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Improved provincial emission inventory and speciation profiles of anthropogenic non-methane volatile organic compounds: a case study for Jiangsu, China

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SUPPLEMENT

Number of tables: 9

Table S1 Estimations of anthropogenic NMVOC emissions for China (Tg).

Table S2 Sampling locations and numbers of field measurements by chemical product.

Table S3 Uncertainties of activity levels by source.

Table S4 Uncertainties of NMVOC emission factors by source. For uniform distributions, the values in parentheses are expressed as g/kg.

Table S5 Source profiles of measured stack emissions (weight percentage, wt %). Numbers in parentheses are standard deviations.

Table S6 Source profiles of measured fugitive emissions (weight percentage, wt %). Numbers in parentheses are standard deviations.

Table S7 Anthropogenic NMVOC emissions for Jiangsu by city in 2005 and 2014 (Gg).

Table S8 NMVOC emissions from refinery and chemical industry in Nanjing 2011, estimated using three methods with various data sources.

Table S9 NMVOC emissions from Nanjing's 15 key chemical and refinery enterprises for 2014, estimated using two methods

Number of figures: 6

Figure S1 Locations of Jiangsu province and the major point sources of NMVOC emissions for 2014.

Figure S2. The flow diagrams for sampling (a) and analysis (b) of NMOVC species from chemical industry.

Figure S3 NMVOC emissions from industrial processes (a), solvent use (b) and on-road transportation (c) in Jiangsu 2005-2014.

Figure S4 Emissions and OFPs of the 25 species with the largest OFPs in Jiangsu for 2005 (a) and 2014 (b).

Figure S5 Spatial distributions of NMVOC emissions from refinery and chemical industry in Nanjing for 2014, with the emissions of the 15 key enterprises calculated using device operation based method (a) and emission factor based method (b).

Figure S6 WRF-CMAQ modeling domain and locations of 43 meteorological and 9 Nanjing air quality monitoring sites.

Table S1 Estimations of anthropogenic NMVOC emissions for China (Tg).

Year	Study	Stationary combustion	Industrial process	Solvent use	Transportation	Biomass open burning	Oil distribution	Others	Total
1990	Klimont et al., 2002	5.8	0.1	1.2	2.3		0.5	1.2	11.1
1994	Tonooka et al., 2001	11.0		0.9	1.7		0.3		13.9
1995	Klimont et al., 2002	5.5	0.1	1.8	3.6		0.7	1.4	13.1
2000	Streets et al., 2003	5.2	0.2	2.7	5.1		0.9	1.5	15.6
	Bo et al., 2008	4.6	1.3	1.4	3.1	0.8	0.4		11.6
	REAS	2.5	1.2	3.6	8.0			0.5	15.8
2005	Bo et al., 2008	4.9	2.8	1.9	5.5	0.9	0.5		16.5
	Wei et al., 2008	5.5	3.1	3.4	5.6	1.2	0.5	0.1	19.4
	REAS	2.6	1.8	7.2	10.9			0.8	23.3
2006	Zhang et al., 2009								23.2
	REAS	2.7	2.1	8.8	10.8			0.9	25.3
2008	REAS	4.1	2.4	10.2	9.4			1.0	27.1
	MEIC								22.1
2010	MEIC								23.6

Table S2 Sampling locations and numbers of field measurements by chemical product.

Chemical products	Sampling location	No. of samples
Synthetic rubber		
SBR	Stack gas of drying process	3
SIS rubber	Stack gas of drying process	3
SEBS rubber	Stack gas of drying process	3
Cellulose acetate fiber	Stack gas of acetate flake	6
	Stack gas of spinning	11
Polyether		
Polypropylene glycol (PPG)	Stack gas	4
Polyether polyol (POP)	Stack gas	3
Vinyl acetate	Stack gas	3
Ethylene	Stack gas of cracking furnace	6
	Downwind of process device (fugitive)	3
Propylene epoxide	Downwind of process device (fugitive)	2
Glycol	Downwind of process device (fugitive)	3
Polyethylene	Downwind of process device (fugitive)	3
Butanol and octanol	Downwind of process device (fugitive)	3
Total		56

Table S3 Uncertainties of activity levels by source.

Sector	Probability distribution	CV (%)
Fossil fuel stationary combustion		
Power plant	Normal	5
Industrial boiler	Normal	10
Residential	Normal	20
Industrial process		
Point sources	Normal	10
Area sources	Normal	20
Solvent use		
Point sources	Normal	20
Area sources	Lognormal	80
Transportation		
On-road	Normal	30
Off-road	Lognormal	50
Oil distribution	Normal	30
Biomass burning	Normal	30
Others	Normal	30

Table S4 Uncertainties of NMVOC emission factors by source. For uniform distributions, the values in parentheses are expressed as g/kg.

Sector	Subcategory	Distribution
Fossil fuel stationary combustion	All categories	Lognormal (CV: 150%)
Industrial process	Glass fiber/glass work	Lognormal (CV: 500%)
	Food	Lognormal (CV: 500%)
	Other rubber product	Lognormal (CV: 500%)
	Plastic product	Lognormal (CV: 500%)
	Textile	Lognormal (CV: 500%)
	Carbon black	Lognormal (CV: 500%)
	Other industrial process	Lognormal (CV: 300%)
Solvent use	Printing	Uniform (400-1000)
	Exterior wall painting (solvent based)	Uniform (100-1000)
	Interior wall painting	Uniform (100-600)
	New vehicle varnish paint	Uniform (200-800)
	Vehicle refurnish painting	Uniform (300-900)
	Wood-furniture paint (solvent based)	Uniform (300-900)
	Anticorrosive paint	Uniform (150-800)
	Adhesive (shoe making)	Uniform (400-900)
	Synthetic leather coating	Uniform (100-450)
	Agriculture pesticide	Uniform (150-800)
	Other solvent use	Lognormal (CV: 300%)
Transportation	On-road	Lognormal (CV: 150%)
	Off-road	Lognormal (CV: 300%)
Oil distribution	All categories	Lognormal (CV: 150%)
Biomass burning	All categories	Lognormal (CV: 200%)
Others	Garbage disposal	Lognormal (CV: 300%)
	Cooking fume	Lognormal (CV: 500%)

Table S5 Source profiles of measured stack emissions (weight percentage, wt %). Numbers in parentheses are standard deviations.

Species	Synthetic rubber production			Cellulose acetate fiber production		Polyether production		Vinyl acetate	Ethylene
	Butadiene styrene rubber	SIS rubber	SEBS rubber	Acetate flake	Spinning	POP	PPG	production	production
n-Hexane		0.49 (0.84)	0.49 (0.84)		0.42 (1.33)			0.01 (0.01)	0.07 (0.16)
Cyclohexane	69.92 (47.31)	85.33 (21.93)	72.21 (13.81)	27.46 (46.9)	0.12 (0.41)			0.06 (0.1)	
Propane								19.77 (20.99)	
2-Methylpropane						0.51 (0.88)			
n-Butane						0.54 (0.93)	0.1 (0.2)		
2-Methylpentane								0.06 (0.1)	
3-Methylpentane								0.03 (0.05)	
3-Methylhexane									0.88 (2.15)
2,2,4-Trimethylpentane									0.66 (1.61)
Propylene							0.26 (0.49)		
cis-2-Pentene									1.06 (2.6)
Methyl bromide					4.74 (12.27)				
1,1-Dichlorethene					0.03 (0.1)				
Methylene chloride		1.03 (1.78)	1.77 (1.59)	1.27 (2.01)	3.25 (3.82)				
1,1-dichloroethane					0.14 (0.46)				
Chloroform			0.88 (1.52)	0.03 (0.07)	0.75 (2.28)	0.63 (1.08)	2.69 (5.33)		1.53 (2.69)
1,2-Dichloroethane				0.4 (1.06)	0.52 (1.61)		6.07 (12.03)		1.97 (2.26)
1,2-Dichloropropane					0.01 (0.02)				
Trans 1,3-dichloropropylene					0.05 (0.15)				0.26 (0.64)
Dibromochloromethane					0.61 (2)				

Table S9 (continued)

Species	Synthetic rubber production			Cellulose acetate fiber production		Polyether production		Vinyl acetate production	Ethylene production
	Butadiene styrene rubber	SIS rubber	SEBS rubber	Acetate flake	Spinning	POP	PPG		
Tetrachloroethene					0.86 (2.8)				
Chlorobenzene	0.14 (0.24)			1.25 (3.05)	0.21 (0.68)				
Benzylchloride					0.04 (0.11)	2.35 (4.08)	0.87 (1.74)		0.74 (1.81)
1,2-Dichlorobenzene					0.01 (0.05)	1.68 (2.9)	0.82 (1.63)		0.61 (1.5)
Chloromethane							0.35 (0.64)		0.97 (2.36)
Bromodichloromethane									0.24 (0.59)
1,3-Dichlorobenzene						1.43 (2.48)	0.42 (0.84)		0.45 (1.11)
1,4-Dichlorobenzene						1.4 (2.43)	0.49 (0.97)		0.6 (1.08)
1,2,4-Trichlorobenzene						9.37 (16.22)	2.72 (5.44)	0.03 (0.04)	13.56 (16.2)
Benzene	0.64 (0.73)	0.52 (0.28)	0.44 (0.4)	6.36 (7.5)	0.58 (1.28)	7.84 (12.86)	1.16 (2.28)		21.53 (20.44)
Toluene	0.04 (0.06)			9.01 (21.61)	1.42 (4.37)	0.32 (0.56)			10.94 (16.99)
Ethylbenzene	0.38 (0.61)	0.02 (0.03)		4.37 (7.86)	1.34 (2.54)	2.38 (2.68)	2.63 (5.27)		1.77 (2.74)
m -Xylene	0.38 (0.61)	0.02 (0.03)		3.22 (7.85)	1.33 (2.51)				
p-Xylene	0.4 (0.65)	0.02 (0.03)		4.34 (8.25)	0.59 (1.1)				
Styrene	24.14 (41.59)	0.42 (0.55)	1.48 (1.75)	0.03 (0.09)	0.05 (0.11)		1.28 (2.39)	0 (0.01)	
o-Xylene	0.01 (0.02)			0.27 (0.71)	0.14 (0.32)				
1,3,5-Trimethylbenzene				0.06 (0.16)	0.07 (0.13)				
4-Ethyltoluene				0.06 (0.16)	0.07 (0.13)				
1,2,4-Trimethylbenzene					0.04 (0.11)				
Xylene						4.07 (4.25)			1.47 (2.35)
Isopropylbenzene						1.55 (2.69)			
n-Propylbenzene						3.23 (5.59)	2.23 (4.45)		

Table S9 (continued)

Species	Synthetic rubber production			Cellulose acetate fiber production		Polyether production		Vinyl acetate production	Ethylene production
	Butadiene styrene rubber	SIS rubber	SEBS rubber	Acetate flake	Spinning	POP	PPG		
p-Ethyltoluene									0.17 (0.41)
m-Diethylbenzene									1.51 (3.69)
Naphthalene						0.72 (1.09)	0.01 (0.02)		0.01 (0.02)
Acetone	3.19 (4.94)	10.85 (16.82)	19.14 (10.96)	33.89 (41.42)	70.41 (27.28)	0.56 (0.75)	5.37 (8.84)		16 (21.25)
2-Propanol	0.74 (1.22)	0.53 (0.62)	1.35 (0.93)	4.29 (6.55)	5.39 (4.84)	9.74 (10.43)			
Vinyl acetate					0.23 (0.49)			0 (0.01)	
2-Butanone		0.28 (0.49)	0.28 (0.49)	2.13 (3.79)	0.2 (0.48)				
Ethyl acetate					0.06 (0.2)	0.41 (0.62)	0.02 (0.03)		0.76 (1.61)
Tetrahydrofuran	0.02 (0.03)			0.02 (0.05)	0.2 (0.53)				
Methyl-2-pentanone				0.03 (0.07)	0.38 (0.83)				
Acrolein						0.13 (0.12)			
Ethylene oxide							63.01 (42.11)		
Propylene oxide							9.45 (6.32)		
Vinylacetate								80.04 (20.94)	
Carbon disulfide		0.51 (0.7)	1.96 (2.35)	1.51 (2.05)	5.74 (9.98)	8.55 (14.74)	0.04 (0.06)		22.26 (17.34)
Acrylontitrile						42.61 (39.32)			

Table S6 Source profiles of measured fugitive emissions (weight percentage, wt %). Numbers in parentheses are standard deviations.

Species	Butanol and octanol production	Propylene oxide production	Polyethylene production	Ethylene production	Glycol production
n-Hexane				20.42 (7.56)	16.3 (14.29)
Propane			1.54 (2.66)		
2-Methylpropane			6.79 (11.76)		
n-Butane			10.95 (18.97)		
iso-Butane				15.11 (16.25）	
Ethylene			42.00 (38.70）	33.03 (22.78）	
Propylene				26.36 (10.12)	
Chloroform	0.63 (1.08)		0.06 (0.11)		
1,2-Dichloroethane	0.76 (1.31)				
1,2-Dichloropropane		64.86			
Benzylchloride	2.35 (4.08)				
1,2-Dichlorobenzene	1.68 (2.9)				
Chloromethane	0.65 (1.13)	4.31			
1,3-Dichlorobenzene	1.48 (2.44)		0.32 (0.55)		
1,4-Dichlorobenzene	1.4 (2.43)				0.17 (0.29)
1,2,4-Trichlorobenzene	19.76 (17.18)	11.74	9.05 (8.07)		1.5 (2.61)
Benzene	12.91 (11.66)	8.37	17.26 (19.52)	5.08 (8.81)	6.97 (8.66)
Toluene					0.14 (0.25)
Ethylbenzene	1.99 (3.45)	2.41	0.28 (0.49)		28.05 (48.59)
Styrene					0.71 (1.23)
Xylene			1.91 (3.3)		35.47 (33.04)
Isopropylbenzene	5.98 (10.36)				
n-Propylbenzene	5.23 (4.67)				
p-Diethylbenzene					9.63 (11.4)
Naphthalene	0.02 (0.03)	0.2	0.01 (0.01)		
Acetone	25.59 (29.95)		4.51 (7.65)		
Ethylacetate	0.38 (0.65)				
Acrolein		1.97			
Carbon disulfide	19.19 (13.03)	6.14	5.32 (6.06)		1.06 (1.83)

Table S7 Anthropogenic NMVOC emissions for Jiangsu by city in 2005 and 2014(Gg).

City	Fossil fuel stationary combustion	Industrial process	Transportation	Solvent use	Oil distribution	Biomass burning	Others	Total
2005								
Nanjing	3.9	53.4	28.7	32.8	8.1	14.1	6.6	147.7
Wuxi	3.2	37.0	22.4	46.7	5.9	9.1	5.4	129.7
Xuzhou	5.1	48.5	31.2	24.9	2.3	56.4	8.5	176.9
Changzhou	2.5	40.8	19.9	22.1	3.6	12.8	4.3	105.9
Suzhou	4.3	59.5	37.3	70.0	5.9	12.8	7.3	197.1
Nantong	4.2	34.8	34.7	53.3	1.4	46.8	7.1	182.4
Lianyungang	2.6	36.5	20.6	11.6	0.8	41.0	4.4	117.5
Huai’an	2.9	23.2	22.9	15.6	1.3	50.6	4.8	121.2
Yancheng	4.5	36.2	34.0	22.2	1.0	83.3	7.5	188.7
Yangzhou	2.6	31.7	20.7	28.7	1.5	34.8	4.4	124.4
Zhenjiang	1.7	20.2	15.2	11.0	1.4	14.9	2.9	67.3
Taizhou	2.7	16.5	18.7	28.4	0.8	37.7	4.5	109.3
Suqian	2.7	22.5	19.1	12.9	0.5	43.5	4.5	105.7
2014								
Nanjing	6.5	126.5	24.7	78.3	5.8	3.4	8.2	253.4
Wuxi	7.4	117.7	19.3	129.9	7	2.2	6.5	290.0
Xuzhou	4.4	71.2	26.9	54.8	3.5	13.6	8.6	183.0
Changzhou	4.4	70.3	17.1	57	4.1	3.1	4.7	160.7
Suzhou	12.5	197.4	32.1	190	11.9	3.1	10.6	457.6
Nantong	5.0	81.1	29.9	143.4	4.3	11.3	7.3	282.3
Lianyungang	1.8	28.2	17.7	24.9	1.3	9.9	4.4	88.2

Table S11 (continued)

City	Fossil fuel stationary combustion	Industrial process	Transportation	Solvent use	Oil distribution	Biomass burning	Others	Total
Huai’an	2.2	35.2	19.7	35.9	1.7	12.2	4.8	111.7
Yancheng	3.6	55.0	29.3	51.7	2.9	20.1	7.2	169.8
Yangzhou	3.4	53.0	17.8	74	3.1	8.4	4.5	164.2
Zhenjiang	3.1	46.6	13.1	25.1	2.8	3.6	3.2	97.5
Taizhou	3.2	48.3	16.1	73	2.7	9.1	4.6	157.0
Suqian	1.9	27.7	16.4	28.5	1.5	10.5	4.8	91.3

Table S8 NMVOC emissions from refinery and chemical industry in Nanjing 2011, estimated using three methods with various data sources.

Sector	NMVOC emissions (metric tons)		
	Method 1 *	Method 2 *	Method 3 *
Refinery	39690	39691	39664
Chemical material industry	14879	7944	2152
Benzene	238		167
Phthalic anhydride	969	1868	
Styrene	254		
Propylene	229		
Acrylic acid	72		
Butanol and octanol	4658	4658	
Ammonia	1062	1062	1062
Propylene epoxide	2452	141	
Ethylene epoxide	1278	51	
Formaldehyde	83	95	
Alkyl benzene	690		
Glycol	1192		
Ethylene	1702	70	923
Synthetic chemical industry	13436	9388	5397
Polypropylene fiber	50		50
Dacron	77		77
Acrylic fiber	6		6
Nylon	20		20
Viscose fiber	875	1066	111
Polystyrene	580		261
Polypropylene	1342		1342
Polyether	5265	2480	
Polyethylene	4263	4263	1570
Synthetic rubber	1959	1579	1959
Fine chemical industry	12159	11790	11772
Printing ink		18	
Adhesive	54		
Dye	7427	7427	7427
Paint	4678	4345	4345
Total	80164	68814	58985

* Methods 1: information from Environmental Statistics, Pollution Source Census, and on-site surveys was applied; Method 2: information from Environmental Statistics was applied; and Method 3: information from economic statistics without any on individual plants was applied.

Table S9 NMVOC emissions from Nanjing's 15 key chemical and refinery enterprises for 2014, estimated using two methods

Enterprise No.	NMVOC emissions (metric tons)		Main materials and products
	Emission factor based [1]	Device operation based [2]	
1	34372	40554	Crude oil
2	15190	13638	Crude oil
3	911	5225	Ammonia, chlorobenzene, cyclohexanone
4	9353	4830	Ethylene, glycol, butanol
5	5684	4279	Methanol, butanol and octanol
6	42	3355	Alkyl benzene
7	0	679	Tert-butylanmine
8	2357	594	Polyether
9	1322	432	Vinyl acetate
10	0	265	Caprolactam
11	0	261	Polyvinyl acetate
12	0	222	Hydroxyethyl celluslose
13	1919	205	Acetic acid
14	3704	115	Propylene epoxide, polyether
15	0	114	Surfactant
Total	74854	74771	

1 As described in Section 2 in the main text.

2 The NMVOC emissions in chemical and refinery enterprises include leaks of hydrocarbon vapors from individual device (valves, flanges, seals, etc.), storage, loading and unloading of organic liquid, waste water treatment, and cooling tower. The emissions of each process are separately calculated and then added up for the total emissions of a plant, as following equation:

$$E = \sum E(m) \quad \text{(S1)}$$

Where E is VOCs emissions in chemical and refinery enterprises, E(m) is the emission from the emission source m.

Emissions of leaks from devices were calculated as Eq. (S2):

$$E(L) = \sum_r [e(r) \times t(r)] \quad \text{(S2)}$$

where *E(L)* is emissions of leaks from devices; *e(r)* is NMVOC emission rate of equipment seal point *r* and is determined by the type of seal point according to USEPA (2002); *t(r)* is operation time of seal point *r* in a year.

Emissions from storage of organic liquid were estimated by the software Tanks 4.0.9d (available at https://www3.epa.gov/ttn/chief/software/tanks/), with detailed information on the type and size of tank, the characteristics of organic liquids, and the annual turnover incorporated.

For emissions from loading & unloading of organic liquids, waste water treatment and cooling tower, emission factors from MOFC (2015) were employed to calculate the emissions.

Figure S1 Locations of Jiangsu province and the major point sources of NMVOC emissions for 2014.

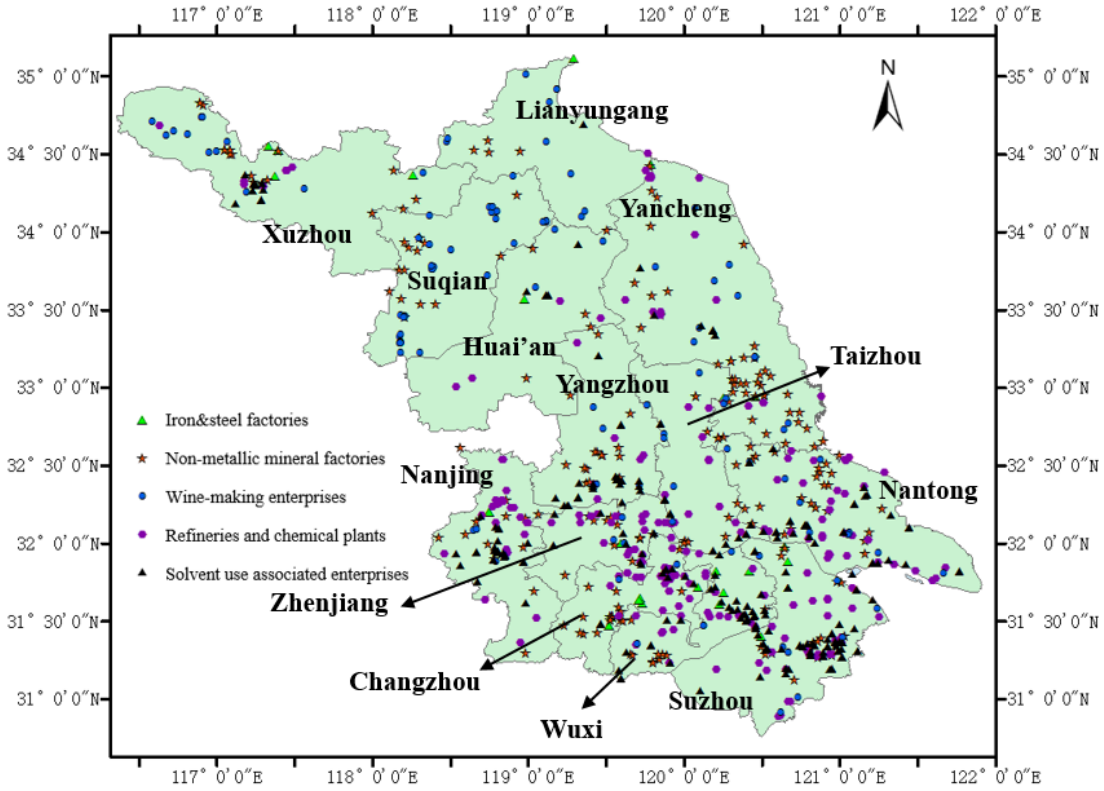
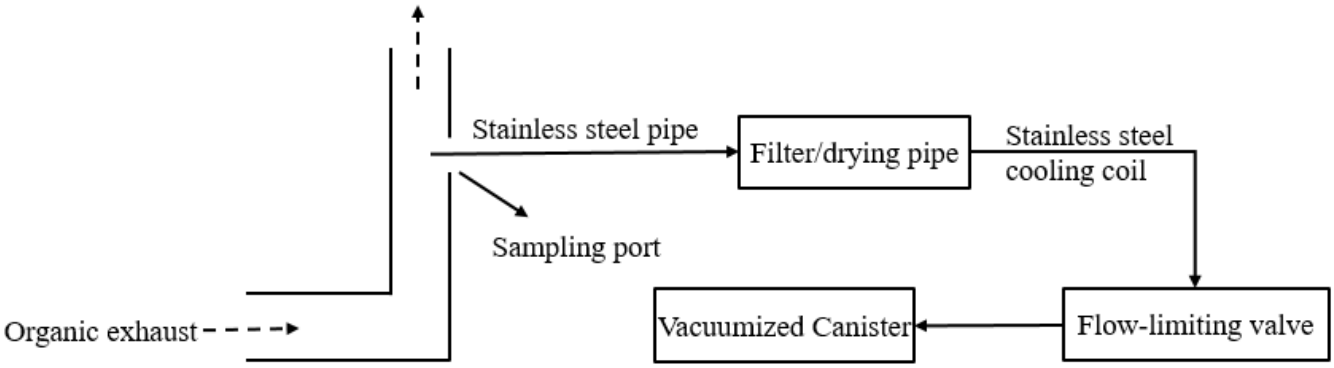
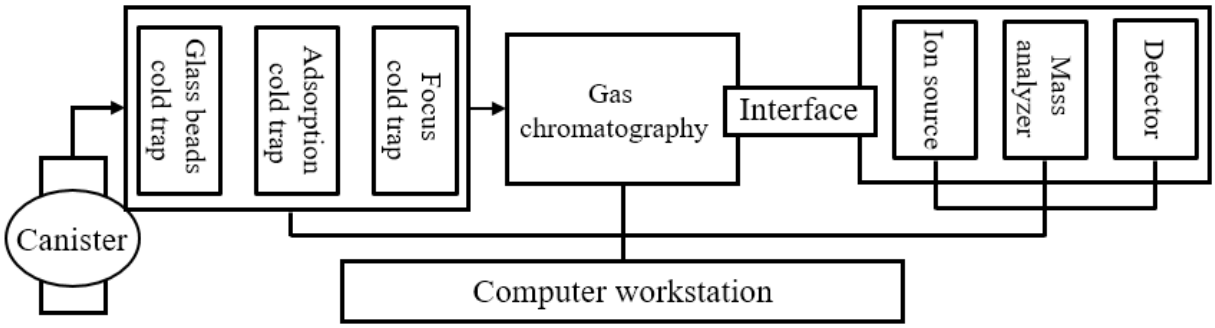


Figure S2 The flow diagrams for sampling (a) and analysis (b) of NMOVC species from chemical industry.

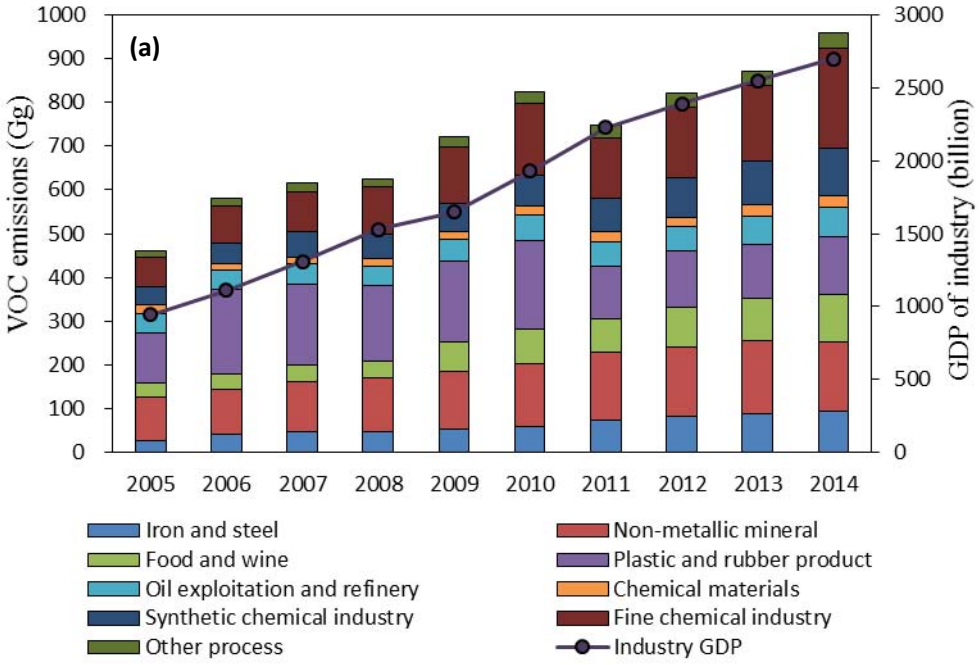


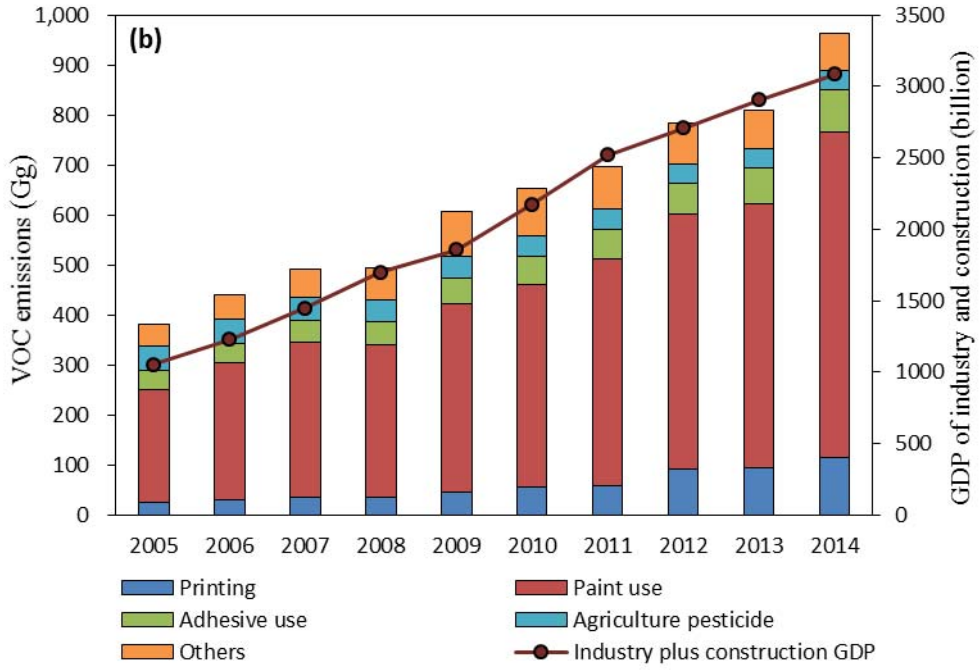
(a)



(b)

Figure S3 NMVOC emissions from industrial processes (a), solvent use (b) and on-road transportation (c) in Jiangsu 2005-2014.





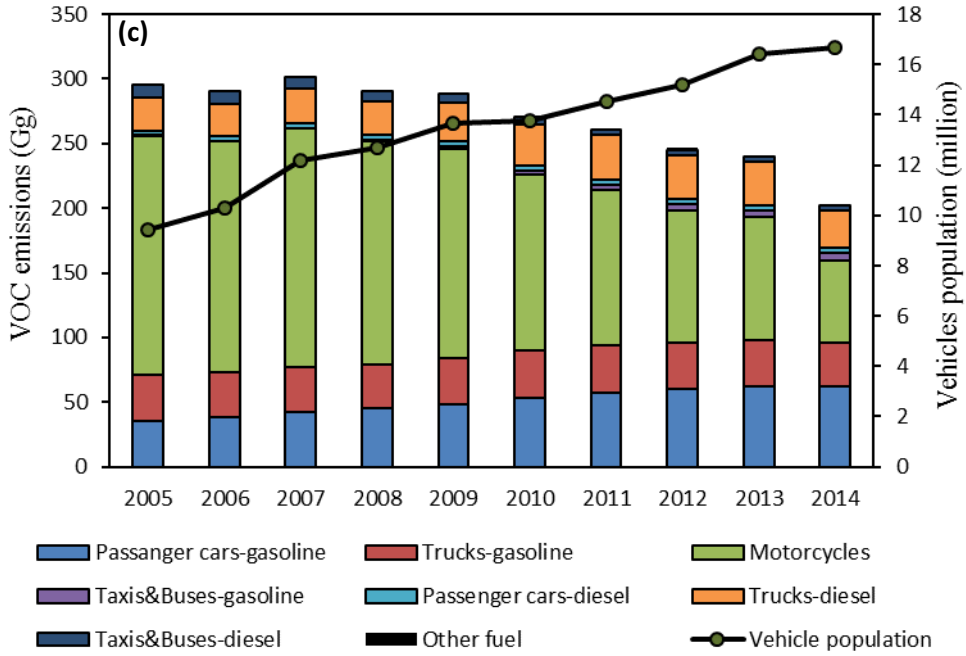
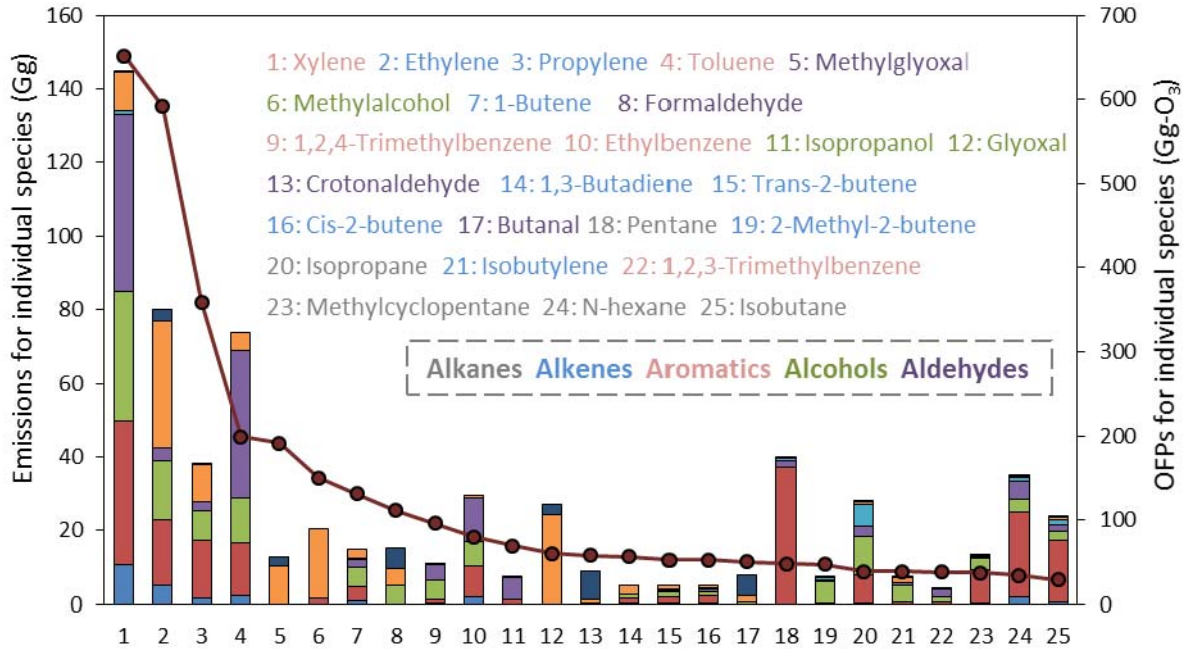

(c)
VOC emissions (Gg)
Vehicles population (million)
350
300
250
200
150
100
50
0
18
16
14
12
10
8
6
4
2
0
2005
2006
2007
2008
2009
2010
2011
2012
2013
2014
Passanger cars-gasoline
Trucks-gasoline
Motorcycles
Taxis&Buses-gasoline
Passenger cars-diesel
Trucks-diesel
Taxis&Buses-diesel
Other fuel
Vehicle population

Figure S4 Emissions and OFPs of the 25 species with the largest OFPs in Jiangsu for 2005 (a) and 2014 (b).

(a)



(b)

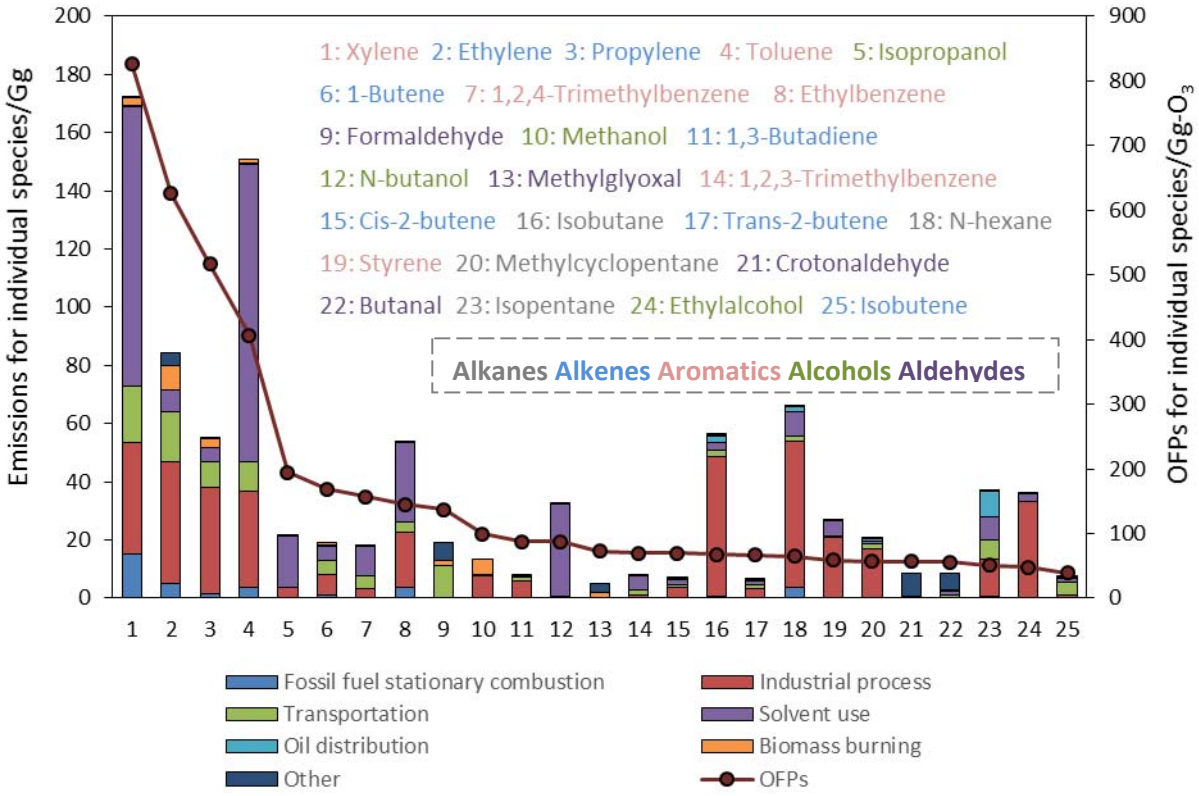
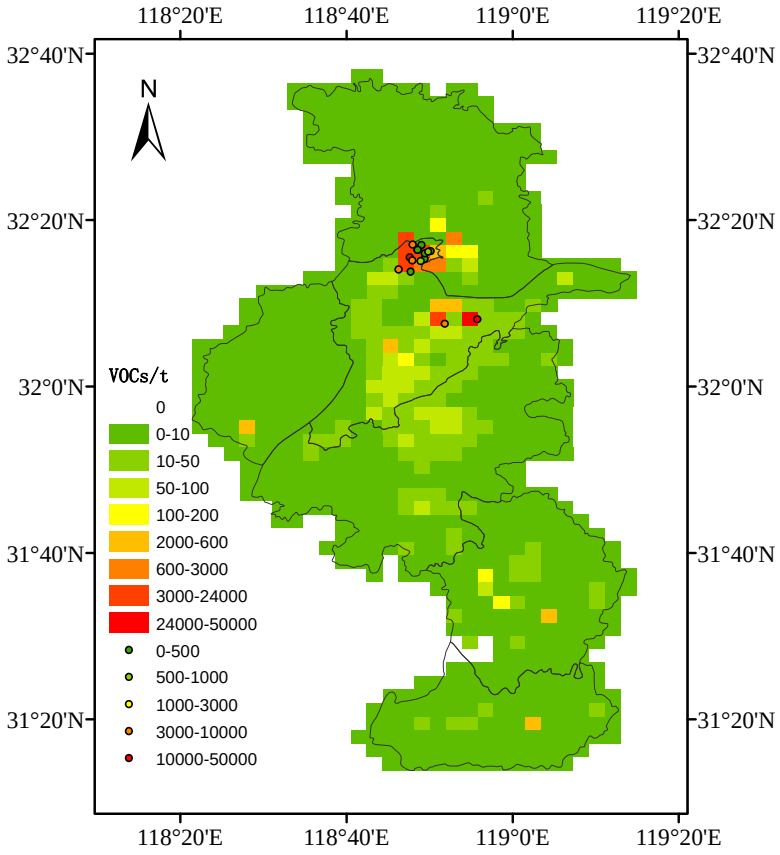
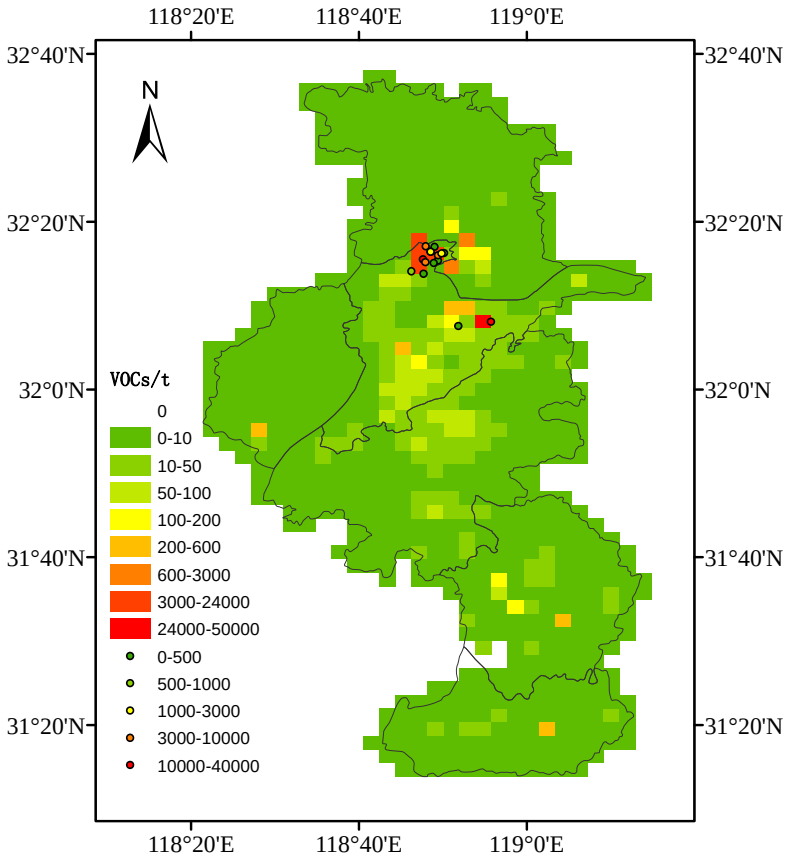


Figure S5 Spatial distributions of NMVOC emissions from refinery and chemical industry in Nanjing for 2014, with the emissions of the 15 key enterprises calculated using device operation based method (a) and emission factor based method (b).

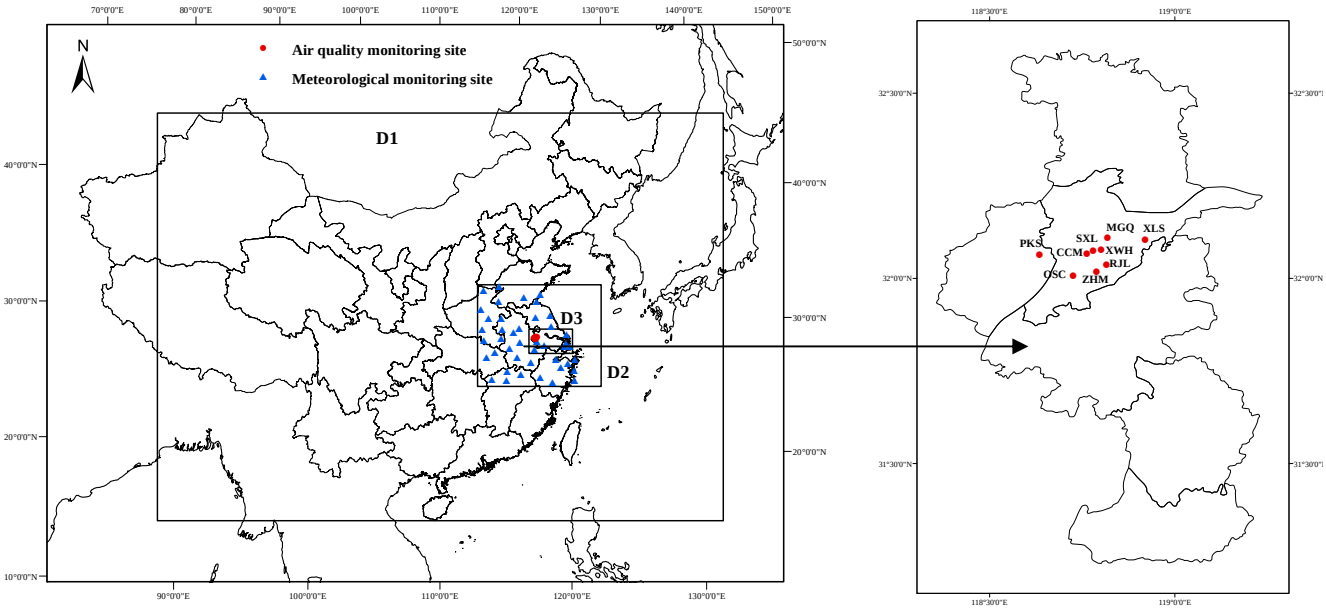


(a)



(b)

Figure S6 WRF-CMAQ modeling domain and locations of 43 meteorological and 9 Nanjing air quality monitoring sites.



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