1   Supporting Information for

2   **Development and evaluation of E3SM-MOSAIC: Spatial distributions**

3   **and radiative effects of nitrate aerosol**

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Introduction

In the Supporting Information section, we prepared 13 figures and 4 tables to support the main manuscript. Table S1 gives the mass budget of nitrate in MTC_SPLAC and MTC_FAST. Table S2 gives the mass budget of sulfate from 5 E3SM experiments. Table S3 lists the tropospheric HNO_3 burden in the three MOSAIC experiments compared with other studies. Table S4 lists the mean surface molar concentrations of aerosols and gases for Figures 4 and 5 in the main manuscript. Figure S1 evaluates the modeled tropospheric column ozone against OMI/MLS retrievals. Figure S2 compares modeled tropospheric C_2H_6, C_3H_8, C_2H_2, and C_2H_4 with summaries of observations from aircraft campaigns. Figure S3 shows the spatial distributions of nitrate column mass tendencies due to aqueous chemistry and gas-aerosol exchange. Figure S4 shows the spatial distributions of ammonium burden. Figure S5 shows the latitude-altitude cross sections of nitrate concentrations. Figure S6 evaluates modeled surface concentrations of nitrate aerosols and HNO_3 from Zaveri et al. (2021) and Lu et al. (2021) against observations at CASTNET, EMEP, and EANET network sites. Figure S7 evaluates modeled surface concentrations of NO_x against observations at AQS, EMEP, and EANET network sites.

48   Figures S8-S10 show the seasonal variations of simulated and observed sulfate, HNO_3,

49   and ammonium surface concentrations at 6 CASTNET sites. Figures S11 and S12

50   compare vertical profiles of HNO_3 concentrations from model simulations with

51   observations from INTEX-B, ARCTAS, DC3, SEAC[4]RS, and ATom campaigns. Figure

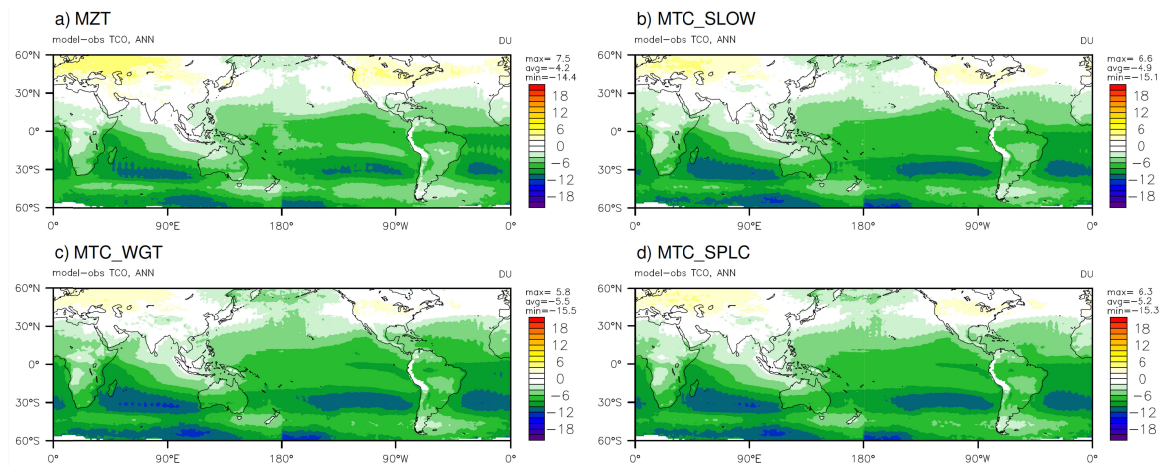52   S13 shows the spatial distributions of RFari of ammonium aerosols.

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55   **Figure S1.** Spatial distributions of global annual mean tropospheric column ozone

56   differences compared with OMI/MLS retrievals.
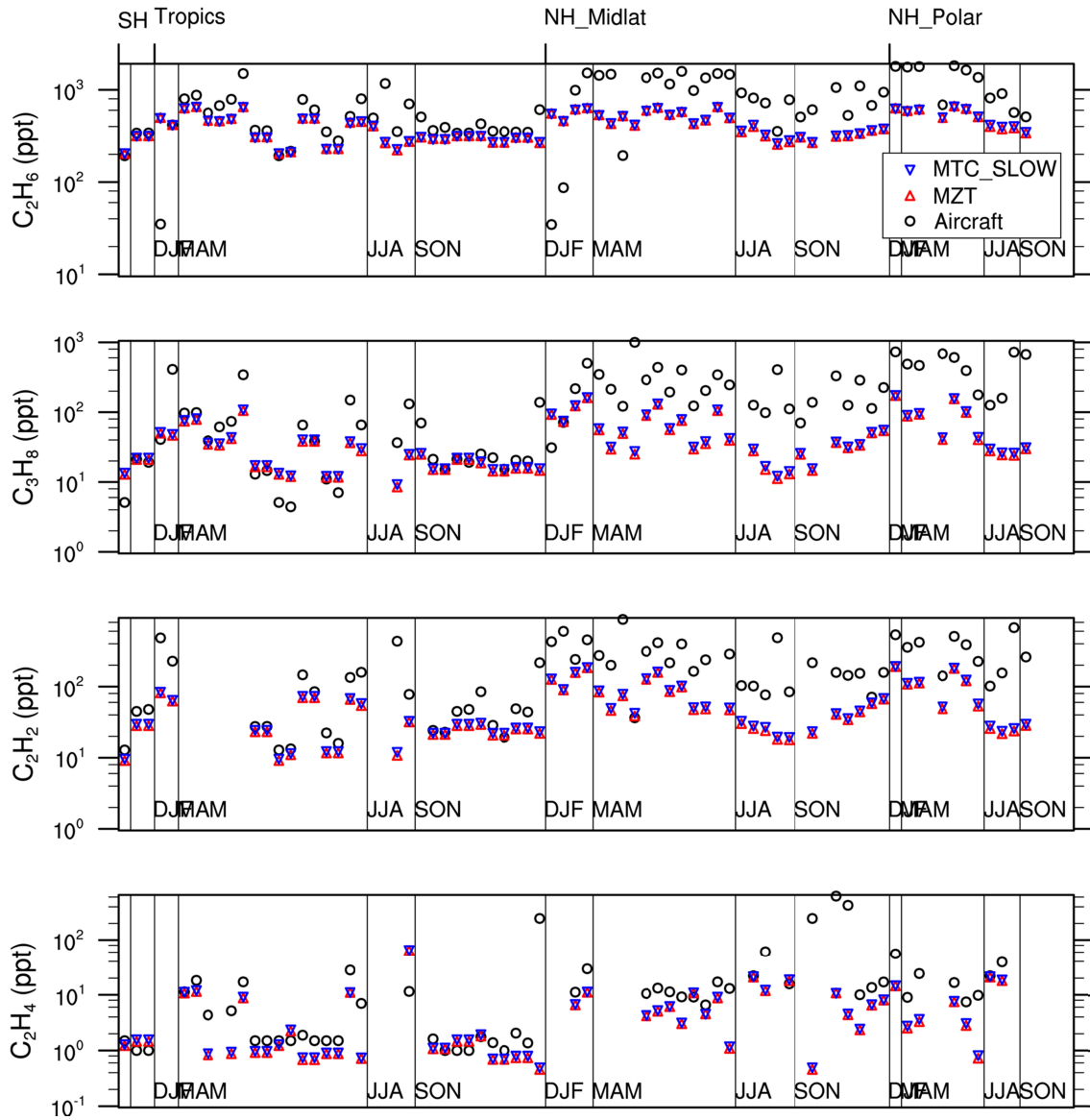
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Figure S2. Evaluation of modeled tropospheric C_2H_6, C_3H_8, C_2H_2, and C_2H_4 (averaged over 2005-2014) with summaries of observations from aircraft campaigns (operated during 1995-2010), averaged over 2-7 km.
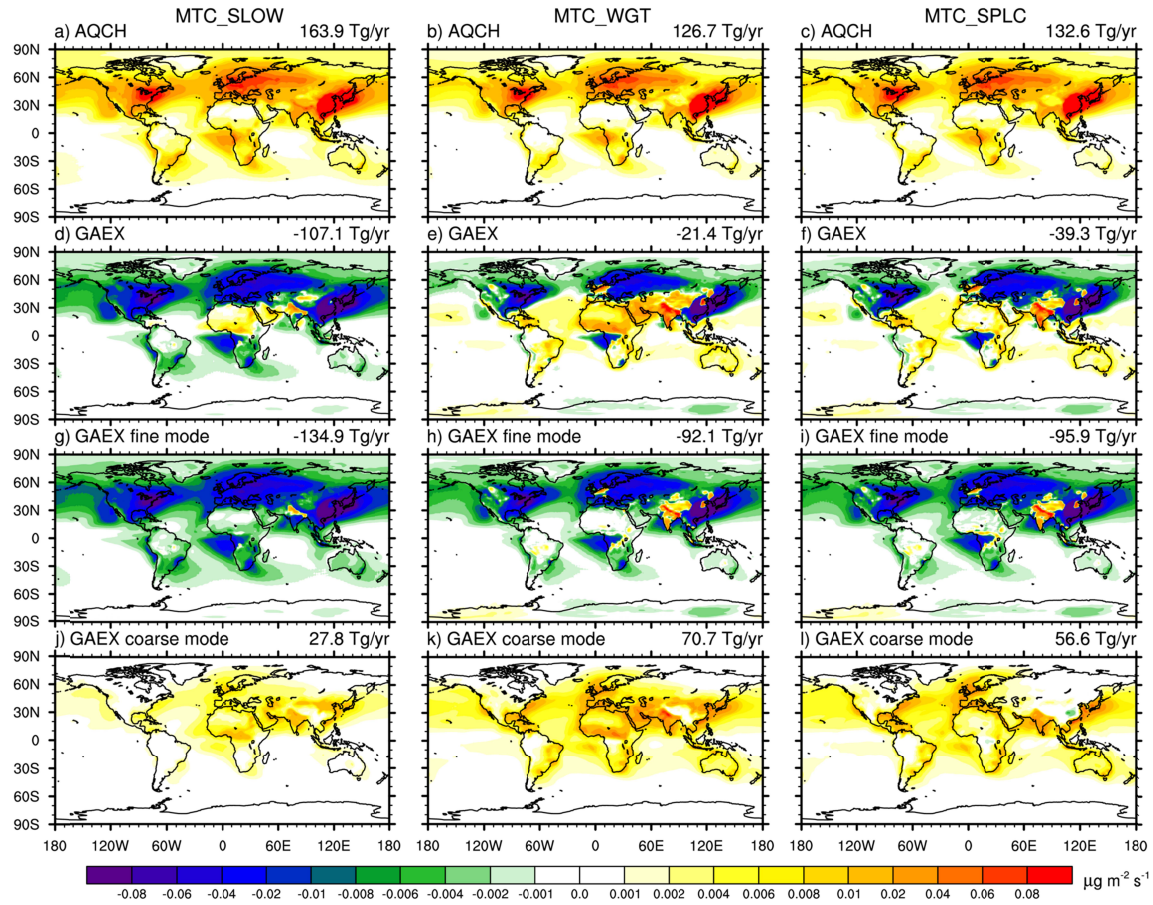
Figure S3. Spatial distributions of nitrate column mass tendencies ($\mu g\ m^{-2}\ s^{-1}$) due to (a-c) aqueous chemistry (AQCH), (d-f) gas-aerosol exchange (GAEX), (g-i) gas-aerosol exchange in the fine mode, and (j-l) gas-aerosol exchange in the coarse mode.
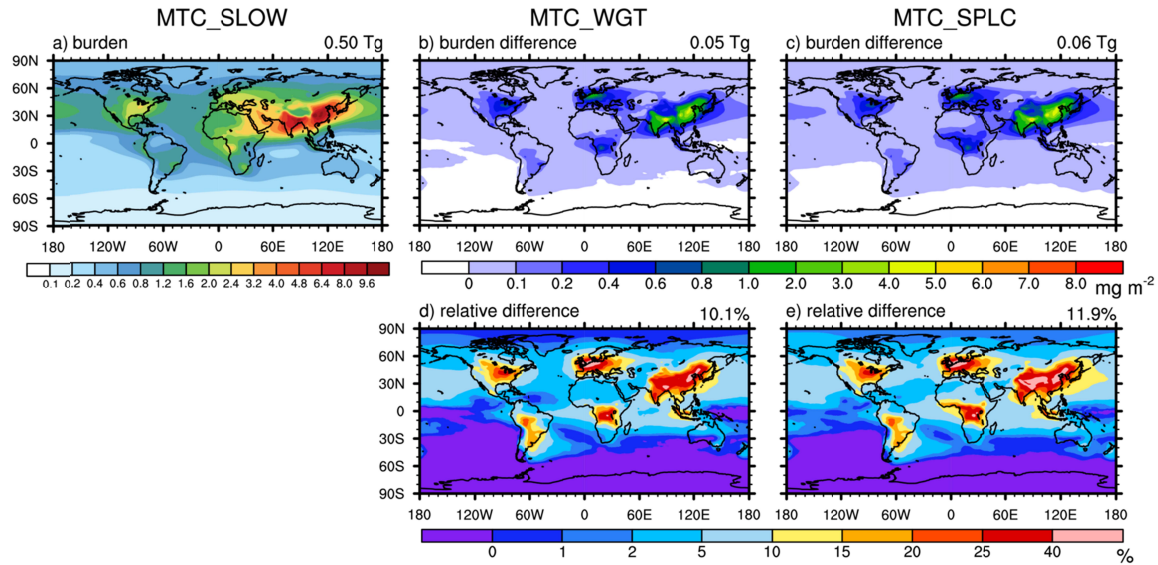
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Figure S4. Spatial distributions of global annual mean (a) ammonium burden (MTC_SLOW), (b-c) ammonium burden differences (MTC_WGT and MTC_SPLC) compared to MTC_SLOW, and (d-e) relative differences of ammonium burden compared to MTC_SLOW. Numbers at the top-right of each panel are global annual mean values.
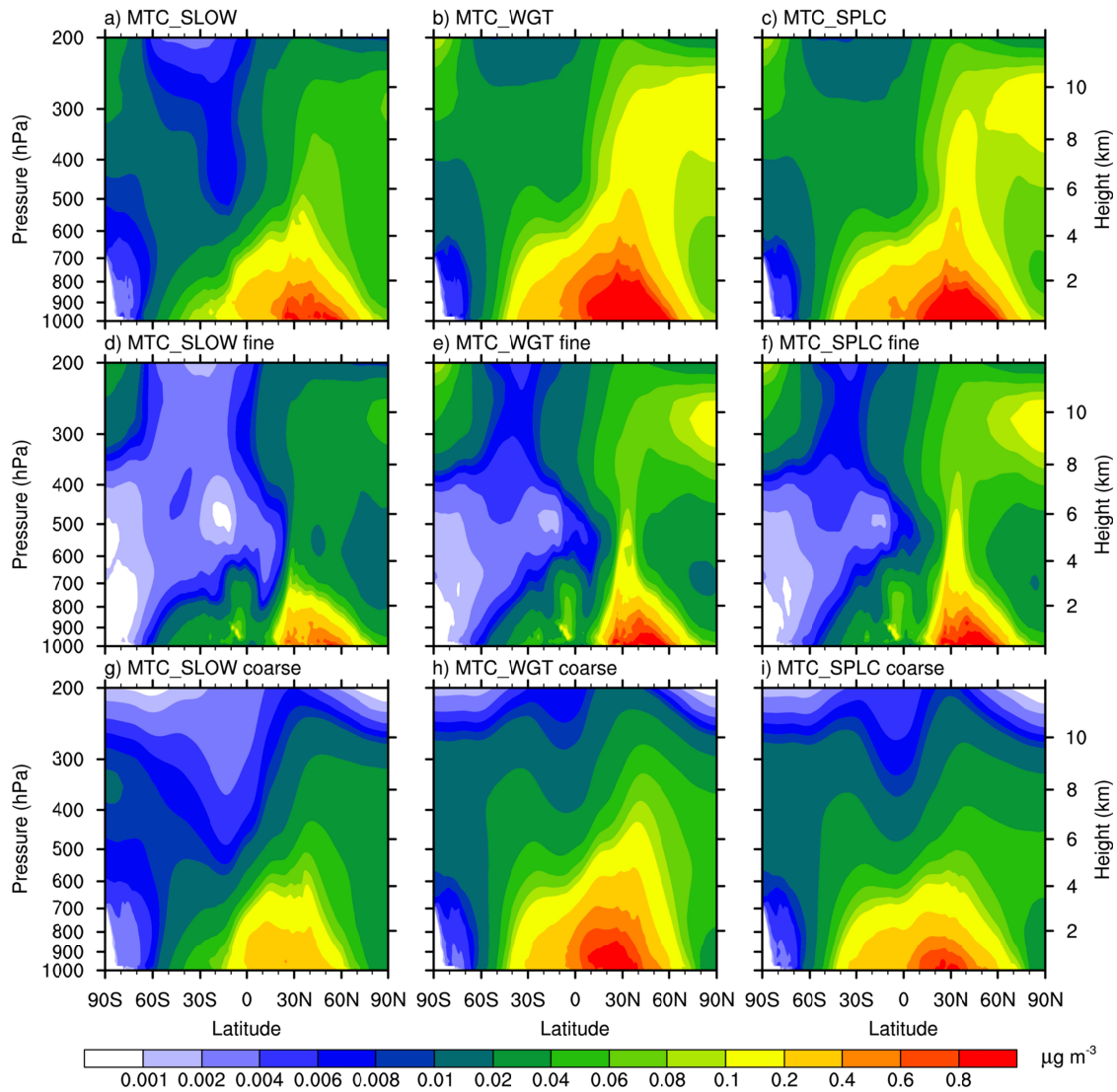
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Figure S5. Latitude-altitude cross sections of annually averaged zonal mean (a-c) total, (d-f) fine mode, and (g-i) coarse mode nitrate concentrations ($\mu g\ m^{-3}$).
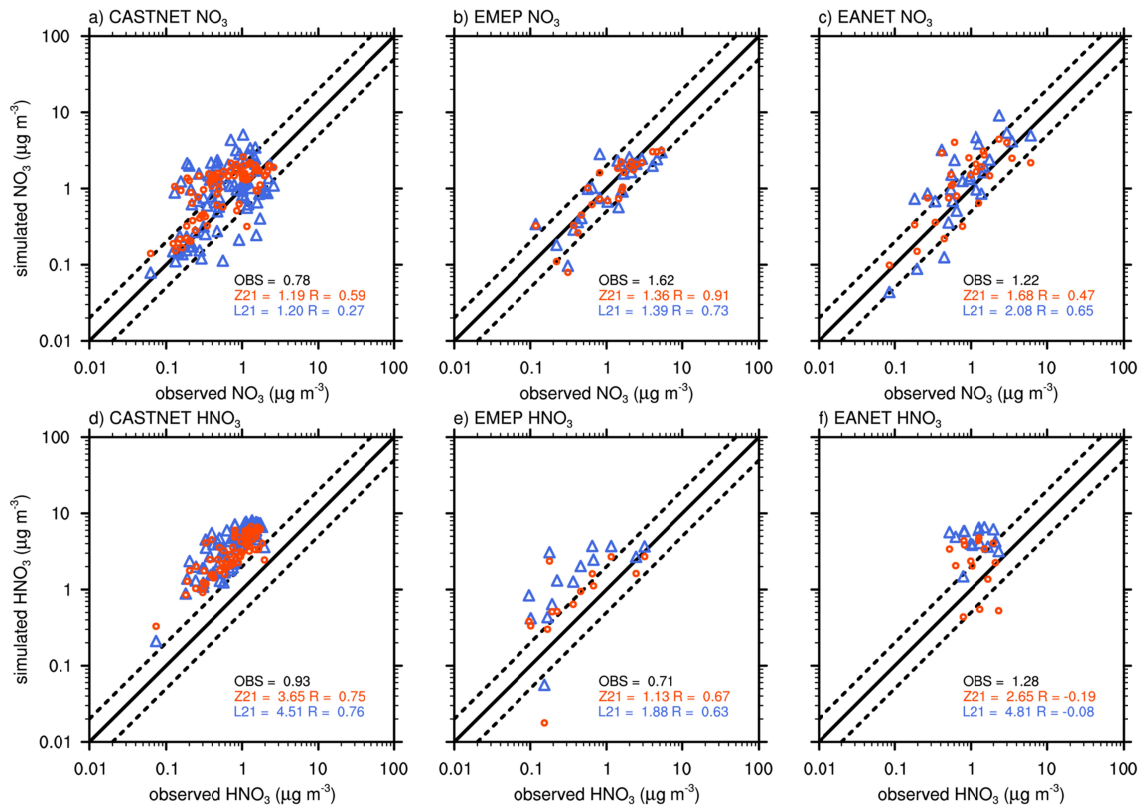
Figure S6. Scatter plots of modeled annual mean surface concentrations ($\mu g\ m^{-3}$) of nitrate aerosols (top row) and HNO_3 (bottom row) from Zaveri et al. (2021) (2000-2005) and Lu et al. (2021) (2005-2007) compared to observations at CASTNET (left column), EMEP (middle column), and EANET (right column) network sites during 2005-2014. The numbers are mean concentrations and correlation coefficients.
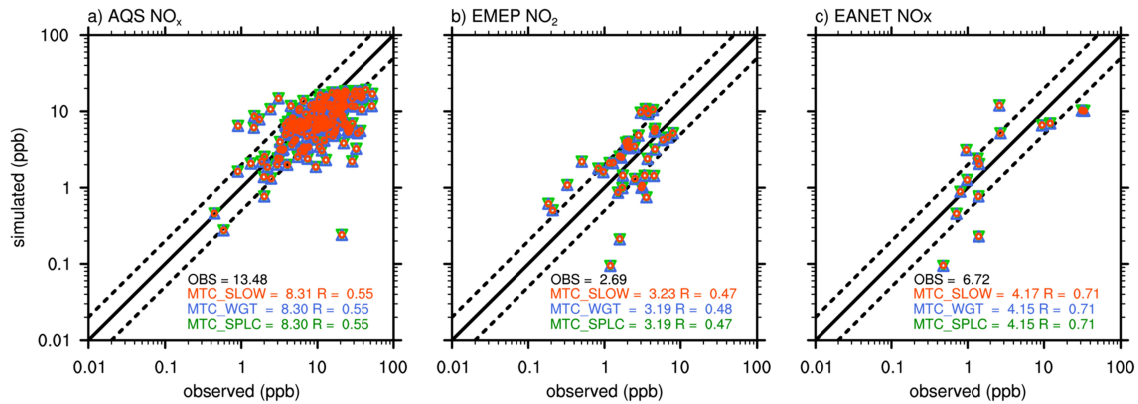
Figure S7. Scatter plots of modeled annual mean surface concentrations of NO_x (ppb) compared to observations at AQS and EANET network sites and NO_2 (ppb in STP; converted from $\mu g\ m^{-3}$) compared to observations at EMEP network sites. We only select rural and suburban sites for AQS network. The numbers are mean concentrations and correlation coefficients.

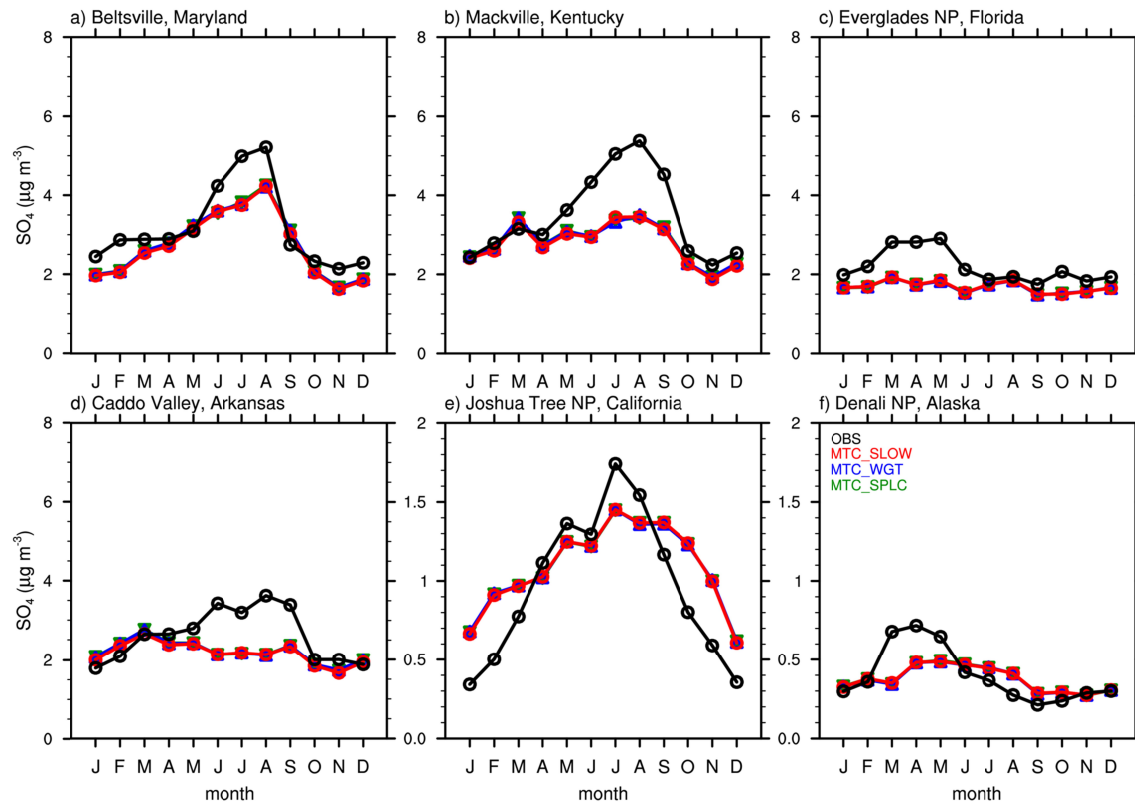**Figure S8.** Seasonal variations of simulated (color lines and symbols) and observed (black lines and circles) sulfate surface concentrations ($\mu g\ m^{-3}$) at six CASTNET sites.

Figure S9. Seasonal variations of simulated (color lines and symbols) and observed (black lines and circles) HNO_3 surface concentrations ($\mu g\ m^{-3}$) at six CASTNET sites.

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Figure S10. Seasonal variations of simulated (color lines and symbols) and observed (black lines and circles) ammonium surface concentrations ($\mu g\ m^{-3}$) at six CASTNET sites.
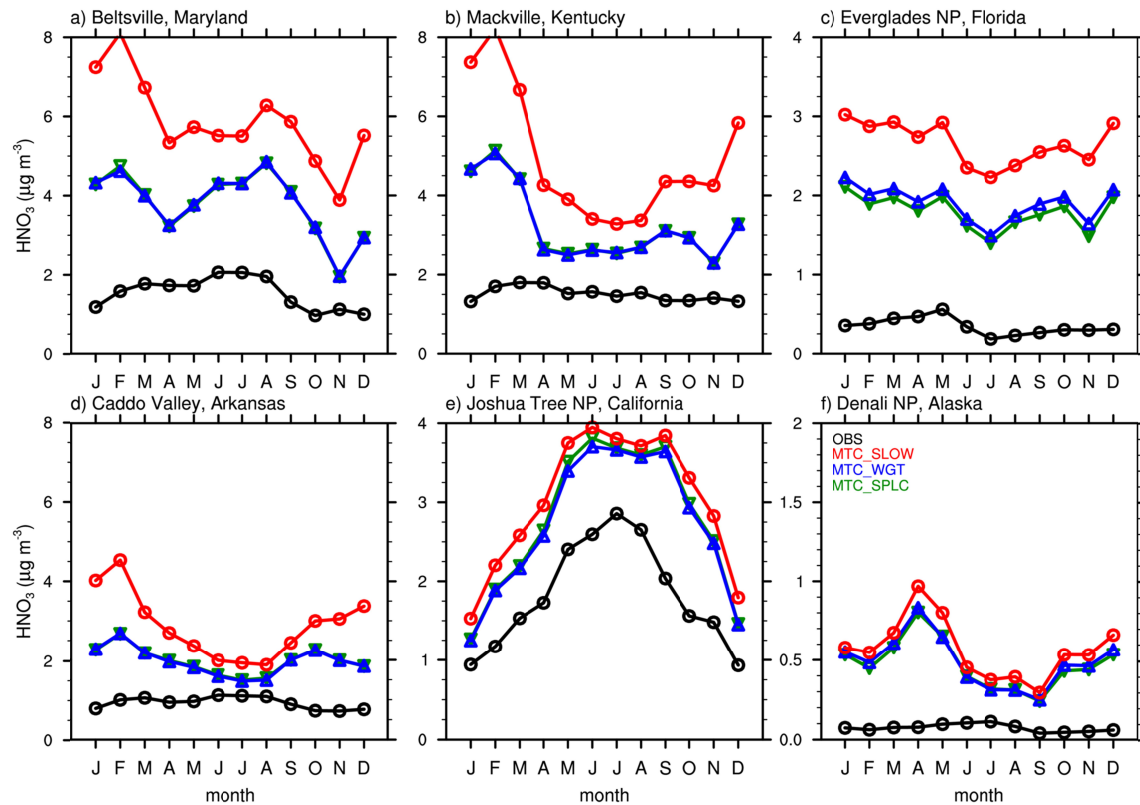
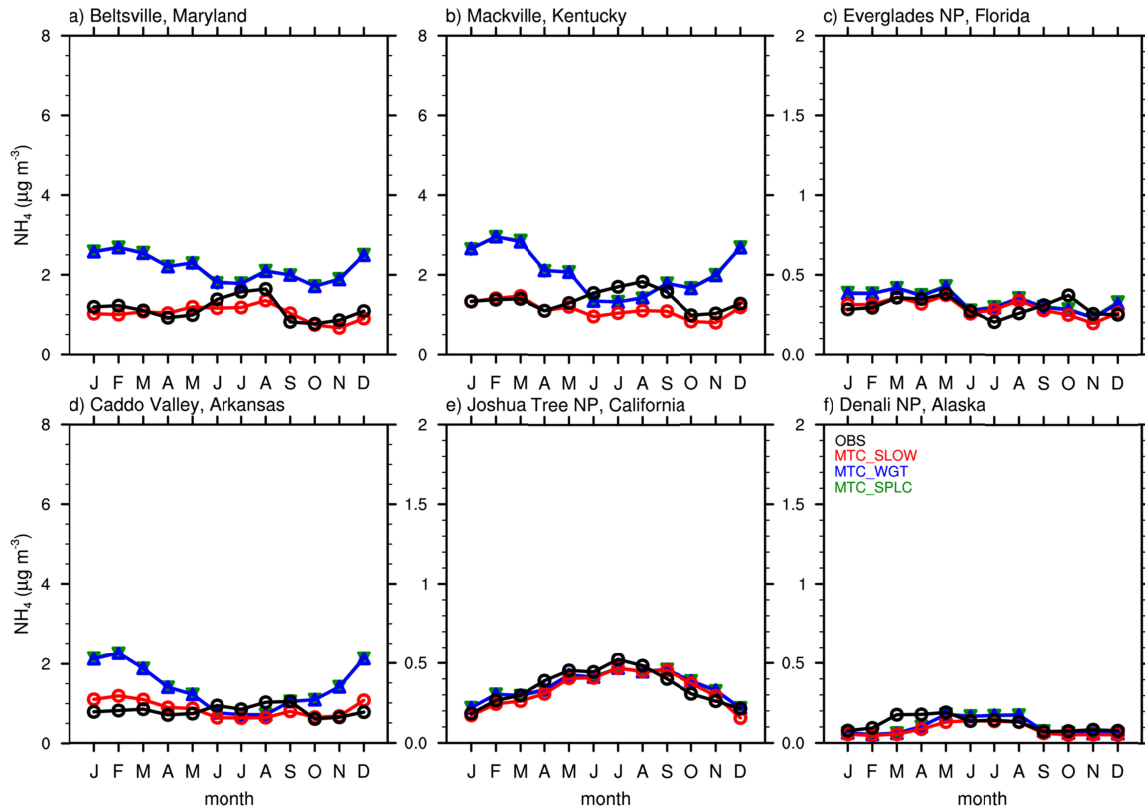**Figure S11.** Vertical profiles of HNO_3 concentrations ($\mu g\ m^{-3}$ in STP) from model simulations (colored lines) and four aircraft campaigns (dark solid lines for mean values; shaded areas for plus/minus one standard deviation of observations).
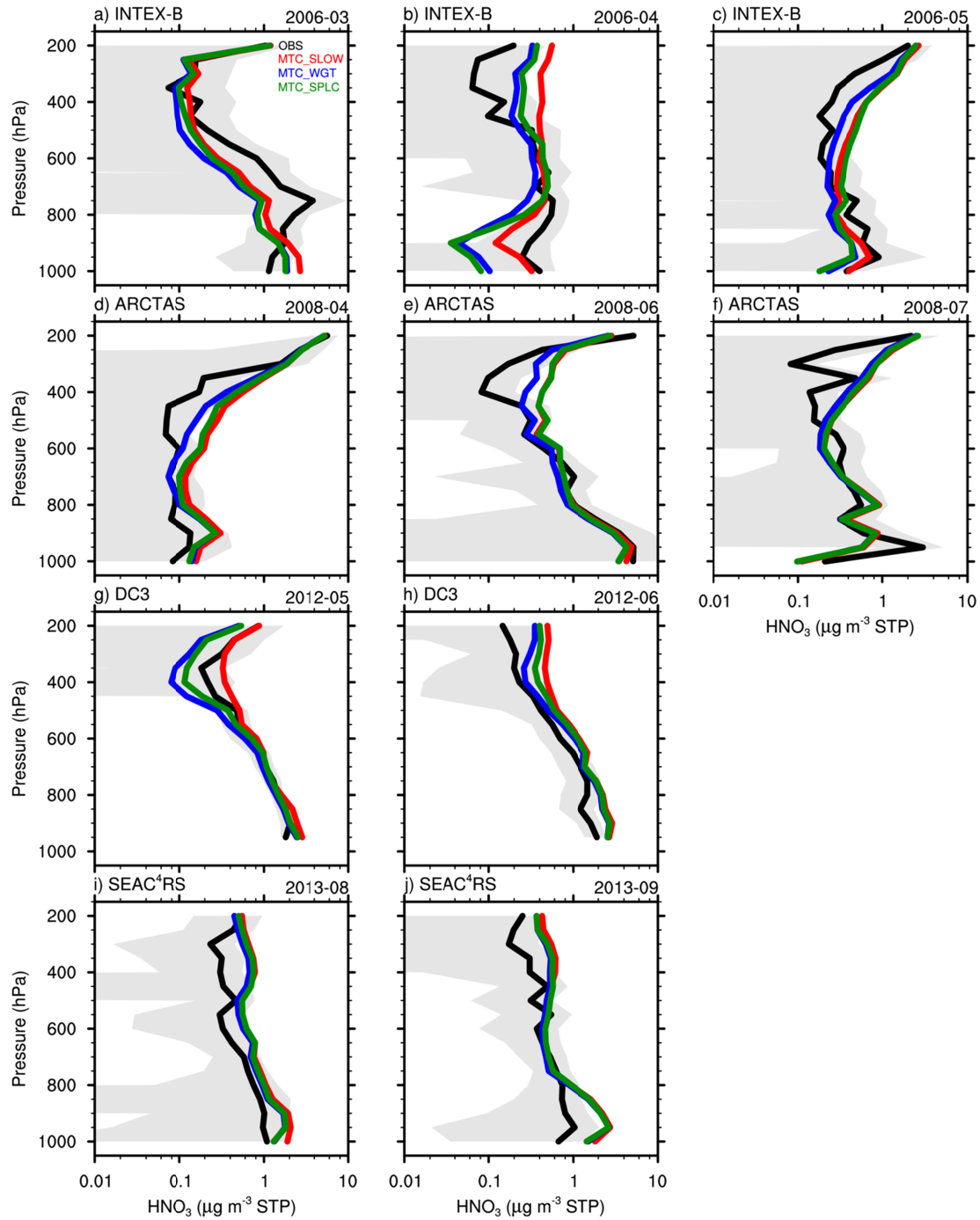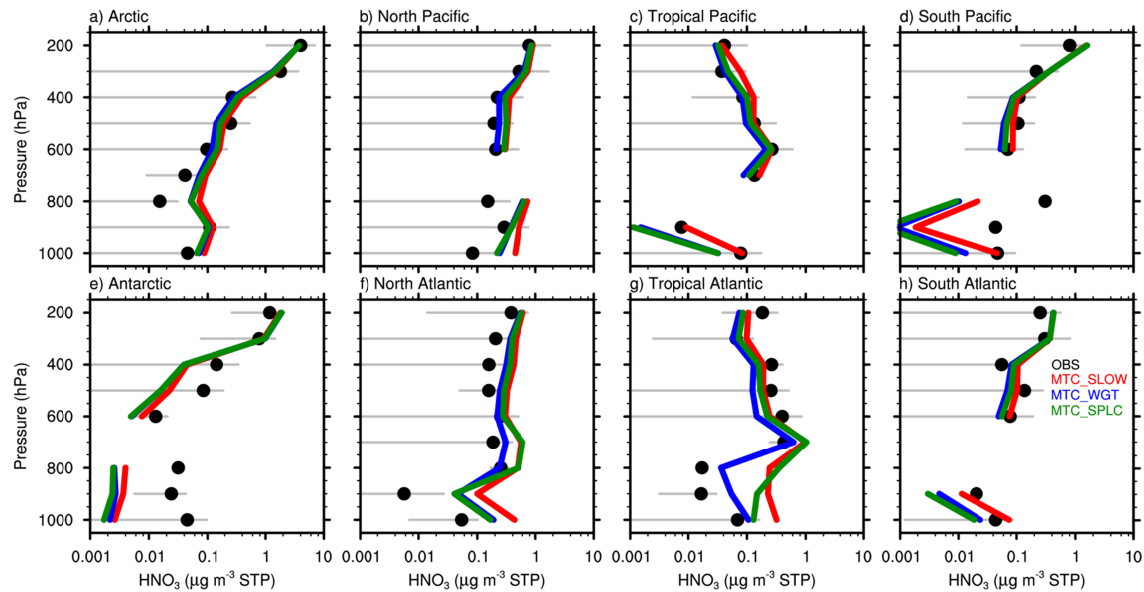
Figure S12. Vertical profiles of HNO₃ concentrations ($\mu g\ m^{-3}$ in STP) from model simulations (colored lines) and ATom campaign (black dots for mean values; grey lines for plus/minus one standard deviation of observations).
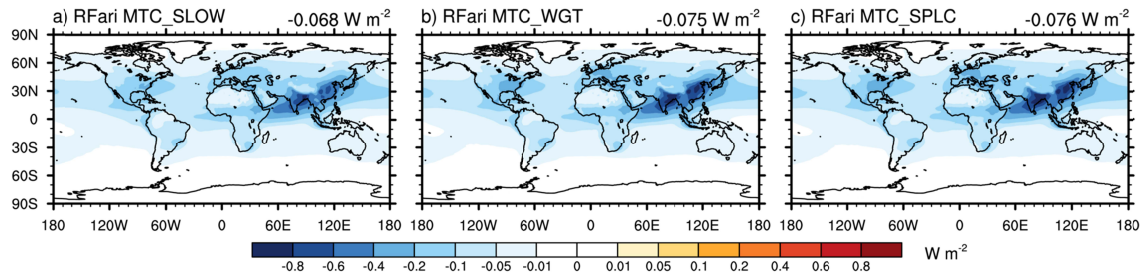
Figure S13. Spatial distributions of RFari of ammonium aerosols between 1850 and 2010.

200 **Table S1.** Mass Budgets of Nitrate in MTC_SPLAC and MTC_FAST

NO_3	MTC_FAST	MTC_SPLAC
Aqueous Chemistry (Tg N a^{-1})	26.8 (26.8, 0.1)	29.4 (29.3, 0.1)
Gas-aerosol Exchange (Tg N a^{-1})	–0.4 (–19.7, 19.2)	–7.9 (–21.0, 13.1)
Net Chemistry Production (Tg N a^{-1})	26.4 (7.1, 19.3)	21.1 (8.3, 13.2)
Dry Deposition (Tg N a^{-1})	11.3 (1.5, 9.8)	8.4 (1.7, 6.7)
Wet Deposition (Tg N a^{-1})	15.0 (5.6, 9.5)	13.1 (6.6, 6.5)
Burden (Tg N)	0.256 (0.061, 0.194)	0.191 (0.080, 0.111)
Lifetime (day)	3.54 (3.18, 3.68)	3.25 (3.51, 3.08)

201 Note. Values in parentheses are for the fine (accumulation and Aitken mode) and coarse
202 modes, respectively.

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204 **Table S2.** Mass Budgets of nss-Sulfate

nss-SO_4	Default	MZT	MTC_SLOW	MTC_WGT	MTC_SPLC
Emission (Tg S a^{-1})	1.81	1.81	1.81 (61.89)	1.81 (61.92)	1.81 (61.89)
Aqueous chemistry (Tg S a^{-1})	24.18	19.29	20.08	20.09	19.41
Gas-aerosol exchange (Tg S a^{-1})	15.80	12.93	12.65	12.51	12.60
Dry deposition (Tg S a^{-1})	8.79	8.11	8.37 (52.88)	8.24 (52.82)	8.24 (52.80)
Wet deposition (Tg S a^{-1})	33.12	26.36	26.99 (42.58)	26.96 (42.53)	27.01 (42.57)
Burden (Tg S)	0.784	0.703	0.706 (0.844)	0.702 (0.840)	0.705 (0.843)
Life time (day)	6.83	7.45	7.29 (3.23)	7.27 (3.22)	7.29 (3.23)

205 Note. Values in parentheses are for total sulfate, including ss-sulfate.

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Table S3. Tropospheric HNO_3 burden in the Three E3SM Experiments Compared with

Other Studies

	Burden (Tg N)[a]
MTC_SLOW	0.425
MTC_WGT	0.353
MTC_SPLC	0.389
Bian et al. (2017)	0.56 [0.15, 1.3][b]
	0.39 [0.15, 0.69][c]
Lu et al. (2021)	0.637
Zaveri et al. (2021)	0.422
Xu & Penner (2012)	0.30
Feng & Penner (2007)	0.37

[a]Values are calculated as pressure > 100 hPa for Bian et al. (2017), Lu et al. (2021), Zaveri et al. (2021), and this study; pressure > 150 hPa for Xu and Penner (2012); and pressure > 200 hPa for Feng and Penner (2007). [b]Values in brackets are minimum and maximum values, respectively. [c]We select 4 GCMs which simulate the formation of nitrate aerosols in both the fine and coarse modes and consider the heterogeneous reactions on dust and sea salt particles.

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232 **Table S4.** Mean Surface Molar Concentrations (ppb in STP; converted from µg m^{-3}) of

233 Aerosols and Precursor Gases for Figures 4 and 5.

Region	Specie	Observation	MTC_SLOW	MTC_WGT	MTC_SPLC
U.S.	NO_3	0.28	0.43	1.01	1.01
	HNO_3	0.33	1.30	0.97	0.97
	NO_3+HNO_3	0.61	1.72	1.98	1.98
	NH_4	0.97	0.92	1.54	1.54
	NH_3	2.17	1.90	1.47	1.47
	NH_4+NH_3	3.14	2.82	3.01	3.02
	SO_4	0.49	0.46	0.46	0.46
	SO_2	0.72	1.32	1.33	1.33
Europe	NO_3	0.56	0.53	1.18	1.17
	HNO_3	0.25	0.70	0.44	0.44
	NO_3+HNO_3	0.82	1.23	1.62	1.61
	NH_4	1.06	0.77	1.60	1.60
	NH_3	1.70	2.45	1.87	1.87
	NH_4+NH_3	2.76	3.22	3.47	3.47
	SO_4	0.42	0.34	0.35	0.35
	SO_2	0.47	1.00	1.00	1.00
East Asia	NO_3	0.44	0.53	1.21	1.19
	HNO_3	0.45	1.92	1.37	1.38
	NO_3+HNO_3	0.89	2.44	2.58	2.57
	NH_4	1.28	1.43	2.16	2.19
	NH_3	2.48	2.13	1.69	1.69
	NH_4+NH_3	3.76	3.56	3.84	3.87
	SO_4	0.87	0.83	0.84	0.84
	SO_2	2.46	2.37	2.38	2.39

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