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Source apportionment of atmospheric ammonia before, during, and after the 2014 APEC summit in Beijing using stable nitrogen isotope signatures

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Table

Table S1. Source information, sampling time (from June 2014 to January 2015), mass concentration ($\mu\text{g m}^{-3}$) and $\delta^{15}\text{N}$ value (‰) of the 44 NH_3 samples used in this study.

Category	Sub-category	Sampling Site	Time on	Time off	Season	NH_3 conc.	$\delta^{15}\text{N-NH}_3$	Method	Comments
Livestock	pig	piglet sty-SHPF	9 th 12, 7:15	9 th 12, 19:15	winter	520.3	-27.2	PT-IRMS	a
		piglet sty-SHPF	10 th 12, 7:10	10 th 12, 19:10	winter	667.5	-28.2	PT-IRMS	a
		piglet sty-SHPF	3 rd 6, 7:10	3 rd 6, 19:10	summer	1334.7	-29.3	PT-IRMS	a
		piglet sty-SHPF	4 th 6, 7:00	4 th 6, 19:00	summer	1151.4	-31.7	PT-IRMS	a
		adult pig sty-SHPF	9 th 12, 7:25	9 th 12, 19:25	winter	462.2	-27.1	PT-IRMS	a
		adult pig sty-SHPF	10 th 12, 7:20	9 th 12, 19:20	winter	696.2	-30.3	PT-IRMS	a
		adult pig sty-SHPF	4 th 6, 7:10	4 th 6, 19:10	summer	1502.8	-29.8	PT-IRMS	a
On-road traffic	vehicle	Handan Tunnel	1 st 7, 19:00	1 st 8, 7:40	summer	92.1	-9.6	PT-IRMS	b
		Handan Tunnel	1 st 8, 7:40	1 st 9, 15:30	summer	87.4	-12.2	PT-IRMS	b
		Handan Tunnel	3 rd 12, 11:18	9 th 1, 9:43	winter	56.5	-16.7	PT-IRMS	b
		Handan Tunnel	9 th 1, 9:43	5 th 2, 10:15	winter	61.2	-17.8	PT-IRMS	b
		Handan Tunnel	1 st 7, 19:05	1 st 8, 7:45	summer	83.7	-11.9	PT-IRMS	b
		Handan Tunnel	1 st 8, 7:45	1 st 9, 15:35	summer	77.5	-14.1	PT-IRMS	b
		Handan Tunnel	3 rd 12, 11:22	9 th 1, 9:47	winter	35.7	-16.4	PT-IRMS	b
		Handan Tunnel	9 th 1, 9:47	5 th 2, 10:20	winter	33.2	-15.1	PT-IRMS	b
Waste	solid waste	Liangcheng Estate WTS	27 th 5, 12:10	4 th 6, 10:17	summer	329.6	-29.9	EA-IRMS	c
		Liangcheng Estate WTS	4 th 6, 10:20	11 th 6, 9:45	summer	412.8	-31.4	EA-IRMS	f
		Liangcheng Estate WTS	5 th 1, 11:43	12 th 1, 9:40	winter	363.8	-37.2	EA-IRMS	f
		Liangcheng Estate WTS	12 th 1, 9:40	12 th 1, 21:40	winter	271.2	-36.0	PT-IRMS	f
		Fudan campus WTS	27 th 5, 10:53	4 th 6, 9:20	summer	488.6	-32.2	EA-IRMS	f
		Fudan campus WTS	4 th 6, 9:30	11 th 6, 8:34	summer	542.4	-30.7	EA-IRMS	f
		Fudan campus WTS	6 th 1, 8:50	6 th 1, 20:50	winter	275.5	-35.7	PT-IRMS	f
		Fudan campus WTS	13 th 1, 7:30	13 th 1, 19:30	winter	349.4	-37.6	PT-IRMS	f
	wastewater	Quyong WWTP	5 th 7, 14:20	6 th 7, 14:20	summer	258.5	-40.7	PT-IRMS	d
		Quyong WWTP	5 th 7, 14:20	6 th 7, 14:20	summer	247.0	-41.9	PT-IRMS	g
		Quyong WWTP	10 th 1, 10:55	11 th 1, 10:55	winter	127.2	-40.6	PT-IRMS	g
		Quyong WWTP	10 th 1, 10:55	11 th 1, 10:55	winter	151.0	-40.9	PT-IRMS	g
		Quyong WWTP	5 th 7, 14:23	6 th 7, 14:23	summer	236.2	-40.7	PT-IRMS	g
		Quyong WWTP	5 th 7, 14:23	6 th 7, 14:23	summer	243.3	-42.0	PT-IRMS	g
		Quyong WWTP	10 th 1, 10:58	11 th 1, 10:58	winter	154.8	-39.2	PT-IRMS	g
human excreta	Quyong WWTP	10 th 1, 10:58	11 th 1, 10:58	winter	142.1	-41.9	PT-IRMS	g	
	Dongfang Estate	16 th 7, 14:25	16 th 7, 16:25	summer	3295.8	-39.3	PT-IRMS	e	
	Dongfang Estate	7 th 8, 14:40	7 th 8, 16:40	summer	3534.3	-38.1	PT-IRMS	h	
	Dongfang Estate	9 th 1, 9:10	9 th 1, 11:10	winter	3415.5	-37.9	PT-IRMS	h	
	Dongfang Estate	12 th 1, 9:35	12 th 1, 11:35	winter	3238.0	-39.2	PT-IRMS	h	
	Fudan teaching building	13 th 7, 9:15	13 th 7, 11:15	summer	5272.5	-37.3	PT-IRMS	h	
	Fudan teaching building	5 th 8, 13:50	5 th 8, 15:50	summer	6211.0	-39.0	PT-IRMS	h	
	Fudan teaching building	7 th 1, 8:40	7 th 1, 10:40	winter	5492.2	-37.4	PT-IRMS	h	
Fudan teaching building	14 th 1, 8:30	14 th 1, 10:30	winter	5614.7	-39.6	PT-IRMS	h		

Fertilizer	urea	Fudan laboratory	10 th 5, 8:50	10 th 5, 14:50	/	165.6	-52.0	EA-IRMS	f
		Fudan laboratory	10 th 5, 8:50	10 th 5, 17:50	/	247.5	-51.4	EA-IRMS	i
		Fudan laboratory	10 th 5, 8:50	10 th 5, 20:50	/	369.1	-50.2	EA-IRMS	i
		Fudan laboratory	10 th 5, 8:50	10 th 5, 23:50	/	575.4	-47.6	EA-IRMS	i
		Fudan laboratory	10 th 5, 8:50	11 th 5, 1:00	/	623.7	-48.7	EA-IRMS	

- a. Pigs dominate the livestock breeding in Eastern China, so as to Shanghai and its surrounding regions where almost all livestock farms are pigs. Therefore, Ogawa PSDs were deployed in the Shanghai pig breeding farm (SHPF; 121.582°E, 30.856°N), a leading pig production unit in and around the region. Specifically, four Ogawa NH₃ PSDs were attached to the center pillars (~1.5 m above ground level) of two pigsties, a 66-head piglet sty and a 25-head adult pig sty. Both sites are in 38.5 m² without the operation of ventilation fan during our sampling period, which could significantly reduce the interference of open air.
- b. Tests were conducted in the Handan Road tunnel (121.509°E, 31.302°N; ~2 m above ground level), a city tunnel located in the front of the main gate of Fudan University. As a part of the Mid-ring Road in Shanghai and one of the busiest traffic sections, the average traffic density in the tunnel is around 150000 vehicles per day, most of which are personal and light-duty gasoline vehicles. The tunnel is 760 m long with two bores of four lanes in each direction, and each bore has a cross-sectional area of 66.4 m² (14.75 m × 4.5 m).
- c. The production of odors (including NH₃) from municipal solid waste (MSW) is an inevitable consequence of urbanization, and this is particularly true in the waste transfer stations (WTS) where MSW is unloaded from garbage collection trucks and temporarily stored in a compacting container before being reloaded onto larger long-distance transport vehicles for shipment to landfills or other treatment facilities. Two WTSs in the metropolitan areas of Shanghai were selected, one is adjacent to a densely populated residential community called Zhongxing Liangcheng Estate (121.479°E, 31.305°N) and another is in the teaching zone of Fudan University (121.511°E, 31.306°N). At each site, two Ogawa NH₃ PSDs were intruded into the garbage compacting container for one week (EA-IRMS) or 12 hr (PT-IRMS).
- d. The storage and treatment of municipal wastewater and domestic sewage in wastewater treatment plants (WWTPs) appears to be a source NH₃ in urban areas. To collect wastewater-originated NH₃, four Ogawa NH₃ PSDs were hanged on two ropes through a protective shield (designed mainly to avoid the escape of toxic H₂S) over the influent sewer of Quyang WWTP (121.492°E, 31.285°N), in which a large amount of NH₃ can be produced through the anaerobic decomposition of urea and other organic N compounds in wastewater.
- e. In urban China, human excreta are typically stored in a three-grille septic tank under the building at first. After a series of anaerobic decomposition processes, a substantial amount of odors (including NH₃) will be generated and emitted through a ceiling duct. The liquid supernatant will then flow away from the third grille to the municipal sewage system. Samples were collected from the ceiling ducts of two buildings (a residential building called Dongfang Estate with 21 residents and a five-floor teaching building in Fudan campus, respectively). To prevent potential perturbation of the outdoor environment, measurements were taken approximately 1.5 m from the exhaust end of the three ceiling ducts.
- f. China consumed 33.6 Mt or 33% of the world's N-fertilizer in 2009, 66.7% of them were urea, and the rest. The total amount of N-fertilizer application in Beijing is around 100 kt annually, in which urea accounts for nearly 70%. Extensive laboratory simulations and in situ micrometeorological measurements have confirmed that the NH₃ volatilization rates are of highly variable and controlled by various factors such as soil pH, moisture, and temperature. While quantification of the emission factor from urea-N application was beyond the scope of this study. To collect NH₃ solely from urea volatilization, a simplified laboratory volatilization experiment, similar to the one described by [Roelcke *et al.*, Laboratory measurements and simulations of ammonia volatilization from urea applied to calcareous Chinese loess soils, *Plant & Soil*, 1996, 181:123-129] was performed. Specifically, five volatilization apparatuses were running in parallel. 1 L soils collected directly from a vegetable field (top layer of 0-20 cm) were poured into a glass breaker (2 L) first, and then 100 g urea was spread evenly over the soil surface together with a fume cupboard for each measurement; a clean tray and two Ogawa NH₃ PSDs (consisting four filters) were then deposited into it in sequence. After that, the mouth of the breaker was sealed with plastic film for half hour, leaving a little opening to decrease the air pressure in it. Four

filters in two PSDs: a filter was used to determine NH_3 concentration; the other three were weighed and enclosed in tin capsules to be analyzed by an elemental analyzer combustion coupled with IRMS.

Figures

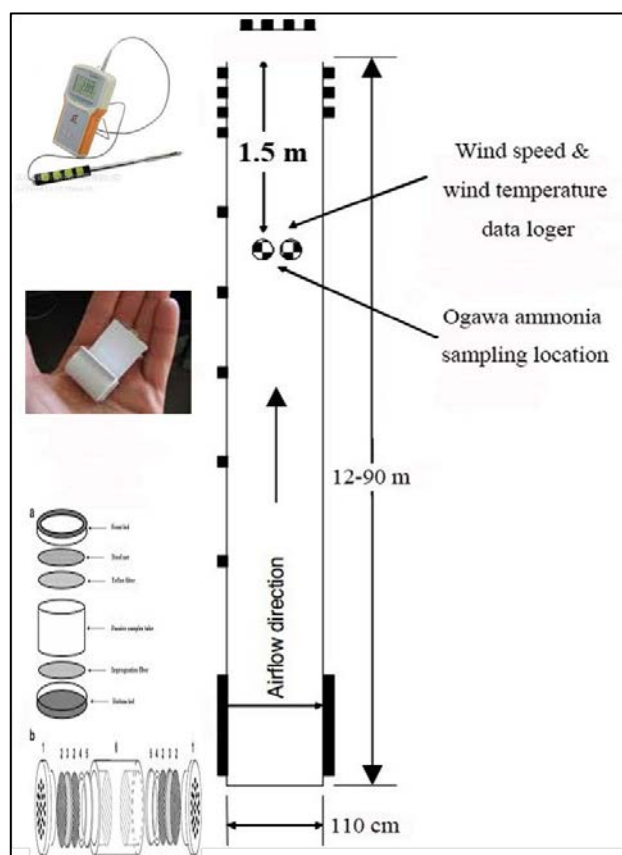


Figure S1. A schematic of the sampling in the exhaust of ceiling duct. According to China’s “Code for planning of urban environmental sanitation facilities” (GB 50337-2003), “fecal sewage front-end processing facility”, also known as septic tanks, are recommended in building construction for human excreta pre-treatment to avoid direct discharging of fecal sewage into the urban sewage pipe network. Although it is not mandatory to install septic tanks, to reduce the burden of wastewater treatment in sewage treatment plants, almost all cities in China have adopted the septic tank system (Cheng, H., H. Qiu, and M. Liu (2010), Review of setting septic tank in building outdoor drainage. *Fujian Architecture & Construction*, 129(150), 4-5. In Chinese with English abstract). In Shanghai, human excreta are also stored in a three-grille septic tank under the building at first. After a series of anaerobic decomposition processes, a substantial amount of odors (including NH_3) will be generated and emitted through a ceiling duct (polyvinyl chloride pipe). In our study, to avoid the inference of ambient air, samplings were taken approximately 1.5 m from the exhaust end of the ceiling duct. Moreover, although the mouth of the ceiling duct was open, the ceiling duct can be viewed as a one-way outflow channel of NH_3 emissions (from septic tank to open atmosphere).

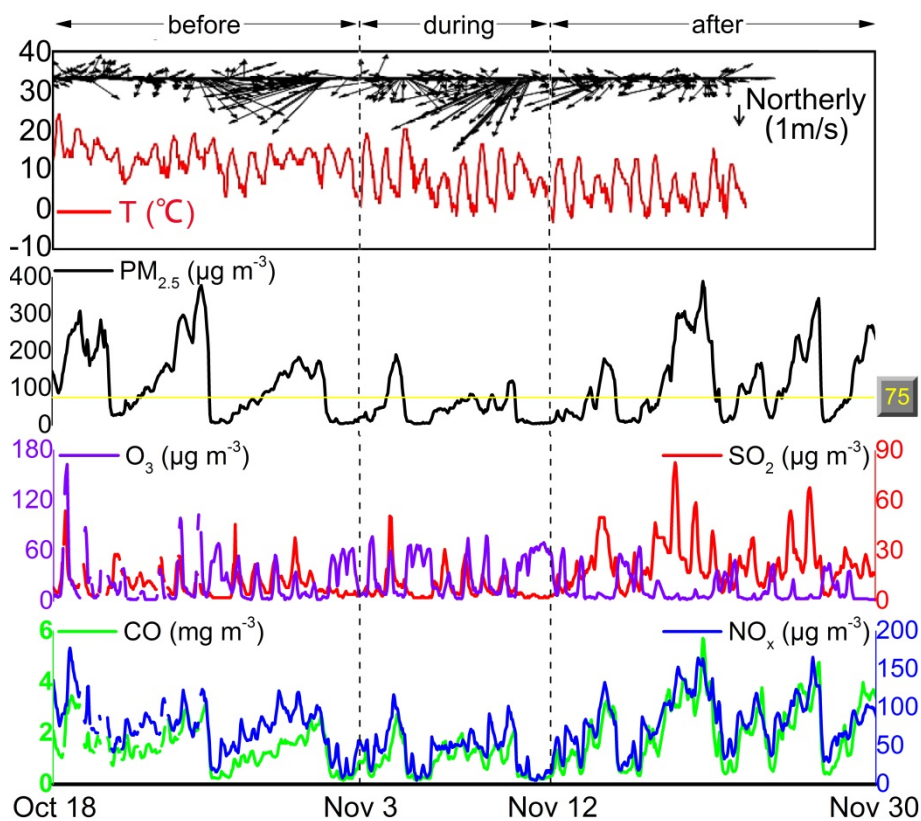


Figure S2. Time-series evolutions of hourly meteorological parameters (temperature, wind speed & direction; modified from *Li, et al. ACP, 2015, 15(14): 7945-7959. Doi: 10.5194/acp-15-7945-2015*) and air pollutants and before, during, and after the 2014 APEC summit in Beijing.

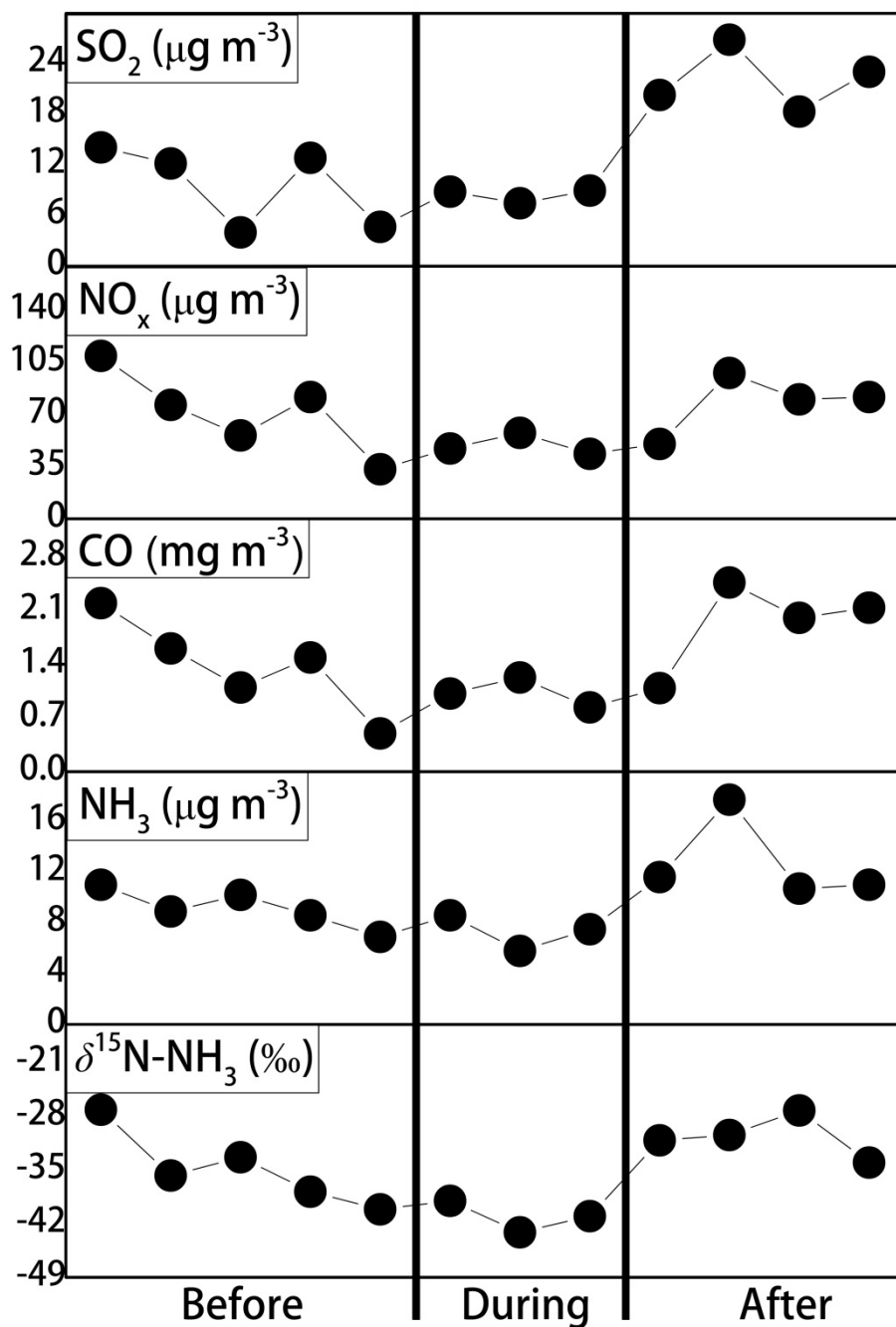


Figure S3. Time-series evolutions of mass concentrations of SO₂, NO_x, CO, NH₃, and isotopic composition of NH₃ (δ¹⁵N-NH₃) for each NH₃ sampling event in Beijing before, during, and after the 2014 APEC summit.