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Supplement of

Role of ammonia in European air quality with changing land and ship emissions between 1990 and 2030

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Supplementary

Table S1: Information about the measurement sites for ammonia used in this study ((http://ebas.nilu.no, last access: 10 July 2020). The first two letters of the stations refer to the country names; AT: Austria, CH: Switzerland, DE: Germany, DK: Denmark, ES: Spain, GB: Great Britain, HU: Hungary, IT: Italy, MD: Moldavia, NL: The Netherlands, NO: Norway, PL: Poland, SE: Sweden, SK: Slovakia

Station	Station name	Instrument type	Resolution	Start time	End time
AT0002R	Illmitz	filter_3pack	1d	1/2/2010	12/31/2010
CH0002R	Payerne	CEH_denuder	2w	1/27/2010	12/15/2010
CH0005R	Rigi	CEH_denuder	2w	1/26/2010	12/14/2010
DE0001R	Westerland	filter_3pack	1d	1/2/2010	12/31/2010
DE0009R	Zingst	filter_3pack	1d	1/2/2010	12/31/2010
DK0003R	Tange	filter_3pack	1d	1/2/2010	12/31/2010
DK0005R	Keldsnor	filter_3pack	1d	1/2/2010	7/4/2010
DK0008R	Anholt	filter_3pack	1d	1/2/2010	12/31/2010
DK0012R	Risoe	filter_3pack	1d	7/30/2010	12/31/2010
DK0031R	Ulborg	filter_3pack	1d	1/2/2005	12/31/2010
ES0008R	Niembro	passive_sampler	1w	1/11/2010	12/20/2010
ES0009R	Campisabalos	passive_sampler	1w	1/11/2010	12/20/2010
GB0006R	Lough Navar	CEH_denuder	1mo	1/31/2010	11/30/2010
GB0013R	Yarner Wood	CEH_denuder	1mo	1/31/2010	11/30/2010
GB0014R	High Muffles	CEH_denuder	1mo	1/31/2010	11/30/2010
GB0048R	Auchencorth Moss	online_IC	1h	1/1/2010	12/31/2010
GB0054R	Glen Saugh	CEH_denuder	1mo	1/31/2010	11/30/2010
HU0002R	K-puszta	filter_3pack	1d	1/2/2010	12/31/2010
IT0001R	Montelibretti	denuder	1w	1/5/2010	12/30/2013
MD0013R	Leova II	filter_3pack	1d	1/2/2010	12/31/2010
NL0007R	Eibergen	tub_denuder	1h	1/1/2010	12/31/2010
NL0091R	De Zilk	denuder	1h	1/1/2010	12/31/2010
NO0001R	Birkenes	filter_3pack	1d	1/2/2010	1/31/2010
NO0002R	Birkenes II	filter_3pack	1d	1/2/2010	12/31/2010
NO0015R	Tustervatn	filter_3pack	1d	1/2/2010	12/31/2010
NO0039R	Kårvatn	filter_3pack	1d	1/2/2010	12/31/2010
NO0055R	Karasjok	filter_3pack	1d	1/2/2010	2/28/2010
NO0056R	Hurdal	filter_3pack	1d	1/2/2010	12/31/2010
PL0005R	Diabla Gora	filter_3pack	1d	1/2/2010	12/31/2010
SE0005R	Bredkälen	filter_3pack	1d	1/2/2010	12/31/2010
SE0011R	Vavihill	filter_3pack	1d	1/2/2010	12/31/2010
SE0012R	Aspvreten	filter_3pack	1d	1/2/2010	12/31/2010
SE0014R	Råö	filter_3pack	1d	1/2/2010	12/31/2010
SK0006R	Starina	filter_2pack	1d	1/2/2010	12/31/2010

Table S2: Model performance for annual mean concentrations of ammonia (NH_3), total ammonium (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	34	1.2	2.9	4.3	37.9	95.7	0.42
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: µgN m^{-3}, PNH_4, PNO_3 and PSO_4: µg m^{-3}, WNH_x and WNO_x: mg N m^{-2}, WSO_x : mg S m^{-2}

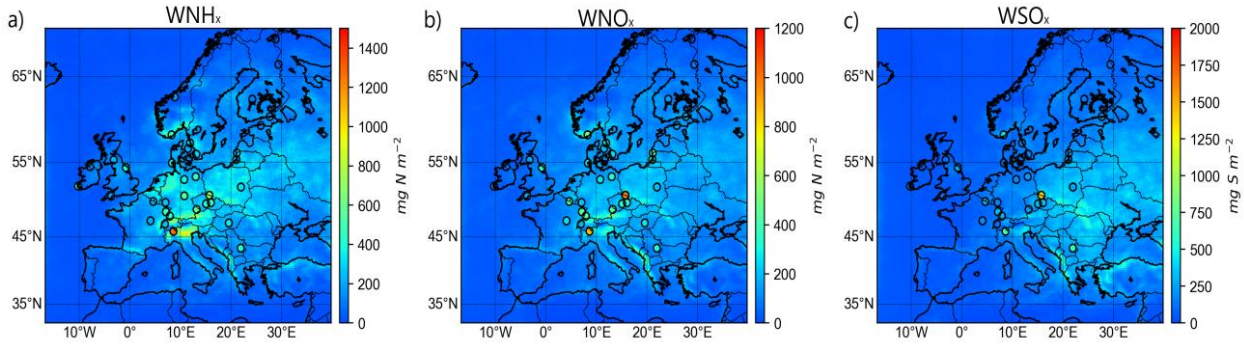


Figure S1: Accumulated annual wet deposition estimated for a) WNH_x (mg N m^{-2}), b) WNO_x (mg N m^{-2}), c) WSO_x (mg S m^{-2}) in 2010. The colored circles show the measurements.

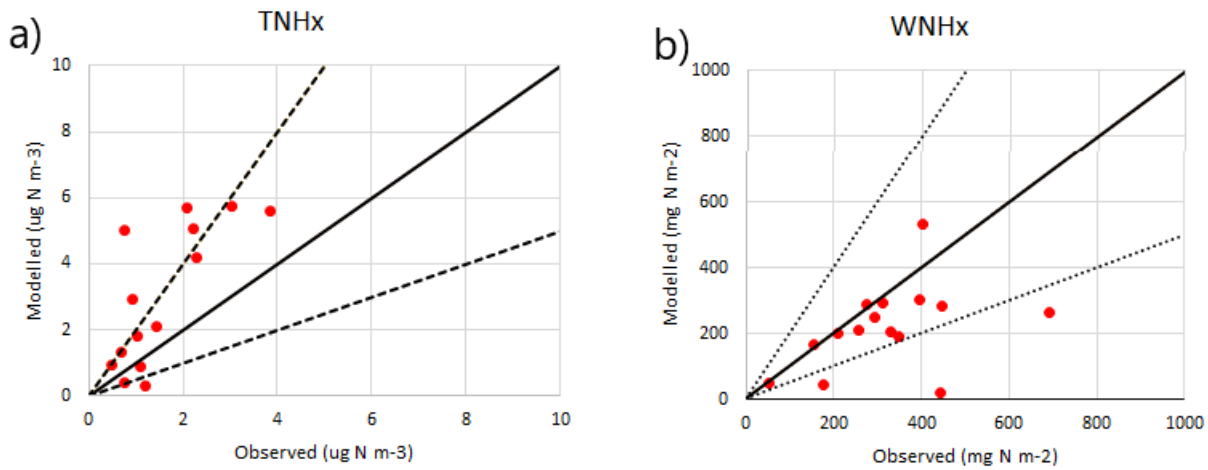
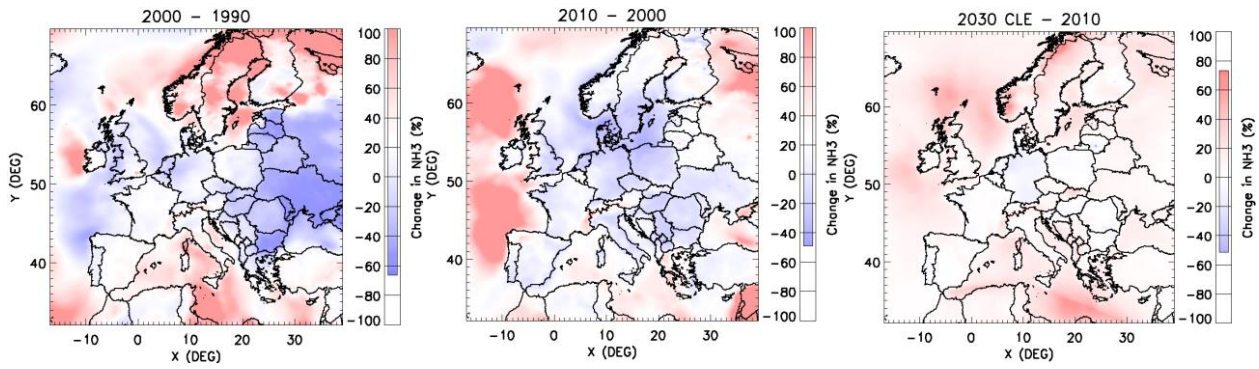
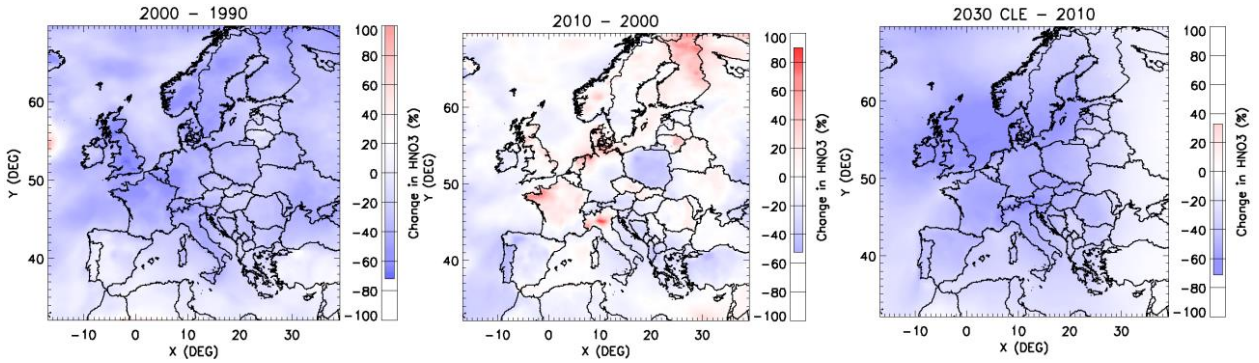


Figure S2: Modelled versus measured a) annual concentrations of TNH_x (µgN m^{-3}), b) accumulated annual wet deposition of WNH_x (mg N m^{-2}) at common sites in 2010. Measurements are from the EMEP network (see Section 3.1 in the main text).

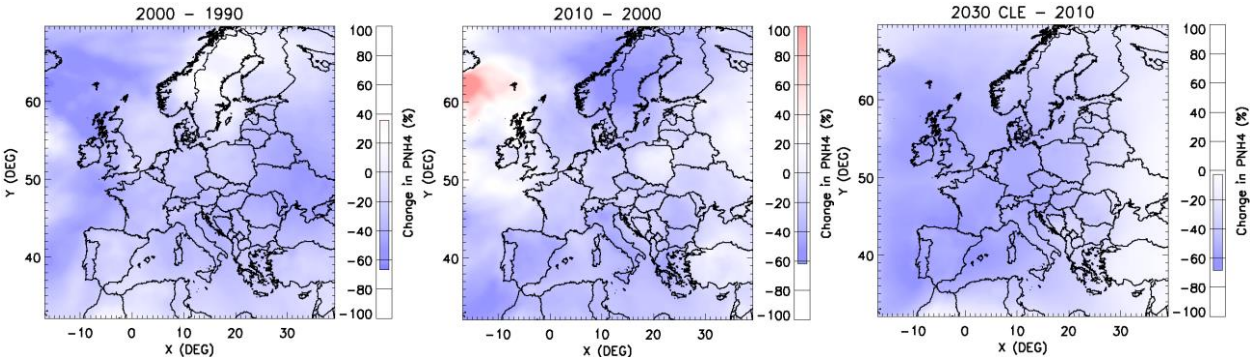
a)


2000 – 1990
2010 – 2000
2030 CLE – 2010
Y (DEG)
X (DEG)
Change in NH3 (%)

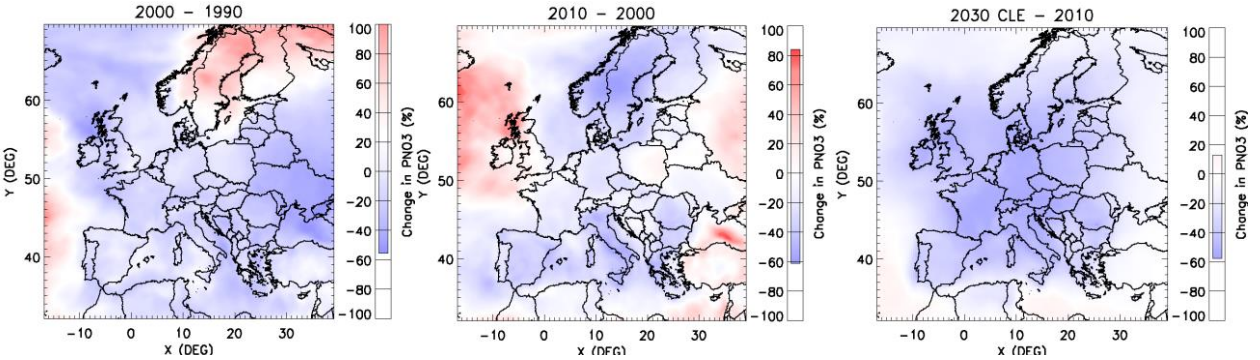
b)


2000 – 1990
2010 – 2000
2030 CLE – 2010
Y (DEG)
X (DEG)
Change in HNO3 (%)

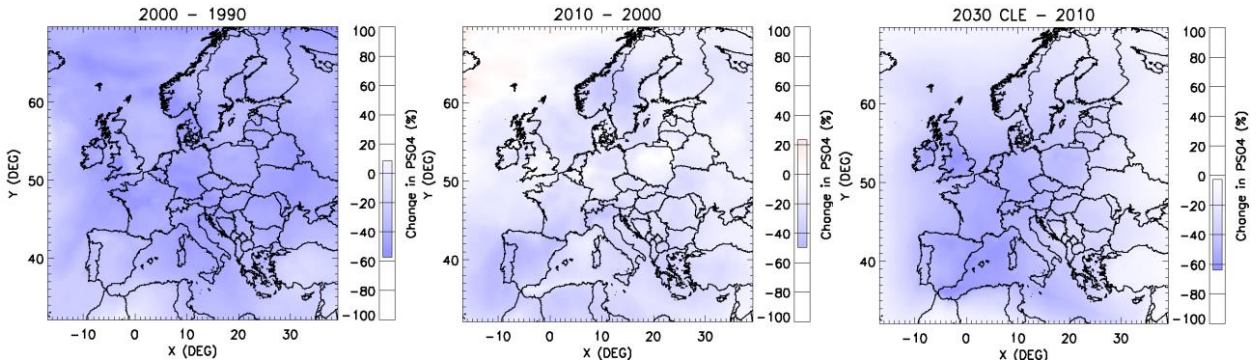
c)


2000 – 1990
2010 – 2000
2030 CLE – 2010
Y (DEG)
X (DEG)
Change in PNH4 (%)

d)


2000 – 1990
2010 – 2000
2030 CLE – 2010
Y (DEG)
X (DEG)
Change in PNO3 (%)

e)


2000 – 1990
2010 – 2000
2030 CLE – 2010
Y (DEG)
X (DEG)
Change in PSO4 (%)

f)

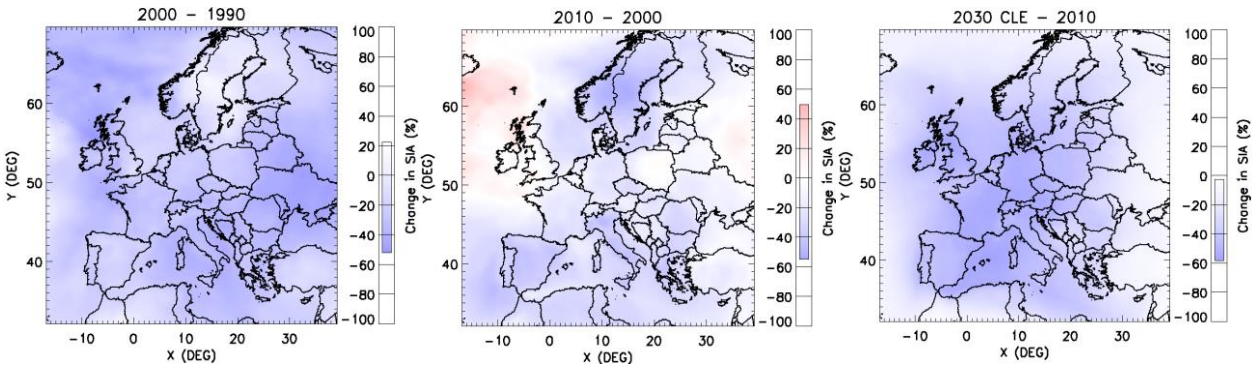


Figure S3: Relative changes (%) in annual average concentrations of a) NH_3, b) HNO_3, c) PNH_4, d) PNO_3, e) PSO_4 and f) SIA between 1990 and 2000 (left), 2000 and 2010 (middle) and 2010 and 2030_CLE (right).

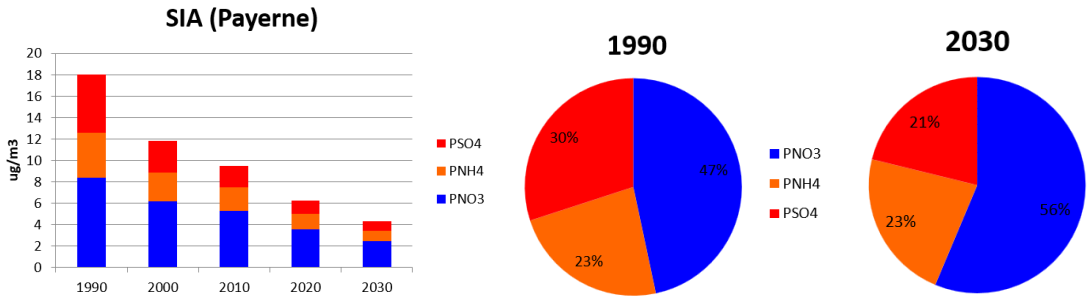
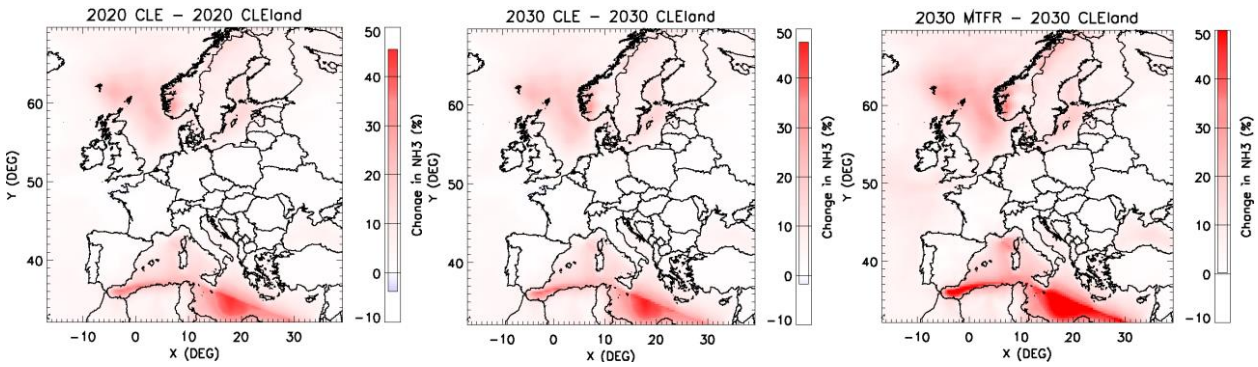
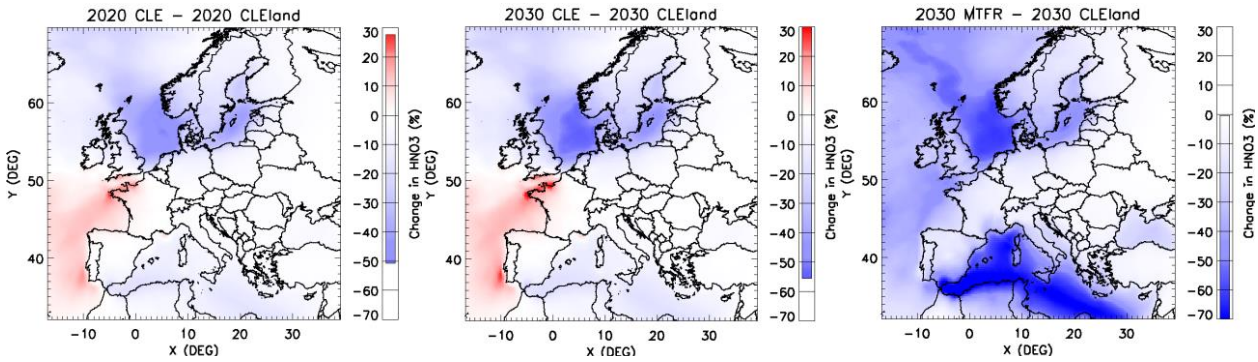


Figure S4: The modeled annual concentrations of SIA components between 1990 and 2030_CLE (left) and relative composition of SIA in 1990 (middle) and 2030_CLE (right) at Payerne (Switzerland).

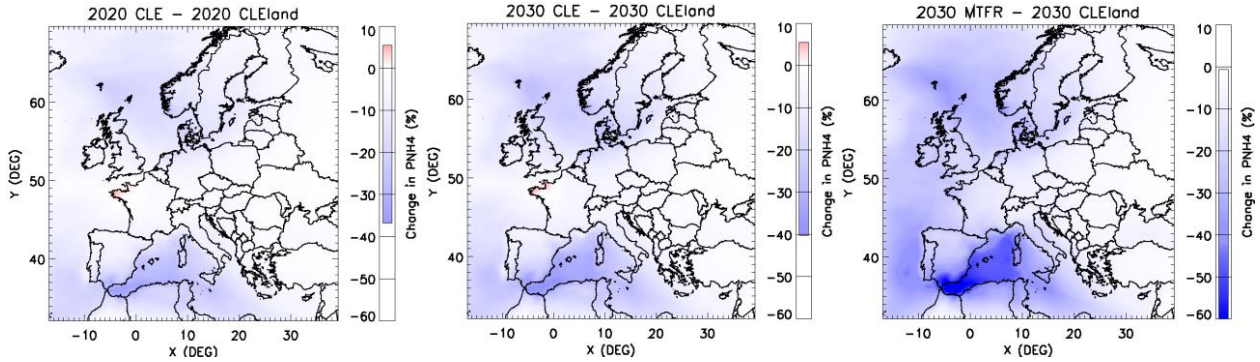
a)



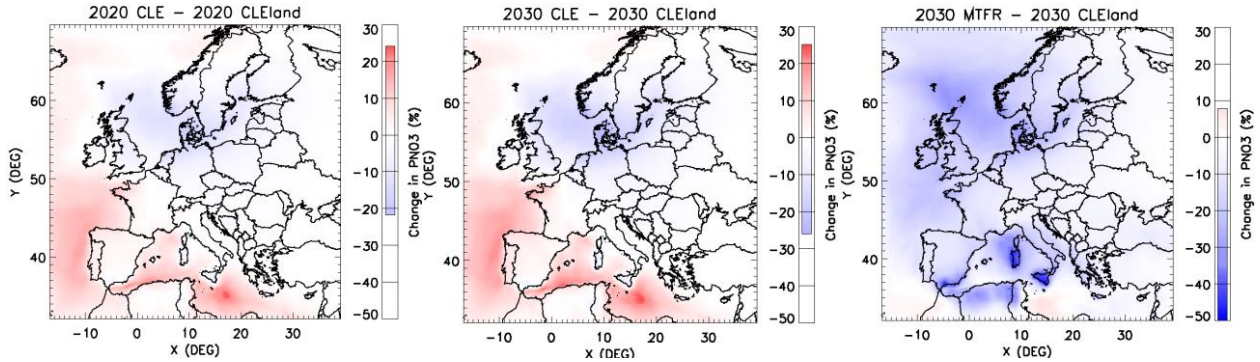
b)



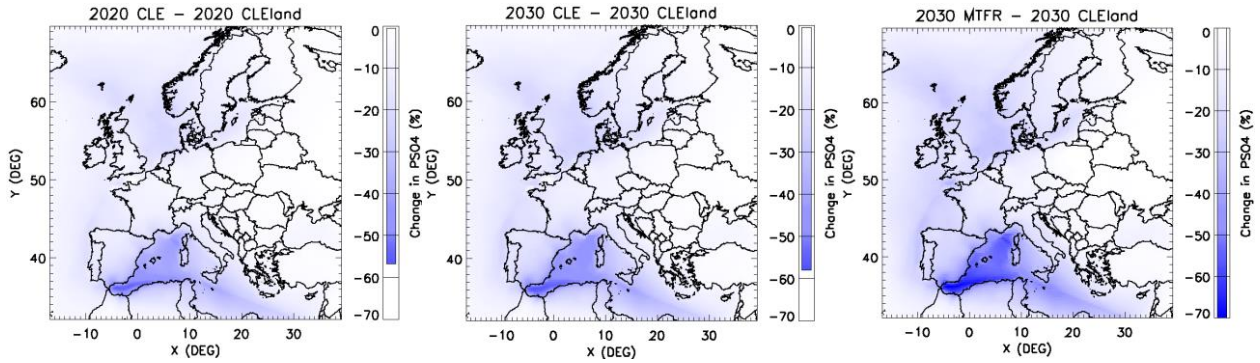
c)


2020 CLE – 2020 CLEland
2030 CLE – 2030 CLEland
2030 MTFR – 2030 CLEland
Change in PNH4 (%)
X (DEG)
Y (DEG)

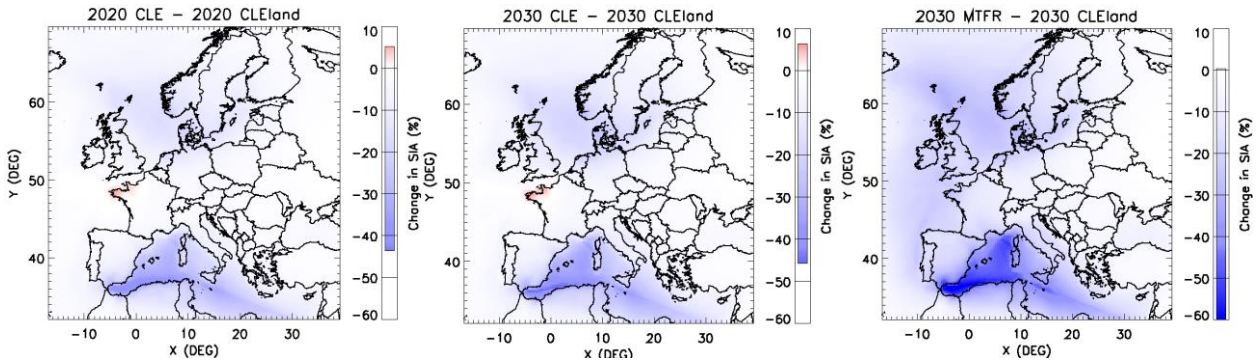
d)


2020 CLE – 2020 CLEland
2030 CLE – 2030 CLEland
2030 MTFR – 2030 CLEland
Change in PNO3 (%)
X (DEG)
Y (DEG)

e)


2020 CLE – 2020 CLEland
2030 CLE – 2030 CLEland
2030 MTFR – 2030 CLEland
Change in PSO4 (%)
X (DEG)
Y (DEG)

f)


2020 CLE – 2020 CLEland
2030 CLE – 2030 CLEland
2030 MTFR – 2030 CLEland
Change in SIA (%)
X (DEG)
Y (DEG)

g)

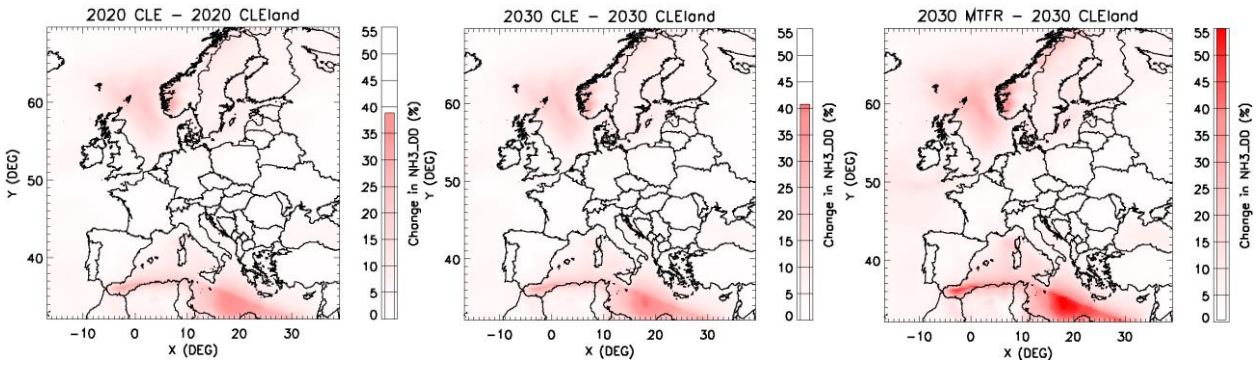


Figure S5: Effect of ship emissions (%) on annual average concentrations of a) ammonia (NH_3), b) nitric acid (HNO_3), c) ammonium (PNH_4), d) particulate nitrate (PNO_3), e) sulfate (PSO_4), f) secondary inorganic aerosols (SIA) and g) on annual dry deposition of ammonia in scenarios 2020_CLE (left),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

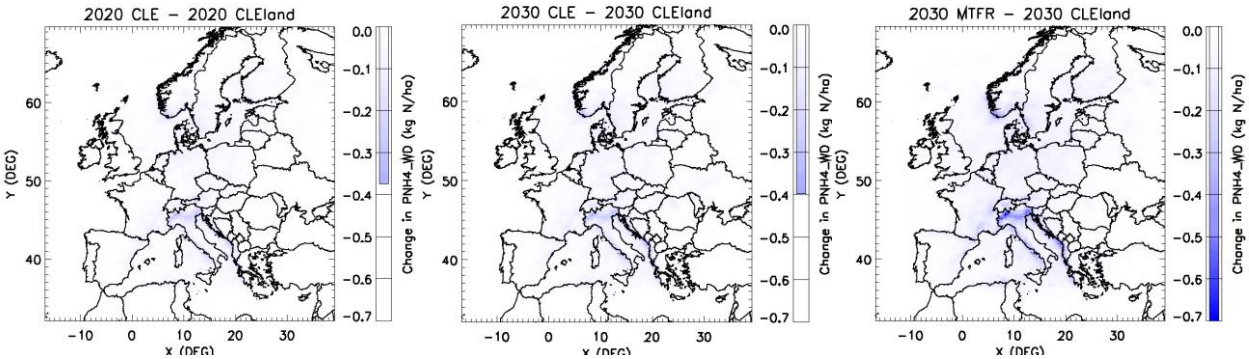


Figure S6: Effect of ship emissions on wet deposition of ammonium (kg N ha^{-1}) in scenarios 2020_CLE (left), 2030_CLE (middle) and 2030_MTFR (right).