Supplement of Atmos. Chem. Phys., 23, 3819–3828, 2023
https://doi.org/10.5194/acp-23-3819-2023-supplement

Atmospheric Chemistry and Physics
Open Access

Supplement of

Vehicular ammonia emissions: an underappreciated emission source in densely populated areas

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Part I. Supplementary Figures

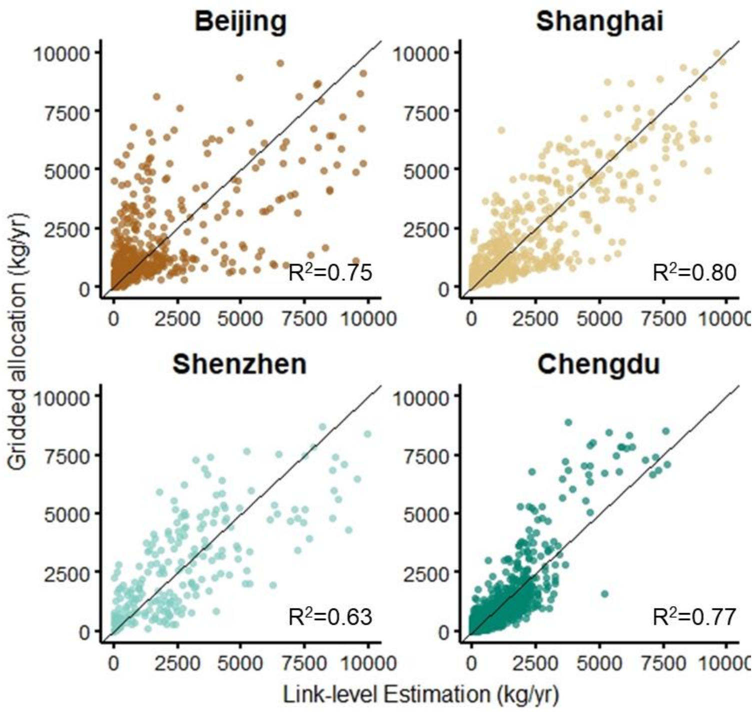

Fig S1 Comparison of the gridded allocations of on-road NH_3 emissions with the estimations based on link-level inventories.

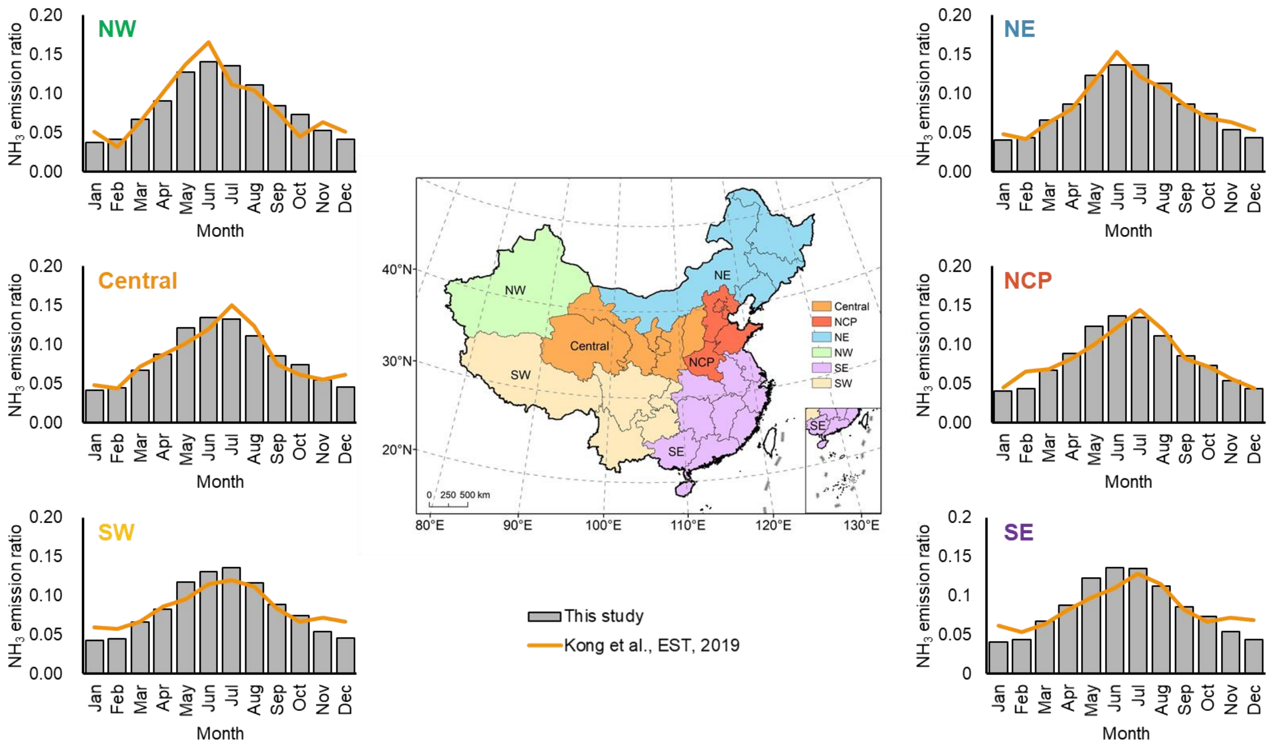

Fig S2 Comparison of the monthly variations of total anthropogenic NH_3 emissions derived in this study with surface

NH_3 observations obtained from the Ammonia Monitoring Network in different regions of China in Kong et al (Kong et al., 2019). NH_3 emissions are aggregated into six different regions of China, i.e., the North China Plain (NCP), Northeast China (NE), Southwest China (SW), Southeast China (SE), Northwest China (NW), and Central regions, denoted by different colors.

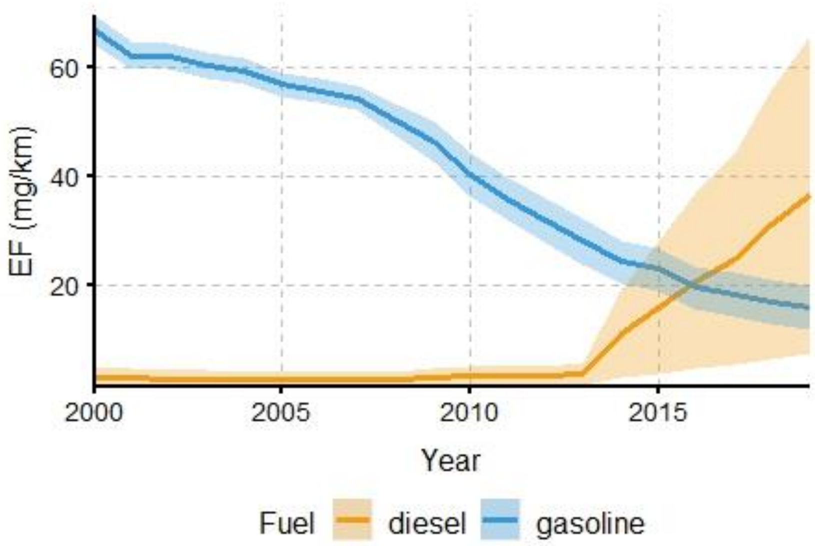

Fig S3 Trends of fleet average NH_3 EFs for gasoline and diesel vehicles in China, 2000-2019. Shadows show the uncertainty ranges.

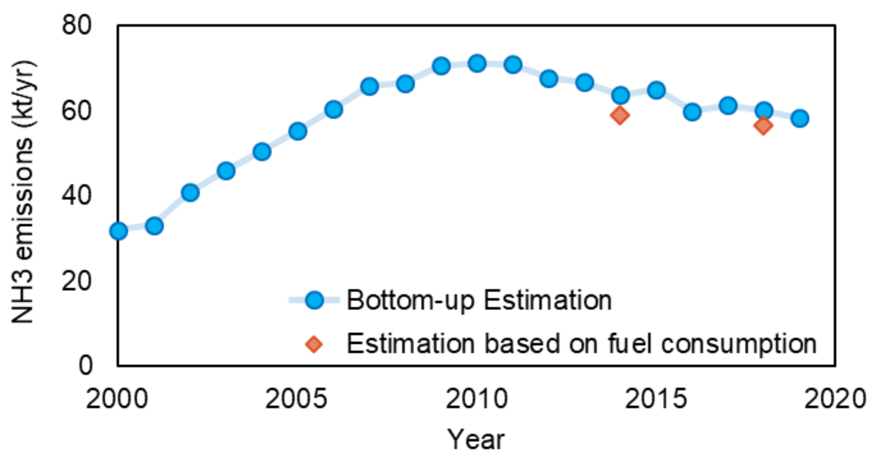

Fig S4 Validation of the national NH_3 emissions of gasoline fleet estimated by the bottom-up method in this study and top-down estimations based on annual gasoline consumptions.

Note: The national average NH_3 EFs for top-down estimation in 2014 and 2018 referred to the on-road measurements in developing cities and developed cities measured in 2014 by Sun et al. (2017) (Sun et al., 2017).

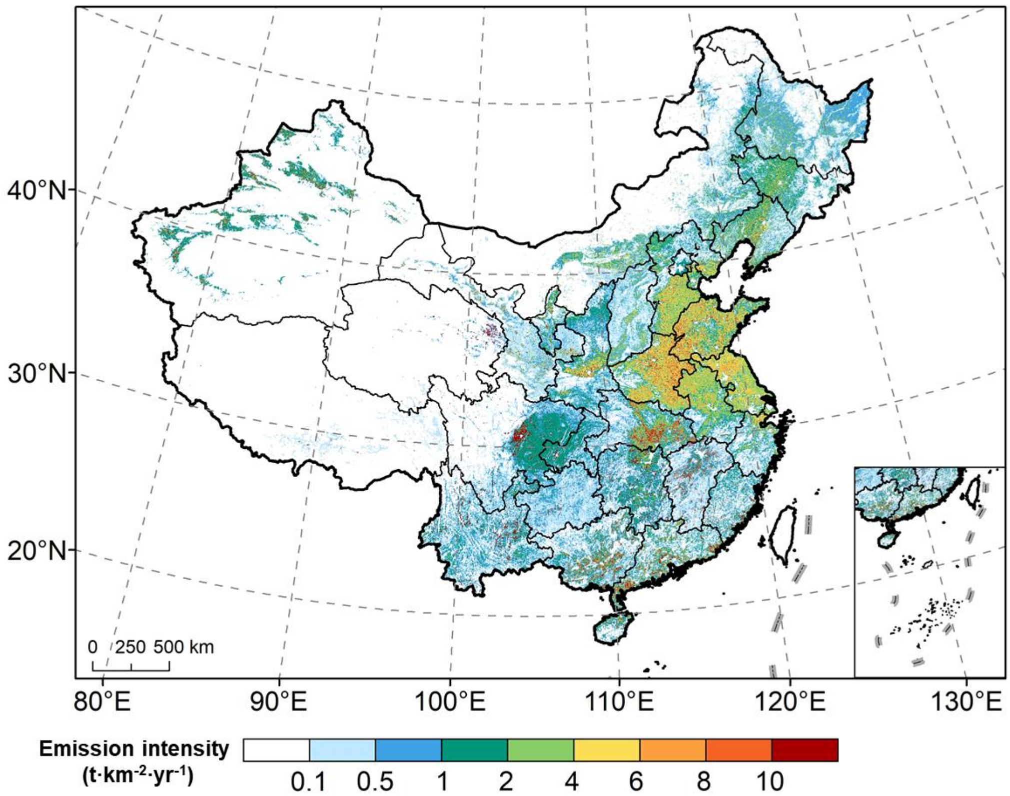

Fig S5 Highly spatial-resolved (3 km×3 km) agricultural NH_3 emission intensities in mainland China, 2019.

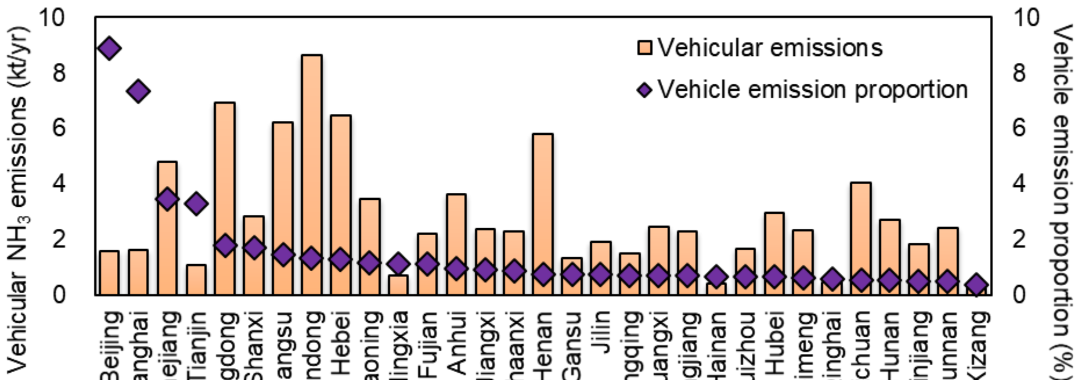

Fig S6 Annual vehicular NH_3 emissions and the proportions in total anthropogenic NH_3 emissions by province in China, 2019.

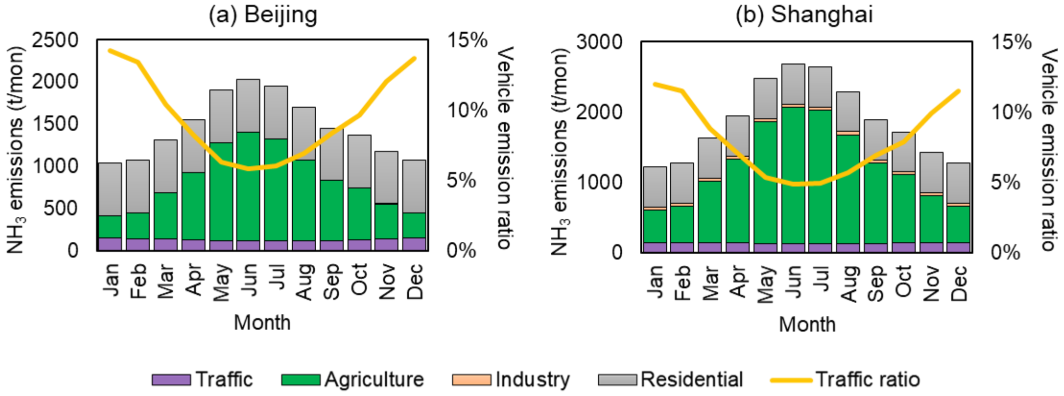

Fig S7 Monthly variation in anthropogenic NH_3 emissions and vehicle emission proportions in (a) Beijing, and (b) Shanghai in 2019.

Part II. Supplementary Tables

Table S1 Definition of vehicle categories in this study.

Vehicle catagory	Abbreviation	Discription	Feul type	Emission standard
Light-duty passenger vehicle	LDV	Length ≤3.5 m Passenger capacity ≤9	Gasoline/diesel/electric	China I to China VI
Medium-duty passenger vehicle	MDV	Length ＜6 m 9＜Passenger capacity ＜20	Gasoline/diesel/electric	China I to China VI
Heavy-duty passenger vehicle	HDV	Length ≥ 6 m Passengers capacity ≥20	Gasoline/diesel/electric	China I to China VI
Light-duty truck	LDT	Length＜6 m GVW[a] ≤ 4500 kg	Gasoline/diesel/electric	China I to China VI
Medium-duty trunk	MDT	Length ≥ 6 m 4500＜GVW ≤12000 kg	Gasoline/diesel/electric	China I to China VI
Heavy-duty truck	HDT	Length ≥ 6 m GVW＞12000 kg	Gasoline/diesel/electric	China I to China VI
Motorbike	Motorbike	Two-wheel motorbike	Gasoline	China I to China IV
Taxi	Taxi		Gasoline/electric	China I to China VI
Bus	Bus		Diesel/electric	China I to China VI

Note: [a] GVW: Gross vehicle weight.

Table S2 NH_3 EFs of LDGVs or LDGV-dominated fleets reported by various studies.

Model year	Location	Experimental Method	Fleet	Original value	NH_3 EF (mg/km)	Reference
2000-2019	China	Estiamtion based on average engine combustion efficiency	China 1 LDGV		86.0	This study
			China 2 LDGV		60.7	
			China 3 LDGV		25.5	
			China 4 LDGV		19.1	
			China 5 LDGV		10.9	
			China 6 LDGV		8.0	
2001	Empa, EU	Dynamometer (EDC)	Euro 3 LDGV	22 mg/km	22	(Heeb et al., 2006)
2000-2004	Swiss, EU	Dynamometer (EDC)	Euro 3 LDGV	16±12 mg/km	16±12	(Heeb et al., 2008)
			Euro 4 LDGV	10±7 mg/km	10±7	
2008	Italy, EU	Dynamometer (NEDC)	Euro 5 LDGV	13.9±11.3 mg/km	13.9±11.3	(Suarez-Bertoa et al., 2014)
2008	Italy, EU	Dynamometer (WLTC)	Euro 5 LDGV	11±1 mg/km	11±1	(Suarez-Bertoa et al., 2015)
2006-2016	Shanghai, China	Dynamometer (WLTC)	China 2 LDGV	30.8 mg/km	30.8	(Huang et al., 2018)
			China 3 LDGV	18.8 mg/km	18.8	
			China 4 LDGV	12.6 mg/km	12.6	
			China 5 LDGV	10.8 mg/km	10.8	
2020	UK, EU	Dynamometer (WLTC)	Euro 6 LDGV	13.0 mg/km	13.0	(Selleri et al., 2022)
	Beijing, China	Dynamometer (WLTC)	China 6 LDGV	0.65±0.38 ~ 8.01±3.12mg/km	0.7±0.4 ~8.0±3.1	(Wang et al., 2018)
1985-2012	UK, EU	Remote sensing	Euro 1 LDGV	0.93±0.12 ppbv/ppmv CO_2	59.6±7.7	(Carslaw and Rhys-Tyler, 2013)
			Euro 2 LDGV	0.94±0.04 ppbv/ppmv CO_2	60.2±2.6	
			Euro 3 LDGV	0.78±0.03 ppbv/ppmv CO_2	50.0±1.9	
			Euro 4 LDGV	0.54±0.02 ppbv/ppmv CO_2	34.6±1.3	
			Euro 5 LDGV	0.34±0.04 ppbv/ppmv CO_2	21.8±2.6	
2000-2019	UK, EU	Remote sensing	Euro 2 LDGV	1.13±0.04 g/kg fuel	63.3±2.2	(Farren et al., 2021)
			Euro 3 LDGV	0.93±0.02 g/kg fuel	52.1±1.1	
			Euro 4 LDGV	0.78±0.01 g/kg	43.7±0.6	

Model year	Location	Experimental Method	Fleet	Original value	NH_3 EF (mg/km)	Reference
				fuel		
			Euro 5 LDGV	0.64±0.03 g/kg fuel	35.8±1.7	
			Euro 6 LDGV	0.49±0.01 g/kg fuel	27.4±0.6	
2000 in average	San Jose, US	Remote sensing	LDGV-dominated	0.49±0.01 g/kg fuel	27.4±0.6	(Bishop et al., 2010)
	Fresno, US			0.49±0.01 g/kg fuel	27.4±0.6	
	LA, US			0.79±0.02 g/kg fuel	44.2±1.1	
2005 in average	Denver, US	Remote sensing	LDGV	0.45±0.02 g/kg fuel	25.2±1.1	(Bishop and Stedman, 2015)
2004 in average	LA, US		LDGV	0.59±0.02 g/kg fuel	33.0±1.1	
2006 in average	Tulsa, US		LDGV	0.44±0.01 g/kg fuel	24.6±0.6	
Pre 1999	CA, US	Tunnel Test	LDGV-dominated	78±6.0 mg/km	78±6.0	(Kean et al., 2000)
Pre 1995	San Francisco, US	Tunnel Test	LDGV-dominated	74.3±4.5 mg/km	74.3±4.5	(Kean et al., 2009)
1996-2000				45.1±3.0 mg/km	45.1±3.0	
Pre 2011	São Paulo, Brazil	Tunnel Test	LDGV-dominated	44±22 mg/km	44±22	(Vieira et al., 2016)
Pre 2015	Shanghai, China	Tunnel Test	LDGV-dominated	28±5 mg/km	28±5	(Chang et al., 2016)
2000-2007	CA, US	On-road mobile measurement	LDGV-dominated	0.49±0.06 g/kg fuel	27.4±3.4	(Sun et al., 2014)
Pre 2014	Developed cities in China	On-road mobile measurement	LDGV-dominated	0.37±0.06 ppbv/ppmv CO_2	23.7±3.8	(Sun et al., 2017)
	Developing cities in China			0.50±0.08 ppbv/ppmv CO_2	32.0±5.1	

Note: To compare NH_3 EFs from various literatures, the units were converted to mg/km based on the following rules: 1) For original value in units of ppbv/ppmv CO_2, the NH_3:CO_2 emission ratio can be converted to NH_3 EF (mg/kg fuel) using Eq. S(1) (Sun et al., 2017).

$$EF\left(\text{mg/kg fuel}\right) = \frac{ER(\text{in ppbv/ppmv CO}_2)\cdot 17 W_C F_{CO_2}}{12} \quad (1)$$

where W_C = 0.85, is the mass fraction of carbon in gasoline. F_{CO2} = 95%~99%, is the molar fraction of CO_2 in the total combusted carbon, which can be approximated by Eq. S(2) (the contributions of hydrocarbons/soots are negligible).

$$F_{CO_2} = \frac{[CO_2]}{[CO_2]+[CO]} \quad (2)$$

2) The original value in units of g/kg fuel can be converted to mg/km by Eq. S(3).

$$EF(\text{mg/km}) = EF(\text{g/kg fuel})\cdot FC(\text{g/km}) \quad (3)$$

where FC is the fleet average fuel consumption. FC= 56 g/km for LDGVs according to COPERT model (Ntziachristos et al., 2009).

Table S3 NH_3 EFs of HDDVs reported by various studies.

Reference	Experimental Method	Emission standard	After-treatment	Original value	NH_3 EF (mg/km) [a]
This study	PEMS and Dynamometer (C-WTVC)	China III	Without SCR	0.02±0.01 g/kg fuel	4.4±2.4
		China IV/V	SCR-equipped	0.31±0.50 g/kg fuel	73.9±118.7
(Suarez-Bertoa et al., 2016)	PEMS	Euro V	SCR-equipped	41.0±17.4 mg/kWh	102.5±43.5
(Tadano et al., 2014)	Dynamometer (ESC)	Euro V	Without SCR	4±2 mg/kWh	10±5
			SCR-equipped	60±20 mg/kWh	150±50
(Preble et al., 2019)	Field sampling	2010-2018 MY	SCR-equipped	0.18±0.07 g/kg fuel	43.2±16.8
(Jeon et al., 2016)	Dynamometer (WTHC)	Euro VI	SCR+ASC [b]	5 mg/kWh	12.5
(Khalek et al., 2015)	Dynamometer (FTP)	2010-compliant engines	SCR+ASC	3.4±1.9 mg/kWh	8.4±4.7
(Mendoza-Villafuerte et al., 2017)	PEMS	Euro VI	SCR+ASC	13.5±4.5 mg/kWh	33.8±11.3

Note: [a] The units of EF were converted to mg/km based on the following rules:

1) The original value in units of g/kg fuel can be converted to mg/km by Eq. S(4).

$$EF(\text{mg/km}) = EF(\text{g/kg fuel})\cdot FC(\text{g/km}) \quad (4)$$

where FC is the fleet average fuel consumption. FC= 240 g/km for HDDVs according to COPERT model (Ntziachristos et al., 2009).

2) The original value in units of mg/kWh can be converted to mg/km by Eq. S(5). based on the energy consumption value (EC=9 MJ/km for HDDVs) according to COPERT model (Ntziachristos et al., 2009).

$$EF\,(\text{mg/km}) = EF\,(\text{mg/kWh})/3.6 \cdot EC(\text{MJ/km}) \tag{5}$$

where EC is the fleet average energy consumption. EC=9 MJ/km for HDDVs according to COPERT model (Ntziachristos et al., 2009).

[b] Ammonia Slip Catalyst (ASC).

Table S4 Uncertainty ranges of NH_3 emission factors used in this study.

Vehicle types	Emission standards	Uncertainty ranges
LDGV	Euro/China 2	4%
LDGV	Euro/China 3	27%
LDGV	Euro/China 4	25%
LDGV	Euro/China 5	33%
LDGV	Euro/China 6	38%
HDDV	Without SCR	52%
HDDV	SCR-equipped	81%
HDDV	SCR+AMOX	45%

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