Supplementary Material

Text S1 Source implication

Figure S1 shows the scatter plots of toluene to benzene ratio at ZY and JX. The toluene to benzene (T/B) ratio is often used to indicate the possible sources of AHs [29,35,76]. For example, the T/B value of ~2.0 implies vehicle exhaust [23,77], 0.6 indicates biomass burning [78], 0.7 indicates coal burning [31], and 4.3 indicates industrial solvent usage [76,79]. Significant correlations ($p < 0.01$) were observed between the two species at both sites. As shown in Figure S1, at both sites, all the samples were clustered near the lines of industrial solvent, vehicle exhaust, and biomass/coal burning. This indicated that AHs at both sites have the same emission sources of industrial solvent, vehicle exhaust, and biomass/coal burning. The average T/B ratios were 2.5 ± 1.2 and 1.9 ± 1.1 at ZY and JX, respectively, suggesting that vehicular emissions were the primary sources for AHs at the two sites.

Text S2 Identification of factor number

According to previous discussion, industrial solvent, vehicle exhaust, and biomass/coal burning were determined to be the dominant sources of AHs at ZY and JX. Then, three to six factors were tested with the input data, and bootstrap (BS) and displacement (DISP) analyses were run to evaluate the model results. The PMF model error estimation results are listed in Table S1. In PMF model, BS analysis was applied to identify whether a small set of observations existed that could disproportionately influence the solution [45]. The BS analysis was run for 100 times, and mapping over 80% of the runs indicated that the number of factors was appropriate. As shown in Table S1, when the factor numbers were three and four, all factors mapped for more than 80%. Moreover, no swap occurred with DISP, indicating that the solutions of three and four factors were stable and acceptable. For the solution of four factors, Q(robust) value (6906.4) was lower than that of three factors (7974.7), and the source profiles were more reasonable and interpretable. Hence, the solution of four factors was determined to be optimum.

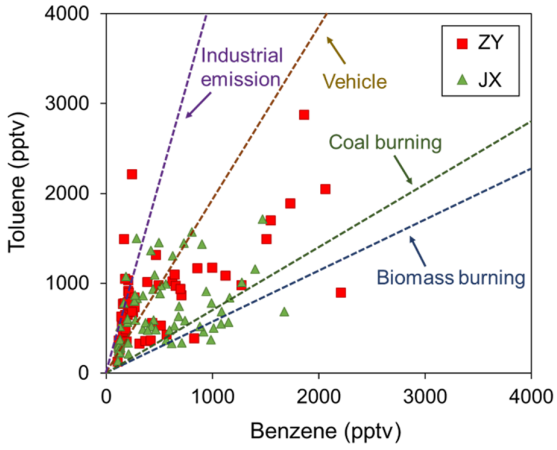

Figure S1. Scatter plots of toluene to benzene ratio at ZY and JX.

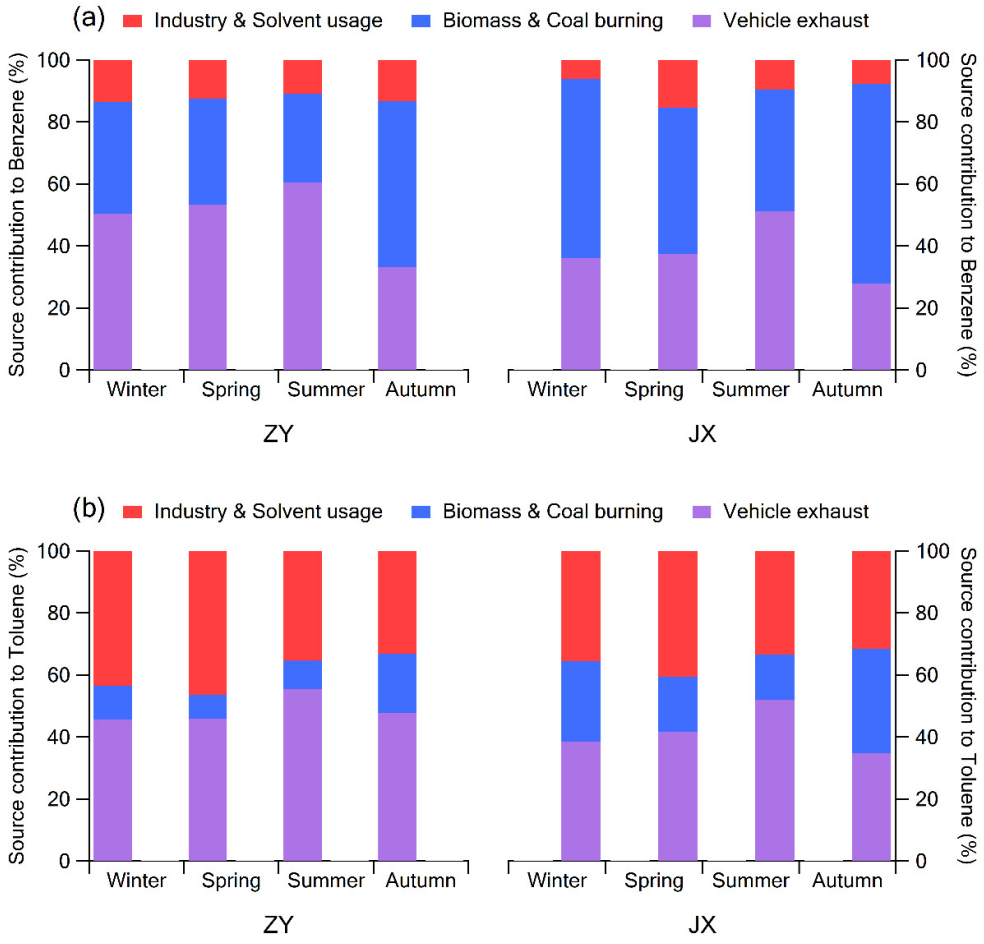

Figure S2. Source contributions to (a) benzene and (b) toluene in various seasons at ZY and JX.

Table S1. Method detection limit (MDL), range (minimum-maximum) and average concentration ± SD of VOC species other than AHs at ZY and JX during the sampling period (Units: pptv), ND = below detection limit.

Species	MDL	ZY (N = 66) *		JX (N = 67) *	
		Range	mean ± SD	Range	mean ± SD
ethene	39	217–8754	3456 ± 2599	496–8327	3372 ± 2277
ethyne	57	728–9669	3669 ± 3071	428–9865	3780 ± 3029
ethane	41	706–8378	2808 ± 1490	787–6057	2363 ± 1098
propene	31	79–1726	510 ± 384	118–1612	494 ± 317
propane	31	509–9735	3148 ± 2077	467–7011	2521 ± 1338
i-butane	21	385–7679	2469 ± 1863	111–6585	2030 ± 1710
1-butene	17	222–1611	552 ± 284	232–2490	673 ± 418
1,3-butadiene	7	14–746	191 ± 147	10–835	169 ± 141
n-butane	21	102–6621	2909 ± 1776	143–7461	1759 ± 1558
trans-2-butene	13	78–580	159 ± 102	27–254	128 ± 39
cis-2-butene	11	114–418	182 ± 62	87–232	160 ± 36
3-methyl-1-butene	2	19–146	74 ± 33	19–115	71 ± 31
i-pentane	14	99–5500	1121 ± 1097	15–4518	918 ± 683
1-pentene	20	22–632	227 ± 178	ND–217	77 ± 44
2-methyl-1-butene	3	25–1240	288 ± 145	13–577	131 ± 185
n-pentane	8	169–5173	1459 ± 1212	188–5087	1067 ± 994

isoprene	13	10–6828	1762 ± 1007	14–7828	2198 ± 1907
trans-2-pentene	10	9–1258	250 ± 158	23–1118	289 ± 328
cis-2-pentene	6	23–1419	353 ± 348	15–1088	354 ± 341
2-methyl-2-butene	2	5–298	85 ± 81	34–70	51 ± 9
2,2-dimethylbutane	14	35–97	57 ± 13	17–224	48 ± 41
cyclopentene	8	9–144	50 ± 34	15–41	27 ± 11
4-methyl-1-pentene	5	15–88	31 ± 17	17–438	80 ± 76
cyclopentane	8	27–279	86 ± 62	25–800	103 ± 154
2,3-dimethylbutane	12	31–1271	250 ± 371	12–1247	357 ± 235
2-methylpentane	8	98–1771	420 ± 307	9–1788	304 ± 332
3-methylpentane	7	7–2494	367 ± 422	22–1264	335 ± 202
2-methyl-1-pentene	5	18–629	238 ± 125	58–776	278 ± 136
n-hexane	6	104–895	353 ± 203	41–251	121 ± 41
methylcyclopentane	9	70–2783	950 ± 415	159–1109	297 ± 205
2,4-dimethylpentane	4	159–1106	281 ± 176	121–892	262 ± 159
cyclohexane	16	121–941	274 ± 154	21–1536	272 ± 262
2-methylhexane	6	21–629	207 ± 163	69–283	192 ± 78
2,3-dimethylpentane	9	178–771	394 ± 216	169–1150	535 ± 401
3-methylhexane	6	169–1098	553 ± 390	154–2528	458 ± 465
2,2,4-trimethylpentane	9	160–974	367 ± 236	163–353	194 ± 28
n-heptane	10	162–489	203 ± 58	146–242	190 ± 17
methylcyclohexane	8	156–786	225 ± 100	114–558	203 ± 88
2,3,4-trimethylpentane	6	114–339	194 ± 60	184–362	211 ± 25
2-methylheptane	4	184–253	210 ± 19	182–314	210 ± 26
3-methylheptane	5	183–244	207 ± 16	183–747	236 ± 82
n-octane	6	183–523	231 ± 62	80–437	172 ± 92
n-nonane	6	80–330	166 ± 85	91–771	213 ± 115
n-decane	6	91–760	204 ± 110	79–422	149 ± 88
α-pinene	6	81–645	151 ± 113	30–249	133 ± 63
β-pinene	6	30–372	141 ± 74	34–633	225 ± 145
HFC-134a	7	58–129	96 ± 17	30–161	87 ± 30
HFC-152a	7	ND–55	19 ± 12	ND–46	15 ± 13
HCFC-22	21	152–537	298 ± 164	61–530	196 ± 109
CFC-12	16	29–887	452 ± 219	116–1845	828 ± 464
CH_3Cl	20	131–2266	719 ± 440	316–2208	822 ± 577
CFC-114	2	ND–129	35 ± 27	ND–132	43 ± 32
H-1211	2	ND–12	4 ± 2	ND–25	7 ± 5
CH_3Br	2	ND–19	8 ± 4	ND–33	17 ± 8
CFC-11	16	51–479	276 ± 125	60–319	194 ± 153
HCFC-141b	16	32–256	94 ± 60	17–242	89 ± 56
CH_3I	6	ND–29	12 ± 4	ND–46	11 ± 9
CH_2Cl_2	3	229–4883	1480 ± 900	142–3524	1087 ± 607
CFC-113	7	58–104	82 ± 10	46–112	79 ± 12
$CHCl_3$	5	50–1205	352 ± 147	47–1916	297 ± 196
CH_3CCl_3	2	44–129	83 ± 22	69–138	89 ± 14
1,2-DCE	5	7–294	85 ± 49	6–331	97 ± 53
CCl_4	2	3–172	76 ± 43	3–165	69 ± 35
CH_2Br_2	2	3–165	71 ± 51	3–178	78 ± 44
C_2HCl_3	2	ND–45	9 ± 12	ND–42	10 ± 9
$CHBr_2Cl$	2	ND–88	19 ± 14	ND–61	18 ± 11
C_2Cl_4	3	4–354	83 ± 79	5–396	84 ± 99

* N = number of samples.

Table S2. Summary of error estimation results of PMF model.

Diagnostic	3 factors	4 factors	5 factors	6 factors
Q(true)	8384.5	7075.8	6002.1	5089.8
Q(robust)	7974.7	6906.4	5873.8	5004.9
Factors with BS mapping < 80%	NO	NO	Yes	Yes
DISP %dQ	< 0.1%	< 0.1%	< 0.1%	< 0.1%
DISP swaps	0	0	0	0

Table S3. Seasonal mean values of meteorological parameters in Wuhan during the sampling period (mean ± SD).

Season	Temperature (℃)	Relative Humidity (%)	Wind Speed (m/s)	Sea Level Pressure (hPa)	Boundary Layer Height (m)
Winter	13.0 ± 5.0	51.3 ± 17.1	3.5 ± 1.4	1020.6 ± 5.8	1023.8 ± 268.6
Spring	21.3 ± 4.7	58.0 ± 20.5	3.7 ± 1.4	1009.2 ± 4.0	1343.5 ± 544.6
Summer	33.0 ± 3.5	66.0 ± 11.8	4.1 ± 1.9	998.5 ± 2.1	1223.0 ± 302.3
Autumn	16.0 ± 4.7	58.8 ± 19.7	2.4 ± 1.6	1020.3 ± 5.2	905.3 ± 176.6

Table S4. Atmospheric lifetime of BTEX with respect to reaction with hydroxyl radical (OH) [75].

Species	Atmospheric lifetime *
Benzene	9.4 days
Toluene	1.9 days
Ethylbenzene	1.6 days
m-xylene	11.8 h
p-xylene	19.4 h
o-xylene	20.3 h

* Concentration used in calculations: [OH] = 10^6 molecule cm^{-3}.

Table S5. Seasonal mean values of methyl chloride (CH_3Cl) at ZY and JX during the sampling period (mean ± 95% C.I.).

Season	ZY (pptv)	JX (pptv)
Winter	814 ± 50	1172 ± 172
Spring	698 ± 49	820 ± 51
Summer	340 ± 197	379 ± 201
Autumn	1360 ± 448	1046 ± 173