Supplementary Information
Verification of anthropogenic VOC emission inventory through ambient measurements and satellite retrievals

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Table S1. Source classification and EFs of anthropogenic VOC sources.

Level1	Level 2	Level 3	Level 4	EF	Unit	Reference
Biomass burning	Biofuel	Crop residue		8.27	g kg^{-1}	Zhang et al., 2013
		Firewood		3.13	g kg^{-1}	Zhang et al., 2013
	Crop residue burning in fields	Crop residue	Maize	6.05	g kg^{-1}	Zhang et al., 2013
			Wheat	7.48	g kg^{-1}	Li et al., 2007
			Rice	10.4	g kg^{-1}	Li et al., 2007
			Others	8.45	g kg^{-1}	Li et al., 2016
Stationary fossil fuel combustion	Industrial and commercial consumption	Coal	Manufacturing	0.39	g kg^{-1} fuel	PKU,2001
			Catering	0.5	g kg^{-1} fuel	PKU,2001
		Fuel oil		0.04	g kg^{-1} fuel	AP42
		Coal gas		0.00044	g m^{-3} fuel	Bo et al., 2008
		Liquefied petroleum gas		108	g m^{-3} fuel	AP42
		Natural gas		0.088	g m^{-3} fuel	AP42
	Thermal power generation	Coal		0.15	g kg^{-1} fuel	PKU,2001
		Liquefied petroleum gas		108	g m^{-3} fuel	AP42
		Natural gas		0.088	g m^{-3} fuel	AP42
		Coal		0.39	g kg-1 fuel	PKU,2001
		Liquefied petroleum gas		108	g m^{-3} fuel	AP42
		Natural gas		0.088	g m^{-3} fuel	AP42
	Resident consumption	Coal	Urban/Rural	0.67	g kg^{-1} fuel	Zhang et al., 2000
		Coal gas	Urban/Rural	0.00044	g m^{-3} fuel	Bo et al., 2008
		Liquefied petroleum gas	Urban/Rural	96	g m^{-3} fuel	AP42
		Natural gas	Urban/Rural	0.13	g m^{-3} fuel	Tsai et al.,2003
Transportation	On-road vehicles			COPERT 4		
	Off-road transportation	Aircrafts		1.95	kg LTO^{-1}	Bo et al., 2008
		Trains	Diesel	6.14	g kg^{-1}	Bo et al., 2008
		Marine vessels	Diesel	8.0	g kg^{-1}	EEA,2013
		Agricultural machinery	Gasoline/Diesel	91.5/18.3	g kg^{-1}	EEA,2013
		Construction machinery	Diesel	18.3	g kg^{-1}	EEA,2013
Solvent utilization	Printing	Offset printing ink	Thermosetting/ Cold setting wheel	100/10	g kg^{-1}	DB11/1201-2015
		Flexo ink	Solvent-based ink/ Water-based ink	350/100	g kg^{-1}	DB11/1201-2015
		Gravure /Letterpress ink		300	g kg^{-1}	DB11/1201-2015
	Asphalt pavement	Asphalt		26	g kg^{-1}	AP42
	Surface	Building	Interior wall	100/30	g l^{-1}	GB18582-2008

coating	painting	coating (solvent-based/water-based)			
		Exterior wall coating (solvent-based/water-based)	430/40	g l^{-1}	GB18582-2008
	Furniture manufacturing		0.4	g $piece^{-1}$	AP42
	Vehicle coating and maintenance	Car	4	kg $vehicle^{-1}$	Local Standard
		Truck	10	kg $vehicle^{-1}$	Local Standard
		Buses	60	kg $vehicle^{-1}$	Local Standard
		Motorcycles	1	kg $vehicle^{-1}$	AP42
		Bicycles	0.3	kg $bike^{-1}$	AP42
	Vehicle repair		0.46	g $yr^{-1}capita^{-1}$	EEA,2013
	Machinery coating	Machine tool equipment/ Agricultural machine/Commercial machine	0.4	kg $piece^{-1}$	AP42
	Cans manufacturing		1	g $piece^{-1}$	This study
	electronics manufacturing coating	Enameled wire	11	g kg^{-1}	This study
		Electric cable	15.46	g kg^{-1}	AP42
		Circuit board	0.1	kg m^3	Qiu et al., 2014
		Mobile phone	0.01	g $piece^{-1}$	Taiwan, 2009
	Domestic appliance coating		0.4	kg $piece^{-1}$	AP42
	Office Supply coating	Typewriter/ Printer/Others	0.2	kg $piece^{-1}$	AP42
Pesticide utilization	Insecticides	Dichlorvos/Omethoate/Cypermethrin	567	g kg^{-1}	AP42
	Herbicides	Paraquat/ Glyphosate	316	g kg^{-1}	AP42
	Fungicides	Carbendazim/ Kitazine	475	g kg^{-1}	AP42
	Other pesticides		470	g kg^{-1}	AP42
Residents living consumption	Dry cleaning agent	Trichloroethylene/ettrachloroethylene	1000	g kg^{-1}	AP42
	Cooking		0.0035	kg $yr^{-1}capita^{-1}$	AP42
	Commercial/Consumer Solvent Use	Aerosol products	0.8	kg $yr^{-1}capita^{-1}$	AP42
		Household products	0.43	kg $yr^{-1}capita^{-1}$	AP42

			Toiletries	0.16	kg $yr^{-1}capita^{-1}$	AP42
			Nonindustrial adhesives	0.07	kg $yr^{-1}capita^{-1}$	AP42
			Space deodorant	0.05	kg $yr^{-1}capita^{-1}$	AP42
			Moth control	0.04	kg $yr^{-1}capita^{-1}$	AP42
			Laundry detergent	0.01	kg $yr^{-1}capita^{-1}$	AP42
Industrial processes	Petroleum & related industry	Crude oil and natural gas extraction	Crude oil	0.2	g kg^{-1}	AP42
			natural gas	0.1	g m^{-3}	EEA,2013
		Raw chemicals manufacturing	Ethylene	0.6	g kg^{-1}	EEA,2013
			Propylene	0.45	g kg^{-1}	EEA,2013
			Benzene	0.55	g kg-1	Taiwan,2009
			Ethylbenzene	0.1	g kg^{-1}	EEA,2013
			Butadiene	5.03	g kg^{-1}	AP42
			Styrene	1	g kg^{-1}	EEA,2013
		Fertilizer manufacturing	Synthesis ammonia	4.72	g kg^{-1}	AP42
			Urea	0.01	g kg^{-1}	AP42
		Pharmaceuticals production		300	g kg^{-1}	EEA,2013
		Paint & related manufacturing	Paint	60	g kg^{-1}	AP42
			Ink	15	g kg^{-1}	AP42
			Carbon black	52	g kg^{-1}	AP42
			Dyeing cloth	81.4	g kg^{-1}	AP42
		Synthetic resin materials manufacturing	PVC	0.77	g kg^{-1}	AP42
			PP	4	g kg^{-1}	EEA,2013
			PS	3.34	g kg^{-1}	AP42
			PE	3.75	g kg^{-1}	AP42
		Synthetic rubber manufacturing	Synthetic rubber	7.17	g kg^{-1}	AP42
		Synthetic fiber manufacturing	Cellulosic fiber	112	g kg^{-1}	AP42
			Viscose	14.5	g kg^{-1}	AP42
			Acrylonitrile	1	g kg^{-1}	EEA,2013
			Nylon	3.3	g kg^{-1}	AP42
			Dacron	0.7	g kg^{-1}	AP42
			Acrylon	37	g kg^{-1}	AP42
			Vinylon	7.7	g kg^{-1}	AP42
			Polypropylene fiber	37	g kg^{-1}	AP42
		Petroleum refining		1.78	g kg^{-1}	AP42
		Petroleum products transport	Crude oil/ Gasoline/ Others	0.54/1.5/ 1	g kg^{-1}	GB11085-89
		Petroleum products storage	Crude oil/ Gasoline/Diesel /Others	0.045/0. 295/0.17 5/0.045	g kg^{-1}	Qiu et al., 2014
		Service station	Gasoline /Diesel	1.78/0.5 4	g kg^{-1}	AP42

	Rubber products manufacturing	Tire	0.3	kg piece^{-1}	AP42
	Polyfoam manufacturing	Polyfoam	770	g kg^{-1}	AP42
	Artificial leather manufacturing	PVC	2.27	g m^{-3}	AP42
Other industrial processes	Cement manufacturing		0.014	g kg^{-1}	AP42
	Tile clay manufacturing		0.012	g kg^{-1}	AP42
	Glass products manufacturing	Plate glass	0.1	g kg^{-1}	AP42
	Ceramics manufacturing		29	g kg^{-1}	AP42
	Non-metal mineral product	Asphalt	0.018	g kg^{-1}	AP42
		Charcoal	135	g kg^{-1}	AP42
	Iron And Steel Production	Sinter production	0.138	g kg^{-1}	EEA,2013
		Pellet plant	0.014	g kg^{-1}	EEA,2013
		Blast furnace	0	g kg^{-1}	EEA,2013
		Converter / electric steelmaking	0/0.046	g kg^{-1}	EEA,2013
		Cold/hot rolled steel	0/0.007	g kg^{-1}	EEA,2013
	Coke production	Heap coking	2.96	g kg^{-1}	AP42
	Sugar production		10	g kg^{-1}	EEA,2013
	Vegetable oil processing	Extraction process	9	g kg^{-1}	AP42
	Wine production	White spirit/red/wine/ beer	40/0.8/0.35	g l^{-1}	AP42
	Artificial panels		3	kg m^{-3}	AP42
	Pulp manufacturing	Kraft process	3.1	kg m^{-3}	AP42
	Coal mining and dressing	Coal washing	0.196	g kg^{-1}	AP42
	Municipal services	Sewage treatment	0.0011	g/kg	EEA, 2013
		Solid waste incineration/ landfill	1.23/1.56	g/kg	EEA, 2013
		Municipal waste incineration	0.02	g/kg	EEA, 2013

Table S2. VOC emission ratios (ppmv CO-1) and annual emissions (ton yr-1) for individual VOC species determined by the measurements and emission inventory for the PKU site (0.25 °×0.25 °grid).

Species	ER	Emissions		Species	ER	Emissions	
		ER	EI			ER	EI
Alkanes	--	--	--	**Halocarbon**	--	--	--
Ethane	5.93	1885.20	662.42	Chloromethane	0.51	313.83	17.08
Propane	3.15	1694.73	398.51	Chloroethylene	0.04	31.60	1.11
i-Butane	0.81	570.55	355.16	Bromomethane	0.00	5.54	0.38
n-Butane	1.19	843.33	465.99	Chloroethane	0.01	10.19	0.50
Cyclopentane	0.08	68.12	89.58	1,1-Dichloroethylene	0.00	3.05	1.11
i-Pentane	0.67	594.83	703.17	Dichloromethane	0.85	884.58	118.95
Pentane	0.44	386.99	478.85	1,1-Dichloroethane	0.05	55.37	1.11
2,2- Dimethylbutane	0.01	15.26	60.41	cis-1,2-dichloroethylene	0.02	22.43	2.90
2,3- Dimethylbutane	0.09	91.86	86.76	Chloroform	0.31	448.97	5.96
2- Methylpentane	0.18	185.45	418.12	1,1,1-Trichloroethane	0.00	2.49	0.88
3- Methylpentane	0.10	101.46	290.88	Carbon tetrachloride	0.06	118.09	17.80
n-Hexane	0.25	260.52	437.70	1,2- Dichloroethane	0.28	334.18	65.19
2,4-Dimethyl pentane	0.01	13.36	210.23	Trichloroethylene	0.04	62.77	418.53
Methylcyclopentane	0.10	104.64	182.83	1,2-Dichloropropane	0.24	330.41	41.10
2- methylhexane	0.04	51.41	226.73	tran-1,3-Dichloropropene	0.00	4.00	9.44
2,3- Dimethyl pentane	0.03	37.79	83.55	cis-1,3-Dichloropropene	0.00	1.71	13.52
3- methylhexane	0.05	60.53	186.69	1,1,2-Trichloroethane	0.02	25.19	3.75
2,2,4-Trimethylpentane	0.04	58.70	73.67	Tetrachloroethylene	0.04	72.83	604.10
n-Heptane	0.07	86.74	198.46	Chlorobenzene	0.01	16.63	4.23
Methylcyclohexane	0.04	49.54	168.08	1,3-Dichlorobenzene	0.00	3.10	4.35
2,3,4-Trimethylpentane	0.02	25.47	28.94	1,4-Dichlorobenzene	0.05	82.24	5.41
2- Methylheptane	0.02	30.22	112.59	Benzyl chloride	0.00	1.60	0.58
3- Methylheptane	0.01	13.71	172.36	1,2-Dichlorobenzene	0.00	4.27	3.85
Octane	0.04	53.59	179.01	**OVOCs**	--	--	--
Nonane	0.03	44.01	158.39	Acrolein	0.12	78.72	7.37
n-Decane	0.03	55.68	158.59	Propanal	0.12	85.76	37.96
Undecane	0.02	48.01	101.29	Isobutylene aldehyde	0.02	18.39	10.81
Alkenes	--	--	--	n-Butanal	0.05	42.57	82.25
Ethylene	5.38	1839.4	888.05	n-Pentanal	0.03	30.60	3.22
Propylene	1.24	635.64	517.20	Hexanal	0.10	122.55	25.96
tran-2-Butene	0.07	44.95	99.02	Acetone	1.61	1139.32	356.46
1-Butene	0.20	135.26	198.21	Methyl vinyl ketone	0.11	91.81	6.47
cis-2-Butene	0.04	26.43	91.90	2-Butanone	0.62	543.43	103.36
1,3-Butadiene	0.15	101.44	102.92	2-Pentanone	0.02	22.88	2.24
1-Pentene	0.03	23.48	82.68	3-Pentanone	0.03	33.78	0.38

tran-2-Pentene	0.02	13.65	82.26	Methyl acetate		0.24	216.87	1.52
Isoprene	0.03	24.70	24.22	MTBE		0.15	163.95	27.07
cis-2-Pentene	0.01	6.88	91.90	Vinyl acetate		0.01	6.24	11.21
1-Hexene	0.02	18.72	43.35	Ethyl acetate		0.65	697.92	531.73
Aromatics	--	--	--	Methyl methacrylate	0.58	704.38	0.12	
Benzene	0.78	741.09	940.01	Butyl acetate	0.32	452.31	3.04	
Toluene	0.86	968.52	1506.05	**Nitriles**	--	--	--	
Ethylbenzene	0.22	289.34	1441.41	Acetonitrile	0.21	0.21	16.52	
m/p-Xylene	0.25	318.34	2289.14	**Alkyne**	--	--	--	
o-Xylene	0.19	241.11	887.66	Acetylene	3.62	1326.39	390.19	
Styrene	0.08	99.30	313.55					
i-Propylbenzene	0.01	17.47	93.46					
n-Propylbenzene	0.02	27.69	110.55					
3-Ethyl toluene	0.05	75.91	140.27					
4-Ethyl toluene	0.03	40.44	176.27					
1,3,5-Trimethylbenzene	0.02	33.82	8.45					
2-Ethyl toluene	0.02	33.91	140.27					
1,2,4-Trimethylbenzene	0.07	95.59	520.92					
1,2,3-trimethylbenzene	0.02	35.52	185.99					
1,3-Diethyl benzene	0.00	8.07	40.78					
1,4-Diethyl benzene	0.02	25.15	69.66					

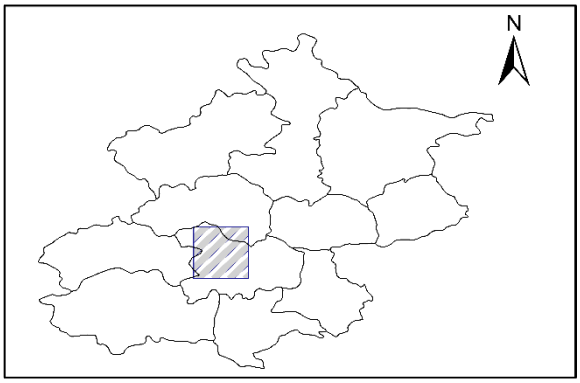

Figure S1 The emission grid PKU site loacted in (Black square).

Source identification

The first factor was identified as vehicle-related source. This factor explained 54% of MTBE, which is a widely used gasoline additive, used as an oxygenate to raise the octane number. This source was also dominated by a strong presence of C3-C5 alkane and alkene (propane, 61%; isobutane, 68%; n-butane, 75; isopentane, 76%; n-pentane, 68%; 1,3-butadiene, 64%) which can release from vehicle exhaust and fuel evaporation. Tunnel studies shown the toluene/benzene ratio for vehicular exhaust was about 1.6 (Kuster et al., 2004), and the mean toluene/benzene ratio of this source profile was 1.70. Acetylene, the combustion tracer, was explained 27% by this factor.

Factor two was the only factor with a distinct maximum in winter (January) as shown in figure 7. This factor contained rather short-lived alkenes such as ethylene (contained 62% of ethylene), propene (64%), trans-2-butene (49%), 1-butene (46%), typical for incomplete combustion processes (Leuchner et al., 2015). Large parts of the acetylene (49%) were also explained in this factor, which was major species emitted from combustion process (Liu et al., 2008). This factor could be attributed to residential heating and other combustion processes, and was identified as fuel combustion source.

The third source profile contains 65% of the total Freon113, which has a long lifespan in the atmosphere. This factor was characterized by high values of unreactive species, such as ethane, acetone, and benzene. Thus, this source was concluded as VOCs in aged air masses. The ratio between benzene and toluene is a useful indicator of the age of air masses. The mean benzene/toluene ratio of this source profile is 2.3, which is much higher than the typical values from vehicle exhaust (0.5) and solvent utilization (0.3). In aged air masses, the benzene/toluene ratios are higher than in fresh air (Wu et al., 2016). Moreover, the third factor contains large amount of acetonitrile (41%) and chloromethane (48%), which were regarded as the fingerprint of biomass burning. Therefore, this source was considered to represent aged air mass and biomass burning originating from air mass transport.

The fourth source was distinguished by significant amounts of chlorinated organic compounds, including 1,1-dichloroethane (65%), tetrachloroethylene (64%), chloroform (59%), 1,2-dichloropropane (54%), 1,2-dichloropropane (54%), which are tracers of industrial processes (Scheff and Wadden, 1993). This source was also characterized by significant percentage of esters (ethylacetate, 60%; n-butylacetate, 60%; methylacetate, 59%). Thus, the fourth source was identified as industrial processes source.

Factor five explained 66% and 75% of the measured α-pinene and β-pinene, respectively, which were indicators of biogenic emissions. Figure 7 reveals that the contribution of the fifth factor were presented higher levels in summer (July), and lower levels in winter (January). Biogenic emissions are strongly influenced by temperature and solar radiation (Guenther et al., 2012). Therefore, this factor was identified as biogenic emissions. The sixth factor shown in fig.6 was rich in aromatic species and C5-C8 substituted/cyclo-alkanes, which are markers for solvent utilization sources, including painting, printing, surface coating, and solvent emissions of household and consumer products (Liao et al., 2015). We therefore concluded it is from solvent utilization source.

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