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Trends of non-methane hydrocarbons (NMHC) emissions in Beijing during 2002–2013

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Trends of non-methane hydrocarbons (NMHC) emissions in Beijing during 2002–2013

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1. NMHC species measured at the PKU site

Table S1 NMHC species measured at the PKU site during 2004–2012.

Species	**Species**
Alkanes	***Aromatics***
ethane	benzene
propane	toluene
i-butane	ethylbenzne
n-butane	*m*,*p*-xylene
i-pentane	*o*-xylene
n-pentane	***Alkenes***
n-hexane	ethylene
Alkynes	propylene
acetylene	1-butene

2. NMHC emission standards for vehicle exhaust and paint & solvent use

Table S2 The implementation dates of tail pipe emissions regulations (Euro Ⅰ to Euro Ⅴ) for new vehicles in Beijing.

Vehicles	Euro Ⅰ	Euro Ⅱ	Euro Ⅲ	Euro Ⅳ	Euro Ⅴ
LDGV*	1999/1/1	2003/1/1	2005/12/30	2008/3/1	2013/2/1
HDGV*	2002/7/1	2003/9/1	2009/7/1	2010/7/1	--
HDDV*	2000/1/1	2003/1/1	2005/12/30	2008/7/1	2013/2/1
MC*	2001/1/1	2004/1/1	2008/7/1	--	--

* LDGV: light-duty gasoline vehicles; HDGV: high-duty gasoline vehicles; HDDV: high-duty gasoline vehicles; MC: motorcycles.

Table S3 The emission limits of total hydrocarbon (THC) for vehicles from Euro Ⅰ to Euro Ⅴ standards.

Vehicles	Euro Ⅰ	Euro Ⅱ	Euro Ⅲ	Euro Ⅳ	Euro Ⅴ
LDGV (g/km)	0.97*	0.5*	0.2	0.1	0.1
HDGV (g/kW·h)	14*	4.1*	0.35	0.29	--
HDDV (g/kW·h)	1.1	1.1	0.66	0.46	0.46
MC (g/km)	4	1.2	0.8	--	--

Table S4 Limits of benzene and other aromatics in national standards for paint and solvents in China.

Standard No.	National standard name	Limits of benzene	Limits for aromatics	Implementation Date
GB 24613-2009	Limit of harmful substances of coatings for toys	≤ 0.3%	TEX ≤ 30%	2010.10.01
GB 18581-2009	Indoor decorating and refurbishing materials-Limit of harmful substances of solvent based coatings for woodenware	≤ 0.3%	TEX ≤ 30%	2010.06.01
GB 24408-2009	Limit of harmful substances of exterior wall coatings	≤ 0.3%	TEX ≤ 40%	2010.06.01
GB 24409-2009	Limit of harmful substances of automobile coatings	≤ 0.3%	TEX ≤ 40%	2010.06.01
GB/T 23446-2009	Spray polyurea waterproofing coatings	≤ 200 mg/kg	TEX ≤ 1.0 g/kg	2010.01.01
GB/T 22374-2008	Floor coatings	≤ 0.1 g/kg (water based) ≤ 1.0% (solvent based)	TX ≤ 10 g/kg (water based) TX ≤ 200 g/kg (solvent based)	2009.05.01
HJ 457-2009	Technical requirement for environmental labeling products-Waterproof coatings	≤ 0.5 g/kg	TEX ≤ 80 g/kg (Epoxy and polyurethane coatings) TEX ≤ 50 g/kg (Polyurea coating)	2009.05.01
GB 18582-2008	Indoor decorating and refurbishing materials-Limit of harmful substances of interior architectural coatings	--	BTEX ≤300 mg/kg	2008.10.01
HG/T 3950-2007	Antibacterial coating	≤ 0.5%	TX ≤ 45% (Nitrocellulose); TX ≤ 40% (polyurethane); TX ≤ 5% (Alkyd paint)	2008.01.01
JC 1066-2008	Limit of harmful substances of building waterproof coatings	-- (water based) ≤ 200 mg/kg (reactive waterproof coating) ≤ 2 g/kg (solvent based)	BTEX ≤ 300 mg/kg (water based) TEX ≤ 1.0 g/kg (reactive coating A) TEX ≤ 5.0 g/kg (reactive coating B) TEX ≤ 400 g/kg (solvent based)	2008.07.01
HJ/T 414-2007	Technique requirement for Environmental labelling products-Solvent based wood coatings for indoor decoration and refurbishing	≤ 0.05%	TEX ≤ 25% (Nitrocellulose and polyurethane coatings); TEX ≤ 5% (Alkyd paint)	2008.04.01
	Cleaner production standard–Automobile manufacturing (Painting)	=0		2006.12.01
HJ/T 201-2005	Technical requirement for environmental labeling products-Water based coatings	--	TEX ≤ 300 mg/kg	2006.01.01
GB 50325-2001	Code for indoor environmental pollution control of civil building engineering	5 g/L	--	2002.01.01

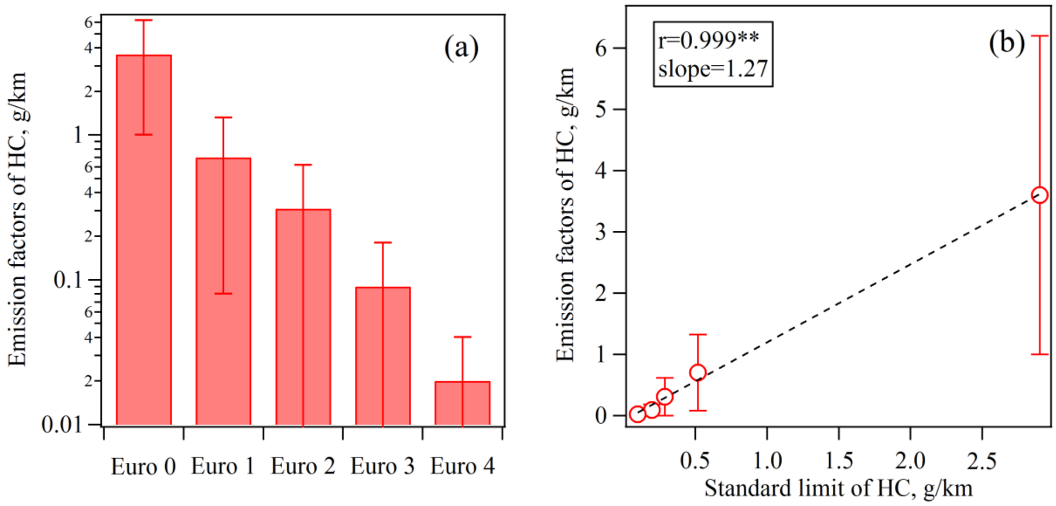

Fig. S1 (a) Emission factors of hydrocarbons for Euro 0–Euro 4 vehicles and (b) their relationship with hydrocarbon limits in vehicular emission standards.

3. Inter-comparison of NMHC measurements

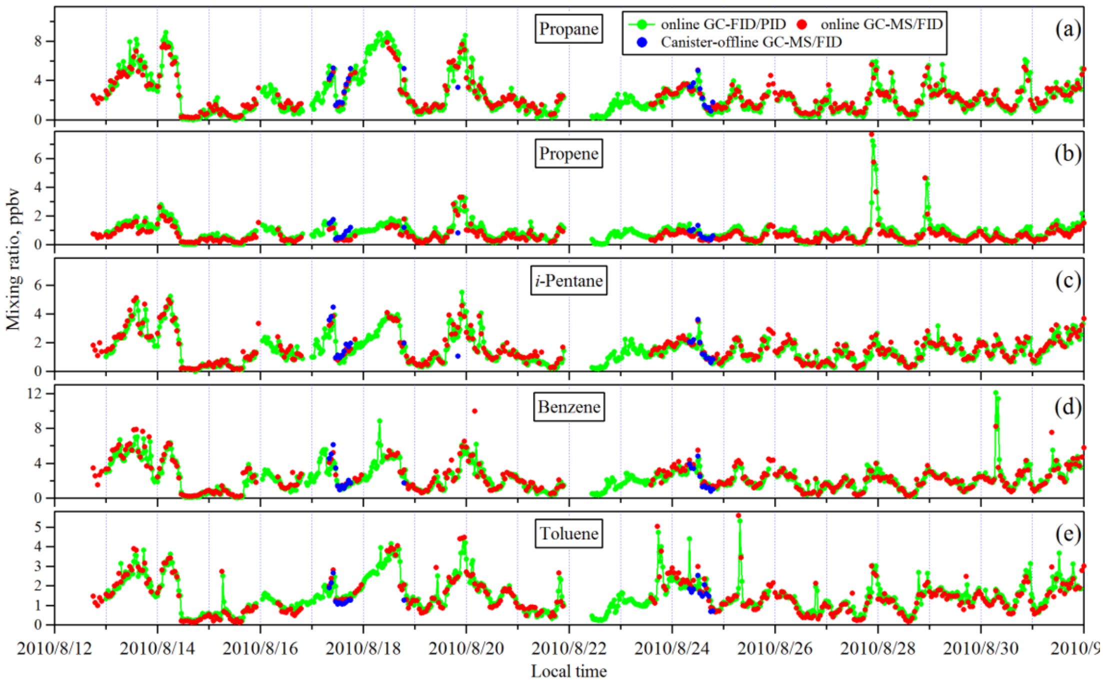

Fig. S2 Inter-comparisons of (a) propane, (b) propene, (c) *i*-pentane, (d) benzene, and (e) toluene measurements among online GC-MS/FID, online GC-FID/PID, and canister-offline GC-MS/FID systems.

4. Temporal changes in wintertime NMHC mixing ratios

Figure S3 compares average mixing ratios of ethane, acetylene, 1,3-butadiene, benzene, and toluene measured in urban areas of Beijing during January–February

2001 (Barletta et al., 2005), December 2002 (Liu et al., 2005) and December 2011–January 2012 (Wang et al., 2014). Average mixing ratios of ethane and acetylene during December 2011–January 2012 were 11.5 ± 6.8 and 7.9 ± 5.5 ppbv, lower than measured values during January–February 2001 by 30% and 77%, respectively. Average level of 1,3-butadiene during December 2011–January 2012 was 0.25±0.23 ppbv, significantly lower than the values of 1.3–1.4 ppbv in 2001 and 2002. Compared with ambient levels in January–February 2001, average mixing ratios of benzene in December 2002 and December 2011–January 2012 decreased by 28% and 59%, respectively. For toluene, the percentages of decline for these two years were 34% and 64%, respectively. It can be found that the relative decline percentages for mixing ratios of these NMHC species except ethane during the past decade were all greater than 50%. Additionally, we found that the relative decline rates for wintertime acetylene (7.0% yr^{-1}), benzene (5.5% yr^{-1}), and toluene (5.8% yr^{-1}) levels were similar with those for summertime observations (6.3% yr^{-1} for acetylene, 5.6% yr^{-1} for benzene, and 4.6% for toluene).

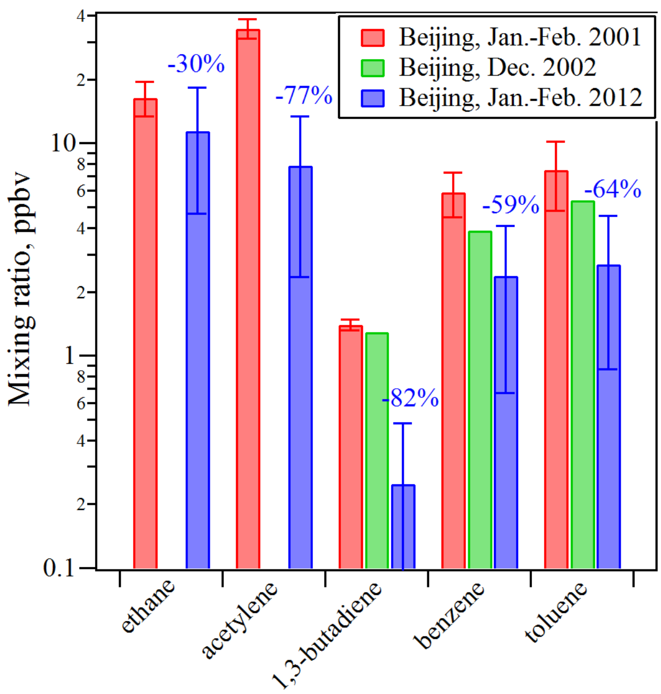

Fig. S3 Comparisons of mixing ratios for ethane, acetylene, 1,3-butadiene, benzene, and toluene measured during January–February 2001 (Barletta et al., 2005), December 2002 (Liu et al., 2005), and December 2011–January 2012 (Wang et al., 2014) in Beijing.

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