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Sensitivity of nitrate aerosols to ammonia emissions and to nitrate chemistry: implications for present and future nitrate optical depth

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Table S1: Heterogeneous reactions and uptake coefficients used in AM3N

	Aerosol	γ	Reference
γ_{HO_2}	all aerosol	1	Mao et al. (2013)
γ_{NO_2}	fine aerosol[a]	10^{-5}	Wong et al. (2011)
γ_{NO_3}	fine aerosol[a]	10^{-3}	Jacob (2000)
$\gamma_{N_2O_5}$	fine aerosol[a]	10^{-2}	Macintyre and Evans (2010)
γ_{NO_3}	dust[b]	0.012	Crowley et al. (2010)
$\gamma_{N_2O_5}$	dust[b]	0.013	Crowley et al. (2010)
γ_{SO_2}	dust	10^{-4}	Ullerstam et al. (2002, 2003)
γ_{HNO_3}	dust[b]	$\frac{8\,RH}{30(1-RH)(1+7RH)}$ for RH<73%	Vlasenko et al. (2006)
		0.118 for RH≥73%	

[a] excludes seasalt and dust (including nitrate and sulfate on dust)
[b] if alkalinity is available

Table S2: Present-day budget of SO_4, NH_x, and NO_y in AM3 and AM3N

	AM3	AM3N	AM3N_fdep_diu	AM3N_diu	AM3N_fdep	AM3N_ns	AM3N_nhet	AM3N_ndust
SO_4[a]								
Production (TgS/yr)	37.3	33.1	33.9	33.3	33.8	33.3	34.0	32.5
OH	10.4	7.7	7.7	7.7	7.7	7.7	8.0	9.1
H_2O_2	26.7	16.2	15.9	16.0	16.1	16.3	15.9	16.5
O_3	0.1	4.5	5.6	4.8	5.2	4.5	5.5	3.9
dust	0.0	1.9	1.9	1.9	1.9	1.9	1.9	0.0
Loss (TgS/yr)	37.4	33.3	34.0	33.4	33.9	33.4	34.1	32.6
Dry deposition	4.7	4.6	4.7	4.6	4.7	4.6	4.7	4.0
SO_4	4.7	3.8	3.9	3.8	3.9	3.8	3.9	4.0
SO_4 on dust	0.0	0.8	0.8	0.8	0.8	0.8	0.8	0.0
Wet deposition	32.7	28.7	29.3	28.8	29.2	28.8	29.4	28.6
SO_4	32.7	27.5	28.2	27.6	28.1	27.6	28.3	28.6
SO_4 on dust	0.0	1.1	1.1	1.1	1.1	1.1	1.1	0.0
Lifetime (days)	4.9	3.8	3.8	3.8	3.8	3.8	3.8	4.3
NH_x								
NH_3 emission (TgN/yr)	54.5	54.5	54.5	54.5	54.5	53.7	54.5	54.5
Loss (TgN/yr)	54.8	55.0	54.8	54.8	55.0	54.2	55.0	55.0
Dry deposition	14.4	23.5	21.3	21.0	23.8	23.2	23.7	23.3
Wet deposition	40.4	30.7	32.6	32.9	30.4	30.3	30.5	31.0
Gas oxidation	0.0	0.8	0.9	0.9	0.8	0.7	0.8	0.7
Lifetime (days)	5.5	2.5	2.7	2.8	2.5	2.5	2.5	2.6
NO_y								
NO emission (TgN/yr)	51.4	51.8	51.8	51.7	51.7	51.8	51.8	51.8
Loss (TgN/yr)	51.3	51.0	51.2	51.0	51.1	51.0	51.1	51.5
Dry deposition	25.4	23.1	25.4	23.1	25.3	23.1	23.4	23.1
HNO_3	18.3	10.7	9.4	10.7	9.5	10.7	10.0	13.9
NO_3 on dust	0.0	3.4	3.3	3.4	3.3	3.4	3.4	0.0
NH_4NO_3	0.7	0.8	4.4	0.8	4.2	0.8	0.5	0.9
Organic nitrogen	3.9	4.0	4.0	4.0	4.0	4.0	4.3	4.0
Wet deposition	25.6	27.6	25.5	27.6	25.5	27.6	27.4	28.1
HNO_3	23.4	17.8	16.4	17.4	16.7	17.7	18.2	21.6
NO_3 on dust	0.0	3.7	3.7	3.7	3.7	3.7	3.7	0.0
NH_4NO_3	0.5	3.5	2.9	3.9	2.5	3.6	2.6	3.9
Organic nitrogen	1.7	2.6	2.6	2.6	2.6	2.6	2.9	2.6
Lifetime (days)	22.7	13.4	13.3	13.4	13.3	13.4	13.6	12.9

[a] SO_2 emissions are 74.0 $TgS\,a^{-1}$ including 16.0 $TgS\,a^{-1}$ from DMS oxidation

Table S3: Summary of model evaluation[a]

	AM3	AM3N	AM3N_fdep_diu	AM3N_diu	AM3N_fdep	AM3N_ns	AM3N_nhet	AM3N_ndust
SO_4								
Aerosol								
US	0.07 (0.81)	-0.11 (0.89)	-0.06 (0.89)	-0.11 (0.89)	-0.05 (0.89)	-0.05 (0.89)	-0.06 (0.89)	-0.11 (0.89)
Europe	-0.43 (0.24)	-0.22 (0.62)	-0.13 (0.64)	-0.22 (0.62)	-0.13 (0.64)	-0.20 (0.67)	-0.13 (0.64)	-0.24 (0.62)
Wet deposition								
US	0.00 (0.42)	-0.07 (0.59)	-0.08 (0.57)	-0.07 (0.58)	-0.07 (0.58)	-0.08 (0.58)	-0.08 (0.58)	-0.07 (0.58)
Europe	-0.18 (0.53)	-0.32 (0.57)	-0.32 (0.53)	-0.32 (0.57)	-0.31 (0.57)	-0.32 (0.58)	-0.31 (0.57)	-0.32 (0.55)
NO_3								
Aerosol								
US	-0.61 (0.64)	1.03 (0.64)	0.17 (0.65)	0.99 (0.64)	0.16 (0.64)	1.38 (0.61)	0.42 (0.62)	1.06 (0.64)
Europe	-0.78 (0.62)	0.32 (0.62)	-0.30 (0.58)	0.29 (0.62)	-0.30 (0.58)	0.31 (0.61)	-0.13 (0.50)	0.17 (0.62)
Gas+Aerosol								
Europe	-0.18 (0.61)	0.17 (0.75)	-0.29 (0.57)	0.16 (0.75)	-0.29 (0.57)	0.17 (0.74)	-0.13 (0.62)	0.08 (0.76)
Wet deposition								
US	0.14 (0.33)	0.23 (0.52)	0.11 (0.54)	0.23 (0.52)	0.11 (0.54)	0.21 (0.52)	0.17 (0.54)	0.27 (0.54)
Europe	-0.32 (0.57)	-0.29 (0.54)	-0.39 (0.54)	-0.29 (0.54)	-0.39 (0.55)	-0.28 (0.58)	-0.32 (0.52)	-0.27 (0.56)
NH_x								
Gas								
US	-0.75 (0.50)	-0.10 (0.54)	-0.22 (0.53)	-0.29 (0.54)	-0.04 (0.53)	-0.12 (0.50)	-0.06 (0.53)	-0.11 (0.53)
Europe	-0.65 (0.48)	0.23 (0.54)	0.17 (0.50)	0.04 (0.53)	0.36 (0.52)	0.25 (0.46)	0.31 (0.53)	0.21 (0.54)
Gas+aerosol								
Europe	0.69 (0.66)	0.18 (0.64)	0.02 (0.64)	0.07 (0.63)	0.12 (0.64)	0.19 (0.60)	0.14 (0.64)	0.19 (0.64)
Wet deposition								
US	-0.20 (0.50)	-0.20 (0.69)	-0.15 (0.69)	-0.14 (0.69)	-0.20 (0.68)	-0.18 (0.55)	-0.20 (0.69)	-0.20 (0.69)
Europe	-0.23 (0.52)	-0.36 (0.58)	-0.32 (0.58)	-0.31 (0.58)	-0.36 (0.58)	-0.36 (0.49)	-0.36 (0.58)	-0.35 (0.58)
AOD								
MODIS								
World	0.09 (0.57)	-0.08 (0.68)	-0.08 (0.68)	-0.07 (0.68)	-0.08 (0.68)	-0.07 (0.67)	-0.08 (0.68)	-0.06 (0.69)
high NO_3	-0.15 (0.83)	0.11 (0.87)	0.09 (0.87)	0.14 (0.86)	0.06 (0.87)	0.13 (0.86)	0.06 (0.87)	0.12 (0.87)
high SO_4	0.57 (0.83)	0.06 (0.87)	0.06 (0.87)	0.08 (0.87)	0.04 (0.88)	0.06 (0.87)	0.04 (0.88)	0.09 (0.88)
MISR								
World	-0.03 (0.53)	-0.16 (0.59)	-0.16 (0.59)	-0.15 (0.58)	-0.17 (0.59)	-0.16 (0.58)	-0.17 (0.59)	-0.14 (0.59)
high NO_3	-0.12 (0.84)	0.21 (0.87)	0.18 (0.87)	0.24 (0.86)	0.16 (0.87)	0.23 (0.86)	0.16 (0.88)	0.24 (0.87)
high SO_4	0.54 (0.86)	0.12 (0.88)	0.12 (0.88)	0.14 (0.87)	0.10 (0.88)	0.12 (0.87)	0.10 (0.88)	0.15 (0.88)
AERONET								
World	-0.03 (0.72)	-0.10 (0.82)	-0.11 (0.82)	-0.08 (0.82)	-0.13 (0.82)	-0.09 (0.82)	-0.12 (0.83)	-0.07 (0.82)
high NO_3	-0.50 (0.87)	-0.01 (0.76)	-0.07 (0.70)	0.02 (0.72)	-0.08 (0.73)	0.03 (0.79)	-0.09 (0.74)	0.03 (0.75)
high SO_4	0.33 (0.47)	-0.10 (0.74)	-0.10 (0.71)	-0.07 (0.72)	-0.12 (0.73)	-0.10 (0.75)	-0.13 (0.72)	-0.05 (0.74)

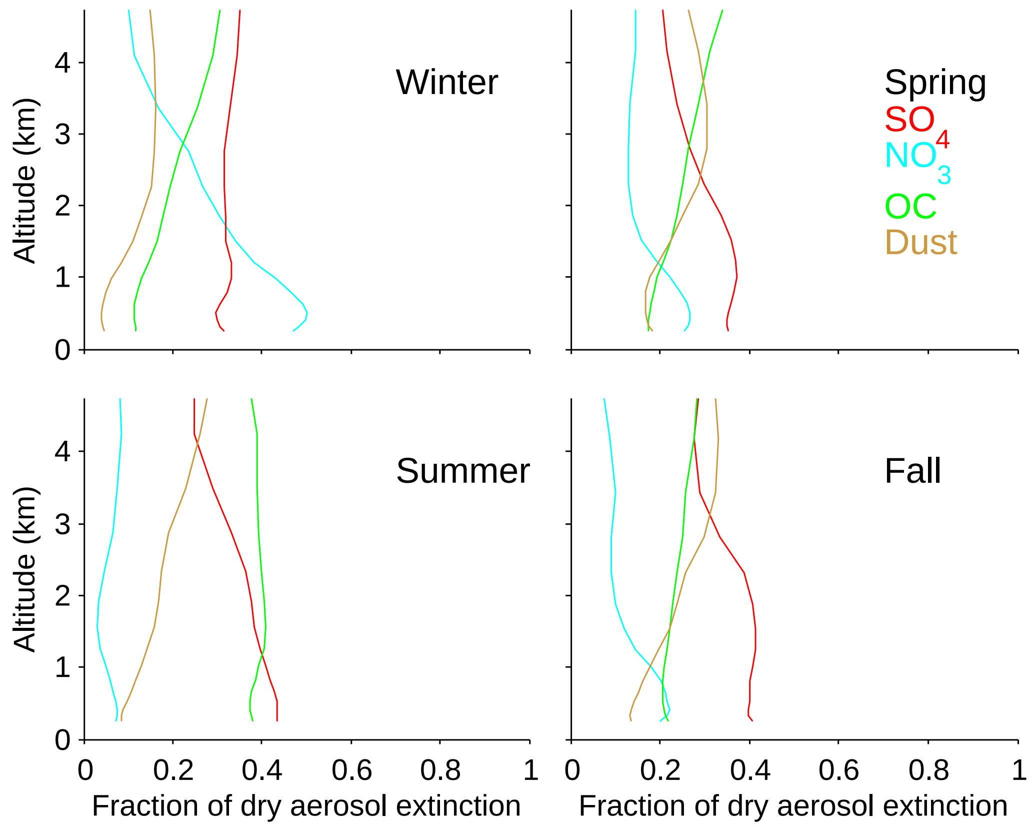

Figure S1: Contribution of SO_4, NO_3, OC, and dust to the simulated dry extinction at Bondville in AM3N_fdep_diu.

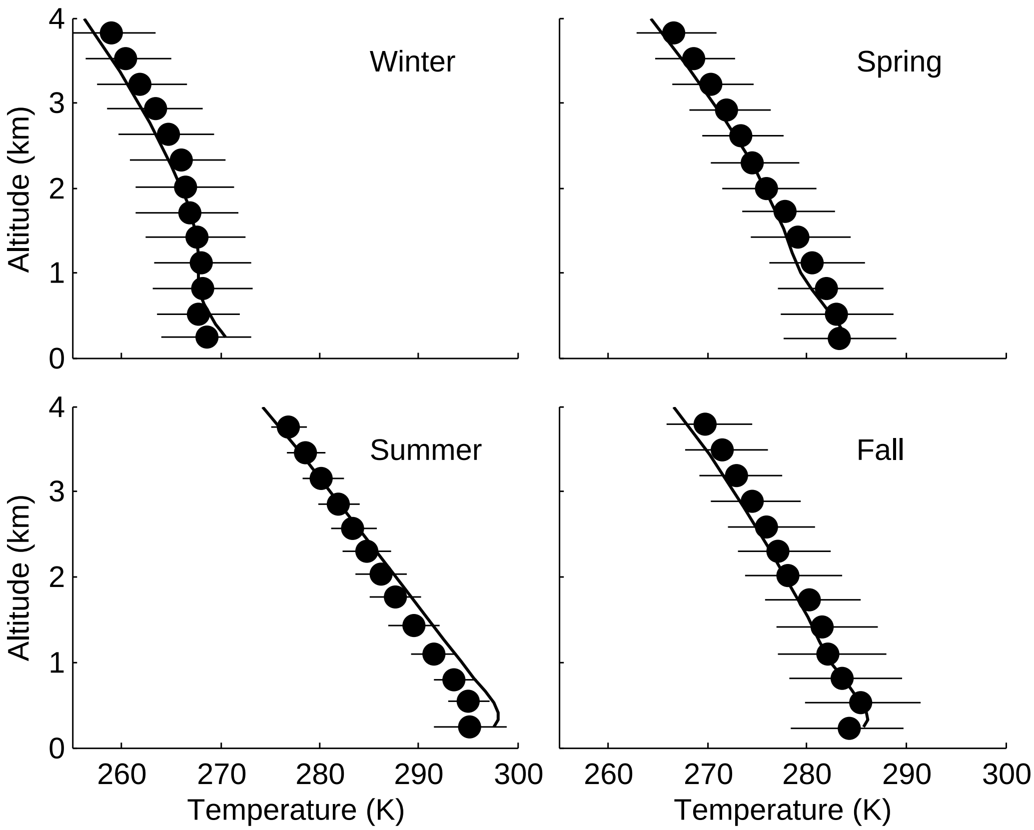

Figure S2: Observed (black dots) and simulated temperature over Bondville. The dots show the average for the each altitude and the bars show the 25^{th} and 75^{th} percentiles. The model is sampled within one hour of the twice daily observations.

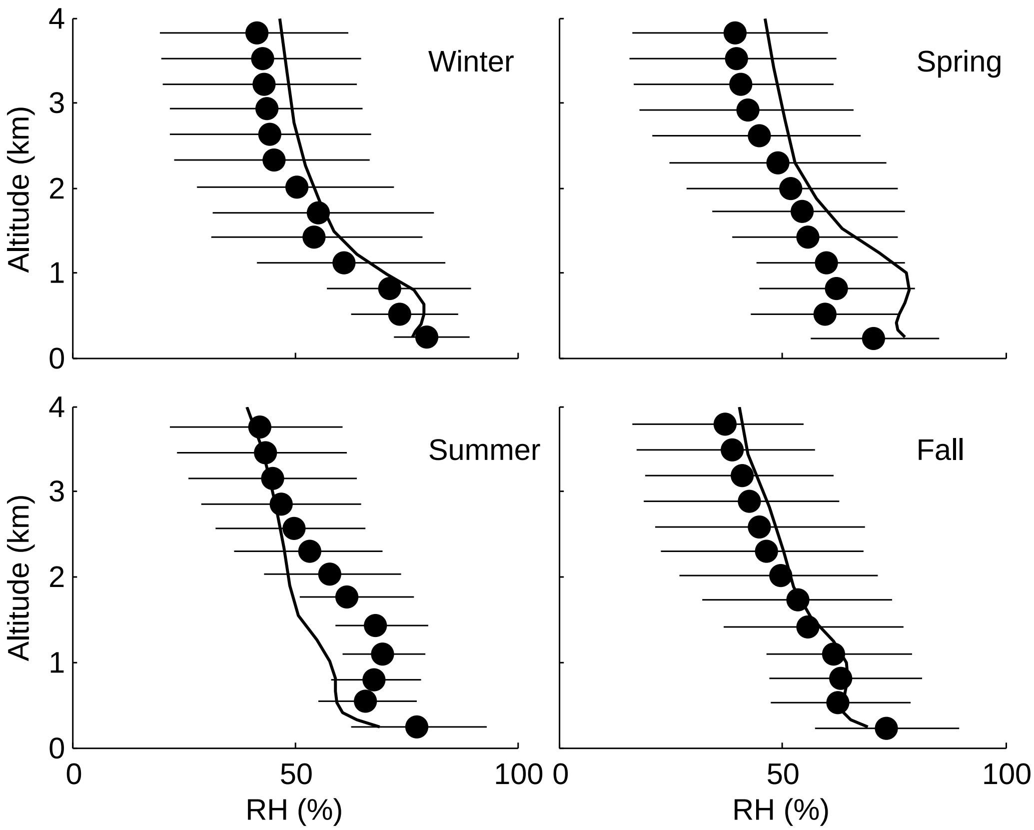

Figure S3: Observed (black dots) and simulated relative humidity over Bondville. The dots show the average for the each altitude and the bars show the 25^{th} and 75^{th} percentiles. The model is sampled within one hour of the twice daily observations.

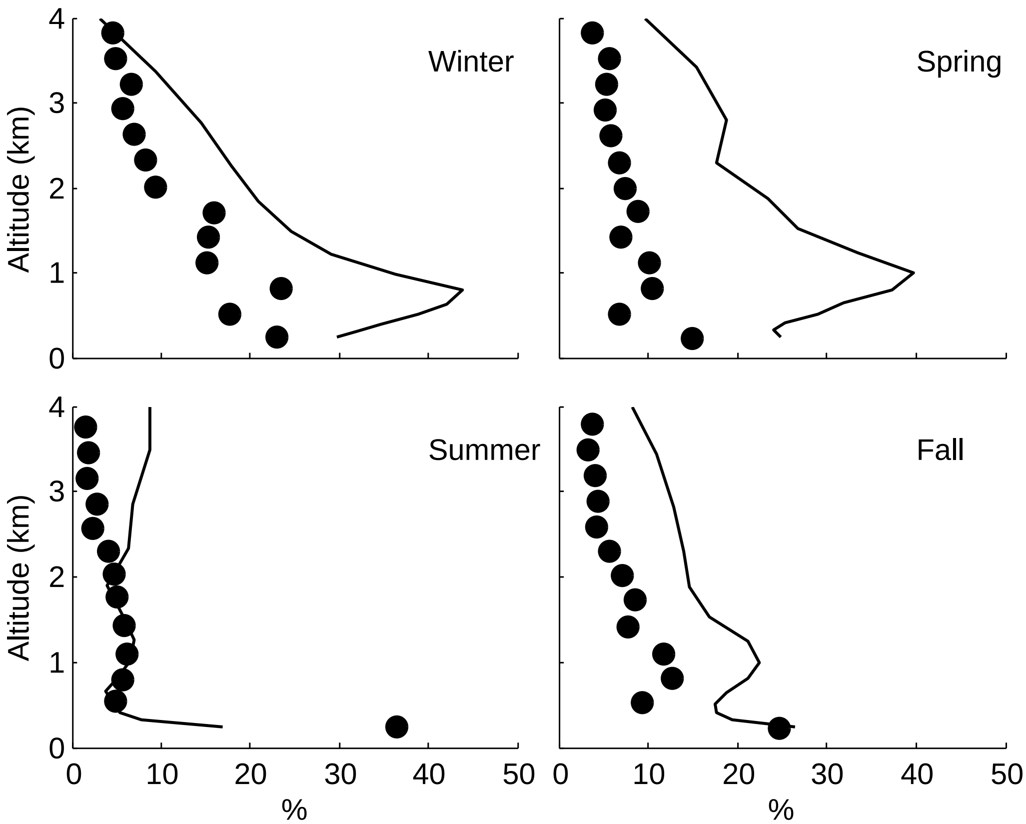

Figure S4: Observed (black dots) and simulated (solid line from AM3N) occurrence of RH exceeding 90% over Bondville from 2008 to 2010. The model is sampled within one hour of the bidaily observations.

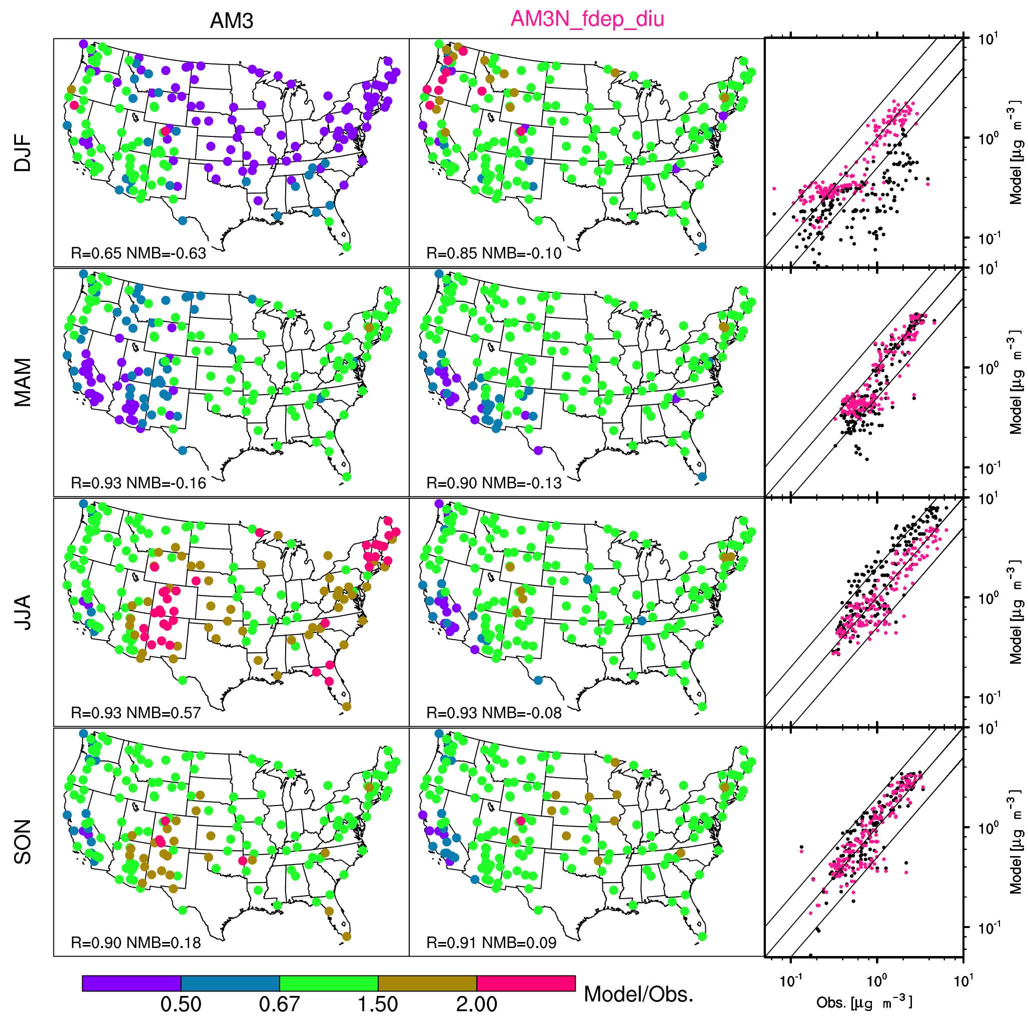

Figure S5: Comparison between observed and simulated SO_4^{2-} for AM3 and AM3N_fdep_diu. The ratio between model and observation is shown for each site and each season for AM3 (left panel) and AM3N_fdep_diu (middle panel). Observations are averaged on a monthly basis over the 2006–2012 period. Scatterplot of seasonal means is shown on the right for AM3 (black) and AM3N_fdep_diu (purple). Note the log scale for the x and y axis.

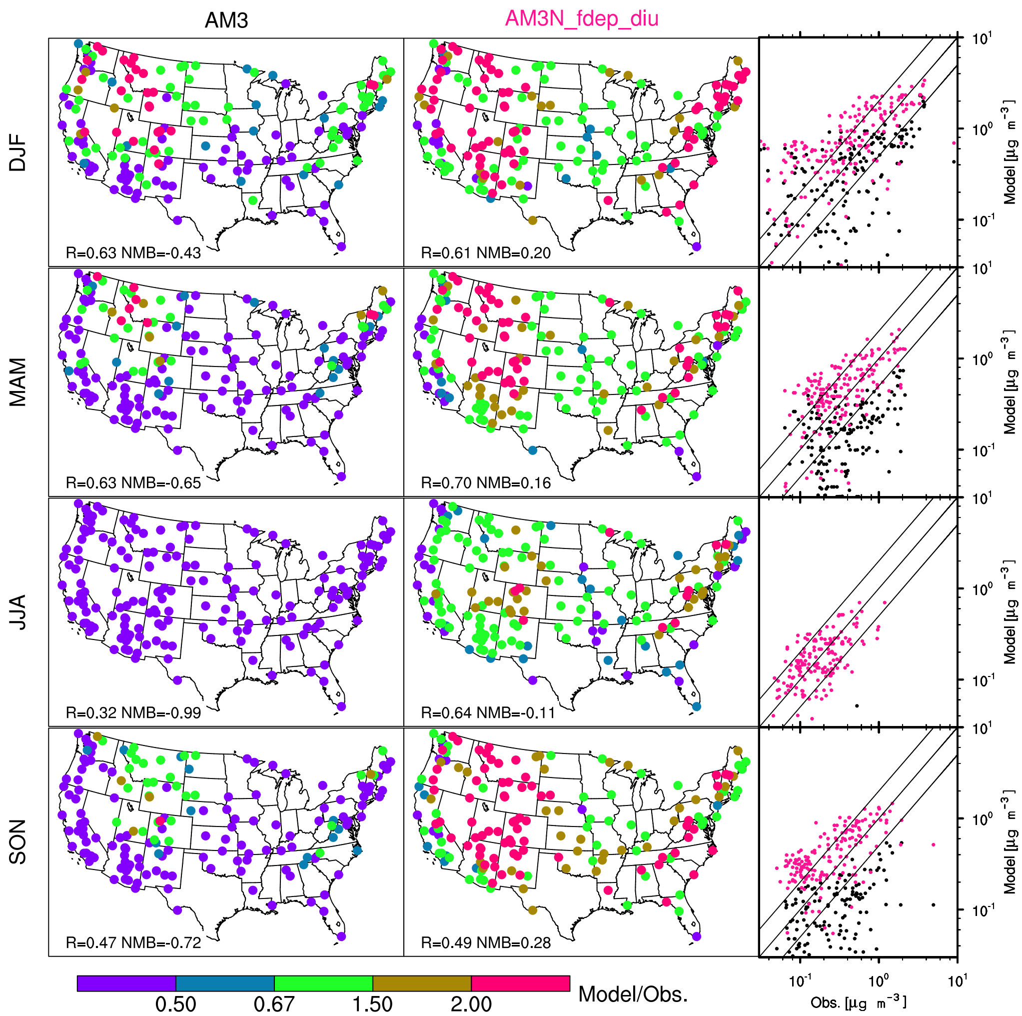

Figure S6: Same as Fig. S5 but for NO_3^- aerosol

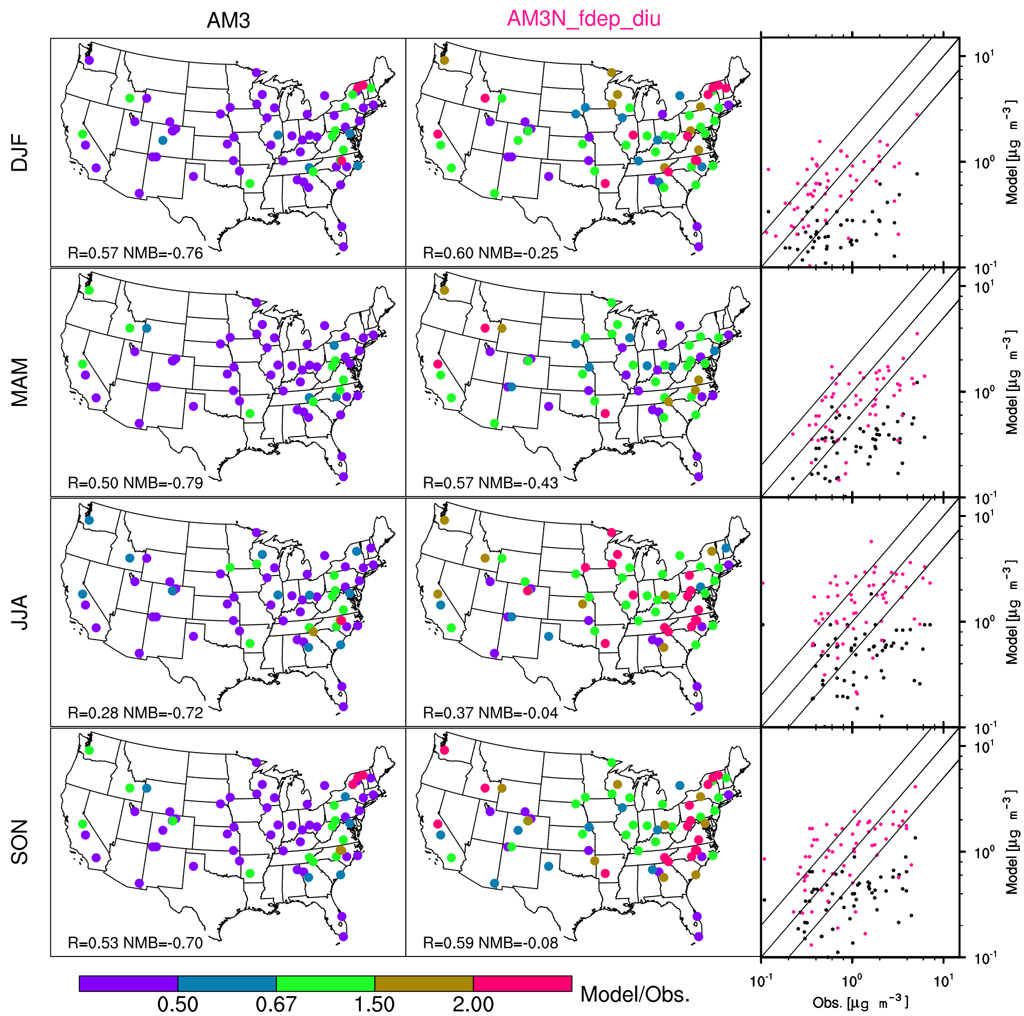

Figure S7: Same as Fig. S5 but for NH_3

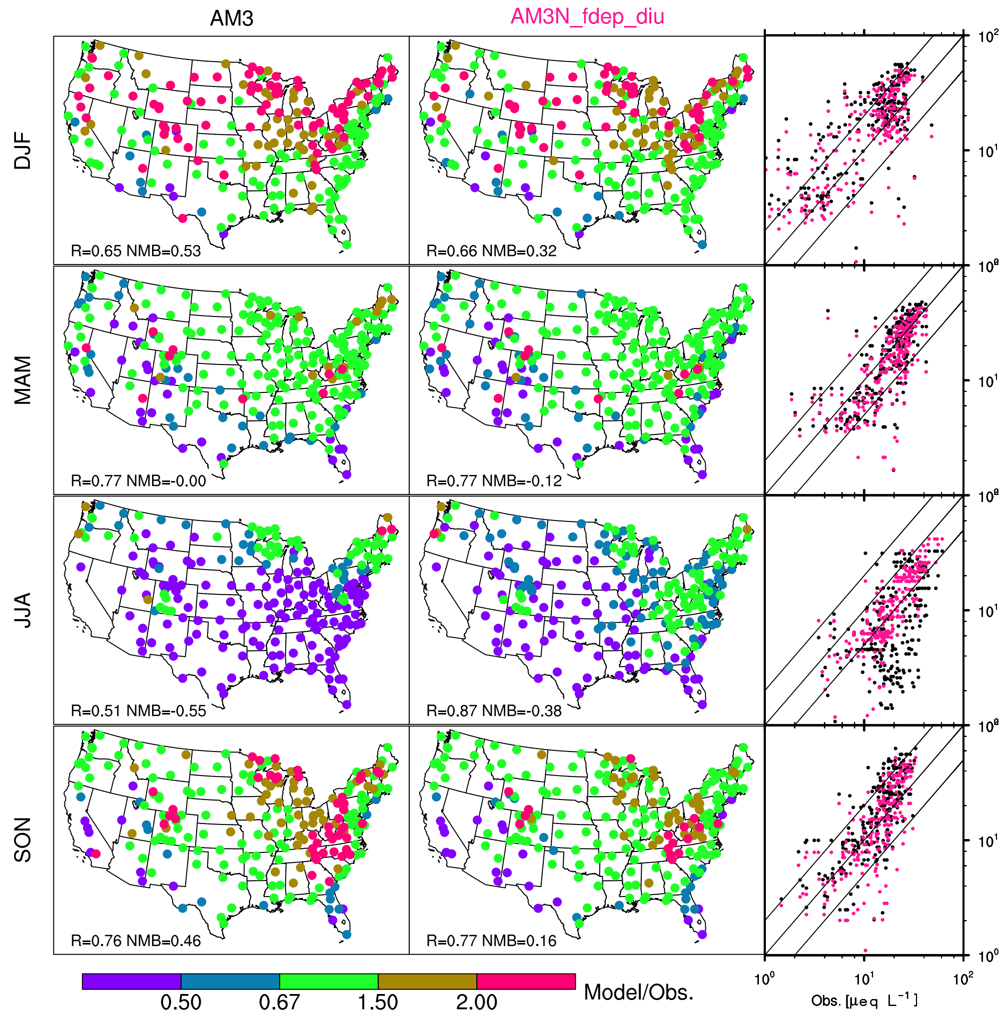

Figure S8: Same as Fig. S5 but for ${SO_4}^{2-}$ concentrations in rain from NADP

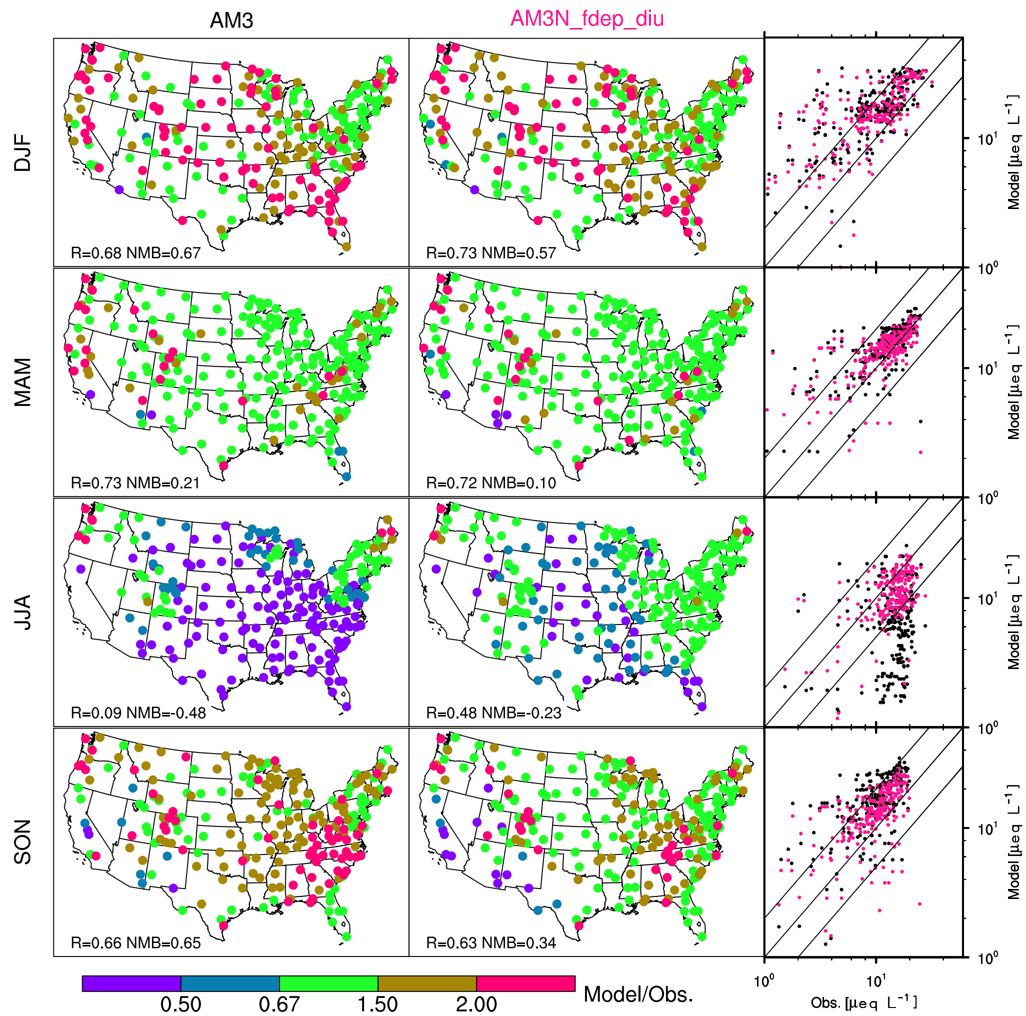

Figure S9: Same as Fig. S5 but for NO_3^- concentration in rain from NADP

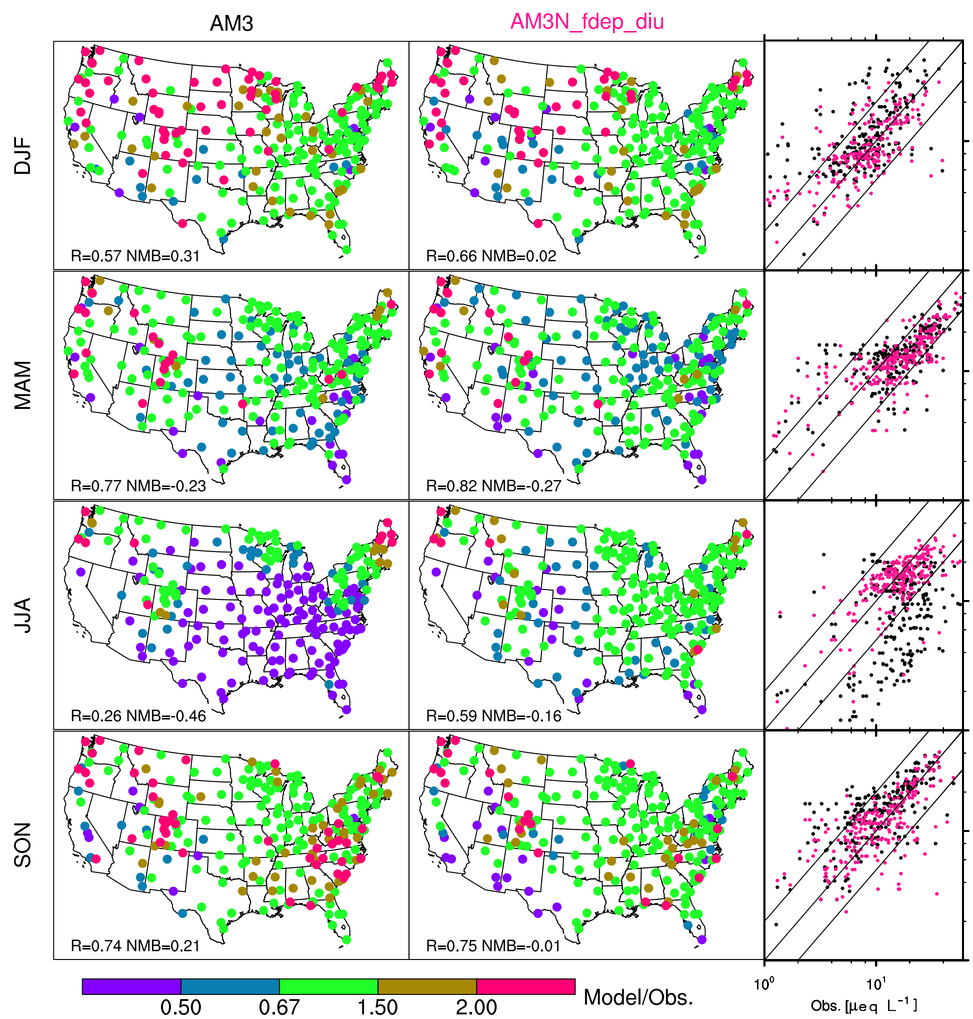

Figure S10: Same as Fig. S5 but for NH_4^+ concentration in rain from NADP

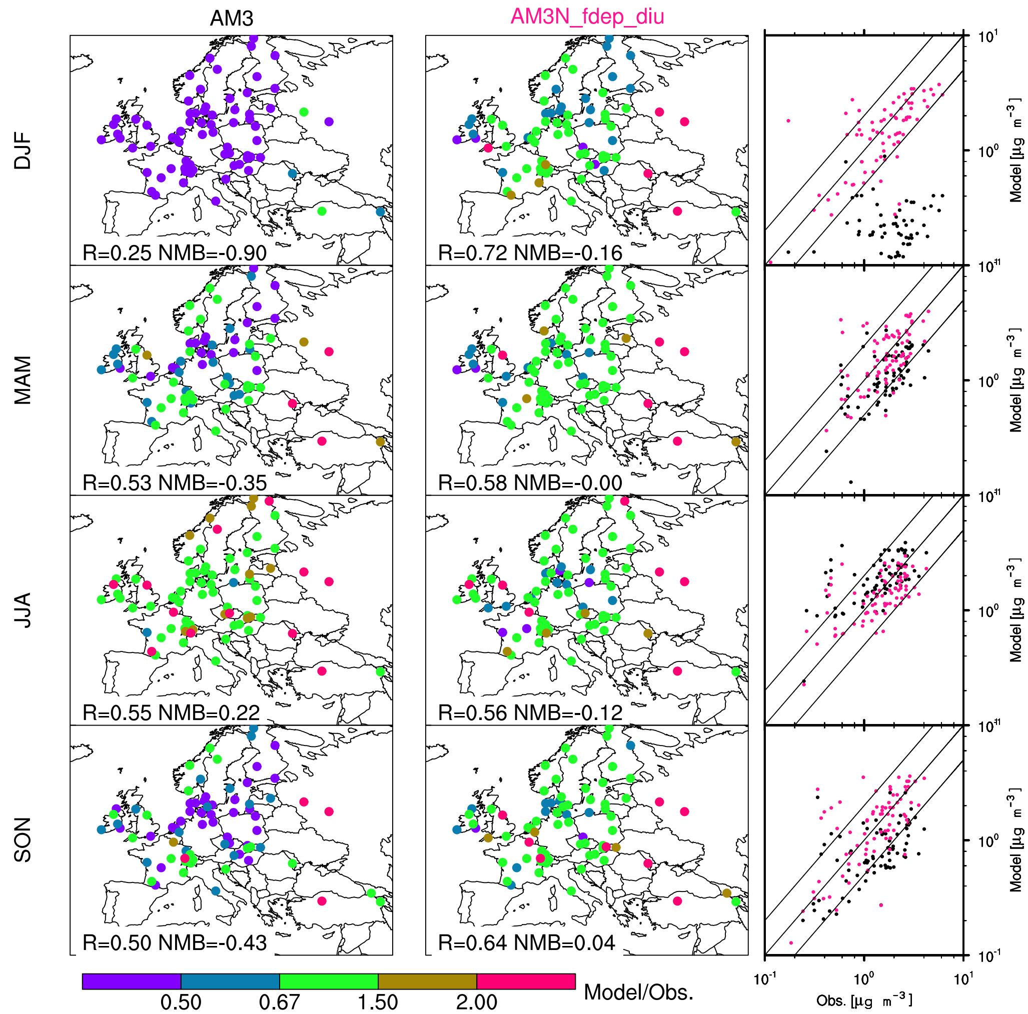

Figure S11: Same as Fig. S5 but for $SO_4{}^{2-}$ surface observations from EMEP.

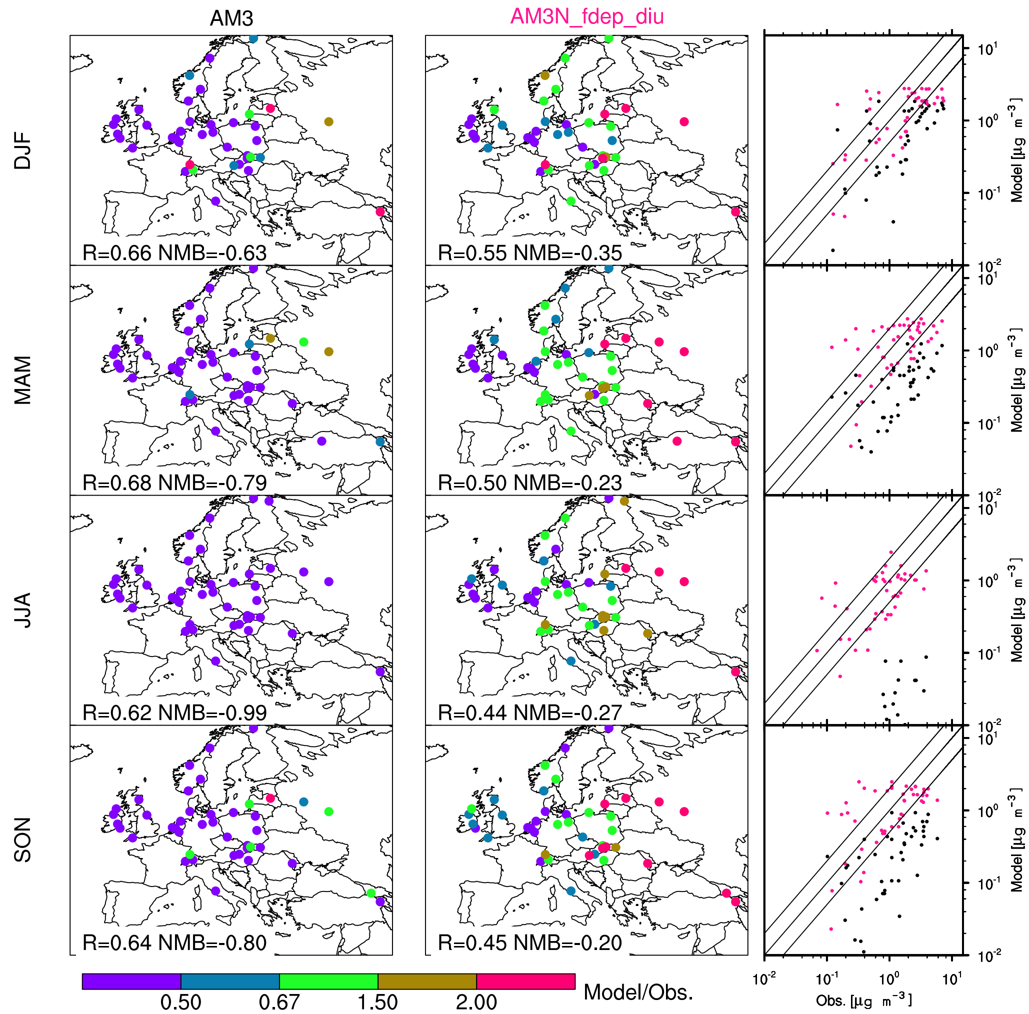

Figure S12: Same as Fig. S5 but for surface NO_3^- from EMEP.

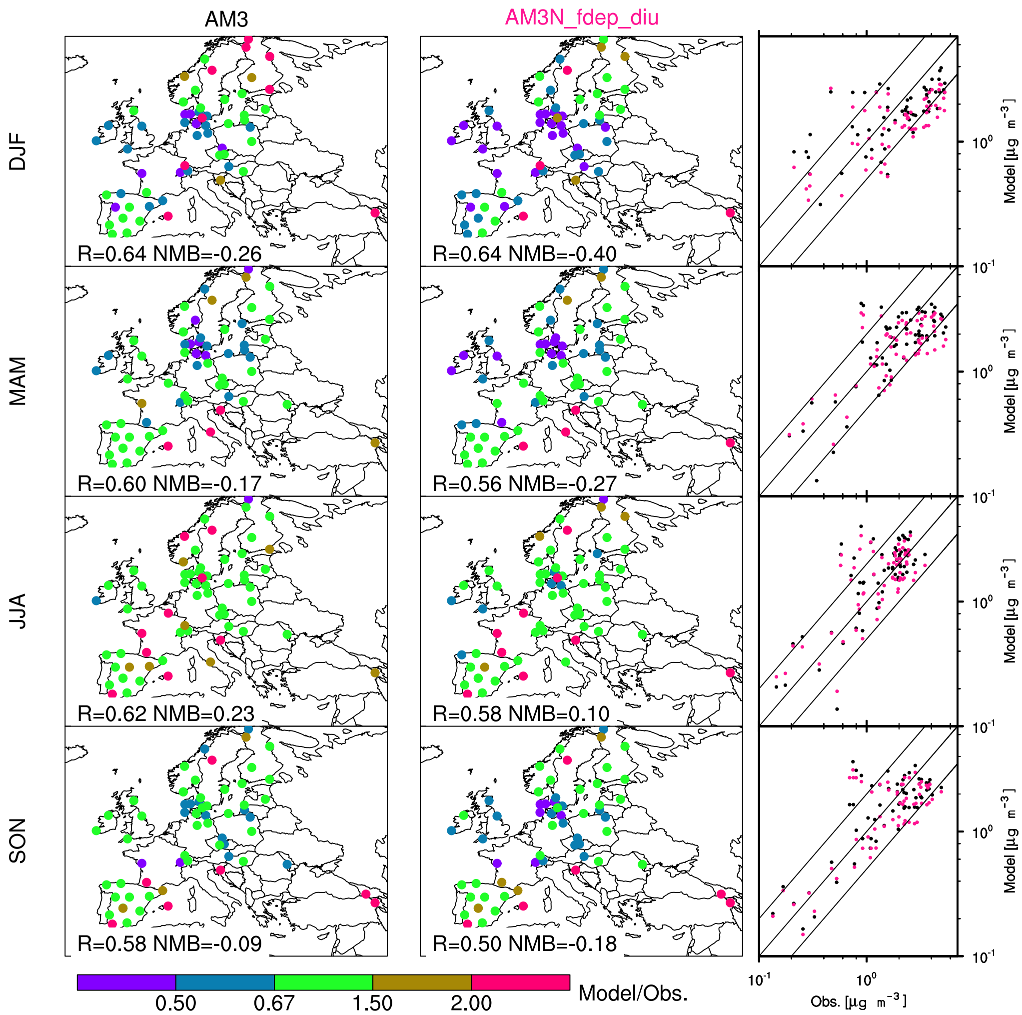

Figure S13: Same as Fig. S5 but for NOy from EMEP.

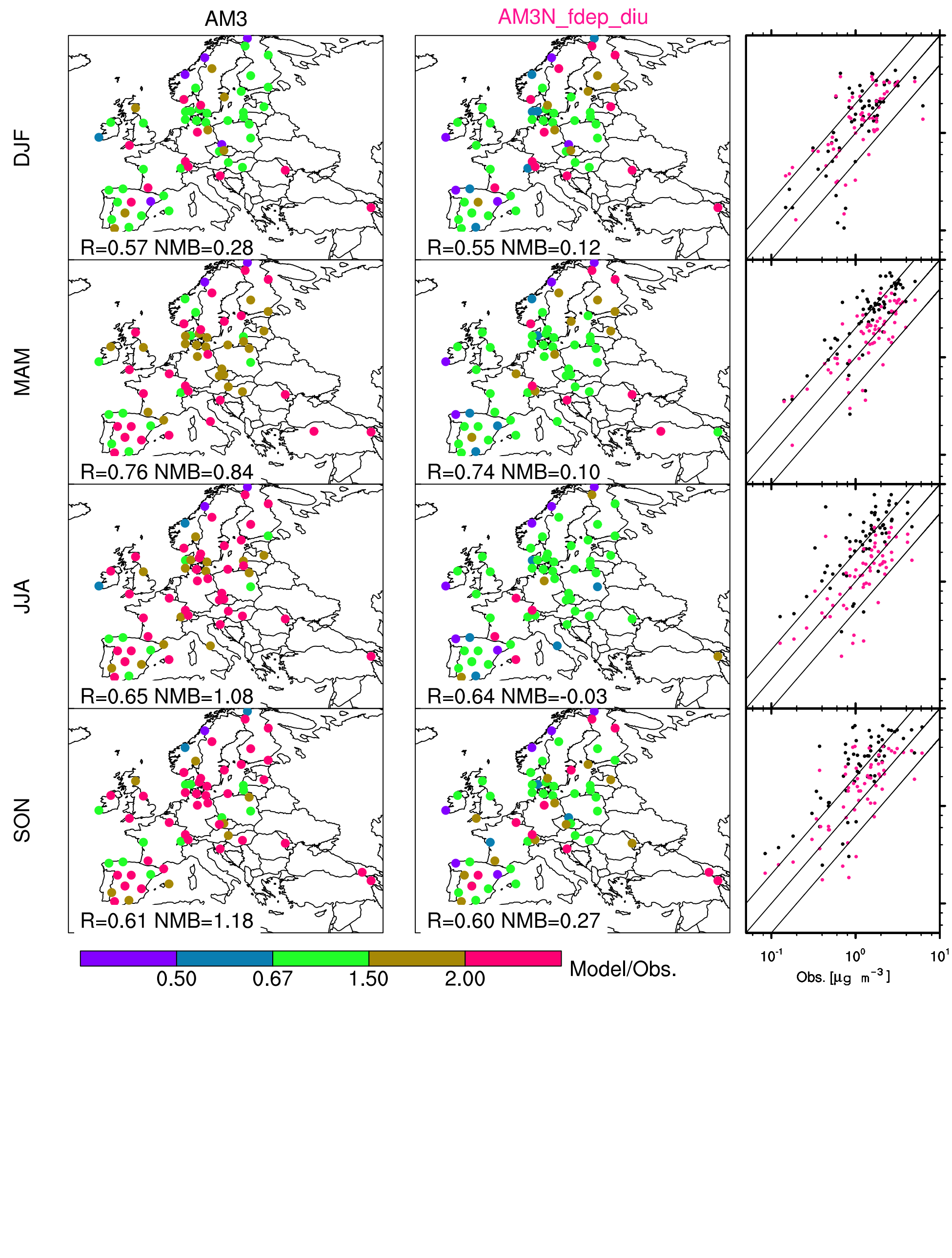

Figure S14: Same as Fig. S5 but for NH_x from EMEP.

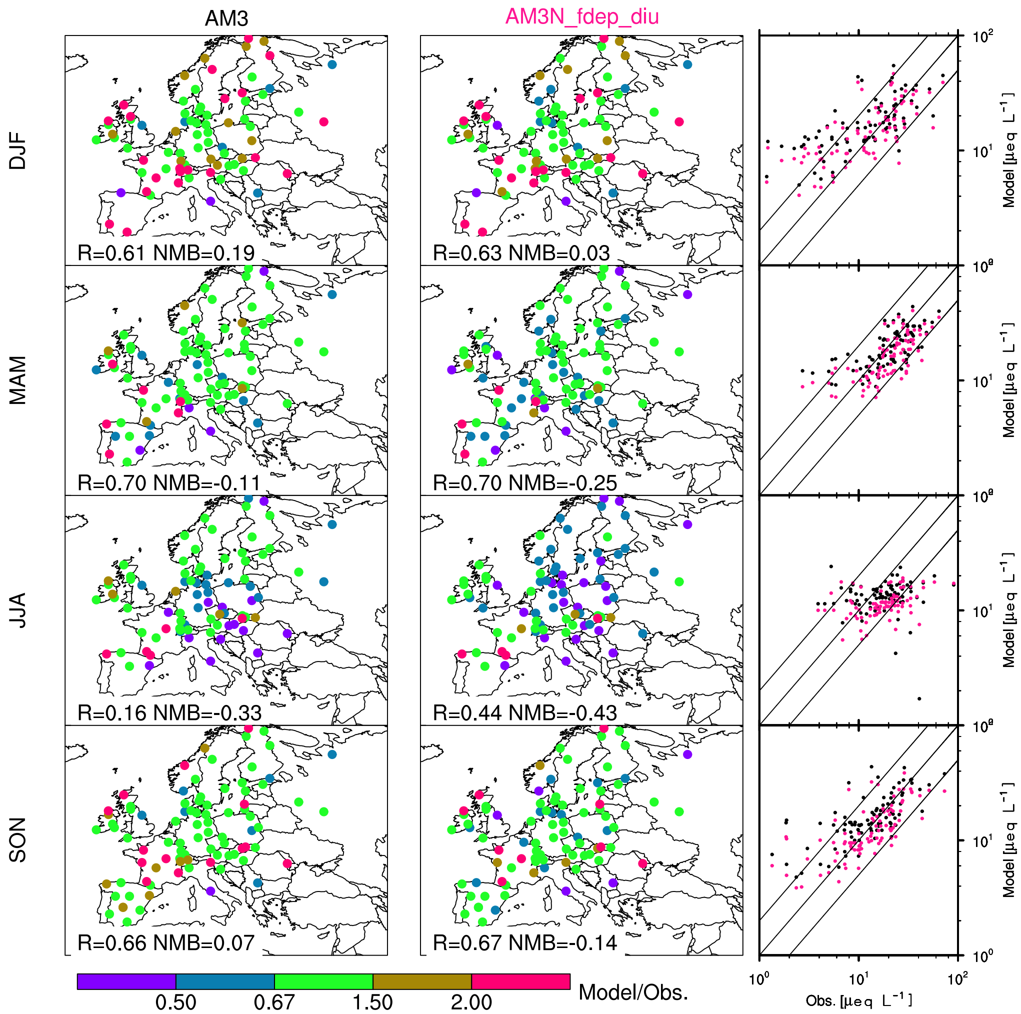

Figure S15: Same as Fig. S5 but for SO_4^{2-} concentration in precipitated water from EMEP

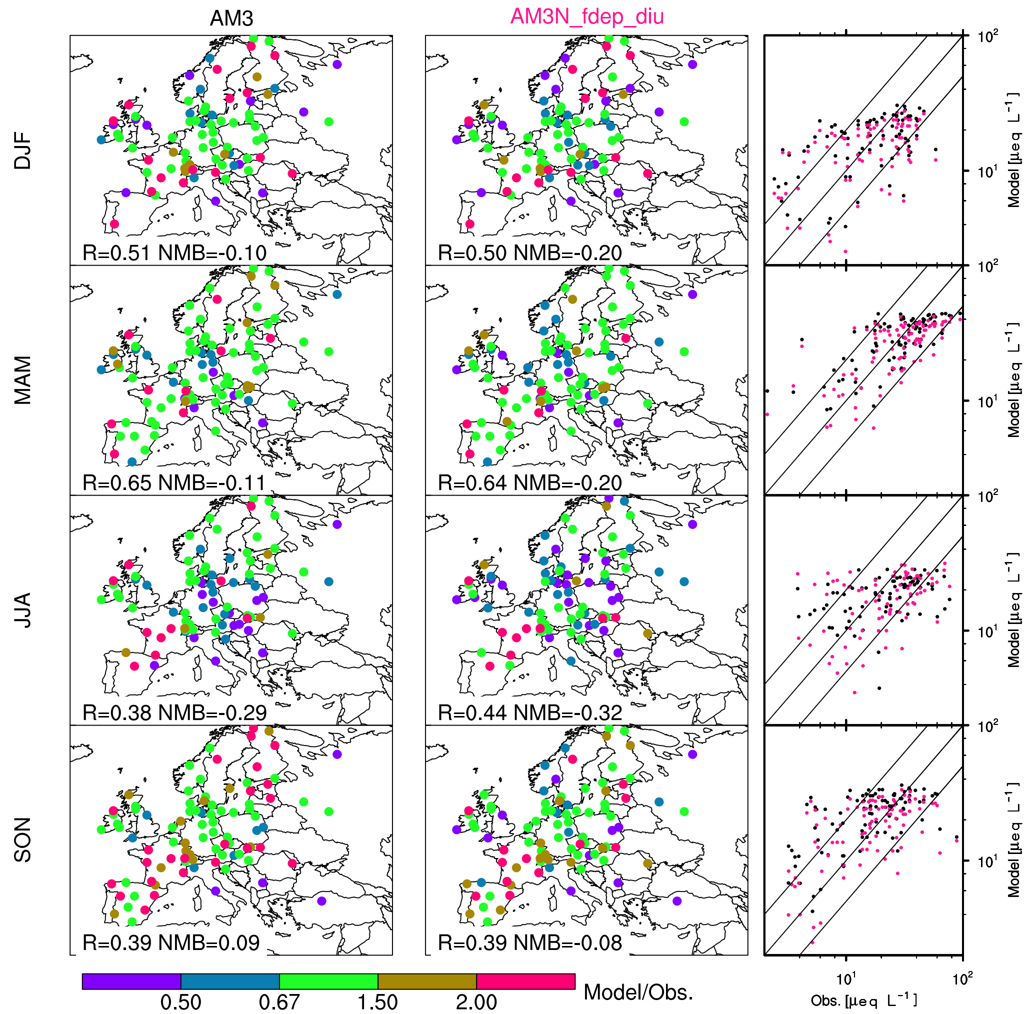

Figure S16: Same as Fig. S5 but for NH_4^+ concentration in precipitated water from EMEP

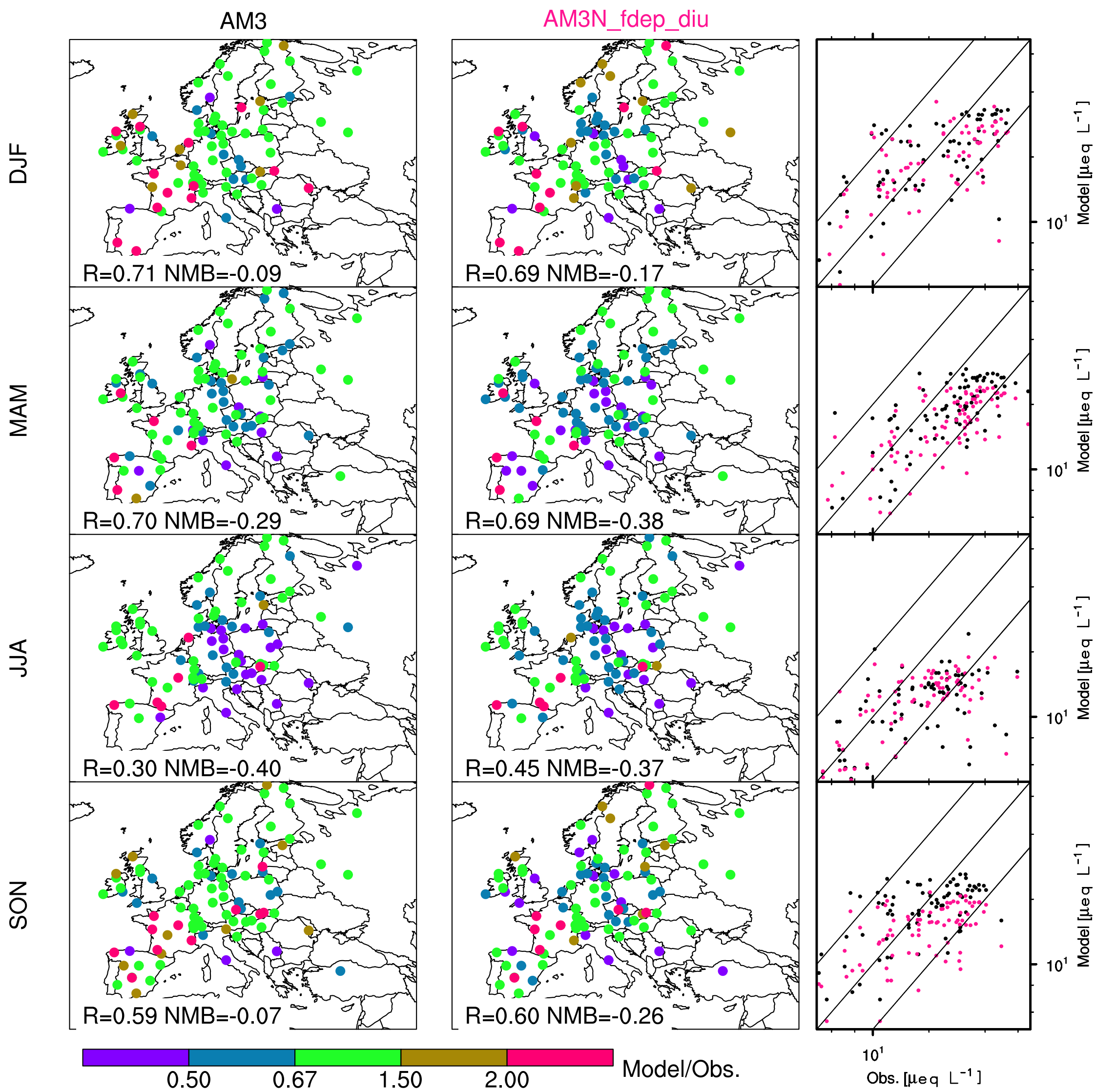

Figure S17: Same as Fig. S5 but for NO_3^- concentration in precipitated water from EMEP

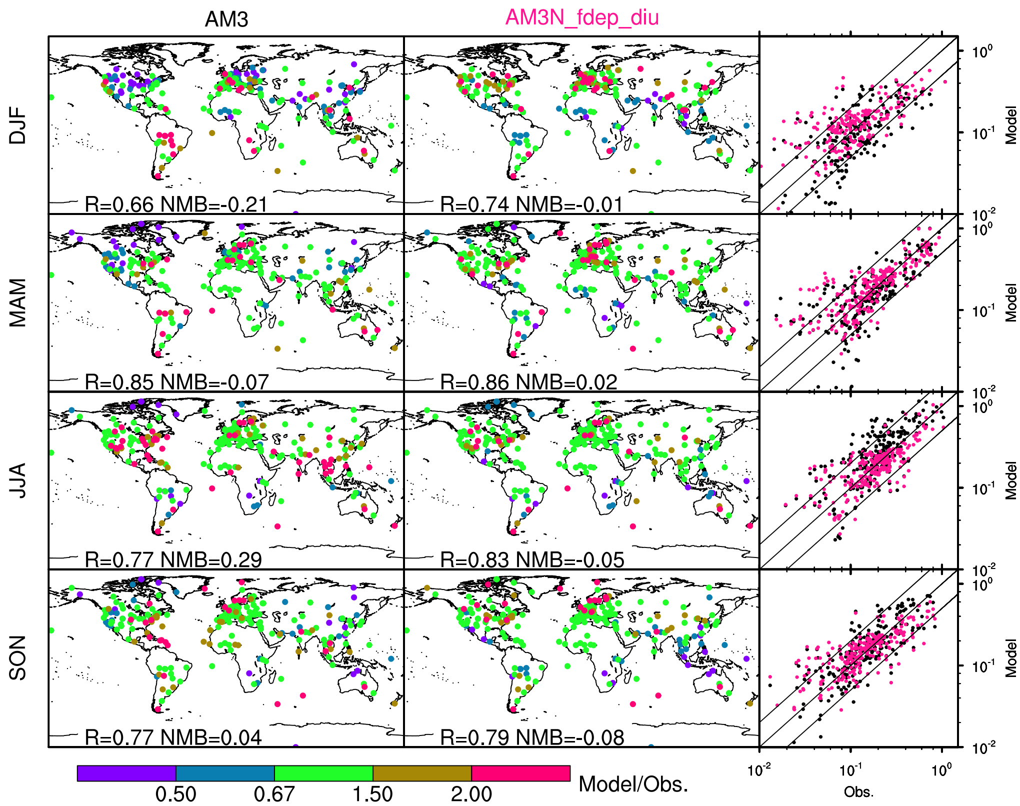

Figure S18: Same as Fig. S5 but for AOD from AERONET

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