Source apportionment of PAHs in road sediments by CMB models: considering migration loss process

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ABSTRACT

Certain gaps always exist between the pollution source apportionment model principles and the actual environmental processes and the efforts to narrow the gaps are always valuable. In this study, it was found that the source profiles calculated by positive matrix factor analysis (PMF) were not much similar to the measured source profiles of collected real pollution sources samples. Results suggested that this difference was due to the changes that happened in the process of pollutant transportation from sources to environmental media such as transportation loss and reactions. To simulate this process, in this study, a modified chemistry mass balance (CMB) application method was proposed. Selective consideration of atmospheric reaction kinetics of certain long-distance migratory sources, such as pollutants from industrial and coke ovens that are far from urban centers, improved the accuracy of CMB model analysis for source diagnostics of polycyclic aromatic hydrocarbons in road sediments. At the same time, sources close to the receptor, such as traffic sources and resident sources, were applied in the CMB model without considering attenuation.

Keywords: Chemistry mass balance; Polycyclic aromatic hydrocarbons; Road sediments; Source analysis

1. Introduction

Road sediments (RS) are common pollution carriers in cities. They are both sources and sinks of pollution [1]. RS is able to migrate to the atmosphere, water, and organisms. When transported to water bodies by runoff, they aggravate water quality due to the pollutants adsorbed on RS from diverse sources [2], which are mostly non-point sources. Among these pollutants, polycyclic aromatic hydrocarbons (PAHs) are particularly concerned because they are harmful to humans and the ecological system and from complex sources.

Various models have been developed to identify and quantify the non-point pollution sources. Receptor models are among the most commonly used ones, using the principle of linear algebra. They divide the combinations of pollutants in the samples into several groups of vectors, which are considered as the composition of possible sources [3]. Receptors models can be divided into source profiles-dependent models (SDM) and source profiles-independent models (SIM). SDMs such as chemistry mass balance (CMB) need to input all possible source information. The advantage of this method is the fact that it can analyze any single sample. SIMs include methods like principal component analysis (PCA), positive matrix factor analysis (PMF) and so on. SIMs do not need any information about sources. The principle of SIMs models is to use statistical principles to extract the potential common factors in the of multiple groups of samples and consider the factors as sources.

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These two kinds of receptor models have been widely used for source apportionment of PAHs in RS. PCA and PMF were used to analyze contributions of PAHs in RS in Iran, China, Brazil, Australia, India, and so on [4–8]. In contrast, few kinds of literature used CMB to analyze PAHs in RS. However, there are some reports of success using the CMB model to analyze PAHs in other sediments such as surface soil, shallow-water, or deep-water sediments [9–11]. Vehicular emission, coke oven, coal burning, and surface pavement are the main sources of PAHs in urban areas according to those studies. Although most studies showed that both SIM and SDM receptor models had good fitting results, there are still some problems in the application. Although SIMs are popular and useful, using SIMs models requires a certain amount of data. Although meaningful results can be obtained with a small amount of data, with the number of samples largerthan the number of sources [12], certain studies have shown that the reliability of analysis results obtained from hundreds of sample data is more acceptable [13,14]. Also, identifying what the source profiles analyzed by SIMs means might also be hard. Those profiles are often not similar to any of the source profiles reported in the literature. Therefore, most studies can only judge the meaning of source profiles by the characteristic molecule, which also introduces uncertainty [15,16]. Meanwhile, SDM is not dependent on the amount of data analyzed, and the uncertainty of each sample will not affect each other. The main problem of using SDM is that this model is based on some assumptions. The key one is to assume that the proportions of multiple pollutants remain unchanged during migration. This is basically impossible for some mixed pollution with different properties and degradation possibilities. In particular, many reports have proved that the degradation of PAHs is mainly caused by photodegradation.

Considering the problems above, many researchers try various ways to improve existing models. One idea is to combine the CMB model with other models. Shi et al. compared the effect of three kinds of SIM combined with CMB [17]. Zelenka et al. [18] combined target transformation factor analysis with CMB. These researches either input source profiles resolved from SIM into CMB to calculate the contribution rate, or use CMB to analyze the unknown source profiles resolved from SIM. The attenuation of pollutants was not considered in those conditions, the inconsistency between SIM and SDM source profiles was attributed to the mixing of source profiles. Another research direction is considering attenuation factors. Venkataraman et al. [19] utilized the changing of elemental carbon concentrations to calculate the migration time of PAHs in the atmospheric particulate matter then import the modified source profiles into CMB models. Wang et al. [20] considered aerosol light extinction to improve the results of the CMB model. Demir and Saral [21] used CMBs to analyze different volatile organic compounds (VOCs) when fully considering the influence of the atmospheric reaction kinetics with different oxidant free radicals, wind directions, and transportation distance. In summary, existing literature either combine CMB with other SIMs to improve the meaningfulness or supplement CMB attenuation factor with other additional monitored environmental factors. Besides, atmospheric aerosols are still the object of most studies.

In this study, the CMB model is applied to PAHs data of road dust. The photodegradation process in atmospheric migration is considered, and the parameters of migration days are calibrated. PMF is also used to verify and compare with CMB results.

2. Materials and methods

2.1. Samples and profiles information

In this study, the data set used includes 16 PAH concentrations in 30 surface street dust samples from Beijing City (Fig. 1). Detailed information about Beijing, sample acquisition and processing can be found in the work before [22]. Since a large number of literature indicate that the degradation of small molecular PAHs is very fast or easy to flourish, therefore only 12 kinds of middle and high rings PAHs are applied in this study, which is given in Table 1. PAHs characters are shown in Table 1. Among these concerning solubility and volatility, associated relative concentrations of BbF and BkF (i.e., the sum of the two isomers) were used as the abbreviation benzofluoranthene (BF)

During the operation of the CMB model, the source profiles imported were derived from our previous studies. Common sources of PAHs adsorbed on RS were studied. About 40 source samples were tested and they were separated into 11 types of sources. The mean PAH concentrations of each type are shown in section 3.1. They can kind of represent the actual potential sources of 30 RS samples used in the study.

2.2. Principle of CMB and PMF model

Both the basic principles of the CMB and PMF model are mass balance. It is assumed that the proportion of some pollutants in the same kind of pollution source is the same, and this combination is called "source profiles". The combination of contamination pollutants in samples taken, that is, the receptor, are the linear additions of these source profiles. Specifically, it can be expressed as the following equation [23,24]:

$$C_{ij} = \sum_{k=1}^{p} F_{ik} S_{kj} + E_{ij}, \left(i = 1, 2, 3, \ldots, m \right)$$ (1)

If there are m kinds of pollutants, n samples, p sources, C_{ij} is the concentration of an ith pollutant in sample j, F_{ik} is the concentration of an ith pollutant in source k, S_{kj} is the contribution rates of source k to sample j, E_{ij} is the error between calculations and measurements.

In CMB model applications, the C_{ij} and each F_{ik} were known values, using the principle of linear regression least square method, we can get the S_{kj} solution values which is close to the reality.

In PMF model applications, the F_{ik} and S_{kj} all need to be calculated, that is why large amounts of samples are needed. Because although S_{kj} is different, F_{ik} that makes up each C_{ij} is the same in a different sample. Then we can use the matrix composed of many groups of vectors to analyze
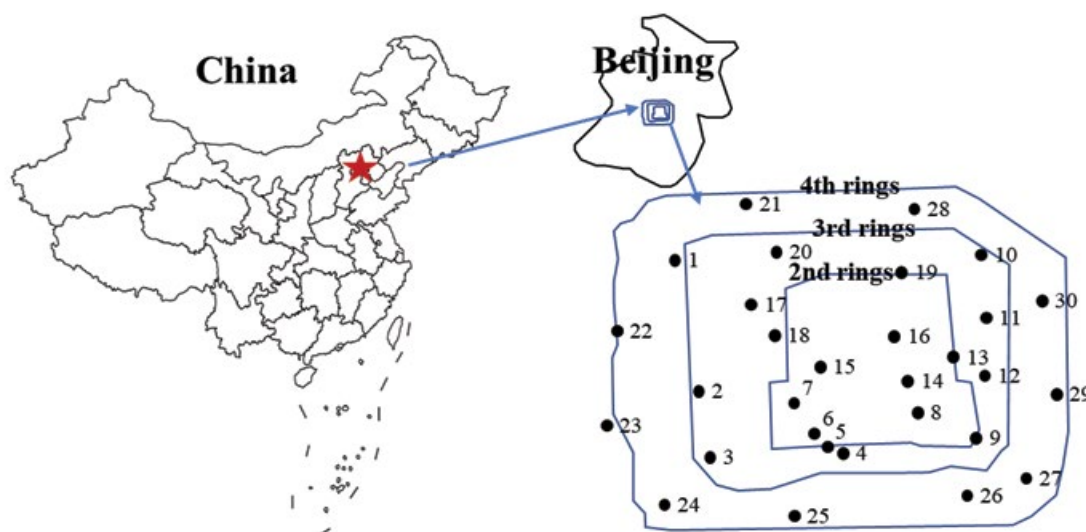
Fig. 1. Overview of sampling points.

Table 1
General information of 12 kinds of PAHs

Name	Abbreviation	Molecular weight (g/mol)	Rings
Fluorene	Flu	166.2	3
Phenanthrene	Phe	178.2	3
Anthracene	Ant	178.2	3
Pyrene	Pyr	202.3	4
Benzo[a]anthracene	BaA	228.3	4
Chrysene	Chr	228.3	4
Benzo[b]fluoranthene	BbF	252.3	5
Benzo[k]fluoranthene	BkF	252	5
Benzo[a]pyrene	BaP	252.3	5
Indeno[1,2,3-cd]pyrene	IND	276	6
Dibenz[a,h]anthracene	DbA	278.4	6
Benzo[ghi]perylene	BghiP/BgP	276	6

the eigenvalues, eigenvectors, and other related steps of factor analysis to obtain the common factors, that is, the source profiles.

The EPA-CMB 8.2 and EPA-PMF 5.0 developed by the Environmental Protection Agency were used to run the models.

2.3. Physical meanings connectivity between CMB and PMF model

CMB algorithm has six well-known assumptions: (1) composition of source emissions is consistent over the period of ambient and source sampling; (2) chemical species do not react with each other, that is, they add linearly; (3) all sources with potential for significantly contributing to the receptor have been identified and have had their "fingerprints" determined; (4) the compositions of different sources are linearly independent of each other; and (5)

measurement uncertainties are random, uncorrelated, and normally distributed. PAHs have many transformation pathways in the environment, especially under the influence of light and precipitation, different PAHs will have different reactions. However, in reality studies, the source samples which were tested to get source profiles are always obtained near or directly from source media. Therefore, once various pollutants react differently, the source components that migrate to the receptor must be inconsistent with the source profiles. Therefore, we call those profiles tested near sources and imported into CMB models as "original profiles (OP)".

On the other hand, similarly, it can be found that all the information inputted in PMF models are just the information related to receptors. This means that the source profiles we calculated by PMF just reflected the combinations of pollutants right in receptors. Those profiles might be lucky and just looked like "CMB Origin", if they don't transport for a while or they don't react much. Or some of the profiles might have been formed after a long period of migration and transformation. Maybe, those ones totally changed from what they used to be. We called all these profiles outputted by SDMs the "arrival profiles (APs)".

The relationship between AP and OP, based on the above theory, can be expressed as Fig. 2.

Considering this relationship, we can find that in AP, OP, and reactions of transporting these three segments, we can calculate one of them by the other two. In this study, we will discuss an improved CMB method considering the migration process of partial source profiles, which convert OPs to APs to make the simulation process closer to reality.

3. Results and discussion

3.1. APs calculated by PMF

As a comparison of the improved methods, a set of all untreated 11 source profiles was inputted into the CMB model to do operations. All 12 PAHs in 30 samples were
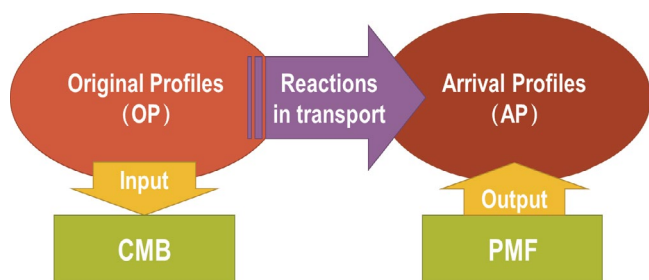
Fig. 2. Relationships between original and arrival profiles.

analyzed. In order to make the analytical results have sufficient physical meaning, we select the source elimination mode in CMB8.2, which can iterate repeatedly to make all contribution rate positive. It was found that all of the samples can be explained by at most seven sources. So, we set the factor number the PMF model dealt to be 7.

Table 2 showed that when the number of factors was 7, the fittings among measurements and calculations were basically good. The *R*-squares of linear fitting were all over 0.9, and the slopes and intercepts were also excellent.

The source profiles resolved by PMF is shown in Fig. 3.

Seven source profiles have been resolved successfully, which is quite a lot. Whereas there were still significant differences among 7 profiles, which may represent totally different 7 source types. This is one of the advantages of PMF. Because of the factor analysis theory that it relies on, the source profiles will be as orthogonal as possible. Those profiles are APs in our study. OPs we already got from our previous studies are shown in Fig. 4

Overall it can be found that generally, Phe and Chr were dominant molecules in PAHs of RS, and this is inseparable from the burning and chemical activities related process involved in a large number of human lives in the city [22]. From Figs. 3 and 4, we can confirm this situation. From Fig. 3, it can be seen that Phe has a high value in any OPs. Coal and biomass combustion mainly produce low ring PAHs, while typical pollution sources including coke

oven coking and other heavy industrial processes will produce more high ring PAHs. Our previous report also found that there was a relationship between sampling points and different monomers. For example, in the samples collected near the main road, the concentrations of pollutants from traffic sources such as BkF, BbF, BaP, IND, BgP, and DbA were much higher.

When those APs are compared with the OPs, some differences can be observed. In Figs. 3 and 4, the same color indicates that the high concentration characteristic molecules of these source profiles are similar, while the gray color means that there is no other source profile, no matter APs or OPs that has high similarity with it.

Firstly, compared with APs, there are many similarities among OPs. Especially, the values of the Phe of all sources are generally high. This is because many sources of PAHs include combustion processes, which are one of the main modes of production of Phe. From this point of view, this situation is in conformity with reality, but it will also lead to the enhancement of collinearity between sources, affecting the linear regression process. Fortunately, collinearity can be reduced by removing some profiles. Secondly, there is some similarity between the APs to the OPs, but it is not obvious. This proves that the hypothesis mentioned above has some truth, that is, there are some differences between AP and OP, probably because of the reaction of pollution, which cannot be ignored.

3.2. How kinetic behavior of non-volatile PAHs affected profiles similarity from APs to OPs?

Similarly, with aerosol, the migration pathways of PAHs in RS are mostly troughing atmospheric wind transmission then accumulating in RS grooves [25]. In this process, PAHs may react with various oxidants in the atmosphere during transportation. A large number of literature show that this reaction follows the first-order reaction law or pseudo-first-order reaction law [26], which can be explained by the following formulas:

$$\ln\left[\text{PAH}_j / \text{PAH}_{j0} \right] = -k \times t \tag{2}$$
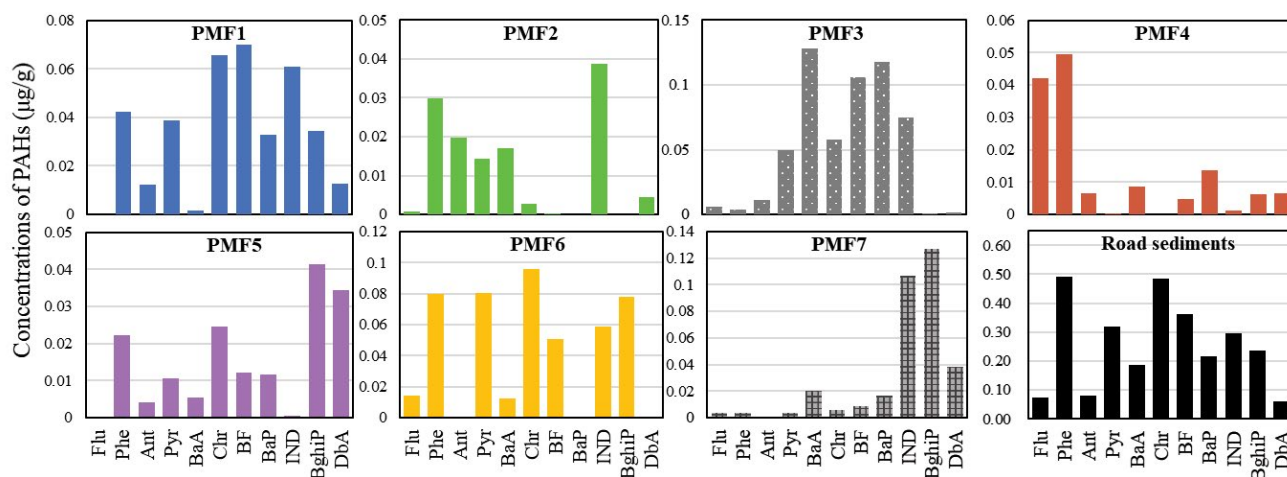

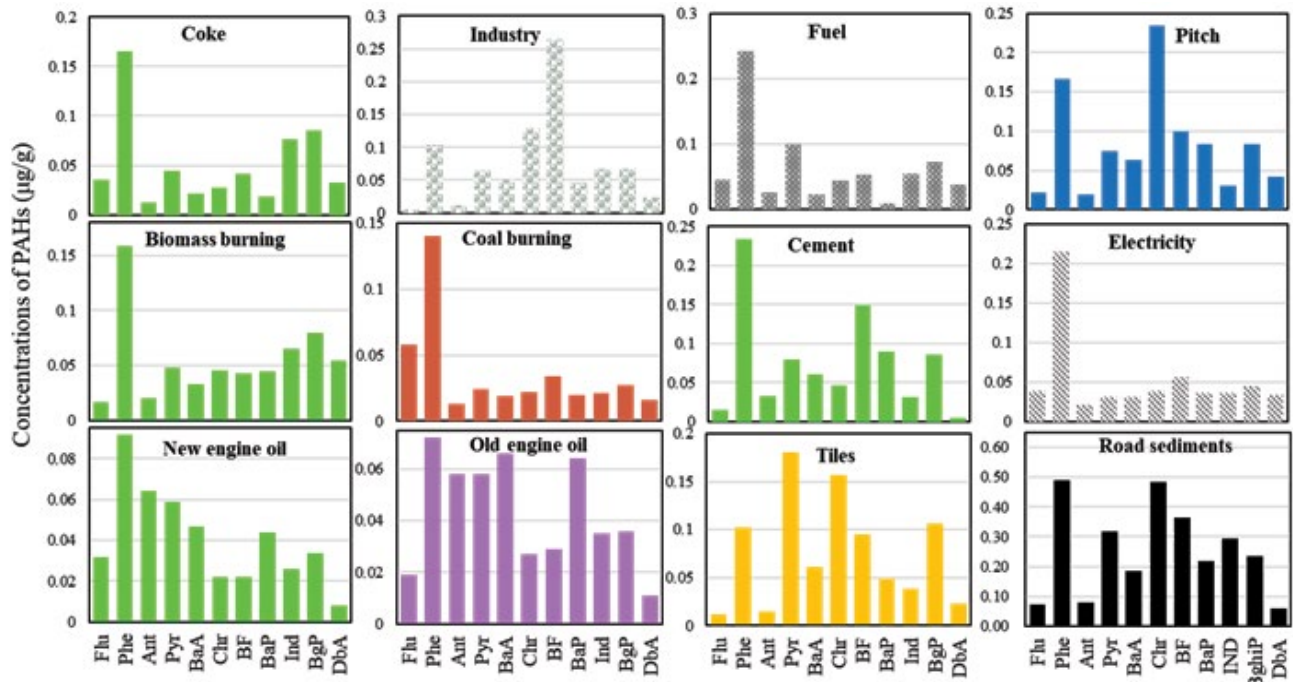Fig. 3. Profiles calculated by PMF models (arrival profiles).

Fig. 4. Profiles got from sample testing of different sources (original profiles).

Table 2
Fitting parameters between calculated and measured values calculated by PMF model

Species	Intercept	Slope	r^2
Flu	0.001	0.982	0.999
Phe	−0.004	1.007	0.943
Ant	0.003	0.942	0.992
Pyr	0.015	0.897	0.959
BaA	0.003	0.985	0.998
Chr	0.005	0.958	0.929
BF	0.003	0.976	0.961
BaP	0.000	0.993	0.994
IND	0.010	0.965	0.989
BghiP	−0.010	1.042	0.977
DbA	0.002	0.981	0.996

PAH_{j0} is the original concentration of the jth PAH. PAH_j is the concentration of jth PAH after t times reactions. The k is the rate constant of the first-order reaction of the jth PAH.

A complex situation is a case that the reaction rates of degradation kinetics of PAHs on aerosols are usually studied in laboratory experiments with pure particulate matter. Besides, the reactive light source conditions were sometimes ultraviolet light, and reactive oxides sometimes used a single species. The measured values under purely experimental conditions are not suitable for simulating natural processes. So, we select the aerosol attenuation coefficient studied under simulated natural conditions [27]. The k we used is shown in Table 3.

We assume that PAHs assemble as the OPs when they start transportation from source, and different PAHs will change after different reactions in the same period of time. Will those combinations change to be closer to the APs calculated by PMF? Therefore, in this part, the trend of OPs under different reaction times was studied and the cosine similarity value was introduced to evaluate the similarity between source profiles.

Cosine similarity is a measure of similarity between two non-zero vectors. Two vectors with the same orientation have a cosine similarity of 1; two vectors at 90° have a similarity of 0. The result of cosine similarity is neatly bounded between 0 and 1. The governing equation is:

$$\text{Cosine similarity} = \frac{A \cdot B}{\|A\| \cdot \|B\|} = \frac{\sum_{i=1}^{n} A_i B_i}{\sqrt{\sum_{i=1}^{n} A_i^2} \sqrt{\sum_{i=1}^{n} B_i^2}} \tag{3}$$

A and B are two vectors with the same dimension (n), A_i and B_i are the ith elements of each vector.

All 11 OPs were calculated changing 8 h interval so that not to miss key changes. The half-life period of 80% of the non-volatile PAH is less than 150 h [27], hence, we analyze the trend from 0 to 144 h. Because the reaction follows a pseudo-first-order reaction rate, it can be observed that the concentration of PAHs decreases regularly without the mutation. We choose some of the key time points for representative analysis. Cosine similarity analysis (Cos SA) was performed between OPs changed after 8, 48, 72, and 144 h, and the 7 APs calculated by PMF one by one. Table 4 shows the maximum cosine similarities of each

time group OP and the OP considere PAHs degradation with different reacting times (ODP). The OP types corresponding to the maximum Cos SA are also listed.

It can be found that with the increase of migration time, the value of the industrial source is closer and closer to PMF1. On the contrary, most other sources become more and more different from the APs as the migration time increases.

Most source similarity types have not changed except for PMF2. It can be seen that there is a certain similarity between the original coke and oil source profiles. At the same time, the cosine similarity of PMF7 and coke is getting lower, conversely. That is much lower than that of PMF2 and coke. At the same time, the similarity between Coke and PMF2 is increasing with migration, as shown in Fig. 5.

This indicates the possibility that PMF2 represents the true coke source, while PMF7 is not similar to any of the 11 source profiles considered in the study. It is temporarily recorded as unknown. Although most of the pollution sources have been covered by the OPs in our previous research, there are still unavoidable possibilities for some other sources. For example, the local soil parent material, nearby plant litter, and other natural substances may also mix into the RS samples, but that information was lacked.

This phenomenon can be reasonably explained. Some sources such as traffic sources even occur directly near the sampling points on the central roads of Beijing downtown. Besides, street sweeping is frequent, so these parts of PAHs

samples had not experienced degradation much. However, the industrial area is far away from the city center, and the emission positions are always top of high chimneys. It is more likely that the dust will fall to the sampling point through atmospheric migration. Therefore, the source profiles considering degradation is closer to the APs.

In terms of PAH monomers, taking coking coal and industry as examples, we analyze the concentration changes of different molecules with the set decay time as Fig. 6 shows.

It can be found that due to the reaction considered being a pseudo-first-order reaction, the attenuation process of both the total amount and monomer conforms to the logarithmic curve. Because of the different properties of PAHs, it can be found that the proportion of some monomers has changed a lot. For example, in the beginning, BgP had higher concentration than IND. Phe was definitely most notably in Coke source but after 144 h degradation, the concentration of IND was higher than BgP and even close to Phe. This means that if only using the original way to determine the meaning of the source according to the peak value molecule, it is very easy to get misjudgment without considering the attenuation.

Though the sum of 11 PAHs changed from 0.3922–0.9197 μg/g to 01229–0.3102 ng/μg/g, it can be found that max cosine similarities only changed about 1.4%–12.6%.

Table 3
Rate constants of the first-order reaction of some PAH

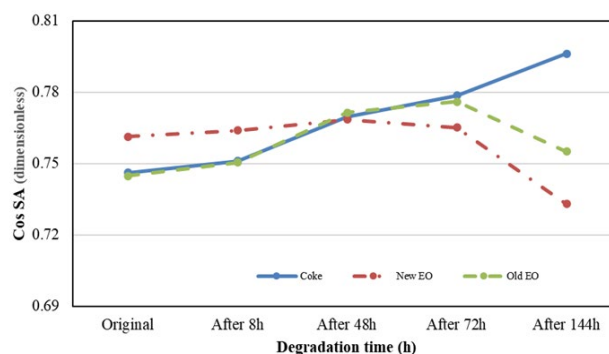| PAHs	k (h^{-1})	PAHs	k (h^{-1})
Flu	0.0067	BbF	0.0072
Phe	0.0083	BkF	0.0114
Ant	0.0132	BaP	0.0135
Pyr	0.0047	Ind	0.0065
BaA	0.0125	BgP	0.012
Chr	0.0051	DbA	0.007

Fig. 5. The tendency of cosine similarities (dimensionless) changing between some field measured sources and PMF2 profiles.

Table 4
Cos SA of origin profiles, degradable profiles and PMF profiles

Profiles from PMF results		PMF1	PMF2	PMF3	PMF4	PMF5	PMF6	PMF7
Original	Max Cos SA	0.9062	0.7614	0.7472	0.9182	0.7945	0.9359	0.5933
	Type	Industry	New EO	Old EO*	Coal	Biomass	Tiles	Coke
After 8 h	Max Cos SA	0.908	0.764	0.7447	0.9181	0.7918	0.9354	0.5884
	Type	Industry	New EO	Old EO	Coal	Biomass	Tiles	Coke
After 48 h	Max Cos SA	0.9148	0.7714	0.7249	0.9165	0.7763	0.9262	0.5657
	Type	Industry	Old EO	Old EO	Coal	Biomass	Tiles	Coke
After 72 h	Max Cos SA	0.9175	0.7787	0.708	0.9144	0.7661	0.9164	0.5524
	Type	Industry	Coke	Old EO	Coal	Biomass	Tiles	Coke
After 144 h	Max Cos SA	0.9192	0.7964	0.6641	0.9023	0.7333	0.8787	0.5183
	Type	Industry	Coke	Industry	Coal	Biomass	Tiles	Coke
Final source	Source	Industry	Coke	Old EO	Coal	Biomass	Tiles	Unknown

*engine oil (EO))

Therefore, although considering degradation will cause a great change in the shape of the source profiles, it has little effect on the similarity between most arrival and degradation profiles.

3.3. What performance of original/degradation/APs were applied in the CMB model?

Considering the above problems, we believe that if we consider the degradation factor of the industry source and coke oven source, which indicates that the source may have experienced long-term migration before reaching the receptor location, it will be possible to effectively enhance the degree of simulation.

Therefore, we replace the corresponding OP with industrial source and coke oven source after 240 h degradation reaction and nine other OPs, input them into CMB models. According to the literatures, the degradation of more than 90% PAHs in the atmosphere is less than 250 h, no matter in light reaction or dark reaction. At the same time, we also find that the Cos SA value of Coke and Industry increases with the decay time increase, and there is no turning point within 240 h. So, under this setting, the results were recorded as "CMB degradation", also we inputted 7 AP calculated by PMF into CMB model, the results of them were recorded as "PMF input CMB".

Contribution rates of PAHs from 30 samples analyzed using the three methods mentioned above are recorded in Fig. 7. In order to evaluate the effect of each method, we use the R-square of the CMB model to evaluate the results. The R-square corresponding to each method is shown in Fig. 8.

The source contribution rates of all 30 sample points can be calculated by any set of the source profile combinations mentioned above (Fig. 7). At the same time, the fitting rate is reasonable (Fig. 8).

Firstly, in the analysis of source contribution rate, the similarities among "CMB origin" and "CMB degradation" were much more than that of those two with PMF input with CMB. For differences among CMB origin and CMB degradation part, the difference between the two can be said to be very small, especially in the category of contributing

sources, the contribution rates of most of the samples did not change too much. Some samples like 5, 7, 23, the substitution of the source species may be due to the changes of collinearity after the degradation of the source profiles being considered. While the contributions rates in the "PMF input CMB" group, most samples were different from those contributions in former CMB methods. This problem can be explained with Table 3 and there was still some divergence between most APs and OPs, even considering degradations. The coal-burning and industry sources with the largest contribution rate have little difference, while the coke sources have changed greatly. From the change of PMF2 similar source, we can also find that there is a certain similar relationship between coke source and other sources, that kind of confusion may be the reason.

Secondly, comparing those R-square of three methods, in the aspect of all samples' fittings, CMB Degradation was the one performed best with no doubt. The R-square of PMF input CMB was mostly higher than CMB origin. Moreover, although the R-value is more discrete, there were not much low abnormal values, unlike the two methods of input OPs. However, in any case, the fitting degree of these three methods are all above 0.75, and most of them are above 0.9, which proved that all three methods can simulate the actual situation well. The existed differences among them were due to similarities in diverse profiles. Also, the mixture sources in APs run by PMF and uncertainties in the simulations of degradation could also be the reason.

Comparing with other researchers' conclusions about the combined receptor model, Shi et al. have been committed to exploring the direction of the PMF-CMB combined model [17,28,29]. Their original intention of developing such models is to solve the unsolved problem caused by the high incidence of collinearity in CMB. Our results confirm their view that PMF-CMB is easier to get the optimal solution than CMB alone. There are still some problems in PMF-CMB combined model. The PMF-CMB solutions with negative or large positive F-peaks often got unsatisfactory results. This is due to the unreasonable extracted profile of mixed sources on the first stage. F-peak, as a process of finding local extremum, its result depends on the data set being analyzed. Therefore, the PMF-CMB method still requires
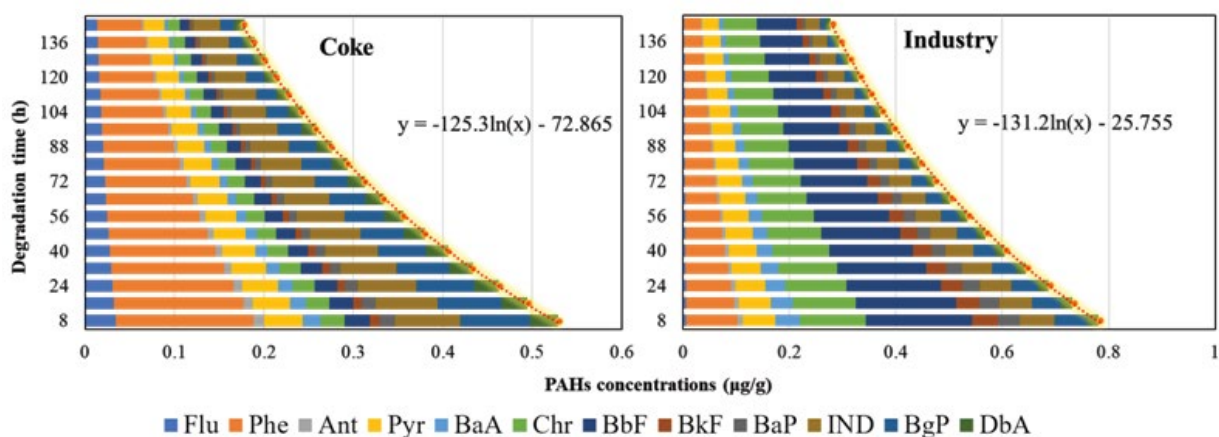

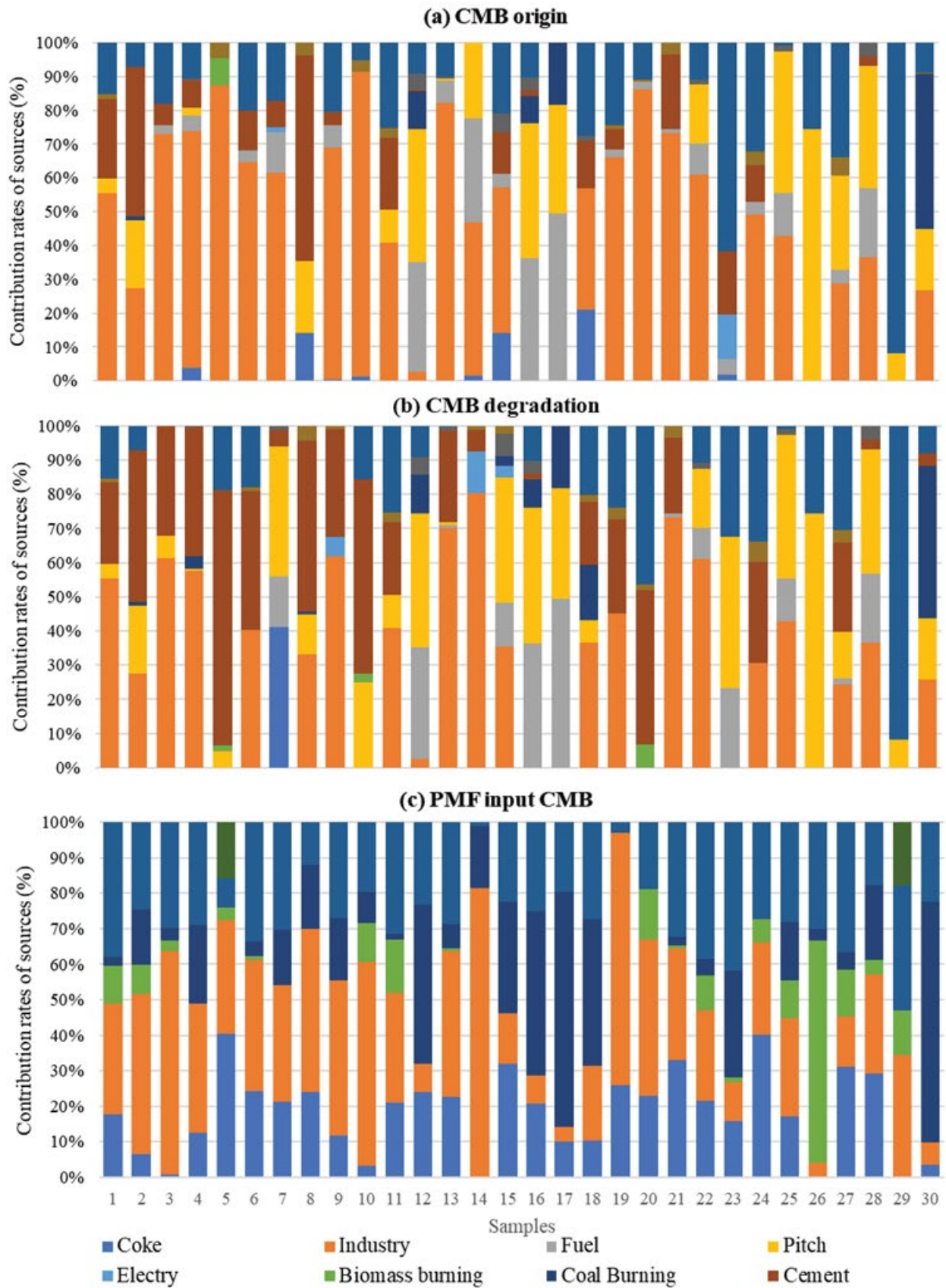Fig. 6. Trends of different PAHs changing with decay time.

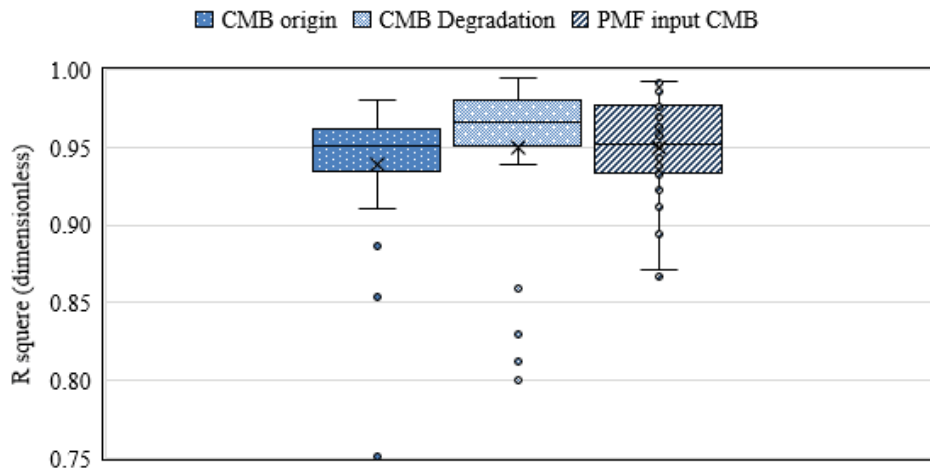Fig. 7. Source contributions calculated by three different CMB treatment methods.

Fig. 8. The fitting degree between measurements and simulations obtained by three different CMB treatment methods.

data quality, especially the relationship between samples cannot be affected by too much uncertainty. However, the CMB degradation model proposed in this paper has no requirements for the relationship between samples while improving the solution optimization and avoiding collinearity. Of course, we recognized that more cases are needed to confirm the reliability of this method. However, we still believe that the SDM model CMB has the part of the potential to be developed that the SIM model does not have.

4. Conclusions

There were much differences between the AP obtained by PMF and OPs inputted into CMB. This is partially due to the migration and transformation of various pollutants in the processes. The differences among APs were obvious, while the similarity among OPs was high, and the existence of collinearity was not conducive to the process of source analysis, but in fact, OPs were closer to the actual situation. Selecting some profiles which migrate from a source in real process and consider the first-order reaction process of PAHs will effectively improve the similarity between the original and APs of PAHs. In the case of this paper, we consider the industrial and coke oven sources which are far from the urban central road. And in practice, when we use CMB to calculate the source contribution rates of each single sample point, we can effectively improve the accuracy of the source resolution.

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