S1. Additional descriptions on the parameters involved in equations

Additional details on the parameters involved in the estimation of S/IVOC emissions are listed as below:

$$E_{S/IVOCs,j}=\sum_{j,k} A_{j,k} \times EF_{S/IVOCs,j} \times (1-\mu) \times 10^{-3} \quad (S1)$$

where j and k denote the specific sector and city, respectively; $E_{S/IVOCs}$ denotes the annual emissions of S/IVOCs; A, EF, and μ represent the activity level, average emission factor, and removal efficiency, respectively.

$$E_{S/IVOCs,j}=\sum_{j,k} A_{j,k} \times EF_{PM_{2.5},j} \times F_{OC,j} \times \frac{OM}{OC_j} \times \left(\frac{E_{SVOCs,j}}{E_{POA,j}}+\frac{E_{IVOCs,j}}{E_{POA,j}}\right) \times (1-\mu) \times 10^{-3} \quad (S2)$$

$$E_{S/IVOCs,j}=E_{PM_{2.5},j} \times F_{OC,j} \times \frac{OM}{OC_j} \times \left(\frac{E_{SVOCs,j}}{E_{POA,j}}+\frac{E_{IVOCs,j}}{E_{POA,j}}\right) \quad (S3)$$

where $E_{PM_{2.5}}$ denotes the annual emissions of $PM_{2.5}$; E_{SVOCs}, E_{IVOCs}, and E_{POA} denote the emissions or emission factors of S/IVOCs and POA; F_{OC} denotes the mass fractions of OC to $PM_{2.5}$. Note that SVOC emissions in the present study were assumed to have been included in traditional POA emissions (Zheng et al., 2010; Liu et al., 2015; Dr. Qi Chen, Peking University, Personal Communication), while IVOC emissions have not been reported in current inventories. Thus, SVOC emissions were directly extracted from the traditional POA emissions. Additionally, in order to obtain localized parameters as much as possible, data of ratios of O/C, H/C, N/C, and OM/OC were extracted from field measurements conducted in China. The observed data of HOA (Hydrocarbon-like Organic Aerosols), COA (Cooking-like Organic Aerosols), and BBOA (Biomass burning Organic Aerosols) were used to calculate the four abovementioned ratios for on-road/off-road mobile sources, residential sources, and biomass burning, respectively, while data of field measured OA were used for industrial emissions and dust because measured OA for these two sources could not be identified in measurement data. The ratio of S/IVOCs to POA for all emission sources, except for biomass burning, were based on the relationship between the emissions or emission factors of S/IVOCs and POA measured from vehicle emissions owing to the lack of relevant information.

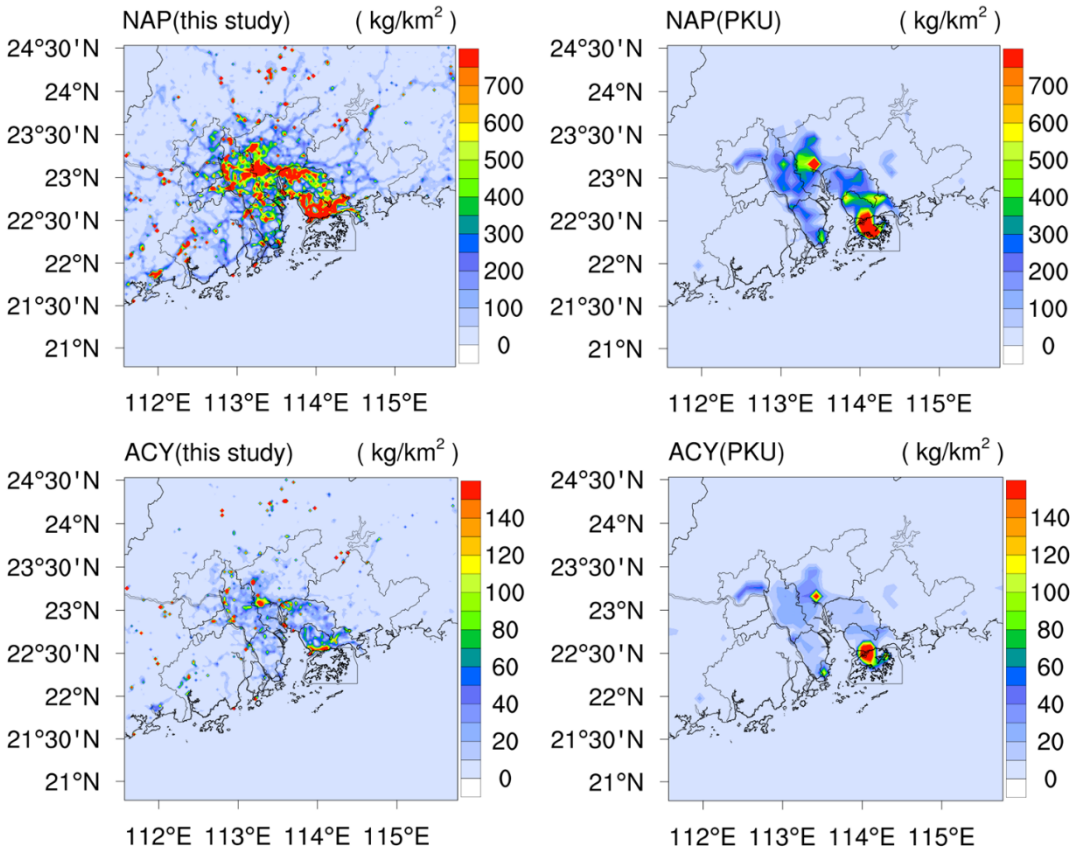
Additional details on the parameters involved in the VBS approach are listed as below:

$$S/IVOC(g)_{2,e,c} + OH \rightarrow SI\text{-}SOA(g)_{1,e,c} + 0.5\ SI\text{-}SOA(g)_{1,e,o} \quad (S4)$$

$$S/IVOC(g)_{2,e,o} + OH \rightarrow SI\text{-}SOA(g)_{1,e,o} + OH \quad (S5)$$

where (g) denotes gas phase; the subscript *1* denotes the low-volatility species (C* = 0.01 μg/m^3 at 298K and 1 atm), *2* denotes the high-volatility species (C* = 10^5 μg/m^3 at 298K and 1 atm); *e* denotes the emission categories, including biomass burning and other anthropogenic emissions; *c* denotes the non-oxygen (C, H, N) component of the species; *o* represents the oxygen component. Note that the addition of oxygen was only considered in Eq. (S4) in order to ensure that the mass growth was not calculated twice by the oxidation of non-oxygen and oxygen components of the same species. Furthermore, the non-oxygen fraction of each species was calculated on the basis of the ratio of non-oxygen component to carbon component, which was derived from elemental ratios of O/C, H/C and N/C (Table 2 in the main text).

S2. Supplementary figures and tables



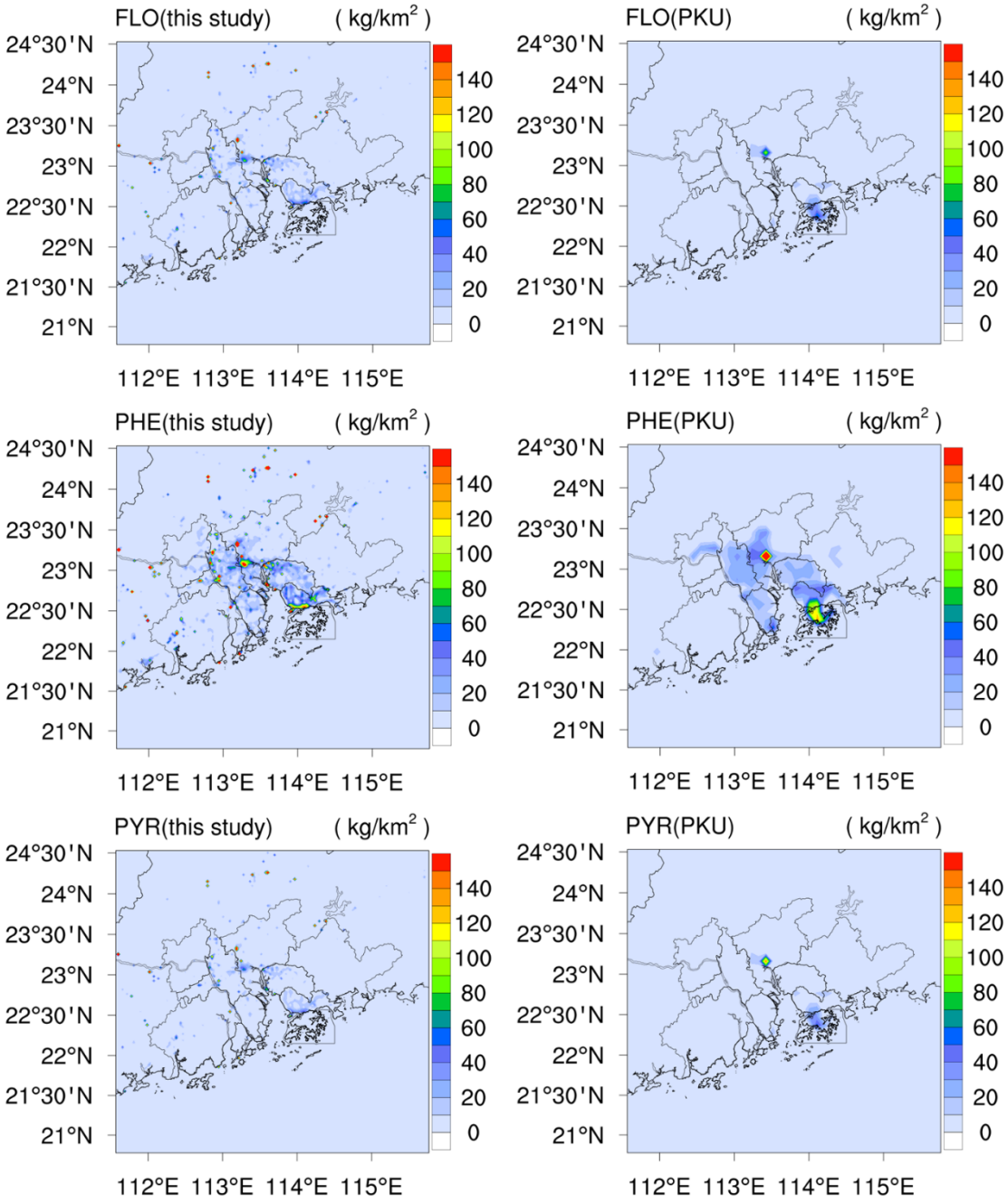


Figure S1: Comparisons of spatial distribution of five individual PAH between emission inventory of this study and PKU (denoted as Shen's inventory).

Table S1: Comparisons of emissions of five individual PAH and total of them in the PRD region between emission inventory of this study and Shen's inventory (unit: t/year).

inventory	Guangzhou	Shenzhen	Zhuhai	Foshan	Jiangmen	Dongguan	Zhongshan	Huizhou	Zhaoqing	PRD region
					naphthalene					
this study	2524.5	1585.7	392.9	1979.0	1214.1	1249.4	469.5	596.0	496.7	10507.8
PKU	840.2	211.5	23.9	297.4	84.2	360.5	142.1	437.8	106.9	2504.5
					acenaphthylene					
this study	134.8	84.7	21.0	105.7	64.8	66.7	25.1	31.8	26.5	561
PKU	126.4	52.7	3.4	44.2	28.1	30.5	18.3	52.2	38	393.8
					fluorene					
this study	38.3	24.0	6.0	30.0	18.4	18.9	7.1	9.0	7.5	159.3
PKU	36.2	8.6	0.8	10.4	4.7	10.2	4.7	14.1	6.1	95.8
					phenanthrene					
this study	95.1	59.7	14.8	74.6	45.7	47.1	17.7	22.5	18.7	395.9
PKU	139.8	35.4	4.0	43.6	18.1	41.3	18.3	57.1	23.8	381.4
					pyrene					
this study	29.5	18.5	4.6	23.1	14.2	14.6	5.5	7.0	5.8	122.6
PKU	41.2	8.5	0.8	10.0	4.4	9.8	4.5	13.4	5.6	98.2
					total of five PAHs					
this study	2822.1	1772.6	439.2	2212.3	1357.2	1396.7	524.9	666.3	555.3	11746.6
PKU	1183.8	316.7	32.9	405.6	139.5	452.3	187.9	574.6	180.4	3473.7

References

Liu, F., Zhang, Q., Tong, D., Zheng, B., Li, M., Huo, H. and He, K. B.: High-resolution inventory of technologies, activities, and emissions of coal-fired power plants in China from 1990 to 2010, Atmos. Chem. Phys., 15(23), 13299–13317, doi:10.5194/acp-15-13299-2015, 2015.

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