

Supplementary

Table S1: Model performance for annual mean concentrations of ammonia (NH_3), total ammonia (TNH_x), total nitrate (TNO_3), ammonium (PNH_4), nitrate (PNO_3), sulfate (PSO_4) and accumulated annual wet deposition of WNH_x , WNO_x and WSO_x in 2010. MB: mean bias; ME: mean error; RMSE: root-mean-square error; MFB: mean fractional bias; MFE: mean fractional error; r : Pearson correlation coefficient.

	number of stations	MB*	ME*	RMSE*	MFB (%)	MFE (%)	r
NH_3	29	0.5	2.7	4.3	19.3	92.1	0.47
TNH_x	51	1.1	1.6	2.3	35.5	65.7	0.34
TNO_3	52	0.0	0.6	1.5	26.0	51.6	-0.04
PNH_4	32	0.5	0.5	0.7	45.7	47.5	0.80
PNO_3	25	1.5	2.7	3.2	54.6	104.1	-0.20
PSO_4	43	0.1	0.6	0.8	4.7	30.6	0.60
WNH_x	32	-120	133	178	-40.1	45.5	0.81
WNO_x	36	-142	143	206	-58.2	62.0	0.75
WSO_x	33	-115	117	227	-50.1	51.2	0.62

* Units: NH_3 : ppb, TNH_x and TNO_3 : $\mu\text{g N m}^{-3}$, PNH_4 , PNO_3 and PSO_4 : $\mu\text{g m}^{-3}$, WNH_x and WNO_x : mg N m^{-2} , WSO_x : mg S m^{-2}

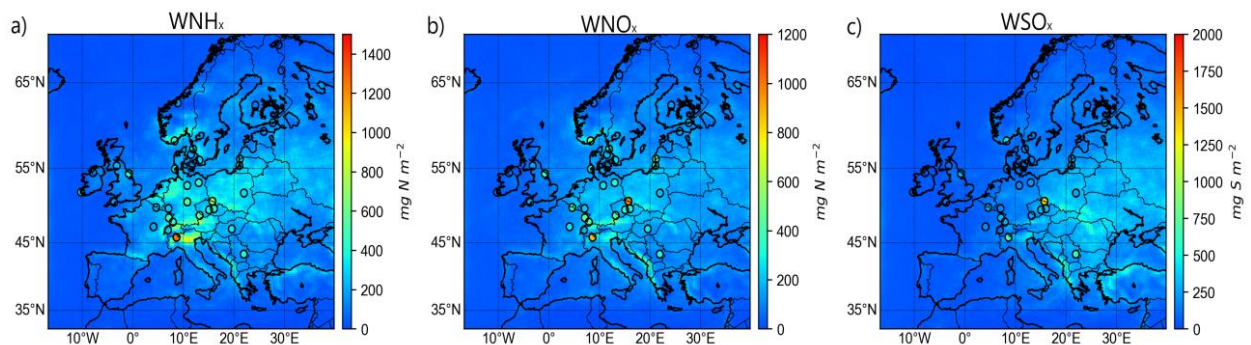


Figure S1: Accumulated annual wet deposition estimated for a) WNH_x (mg N m^{-2}), b) WNO_x (mg N m^{-2}), c) WSO_x (mg S m^{-2}) in 2010. The colored circles show the measurements.

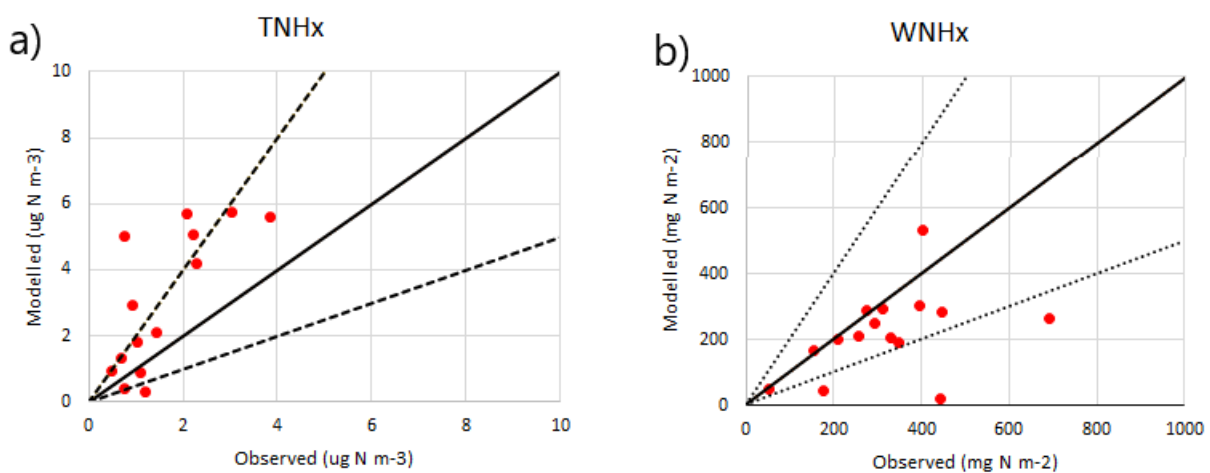
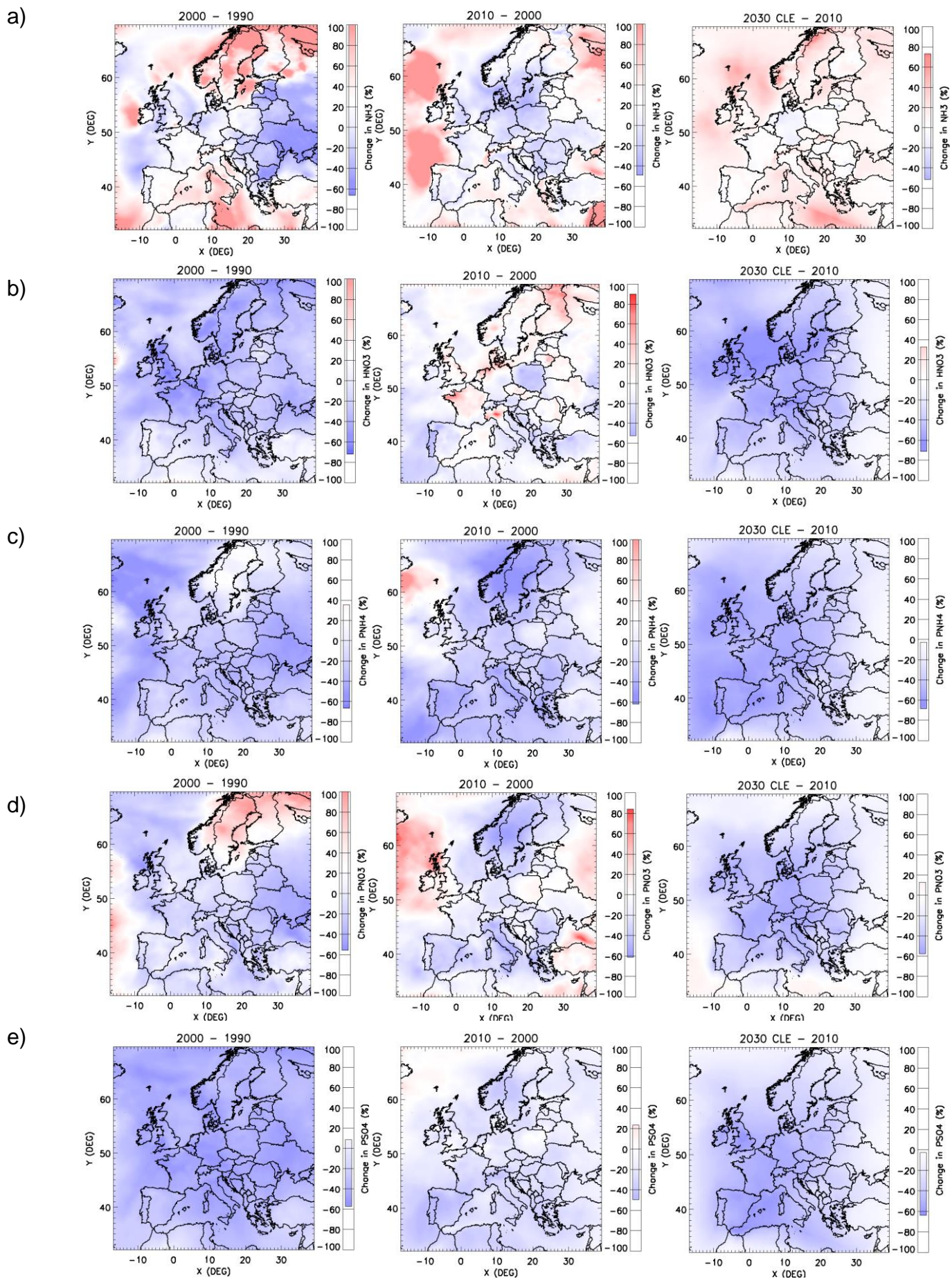


Figure S2: Modelled versus measured a) annual concentrations of TNH_x (mg N m^{-2}), b) accumulated annual wet deposition of WNH_x (mg N m^{-2}) at common sites in 2010.



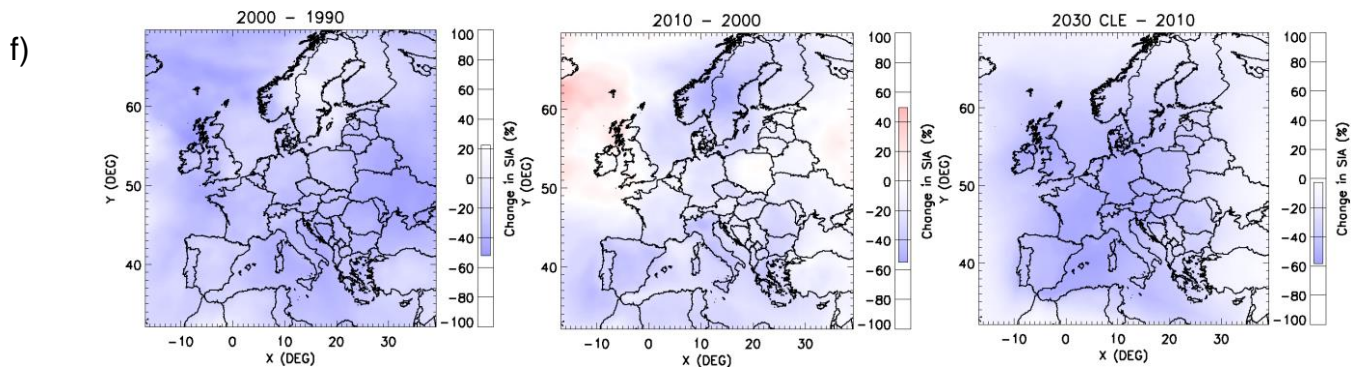


Figure S3: Relative changes (%) in annual average concentrations of a) NH_3 , b) HNO_3 , c) PNH_4 , d) PNO_3 , e) PSO_4 and f) SIA between 1990 and 2000 (left), 2000 and 2010 (middle) and 2010 and 2030_CLE (right).

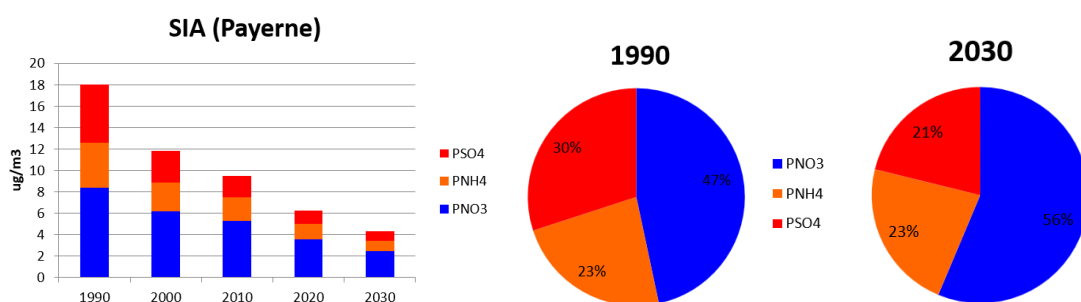
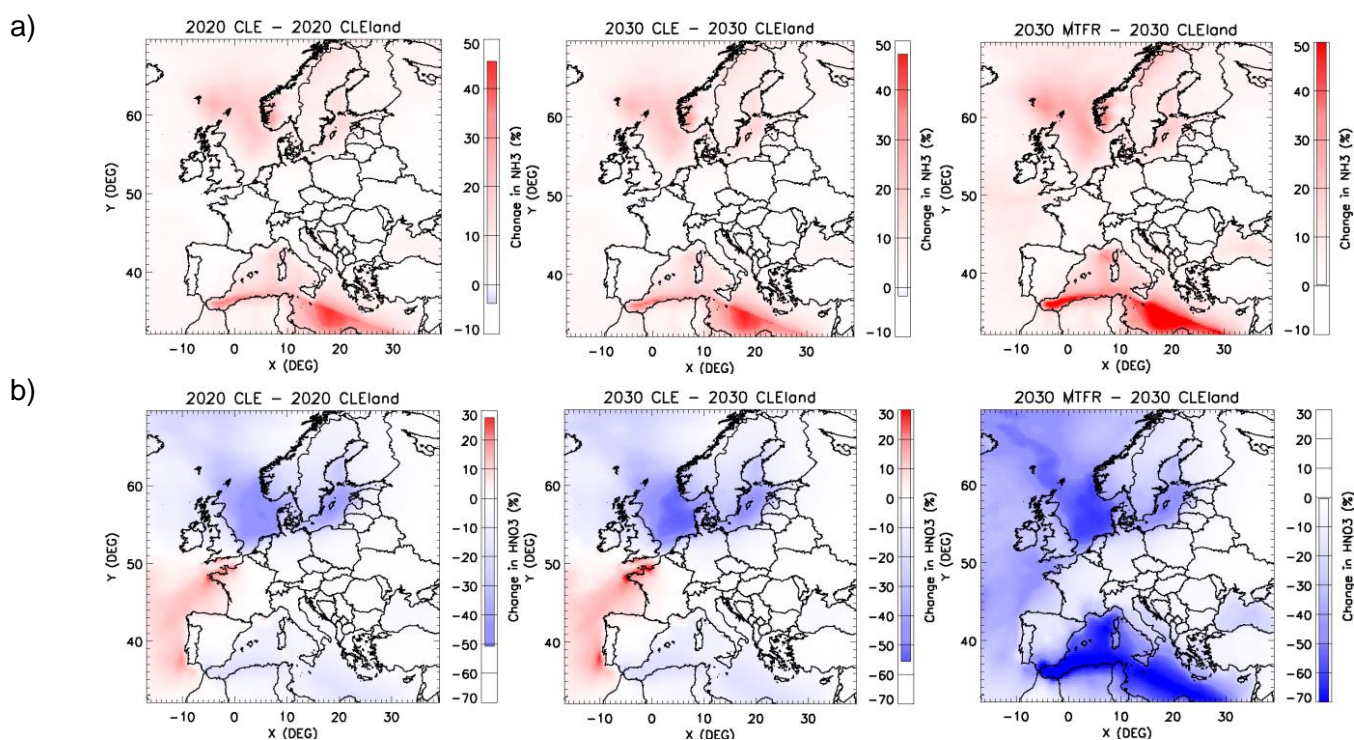
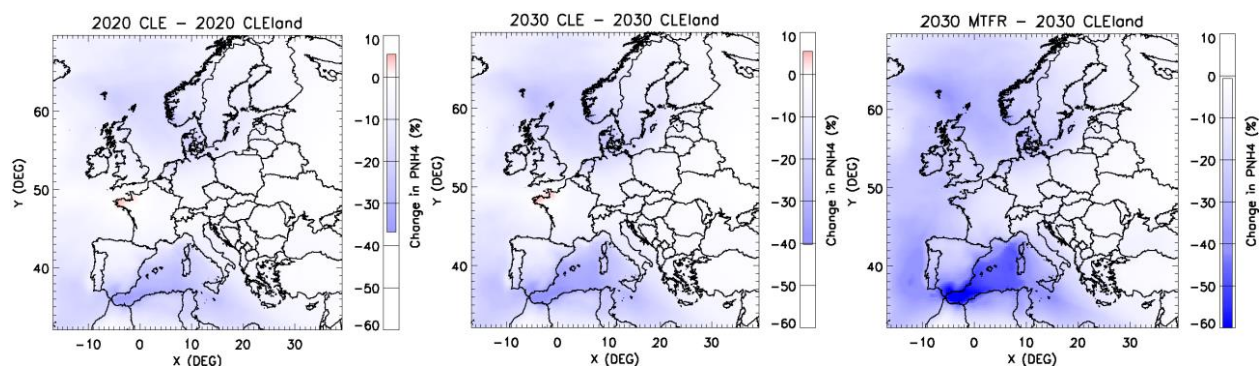


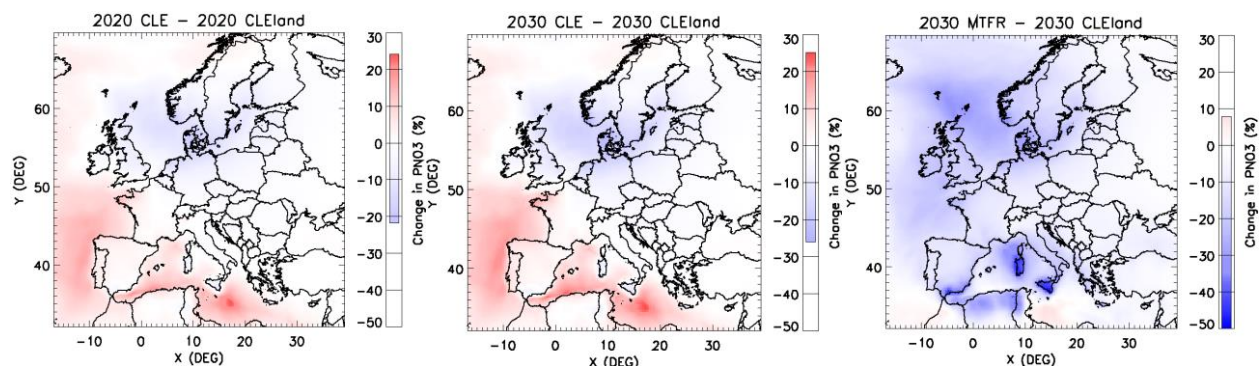
Figure S4: The modeled annual concentrations of SIA components between 1990 and 2030_CLE (left) and relative composition of SIA in 1990 (middle) and 2030_CLE (right) at Payerne (Switzerland).



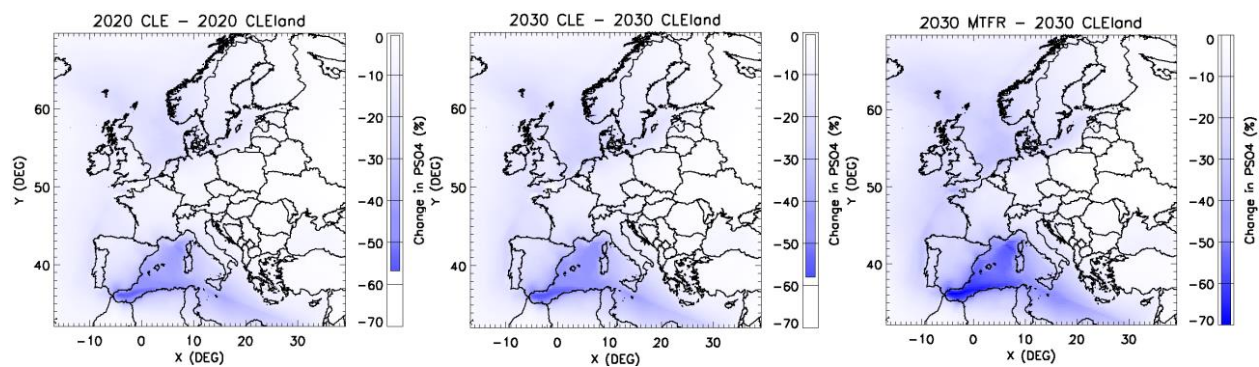
c)



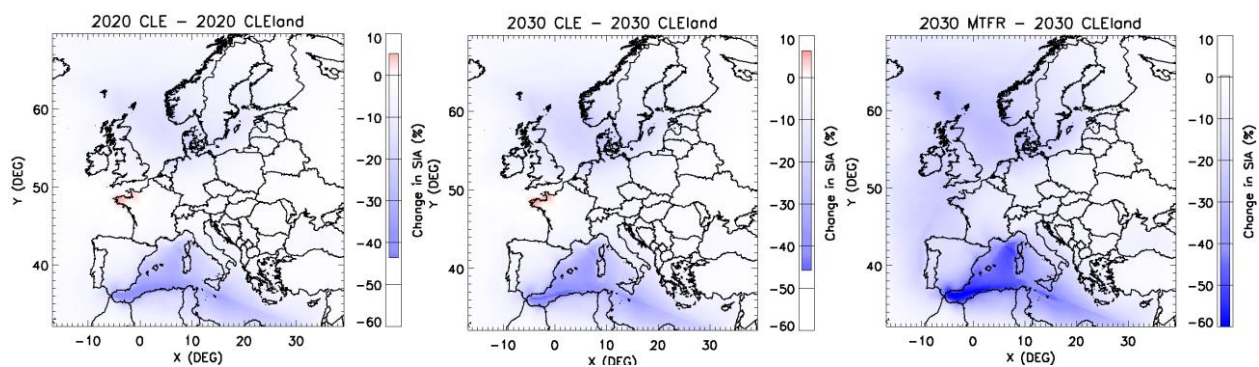
d)



e)



f)



g)

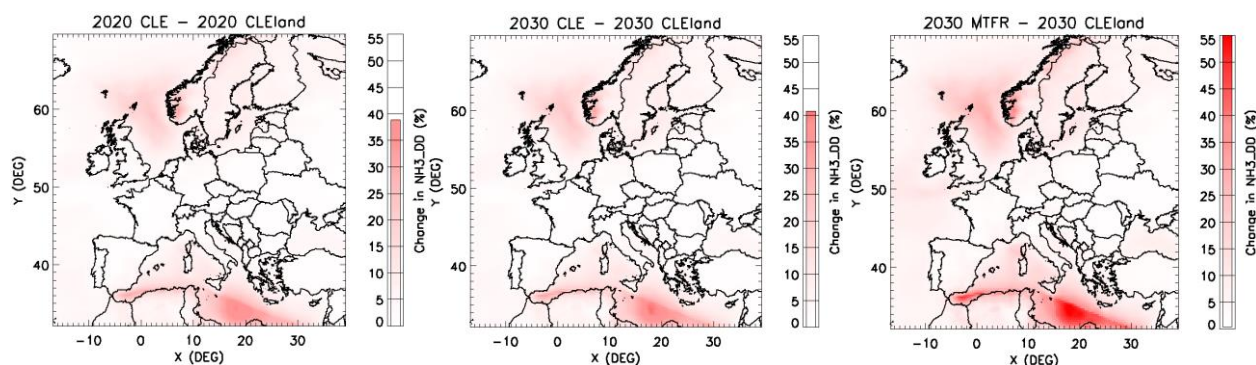


Figure S5: Effect of ship emissions (%) on annual average concentrations of a) ammonia (NH_3), b) nitric acid (HNO_3), c) ammonium (PNH_4), d) particulate nitrate (PNO_3), e) sulfate (PSO_4), f) secondary inorganic aerosols (SIA) and g) on annual dry deposition of ammonia in scenarios 2020_CLE (left), 2030_CLE (middle) and 2030_MTFR (right).

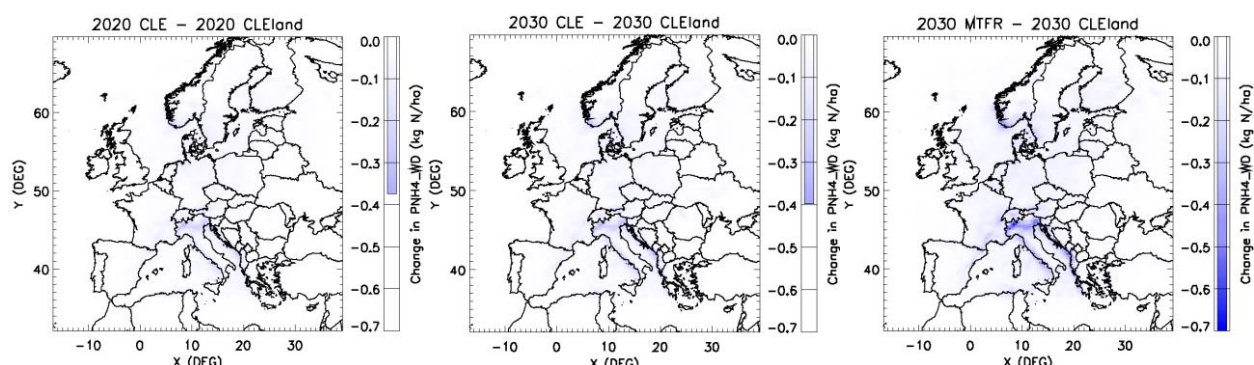


Figure S6: Effect of ship emissions on wet deposition of ammonium (kg N ha^{-1}) in scenarios 2020_CLE (left), 2030_CLE (middle) and 2030_MTFR (right).