

Levels and Sources of PAHs in Air-borne PM_{2.5} of Hefei City, China

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Abstract This work studied the concentrations and sources of polycyclic aromatic hydrocarbons (PAHs) in air-borne particulate matter of Hefei, China. Samples of PM_{2.5} were collected daily at two sites during May, 2014, and January, 2015. The average daily concentration of PM_{2.5} was 96.88 $\mu\text{g m}^{-3}$, which is higher than the 2012 China Ambient Air Quality Standard (GB3095-2012 24-h grade II) of 75 $\mu\text{g m}^{-3}$. The concentrations of 16 EPA priority polycyclic aromatic hydrocarbons (PAHs) were analyzed by gas chromatography-mass spectrometry. The PM_{2.5}-bound PAH concentrations ranged from 4.92 to 71.00 ng m^{-3} (mean = 21.34 ng m^{-3}), and exhibited obvious seasonal (31.38 ng m^{-3} in winter and 14.05 ng m^{-3} in summer) and spatial variability (27.23 ng m^{-3} at site ME and 18.20 ng m^{-3} at site MS). Meteorological conditions such as ambient temperature, wind speed and humidity had influences on the concentrations of PAHs. As an index for PAH carcinogenicity, the annual average concentration of benzo(a)pyrene ranged from 0.46 to 2.31 ng m^{-3} , with a mean of 1.15 ng m^{-3} . This mean was lower than the China Ambient Air Quality Standard (GB3095-2012) of 2.5 ng m^{-3} . The diagnostic PAH ratios and principal component analysis (PCA) suggested that combustion of coal and vehicle emissions were the main sources of PAHs in PM_{2.5}.

Keywords Polycyclic aromatic hydrocarbons · PM_{2.5} · Meteorology · Sources apportionment

Air-borne particulate matter plays an important role in climate and atmospheric chemistry, as it contains various harmful species that may affect human health (Talifou et al. 2015). In particular, the particulate matter with aerodynamic diameter less than 2.5 μm (PM_{2.5}), attracts public attention around the world, because it is deposited throughout the human respiratory tract, causing heart and lung diseases (Pui et al. 2014). Numerous publications have revealed that polycyclic aromatic hydrocarbons (PAHs) can be highly enriched in PM_{2.5} (Masih et al. 2012), due to their low solubility, low reactivity and moderate vapor pressure (Chang et al. 2006). Particulate-phase PAHs are considered to be a significant hazard to human health (Hong et al. 2007).

PAHs are compounds with two or more aromatic rings, which are mainly derived from the incomplete combustion of carbon enriched materials (Ma et al. 2010a). Although some natural sources (volcanoes and forest fires) could contribute to the PAH burden, anthropogenic sources, such as combustion of fossil fuels, residential heating, and coke production are by far the major sources of PAHs (Li et al. 2014).

Hefei is the capital city of Anhui province, China. In recent decades, Hefei has faced accelerated development of economy, industrial facilities, urban construction and public transportation. In this study, PM_{2.5} samples were collected systematically from Hefei city and their PAHs were measured. The main aims of this study are to: (1) investigate the spatial and seasonal variations of PAHs, (2) understand the controls of meteorological parameters on the enrichment and distribution of PAHs, and (3) identify

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the possible sources of PAHs. We aimed to provide a case to understand the organic pollution in particulate matter at developing cities.

Materials and Methods

Hefei City (E117°27', N 31°86') is the economic and cultural center of Anhui province. It is located in the middle of the province, covering an urban area of 1,100 km². It had 4.86 million residents and more than 1.2 million vehicles in 2015 (Deng et al. 2016). Hefei has a subtropical humid monsoon climate, with four seasons defined as follows: spring (March–April), summer (May–August), fall (September–November) and winter (December–February). PM_{2.5} was sampled simultaneously at two locations of Hefei: Site A (MS) located at a middle school, northeast of First Ring Rd, Yaohai district, adjacent to the residential area; Site B (ME) located at west section of Fanhua Avenue, Shushan district, adjacent to a road with a very high traffic volume (Fig. 1).

A middle volume air sampler (LaoYing 2030, Laoshan Electronic Instrument, Qingdao, CN) was used to collect the PM_{2.5} on a quartz fiber filter (Whatman EPM2000, UK, diameter=90 mm), at a flow rate of 100–120 L min⁻¹ for approximately 20 h. In total, samples were collected at the time intervals of 20–28 May 2014 in summer, and 16–23 January 2015 in winter. Before sampling, all filters were baked in a muffle furnace at 500°C for 4 h to eliminate

the background organic matter. Filters were weighed on a microbalance to ±0.001 mg (Sartorius MSA 3.6P-000-DM, Gottingen, DE) on a clean bench before and after sampling to calculate the mass of collected PM_{2.5} samples. Upon collection, the filters were stored in clean containers kept at 4°C.

Each filter was cut into small pieces (1 cm × 1 cm) using stainless steel scissors, and then was extracted for PAHs by dichloromethane (DCM) and acetone (chromatographically pure grade) in an accelerated solvent extraction apparatus (Dionex-ASE 350, Sunnyvale, CA, USA). The extracts were concentrated to 1 mL by automatic parallel concentrator (MultiVap-8, LabTech, Hopkinton, MA, USA) before analysis with a gas chromatograph (GC, Agilent Technologies 7890, Wilmington, DE, USA) equipped with a gas chromatograph Agilent HP-5MS, non-polar 30 m × 0.25 mm capillary column in tandem with a mass spectrometer (MS, 5975 N).

The helium carrier gas was set at a flow rate of 1.0 mL min⁻¹. Each sample run was programmed as follows: starting temperature of 100°C for 2 min, ramping to 200°C at a rate of 20°C min⁻¹, ramping to 300°C at a rate of 4°C min⁻¹ and keeping for 10 min.

Sixteen priority PAH compounds specified by the Environmental Protection Agency, USA (USEPA), were analyzed, including: NAP—naphthalene; ANY—acenaphthylene; ANA—acenaphthene; FL—fluorene; PHE—phenanthrene; An—anthracene; FLU—Fluoranthene; PYR—pyrene; BaA—benz[a]anthracene; CHR—chrysene;

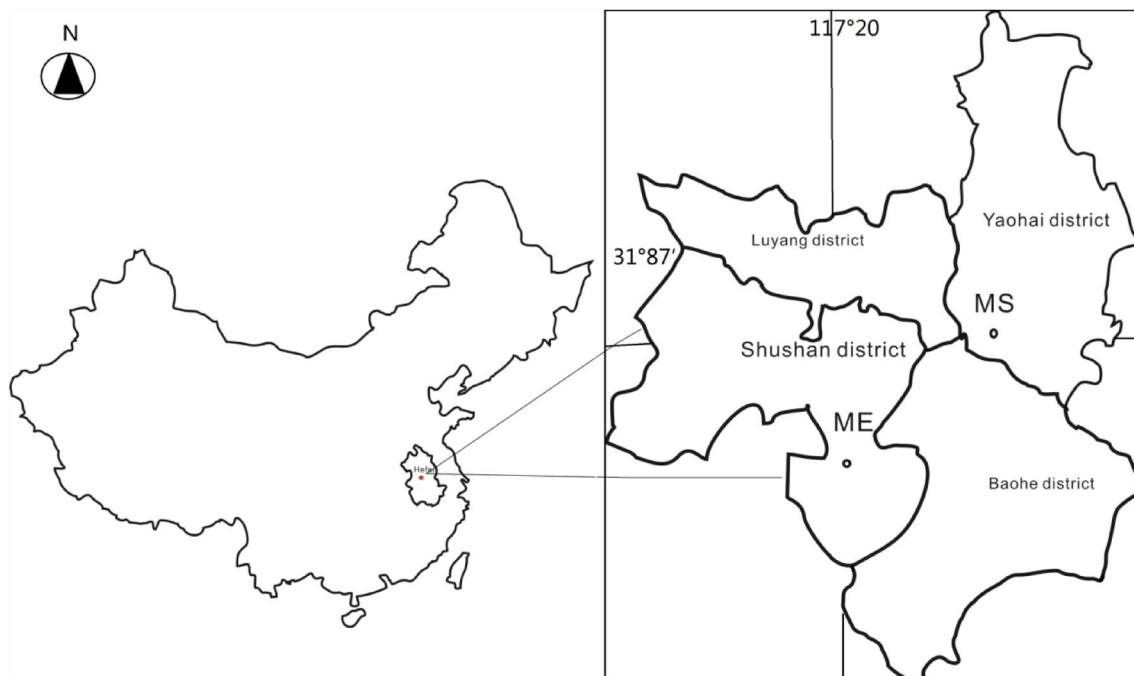


Fig. 1 Sketch map showing the PM_{2.5} sampling locations (ME, MS), Hefei, Anhui, China

BbF—benzo[b]fluoranthene; BkF—benzo[k]fluoranthene; BaP—benzo[a]pyrene; IcdP—indeno[1,2,3-cd]pyrene; DBA—dibenzo[a,h]anthracene; and BghiP—benzo[ghi]perylene.

Field blank, method blanks (solvent) and spiked blanks (standards spiked into solvent) were analyzed by the same procedure. There were no significant background interferences. In addition, surrogate standards were added to all the samples to monitor procedural performance and matrix effects consisting of phenanthrene-d10, chrysene-d12, and perylene-d12 (Aldrich Chemical, Gillingham, Dorset, UK). The mean recoveries (%) of the three surrogates in field samples were within the acceptable range (75%–128%). The calibration curves were linearly fitted with correlation coefficients (R^2) from 0.996 to 1 for all of the PAHs when conducting the external calibration. PAH concentrations were corrected for recovery efficiency during extraction.

Pearson correlation and principal component analysis (PCA) and correlation analysis were performed using SPSS version 17.0 software packages (IBM, Somers, NY, USA). Meteorological data were obtained from the Weather Bureau of Anhui Province, and are listed in Table 1.

Results and Discussion

At ME and MS sampling sites, the daily concentrations of $PM_{2.5}$ reached 121.75 ± 26.96 and $104 \pm 21.75 \mu\text{g m}^{-3}$ in winter, whereas in summer, they were 100.44 ± 44.37

and $64.89 \pm 30.78 \mu\text{g m}^{-3}$, respectively. The average daily value of $PM_{2.5}$ concentration was $96.88 \pm 32.46 \mu\text{g m}^{-3}$, higher than the National Ambient Air Quality standard for $PM_{2.5}$ (NAAQS-2012 24-h) of $35 \mu\text{g m}^{-3}$ (Chen et al. 2015; You 2014). It was also higher than the China Ambient Air Quality Standard (GB3095-2012 24-h grade II) of $75 \mu\text{g m}^{-3}$ (Brunner 1985; China 2012).

Mean concentrations of $PM_{2.5}$ -bound individual PAHs are shown in Table 2. During sampling times, the total mean \sum PAH concentrations ranged from 4.92 to 71.00 ng m^{-3} in Hefei, with a mean of 21.34 ng m^{-3} . This mean is lower than those in other large Chinese cities such as Beijing (112.7 ng m^{-3} in 2003), Qingdao (87.5 ng m^{-3}) and Guangzhou (117.4 ng m^{-3}), but is obviously higher than those in the developed countries/regions such as London (17.23 ng m^{-3}), Hong Kong (7.42 ng m^{-3}), and Los Angeles (2.46 ng m^{-3}) (Baek et al. 1992; Fraser et al. 1998; He et al. 2006; Zheng and Fang 2000). Benzo(a)pyrene has been categorized as a carcinogenic PAH, and is often used as an indicator of human PAH exposure. In this study, the mean BaP concentration was 0.46 to 2.31 ng m^{-3} (mean = 1.15 ng m^{-3}), which was lower than the maximum allowable annual average BaP concentration of 2.5 ng m^{-3} , according to the China Ambient Air Quality Standard (GB3095-2012) (China 2012). Our BaP concentration was comparable to the published results for other Chinese cities, and was lower than value for Beijing (17.8 ng m^{-3}), Guangzhou (2.98 ng m^{-3}) and Harbin (2.5 ng m^{-3}) (Liu et al. 2015; Ma et al. 2010b; Wang et al. 2008).

The spatial and seasonal variations in PAH concentrations for the two sampling sites are shown in Table 2. The overall mean \sum PAHs in the $PM_{2.5}$ at ME site (27.23 ng m^{-3}) was higher than that at MS site (18.20 ng m^{-3}). This was largely due to the 2.2-fold higher winter values at site ME. This might relate to the locations of the sampling sites. The ME site is located in the economic development zone, the major industrial area of Hefei. The sampling site was adjacent to a road with a high volume of traffic. Miguel and Pereira (1989) noted that PAHs were the major components of vehicle exhausts, especially gasoline- and diesel-powered vehicles. On the other hand, the MS site was located at a middle school, where there was a mixture of residential, commercial and industrial areas, with less traffic volume and industrial activity than at site ME.

The seasonal variations of PAHs in $PM_{2.5}$ of Hefei are presented in Table 2. At both sites combined, the mean concentration for \sum PAHs was higher in winter (31.38 ng m^{-3}) than in summer (14.05 ng m^{-3}). Similar seasonal contrasts in $PM_{2.5}$ -bound PAHs have been reported in other areas (Guo et al. 2003; Wang et al. 2015). For example, Guo et al. (2003) found that the ratio of PAHs in winter to summer was in the range of 7.5–8.6. We speculate that this variation might relate to meteorological conditions

Table 1 Meteorological parameters during $PM_{2.5}$ sampling periods

Date	Temperature (°C)	Relative humidity (%)	Wind speed (m s^{-1})
20/05/2014	22.2	93.0	1.5
21/05/2014	24.0	74.0	1.0
22/05/2014	26.2	59.8	2.4
23/05/2014	24.0	66.0	2.0
24/05/2014	24.3	80.0	2.0
25/05/2014	26	82.1	2.1
26/05/2014	27.2	49.8	2.0
27/05/2014	26.5	51.9	2.4
28/05/2014	28	58.2	1.5
16/01/2015	6.1	76.5	1.6
17/01/2015	3.2	53.0	1.0
18/01/2015	4.0	62.1	1.5
19/01/2015	5.5	60.1	1.2
20/01/2015	9.4	44.9	1.6
21/01/2015	6.1	60.0	2.0
22/01/2015	4.1	55.0	1.0
23/01/2015	3.0	71.3	1.0

Table 2 Mean concentrations (ng m⁻³) of individual PAHs in PM_{2.5}

	MS		ME	
	Summer (n = 18)	Winter (n = 16)	Summer (n = 18)	Winter (n = 16)
NAP	0.92 (0.92–1.00)	2.17 (0.75–2.92)	0.93 (0.12–1.50)	2.47 (2.25–2.83)
ANY	nd (nd-nd)	0.03 (nd–0.08)	nd (nd-nd)	0.08 (nd–0.17)
ANA	0.02 (nd–0.08)	0.22 (nd–0.08)	0.02 (nd–0.08)	0.03 (nd–0.08)
FL	0.10 (0.08–0.17)	0.11 (0.08–0.17)	0.10 (0.08–0.17)	0.25 (0.08–0.50)
PHE	0.77 (0.58–0.92)	1.05 (0.58–1.75)	0.69 (0.42–1.00)	2.28 (0.50–5.08)
An	0.04 (nd–0.08)	0.08 (0.08–0.08)	0.02 (nd–0.08)	0.11 (nd–0.25)
FLU	0.73 (0.50–1.00)	1.55 (0.83–2.08)	0.69 (0.25–1.25)	4.81 (0.58–11.17)
PYR	0.63 (0.5–0.83)	1.31 (0.67–1.83)	0.54 (0.33–1.00)	3.58 (0.50–7.83)
BaA	0.36 (0.25–0.43)	1.03 (0.42–2.00)	0.21 (0.17–0.33)	1.03 (0.42–2.00)
CHR	0.77 (0.58–0.92)	1.69 (0.83–2.75)	0.61 (0.25–0.92)	4.25 (0.50–7.75)
BbF	2.88 (1.08–4.08)	2.33 (1.08–4.25)	2.29 (0.25–3.75)	4.81 (2.67–7.75)
BkF	2.88 (1.08–4.09)	2.33 (1.08–4.25)	2.29 (0.25–3.75)	4.81 (2.67–7.75)
BaP	0.88 (0.50–1.33)	0.94 (0.67–1.33)	0.46 (0.25–0.67)	2.31 (0.67–3.33)
IcdP	2.77 (2.42–3.50)	2.58 (1.92–3.00)	1.31 (0.75–1.83)	7.22 (1.92–1.75)
DBA	0.85 (0.33–2.50)	0.31 (0.17–0.42)	0.19 (0.17–0.25)	0.67 (0.17–1.08)
BghiP	2.00 (1.83–2.5)	1.81 (1.42–2.08)	0.96 (0.50–1.33)	3.04 (1.08–6.62)
∑PAHs	16.68 (12.49–19.82)	19.71 (5.32–23.75)	11.41 (4.92–16.07)	43.04 (14.19–71.00)

nd non-detected, NAP naphthalene, ANY acenaphthylene, ANA acenaphthene, FL fluorene, PHE phenanthrene, An anthracene, FLU Fluoranthene, PYR pyrene, BaA benz[a]anthracene, CHR chrysene, BbF benzo[b]fluoranthene, BkF benzo[k]fluoranthene, BaP benzo[a]pyrene, IcdP indeno[1,2,3-cd]pyrene, DBA dibenzo[a,h]anthracene, BghiP—benzo[ghi]perylene

and human activities. In summer, the high temperature and intense sunlight could enhance the degradation of PAHs in PM_{2.5}, while in winter, the increased domestic heating and consumption of fuels could release PAHs to the atmosphere (He et al. 2006; Tang et al. 2005).

Correlations between meteorological parameters and the concentrations of individual PAHs were calculated to investigate possible relationships (Table 3), in order to investigate the influence of meteorological conditions on the distribution of PAHs. We found a negative correlation between the concentrations of PAHs and ambient temperature. This was largely attributed to the influence of increased emissions during the winter season, with particulate-bound PAH concentrations increasing due to the consumption of fuels associated with domestic heating (Tham et al. 2008). This also can be ascribed to the degradation rate of PAHs commonly being faster under higher temperatures and stronger solar radiation. In this study, the concentrations of PAHs have no significance correlations with the wind speed and relative humidity.

The PAHs of PM_{2.5} could derive from various emission sources. The possible sources of PAHs in air can be identified by diagnostic concentrations and PCA (principal component analysis) of some marker PAH compounds.

The concentrations of a group of PAHs, or specific PAHs compounds, have been used to identify the emission sources (Yuan et al. 2015), such as BaA and CHR, An,

Table 3 Correlations between concentrations of PAHs (ng m⁻³) and meteorological parameters

R	All samples(n=28)		
	Temperature	Wind speed	Relative humidity
NAP	-0.73**	-0.39	-0.10
ANY	-0.47*	-0.30	-0.46
ANA	-0.26	-0.13	-0.30
FL	-0.30	-0.07	-0.21
PHE	-0.31	-0.09	-0.45
An	-0.42*	-0.15	-0.38
FLU	-0.38	-0.19	-0.41
PYR	-0.42	-0.24	-0.41
BaA	-0.58*	-0.44	-0.38
CHR	-0.51	-0.35	-0.41
BbF	-0.15	-0.06	-0.51*
BkF	-0.15	-0.06	-0.51*
BaP	-0.49*	-0.33	-0.39
IcdP	-0.46*	-0.28	-0.39
DBA	-0.09	-0.26	-0.22
BghiP	-0.36	-0.25	-0.16
∑PAHs	-0.40*	-0.01	-0.28

ns: not significant; ** $p < 0.01$; * $p < 0.05$

FLU, PYR, for coal combustion; An, BaP and BghiP for coke production(Cai et al. 2014); FLU and PYR for combustion of wood (Tekasakul et al. 2008); and FLU for oil burning (Khalili et al. 1995). However, in different types of emission source profiles, some overlaps in similarity have been noted for these marker PAHs. (Hu et al. 2012).

Thus, we have used diagnostic ratios, such as (FLU/[FLU + PYR], BaA/[BaA + CHR], An/[An + PHE], BaP/BghiP and IcdP/[IcdP + BghiP]) to appoint the sources of PAHs in our case study (Table 4). Different values of each diagnostic ratio correspond to characterized emission sources. These diagnostic ratios are all indicators of human activities, with coal combustion and motor vehicle emissions being the primary sources of PAHs in PM_{2.5} of Hefei.

Principal component analysis was used to aid in the interpretation of PAH emission sources by reducing the large number of variables to a smaller number of factors. Each of the factor from the PCA is associated with source characterization by its most representative PAH compound(s) (Saxena et al. 2014). Source groupings were determined by PCA with varimax rotation, and the retention of principal components having eigen values higher than 1 of the complete data set of PAHs concentrations. In addition, loadings determined the most representative PAH compounds in each factor, with values higher than 0.5 being selected (Hu et al. 2012).

Three factors accounted for 84.03% of the total variance of the data (Table 5), with factor 1 explaining 63.26% of the variance. This factor showed high loading for ANY, FL, PYR, PHE, CHR, FLU, An and IcdP. Factor 2 explained 11.82% of the variance, and showed a high loading for BghiP. As summarized by Harrison et al. (1996), FLU, PYR and CHR are considered to be the tracers of coal combustion; and IcdP, PHE and BghiP to be the tracers of vehicle exhaust(Harrison et al. 1996). The lower molecular weight PAHs, such as FL and An, might be formed in processes during the pyrolysis of fossil fuels (Dachs et al. 2002; Jung et al. 2015). Therefore, this suggested that Factor 1 was representative of pyrogenic sources, including coal burning (power plants and space heating) and vehicle emissions, and Factor 2 was representative of vehicle

exhaust. The appointed sources of PCA analysis are consistent with diagnostic ratios. Factor 3 only explained 8.95% the variance. It showed a high loading for DbA, a tracer for coke oven sources (Simcik et al. 1999).

In conclusion, concentrations of 16 PAHs were measured in the PM_{2.5} of Hefei during the periods of 20–28 May, 2014, and 16–20 January, 2015. The concentration of PAHs ranged from 12.49 to 23.75 ng m⁻³ at site MS and from 4.92 to 71.00 ng m⁻³ at site ME. A strong seasonal trend in PAH concentration was observed during the sampling period, with a higher level in winter than in summer. Meteorological conditions influenced the PAH

Table 5 Factorial weights matrix for the source analysis of PAHs in Hefei

PAHs	Factor1	Factor2	Factor3
ANY	0.973	-0.026	-0.52
FL	0.967	-0.020	0.116
PYR	0.966	0.550	0.120
PHE	0.964	-0.152	0.084
CHR	0.940	0.280	0.079
FLU	0.889	-0.004	0.200
An	0.881	0.081	-0.118
IcdP	0.87	0.459	0.127
BaA	0.683	0.421	0.03
BaP	0.846	0.507	0.106
BbF	0.771	-0.206	0.467
BkF	0.771	-0.206	0.467
NAP	0.597	-0.081	-0.487
BghiP	0.172	0.809	-0.079
ANA	0.287	-0.614	-0.375
DBA	0.224	0.096	0.697
Variance%	63.26	11.82	8.95
Cumulative%	63.26	75.07	84.03
Possible source	Pyrogenic sources	Vehicle exhaust	Coke oven source

Extraction Method: Principal Component Analysis

Rotation Method: Varimax with Kaiser Normalization

Eigenvalue > 1.00

Table 4 Diagnostic ratios of PAHs in PM_{2.5} of Hefei

Diagnostic ratios	Summer MS	Summer ME	Winter MS	Winter ME
FLU/(FLU + PYR)	0.54	0.56	0.54	0.57
BaA/(BaA + CHR)	0.32	0.26	0.38	0.34
An/(An + PHE)	0.09	0.1	0.07	0.07
BaP/BghiP	0.44	0.48	0.52	0.76
IcdP/(IcdP + BghiP)	0.58	0.57	0.59	0.7

Reference source emission:FLU/(FLU + PYR) < 0.4: unburned petroleum, 0.4–0.5:combustion of fuel, >0.5 coal and wood; BaA/(BaA + CHR) > 0.35: combustion, 0.2–0.35:mixed sources, <0.2 petrogenic; An/(An + PHE) > 0.1: combustion, <0.1:petroleum sources; BaP/BghiP 0.3–0.4:traffic, 0.4–0.9 traffic and combustion, 0.9–6.6 coal combustion; IcdP/(IcdP + BghiP) < 0.4:traffic, >0.5:coal combustion

concentrations, with air temperature and PAH concentration being inversely related. According to the analysis of emission source of PAHs based on diagnostic ratios and PCA, it was concluded that combustion of coal and vehicle exhaust were the predominant sources of PAHs in PM_{2.5} of Hefei.

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