

Relating atmospheric N₂O concentration to N₂O emission strength in the U. S. Corn Belt

Congsheng Fu^{1,2}, Xuhui Lee^{1,2}, Timothy J. Griffis³, Edward J. Dlugokencky⁴, Arlyn E. Andrews⁴

Supplementary Information

A numerical example on how to constrain the emission multiplier

The concentration observed at NWR in south winds during June 1-20 is 323.981 ppb and that at the height of 185 m on the KCMP tower is 329.424 for the same period, giving an (unadjusted) concentration enhancement of

$$\text{Term 1} = 329.424 - 323.981 = 5.443 \text{ ppb.}$$

The concentration given by the background simulation for the height of 185 m at the KCMP grid point is 321.352 ppb. In this simulation, the agricultural emission is set to zero in the Corn Belt. The concentration given by the default simulation for the same height in this grid point is 321.606 ppb. In the default simulation, the emission intensity is the prior (that is, the flux given by EDGAR42). The modeled concentration enhancement is

$$\text{Term 2} = 321.606 - 321.352 = 0.254 \text{ ppb.}$$

If EDGAR42 were perfect (and the model had no errors), Term 2 would match Term 1 perfectly. However, the fact that Term 2 is much smaller than Term 1 indicates that the prior emission is biased too low.

We use the difference between the observed and the model default concentration enhancement to find an optimal flux multiplier. But before doing so, we should adjust Term 1 slightly to account for the fact the observed background site is NWR but the modeled background is at the KCMP grid. This adjustment for this spatial mismatch is made using the background simulation results. It is worth mentioning that the influence of emissions from the Corn Belt on the atmospheric N₂O concentration at the NWR site is very limited, for example, the modeled N₂O concentrations at the NWR site from the background and scaled simulations are quite close (Table S1). According to the background simulation, the mean concentration at the NWR and the KCMP model grids is 321.257 and 321.352 ppb, respectively. So the amount of adjustment is

$$\text{Term 3} = 321.352 - 321.257 = 0.095 \text{ ppb.}$$

The concentration multiplier constrained by the observation is

$$M_C = (\text{Term 1} - \text{Term 3}) / \text{Term 2} = 21.055$$

Substituting this value in the inversion Equation (1) and noting that $a = 0.740$ for south winds, we obtain a constrained flux multiplier $M_F = 28.102$

Table S1. Modeled N₂O mixing ratio increases (ppb, ‘scaled simulation’ – ‘background simulation’) at the NWR tower site. Values shown in this table are the mean value for each modeling period. Numbers in the brackets are experimental multiples.

Height	June (25-fold)	August (12-fold)	October (3-fold)	December (6-fold)
32 m	0.11	0.08	0.08	0.04
100 m	0.10	0.07	0.07	0.03
185 m	0.09	0.06	0.06	0.03

Table S2. Experimental and optimized flux multiplier M_F using enlarged agricultural emissions for the whole modeling domain. Values in brackets are constrained agricultural emission flux in units of nmol m⁻² s⁻¹.

Time	June 1 – 20	August 1 – 20	October 1 – 20	December 1 – 20
Experimental	0, 1, 25			
Optimized ^a	16.8 (2.58)	8.4 (1.28)	3.1 (0.47)	2.8 (0.43)
Optimized ^b	19.4 (2.97)	10.1 (1.54)	3.4 (0.52)	3.2 (0.49)
Optimized ^c	24.1 (3.68)	11.3 (1.72)	4.2 (0.64)	4.3 (0.66)

Notes: a, b, c: using observation data at heights of 32, 100, and 185 m, respectively.

Table S3. Experimental and optimized flux multiplier M_F using spatial distribution of a prior emission in proportion to fertilization. Values in brackets are constrained agricultural emission flux in units of nmol m⁻² s⁻¹.

Time	June 1 – 20	August 1 – 20	October 1 – 20	December 1 – 20
Experimental	0, 1, 25			
Optimized ^a	17.7 (2.70)	9.2 (1.41)	3.3 (0.51)	3.0 (0.46)
Optimized ^b	20.9 (3.19)	11.2 (1.71)	3.7 (0.57)	3.5 (0.53)
Optimized ^c	25.3 (3.86)	12.5 (1.92)	4.6 (0.70)	4.2 (0.65)

Notes: a, b, c: using observation data at heights of 32, 100, and 185 m, respectively.

Table S4. Experimental and optimized flux multiplier M_F using AMT as the background site. Values in brackets are constrained agricultural emission flux in units of nmol m⁻² s⁻¹.

Time	June 1 – 20	August 1 – 20	October 1 – 20	December 1 – 20
Experimental	0, 1, 25	0, 1, 12	0, 1, 3	0, 1, 6
Optimized ^a	20.4 (3.12)	11.2 (1.71)	4.5 (0.68)	3.0 (0.47)
Optimized ^b	24.3 (3.71)	14.0 (2.15)	5.2 (0.80)	3.7 (0.57)
Optimized ^c	30.0 (4.59)	16.3 (2.49)	6.5 (0.99)	4.4 (0.67)

Notes: a, b, c: using observation data at heights of 32, 100, and 185 m, respectively.

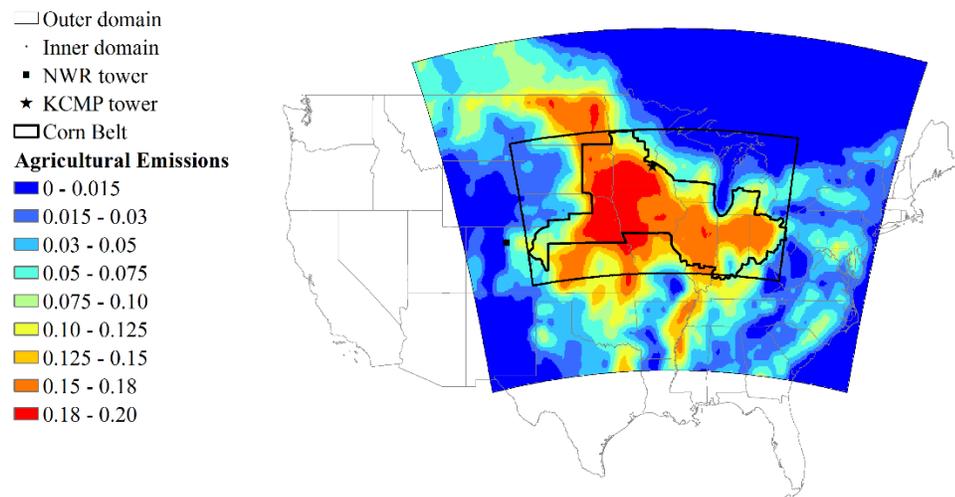


Figure S1. Total EDGAR42 agricultural emission (units: $\text{nmol m}^{-2} \text{s}^{-1}$).

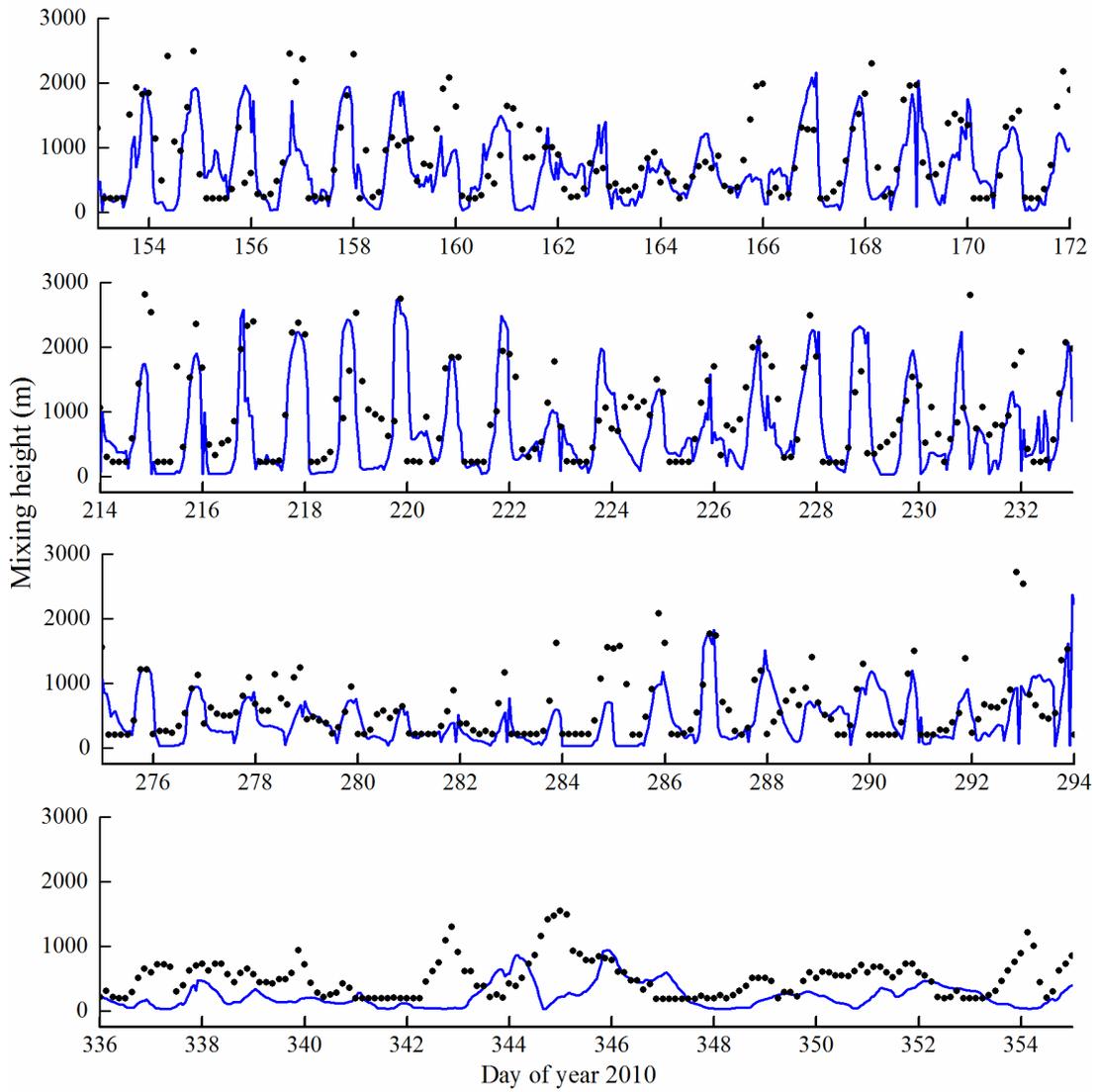


Figure S2. Simulated mixing height and the NCEP-NARR data (dots) for the four simulation periods.

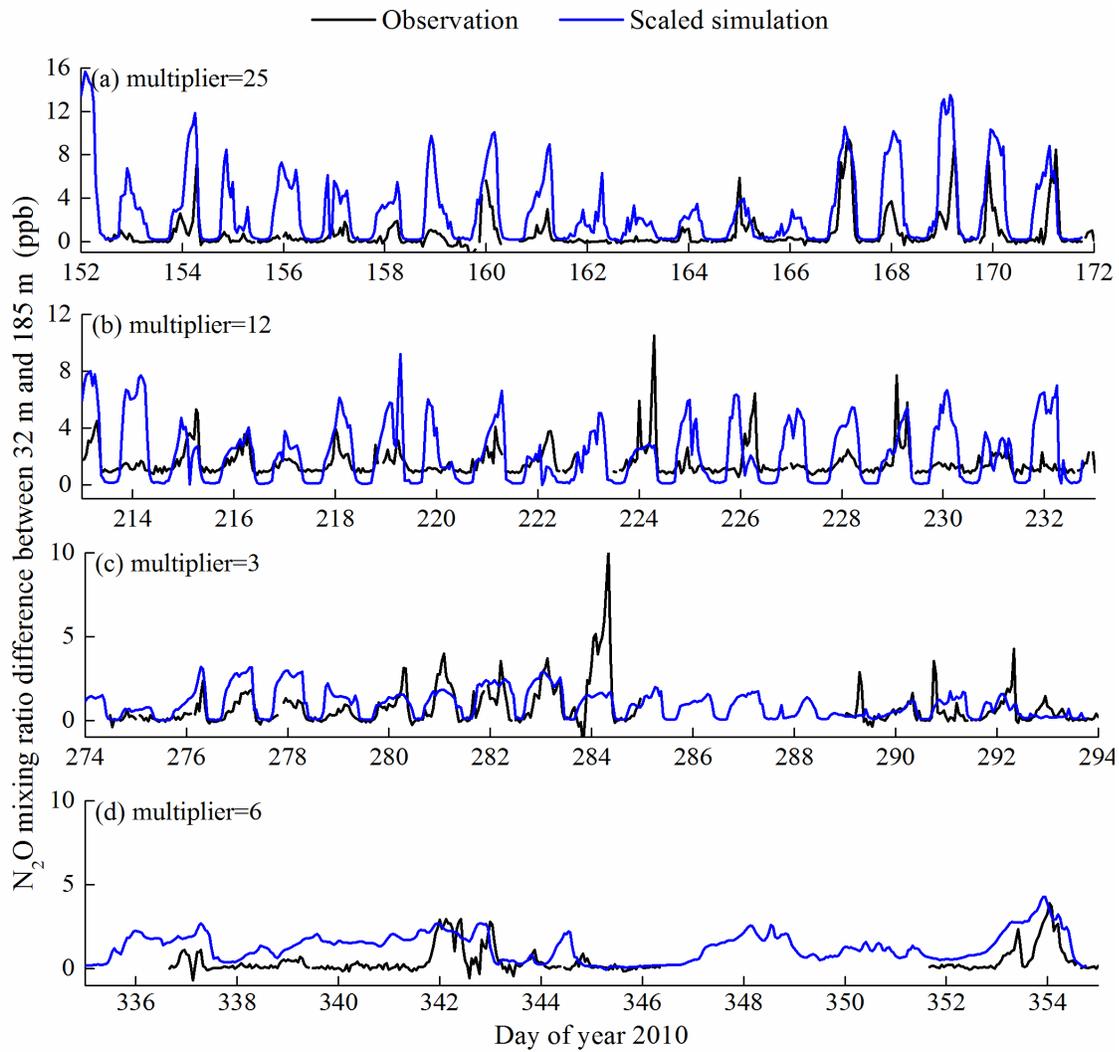


Figure S3. Observed and modeled vertical N_2O mixing ratio gradients (32 m minus 185 m) at the KCMP tower site.

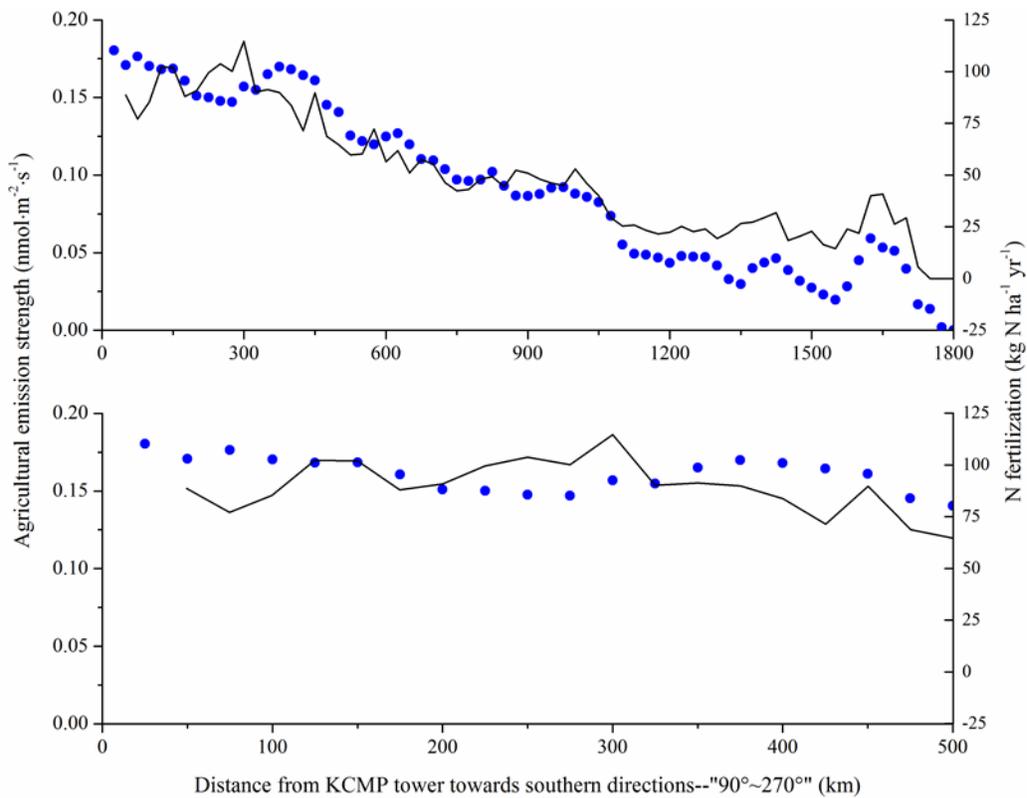


Figure S4. Agricultural N₂O emission strength and N fertilization rate as functions of distance from the KCMP tower in the south wind sector (90° – 270°).

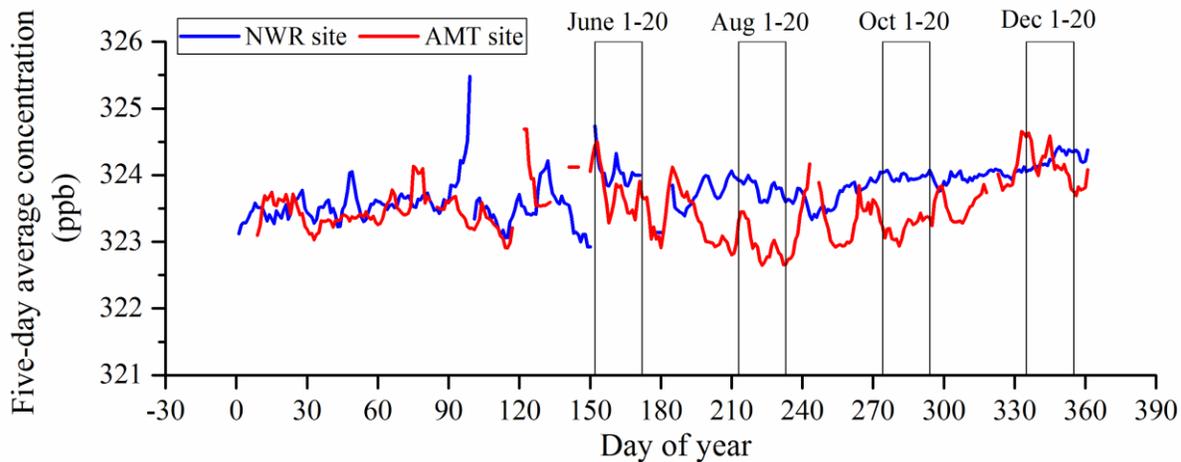


Figure S5. Observed N₂O mixing ratio at Niwot Ridge, Colorado (NWR) and Argyle, Maine (AMT).