Supporting Information for "Modulation of nitrogen deposition by natural and man-made surface heterogeneities in North America"

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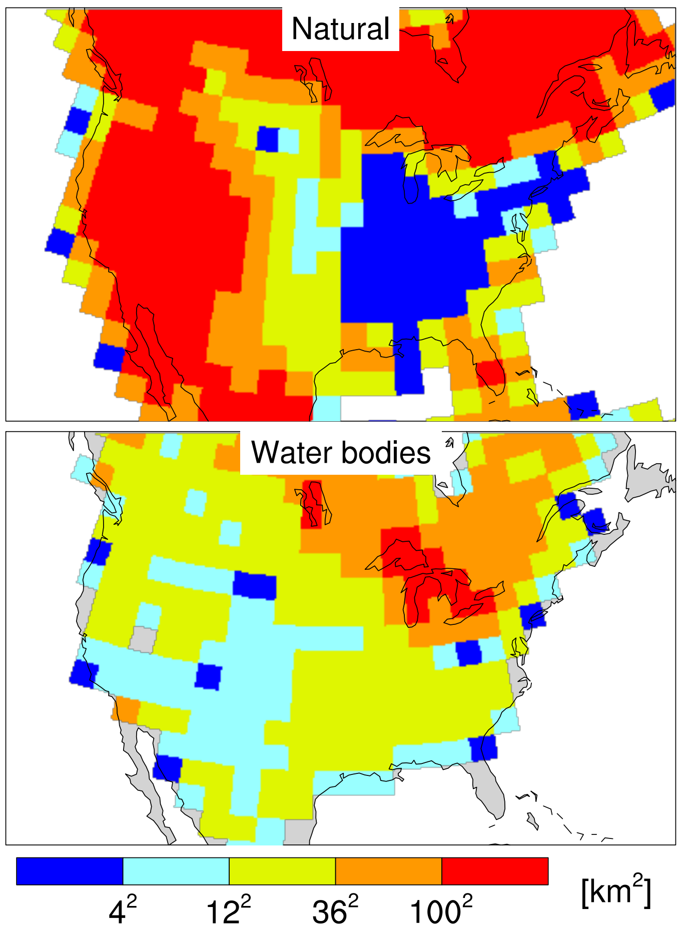

Figure S1: Average area covered by natural vegetation and water bodies from 2008-2010.

Sensitivity of $v_d(HNO_3)$ and $v_d(NH_3)$ to surface properties

NH_3 and HNO_3 are the most important contributors to N dry deposition in the US(Zhang et al., 2012) and we focus here on the impact of surface properties on $v_d(NH_3)$ and $v_d(HNO_3)$. Fig. S2 shows the simulated $v_d(HNO_3)$ and $v_d(NH_3)$ over natural vegetation (green solid line) in two grid cells located in North America: (35°N, -83.75°W) and (51°N, -116.25°W). These grid boxes encompass the Great Smoky Mountains National Park (GSM) in the US and the Banff National Park in Canada. Natural vegetation is comprised of deciduous forest at GSM and coniferous forest at Banff with similar vegetation height ($\sim 20m$) and summer LAI (5–6).

Simulated $v_d(HNO_3)$ is 50% faster at Banff than GSM. Such enhancement over coniferous forests is consistent with observations (Meyers et al., 1989; Sievering et al., 2001; Pryor and Klemm, 2004) and reflects the lower laminar resistance associated with needles relative to deciduous leaves (eq. 1). In contrast, the widely used parameterization of Hicks et al. (1987) (Hicks et al., 1987) suggests a negligible impact of species changes on $v_d(HNO_3)$.

Unlike HNO_3, $R_{sf,v}(NH_3)$ can be large, which exacerbates the sensitivity of $v_d(NH_3)$ to surface properties. This results in a more pronounced seasonality for $v_d(NH_3)$ than $v_d(HNO_3)$, with a maximum in summer and a large impact of acidity and canopy wetness. For instance, $v_d(NH_3)$ in the fall can be twice as large at Banff when canopy wetness is accounted for. At GSM and Banff, the dry deposition of acids is predicted to exceed that of NH_x and co-deposition is simulated to increase $v_d(NH_3)$ by up to 100%.

Fig. S2 also shows that $v_d(HNO_3)$ and $v_d(NH_3)$ over pasture (magenta) and cropland (red) at GSM are slower than over the collocated forest. For HNO_3, this reflects the lower vegetation height, which results in slower $u_\star$ and greater $R_{b,v}$(Hicks, 2006). For NH_3, the lower LAI contributes to the slower $v_d(NH_3)$. The largest reduction relative to natural vegetation is for cropland before and after harvest highlighting the importance of management practices (e.g., grazing intensity, cropping schedule) for N deposition. AM3–LM3 simulates faster $v_d(NH_3)$ over water bodies relative to vegetated surfaces, especially outside of the growing season, but slower $v_d(HNO_3)$. These differences can be attributed to the large effective solubility of NH_3 in freshwater and to the low roughness height of water bodies respectively.

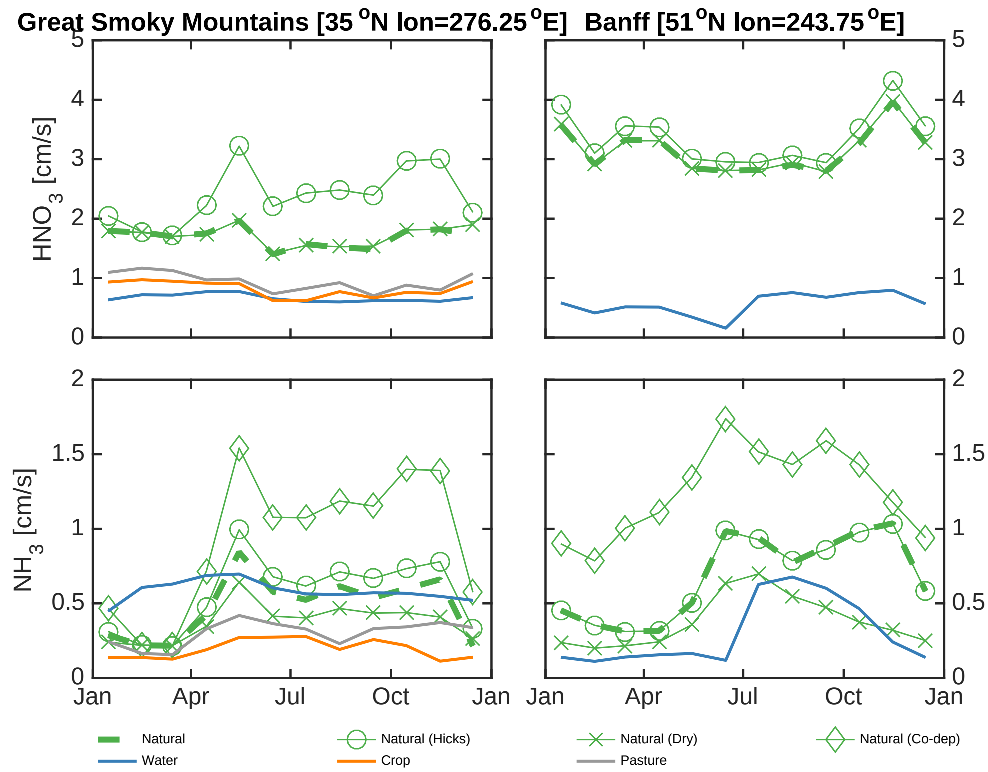

Figure S2: Simulated monthly deposition velocities of HNO_3 and NH_3 for natural vegetation (green), cropland (red), pasture (purple), and water bodies (blue) in the model grid cells that encompass Great Smoky Mountains National Park (GSM, left column) and Banff (right column). The simulated natural vegetation is deciduous tree at GSM and coniferous tree at Banff The green circles show the simulated deposition velocities over natural vegetation when the laminar resistance depends solely on the friction velocity ($u_\star$) (Hicks et al., 1987), neglecting the impact of leaf width(Jensen and Hummelshøj, 1997). Green stars show the simulated deposition velocity when the canopy is assumed to be dry. The green dash line shows the simulated deposition velocity of NH_3 when $R_{ns}(NH_3)$ is allowed to decrease with increasing acid deposition (co-deposition).

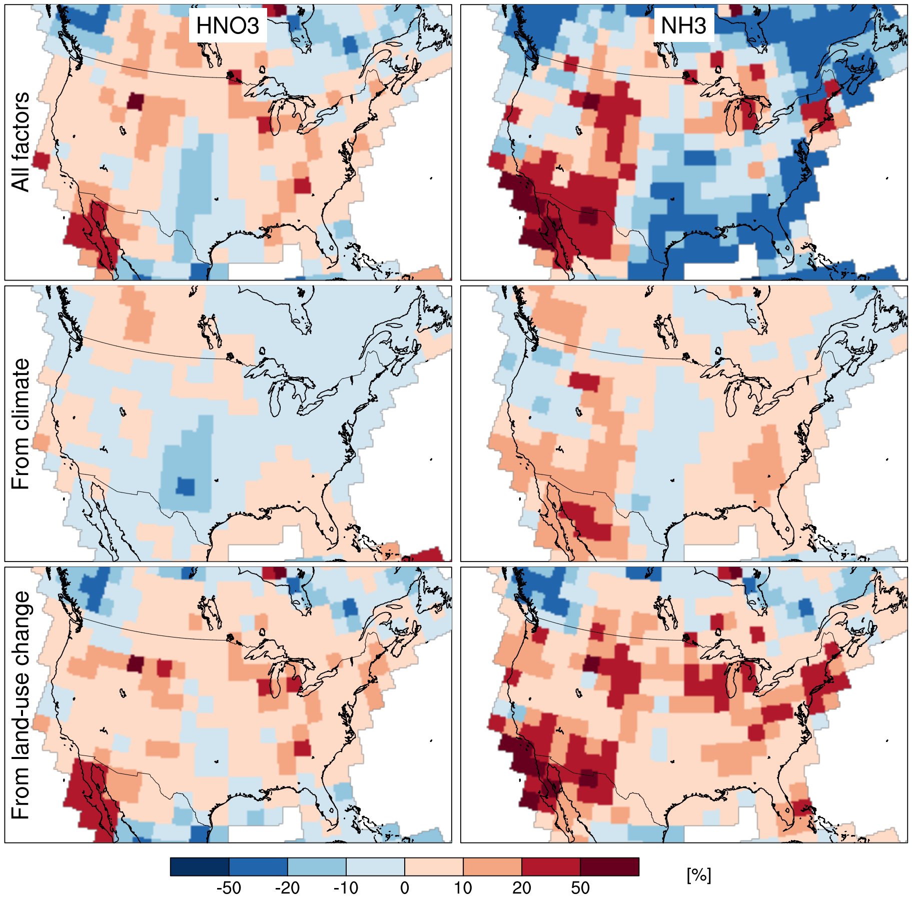

Figure S3: Simulated change in deposition velocity between 2010 and 2050 for HNO_3 and NH_3 (top panel). The contribution of climate change and land-use change to the overall change are shown in the middle and bottom panels respectively.

Table S1: Parameters used for the different LM3 land and vegetation types

	pasture	crop	temperate deciduous	tropical	evergreen	c3 grass	c4 grass	wetland	lake	glacier
r_{cuS} [a]	1000	1500	2500	2500	2000	1000	1000	1500		
r_{cuO} [a]	4000	4000	4000	4000	4000	4000	4000	4000		
r_{cuSw} [a]	100	100	100	100	100	100	100	100		
r_{cuOw} [a]	200	200	400	400	200	200	200	300		
r_{gS} [a,b]	200	200	200	100	200	200	200	50	20	70
r_{gO} [a,b]	200	200	200	100	200	200	200	500	500	2000
r_{bkS} [c]	1000	1000	1000	1000	1000	1000	1000	1000		
r_{bkO} [d]	4000	4000	4000	4000	4000	4000	4000	4000	4000	4000
lw (cm)[e]	1	5	3	5	0.15	1	1	see note (f)		

[a] resistances from Zhang et al. (2003)
[b] the surface resistance is set to 700 s/m (desert) for vegetated tiles with above ground biomass lower than 0.25 $kg\,m^{-2}$
[c] from Padro et al. (1993)
[d] assuming $r_{bkO} = 4r_{bkS}$
[e] from Petroff and Zhang (2010)
[f] based on the tile vegetation type

Table S2: Compound specific adjustment

Name	α	β	MW (kg/mol)
H_2O_2[a,b]	100000	1	0.034
Hydroxyacetone[a]	5	1	0.149
HCOOH[a]	0.056	1	0.045
HNO_3[a]	100000	1	0.063
IEPOX[a]	20	1	0.118
ISOPNB[a]	7	1	0.147
ISOPND[a]	7	1	0.147
$ISOPNO_3$[a]	7	1	0.147
ISOPOOH[a]	20	1	0.118
MACRN[a]	7	1	0.149
MACROOH[a]	20	0	0.12
MPAN[c]	0	1	0.147
MVKN[a]	7	1	0.149
N_2O_5[d]	100000	1	0.063
PAN[c]	0	2	0.121
PROPNN[a]	7	1	0.119

[a] α and β tuned to capture Nguyen et al. (2015) measurements
[b] no adjustment for cold temperature is done following Wesely (1989)
[c] following Wu et al. (2012)
[d] assumed to follow HNO_3

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