



*Supplement of*

## **A comprehensive global modeling assessment of nitrate heterogeneous formation on desert dust**

**Rubén Soussé Villa et al.**

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## S1 Model emissions, size bins and alkalinity calculations

**Table S1.** Total emissions (anthropogenic, biogenic and biomass burning) for 2018 used in this study.

Species	Emission (Tgy-1)
<b>NO</b>	75.93
<b>NO<sub>2</sub></b>	17.86
<b>HONO</b>	0.51
<b>NH<sub>3</sub></b>	61.81
<b>SO<sub>2</sub></b>	104.40
<b>PM<sub>2.5</sub>SO<sub>4</sub></b>	0.53

**Table S2:** Density, volumetric radius, effective radius, pm2.5 and pm10 fractions of each bin of the studied species.

DUST									
	units	bin 1	bin 2	bin 3	bin 4	bin 5	bin 6	bin 7	bin 8
<b>Density</b>	kgm-3	2500	2500	2500	2500	2650	2650	2650	2650
<b>Radius vol.</b>	$\mu m$	0.15	0.25	0.47	0.8	1.36	2.29	3.93	7.24
<b>Radius eff.</b>	$\mu m$	0.15	0.25	0.45	0.78	1.32	2.24	3.80	7.11
<b>PM2.5 frac.</b>		1	1	1	1	0.38	0	0	0
<b>PM10 frac.</b>		1	1	1	1	1	1	0.87	0
SEA_SALT									
	units	bin 1	bin 2	bin 3	bin 4	bin 5	bin 6	bin 7	bin 8
<b>Density</b>	kgm-3	2160	2160	2160	2160	2160	2160	2160	2160
<b>Radius vol.</b>	$\mu m$	0.15	0.25	0.47	0.81	1.40	2.37	4.30	9.23
<b>Radius eff.</b>	$\mu m$	0.14	0.24	0.45	0.79	1.36	2.32	4.13	8.64
<b>PM2.5 frac.</b>		1	1	1	1	0.32	0	0	0
<b>PM10 frac.</b>		1	1	1	1	1	1	0.75	0
NO3									
	units	bin 1	bin 2						
<b>Density</b>	kgm-3	1700	2380						
<b>Radius vol.</b>	$\mu m$	0.35	2.23						
<b>Radius eff.</b>	$\mu m$	0.24	1.79						
<b>PM2.5 frac.</b>		1	0.2						
<b>PM10 frac.</b>		1	1						
NH4									
	units	bin 1	bin 2						
<b>Density</b>	kgm-3	1700	2380						
<b>Radius vol.</b>	$\mu m$	0.35	2.23						
<b>Radius eff.</b>	$\mu m$	0.24	1.79						
<b>PM2.5 frac.</b>		1	0.2						
<b>PM10 frac.</b>		1	1						
SO4									
	units	bin 1	bin 2						
<b>Density</b>	kgm-3	1700	2380						
<b>Radius vol.</b>	$\mu m$	0.35	2.23						
<b>Radius eff.</b>	$\mu m$	0.24	1.79						
<b>PM2.5 frac.</b>		1	0.2						
<b>PM10 frac.</b>		1	1						

Mineral	Bin 1	Bin 2	Bin 3	Bin 4	Bin 5	Bin 6	Bin 7	Bin 8	Soluble in water?	Soluble in acids?	Assumed reactive?	Notes/References
<b>Illite</b>	8.5E-5	4.1E-4	3.1E-3	8.8E-3	2.2E-2	3.2E-2	5.5E-2	1.4E-2	no	yes	yes	1
<b>Montmorillonite</b>	6.2E-5	3.0E-4	2.2E-3	6.4E-3	1.6E-2	2.3E-2	4.0E-2	1.0E-2	-	yes	yes	2
<b>Kaolinite</b>	7.8E-5	3.7E-4	2.8E-3	8.0E-3	2.0E-2	2.9E-2	5.0E-2	1.3E-2	no	yes	no	3
<b>Chlorite</b>	1.2E-5	5.6E-5	4.2E-4	1.2E-3	4.8E-3	1.1E-2	2.6E-2	7.9E-3	no	-	no	4
<b>Vermiculite</b>	7.9E-6	3.8E-5	2.8E-4	8.1E-4	2.1E-3	2.9E-3	5.1E-3	1.3E-3	no	no	no	5
<b>Feldspar</b>	3.4E-6	1.6E-5	1.2E-4	3.5E-4	6.3E-3	2.2E-2	6.0E-2	1.9E-2	no	yes	yes	6
<b>Quartz</b>	1.2E-5	5.8E-5	4.4E-4	1.3E-3	1.9E-2	6.6E-2	1.8E-1	5.6E-2	no	no	no	7
<b>Calcite</b>	3.1E-5	1.5E-4	1.1E-3	3.2E-3	1.0E-2	1.9E-2	3.9E-2	1.1E-2	yes	yes	yes	8
<b>Hematite</b>	3.5E-6	1.7E-5	1.3E-4	3.6E-4	9.2E-4	1.3E-3	2.3E-3	5.9E-4	no	yes	no	9
<b>Goethite</b>	6.2E-6	2.9E-5	2.2E-4	6.3E-4	1.8E-3	3.1E-3	6.2E-3	1.8E-3	no	-	no	10
<b>Gypsum</b>	4.7E-7	2.2E-6	1.7E-5	4.8E-5	1.4E-4	2.5E-4	5.2E-4	1.5E-4	poor	yes	no	11
<b>Mica</b>	0.0E+0	0.0E+0	0.0E+0	0.0E+0	2.7E-3	1.0E-2	2.8E-2	9.0E-3	no	no	no	12
<b>Total</b>	3.0E-4	1.4E-3	1.1E-2	3.1E-2	1.1E-1	2.2E-1	4.9E-1	1.5E-1				

**Table S3.** Globally-averaged fractions (0 to 1) of the minerals attributed to each bin from the Journet et al. (2014) mineral database. The respective references for the assumed solubility of each mineral are:

1. Assumed reactive because relatively soluble in acids (Webmineral, 2008f)
2. Assumed reactive because relatively soluble in acids (Nutting, 1941, 1932)
3. Assumed nonreactive because predominantly containing Al and Si (Yang, 2008).
4. Assumed nonreactive because not soluble in water (Britannica, The Editors of Encyclopaedia, 2018; Mindat.org, a).
5. Assumed nonreactive because not soluble (Huggett, 2015; Schulze, 2005; Bleam, 2017).
6. Assumed reactive because relatively soluble in acids (Blum, 1994; Mindat.org, b).
7. Assumed nonreactive because not soluble (Usher et al., 2003).
8. Assumed reactive because highly soluble (Usher et al., 2003; Krueger et al., 2004; Hodzic et al., 2006).
9. Assumed nonreactive because iron is not considered in ISORROPIA-II (National Center for Biotechnology Information, 2024a; Weast, 1980).
10. Assumed nonreactive because not soluble, although lacking information if soluble in acids. (Essington et al., 2005; National Center for Biotechnology Information. PubChem Compound Database, 2024b).
11. Assumed nonreactive because poorly soluble in water and soluble only in high acidity solutions (Zhang and Muhammed, 1989; National Center for Biotechnology Information. PubChem Compound Database, 2024a; Krueger et al., 2004; Hodzic et al., 2006).
12. Assumed nonreactive because non soluble (National Center for Biotechnology Information, 2024b).

Mineral	Mineral Mw (g/mol)	Soluble element	Element Mw (g/mol)	Element fraction	Bin 1	Bin 2	Bin 3	Bin 4	Total fine	Bin 5	Bin 6	Bin 7	Bin 8	Total coarse	Total	References
Illite	389.34	K	39.10	0.10	2.84%	2.84%	2.84%	2.84%	2.10%	1.45%	1.13%	0.99%	1.41%	2.20%	1	
	389.34	Mg	24.31	0.06	1.76%	1.76%	1.76%	1.76%	1.30%	0.90%	0.70%	0.62%	0.88%	1.37%	1	
<b>Montmorillonite</b>	549.07	Ca	40.08	0.07	1.50%	1.50%	1.50%	1.50%	1.11%	0.76%	0.59%	0.52%	0.75%	1.16%	2	
	549.07	Na	22.99	0.04	0.86%	0.86%	0.86%	0.86%	0.63%	0.44%	0.34%	0.30%	0.43%	0.67%	2	
<b>Feldspar</b>																
-13 Albite	263.02	Na (1/3)	22.99	0.03	0.03%	0.03%	0.03%	0.03%	0.16%	0.27%	0.32%	0.35%	0.27%	0.14%	3	
-1/3 Anorthite	277.41	Ca (1/3)	40.08	0.04	0.05%	0.05%	0.05%	0.05%	0.26%	0.44%	0.53%	0.57%	0.45%	0.23%	4	
-1/3 Orthoclase	278.33	K (1/3)	39.10	0.04	0.05%	0.05%	0.05%	0.05%	0.25%	0.43%	0.52%	0.56%	0.44%	0.22%	5	
Calcite	100.08	Ca	40.08	0.40	4.15%	4.15%	4.15%	4.15%	3.76%	3.41%	3.23%	3.16%	3.39%	3.81%	6	
Gypsum	172.20	Ca	40.08	0.23	0.04%	0.04%	0.04%	0.04%	0.04%	0.03%	0.03%	0.02%	0.02%	0.03%	0.03%	7
<b>Total Ca</b>					5.73%	5.73%	5.73%	5.73%	5.15%	4.64%	4.39%	4.28%	4.61%	5.17%		
<b>Total Na</b>					0.89%	0.89%	0.89%	0.89%	0.89%	0.79%	0.71%	0.66%	0.65%	0.70%	0.79%	
<b>Total K</b>					2.88%	2.88%	2.88%	2.88%	2.88%	2.35%	1.88%	1.64%	1.55%	1.85%	2.37%	
<b>Total Mg</b>					1.76%	1.76%	1.76%	1.76%	1.76%	1.30%	0.90%	0.70%	0.62%	0.88%	1.32%	

**Table S4.** The NVC content from globally-averaged mineralogy of Journet et al. (2014) assumed for each size bin, derived from those minerals assumed to be soluble in water (Supplementary Table S3). The respective references for the ionic composition of each mineral and molar masses are:

1. Webmineral (2008f)
2. Nutting (1941, 1932)
3. Webmineral (2008a)
4. Webmineral (2008b)
5. Webmineral (2008e)
6. Webmineral (2008c); National Center for Biotechnology Information. PubChem Compound Database (2024a)
7. Webmineral (2008d); Hulett and Allen (1902)

Mineral	Bin 1	Bin 2	Bin 3	Bin 4	Bin 5	Bin 6	Bin 7	Bin 8	Soluble in water?	Soluble in acids?	Assumed reactive?	Notes/ References
<b>Illite</b>	1.1E-04	5.4E-04	4.0E-03	1.2E-02	2.9E-02	4.2E-02	7.2E-02	1.9E-02	no	yes	yes	1
<b>Montmorillonite</b>	6.4E-05	3.0E-04	2.3E-03	6.5E-03	1.7E-02	2.4E-02	4.1E-02	1.1E-02	-	yes	yes	2
<b>Kaolinite</b>	7.6E-05	3.6E-04	2.7E-03	7.8E-03	2.0E-02	2.8E-02	4.8E-02	1.3E-02	no	yes	no	3
<b>Calcite</b>	1.3E-05	6.3E-05	4.7E-04	1.4E-03	5.2E-03	1.2E-02	2.7E-02	8.0E-03	yes	yes	yes	4
<b>Quartz</b>	2.0E-05	9.7E-05	7.3E-04	2.1E-03	2.4E-02	8.0E-02	2.1E-01	6.7E-02	no	no	no	5
<b>Feldspar</b>	8.1E-06	3.9E-05	2.9E-04	8.3E-04	8.5E-03	2.8E-02	7.3E-02	2.3E-02	no	yes	yes	6
<b>Iron Oxide</b>	4.6E-06	2.2E-05	1.6E-04	4.7E-04	1.6E-03	3.3E-03	7.3E-03	2.2E-03	-	-	no	7
<b>Gypsum</b>	3.4E-06	1.6E-05	1.2E-04	3.5E-04	1.3E-03	2.8E-03	6.3E-03	1.9E-03	poor	yes	no	8
<b>Total</b>	3.0E-04	1.4E-03	1.1E-02	3.1E-02	1.1E-01	2.2E-01	4.9E-01	1.4E-01				

**Table S5.** Globally-averaged fractions (0 to 1) of the minerals attributed to each bin from the Claequin et al. (1999) mineral database. The respective references for the assumed solubility of each mineral are: References:

1. Assumed reactive because relatively soluble in acids (Webmineral, 2008f).
2. Assumed reactive because relatively soluble in acids (Nutting, 1941, 1932).
3. Assumed nonreactive because predominantly containing Al and Si (Yang, 2008).
4. Assumed reactive because highly soluble (Usher et al., 2003; Krueger et al., 2004; Hodzic et al., 2006).
5. Assumed nonreactive because not soluble (Usher et al., 2003).
6. Assumed reactive because relatively soluble in acids (Blum, 1994; Mindat.org, b).
7. Assumed nonreactive because iron is not considered in ISORROPIA-II (National Center for Biotechnology Information, 2024a; Weast, 1980).
8. Assumed nonreactive because poorly soluble in water and soluble only in high acidity solutions (Zhang and Muhammed, 1989; National Center for Biotechnology Information. PubChem Compound Database, 2024a; Krueger et al., 2004; Hodzic et al., 2006).

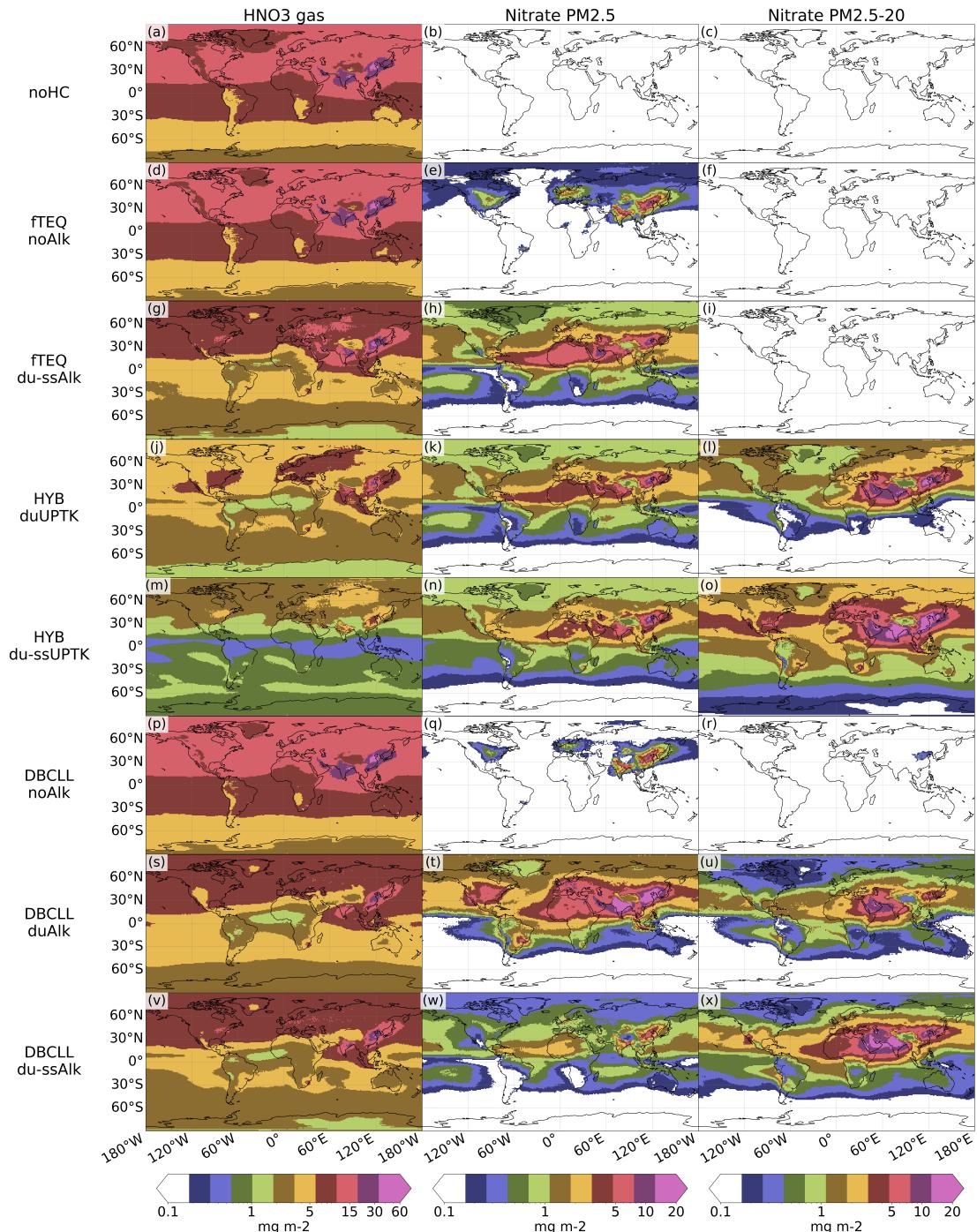
Mineral	Mineral Mw (g/mol)	Soluble element	Element fraction Mw (g/mol)	Element Bin 1	Bin 2	Bin 3	Bin 4	Total fine	Bin 5	Bin 6	Bin 7	Bin 8	Total coarse	Total	References
<b>Illite</b>	389.340	K	39.10	0.10	3.76%	3.76%	3.76%	3.76%	2.78%	1.92%	1.49%	1.31%	1.88%	2.92%	1
	389.340	Mg	24.31	0.06	2.34%	2.34%	2.34%	2.34%	1.73%	1.19%	0.93%	0.82%	1.17%	1.82%	1
<b>Montmorillonite</b>	549.070	Ca	40.08	0.07	1.54%	1.54%	1.54%	1.54%	1.14%	0.78%	0.61%	0.54%	0.77%	1.20%	2
	549.070	Na	22.99	0.04	0.88%	0.88%	0.88%	0.88%	0.88%	0.65%	0.45%	0.35%	0.31%	0.44%	0.69%
<b>Feldspar</b>															
-1/3 Albite	263.020	Na (1/3)	22.99	0.03	0.07%	0.07%	0.07%	0.07%	0.21%	0.33%	0.39%	0.42%	0.34%	0.19%	3
-1/3 Anorthite	277.410	Ca (1/3)	40.08	0.04	0.12%	0.12%	0.12%	0.12%	0.35%	0.55%	0.65%	0.70%	0.56%	0.31%	4
-1/3 Orthoclase	278.330	K (1/3)	39.10	0.04	0.11%	0.11%	0.11%	0.11%	0.34%	0.54%	0.63%	0.68%	0.55%	0.31%	5
Calcite	100.090	Ca	40.08	0.40	1.76%	1.76%	1.76%	1.76%	1.95%	2.11%	2.19%	2.22%	2.12%	1.92%	6
<b>Gypsum</b>															
Total Ca				0.23	0.26%	0.26%	0.26%	0.26%	0.26%	0.28%	0.30%	0.31%	0.30%	0.28%	7
Total Na					0.95%	0.95%	0.95%	0.95%	3.68%	3.68%	3.71%	3.74%	3.76%	3.76%	3.71%
Total K					3.87%	3.87%	3.87%	3.87%	3.87%	3.87%	3.87%	3.87%	0.75%	0.73%	0.87%
Total Mg					2.34%	2.34%	2.34%	2.34%	2.34%	2.34%	2.34%	2.34%	1.99%	2.42%	3.19%

**Table S6.** The NVC content from globally-averaged mineralogy of Journet et al. (2014) assumed for each size bin, derived from those minerals assumed to be soluble in water (Supplementary Table S5). The respective references for the ionic composition of each mineral and molar masses are:

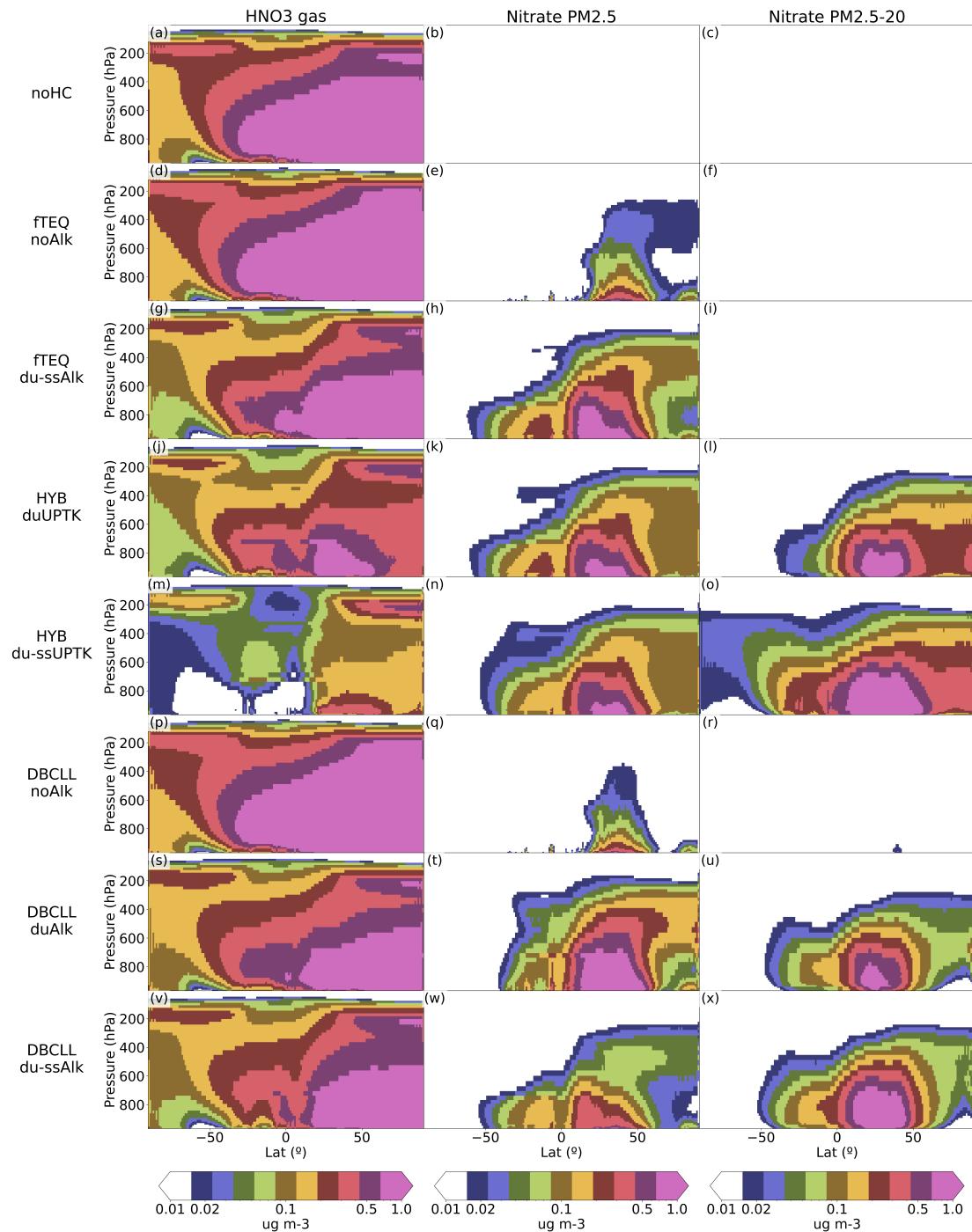
1. Webmineral (2008f)
2. Nutting (1941, 1932)
3. Webmineral (2008a)
4. Webmineral (2008b)
5. Webmineral (2008e)
6. Webmineral (2008c); National Center for Biotechnology Information. PubChem Compound Database (2024a)
7. Webmineral (2008d); Hulett and Allen (1902)



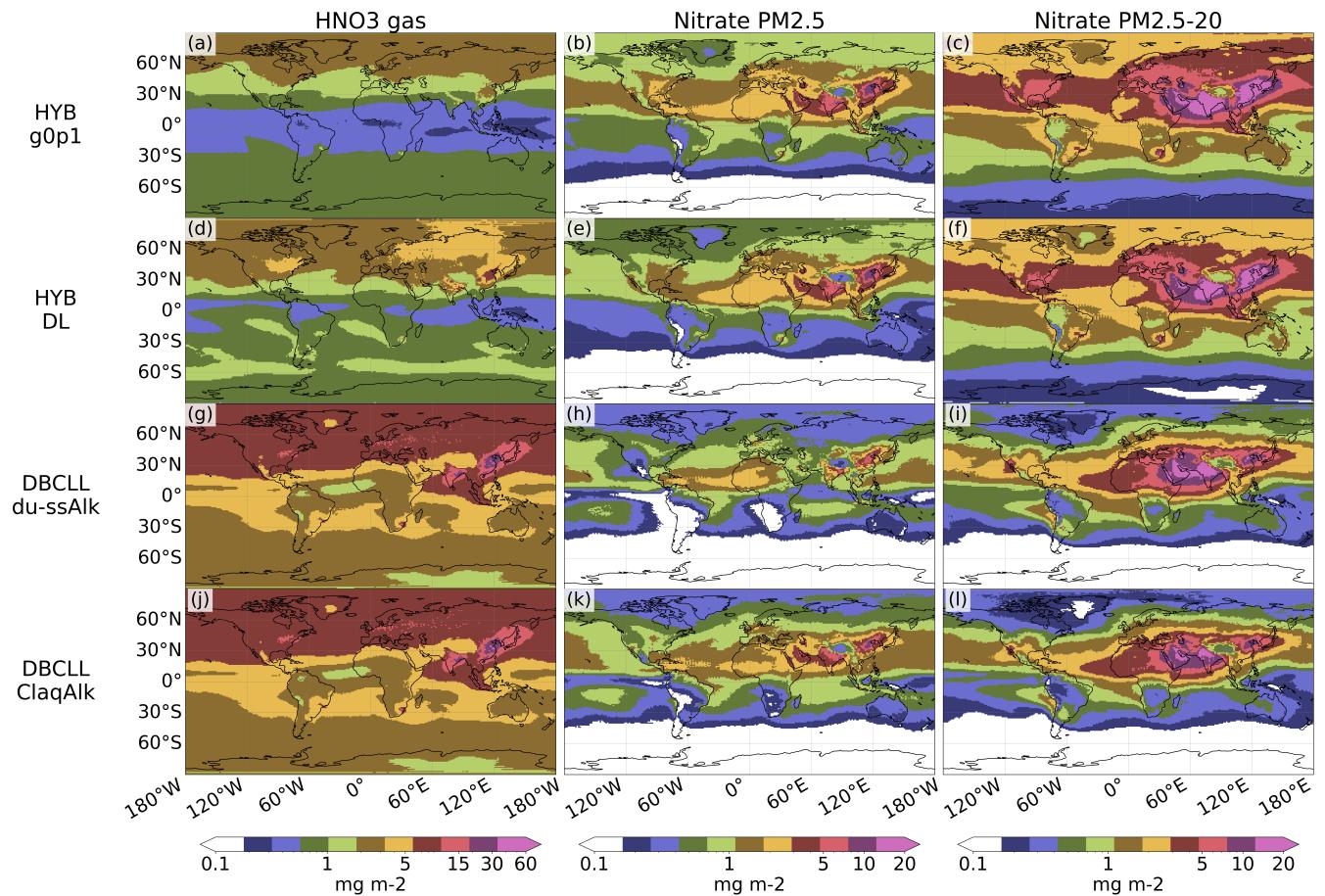
## S2 Spatial distributions



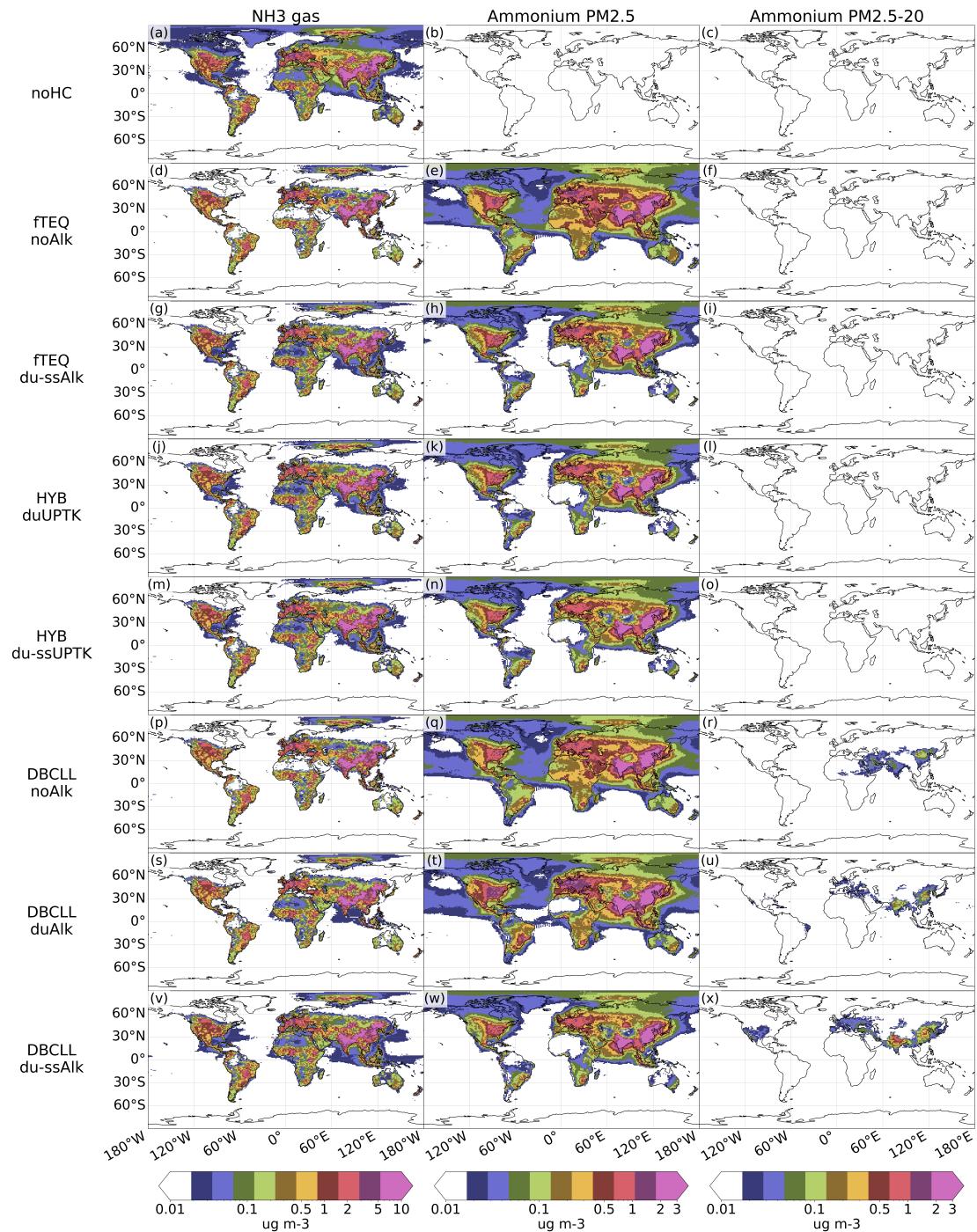
**Figure S1.** Column loads ( $mg\ m^{-2}$ ) of  $HNO_3(g)$ , fine and coarse particulate nitrate simulated by the different mechanisms, averaged for 2018.



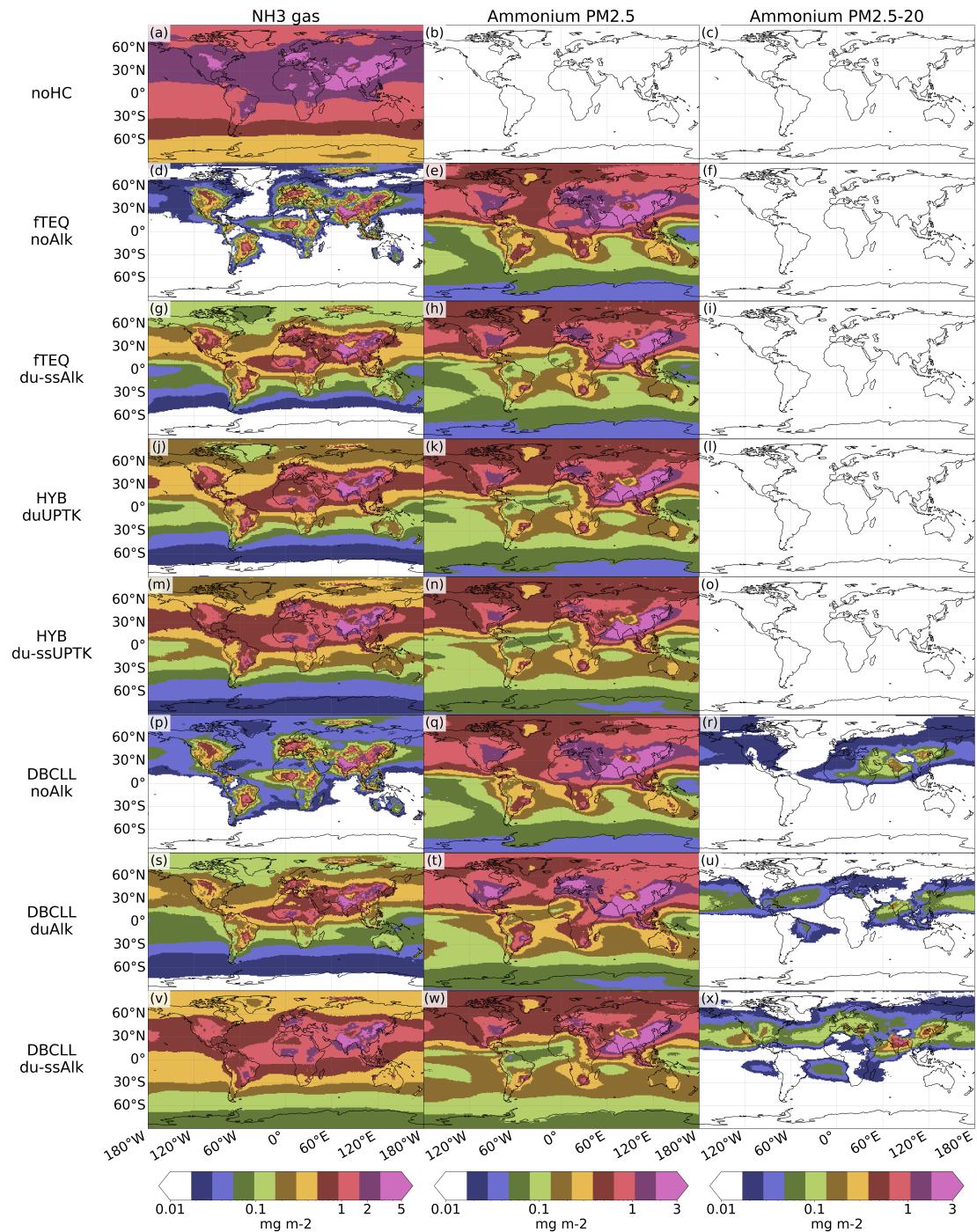
**Figure S2.** Zonal average concentrations ( $\mu\text{g m}^{-3}$ ) of  $\text{HNO}_3$ , fine and coarse particulate nitrate simulated by the different mechanisms, averaged for 2018.



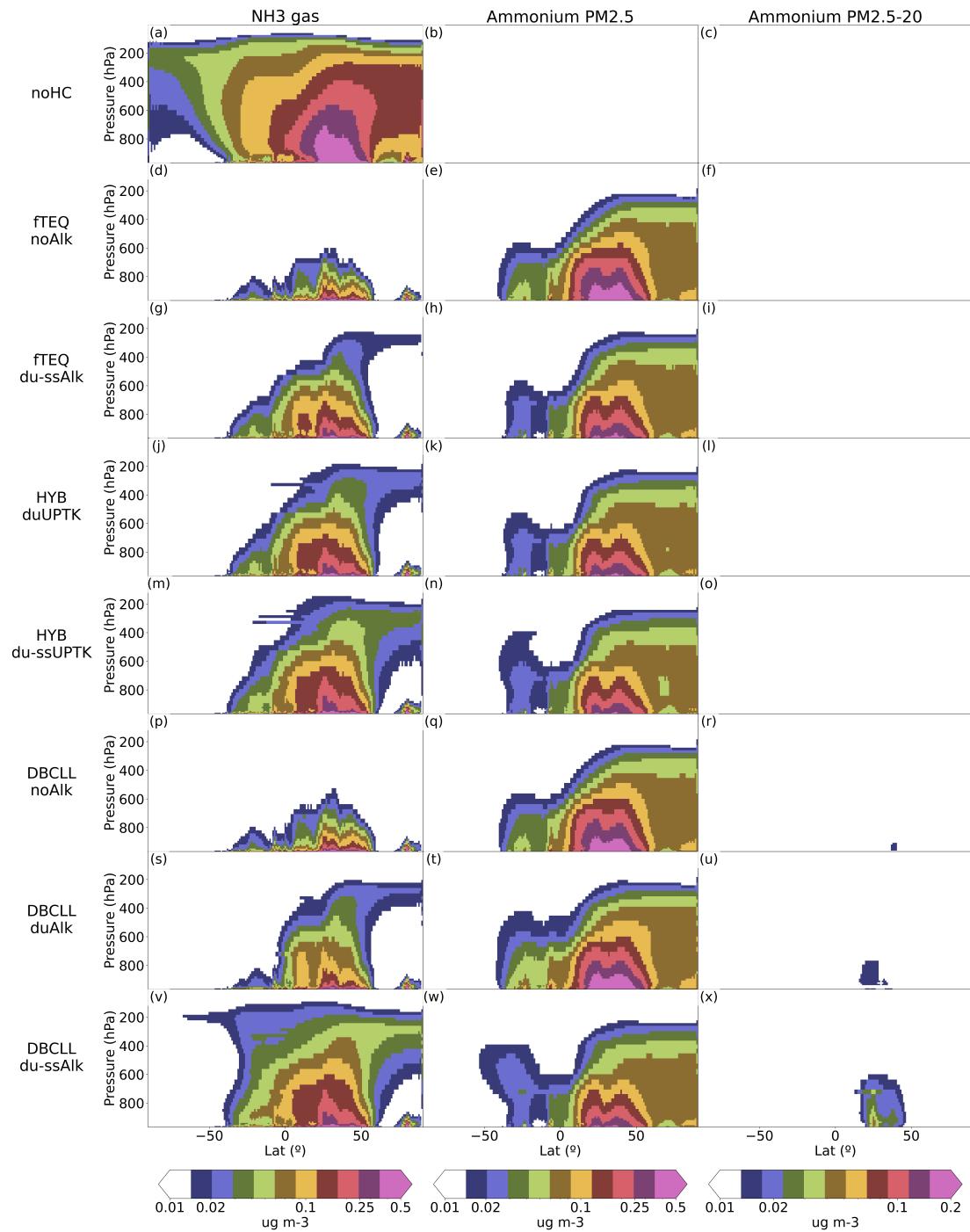
**Figure S3.** Column loads ( $\text{mg m}^{-2}$ ) of  $\text{HNO}_{3(g)}$ , fine and coarse particulate nitrate simulated by additional mechanisms, averaged for 2018.



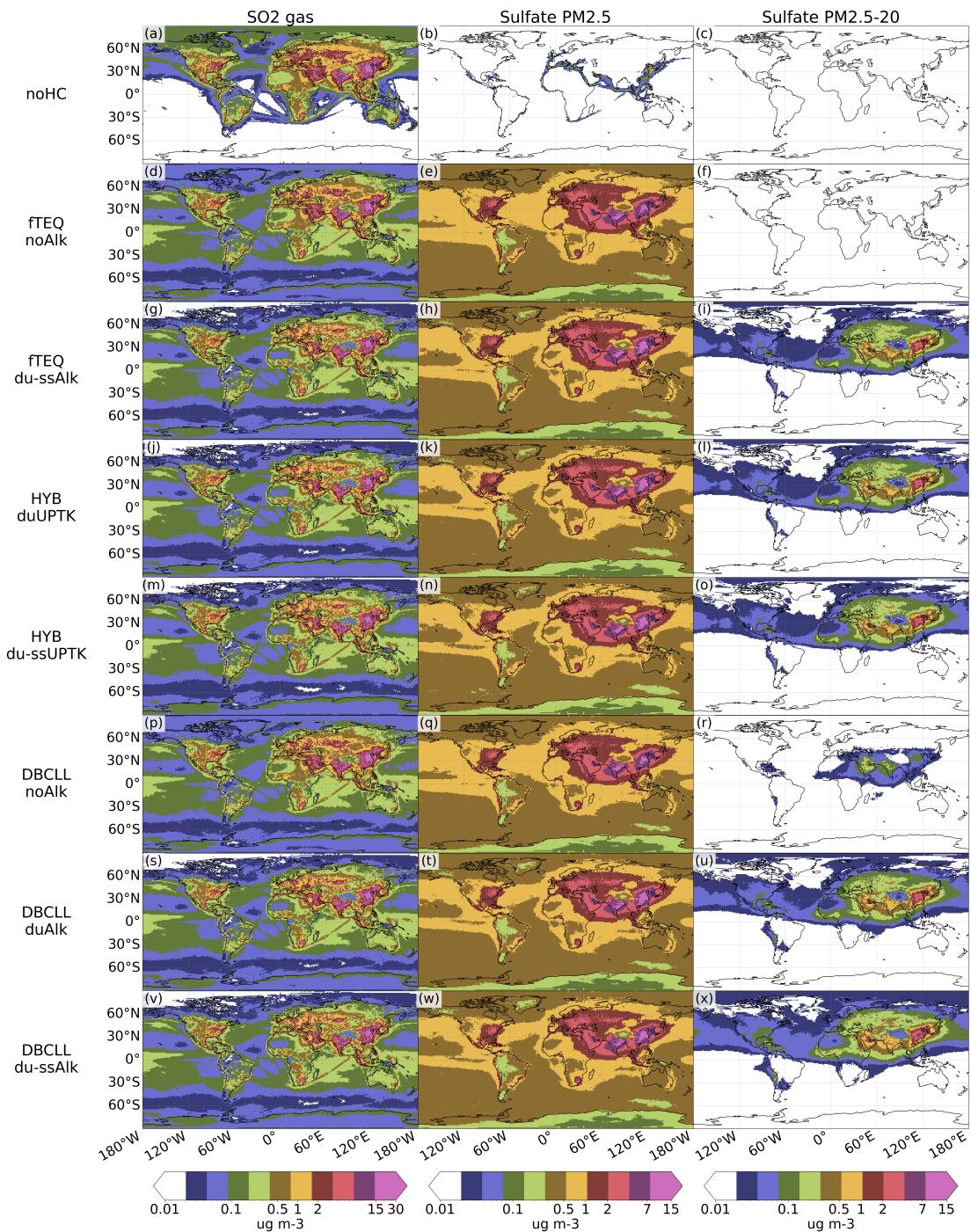
**Figure S4.** Surface concentrations ( $\mu\text{g m}^{-3}$ ) of  $\text{NH}_3(g)$ , fine and coarse particulate ammonium simulated by the different mechanisms, averaged for 2018.



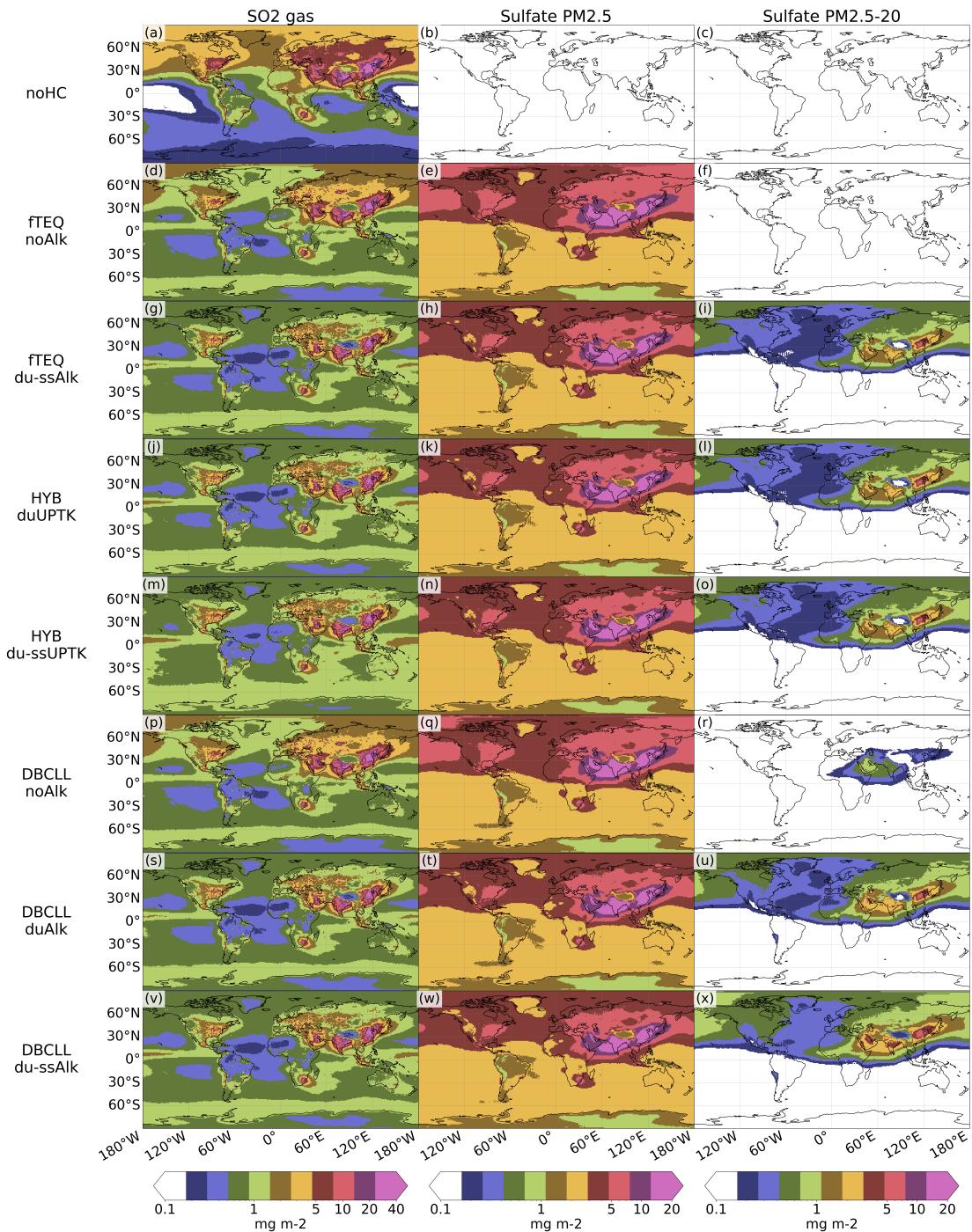
**Figure S5.** Column loads ( $mg\ m^{-2}$ ) of  $NH_3(g)$ , fine and coarse particulate ammonium simulated by different mechanisms, averaged for 2018.



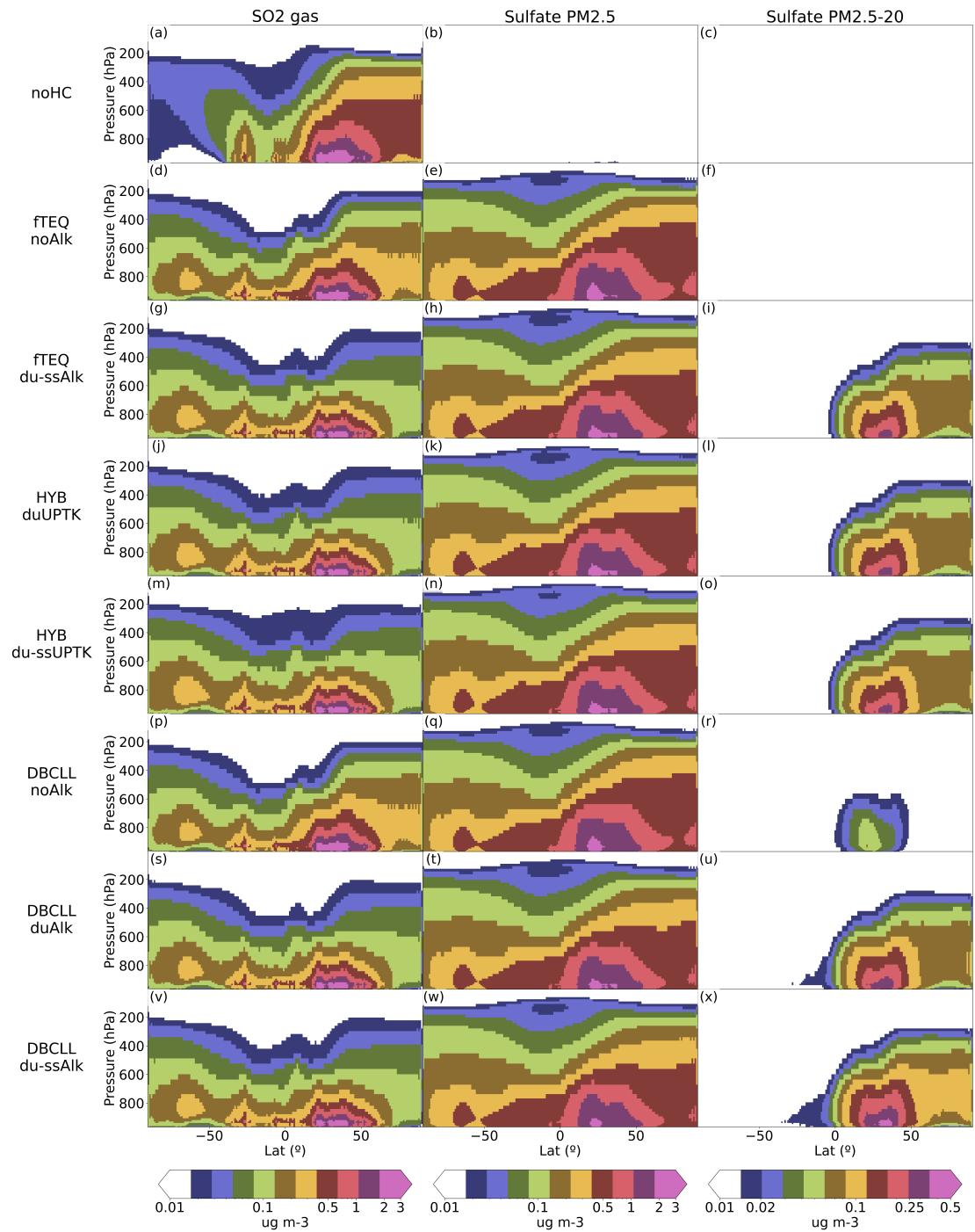
**Figure S6.** Zonal average concentrations ( $\mu\text{g m}^{-3}$ ) of  $\text{NH}_3(g)$ , fine and coarse particulate ammonium simulated by the different mechanisms, averaged for 2018.



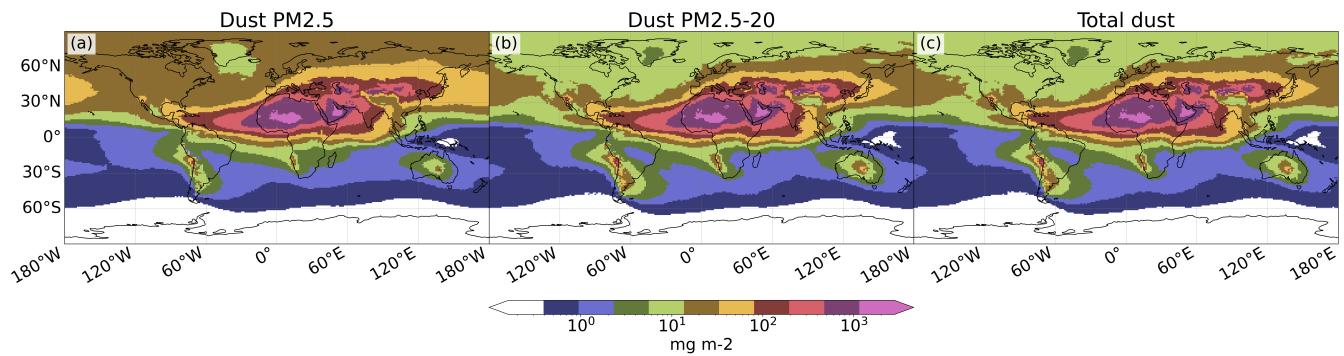
**Figure S7.** Surface concentration ( $\mu\text{g m}^{-3}$ ) of  $\text{SO}_{2(\text{g})}$ , fine and coarse particulate sulfate  $\text{SO}_4^{2-}$  simulated by the different mechanisms, averaged for 2018.



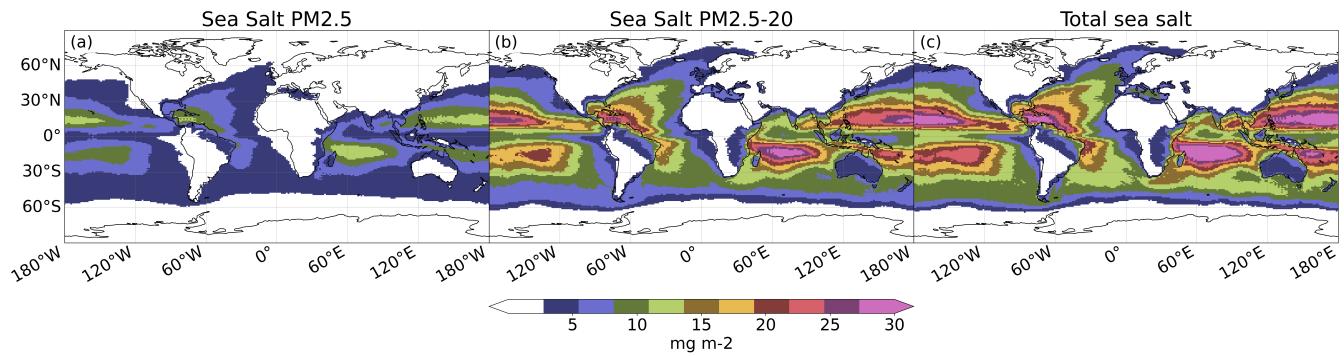
**Figure S8.** Column load ( $\text{mg m}^{-2}$ ) of  $\text{SO}_{2(\text{g})}$ , fine and coarse particulate sulfate  $\text{SO}_4^{2-}$  simulated by the different mechanisms, averaged for 2018.



**Figure S9.** Zonal average concentration ( $\mu\text{g m}^{-3}$ ) of  $\text{SO}_{2(g)}$ , fine and coarse particulate sulfate  $\text{SO}_4^{2-}$  simulated by the different mechanisms, averaged for 2018.



**Figure S10.** Dust average column load ( $mgm^{-2}$ ) for 2018.



**Figure S11.** Sea salt average column load ( $mgm^{-2}$ ) for 2018.

### S3 Statistical metrics and supplementary evaluation results

Each experiment is evaluated against observations in terms of correlation coefficient (corr), mean bias (bias) and root mean

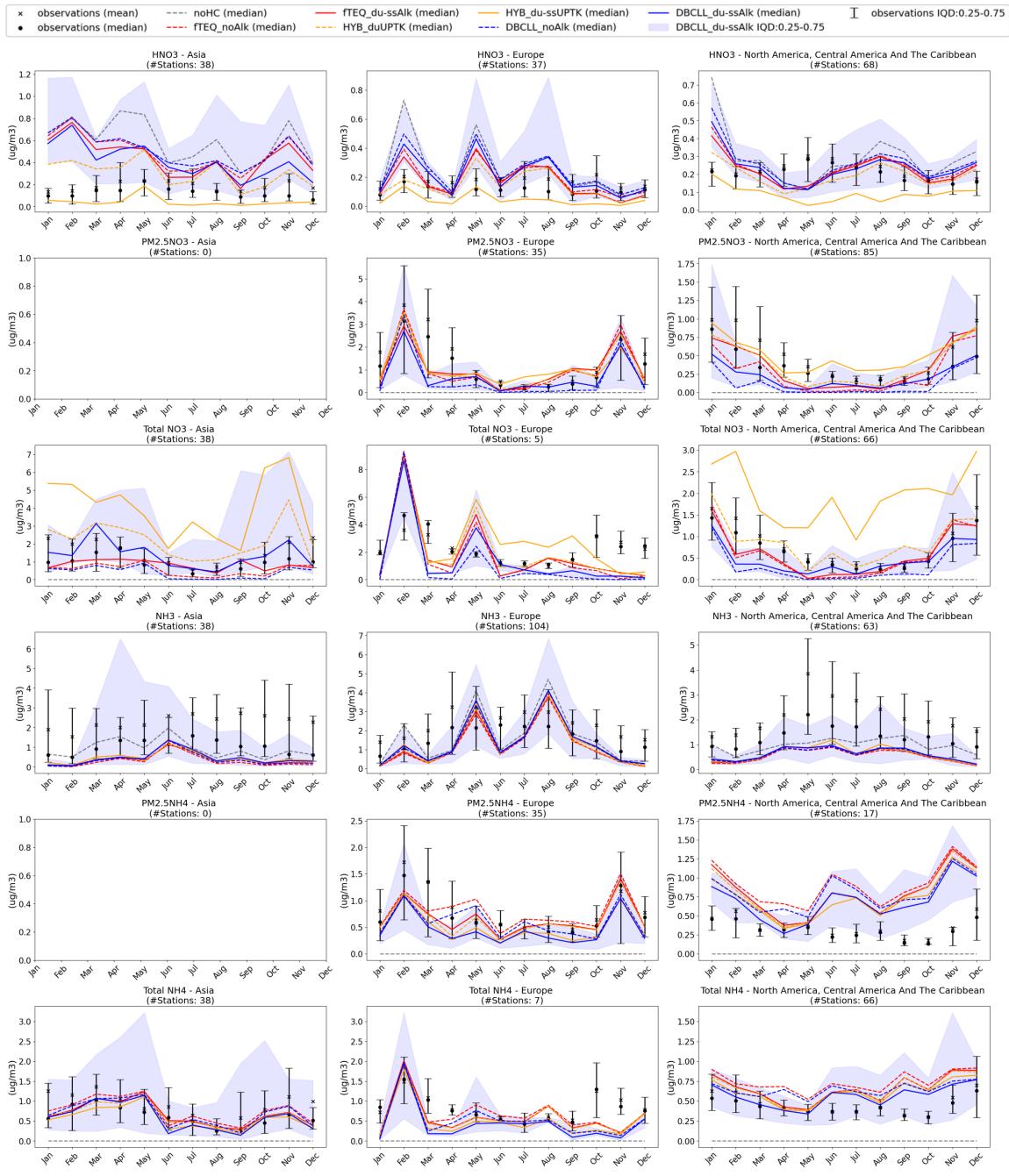
5 square error (rmse), formulated as follows:

$$corr = \frac{\sum_{i=1}^N (S_i - \bar{S})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^N (S_i - \bar{S})^2} \sqrt{\sum_{i=1}^N (O_i - \bar{O})^2}} \quad (1)$$

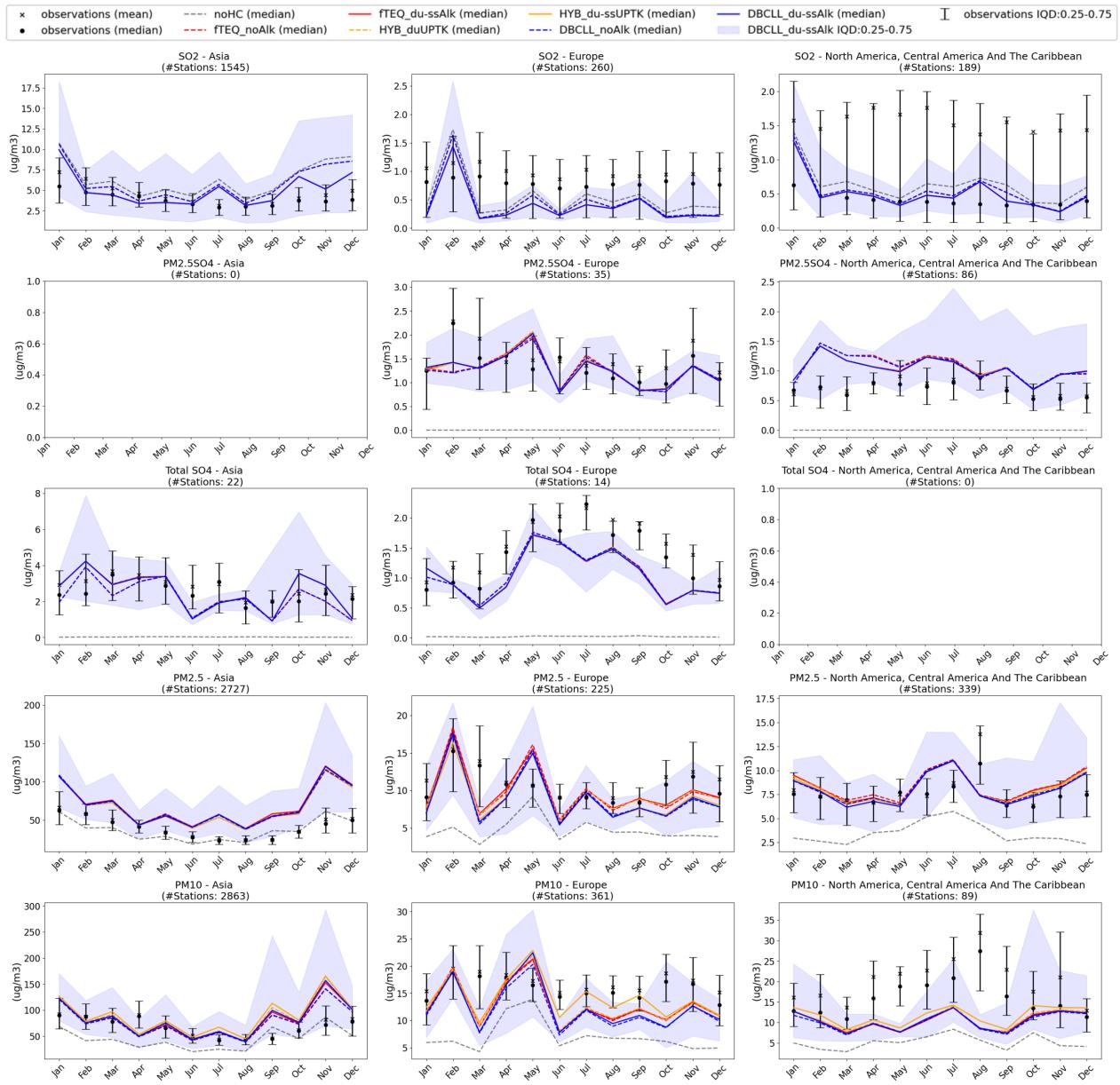
$$bias = \frac{\sum_{i=1}^N (S_i - O_i)}{N} \quad (2)$$

$$rmse = \sqrt{\frac{\sum_{i=1}^N (S_i - O_i)^2}{N}} \quad (3)$$

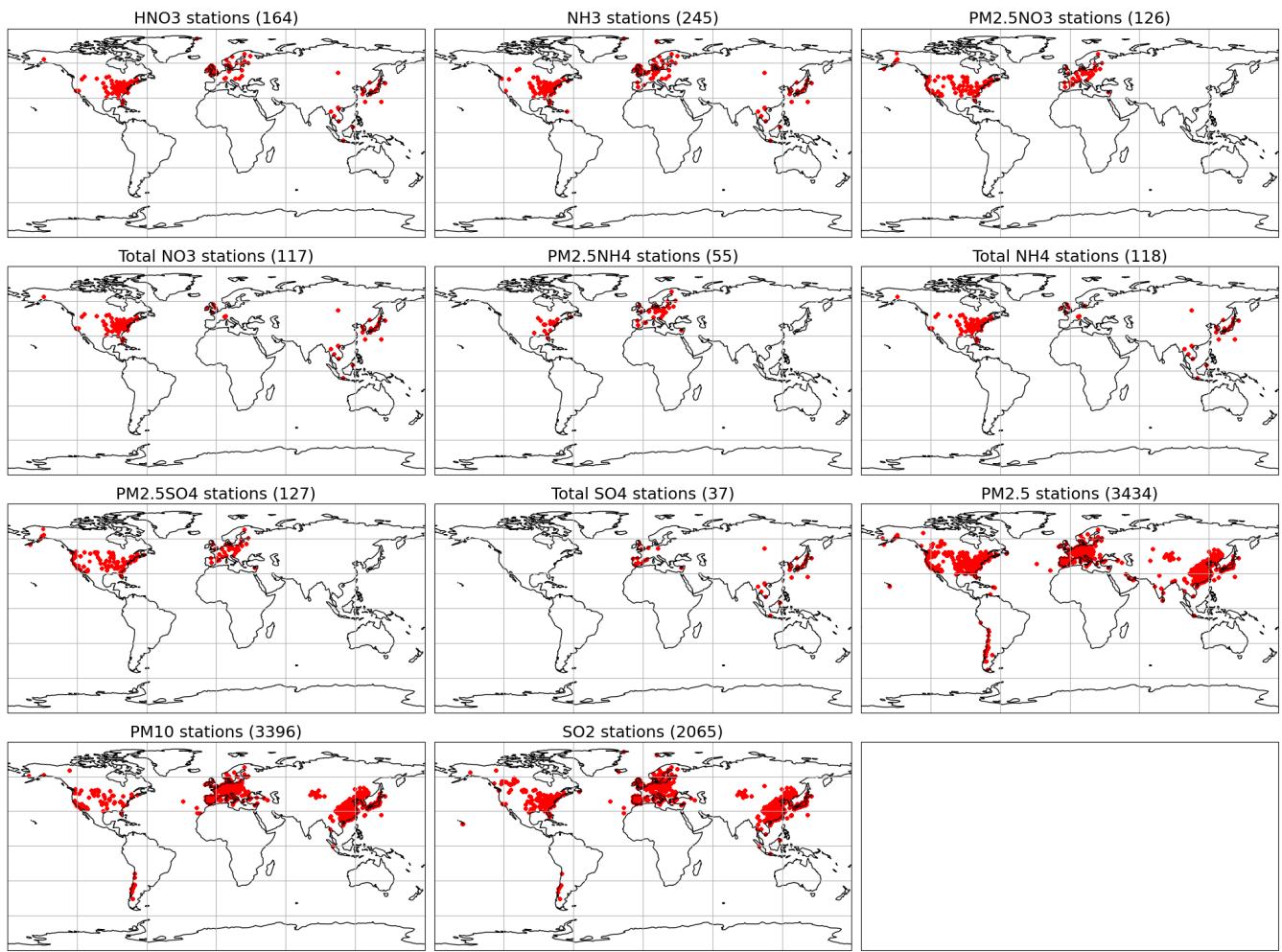
Where  $S$  are the simulation results,  $O$  the observations and  $N$  the number of observations.



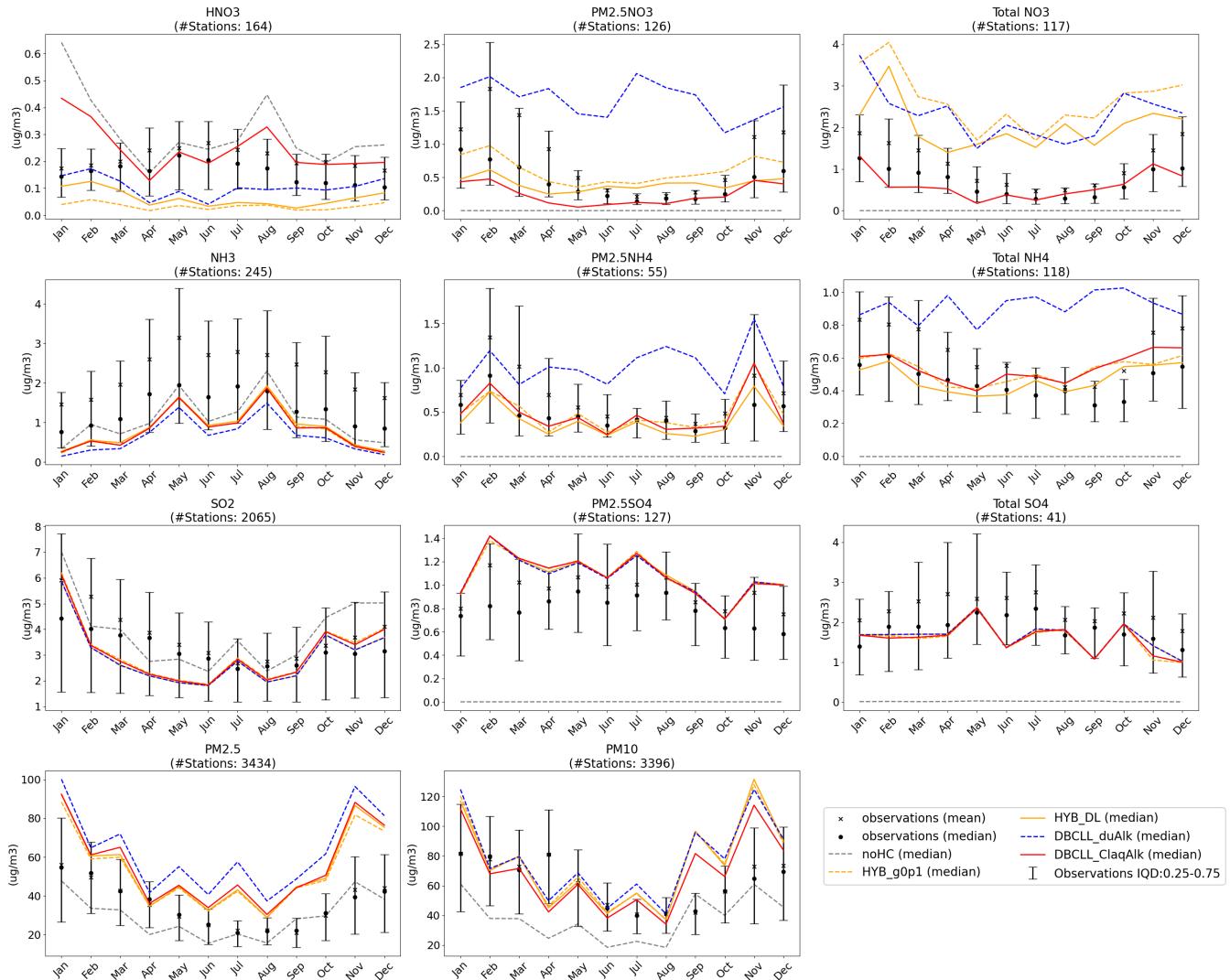
**Figure S12.** Yearly surface concentration evaluation for Asia, Europe and North-Central America continents. Species are in rows and continents in columns. Black solid dots and crosses represent observational mean and median, respectively. Error bars are the interquantile 0.25 to 0.75 distance. Coloured lines represent medians obtained from the most representative configurations. Blue shade is the interquantile 0.25 - 0.75 distance for the DBCLL\_du-ssAlk simulation. Observational PM<sub>10</sub> from EANET (Asia) and US-EPA-CASTNET (North America) refers to total particle concentration, while GHOST data (Europe) is limited to PM<sub>10</sub>. (Continues in the next page)



**Figure S12.** (Continuation) Yearly surface concentration evaluation for Asia, Europe and North-Central America continents. Species are in rows and continents in columns. Black solid dots and crosses represent observational mean and median, respectively. Error bars are the interquartile 0.25 to 0.75 distance. Coloured lines represent medians obtained from the most representative configurations. Blue shade is the interquartile 0.25 - 0.75 distance for the DBCLL\_du-ssAlk simulation. Observational PM<sub>10</sub> from EANET (Asia) and US-EPA-CASTNET (North America) for sulfate refers to total particle concentration, while GHOST data (Europe) is limited to PM<sub>10</sub>.



**Figure S13.** Stations used in the observational evaluation for each species from the main text Fig. 5 and Supplement Fig. S12.



**Figure S14.** Observational evaluation of gas and particulate species of additional sensitivity experiments. Black solid dots and crosses represent monthly mean and median of observations, respectively. Colour lines represent each configuration's monthly median surface concentrations over observational points. Error bars are the observational interquantile 0.25 to 0.75 distance. For the Total NO<sub>3</sub> modes, data from Europe, Asia and North-Central America has been averaged, despite data from Asia and North-Central America refers to total particle concentration, while data from Europe is limited strictly to 10  $\mu\text{m}$  particle diameter.

**Table S7:** Correlation coefficients, bias ( $\mu\text{g m}^{-3}$ ) and root mean square error (rmse,  $\mu\text{g m}^{-3}$ ) of additional sensitivity experiments. The evaluation corresponds to the configurations' median with respect to the median of observations, corresponding to the timeseries shown in Fig. S14.

	HNO3			PM2.5NO3			Total NO3		
	corr	bias	rmse	corr	bias	rmse	corr	bias	rmse
<b>noHC</b>	0.03	0.15	0.20	0.00	-0.43	0.49	0.00	-0.70	0.77
<b>HYB_g0p1</b>	0.05	-0.13	0.13	0.83	0.18	0.23	0.81	1.96	2.01
<b>HYB_DL</b>	-0.08	-0.10	0.11	0.61	-0.03	0.21	0.53	1.32	1.40
<b>DBCCLL_duAlk</b>	-0.34	-0.05	0.08	0.21	1.24	1.28	0.80	1.61	1.65
<b>DBCCLL_ClaqAlk</b>	0.14	0.09	0.12	0.82	-0.18	0.24	0.81	-0.09	0.22

	NH3			PM2.5NH4			Total NH4		
	corr	bias	rmse	corr	bias	rmse	corr	bias	rmse
<b>noHC</b>	0.80	-0.29	0.44	0.00	-0.48	0.51	0.00	-0.45	0.46
<b>HYB_g0p1</b>	0.85	-0.53	0.59	0.64	0.01	0.16	0.47	0.07	0.11
<b>HYB_DL</b>	0.85	-0.49	0.56	0.77	-0.09	0.14	0.50	0.02	0.09
<b>DBCCLL_duAlk</b>	0.88	-0.71	0.73	0.27	0.53	0.58	-0.46	0.46	0.48
<b>DBCCLL_ClaqAlk</b>	0.85	-0.53	0.59	0.70	-0.01	0.16	0.50	0.09	0.12

	SO2			PM2.5SO4			Total SO4		
	corr	bias	rmse	corr	bias	rmse	corr	bias	rmse
<b>noHC</b>	0.61	0.65	1.24	0.59	-0.79	0.80	0.62	-1.91	1.93
<b>HYB_g0p1</b>	0.62	-0.12	0.94	0.59	0.29	0.32	0.53	-0.42	0.54
<b>HYB_DL</b>	0.63	-0.29	0.91	0.57	0.29	0.33	0.60	-0.39	0.50
<b>DBCCLL_duAlk</b>	0.63	-0.29	0.91	0.55	0.29	0.32	0.59	-0.38	0.49
<b>DBCCLL_ClaqAlk</b>	0.62	-0.16	0.94	0.57	0.29	0.33	0.55	-0.41	0.52

	PM2.5			PM10		
	corr	bias	rmse	corr	bias	rmse
<b>noHC</b>	0.75	-5.62	9.55	0.40	-23.16	28.42
<b>HYB_g0p1</b>	0.76	18.02	21.95	0.38	14.27	29.96
<b>HYB_DL</b>	0.76	19.44	23.81	0.36	13.87	30.41
<b>DBCCLL_duAlk</b>	0.71	28.07	31.58	0.38	16.47	30.10

**Table S7 continued from previous page**

<b>DBCLL_ClaqAlk</b>	0.76	20.75	24.88	0.43	7.45	24.20	
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**Table S8.** GHOST quality flags used for the observational evaluation.

Code	Flag	Description
0	Missing Measurement	i.e. NaN.
1	Infinite Value	Value is infinite – occurs when data values are outside of the range that <i>float32</i> data type can handle (-3.4E+38 to +3.4E+38).
2	Negative Measurement	Measurement is negative in absolute terms.
6	Invalid Data Provider Flags - GHOST Decreed	Measurements are associated with data quality flags given by the data provider which have been decreed by the GHOST project architects as being associated with substantial uncertainty/bias.
8	No Valid Data to Average	After screening by key QA flags, no valid data remains to average in the temporal window.
20	Erroneous Primary Sampling	The primary sampling is not appropriate to prepare the specific parameter for subsequent measurement.
21	Erroneous Sample Preparation	The sample preparation is not appropriate to prepare the specific parameter for subsequent measurement.
22	Erroneous Measurement Methodology	The measurement methodology used is not known to be able to measure the specific parameter.
72	Below Preferential Lower Limit of Detection	Measurement is below or equal to the preferential lower limit of detection.
75	Above Preferential Upper Limit of Detection	Measurement is above or equal to the preferential upper limit of detection.
82	Insufficient Measurement Resolution - Preferential	The preferential resolution for the measurement is coarser than a set limit (variable by measured parameter).
83	Insufficient Measurement Resolution - Empirical	The resolution of the measurement is analysed month by month. If the minimum difference between observations is coarser than a set limit (variable by measured parameter), measurements are flagged.
110	Data Outlier - Exceeds Scientifically Decreed Lower/Upper Limit	The measured value is below or greater than scientifically feasible lower/upper limits (variable by parameter).
111	Data Outlier - Monthly Median Exceeds Scientifically Decreed Upper Limit	The median of the measurements in a month is greater than a scientifically feasible limit (variable by parameter).
112	Data Outlier - Network Decreed	Data has been reported to be an outlier through data flags by the network data reporters (and not manually checked and verified as valid).
113	Data Outlier - Manually Decreed	Data has been found and decreed manually to be an outlier.
115	Probable Data Outlier - Monthly Adjusted Boxplot	Measured value exceeds adjusted boxplot outer fence (lower or upper) of monthly data, therefore is a probable data outlier.
132	Systematic Inconsistent Monthly Distributions - 4/6 Months >= Zone 6	4 out of 6 months' distributions are classed as Zone 6 or higher, suggesting there are potentially systematic reasons for the inconsistent distributions across the 6 months.
133	Systematic Inconsistent Monthly Distributions - 8/12 Months >= Zone 6	8 out of 12 months' distributions are classed as Zone 6 or higher, suggesting there are potentially systematic reasons for the inconsistent distributions across the 12 months.



10 **S4 Budget tables**

**Table S9.** Results for gas  $\text{HNO}_{3(g)}$  obtained with the studied heterogeneous chemistry mechanisms. Results from the literature references are reported at the end of the table: The average of all the participating models in the intercomparison AeroCom phase III nitrate experiment for 2008 (*AeroCom*), specifying their standard deviation (*STD AeroCom*), results from the GMI model (*GMI*, using a similar HYB approach with UPTK reactions on dust and SS), and results from the EMAC model (*EMAC 2008*, using a similar approach to DBCLL\_du-ssAlk). Also using the EMAC model, results obtained by the Karydis et al. (2016) study are reported as *EMAC 2005-2008*. Results from models using a similar approach to HYB\_du-ssUPTK are reported as *IFS* for Rémy et al. (2022), *LMDz-INCA* for Hauglustaine et al. (2014) and *MetOffice UM* for Jones et al. (2021).

$\text{HNO}_{3(g)}$	Burden (Tg)	Wet Dep. (Tg y-1)	Dry Dep. (Tg y-1)	Total Dep. (Tg y-1)	Production (Tg y-1)	Lifetime (days)
<b>noHC</b>	4.79	47.1	110.4	157.5	158.3	5.5
<b>fTEQ_noAlk</b>	4.99	48.6	107.3	155.9	156.7	5.8
<b>fTEQ_du-ssAlk</b>	2.93	41.4	77.4	118.8	119.3	4.5
<b>HYB_duUPTK</b>	1.96	29.9	52.0	81.9	82.4	4.4
<b>HYB_du-ssUPTK</b>	0.69	7.2	13.7	20.9	21.4	5.9
<b>HYB_gDU=0.1</b>	0.46	5.9	5.9	11.8	12.2	7.1
<b>HYB_DL</b>	0.72	6.7	14.0	20.7	21.2	6.2
<b>DBCLL_noAlk</b>	5.13	49.1	109.1	158.2	159.0	5.9
<b>DBCLL_duAlk</b>	2.72	45.4	59.9	105.3	105.7	4.7
<b>DBCLL_du-ssAlk</b>	2.53	36.1	61.9	98.0	98.6	4.7
<b>DBCLL_ClaqAlk</b>	2.58	36.2	62.1	98.3	98.9	4.8
<b>AeroCom</b>	<b>2.50</b>	<b>108.7</b>	<b>45.8</b>	<b>154.5</b>	<b>179.0</b>	<b>4.6</b>
<b>STD AeroCom</b>	<b><math>\pm 1.83</math></b>	<b><math>\pm 39.8</math></b>	<b><math>\pm 13.0</math></b>	<b><math>\pm 52.8</math></b>	<b><math>\pm 89.9</math></b>	<b><math>\pm 1.6</math></b>
<b>GMI</b>	<b>2.50</b>	<b>108.7</b>	<b>45.8</b>	<b>154.5</b>	<b>110.0</b>	<b>3.5</b>
<b>EMAC 2008<sup>1</sup></b>	<b>3.10</b>	<b>136.0</b>	<b>56.1</b>	<b>192.1</b>	-	-
<b>EMAC 2005-2008<sup>2</sup></b>	<b>1.65</b>	-	-	-	-	-
<b>IFS</b>	-	-	-	-	-	-
<b>LMDz-INCA</b>	<b>1.35</b>	<b>76.6</b>	<b>66.0</b>	<b>142.6</b>	<b>218.3</b>	<b>2.3</b>
<b>MetOffice UM<sup>3</sup></b>	<b>2.16</b>	<b>67.1</b>	<b>27.0</b>	<b>94.1</b>	<b>242.1</b>	<b>3.2</b>

<sup>1</sup> From Bian et al. (2017).

<sup>2</sup> From Karydis et al. (2016).

<sup>3</sup> Jones et al. (2021) performs two sensitivity tests to the accommodation coefficient used for  $\text{NO}_3^-$  formation in the fine mode: *FAST* with 0.193 and *SLOW* with 0.001. Here, results from the *FAST* test are reported on the basis that they present similar fine  $\text{NO}_3^-$  formation rates to our average fine nitrate results

**Table S10.** Results for gas NH<sub>3(g)</sub> obtained with the studied heterogeneous chemistry mechanisms. Results from the literature references are reported at the end of the table: The average of all the participating models in the intercomparison AeroCom phase III nitrate experiment for 2008 (*AeroCom*), specifying their standard deviation (*STD AeroCom*), results from the GMI model (*GMI*, using a similar HYB approach with UPTK reactions on dust and SS), and results from the EMAC model (*EMAC 2008*, using a similar approach to DBCLL\_du-ssAlk). Also using the EMAC model, results obtained by the Karydis et al. (2016) study are reported as *EMAC 2005-2008*. Results from models using a similar approach to HYB\_du-ssUPTK are reported as *IFS* for Rémy et al. (2022), *LMDz-INCA* for Hauglustaine et al. (2014) and *MetOffice UM* for Jones et al. (2021).

NH <sub>3(g)</sub>	Emissions (Tg y-1)	Burden (Tg)	Wet Dep. (Tg y-1)	Dry Dep. (Tg y-1)	Total Dep. (Tg y-1)	Loss (Tg y-1)	Lifetime (days)
<b>noHC</b>	61.8	0.86	7.1	54.5	61.7	0.0	5.1
<b>fTEQ_noAlk</b>	61.8	0.05	1.3	39.6	40.9	-20.9	0.3
<b>fTEQ_du-ssAlk</b>	61.8	0.16	1.8	44.2	45.9	-15.9	0.9
<b>HYB_duUPTK</b>	61.8	0.21	1.9	44.7	46.6	-15.1	1.2
<b>HYB_du-ssUPTK</b>	61.8	0.26	2.2	45.6	47.8	-14.0	1.5
<b>HYB_gDU=0.1</b>	61.8	0.28	2.2	46.0	48.3	-13.5	1.7
<b>HYB_DL</b>	61.8	0.38	2.9	46.7	49.6	-12.1	2.2
<b>DBCLL_noAlk</b>	61.8	0.06	1.6	40.4	42.0	-19.8	0.4
<b>DBCLL_duAlk</b>	61.8	0.14	1.8	39.8	41.7	-20.2	0.8
<b>DBCLL_du-ssAlk</b>	61.8	0.35	2.8	45.9	48.6	-13.2	2.0
<b>DBCLL_ClaqAlk</b>	61.8	0.31	2.7	45.5	48.2	-13.6	1.8
<b>AeroCom</b>	<b>62.9</b>	<b>0.20</b>	<b>13.4</b>	<b>18.7</b>	<b>32.1</b>	<b>-32.1</b>	<b>0.7</b>
<b>STD AeroCom</b>	<b>±3.9</b>	<b>±0.20</b>	<b>±5.1</b>	<b>±5.1</b>	<b>±10.2</b>	<b>±12.0</b>	<b>±0.3</b>
<b>GMI</b>	<b>60.4</b>	<b>0.85</b>	<b>1.1</b>	<b>8.7</b>	<b>9.8</b>	<b>-50.1</b>	<b>5.2</b>
<b>EMAC 2008<sup>1</sup></b>	<b>59.3</b>	<b>0.85</b>	<b>0.0</b>	<b>15.5</b>	<b>15.5</b>	-	-
<b>EMAC 2005-2008<sup>2</sup></b>	-	<b>0.82</b>	-	-	-	-	-
<b>IFS</b>	-	-	-	-	-	-	-
<b>LMDz-INCA</b>	<b>61.3</b>	<b>0.11</b>	<b>13.4</b>	<b>25.9</b>	<b>39.3</b>	<b>-21.2</b>	<b>0.6</b>
<b>MetOffice UM<sup>3</sup></b>	<b>64.7</b>	<b>0.05</b>	<b>6.9</b>	<b>21.1</b>	<b>28.0</b>	<b>-36.8</b>	<b>0.3</b>

<sup>1</sup> From Bian et al. (2017).

<sup>2</sup> From Karydis et al. (2016).

<sup>3</sup> Jones et al. (2021) performs two sensitivity tests to the accommodation coefficient used for NO<sub>3</sub><sup>-</sup> formation in the fine mode: *FAST* with 0.193 and *SLOW* with 0.001. Here, results from the *FAST* test are reported on the basis that they present similar fine NO<sub>3</sub><sup>-</sup> formation rates to our average fine nitrate results.

**Table S11.** Results fine, coarse and total particulate  $\text{NH}_4^+$  obtained with the studied heterogeneous chemistry mechanisms. Results from the literature references are reported at the end of the table: The average of all the participating models in the intercomparison AeroCom phase III nitrate experiment for 2008 (*AeroCom*), specifying their standard deviation (*STD AeroCom*), results from the GMI model (*GMI*, using a similar HYB approach with UPTK reactions on dust and SS), and results from the EMAC model (*EMAC 2008*, using a similar approach to *DBCLL\_du-ssALK*). Also using the EMAC model, results obtained by the Karydis et al. (2016) study are reported as *EMAC 2005-2008*. Results from models using a similar approach to *HYB\_du-ssUPTK* are reported as *IFS* for Rémy et al. (2022), *LMDz-INCA* for Hauglustaine et al. (2014) and *MetOffice UM* for Jones et al. (2021).

$\text{NH}_4^+$	Burden (Tg)	Wet Dep. (Tg y-1)			Dry Dep. (Tg y-1)			Total Dep. (Tg y-1)			Production (Tg y-1)			Lifetime (days)				
		Fine	Coarse	Total	Fine	Coarse	Total	Fine	Coarse	Total	Fine	Coarse	Total	Fine	Coarse	Total		
<b>fTEQ_noALK</b>	0.30	0.00	<b>0.30</b>	19.2	0.0	<b>19.2</b>	3.0	0.0	<b>3.0</b>	22.2	0.0	<b>22.2</b>	22.2	0.0	<b>22.2</b>	2.5	0.0	<b>2.5</b>
<b>fTEQ_du-ssALK</b>	0.21	0.00	<b>0.21</b>	14.9	0.0	<b>14.9</b>	2.0	0.0	<b>2.0</b>	16.9	0.0	<b>16.9</b>	16.9	0.0	<b>16.9</b>	2.3	0.0	<b>2.3</b>
<b>HYB_duUPTK</b>	0.21	0.00	<b>0.21</b>	14.3	0.0	<b>14.3</b>	1.9	0.0	<b>1.9</b>	16.2	0.0	<b>16.2</b>	16.2	0.0	<b>16.2</b>	2.3	0.0	<b>2.3</b>
<b>HYB_du-ssUPTK</b>	0.20	0.00	<b>0.20</b>	13.2	0.0	<b>13.2</b>	1.7	0.0	<b>1.7</b>	15.0	0.0	<b>15.0</b>	15.0	0.0	<b>15.0</b>	2.4	0.0	<b>2.4</b>
<b>HYB_gDU=0.1</b>	0.19	0.00	<b>0.19</b>	12.9	0.0	<b>12.9</b>	1.6	0.0	<b>1.6</b>	14.5	0.0	<b>14.5</b>	14.5	0.0	<b>14.5</b>	2.4	0.0	<b>2.4</b>
<b>HYB_DL</b>	0.19	0.00	<b>0.19</b>	11.7	0.0	<b>11.7</b>	1.6	0.0	<b>1.6</b>	13.3	0.0	<b>13.3</b>	13.3	0.0	<b>13.3</b>	2.7	0.0	<b>2.7</b>
<b>DBCLL_noALK</b>	0.29	0.01	<b>0.30</b>	17.8	0.3	<b>18.1</b>	2.8	0.2	<b>2.9</b>	20.5	0.5	<b>21.0</b>	20.5	0.5	<b>21.0</b>	2.6	3.1	<b>2.6</b>
<b>DBCLL_duALK</b>	0.30	0.01	<b>0.31</b>	18.9	0.3	<b>19.2</b>	3.1	0.2	<b>3.3</b>	22.1	0.4	<b>22.5</b>	22.1	0.4	<b>22.5</b>	2.5	4.1	<b>2.5</b>
<b>DBCLL_du-ssALK</b>	0.20	0.02	<b>0.22</b>	12.6	0.2	<b>12.7</b>	1.8	0.1	<b>1.9</b>	14.3	0.3	<b>14.6</b>	14.3	0.3	<b>14.6</b>	2.6	10.8	<b>2.8</b>
<b>DBCLL_ClaqALK</b>	0.22	0.02	<b>0.23</b>	12.9	0.2	<b>13.1</b>	1.8	0.2	<b>2.0</b>	14.7	0.4	<b>15.1</b>	14.7	0.4	<b>15.1</b>	2.7	7.7	<b>2.8</b>
<b>AeroCom</b>	-	-	<b>0.32</b>	-	-	<b>24.4</b>	-	-	<b>5.8</b>	-	-	<b>30.2</b>	-	-	<b>30.4</b>	-	-	<b>4.3</b>
<b>STD AeroCom</b>	-	-	<b>±0.20</b>	-	-	<b>±10.0</b>	-	-	<b>±5.8</b>	-	-	<b>±15.8</b>	-	-	<b>±4.3</b>	-	-	<b>±2.6</b>
<b>GMI</b>	-	-	<b>0.48</b>	-	-	<b>50.7</b>	-	-	<b>1.9</b>	-	-	<b>52.6</b>	-	-	<b>53.0</b>	-	-	<b>3.4</b>
<b>EMAC 2008<sup>1</sup></b>	-	-	<b>0.19</b>	-	-	<b>44.5</b>	-	-	<b>3.6</b>	-	-	<b>48.1</b>	-	-	-	-	-	-
<b>EMAC 2005-2008<sup>2</sup></b>	-	-	<b>0.17</b>	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<b>IFS<sup>3</sup></b>	-	-	<b>0.15</b>	-	-	<b>17.4</b>	-	-	<b>1.4</b>	-	-	<b>18.8</b>	-	-	<b>18.6</b>	-	-	<b>2.9</b>
<b>LMDz-INCA</b>	-	-	<b>0.28</b>	-	-	<b>19.2</b>	-	-	<b>3.2</b>	-	-	<b>22.4</b>	-	-	<b>22.4</b>	-	-	<b>4.5</b>
<b>MetOffice UM<sup>4</sup></b>	-	-	<b>0.54</b>	-	-	<b>32.1</b>	-	-	<b>7.4</b>	-	-	<b>32.1</b>	-	-	<b>39.2</b>	-	-	<b>5.0</b>

1. From Bian et al. (2017).

2. From Karydis et al. (2016).

3. Fine  $\text{NO}_3^-$  reported from neutralization of nitric acid, ammonia and sulfate. Coarse nitrate from heterogeneous chemistry (Rémy et al., 2022).

4. Jones et al. (2021) performs two sensitivity tests to the accommodation coefficient used for  $\text{NO}_3^-$  formation in the fine mode: *FAST* with 0.193 and *SLOW* with 0.001. Here, results from the *FAST* test are reported on the basis that they present similar fine  $\text{NO}_3^-$  formation rates to our average fine nitrate results

**Table S12.** Results for  $\text{SO}_{2(\text{g})}$ , and fine, coarse and total particulate  $\text{SO}_4^{2-}$  obtained with the studied heterogeneous chemistry mechanisms. Results from the literature references are reported at the end of the table: The average of all the participating models in the intercomparison AeroCom phase III nitrate experiment for 2008 (*AeroCom*), specifying their standard deviation (*STD AeroCom*), results from the GMI model (*GMI*, using a similar HYB approach with UPTK reactions on dust and SS), and results from the EMAC model (*EMAC 2008*, using a similar approach to *DBCLL\_du-ssAlk*). Also using the EMAC model, results obtained by the Karydis et al. (2016) study are reported as *EMAC 2005-2008*. Results from models using a similar approach to *HYB\_du-ssUPTK* are reported as *IFS* for Rémy et al. (2022), *LMDz-INCA* for Hauglustaine et al. (2014) and *MetOffice UM* for Jones et al. (2021).

$\text{SO}_4^{2-}$ Emissions ( $\text{Tg y}^{-1}$ )	$\text{SO}_2$ Emissions ( $\text{Tg y}^{-1}$ )	SO4 Burden ( $\text{Tg}$ )			SO4 Wet Dep. ( $\text{Tg y}^{-1}$ )			SO4 Dry Dep. ( $\text{Tg y}^{-1}$ )			SO4 Total Dep. ( $\text{Tg y}^{-1}$ )			SO4 Production ( $\text{Tg y}^{-1}$ )			SO4 Lifetime (days)				
Experiment	Fine	Coarse	Total	Fine	Coarse	Total	Fine	Coarse	Total	Fine	Coarse	Total	Fine	Coarse	Total	Fine	Coarse	Total			
<b>noHC</b>	0.5	0.0	<b>104.4</b>	4.4E-3	0.00	<b>0.00</b>	0.5	0.0	<b>0.5</b>	0.1	0.0	<b>0.1</b>	0.5	0.0	<b>0.5</b>	0.0	0.0	<b>0.0</b>	3.1	0.0	<b>3.0</b>
<b>FTEQ_noAlk</b>	0.5	0.0	<b>104.4</b>	1.98	0.00	<b>1.98</b>	126.9	0.0	<b>126.9</b>	13.5	0.0	<b>13.5</b>	140.5	0.0	<b>140.5</b>	139.9	0.0	<b>139.9</b>	2.6	0.9	<b>2.6</b>
<b>FTEQ_du-ssAlk</b>	0.5	0.0	<b>104.4</b>	1.82	0.14	<b>1.96</b>	116.7	6.9	<b>123.6</b>	13.0	3.5	<b>16.5</b>	129.7	10.4	<b>140.1</b>	129.1	10.4	<b>139.5</b>	2.6	2.4	<b>2.6</b>
<b>HYB_duUPTK</b>	0.5	0.0	<b>104.4</b>	1.82	0.14	<b>1.96</b>	116.5	7.0	<b>123.5</b>	13.0	3.5	<b>16.4</b>	129.4	10.5	<b>139.9</b>	128.9	10.5	<b>139.3</b>	2.6	2.4	<b>2.6</b>
<b>HYB_du-ssUPTK</b>	0.5	0.0	<b>104.4</b>	1.85	0.14	<b>1.98</b>	116.4	7.1	<b>123.5</b>	12.9	3.5	<b>16.4</b>	129.3	10.6	<b>139.9</b>	128.7	10.6	<b>139.3</b>	2.6	2.4	<b>2.6</b>
<b>HYB_gDU=0.1</b>	0.5	0.0	<b>104.4</b>	1.87	0.12	<b>1.99</b>	117.8	6.1	<b>124.0</b>	12.7	2.9	<b>15.7</b>	130.6	9.1	<b>139.7</b>	130.0	9.0	<b>139.1</b>	2.6	2.4	<b>2.6</b>
<b>HYB_DL</b>	0.5	0.0	<b>104.4</b>	1.85	0.14	<b>1.99</b>	116.4	7.1	<b>123.5</b>	12.9	3.5	<b>16.4</b>	129.3	10.6	<b>139.9</b>	128.7	10.6	<b>139.3</b>	2.6	2.4	<b>2.6</b>
<b>DBCLL_noAlk</b>	0.5	0.0	<b>104.4</b>	1.94	0.04	<b>1.98</b>	124.4	2.0	<b>126.4</b>	13.2	0.9	<b>14.0</b>	137.6	2.9	<b>140.5</b>	137.0	2.9	<b>139.9</b>	2.6	2.5	<b>2.6</b>
<b>DBCLL_duAlk</b>	0.5	0.0	<b>104.4</b>	1.78	0.16	<b>1.94</b>	114.8	8.4	<b>123.1</b>	12.7	4.1	<b>16.8</b>	127.5	12.4	<b>139.9</b>	126.9	12.4	<b>139.3</b>	2.6	2.4	<b>2.5</b>
<b>DBCLL_du-ssAlk</b>	0.5	0.0	<b>104.4</b>	1.76	0.20	<b>1.96</b>	114.9	8.3	<b>123.2</b>	12.7	4.1	<b>16.8</b>	127.6	12.4	<b>140.0</b>	127.1	12.3	<b>139.4</b>	2.5	3.0	<b>2.6</b>
<b>DBCLL_ClaqAlk</b>	0.5	0.0	<b>104.4</b>	1.84	0.13	<b>1.97</b>	117.0	6.9	<b>123.8</b>	12.8	3.3	<b>16.1</b>	129.8	10.1	<b>139.9</b>	129.2	10.1	<b>139.3</b>	2.6	2.4	<b>2.6</b>
<b>AeroCom</b>	-	-	<b>122.0</b>	-	-	<b>1.80</b>	-	-	<b>140.0</b>	-	-	<b>14.3</b>	-	-	<b>154.3</b>	-	-	<b>151.0</b>	-	-	<b>4.5</b>
<b>STD AeroCom</b>	-	-	<b>122.0</b>	-	-	<b>±0.81</b>	-	-	<b>±165.3</b>	-	-	<b>±163.3</b>	-	-	<b>±228.6</b>	-	-	<b>±56.0</b>	-	-	<b>±1.9</b>
<b>GMI</b>	-	-	<b>122.0</b>	-	-	<b>3.30</b>	-	-	<b>140.0</b>	-	-	<b>14.3</b>	-	-	<b>154.3</b>	-	-	<b>151.0</b>	-	-	<b>4.5</b>
<b>EMAC 2008<sup>1</sup></b>	-	-	<b>138.0</b>	-	-	<b>1.90</b>	-	-	<b>302.0</b>	-	-	<b>504.0</b>	-	-	<b>806.0</b>	-	-	<b>187.0</b>	-	-	<b>0.9</b>
<b>EMAC 2005-2008<sup>2</sup></b>	-	-	-	-	-	<b>1.78</b>	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
<b>IFS<sup>3</sup></b>	-	-	<b>70.3</b>	-	-	<b>0.367</b>	-	-	<b>397</b>	-	-	<b>1.9</b>	-	-	<b>397</b>	-	-	<b>41.4</b>	-	-	<b>3.2</b>
<b>LMDz-INCA</b>	-	-	-	-	-	<b>1.26</b>	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
<b>MetOffice UM<sup>4</sup></b>	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	

1. From Bian et al. (2017).

2. From Karidis et al. (2016).

3. Fine  $\text{NO}_3^-$  reported from neutralization of nitric acid, ammonium and sulfate. Coarse nitrate from heterogeneous chemistry (Rémy et al., 2022).

4. Jones et al. (2021) performs two sensitivity tests to the accommodation coefficient used for  $\text{NO}_3^-$  formation in the fine mode: *FAST* with 0.193 and *SLOW* with 0.001. Here, results from the *FAST* test are reported on the basis that they present similar fine  $\text{NO}_3^-$  formation rates to our average fine nitrate results

**Table S13.** Nitrogen burdens (TgN) for each experiment and species.

Nitrogen burdens	noHC	fTEQ	fTEQ	HYB	HYB	HYB	DBCLL	DBCLL	DBCLL	DBCLL
	noAlk	du-ssAlk	duUPTK	du-ssUPTK	gDU=0.1	DL	noAlk	duAlk	du-ssAlk	ClaqAlk
HNO <sub>3(g)</sub>	1.06	1.11	0.65	0.44	0.15	0.1	0.16	1.14	0.6	0.56
NH <sub>3(g)</sub>	0.71	0.04	0.13	0.17	0.21	0.23	0.31	0.05	0.11	0.28
NO <sub>(g)</sub>	0.14	0.18	0.14	0.13	0.11	0.1	0.11	0.19	0.14	0.14
NO <sub>2(g)</sub>	0.24	0.27	0.22	0.2	0.17	0.17	0.17	0.28	0.22	0.22
N <sub>2</sub> O <sub>5(g)</sub>	0.04	0.03	0.02	0.02	0.01	0.01	0.01	0.03	0.02	0.02
NTR <sub>(g)</sub>	0.44	0.47	0.48	0.48	0.47	0.47	0.47	0.47	0.48	0.48
PNA <sub>(g)</sub>	0.04	0.03	0.02	0.02	0.02	0.02	0.02	0.03	0.02	0.02
PAN <sub>(g)</sub>	0.8	0.64	0.53	0.49	0.42	0.41	0.42	0.65	0.52	0.52
PAN <sub>x(g)</sub>	0.16	0.13	0.11	0.11	0.09	0.09	0.09	0.14	0.11	0.11
PM <sub>2.5</sub> NO <sub>3(aer)</sub>	0	0.02	0.15	0.13	0.12	0.11	0.09	0.01	0.22	0.06
PM <sub>2.5–10</sub> NO <sub>3(aer)</sub>	0	0	0	0.13	0.27	0.32	0.29	0	0.11	0.18
PM <sub>2.5</sub> NH <sub>4(aer)</sub>	0	0.23	0.17	0.16	0.15	0.15	0.15	0.22	0.24	0.16
PM <sub>2.5–10</sub> NH <sub>4(aer)</sub>	0	0	0	0	0	0	0	0.01	0.01	0.01
NO <sub>(g)</sub> + NO <sub>2(g)</sub> + NO <sub>3(g)</sub>	<b>0.39</b>	<b>0.46</b>	<b>0.36</b>	<b>0.33</b>	<b>0.28</b>	<b>0.28</b>	<b>0.47</b>	<b>0.37</b>	<b>0.36</b>	<b>0.36</b>
NTR <sub>(g)</sub> + PNA <sub>(g)</sub> + PAN <sub>(g)</sub> + PAN <sub>x(g)</sub>	<b>1.44</b>	<b>1.28</b>	<b>1.14</b>	<b>1.09</b>	<b>1.01</b>	<b>0.99</b>	<b>1</b>	<b>1.29</b>	<b>1.13</b>	<b>1.13</b>
Aerosol nitrate + ammonium	<b>0</b>	<b>0.25</b>	<b>0.31</b>	<b>0.42</b>	<b>0.55</b>	<b>0.58</b>	<b>0.53</b>	<b>0.24</b>	<b>0.57</b>	<b>0.41</b>
Total nitrogen	<b>3.63</b>	<b>3.15</b>	<b>2.62</b>	<b>2.48</b>	<b>2.19</b>	<b>2.18</b>	<b>2.29</b>	<b>3.22</b>	<b>2.79</b>	<b>2.76</b>
										<b>2.74</b>

## S5 Discussion of additional sensitivity experiments

### S5.1 HYB\_g0p1

To assess the sensitivity to more efficient coarse  $\text{NO}_3^-$  production rates on dust (reaction R4 in Table 1 ), we perform the HYB\_g0p1 run, that employs a constant value for  $\gamma_{dust}$  of 0.1, rather than utilizing the RH-dependent function proposed by 15 (Fairlie et al., 2010). This experiment is also intended to elucidate the relative role of the dust over SS since, as noted in Section 2.2.1 , the  $\gamma_{dust}$  values used in both HYB\_duUPTK and HYB\_du-ssUPTK runs are well below the value used for  $\gamma_{SS}$  and other studies in the literature (Hauglustaine et al., 2014; Rémy et al., 2022). Results indicate an increase in coarse  $\text{NO}_3^-$  formation in regions rich in dust (Fig. S10), such as the Sahara desert, the Persian Gulf and East Asia (Fig. S3c). Furthermore, a slight enhancement in the transport of coarse  $\text{NO}_3^-$  across northern latitudes is observed, despite lower dust concentrations in 20 that region. This can be attributed to the predominant production of coarse  $\text{NO}_3^-$  on dust across continental Eurasia, followed by its long-range transport at high altitudes to North America.

Among the mechanisms analyzed, HYB\_g0p1 shows the most efficient depletion of  $\text{HNO}_{3(g)}$ , achieved by using a constant value of  $\gamma = 0.1$  instead of the RH-dependent dust uptake coefficient from Fairlie et al. (2010). In this scenario,  $\text{HNO}_{3(g)}$  is further reduced to 0.46 TgN, accompanied by an increase in coarse  $\text{NO}_3^-$  formation up to 1.93 Tg. Furthermore, the deposition 25 rates rise from 140 to 149 Tg/y, as shown in Table 5 in the main text.

### S5.2 DBCLL\_duAlk

The DBCLL\_duAlk configuration only accounts for dust alkalinity in the DBCLL mechanism. It shows that the sole presence of dust, compared to the case with no alkalinity in the DBCLL\_noAlk configuration, significantly increases pH values (i.e. more basic) over the Sahara and continental Asia to a range of 6 to 8 (Fig. 4 g1, g2). Thereby, dust enhances the formation of 30 both fine and coarse  $\text{NO}_3^-$  as well as coarse  $\text{SO}_4^{2-}$  in these regions. Additionally, fine  $\text{NO}_3^-$  formation is amplified in polluted areas, with surface concentrations rising beyond  $5 \mu\text{g m}^{-3}$  (an increase of 2-5  $\mu\text{g m}^{-3}$  relative to DBCLL\_noAlk). This enhanced formation extends to regions rich in coarse dust, such as the Persian Gulf (surface concentrations of  $1\text{-}3 \mu\text{g m}^{-3}$ ) and central/downwind areas of the Saharan desert (surface concentrations of  $0.2\text{-}0.5 \mu\text{g m}^{-3}$  and loads of  $2.0 \text{ mg m}^{-2}$ ), as shown in Figures 3 u and S1u. Over these areas, DBCLL\_duAlk nearly depletes  $\text{HNO}_{3(g)}$  gas (remaining surface concentrations 35 of  $0.01$  to  $0.05 \mu\text{g m}^{-3}$ ). At high altitudes, both fine and coarse  $\text{NO}_3^-$  are distributed across the Atlantic and Pacific oceans with column loads of  $1\text{-}2 \text{ mg m}^{-2}$  (Fig. S1u), resembling the transoceanic patterns also observed in the HYB mechanisms. Conversely, compared to the DBCLL\_noAlk run, fine  $\text{NH}_4^+$  increases over Europe and North-Central America (by at least  $1 \text{ mg m}^{-2}$ , discussed in Section 3.3.1 ), while it decreases over dusty areas (by  $1.0$  to  $2.0 \text{ mg m}^{-2}$ , Fig. S5t, q). This pattern is consistent with the comparison between the fTEQ\_noAlk and the fTEQ\_du-ssAlk runs (Fig. S5h, e). Concurrently, coarse 40  $\text{NH}_4^+$  formation increases significantly in transoceanic areas, reaching  $0.08\text{-}0.1 \text{ mg m}^{-2}$  alongside coarse  $\text{NO}_3^-$  particles, although its formation is halted over dusty regions (i.e. Middle East and Sahara, Fig. S5u). This inhibition can be attributed to the increased alkalinity of dust, which neutralizes  $\text{HNO}_{3(g)}$  and limits the condensation of  $\text{NH}_{3(g)}$ . Lastly, the introduction of dust alkalinity strongly enhances the formation of coarse  $\text{SO}_4^{2-}$ , both over polluted regions ( $5 \text{ mg m}^{-2}$ ) and remote areas ( $0.2\text{-}$

1.0 mg m<sup>-2</sup>), as shown in Fig. S8. The enhanced production of coarse particles (principally coarse NO<sub>3</sub><sup>-</sup>) provides additional

45 surface area for H<sub>2</sub>SO<sub>4(g)</sub> to condense on.

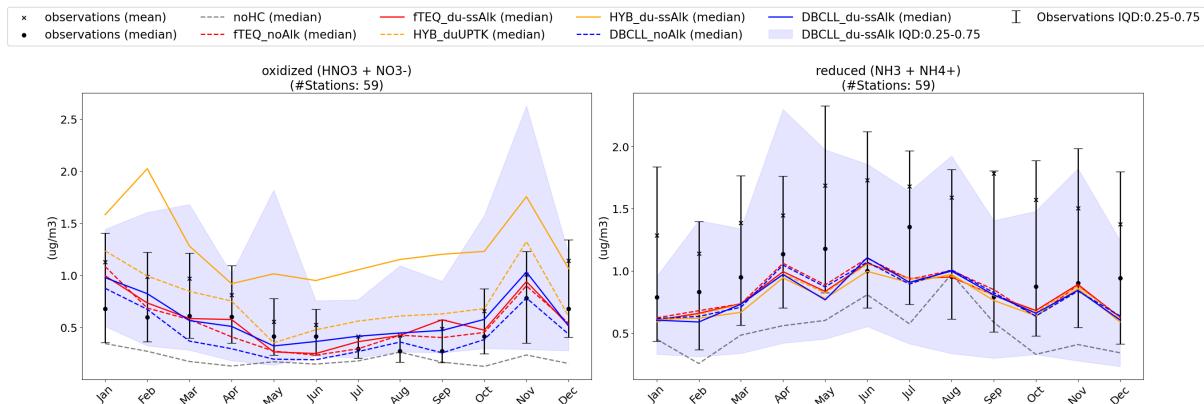
When compared with observations, DBCLL\_duAlk results in opposite results to DBCLL\_noAlk: HNO<sub>3(g)</sub> is underestimated with respect to observations from March to September by -0.1 μg m<sup>-3</sup>, while both fine and total particulate NO<sub>3</sub><sup>-</sup> are overestimated with respect to observations by 1.5 and 2 μg m<sup>-3</sup> respectively (see Fig. S14).

Comparing the obtained NO<sub>3</sub><sup>-</sup> burdens and size distributions with those reported by Rémy et al. (2022), Hauglustaine et al. 50 (2014), and Jones et al. (2021) (Table 5), we observe that the total NO<sub>3</sub><sup>-</sup> values are notably high when only dust alkalinity is considered. This discrepancy can be attributed to excessive fine NO<sub>3</sub><sup>-</sup> formation, as already noted in the observational evaluation. This indicates that while the DBCLL\_duAl approach improves the representation of alkalinity effects compared to the irreversible mechanisms, it still tends to overestimate the fine mode NO<sub>3</sub><sup>-</sup>, suggesting a need for further refinement in the parameterization of NVC effects to achieve more accurate nitrate aerosol predictions. Moreover, deposition rates of total NO<sub>3</sub><sup>-</sup> 55 (54.01 Tg/y) fall below those reported by all the consulted references, suggesting that DBCLL\_duAlk might form too much fine nitrate (with lower deposition rates) and misses coarse NO<sub>3</sub><sup>-</sup> formation (with higher deposition rates).

### S5.3 DBCLL\_ClaqAlk

The DBCLL\_ClaqAlk employs the average alkalinity of dust from Claquin et al. (1999) instead of Journet et al. (2014) with the DBCLL mechanism, and it shows significant differences in specific arid regions compared with DBCLL\_du-ssAlk. Notably, with DBCLL\_ClaqAlk the load of NO<sub>3</sub><sup>-</sup> substantially increases over the Middle East and northern India, while a slight reduction is observed over northern Africa. These changes are significant enough to impact the long-range transport of both fine and coarse NO<sub>3</sub><sup>-</sup> across the northern hemisphere (Fig. S3k, l). Coarse NO<sub>3</sub><sup>-</sup> decreases by about 1-2 mg m<sup>-2</sup> over and downwind of dusty areas, while fine NO<sub>3</sub><sup>-</sup> increases by 2-5 mg m<sup>-2</sup> along the equatorial belt and over polluted regions. Similar differences are found in particulate NH<sub>4</sub><sup>+</sup>, although with an order of magnitude lower. Notably, HNO<sub>3(g)</sub> distributions report 60 negligible differences between both runs. The lower alkalinity derived from the Claquin et al. (1999) dataset accounts for these differences, primarily affecting the formation of NO<sub>3</sub><sup>-</sup> during long-range transport of dust, as discussed in Section 3.4 .

## S6 Observational evaluation of oxidized and reduced species



**Figure S15.** Observational evaluation for oxidized ( $\text{HNO}_{3(g)} + \text{NO}_3^-$ , left) and reduced ( $\text{NH}_{3(g)} + \text{NH}_4^+$ , right) nitrogen species.



**Figure S16.** Stations employed for the observational evaluation of oxidized ( $\text{HNO}_{3(g)} + \text{NO}_3^-$ ) and reduced ( $\text{NH}_{3(g)} + \text{NH}_4^+$ ) nitrogen species.

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