The supplemental material has 9 pages and includes the following items:

Text S1. The reason why the reaction between NH_3 and H_2SO_4 proceeds significantly faster than its reactions with HNO_3 and HCl under normal atmospheric conditions.

Table S1. Deposition rate constant of NH_3 (D_3) and NH_4^+ (D_4) (cm s^{-1}) assessed by previous studies.

Table S2. Generation rate constant of NH_4^+ (G_4, h^{-1}) reported by previous studies.

Figure S1. Sensitivity coefficients of the molar fraction of NH_4^+ to NH_x in the atmosphere to the $\delta^{15}N_{4a\text{-}3s}$ values at 20ºC obtained from six simulation scenarios.

Figure S2. The comparison plots of the $\delta^{15}N_{4a\text{-}3s}$ values calculated by the six fitted equations (FE) against the six model scenarios (MS) simulated by the developed model.

Figure S3. Scatter plots of the $\delta^{15}N_{4a\text{-}3s}$ values calculated by the six fitted equations (FE) against the six model scenarios (MS) simulated by the developed model.

Text S1

The chemical reaction between ammonia gas and acidic aerosols (sulfuric acid (H_2SO_4), nitric acid (HNO_3) and hydrochloric acid (HCl)) (referred to as the gas-to-particle phase transformation process) has been extensively studied. These gas-to-particle phase transformation processes produce ammonium salts (NH_4^+), nitrate (NO_3^-), sulfate (SO_4^{2-}), and chloride (Cl^-), which constitute the main components of secondary $PM_{2.5}$ (Sharma et al., 2007). The formation of these salts depends on the concentrations of ammonia gas and acidic precursor gases (nitrogen dioxide (NO_2) and sulfur dioxide (SO_2)), temperature, relative humidity, solar radiation, and the mixing height of the boundary layer (Behera and Sharma, 2011). Among these influencing factors, gas phase constituent concentration is an important one. Overall, it can be summarized that under normal atmospheric conditions, the reaction between NH_3 and H_2SO_4 is preferred over reactions of NH_3 with HNO_3 and NH_3 with HCl. As reported that the reaction rate constants of NH_3 and H_2SO_4 (k_S) showed relatively greater than k_N (NH_3 with HNO_3) and k_{Cl} (NH_3 with HCl). For instance, an environmental chamber study (Behera and Sharma, 2012) showed that the rate constants of k_S, k_N, and k_{Cl} were estimated as 2.68 (±1.38) × 10^{-4} m^3/μmol/s, 1.59 (±0.897) m^3/μmol/s, and 5.16 (±3.50) × 10^{-5} m^3/μmol/s. A field study (Baek et al., 2004) showed that k_S = 1.14 (±1.25) ×10^{-4} m^3/mmol/s, k_N = 0.73 (±1.49) ×10^{-4} m^3/mmol/s, and k_{Cl} = 0.86 (±1.44) ×10^{-4} m^3/mmol/s, respectively.

Table S1 lists deposition rate constant of NH_3 (D_3) and NH_4^+ (D_4) assessed by previous studies. The average values of the deposition rate constants of D_3 and D_4 reported in the literature were 1.14 ± 0.88 cm s^{-1} and 0.52 ± 0.46 cm s^{-1}, respectively. This indicated that the ratio of NH_3 to NH_4^+ deposition rates was 0.46 ± 0.048. A model study recommended that the deposition rates of NH_3 and NH_4^+ were 7.4 and 2.4 mm s^{-1} (Zhang et al., 2011; Shen et al., 2009). It suggested that the ratio of NH_3 to NH_4^+ deposition rates was 0.32. The average of the two estimates was 0.39. Based on a standard deviation of 0.048, the 95% confidence interval for this ratio was 0.29-0.48. Based on the above statistical analysis, we chose the ratio of D_4 to D_3 to be within the range of 0.2 to 0.5 for further scenario simulation.

As shown in Table S1, the averaged deposition rate constant of NH_3 is 1.14 ± 0.88 cm s^{-1}. Assuming that the deposition heights were 100m and 200m, the deposition rate could be 0.89 h^{-1} and 1.59 h^{-1}. Table S2 lists the generation rate constant of NH_4^+ (G_4) reported by previous studies. The average of G_4 was 0.90 h^{-1}, which was comparable to D_3 when the deposition height was assumed to be 100 m. These previous studies also found that the formation rate of ammonium salt was closely related to the altitude of the atmosphere. On the basis of the statistical analysis, we chose the ratio of G_4 to D_3 to be within the range of 0.5 to 2.0 for further scenario simulation.

Table S1. Deposition rate constant of NH_3 (D3) and NH_4^+ (D4) (cm s^{-1}) assessed by previous studies.

NH_3 (D3)	Receptor	Reference
0.3	Soil	(Krupa, 2003)
0.5	Soil, exposure chamber	(Krupa, 2003)
0.5–0.06	Soil, field conditions	(Krupa, 2003)
1	Soil, maximum	(Krupa, 2003)
0.3–1.3	Different species, during the day	(Krupa, 2003)
0.03–0.13	Different species, at night	(Krupa, 2003)

0.1–0.4	Bogs	(Krupa, 2003)
0.4	Agricultural	(Schrader and Brummer, 2014)
1.9	Heathlands/bogs	(Krupa, 2003)
1.6	Grass (*Lolium multiflorum*)	(Krupa, 2003)
2.2	Forest	(Krupa, 2003)
2.7±0.7	Douglas fir forest	(Krupa, 2003)
3.2	Douglas fir forest	(Krupa, 2003)
3.6	Coniferous forest	(Krupa, 2003)
0.5–5	Exterior surfaces and interior of leaves	(Krupa, 2003)
2.1	Coniferous forest	(Schrader and Brummer, 2014)
1.2	Mixed forest	(Schrader and Brummer, 2014)
0.9	Deciduous forest	(Schrader and Brummer, 2014)
1.5	Mixed forest	(Schrader and Brummer, 2014)
2.2	Coniferous forest	(Schrader and Brummer, 2014)
0.87	Urban	(Schrader and Brummer, 2014)
0.07	In residential areas,	(Yang et al., 2010)
0.74	The North China Plain	(Shen et al., 2009)
0.7	Semi-natural	(Schrader and Brummer, 2014)
0.23	In the field	(Yang et al., 2010)
0.626 ± 0.0976	Sea	(Zhang et al., 2010)
0.7	Water	(Schrader and Brummer, 2014)
0.6	Water	(Schrader and Brummer, 2014)
0.81 ±0.19	Northwestern Pacific	(Zhang et al., 2003)
0.8–2.0	Atlantic Basin	(Renard et al., 2004)
0.83	Australia-Southern Ocean	(Renard et al., 2004)
0.83	NE Pacific	(Renard et al., 2004)
0.88	North and Baltic Seas	(Renard et al., 2004)
0.76	North Sea	(Renard et al., 2004)
0.7	Tampa Bay	(Renard et al., 2004)
0.2–1.5	North Sea	(Renard et al., 2004)
1.14 ± 0.88	Mean	
NH_4^+ (D4)		
1.0	Beech canopy	(Krupa, 2003)
0.0032	Bean	(Krupa, 2003)
0.7–1.3	Beech canopy (throughfall)	(Krupa, 2003)
0.7–2.1	Spruce canopy (throughfall)	(Krupa, 2003)

0.5–1.5	Spruce/beech forest (throughfall)	(Krupa, 2003)
0.44–0.60	*Ceanothus crassifolius* leaves/canopy	(Krupa, 2003)
0.2	Heathlands/bogs	(Krupa, 2003)
0.28±0.05	the Yellow Sea coastal region	(Qi et al., 2013)
0.21	over the Yellow Sea	(Shi et al., 2013)
0.1	Huaniao Island	(Zhu et al., 2013)
0.06 ±0.01	Northwestern Pacifica	(Zhang et al., 2003)
0.52 ± 0.46	Mean	

Table S2. Generation rate constant of NH4+ (G4, h-1) reported by previous studies.

	G4	Reference
Chamber study	0.35±0.21	(Behera and Sharma, 2011)
The Netherlands (< 200 m)	3.6	(Vemetten et al., 1985)
Daytime from vertical concentration distribution	0.36	(Erisman et al., 1988)
Nighttime from vertical concentration distribution	0.18	(Erisman et al., 1988)
A rural site in eastern England	0.014-0.14	(Harrison and Kitto, 1992)
Horizontal concentration distribution	0.01-1.48	(Harrison and Kitto, 1992)
Low layer atmosphere (< 100 m)	0.04-3.60	(Baek et al., 2004)
Atmosphere at 400 m heights	0.08	(Lenhard and Gravenhorst, 1980)
Mean	0.90	

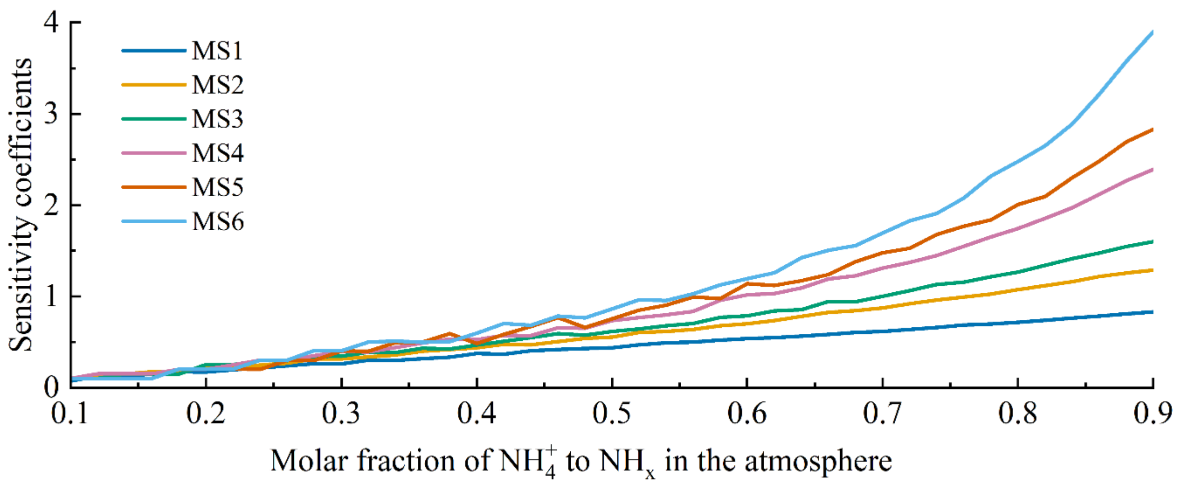

Figure S1. Sensitivity coefficients of the molar fraction of NH_4^+ to NH_x in the atmosphere to the $\delta^{15}N_{4a\text{-}3s}$ values at 20°C obtained from six simulation scenarios.

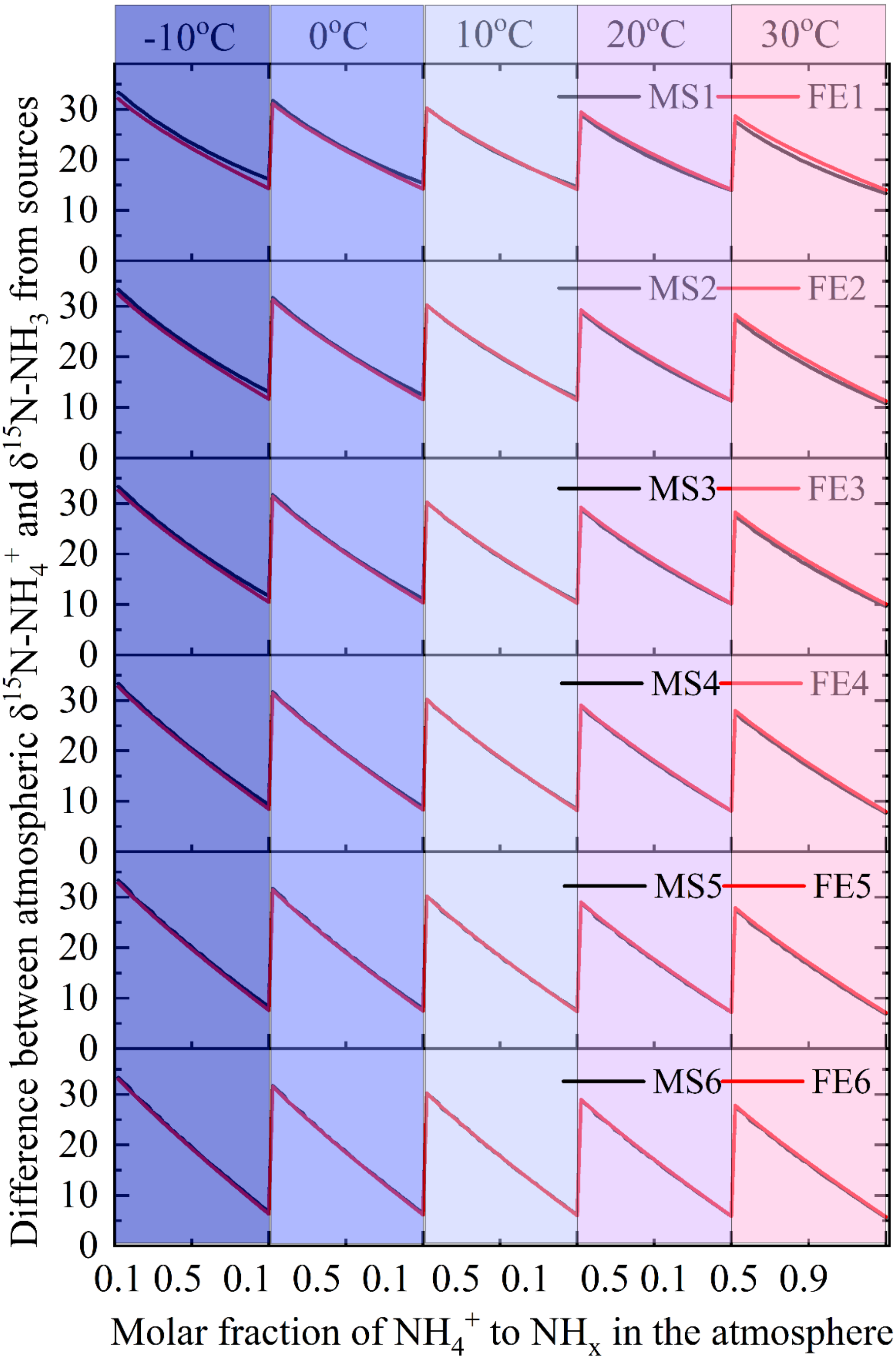

Figure S2. The comparison plots of the $\delta^{15}N_{4a\text{-}3s}$ values calculated by the six fitted equations (FE) against the six model scenarios (MS) simulated by the developed model.

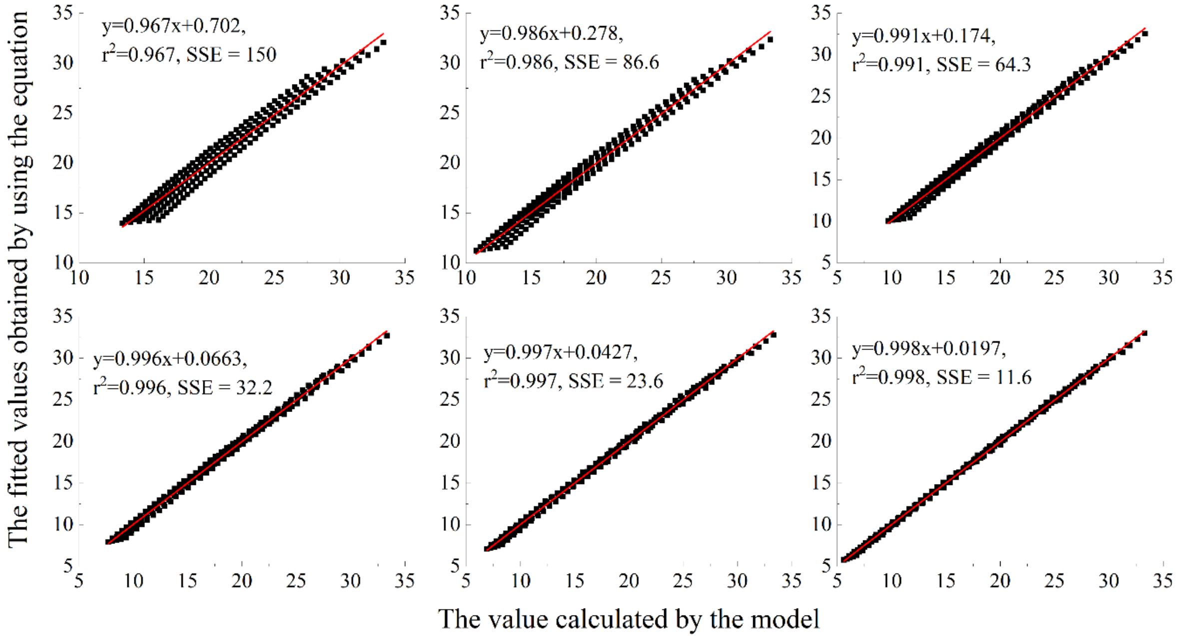

Figure S3. Scatter plots of the $\delta^{15}N_{4a\text{-}3s}$ values calculated by the six fitted equations (FE) against the six model scenarios (MS) simulated by the developed model.

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