Supplementary materials for

Three Northern Regions Shelter Forest contributed to long-term increasing trend of biogenic isoprene emission in Northern China

Xiaodong Zhang[1], Tao Huang[1*], Leiming Zhang[2], Yanjie Shen[1], Yuan Zhao[1], Hong Gao[1], Xiaoxuan Mao[1], Chenhui Jia[1], Jianmin Ma[1,3]*

Affiliation:

[1]Key Laboratory for Environmental Pollution Prediction and Control, Gansu Province College of Earth and Environmental Sciences, Lanzhou University, Lanzhou 730000, P. R. China

[2]Air Quality Research Division, Environment Canada, Toronto, Ontario, M3H 5T4, Canada

[3]CAS Center for Excellence in Tibetan Plateau Earth Sciences, Beijing 100101, China

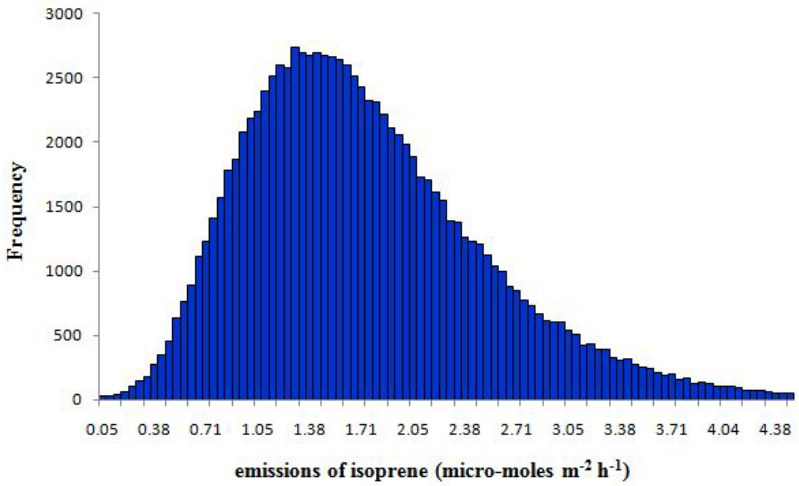

Fig. S1. Frequency distribution of biogenic emission fluxes of isoprene.

Table S1. Rate constants (cm^3 $molecules^{-1}$ s^{-1}) and mean concentrations of OH (molecules cm^{-3}) and O_3 ($\mu g\ m^{-3}$).

OH concentration [1]	O_3 concentration [2]	Rate const with OH [3]	Rate const for O_3 [4]
9.7×10^5	93	10.1×10^{-11}	5.8×10^{-18}

1.Prinn et al., 1995; 2. Annual mean O_3 concentration in Beijing-Tianjin-Hebei; 3. Zhang et al., 4. Li et al., (2013)

Table S2. Coefficients of variation (*CV*, %) of input parameters for the uncertainty analysis (Guenther et al., 1999) in computation of biogenic isoprene emission fluxes using MEGAN2.1 (Guenther et al., 2012), the model run times: 100,000).

Parameter	Light	LAI	E_{opt}	C_{T1}	C_{T2}	Emi_fac
CV （%）	10	30	10	10	10	30

LAI: leaf area index,

E_{opt}: maximum normalized emission capacity

CT1: empirical coefficient for the energy of activation

CT2: empirical coefficient for the energy of deactivation

Emi_fac: emission factor (Guenther et al., 2012)

Table S3. Coefficients of variation (*CV*, %) of input parameters for the uncertainty analysis for isoprene emission fluxes converted from the box model (Eq. 1) in main text (Guenther et al., 1996), the model run times: 100,000.

Parameter	Z_i	C	C_{OH}	C_{O3}
Cv (%)	15	25	30	30

Z_i is the height of the mixed-layer capping inversion, C is measured concentration of isoprene, C_{OH} is OH concentration, and C_{O3} is ozone concentration.

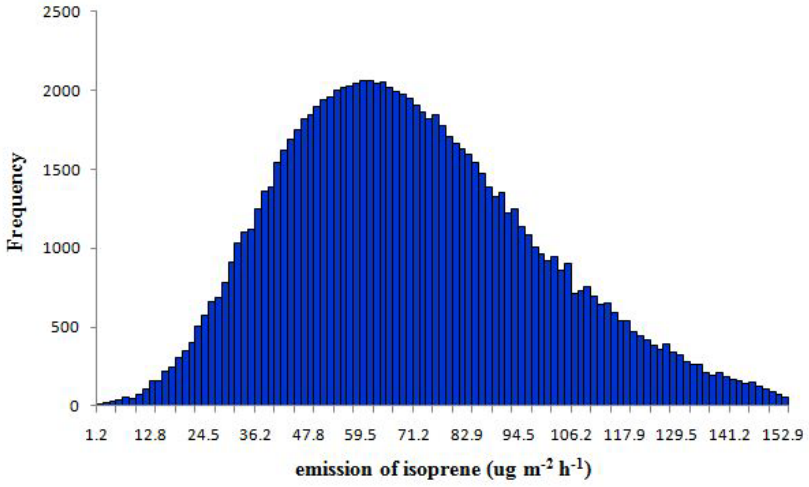

Fig. S2. Frequency distribution of isoprene emission fluxes using the box model (Eq. 1) in main text.

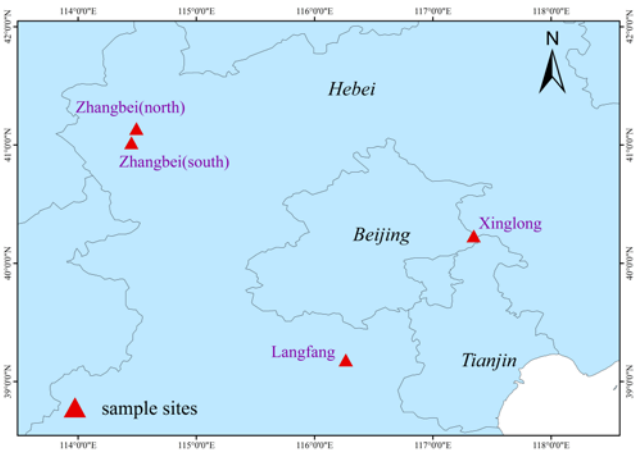

Fig. S3. Sampling sites (red triangle) in the field campaign. Two megacities, Beijing and Tianjin, are also illustrated.

Table S4. Vegetation types and latitude/longitude of sampling sites inside and outside of forest. The forest sites are characterized by poplar and Pinus sylvestris var and the sites outside of forest are characterized by corn and grass.

	Langfang		Zhangbei (south)		Zhangbei (north)		Xinglong	
Vegetation	Poplar	corn	Pinus sylvestris var	grass	Poplar	grass	poplar	corn
Latitude	39.307	39.304	41.034	41.037	41．233	41.235	40.38	40.385
Longitude	116.441	116.437	114.750	114.751	114.819	114.821	17.565	117.557

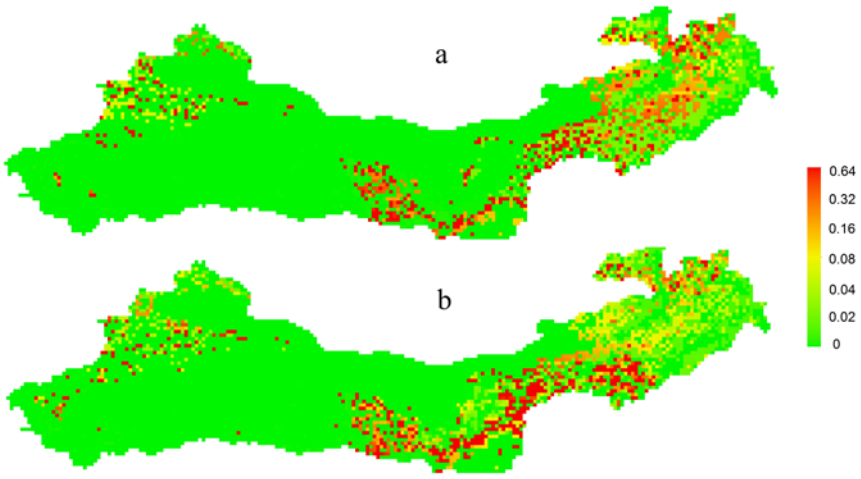

Fig. S4. MEGAN2.1 (Guenther et al., 2012) simulated biogenic isoprene emission fluxes (micro-moles m^{-2} hr^{-1}) in 1982 (a) and 2010 (b) across the TNRSF.

Simplified Gaussian model for an area source

The simplified Gaussian model for an area source, which is obtained from an analytical solution of the atmospheric dispersion equation (Arya, 1999), can be defined as

$$C_{is} = \frac{E_{is}\Delta l}{uh},$$

where E_{is} is the biogenic isoprene emission from area sources, u is wind speed (m s^{-1}), h is the mean canopy height (6 m), Δl is the distance (fetch) upwind to a sampler which alighs with wind direction. Based on the height to fetch ratios in the field sampling of atmospheric wind and scalars proposed by Oak (2004), Δl was taken as 3 km in the present study. The model virtually provides "initial" concentrations derived from the emissions of a chemical species which has been often used in a chemical transport model (Arya, 1999). However, it does not take into consideration of any photochemical process which is important for reactive species such as isoprene. Taking the measure wind speed and TVOCs levels as the area source model input, the mean canopy height of 6 m and the fetch (3 km) upwind to sampling sites, and assuming the isoprene emission as 50% of the TVOCs, we obtained the emission fluxes of 72, 115, 143, and 101 μg m^{-2} h^{-1} at the Langfang, Xinglong, Zhangbei (North), and Zhangbei (South) sites, respectively. Overall these values are higher than the MEGAN2.1 modeled emission fluxes of 36, 41.5, 49, and 47.6 μg m^{-2} h^{-1} but within factors of differences, at the same sites.

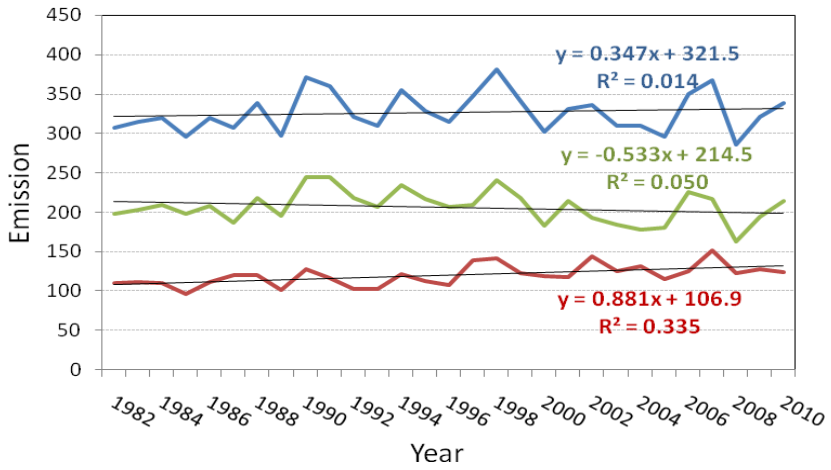

Fig. S5. Annual total biogenic isoprene emission (micro-moles $m^{-2}h^{-1}$) from 1982 to 2010. Blue solid line stands for the emissions in Northern China with the TNRSF included, green soild line represents the emissions in Northern China without the TNRSF included, and red solid line indicates the emission in the TNRSF only. Black solid line stands for their respective linear trend.

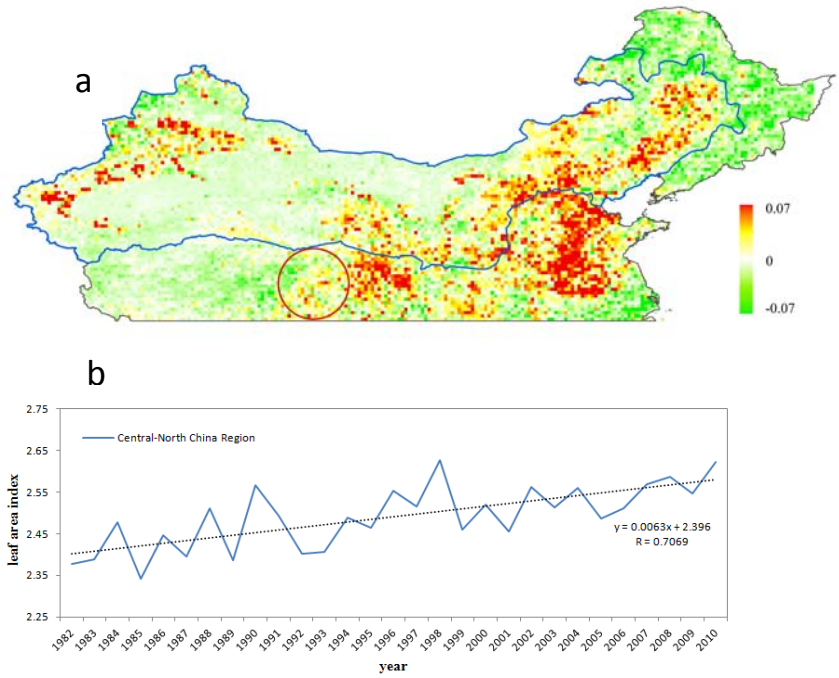

Fig. S6. a) Linear trend of leaf area index (LAI) from 1982 to 2010 over Northern China. Red circle indicates the region extending from Qinghai to Qinling–Ta-Pa Mountains where the higher isoprene emission fluxes in 2000 were found (Fig. 4 in main text); b) Annual mean LAT averaged over Central-North China region of the TNRSF.

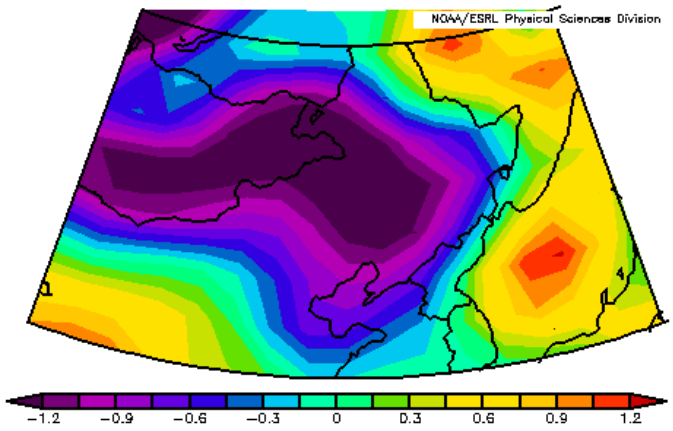

Fig. S7. Differences of annual surface air temperature (SAT) between 1982 and 2010 ($T_{2010} - T_{1982}$)**.** The figure was created from the NCEP reanalysis data and graphic system at http://www.esrl.noaa.gov/psd/cgi-bin/data/composites/printpage.pl.

References:

Arya, S.P.: A*ir Pollution Meteorology and Dispersion*. Oxford University Press, Inc.; New York, New York, 1999.

Guenther, A， Zimmerman, P., Klinger, L., Greenberg, J., Ennis, C., Davis, K., and ait Pollock, W.: Estimates of regional natural volatile organic compound fluxes from

enclosure and ambient measurements, J. Geophys. Res.-Atmos., 101(D1), 1345-1359, 1996.

Guenther, A., Baugh, B., Brasseur, G., Greenberg, J., Harley, P., Klinger, L., Serca, D., and Vierling, L.: Isoprene emission estimates and uncertainties for the Central African EXPRESSO study domain, J. Geophys. Res.-Atmos., 104(D23), 30625–30639, 1999.

Guenther AB, Jiang X, Heald CL et al.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emission, Geosci. Model Dev., **5**, 1471-1492, 2012.

Li, X., Zhao, W., Li, J., Jiang, J., Chen, J,, Chen, J.: Development of a model for predicting reaction rate constants of organic chemicals with ozone at different temperatures, Chemosphere, 92, 1029-1034, 2013.

Oke, T. R. 2004. Sitting and exposure of meteorological instruments at urban sites. in 27th NATO/CCMS International Technical Meeting on Air Pollution Modelling and its Application. Banff, Alberta.

Prinn, R. G., Weiss, R. F., Miller, B. R., Huang, J., Alyea, F. N., Cunnold, D.M., Fraser, P. J., Hartley, D. E., and Simmonds, P.: Atmospheric trends and lifetime of CH_3CCl_3 and global OH concentrations, Science, 269, 187–192, 1995.

Zhang, R., Suh, I., and Lei, W.: Kinetic studies of OH-initiated reactions of isoprene, J. Geophys. Res.-Atmos. 105(D20), 24627-24635, 2000.