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Supplement of

Uncertainties in fertilizer-induced emissions of soil nitrogen oxide and the associated impacts on ground-level ozone and methane

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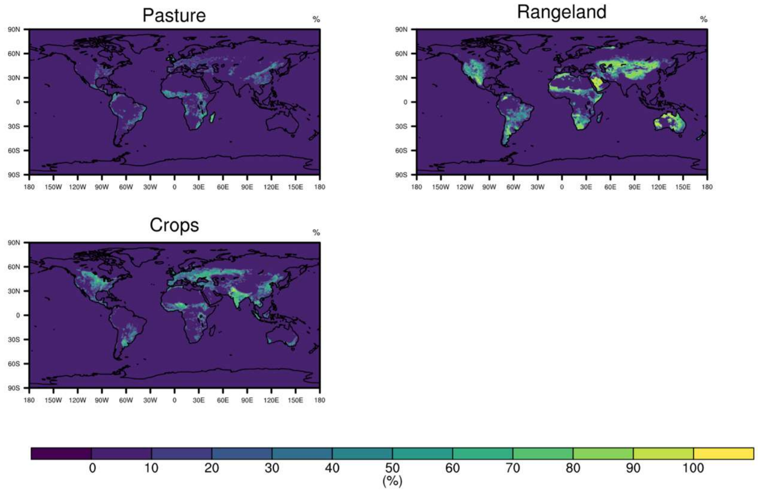

Figure S1. The global pattern of land-use categories used in this study to transform the unit of N fertilizer loadings from kg N per hectare grid area to kg N per hectare pasture, rangeland or crop. The map is from the Land-Use Harmonization (LUH2) (*https://luh.umd.edu/*)

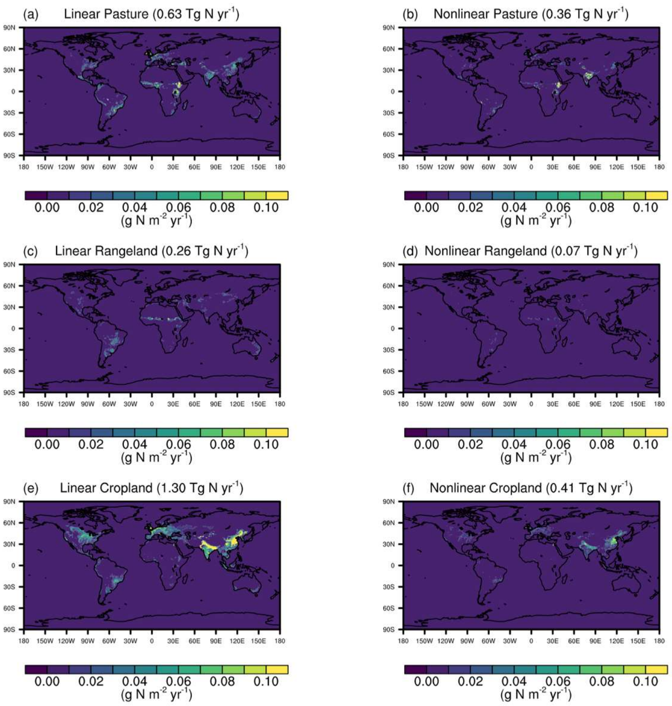

Figure S2. The spatial patterns of SNO_x-Fer in pasture, rangeland and crops estimated by linear EF and non-linear EF approaches in 2019. The global total budget of each estimate is given in the sub-titles.

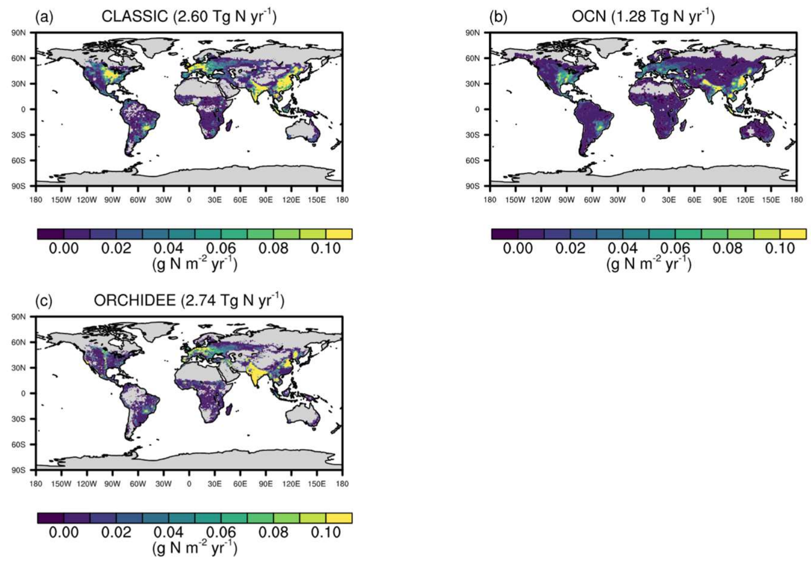

Figure S3. The global pattern of N fertilizer-induced soil NO_x emissions in 2019 simulated by three NMIP2 members. (a) the CLASSIC model; (b) the OCN model and (c) the ORCHIDEE model.

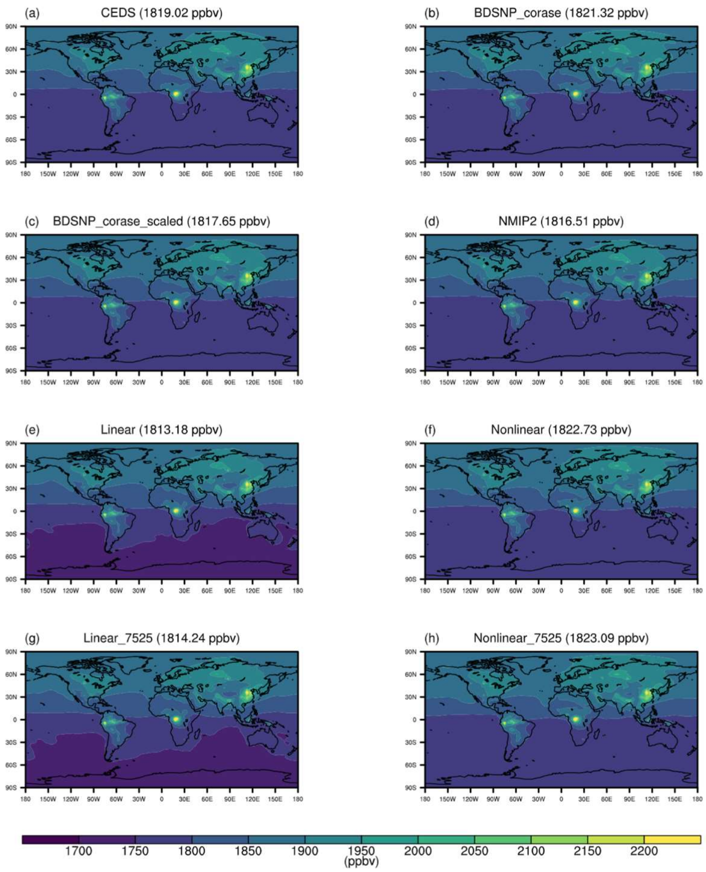

Figure S4. The simulated global surface CH_4 concentrations with different sensitivity experiments. The OH concentrations of each experiment are obtained from the corresponded GEOS-Chem sensitivity experiments with full chemistry mechanisms.

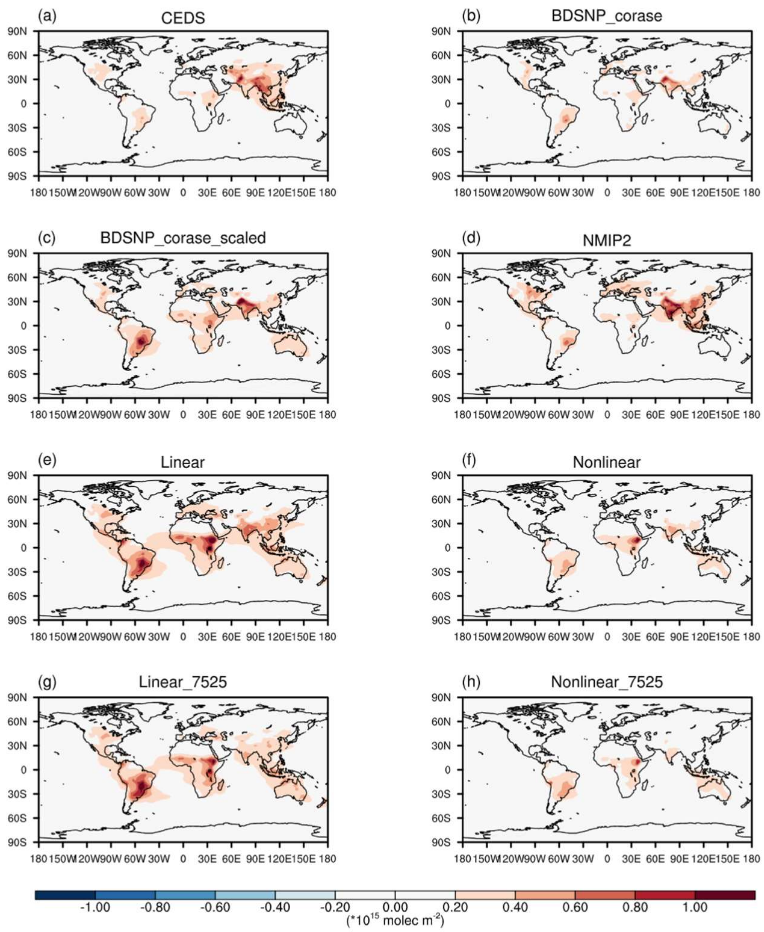

Figure S5. The annual-mean changes in tropospheric column OH concentrations simulated by different GEOS-Chem sensitivity experiments. (a)-(h) The differences of annual-mean tropospheric column OH between varied sensitivity experiments and the Zero experiments.

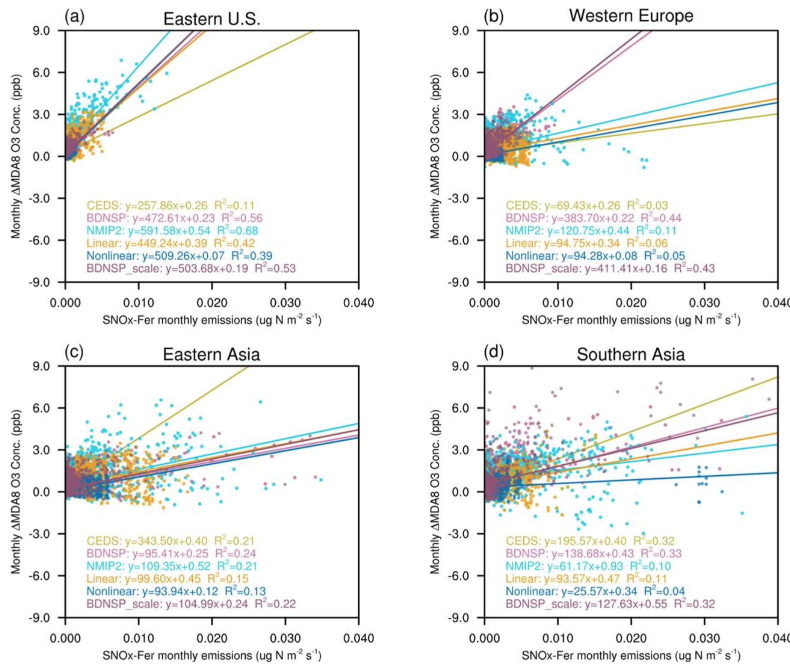

Figure S6. The sensitivity of changes in monthly MDA8 O_3 concentrations (ppbv) to the SNO_x-Fer changes (µg N m^{-2} s^{-1}) among different approaches in four agricultural hotspot regions. Each dot indicates the monthly SNO_x-Fer emissions and associated monthly MDA8 O_3 changes induced by SNO_x-Fer on a simulated grid. The different SNO_x-Fer estimating approaches are indicated by lines with different colors.

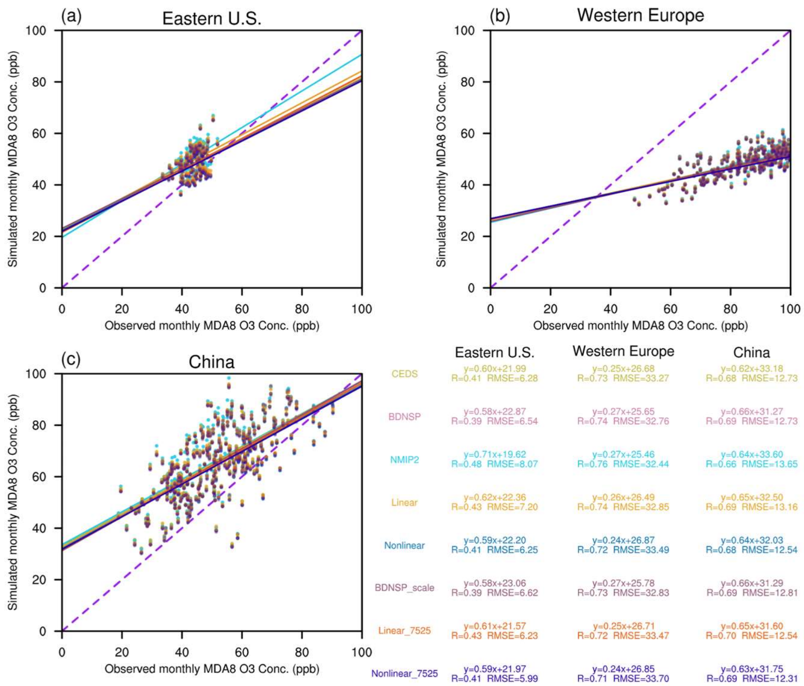

Figure S7. The comparison of monthly-averaged O_3 MDA8 concentrations between site-level observations and the GEOS-Chem simulations. The observational dataset in Eastern U.S., Western Europe and China are assessed via the Air Quality System (AQS, https://www.epa.gov/aqs), European Monitoring and Evaluation Programme (EMEP, https://ebas-data.nilu.no/Default.aspx) and China National Environmental Monitoring Centre (CNEMC, https://www.cnemc.cn/en/), respectively. Each dot indicates one simulated grid, where the observed O_3 concentrations are calculated by averaging all observational sites. The GEOS-Chem sensitivity experiments with different SNO_x-Fer estimating approaches are indicated by different colors.

Table S1. NMIP2 configuration and sensitivity experiments to isolate the contributions of synthetic fertilizer and manure to the soil NO_x emissions. '1850' indicates that the loading of synthetic fertilizer or manure is fixed at the level of 1850, while '1850-2019' indicates transient loadings based on the HaNi dataset. Other environmental forcings, e.g. the climate data, CO_2 concentration and land use change, are followed the historical transient data from 1850 to 2019 in each sensitivity experiment.

Experiment name	Synthetic fertilizer application	Manure application
SH1	1850-2019	1850-2019
SH2	1850	1850-2019
SH3	1850-2019	1850

Model members used in this study: CLASSIC, OCN and ORCHIDEE

Table S2. The sources and sinks of CH_4 (unit: Tg CH_4 yr^{-1}) applied in the GEOS-Chem model and the comparison to the IPCC AR6 budget (Canadell.J.G et al., 2021).

	This study	**IPCC AR6***
	Sources	
Fossil fuels	117.6	115 (114-116)
Agriculture and waste	238.9	208 (192-230)
Livestock	121.7	109 (106-115)
Rice cultivation	36.5	31 (25-37)
Waste water and landfill	80.7	64 (55-77)
Biomass burning and biofuels	17.4	30 (22-39)
Biomass burning	17.4	17 (14-26)
Biofuels		10 (8-13)
Other anthropogenic sources	24	
Wetland	144	149 (102-182)
Other natural sources	104.6**	222 (143-306)
Sum of sources	646.5	727 (581-872)
	Sinks	
Total chemical loss	623	602 (496-754)
Soil absorption	17	30 (11-49)
Sum of sinks	640	632 (507-803)

*Values are from the bottom-up estimates over 2008-2017

** This other natural sources in GEOS-Chem is scaled up from 13.6 to 104.6 Tg CH_4 yr^{-1} to keep the balance of the total CH_4 budget, as the natural sources of CH_4 remained largest uncertainties among all sectors.

References

Canadell.J.G, Monteiro, P. M. S., Costa, M. H., Cunha, L. C. d., Cox, P. M., Eliseev, A. V., Henson, S., Ishii, M., Jaccard, S., Koven, C., Lohila, A., Patra, P. K., Piao, S., Rogelj, J., Syampungani, S., Zaehle, S., and Zickfeld, a. K.: Global Carbon and other Biogeochemical Cycles and Feedbacks. In Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change, 673–816, https://doi.org/10.1017/9781009157896.007, 2021.