2.243	61.758 ± 13.423	3.440 ± 5.220	1.199 ± 0.752	1.145 ± 0.478	4.379 ± 0.716	0.000 ± 0.300
OC	79.647 ± 2.326	15.616 ± 4.537	34.327 ± 11.950	8.117 ± 0.590	11.909 ± 2.531	5.130 ± 0.957	0.000 ± 0.300
$\mathrm{NH4}^+$	0.000 ± 0.300	0.000 ± 0.300	0.000 ± 0.300	0.001 ± 0.000	0.002 ± 0.002	0.052 ± 0.073	0.018 ± 0.005
CL	0.000 ± 0.300	0.000 ± 0.300	0.000 ± 0.300	0.118 ± 0.072	0.127 ± 0.047	0.442 ± 0.625	0.045 ± 0.014
NO ₃	0.000 ± 0.300	0.000 ± 0.300	0.000 ± 0.300	0.014 ± 0.007	0.015 ± 0.017	0.209 ± 0.296	0.000 ± 0.300
SO4 ²⁻	0.000 ± 0.300	0.000 ± 0.300	0.000 ± 0.300	0.690 ± 0.451	0.228 ± 0.084	0.746 ± 1.055	35.300 ± 10.590
Al	0.098 ± 0.044	0.028 ± 0.017	0.022 ± 0.012	9.370 ± 1.330	7.877 ± 0.998	0.470 ± 0.197	0.025 ± 0.008
Si	0.660 ± 0.208	0.431 ± 0.241	0.124 ± 0.113	22.950 ± 2.850	20.133 ± 0.946	1.923 ± 0.598	0.104 ± 0.031
Р	0.091 ± 0.077	0.018 ± 0.011	0.002 ± 0.003	0.037 ± 0.016	0.067 ± 0.037	0.002 ± 0.002	0.000 ± 0.300
S	0.289 ± 0.314	0.365 ± 0.168	0.113 ± 0.076	0.478 ± 0.190	0.423 ± 0.122	0.198 ± 0.101	2.920 ± 0.876
Cl	0.024 ± 0.018	0.014 ± 0.008	0.016 ± 0.013	0.166 ± 0.104	0.264 ± 0.131	0.815 ± 0.179	0.000 ± 0.300
Κ	0.106 ± 0.027	0.001 ± 0.002	0.009 ± 0.007	1.500 ± 0.060	1.827 ± 0.327	0.453 ± 0.008	0.010 ± 0.003
Ca	0.289 ± 0.062	0.055 ± 0.027	0.087 ± 0.078	17.450 ± 1.250	20.200 ± 2.337	3.133 ± 0.342	0.062 ± 0.019
Ti	0.026 ± 0.023	0.002 ± 0.002	0.003 ± 0.003	0.295 ± 0.057	0.267 ± 0.034	0.080 ± 0.019	0.000 ± 0.000
V	0.004 ± 0.003	0.002 ± 0.001	0.002 ± 0.005	0.012 ± 0.003	0.012 ± 0.004	0.007 ± 0.001	1.070 ± 0.321
Cr	0.000 ± 0.300	0.001 ± 0.000	0.002 ± 0.005	0.011 ± 0.001	0.013 ± 0.003	0.043 ± 0.018	0.010 ± 0.003
Mn	0.001 ± 0.001	0.001 ± 0.001	0.001 ± 0.001	0.078 ± 0.022	0.075 ± 0.017	0.606 ± 0.284	0.003 ± 0.001
Fe	0.349 ± 0.181	0.066 ± 0.044	0.017 ± 0.018	3.685 ± 0.685	3.520 ± 0.406	15.700 ± 3.034	0.123 ± 0.037
Cu	0.014 ± 0.005	0.004 ± 0.002	0.003 ± 0.003	0.016 ± 0.004	0.030 ± 0.018	0.106 ± 0.030	0.002 ± 0.001
Ni	0.003 ± 0.002	0.001 ± 0.001	0.001 ± 0.002	0.003 ± 0.000	0.003 ± 0.000	0.033 ± 0.010	0.548 ± 0.164
Zn	0.086 ± 0.098	0.039 ± 0.009	0.022 ± 0.020	0.036 ± 0.011	0.092 ± 0.050	3.913 ± 2.295	0.003 ± 0.001
As	0.000 ± 0.300	0.001 ± 0.001	0.001 ± 0.002	0.003 ± 0.001	0.001 ± 0.002	0.000 ± 0.300	0.004 ± 0.001
Br	0.005 ± 0.002	0.000 ± 0.001	0.001 ± 0.003	0.002 ± 0.001	0.001 ± 0.000	0.011 ± 0.004	0.000 ± 0.300
Ba	0.102 ± 0.116	0.000 ± 0.300	0.068 ± 0.133	0.037 ± 0.005	0.036 ± 0.017	0.033 ± 0.012	0.015 ± 0.004
Pb	0.027 ± 0.010	0.003 ± 0.003	0.011 ± 0.012	0.012 ± 0.005	0.024 ± 0.012	0.448 ± 0.244	0.001 ± 0.000

Table 2: The compositions of emission sources in Bangkok area.

Source: RADIAN (1998)

Table3: Source profiles for the CMB₈ calculations

(%mass)									
	Soil	Road dust	Diesel	Sea salt	Steel mill	Fuel oil	Refuse	Biomass burning	Refuse and Biomass burning
OC	0.1	11.909	10	0	3.3	2.4	1.5	31.609	16.555
EC	0.325	1.145	62	0	3.3	37.1	9.4	5.191	7.296
Na	0.650	NA	0.012	30.42	0.715	3.78	8.83	0.131	4.481
Al	5.55	7.877	0.024	0	1.8	1.425	0.977	0.330	0.654
Κ	3.15	1.827	0.012	1.1	1.15	0.130	10	2.34	6.17
Ca	.0175	20.2	0.325	1.2	4.7	1.45	1.36	0.801	1.081
Ti	0.27	0.267	0	0	0.100	0.043	0.138	0.006	0.072
V	0.004	0.012	0	0	0.037	3.77	0.003	0	0.002
Cr	0.003	0.013	0.005	0	0.423	0.276	0.057	0.004	0.31
Mn	0.037	0.075	0.008	0	2.77	0.050	0.054	0.157	0.106
Fe	1.2	3.52	3.07	0	30.1	2.83	0.590	0.213	0.402
Zn	0.002	0.092	0.116	0	2.6	0.170	0.079	0.012	0.046
Se	0.001	NA	0	0	0.005	0.007	0.003	0	0.002
Br	0.002	0.001	0	0.19	0.014	0.006	0.642	0.023	0.333
Sb	0	NA	0	0	0.009	0.001	0.112	0	0.056
Pb	0.007	0.024	0.049	0	1.05	0.090	5.34	0.012	2.676
Reference	JICA	Radian	JICA	JICA	JICA	JICA	JICA	Tabak	(Refuse+Biomass
	(1991)	(1998)	(1991)	(1991)	(1991)	(1991)	(1991)	(1979)	burning)/2

Ambient Data

A total of 47 data were collected during December 1996 to January 1997. Ambient sampling was conducted at the Office of Environmental Policy and Planning (OEPP) in Bangkok, Thailand (see Figure 1). This receptor site is located in the centre of Bangkok urban area, and surrounded by commercial buildings, government offices, houses, roads and an expressway. Two sets of SIBATA high volume air samplers were placed on the top of a 7-storey building, about 20 meters above the ground. One was loaded with a cellulose nitrate membrane filter for elemental analysis, and the other was loaded with quartz fiber filter for ionic species and carbon analysis. The 24-hour total suspended particulate matter (TSP) was collected.

For elemental compositions, samples were sent to be analyzed by instrumental neutron activation analysis (INAA) at the Japan Environmental Sanitation Centre (JESC), Kawasaki, Japan. Lead content was analyzed by X-ray fluorescence method (XRF), while organic and elemental carbons by CHN analyzer at the Chulalongkorn university, Bangkok, Thailand. Any samples showing tendency to be outliners of some elements were rejected. Finally, only 42 samples, with 14 elements and 2 types of carbons, were used to produce an average sample datum for CMB₈ calculations, as shown in Table 4.

In the CMB modelling, the values of uncertainties of the input data is as equal important as the measurement data themselves. They are needed to weigh the species influences on the calculation results. From previous applications of the CMB model, the U.S. E.P.A. suggested that, typical uncertainties in both source and receptor measurements up to ±30% in each species is tolerable (U.S. E.P.A., 1987). In this study, the actual uncertainties were used for the measurements of source, and those whose uncertainty can not be identified, 10% uncertainty was assigned.



Figure 1: Location of the sampling station in Bangkok.

Component	Mean concentration	Analytical error*
-	(µg/m ³)	(%)
INAA		
Na	0.80833	2.74
Al	3.95714	1.62
K	2.15381	20.39
Ca	11.42381	5.60
Ti	0.29119	18.38
V	0.02912	2.98
Cr	0.01495	10.19
Mn	0.09012	1.57
Fe	3.67381	6.53
Zn	0.38417	5.81
Se	0.00166	28.66
Br	0.01938	18.00
Sb	0.01274	3.06
XRF		
Pb	0.07510	
<u>CHN analyzer</u>		
Organic carbon	8.28595	_
Elemental carbon	11.00714	
<u>Ion chromatography</u>		
NO3 ⁻	6.91512	
SO4 ²⁻	12.16779	_

Table 4: Average concentrations and uncertainties of suspended particulate matter at the receptor site.

* INAA analytical error was determined by the counting of the gamma- ray spectrum.

Results and discussion

Since one assumption in the CMB model is that chemical species should not react with each other, secondary pollutants are assumed zero. Unlike, FA-MR, the CMB model cannot estimate the contributions of secondary pollutants. In reality, secondary pollutants are indeed formed in the atmosphere, most of which are nitrate and sulphate compounds. It is seem impossible to produce a secondary pollutant source profile as the process is complicated and specific. Thus, to reduce this problem, we treated the actual average total mass measured at the receptor site before inputting it into the model by subtracting possible secondary pollutants from the total mass. Data in Table 4 give information on SO₄⁻ and NO₃⁻. Therefore, they were assumed to be secondary pollutants since there are no evidence of sources of these primary pollutants in the vicinity of the receptor. The total mass input was calculated as follows:

Total mass input = Total mass concentration - [(NH 4)2SO4 - NH4NO 3]

The actual average total mass was 189.6 μg . Therefore, the total mass input was calculated to be 162.2 μg .

The result of source contribution estimation calculated by CMBs is shown in Table 5. This table also compares the results from CMBs with FA-MR, previously calculated by Garivait, 1999. These two models yielded similar tendency. The result also shows the advantage of CMB model over FA-MR. That is that each source contribution can be separated (Okamoto, 1990). However, in contrast with FA-MR result, the contribution of refuse and biomass burning was very small (0.82%). To improve the CMB calculation, one requires accurate and precise measurements of the chemical compositions of particulate emission from open burning source which are likely to contribute to PM in Bangkok.

Table 5: Comparison of the CMB₈ and FA-MR mass apportionment calculations.

* % Mass Contribution 54.25
54.25
19.15
al 4.25
ning 11.70
-
10.63

* R² = 0.99, Chi-square = 1.60, d.f. = 4

** Factor analysis and multiple regression model from Garivait (1998), for comparison the secondary pollution being subtracted.

Conclusion

The results from the CMB₈ model calculation revealed that mass contributions to the receptor site in Bangkok urban area mainly came from soil, road dust and vehicles, with little contributions from sea salts, steel mill, refuse and biomass burning, and fuel oil. The result was in the same direction as those calculated by FA-MR model of the same receptor. CMB₈ had advantage over FA-MR in that more source contribution can be identified. However, the selection of a suitable source profiles require indepth knowledge of the source characteristics.

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