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Rapid SO_2 emission reductions significantly increase tropospheric ammonia concentrations over the North China Plain

Mingxu Liu et al.

Correspondence to: Yu Song (songyu@pku.edu.cn), Min Hu (minhu@pku.edu.cn) and Tong Zhu (tzhu@pku.edu.cn)

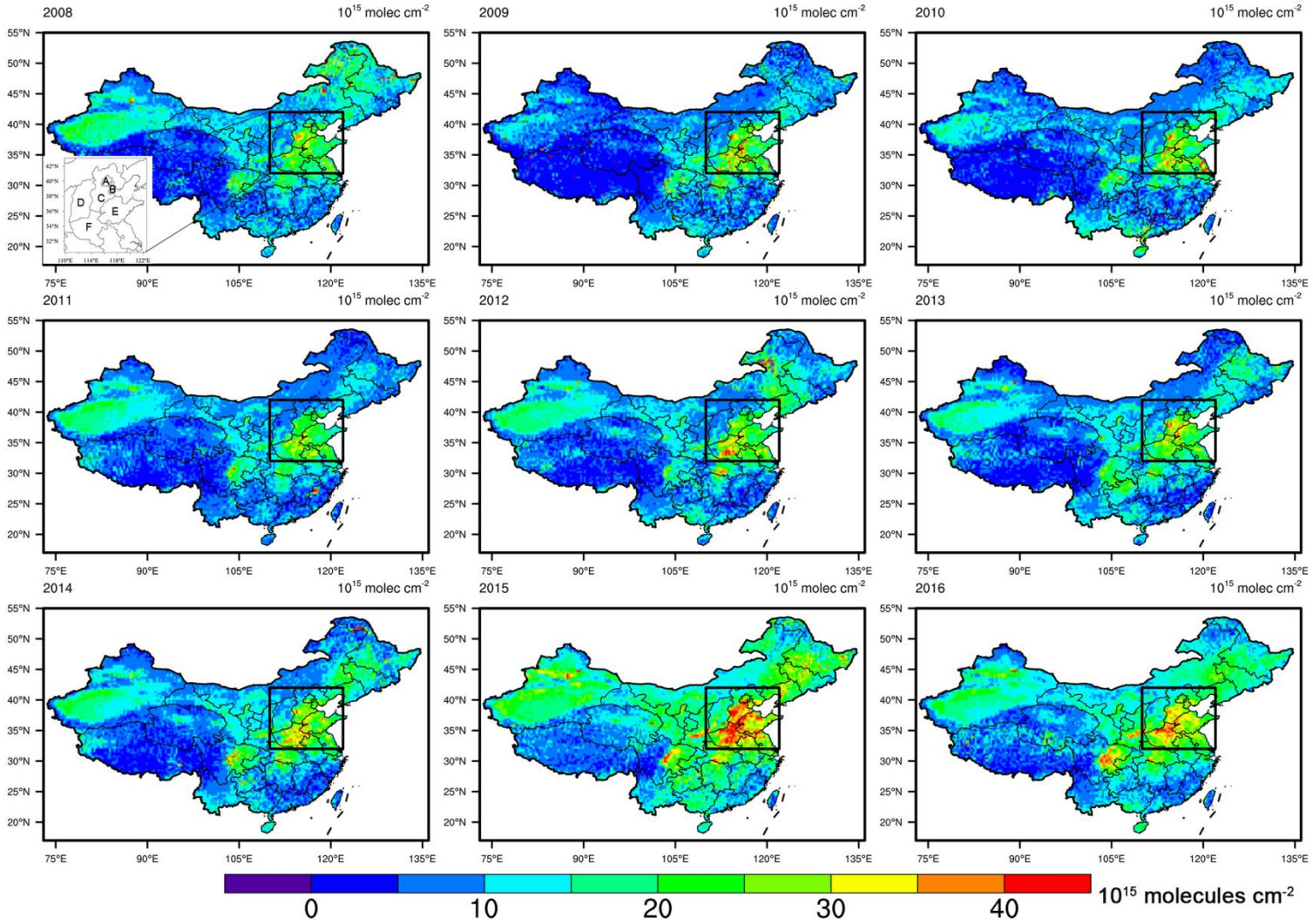

Figure S1. Spatial distribution of NH_3 column densities in mainland China observed by IASI sensor from 2008 to 2016. The area of interest in this study includes Beijing (A), Tianjin (B), Hebei (C), Shanxi (D), Shandong (E) and Henan (F) Provinces, as marked in the black rectangle. The small islands in South China Sea are not included in the figure.

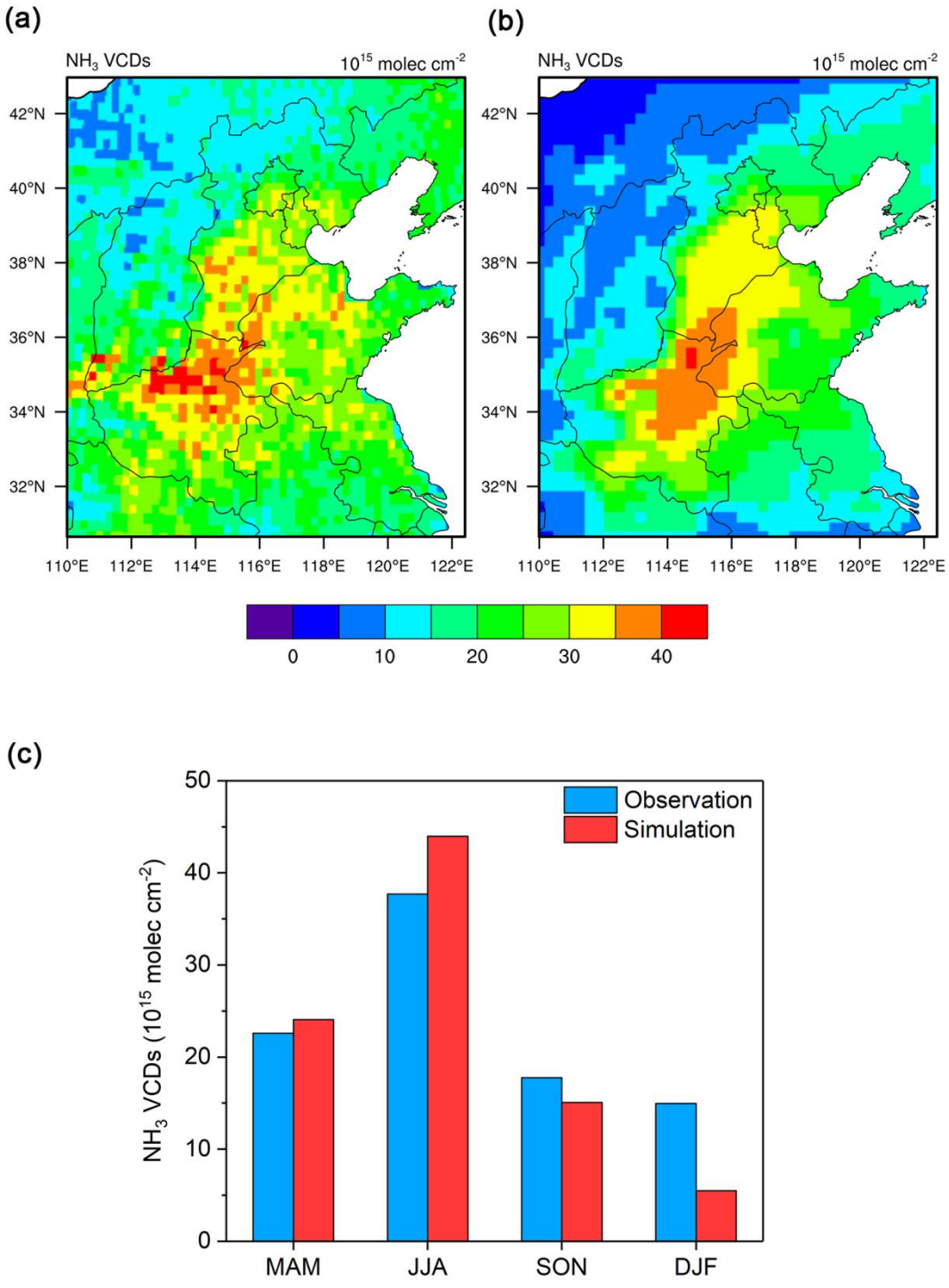

Figure S2. Spatial distribution of satellite observations (a) and modelled tropospheric NH_3 column densities (b) for the year of 2016; (c) comparison of the regional mean NH_3 column densities between observations and simulations among different seasons in 2016.

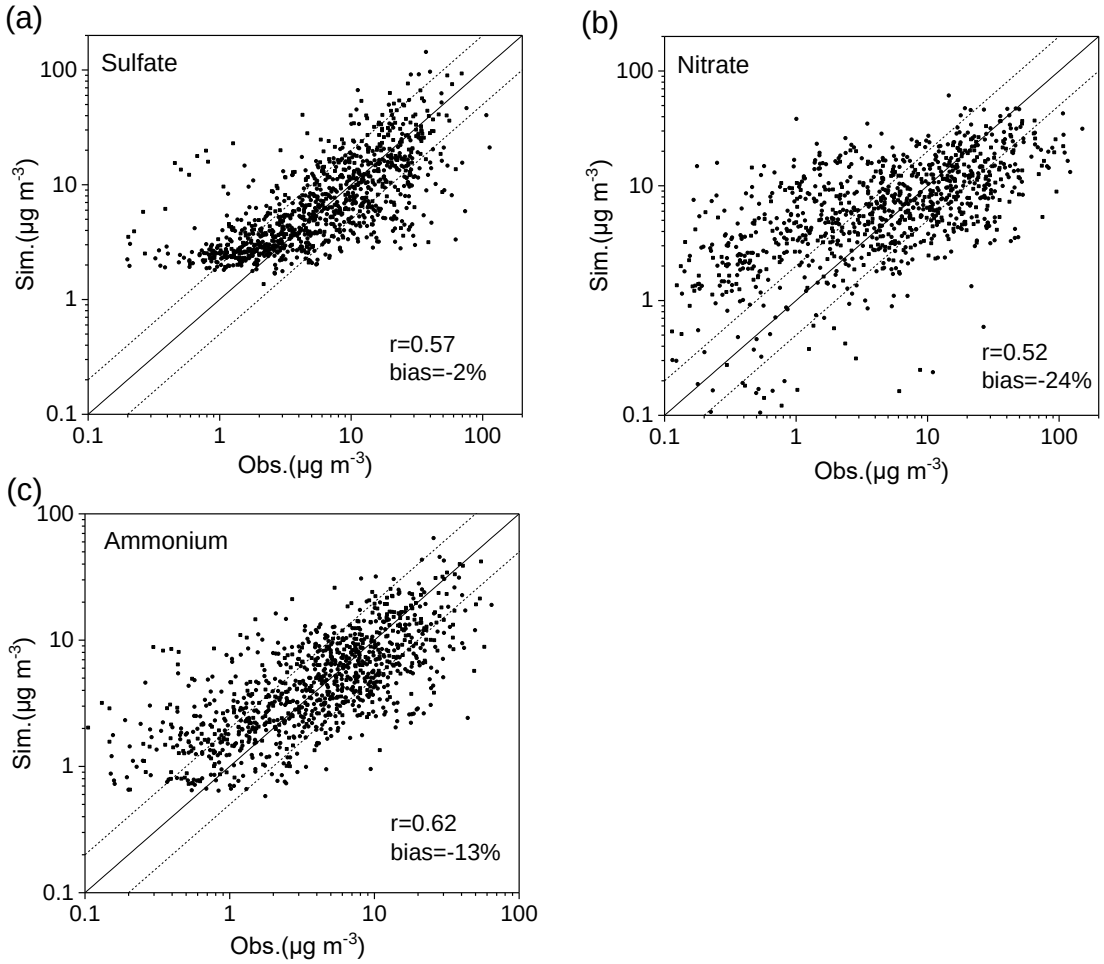

Figure S3. Comparison of modelled $PM_{2.5}$ sulfate (a), nitrate (b) and ammonium (c) concentrations with corresponding daily measurements of $PM_{2.5}$ from 2014 to 2016 over North China Plain. The model results were extracted from the first model level (~25m aboveground) and averaged at one day. The Pearson correlation coefficients and normalized mean bias are shown inset.

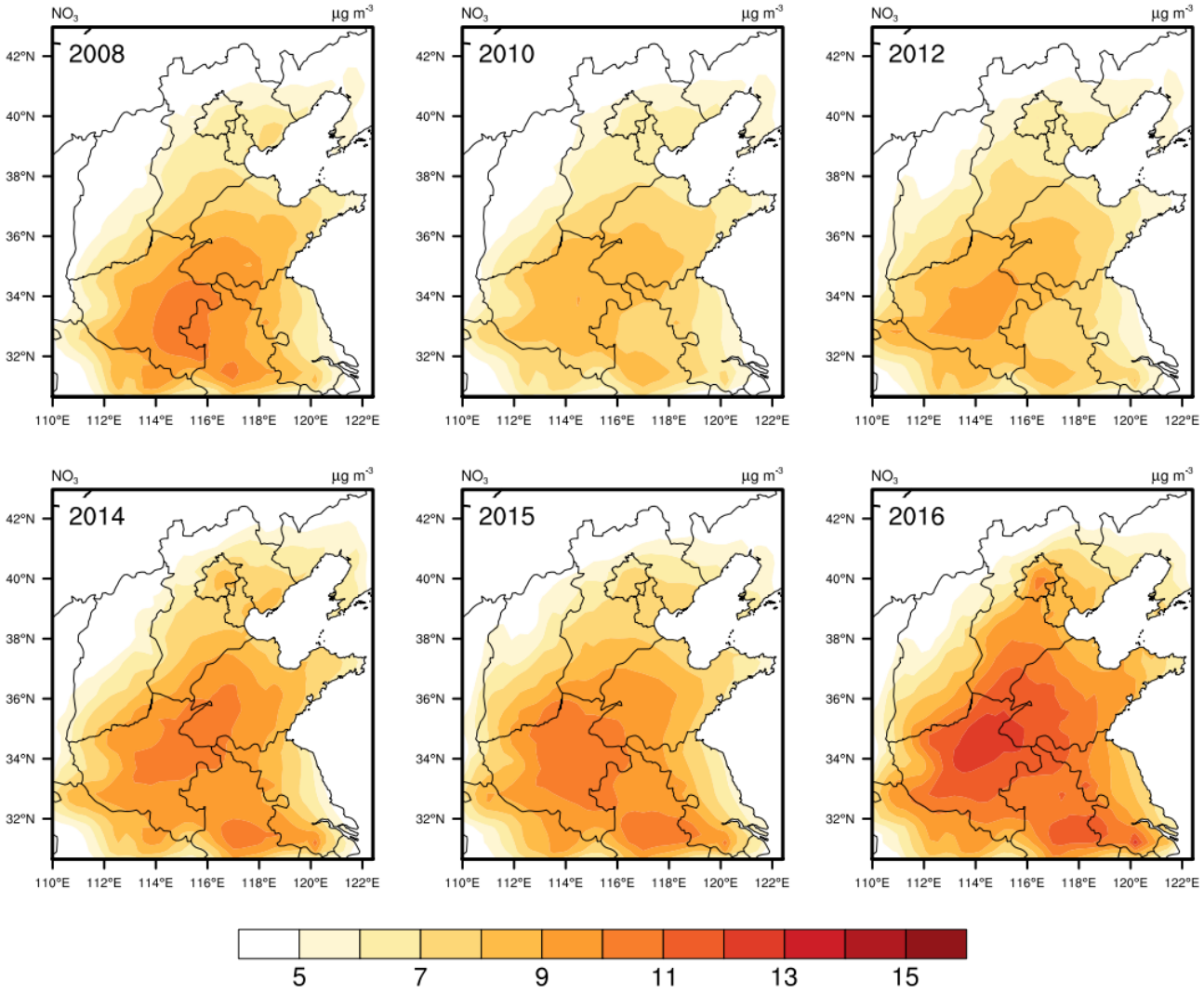

Figure S4. The simulated annual-mean concentrations of $PM_{2.5}$-nitrate (μg m^{-3}) over Northern China in recent years.

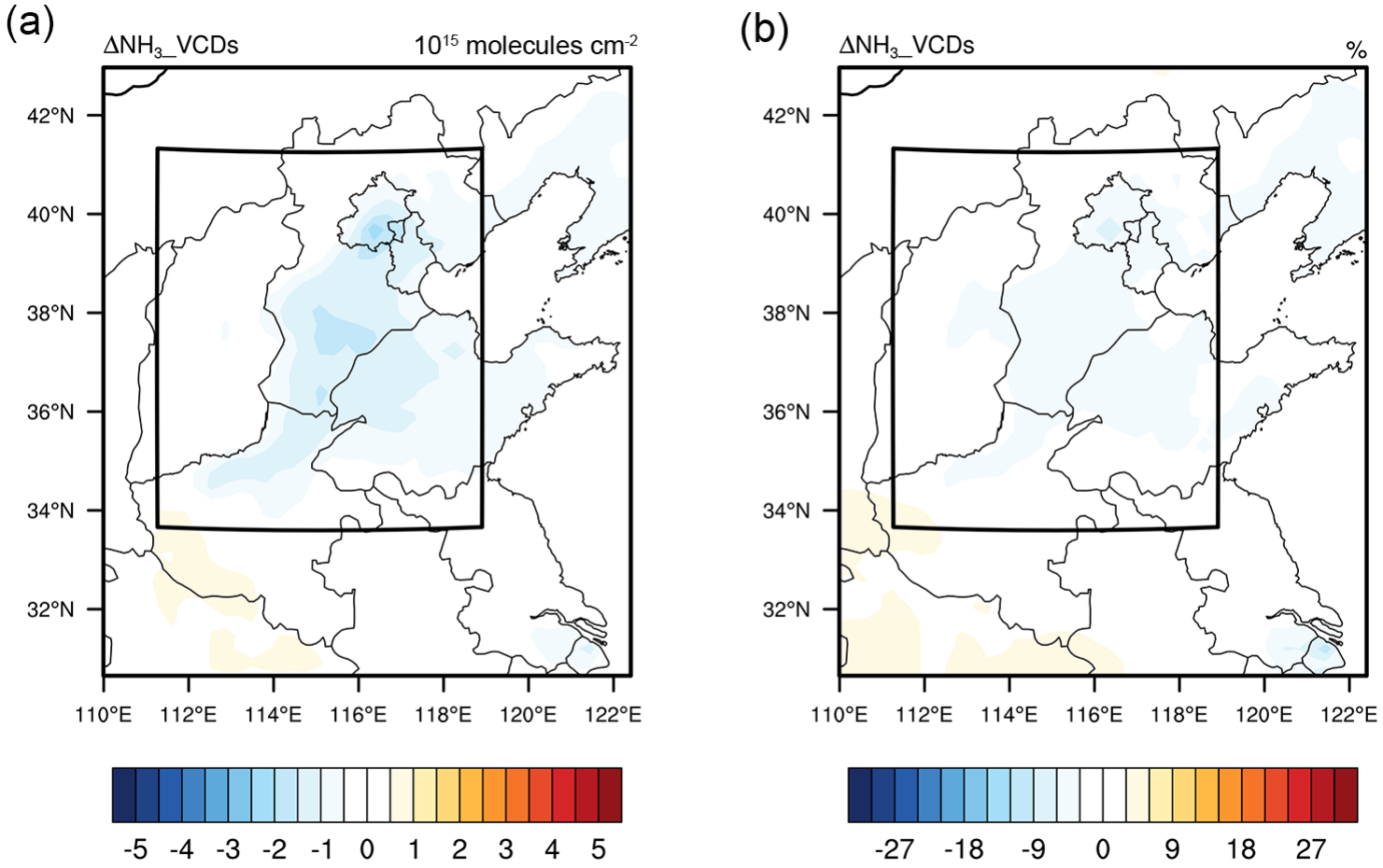

Figure S5. Absolute (a) and percent (b) changes in the simulated column concentrations of NH_3 between the Run_16_N08 (NO_x emissions in 2008) and Run_16 cases. Negative values denote decreases in NH_3 VCDs due to the change in NO_x emissions during 2008−2016. The black box represents the major area of interest in this study.

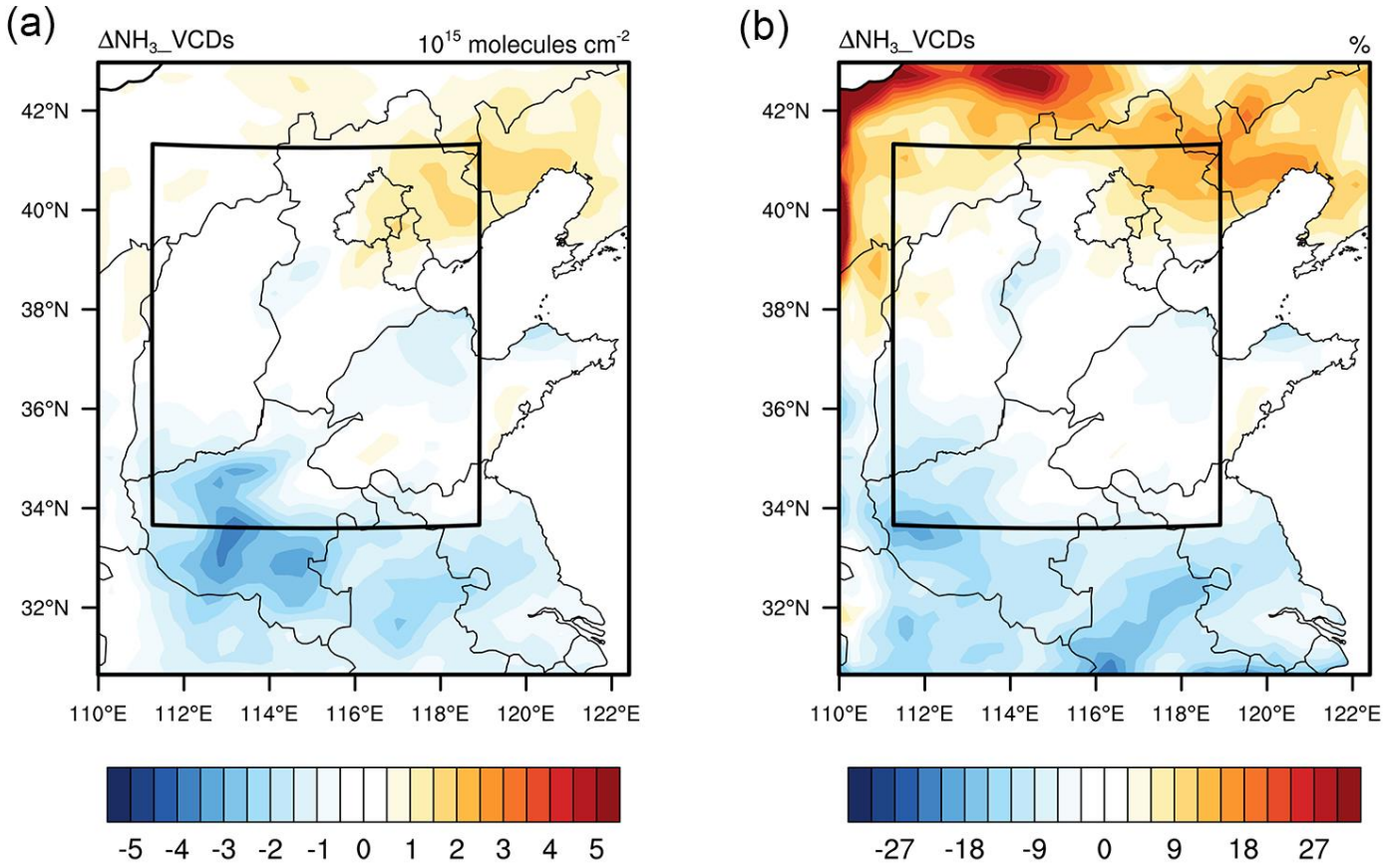

Figure S6. Absolute (a) and percent (b) changes in the simulated column concentrations of NH_3 between the Run_12 and Run_12_16M. Negative values denote decreases due to the change in meteorological fields in the Run_12_16M. The black box represents the major area of interest in this study.

Table S1 Annual consumption of synthetic fertilizers, populations of major livestock animals and the proportions of the intensive system in livestock animal rearing in Northern China from 2008 to 2016.

	2008	2009	2010	2011	2012	2013	2014	2015	2016
Fertilizers (Ton)[1]									
ammonium bicarbonate	146	128	99	108	107	99	61	54	42
Urea	477	489	520	506	507	509	538	532	523
Compound Fertilizer	120	127	135	140	145	149	160	163	168
Animals (Million)[2]									
Beef	18.2	17.4	16.1	15.1	14.7	14.6	14.6	15.0	14.6
Dairy	3.5	3.6	4.3	4.7	4.9	4.8	5.1	5.0	4.8
Goat	49.2	45.4	43.4	42.8	42.1	42.1	43.0	44.2	37.8
Sheep	17.3	19.7	19.3	20.6	21.3	22.1	23.2	23.1	26.9
Pig	132	139	143	142	151	157	164	160	155
Poultry	3056	3141	3209	3369	3658	3605	3424	3504	3635
Proportion of the intensive system in livestock animal rearing (%)[2]	28	32	35	37	39	41	43	46	46

[1] Values were derived from China Agriculture Yearbook (2008−2016) and Cost and Income of Chinese Farm Produce (2008−2016). [2] Values were derived from China Animal Industry Yearbook (2008−2016). The references for these data can be also found in Kang et al. (2016).

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