

Supplementary materials

Table S1. Results of $\delta^{15}\text{N}$ and chemical analyses of individual $\text{PM}_{2.5}$ samples at CRAES site of Beijing and at the background site (Menyuan, Qinghai province) of China.

Site	ID	$\text{PM}_{2.5}$ ($\mu\text{g}/\text{m}^3$)	NO_2 ($\mu\text{g}/\text{m}^3$)	$\delta^{15}\text{N}$ (‰)	TN (%)	$\text{NH}_4^+\text{-N}$ (%)	$\text{NO}_3^-\text{-N}$ (%)	$\text{SO}_4^{2-}\text{-S}$ (%)
Beijing	CRAES001	43.0	57.0	-2.7	14.9	5.9	4.1	2.5
Beijing	CRAES002	101.3	61.5	-2.6	15.0	6.7	8.2	2.4
Beijing	CRAES003	119.4	84.6	10.4	8.2	4.4	0.7	8.3
Beijing	CRAES004	377.7	111.1	13.5	16.5	12.5	1.4	6.8
Beijing	CRAES005	433.6	109.4	12.3	15.2	4.4	1.2	8.0
Beijing	CRAES006	204.1	80.0	8.4	18.4	12.1	4.0	8.2
Beijing	CRAES007	289.0	87.3	8.0	15.1	11.0	3.7	7.5
Beijing	CRAES008	231.7	74.3	3.1	13.5	3.5	4.4	4.0
Beijing	CRAES009	325.6	71.0	2.6	16.0	4.0	3.9	2.4
Beijing	CRAES010	388.9	114.9	-1.9	16.3	4.7	8.7	5.9
Beijing	CRAES011	398.2	122.0	-3.3	19.8	6.0	9.4	4.9
Beijing	CRAES012	270.0	112.0	-4.1	29.3	12.9	9.4	8.3
Beijing	CRAES013	246.0	89.8	-2.4	18.4	6.6	6.6	3.3
Beijing	CRAES014	272.3	74.4	-2.1	17.7	9.3	4.6	4.1
Qinghai	Menyuan001	11.7	6.5	21.2	15.1	9.3	1.9	0.3
Qinghai	Menyuan002	14.6	3.8	27.8	13.8	6.3	2.2	0.2

Qinghai	Menyuan003	10.4	4.6	19.9	3.3	5.6	1.7	0.2
Qinghai	Menyuan004	16.8	6.7	20.3	3.7	5.2	1.6	0.3
Qinghai	Menyuan005	10.0	3.2	20.7	18.7	9.4	1.9	0.2
Qinghai	Menyuan006	7.0	4.5	11.8	1.4	3.1	2.6	0.2
Qinghai	Menyuan007	13.2	4.2	12.3	4.6	5.0	2.0	0.2
Qinghai	Menyuan008	17.8	5.1	8.0	6.2	3.5	2.5	0.2
Qinghai	Menyuan009	15.9	3.0	17.8	6.9	5.4	1.2	0.2
Qinghai	Menyuan010	12.0	2.6	27.9	14.8	6.5	2.3	0.2
Qinghai	Menyuan011	13.9	3.1	16.8	5.1	5.8	1.2	0.3
Qinghai	Menyuan012	9.1	3.5	12.1	3.1	5.1	2.1	0.2
Qinghai	Menyuan013	12.8	5.2	21.0	10.6	5.9	1.7	0.3
Qinghai	Menyuan014	17.0	4.4	21.4	13.0	6.0	1.8	0.3

Table S2. Mean or medium (with *) $\delta^{15}\text{N}$ values reported for major N emissions in the atmosphere. Medium values were calculated using the minimum and maximum values (ranges in parentheses) reported in relevant references.

Sources	N species	$\delta^{15}\text{N}$ / ‰	References
Coal combustion	NO_2	+20.0 (+6.0 ~ +26.0)	Felix et al., 2012 and references cited therein (Moore, 1974, 1977; Heaton, 1990; Kiga et al., 2000; Laffray et al., 2000; Heaton et al., 2004; Elliott et al., 2009)
Coal combustion	NH_3	-6.5* (-15.0 ~ +2.0)	Felix et al., 2013
Vehicle exhausts	NO_2	-2.5 (-19.1 ~ +9.8)	Walters et al., 2015
Vehicle exhausts	NH_3	-4.0* (-5.0 ~ -3.0)	Felix et al., 2013
Wastes & sewage	NH_3	-9.6* (-15.2 ~ -4.0)	Moore, 1977; Heaton, 1986; Freyer, 1991
Biomass burning	NO_2	+13 (+8.0 ~ +15.0)	Hastings et al., 2009; Felix et al., 2013, 2014
Biomass burning	NH_3	+12.0 (n.a)	Kawashima & Kurahashi, 2011
Animal wastes	NO_2	-19.0* (-29.0 ~ -9.0)	Felix et al., 2013; Felix & Elliott, 2014
Animal wastes	NH_3	-39.5* (-56.0 ~ -23.0)	Felix et al., 2013, 2014
Soil N cycle	NO_2	-29.0* (-48.0 ~ -20.0)	Middlecamp & Elliot, 2011; Felix et al., 2013; Felix & Elliott, 2014
Fertilizer application	NH_3	-36.8* (-48.0 ~ -25.6)	Felix et al., 2013, 2014