Supporting Information for

Isotopic Constraints on the Atmospheric Sources and Formation of Nitrogenous Species in Biomass-Burning-Influenced Clouds

Yunhua Chang[1], Yanlin Zhang[1*], Jiarong Li[2], Chongguo Tian[3], Linlin Song[4], Xiaoyao Zhai[1], Wenqi Zhang[1], Tong Huang[1], Yu-Chi Lin[1], Chao Zhu[2], Yunting Fang[4], Moritz F. Lehmann[5], and Jianmin Chen[2*]

[1]Yale-NUIST Center on Atmospheric Environment, Nanjing University of Information Science & Technology, Nanjing 210044, China.

[2]Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention (LAP[3]), Department of Environmental Science & Engineering, Institute of Atmospheric Sciences, Fudan University, Shanghai 200433, China.

[3]Key Laboratory of Coastal Environmental Processes and Ecological Remediation, Yantai Institute of Coastal Zone Research, Chinese Academy of Sciences, Yantai 264003, China.

[4]CAS Key Laboratory of Forest Ecology and Management, Institute of Applied Ecology, Chinese Academy of Sciences, Shenyang 110016, China.

[5]Aquatic and Isotope Biogeochemistry, Department of Environmental Sciences, University of Basel, Basel 4056, Switzerland.

[*]Corresponding authors: Yanlin Zhang (dryanlinzhang@outlook.com) and Jianmin Chen (jmchen@fudan.edu.cn).

Contents of this file

Text S1.

The Bayesian mixing model makes use of stable isotope data to determine the probability distribution of source contributions to a mixture, explicitly accounting for uncertainties associated with multiple sources, their isotopic signatures, and isotope fractionation during transformations. The model has been widely used in ecological studies, such as food-web analyses. In Bayesian theorem, the contribution of each source is calculated based on mixed data and prior information, such that:

$$P(f_q|data) = \theta(data \mid f_q) \times p(f_q) / \sum\theta(data \mid f_q) \times p(f_q)$$

where $\theta(data|f_q)$ and $p(f_q)$ refer to the likelihood of the given mixed isotope signature, and the pre-determined probability of the given state of nature, based on prior information, respectively. The denominator represents the numerical approximation of the marginal probability of the data. In a Bayesian model (stable isotope in R; SIAR), isotope signatures from the mixed data pool are assumed to be normally distributed. Uncertainty in the distribution of isotope sources and associated isotope fractionation during transformations are factored into the model by defining respective mean (μ) and standard deviation (σ) parameters. Prior knowledge about proportional source contributions (f_q) is parametrized using the Dirichlet distribution, with an interval of [0, 1]. To assess the likelihood of the given f_q, the proposed proportional contribution is combined with a user-specified isotope distribution of sources and their associated isotope effects to develop a proposed isotope distribution for the mixture. The probability of fractional source contributions (f_q) is calculated by the Hilborn sampling-importance-resampling method.

.

Table S1. Typical δ^{15}N-NO_x values for coal combustion, transportation and biomass burning, and soils based on literature values.

Source types	Mean (‰)	Standard(‰)	Number	Reference
Coal combustion	13.72	4.57	47	Felix et al., 2012, 2015; Walter et al., 2015
Transportation	-7.25	7.80	151	Walter et al., 2015a, b; Heaton et al., 1997
Biomass burning	1.04	4.13	24	Fibiger et al., 2016; Felix and Elliott, 2013
Biogenic soil	-33.77	12.16	6	Hastings et al., 2009; Felix et al., 2012

Felix, J. D.; Elliott, E. M.; Avery, G. B.; Kieber, R. J.; Mead, R. N.; Willey, J. D.; Mullaugh, K. M., Isotopic composition of nitrate in sequential Hurricane Irene precipitation samples: Implications for changing NO_x sources. *Atmos. Environ.* **2015,** *106*, 191-195.

Felix, J. D.; Elliott, E. M.; Shaw, S. L., Nitrogen isotopic composition of coal-fired power plant NO_x: influence of emission controls and implications for global emission inventories. *Environ. Sci. Technol.* **2012,** *46*, (6), 3528-3535.

Felix, J. D.; Elliott, E. M., Isotopic composition of passively collected nitrogen dioxide emissions: Vehicle, soil and livestock source signatures. *Atmos. Environ.* **2014,** *92*, 359-366.

Felix, J. D.; Elliott, E. M., The agricultural history of human-nitrogen interactions as recorded in ice core δ^{15}N-NO_3^-. *Geophy. Res. Lett.* **2013,** *40*, (8), 1642-1646.

Fibiger, D. L.; Hastings, M. G., First measurements of the nitrogen isotopic composition of NO_x from biomass burning. *Environ. Sci. Technol.* **2016,** *50*, (21), 11569-11574.

Hastings, M. G.; Jarvis, J. C.; Steig, E. J., Anthropogenic impacts on nitrogen isotopes of ice-core nitrate. *Science* **2009,** *324*, (5932), 1288-1288.

Heaton, T. H. E.; Spiro, B.; Madeline, S.; Robertson, C., Potential canopy influences on the isotopic composition of nitrogen and sulphur in atmospheric deposition.

Oecologia **1997,** *109*, (4), 600-607.

Walters, W. W.; Tharp, B. D.; Fang, H.; Kozak, B. J.; Michalski, G., Nitrogen isotope composition of thermally produced NO_x from various fossil-fuel combustion sources. *Environ. Sci. Technol.* **2015,** *49*, (19), 11363-11371.

Walters, W. W.; Goodwin, S. R.; Michalski, G., Nitrogen stable isotope composition ($\delta^{15}N$) of vehicle-emitted NO_x. *Environ. Sci. Technol.* **2015,** *49*, (4), 2278-2285.

6.