Supplement of Atmos. Chem. Phys., 17, 1125–1142, 2017
http://www.atmos-chem-phys.net/17/1125/2017/
doi:10.5194/acp-17-1125-2017-supplement

Atmospheric Chemistry and Physics
Open Access

Supplement of

Chemistry–climate interactions of aerosol nitrate from lightning

Holger Tost

Correspondence to: Holger Tost (tosth@uni-mainz.de)

1 LNO_x emissions

1.1 ARG - preindustrial

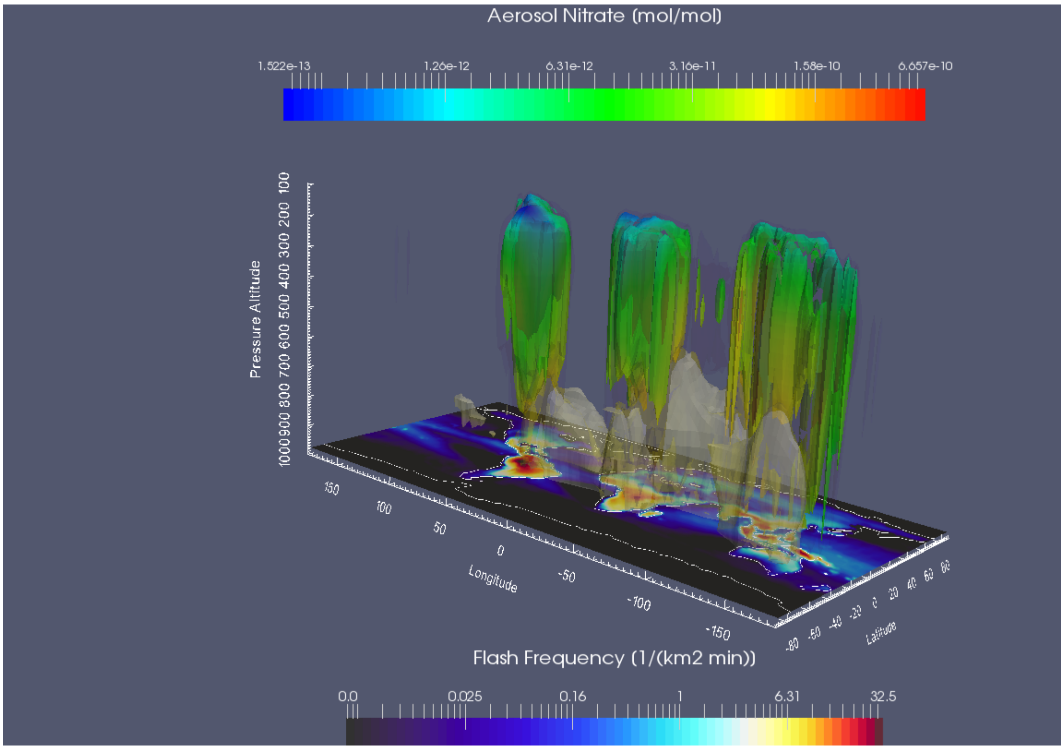

Figure S1: 3D visualisation of LNO_x emissions (coloured isosurface of $1 \cdot 10^{-16}$ kg/(m^{3}s) and darker shaded isosurface of $3 \cdot 10^{-16}$ kg/(m^{3}s)) and the total aerosol nitrate mixing ratios (gray isosurface of 0.1 ppb$_v$). Additionally, the mean flash rate in 1/(km^2 min) is depicted by the 2D slice at the bottom. Note the logarithmic scaling of both colour bars. The figure depicts preindustrial conditions for the ARG model configuration.

1.2 KK - present day

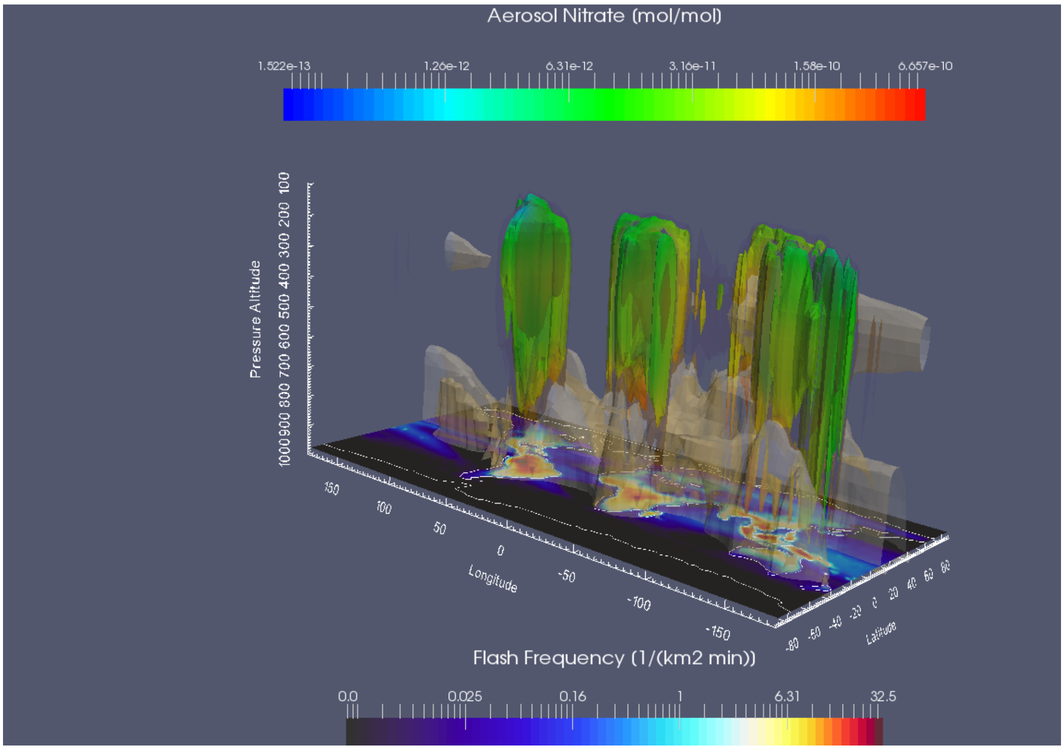

Figure S2: As Fig. 1, but for present day conditions with the KK model configuration.

1.3 KK - preindustrial

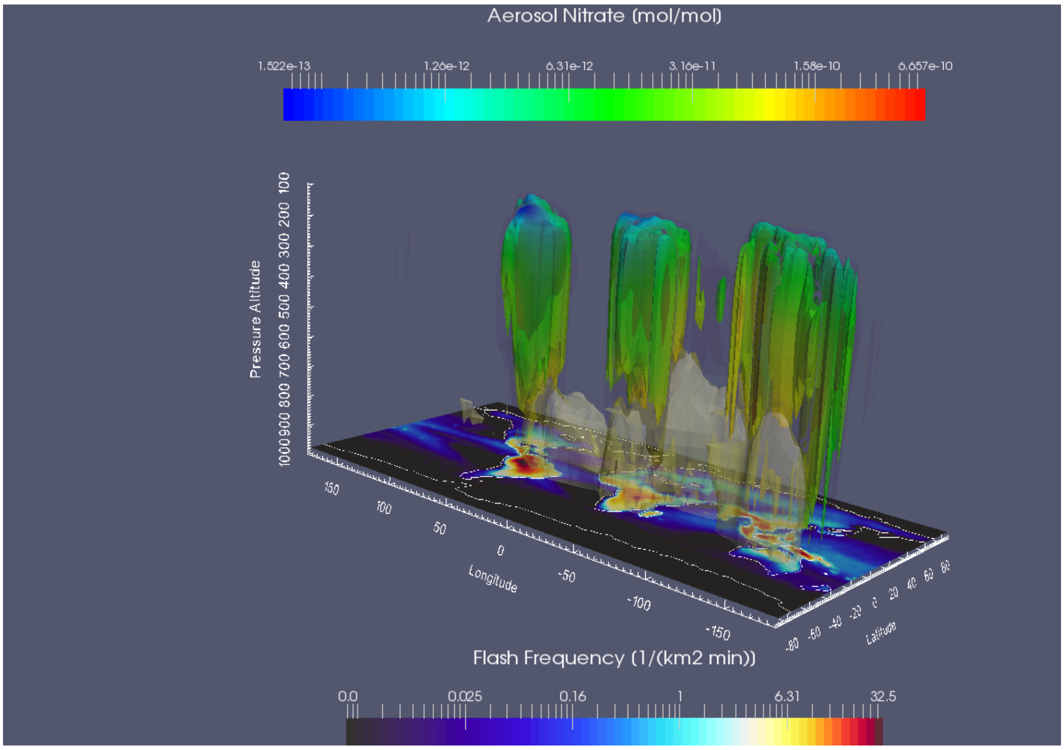

Figure S3: As Fig. 1, but for preindustrial conditions with the KK model configuration.

2 Nitrate distributions

2.1 Budget tables

Table S1: Tropospheric budget in the kappa-koehler simulation of the important highly oxidies nitrogen species, i.e. gaseous HNO_3, gaseous N_2O_5, aerosol NO_3^- and the sum of those three compounds. All values are given in Gg N (except for the relative differences which are provided in %) and are globally and vertically integrated over the whole and the upper troposphere (500hPa up to the tropopause).

	HNO_3	N_2O_5	NO_3^-	Total N(V+)
Present day				
Absolute values (with LNO_x emissions):				
Tropospheric Column burden	407	8.1	143	557
UT Column burden	177	5.1	27.5	209
Absolute differences due to LNO_x emissions:				
Tropospheric Column burden	164	3.9	23.3	191
UT Column burden	109	3.6	8.7	122
Tropospheric Column burden	40.3	48.7	16.4	34.3
UT Column burden	61.9	70.9	31.6	58.2
Preindustrial conditions				
Absolute values (with LNO_x emissions):				
Tropospheric Column burden	237	4.5	67.8	310
UT Column burden	142	4.1	14.9	161
Absolute differences due to LNO_x emissions:				
Tropospheric Column burden	157	3.8	27.8	189
UT Column burden	109	3.5	8.3	121
Relative differences in (%) due to LNO_x emissions:				
Tropospheric Column burden	66.2	82.6	41.0	61.0
UT Column burden	76.7	86.0	55.4	74.9

Table S2: Loss processes for the dominant N(V+) compounds for the present day and preindustrial ARG simulations including and excluding the effect of LNO_x emissions. All values are given in Tg N/yr (except for the relative differences which are provided in %) and are globally integrated.

Sinks	Dry deposition (gaseous HNO_3)	Dry deposition (aerosol NO_3^-)	Wet deposition	Sedi-mentation
Present day				
Absolute values (with LNO_x):	6.03	1.13	25.48	8.24
Absolute change due to LNO_x:	0.13	0.08	4.44	0.70
Relative change due to LNO_x:	2.09	7.37	17.43	8.54
Preindustrial conditions				
Absolute values (with LNO_x):	1.227	0.27	8.90	2.45
Absolute chage due to LNO_x:	0.20	0.08	4.46	0.78
Relative change due to LNO_x:	16.54	29.78	50.07	31.63

Table S3: Loss processes for the dominant N(V+) compounds for the present day and prein-dustrial kappa-koehler simulations including and excluding the effect of LNO_x emissions. All values are given in Tg N/yr (except for the relative differences which are provided in %) and are globally integrated.

Sinks	Dry deposition (gaseous HNO_3)	Dry deposition (aerosol NO_3^-)	Wet deposition	Sedi-mentation
Present day				
Absolute values (with LNO_x):	6.23	1.22	25.48	8.27
Absolute change due to LNO_x:	0.27	0.17	4.48	0.71
Relative change due to LNO_x:	4.35	14.08	17.58	8.64
Preindustrial conditions				
Absolute values (with LNO_x):	1.23	0.27	8.90	2.46
Absolute change due to LNO_x:	0.20	0.08	4.47	0.79
Relative change due to LNO_x:	16.22	29.89	50.15	32.10

2.2 Geographical distributions

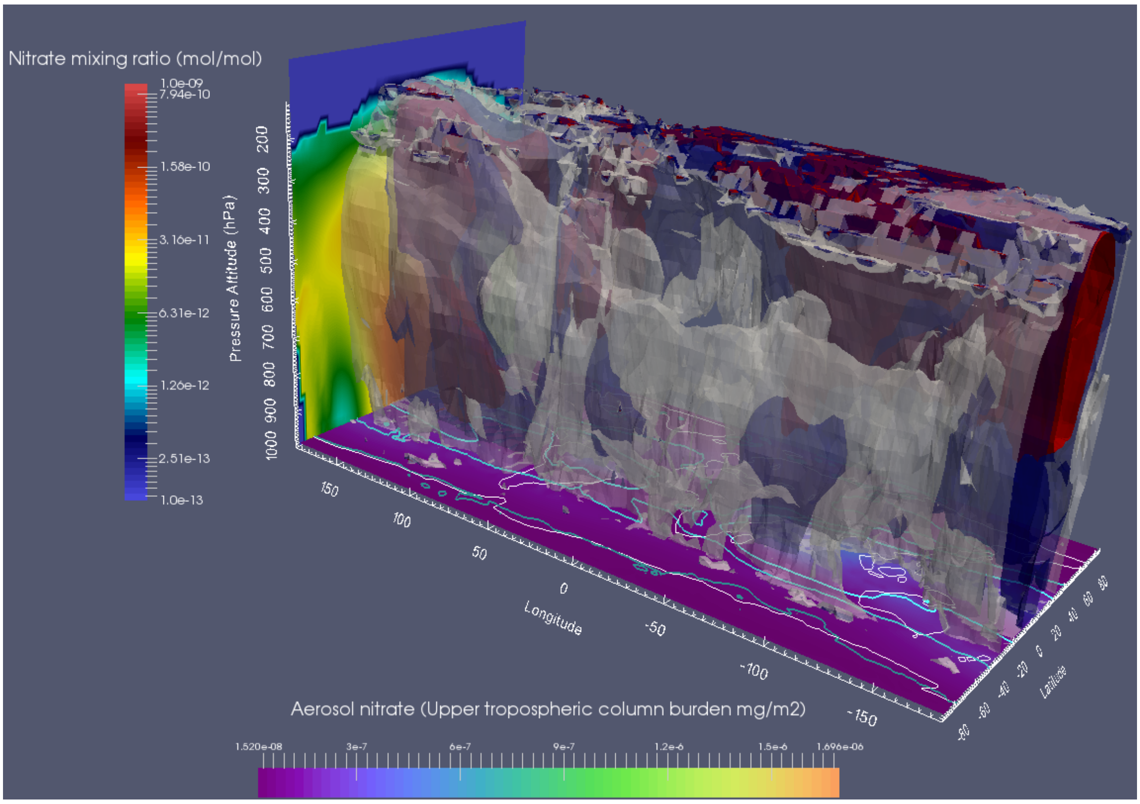

Figure S4: 3D visualisation of the relative differences in tropospheric aerosol nitrate mix-ing ratios between the simulations with and without LNO_x emissions to the simulation including LNO_x emissions. The white isosurface depicts a relative difference of 30%, the blue isosurface of 45%, and the red isosurface of 60%. Additionally, the upper tropospheric aerosol nitrate column burden (in mg/m^2) between 500 hPa and the tropopause is depicted by the coloured panel at the bottom of the graph. The turquoise contour lines depict relative differences of 20%, 40% and 60% difference in this column burden between the two simulations. The differences are calculated with the ARG configuration for preindustrial conditions.

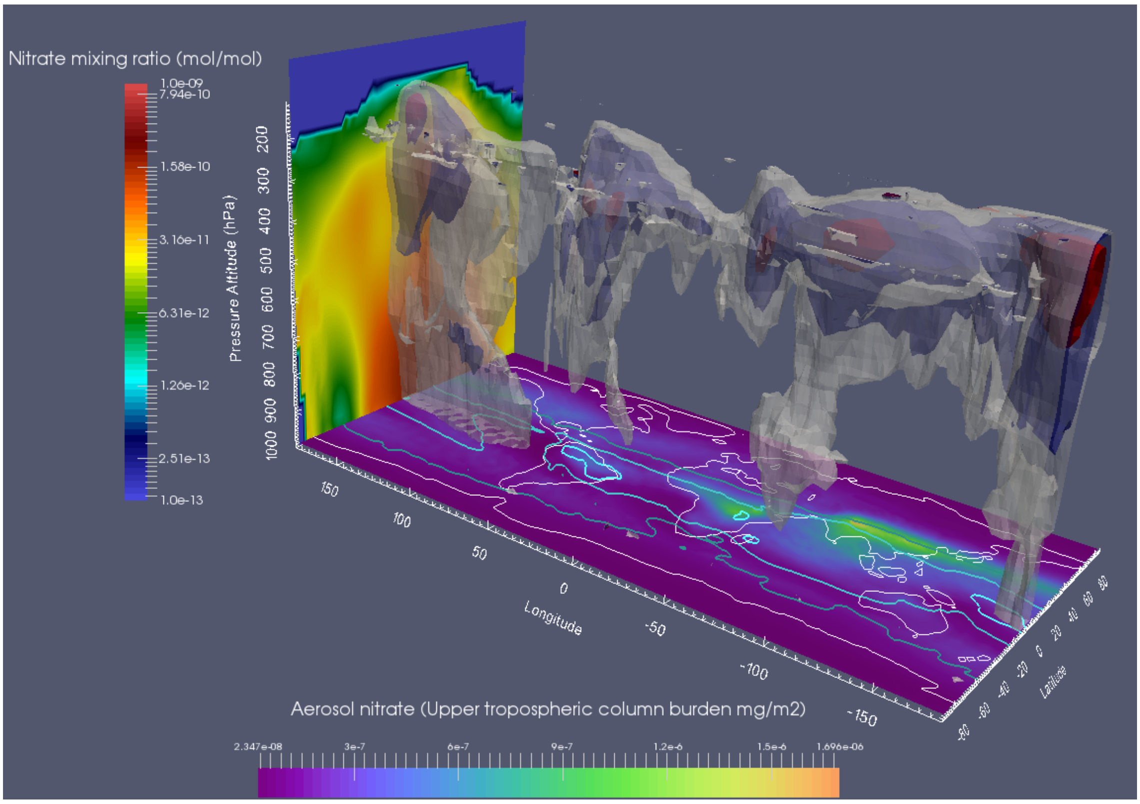

Figure S5: As Fig. 4, but with the KK configuration for present day conditions.

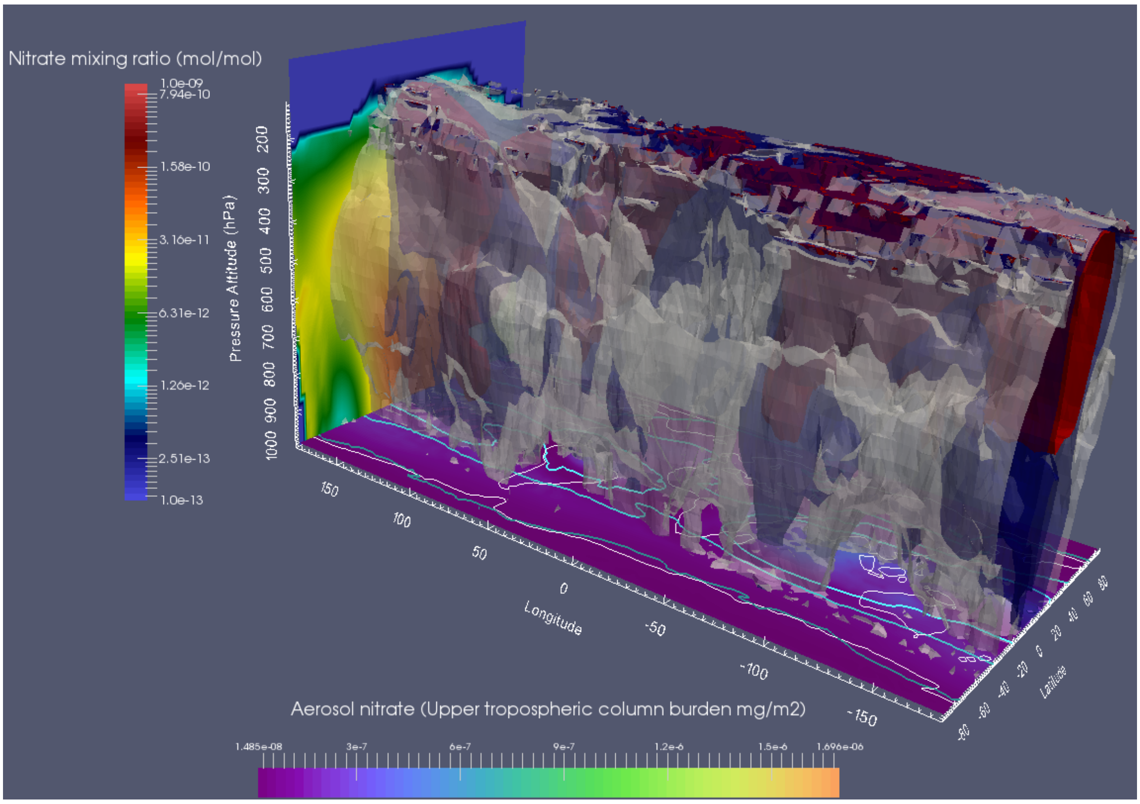

Figure S6: As Fig. 4, but with the KK configuration for preindustrial conditions.

3 Size distributions

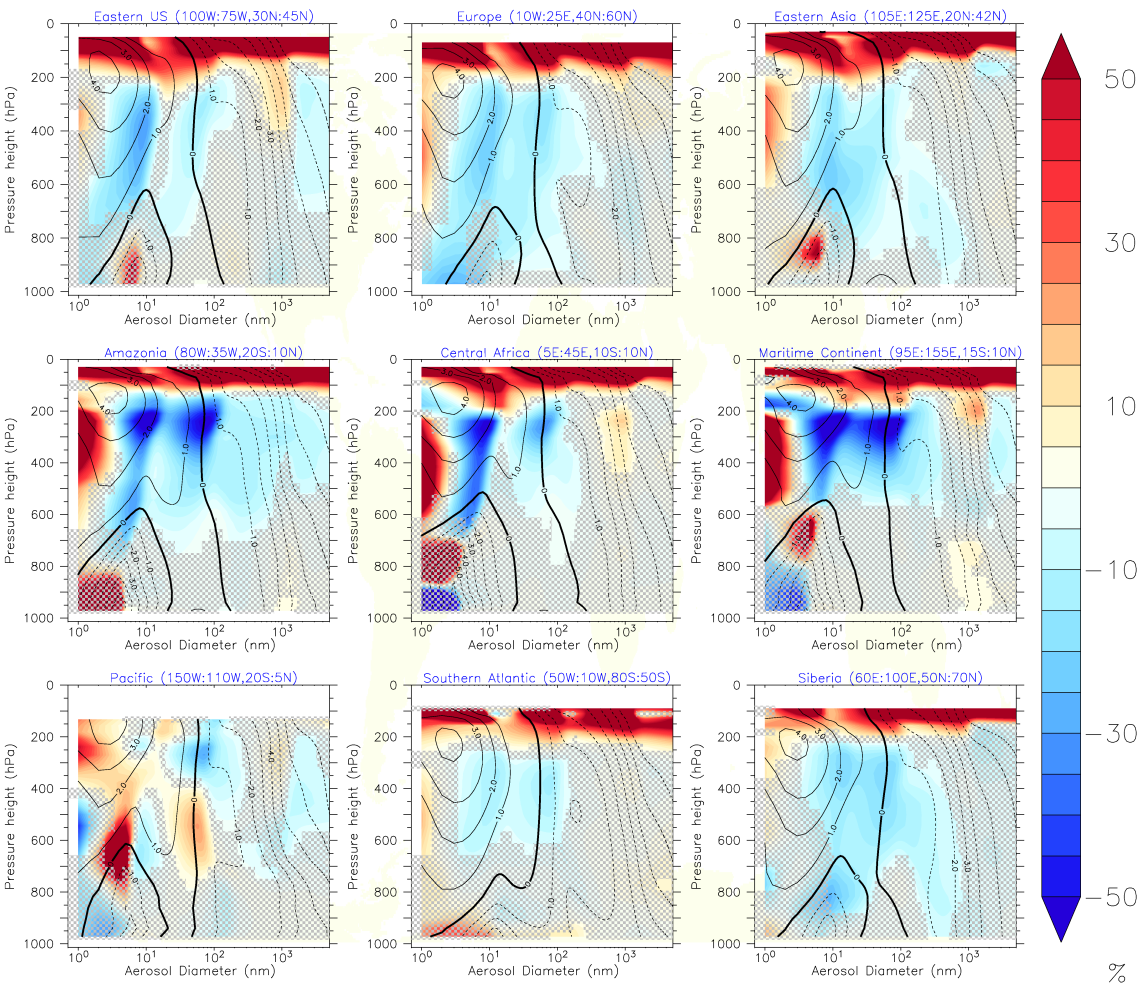

Figure S7: 9 panel plot of the two dimensional aerosol size distribution percentage differences (spatial and regional average for the respective region). Depicted are values (NOLNOX - LNOX)/LNOX * 100. Overlayed are the contours of the absolute values of the size distributions as calculated from the spatial and temporal mean in particles/cm^3. The figure depicts the preindustrial conditions for the ARG configuration.

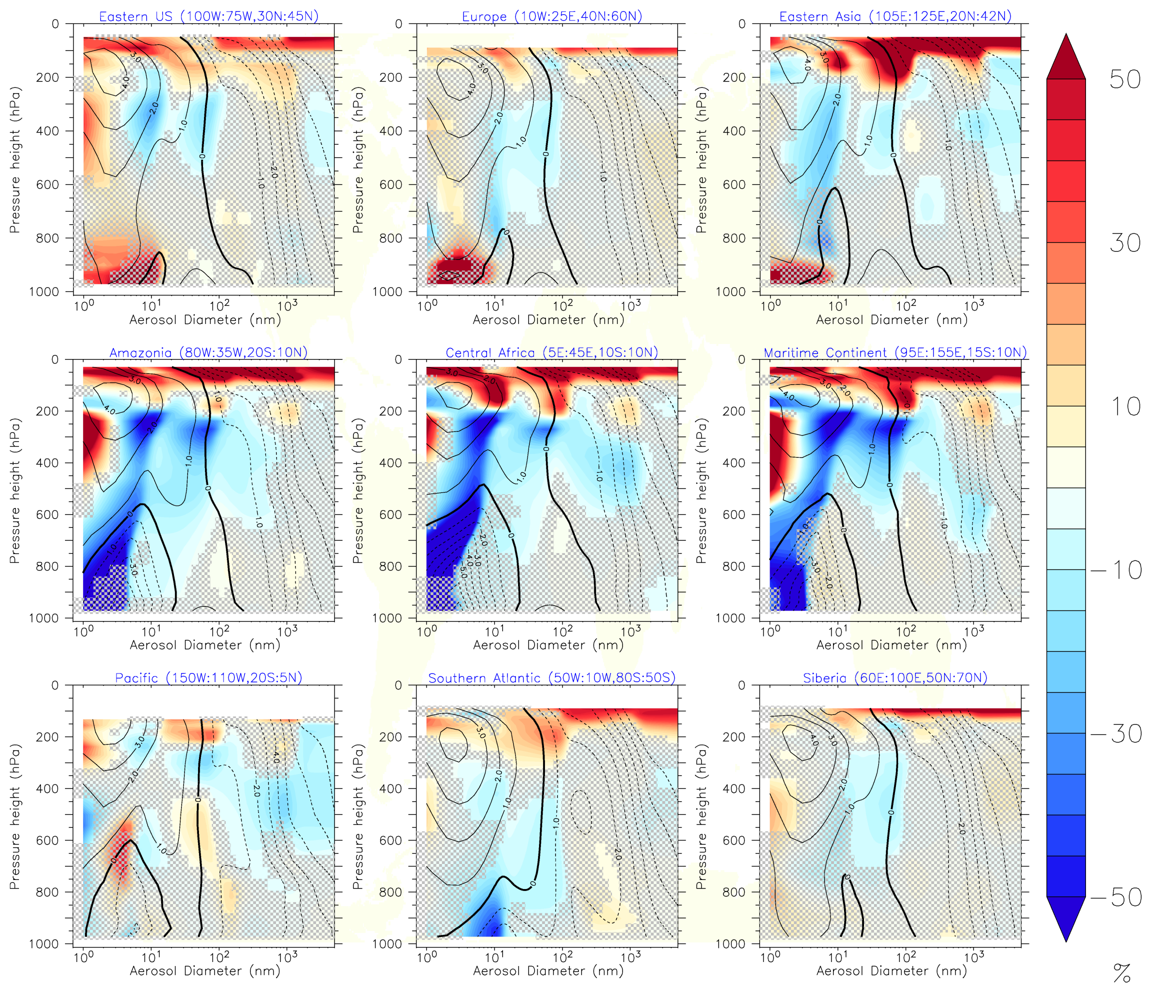

Figure S8: As Fig. 7, but for present day conditions with the KK model configuration.

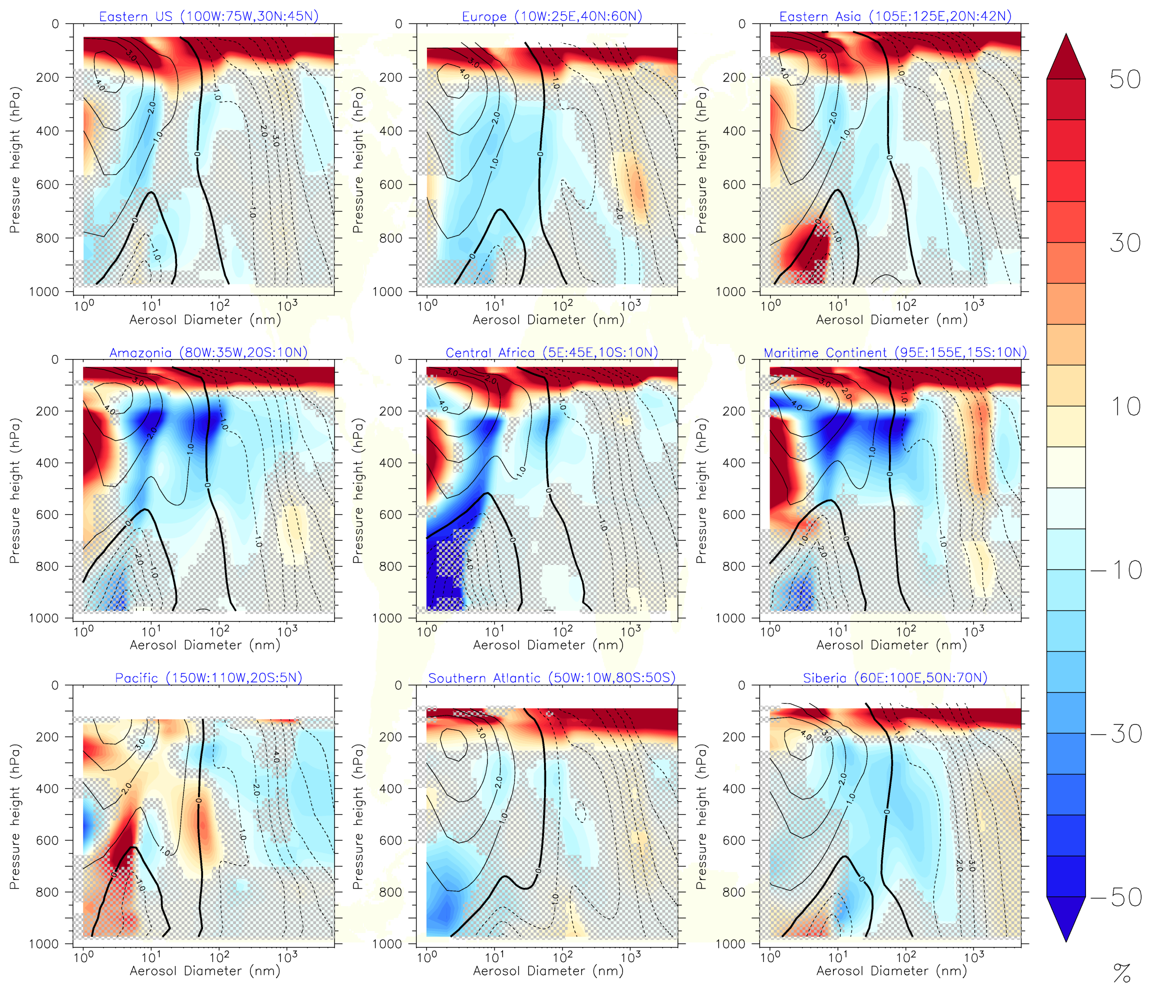

Figure S9: As Fig. 7, but for preindustrial conditions with the KK model configuration.

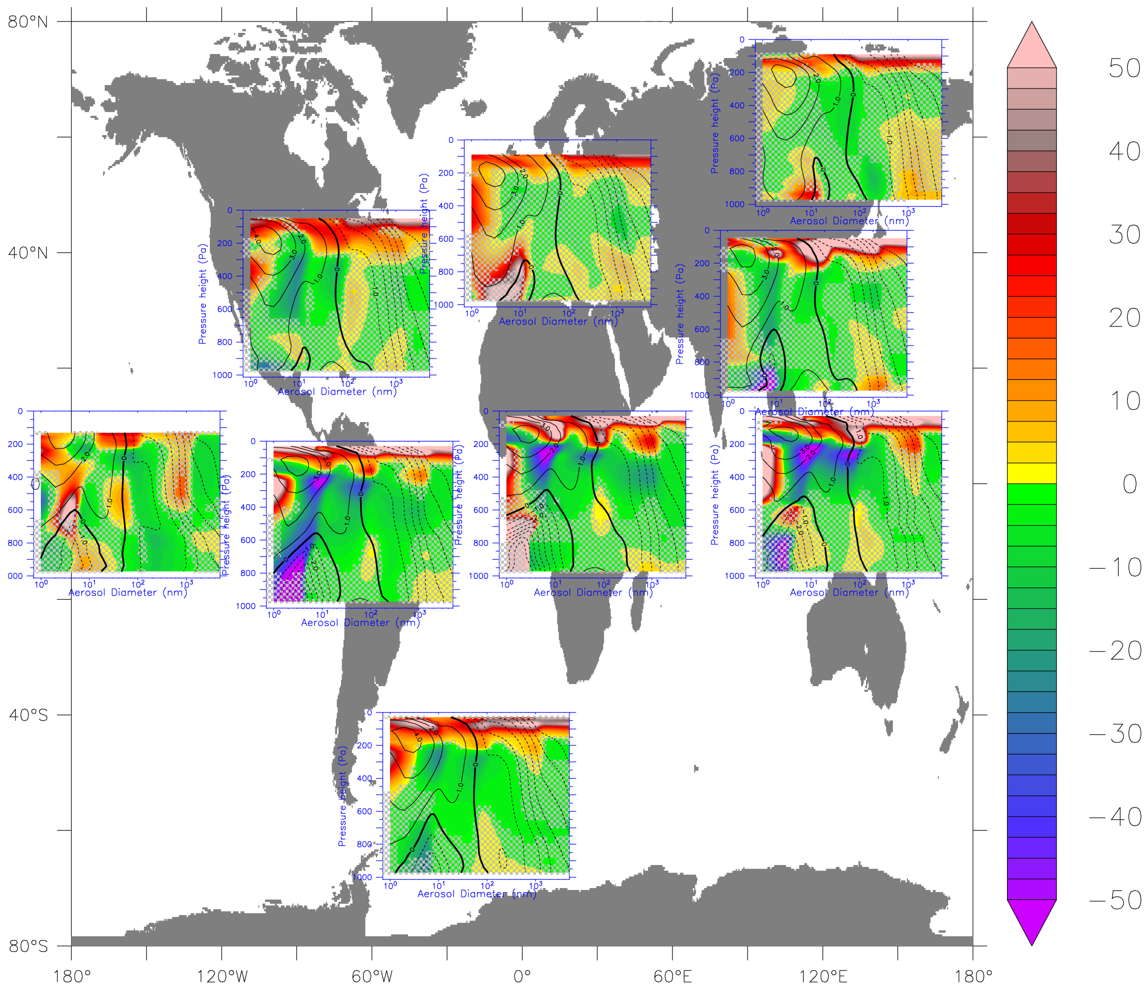

Figure S10: Map of vertically resolved percentage differences (including LNO_x emissions as the reference case) in the aerosol size distributions as spatial and regional average (for the respective regions). Overlayed are the contours of the absolute values of the size distributions as calculated from the spatial and temporal mean in particles/cm^3. The figure depicts the present day conditions for the ARG configuration.

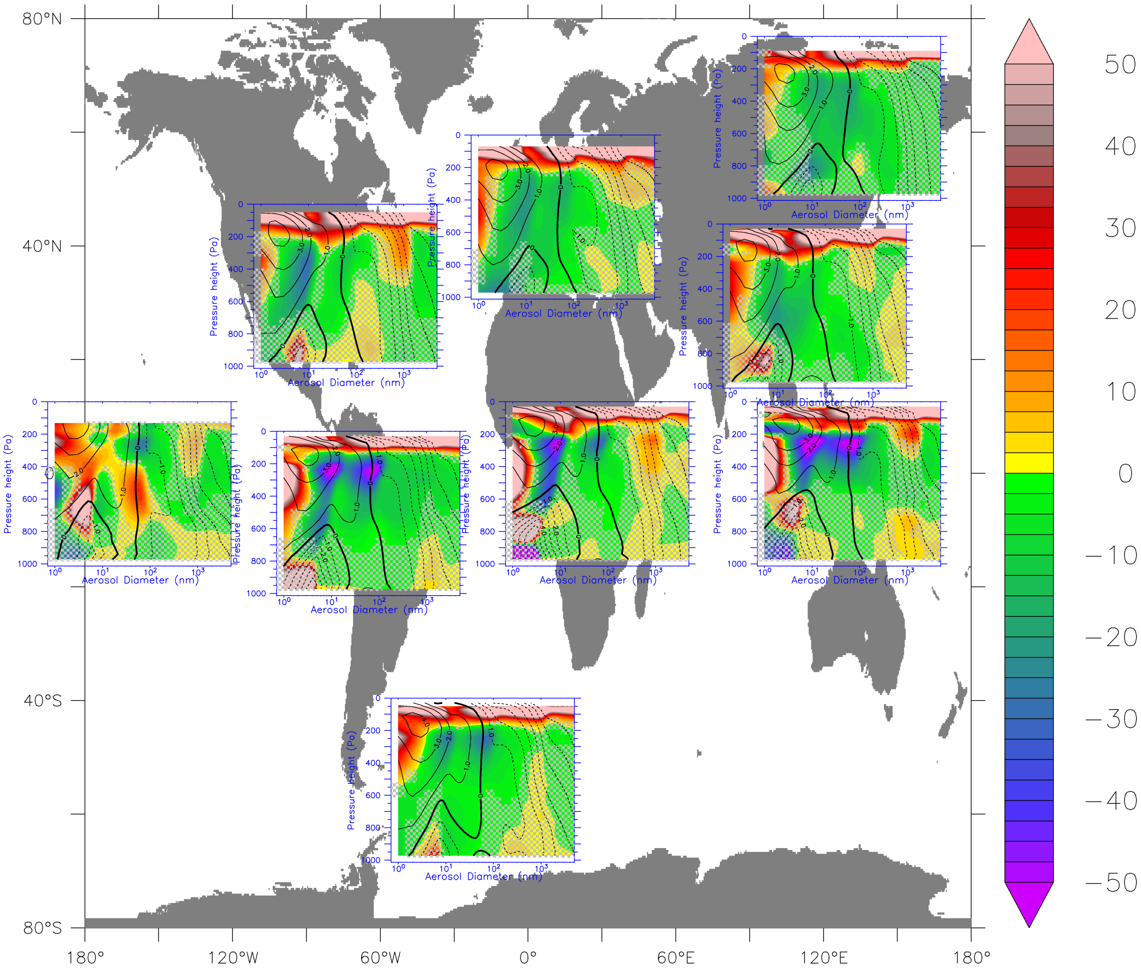

Figure S11: As Fig. 10, but for preindustrial conditions with the ARG model configuration.

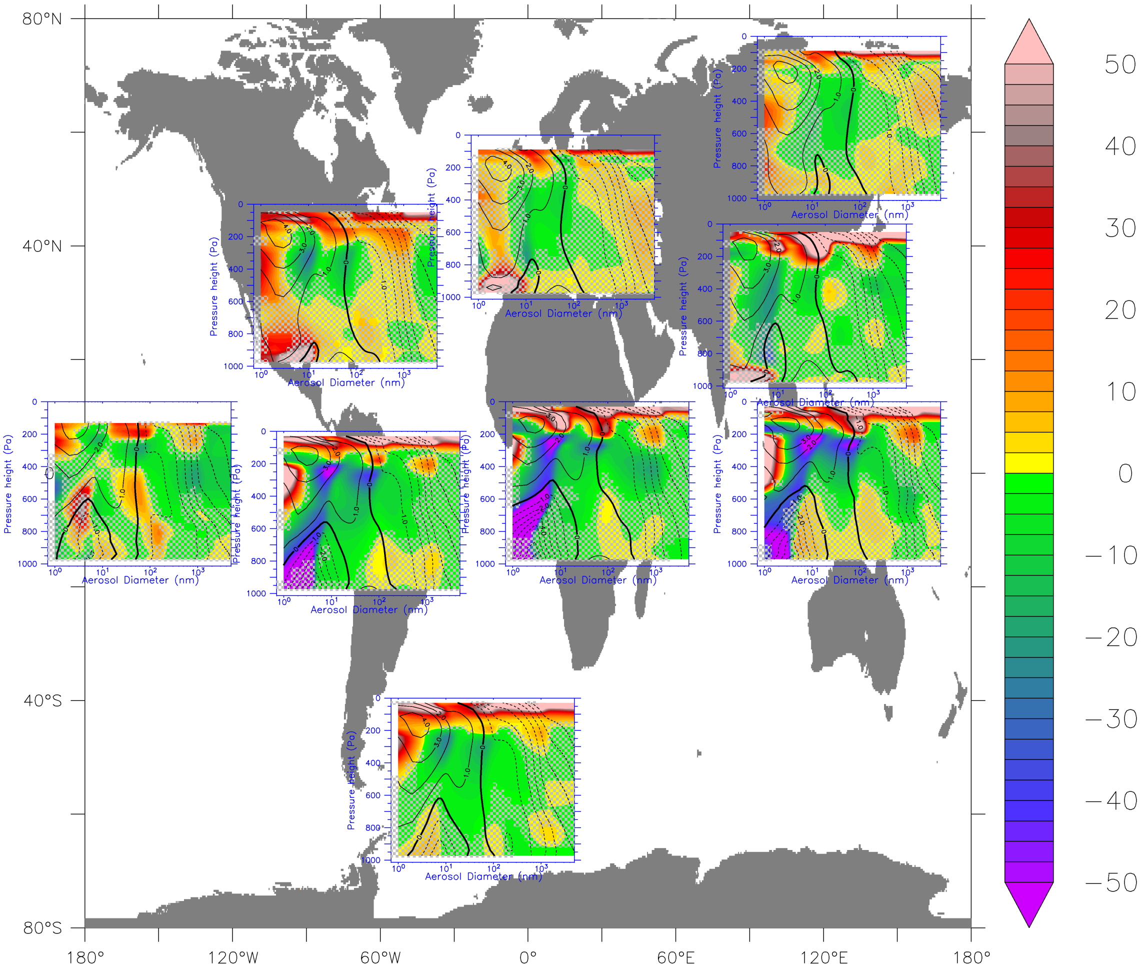

Figure S12: As Fig. 10, but for present day conditions with the KK model configuration.

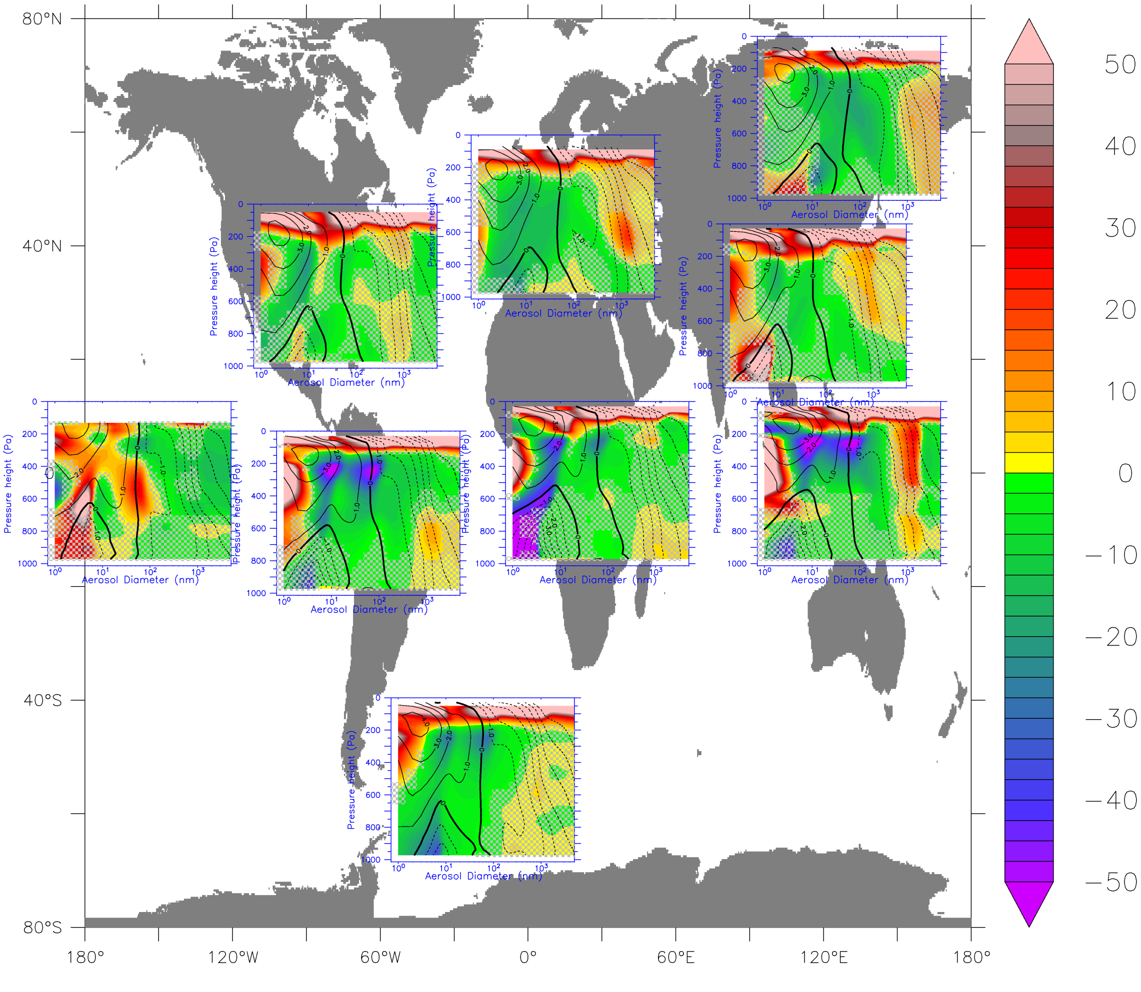

Figure S13: As Fig. 10, but for preindustrial conditions with the KK model configuration.

3.1 Growth Factor

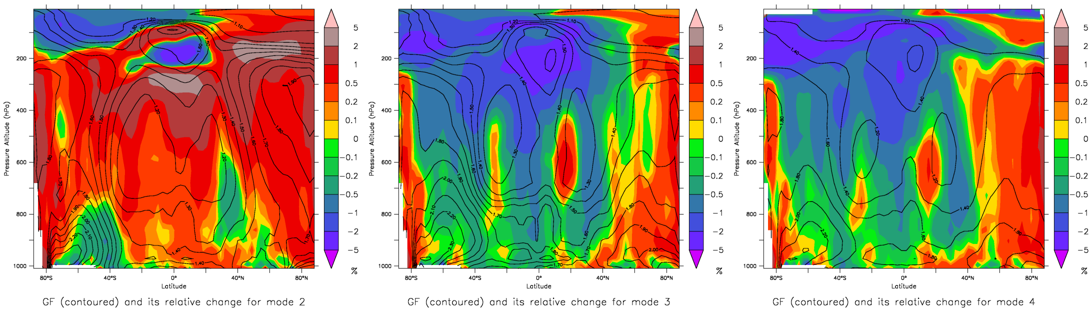

Figure S14: Zonal mean of the change in growth factor (colours) due ot LNO_x emissions and absolute value of the growth factor (wet diameter / dry diameter) for the aitken (left), accumuluation (center) and coarse (right) mode. The figure depicts the ARG configuration for present day conditions.

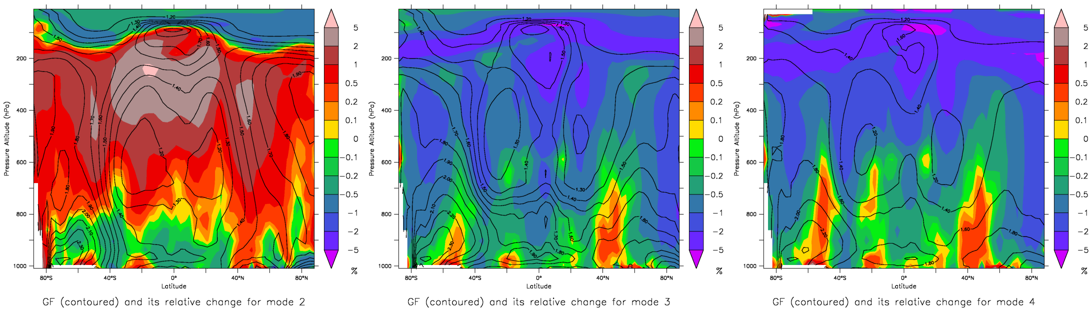

Figure S15: As Fig. 14, but for preindustrial conditions.

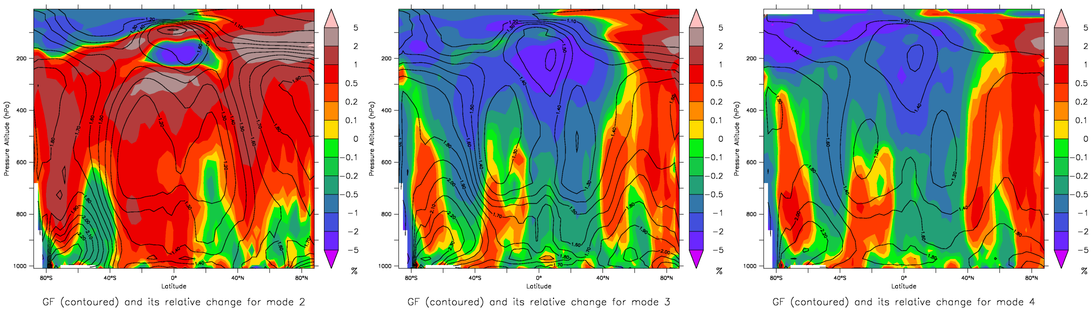

Figure S16: As Fig. 14, but with the KK configuration.

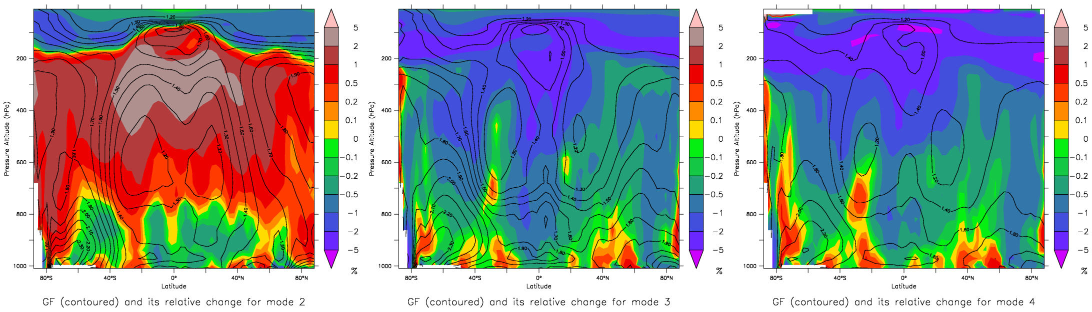

Figure S17: As Fig. 15, but with the KK configuration.

4 Aerosol optical properties

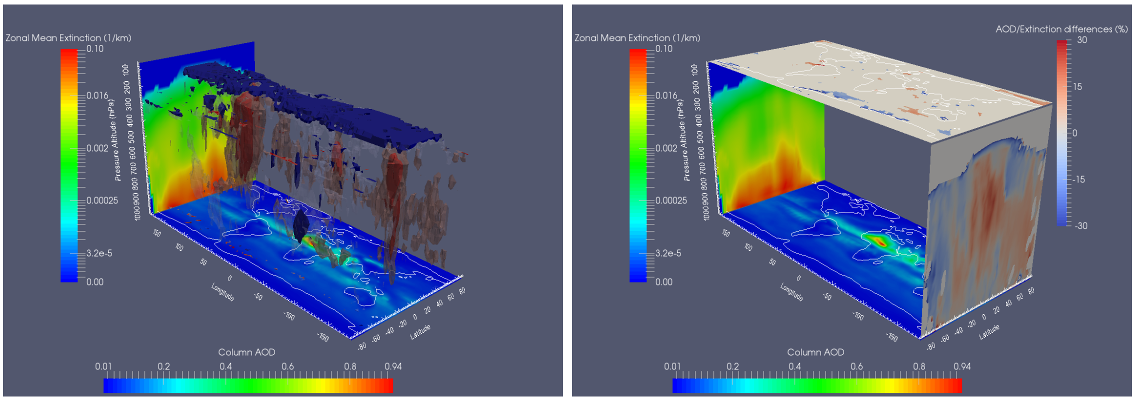

Figure 18: 3d Visualisation of aerosol extinction and the influence of LNO_x emissions. The floor shows a map of the vertically integrated column AOD (at 550 nm) when lightning emissions are included. The ceiling depicts the relative differences of the integrated column AOD between the simulation with lightning emissions minus the simulation without lightning LNO_x for the ARG configuration, but preindustrial conditions. The back panel displays the zonal average aerosol extinction (in 1/km at 550 nm) of the full simulations (Please, note the logarithmic scale.). Additionally, the front panel depicts again relative percentage differences due NO_x emissions from lightning. White areas mark regions without statistical significance.
The right panel also shows AOD (floor) and zonal mean extinction (rear panel), and additionally 3D isosurfaces in the center of the box represent the +10% (pale red) and +20% (dark red) of the enhanced extinction due to active LNO_x emissions, whereas the −10% (pale blue) and −20% (dark blue) isosurfaces mark regions, in which the emissions result in a reduction of the extinction.
.

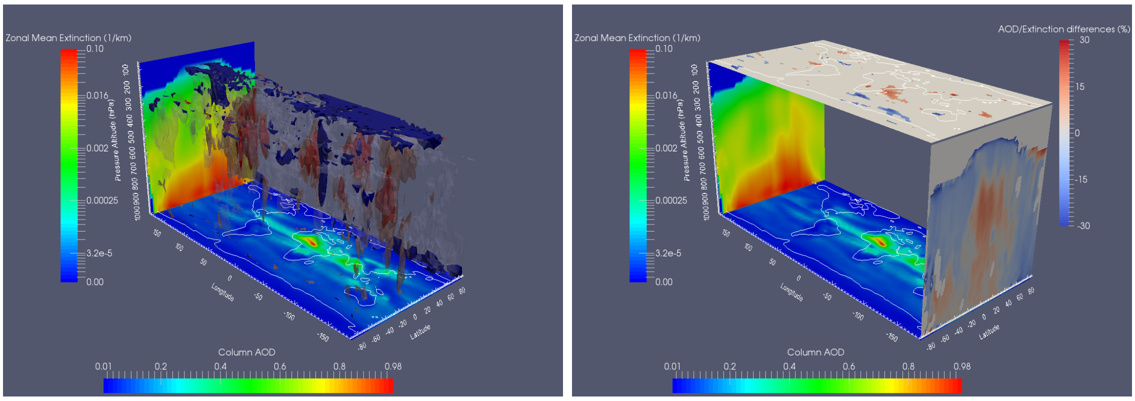

Figure S19: As Fig. 18, but for the KK configuration and present day conditions..

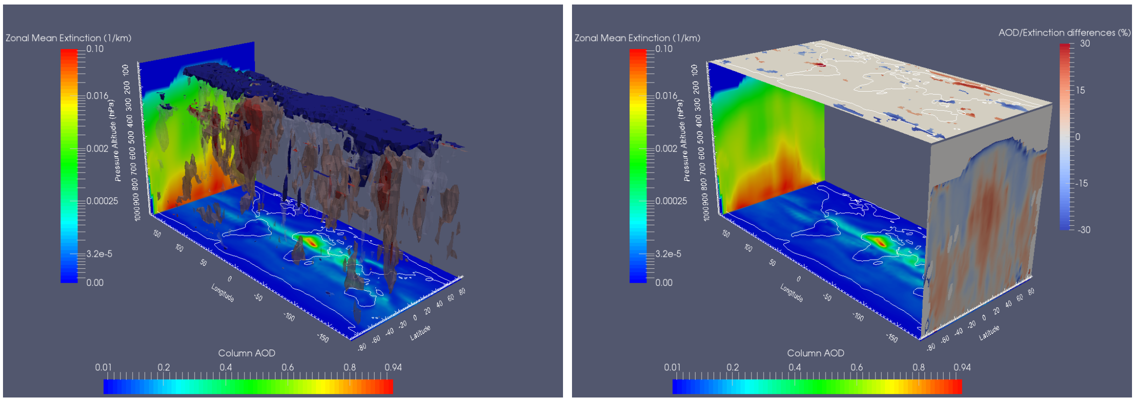

Figure S20: As Fig. 18, but for the KK configuration and preindustrial conditions..

4.1 PDFs of changes in extinction

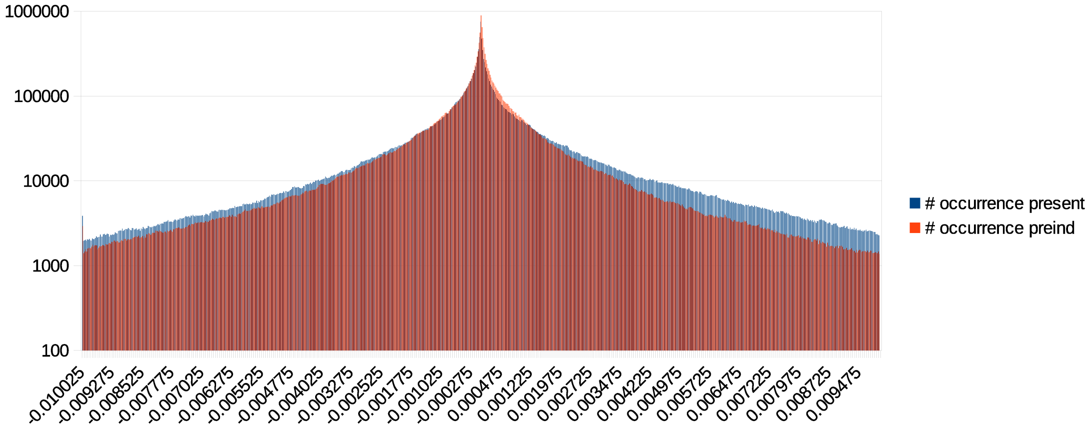

Figure S21: PDF of changes in extinction for present day (blue) and preindustrial conditions (red). Note the logarithmic scaling on the y-axis.

.

5 Aerosol cloud interactions

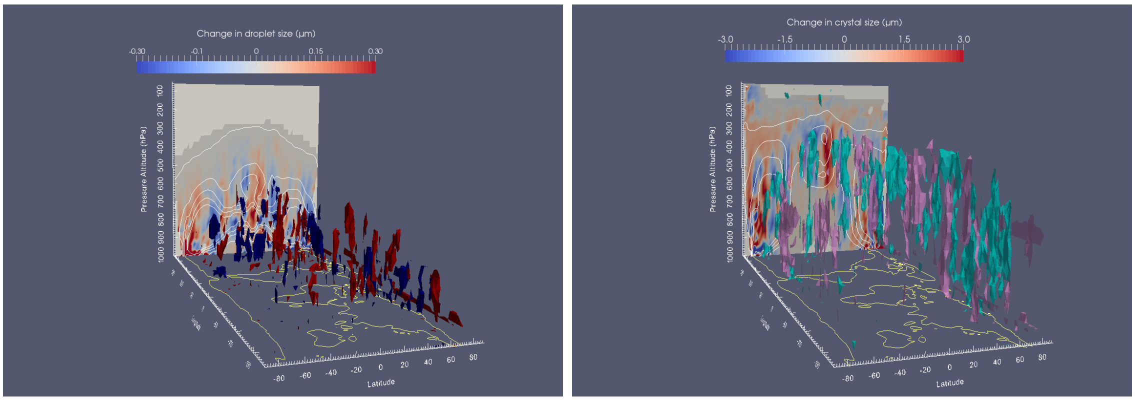

Figure S22: Visualisation of changes in the effective radius of liquid water droplets (left panel) and ice crystals (right panel). The back panel displays the zonal average liquid droplet effect radius (white contours) and ice crystal effective radius (white contours), respectively. The colour coding shows the absolute changes due to the LNO_x emissions. White areas mark regions without statistical significance.

Additionally, the isosurfaces represent the regions for statistical significant absolute changes for the effective radius for water droplets (blue negative, red positive) and effective ice crystal size (turquoise for negative and purple for positive) due the LNO_x emissions. The mean values for the present day conditions in the ARG configuration are shown here.

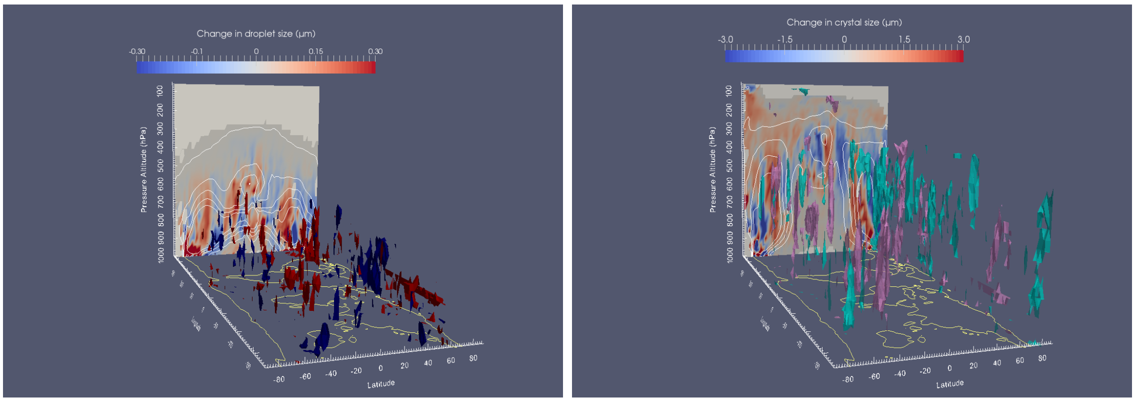

Figure S23: As Fig. 22, but for preindustrial conditions in the ARG configuration.

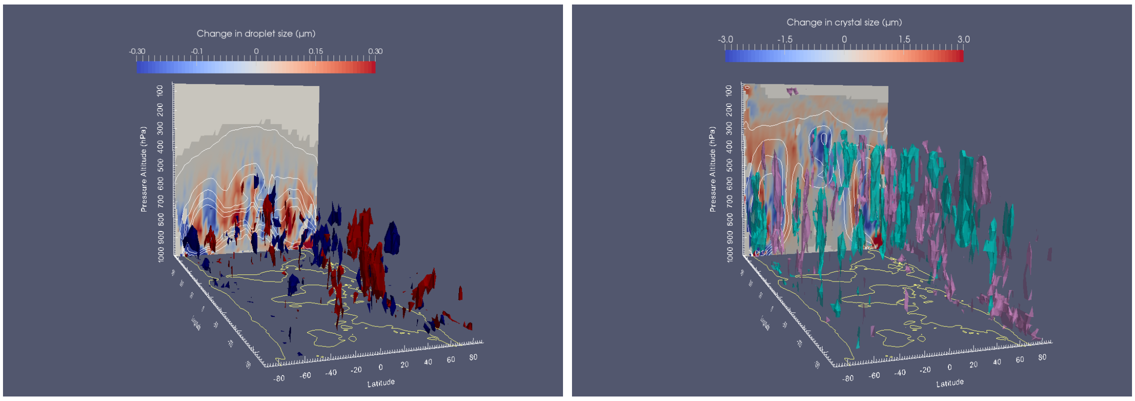

Figure S24: As Fig. 22, but for present day conditions in the KK configuration.

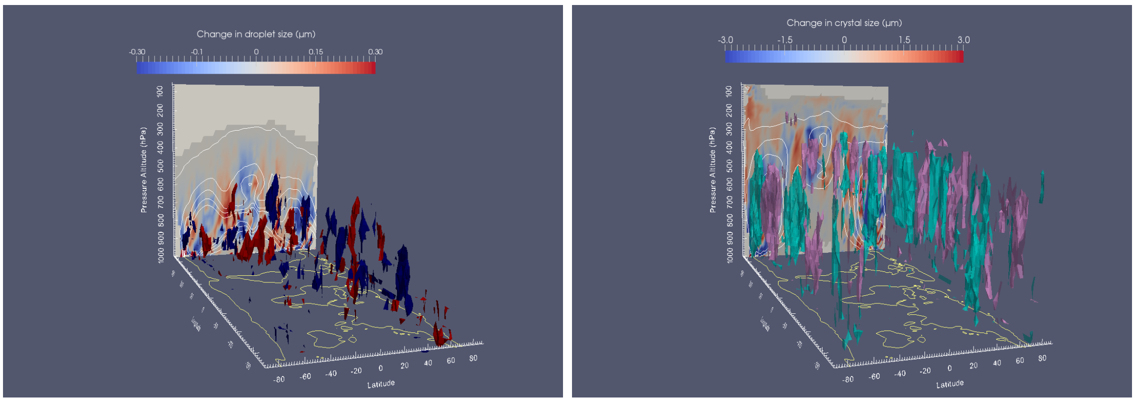

Figure S25: As Fig. 22, but for preindustrial conditions in the KK configuration.

6 Radiative fluxes

6.1 Anthropogenic aerosol effect

6.1.1 ARG configuration

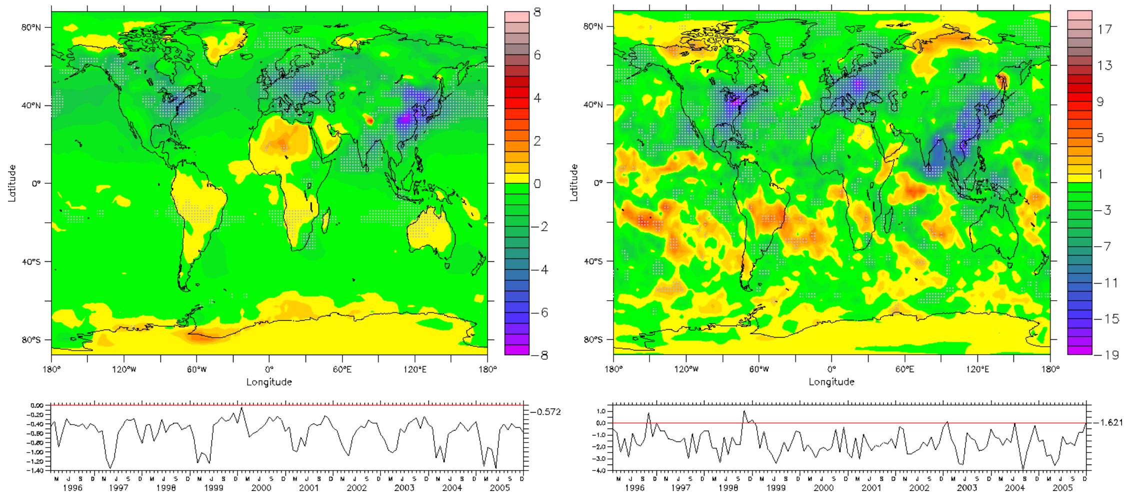

Figure S26: Decadal mean of the anthropogenic aerosol effect in shortwave radiation at the top of the atmosphere in the ARG configuration. The left panel depicts clear sky conditions, the right panel all sky conditions. Hatched regions represent areas with a statistical significance (90% confidence level of a two sided t-test, based on the annual mean
uxes).

6.1.2 KK configuration

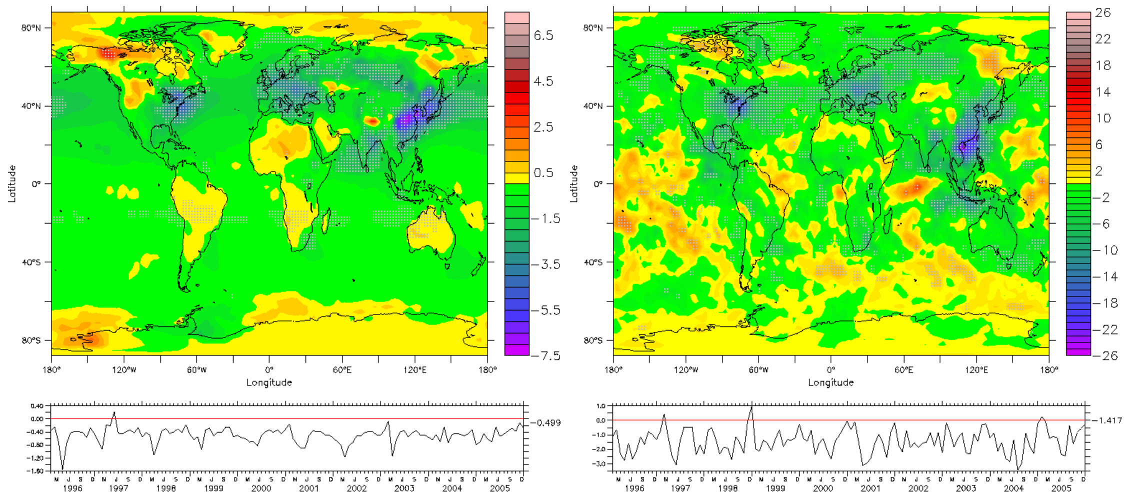

Figure S27: As Fig. 26, but for the KK configuration.

6.2 Lightning NO_x based radiative flux disturbances

6.2.1 ARG - preindustrial

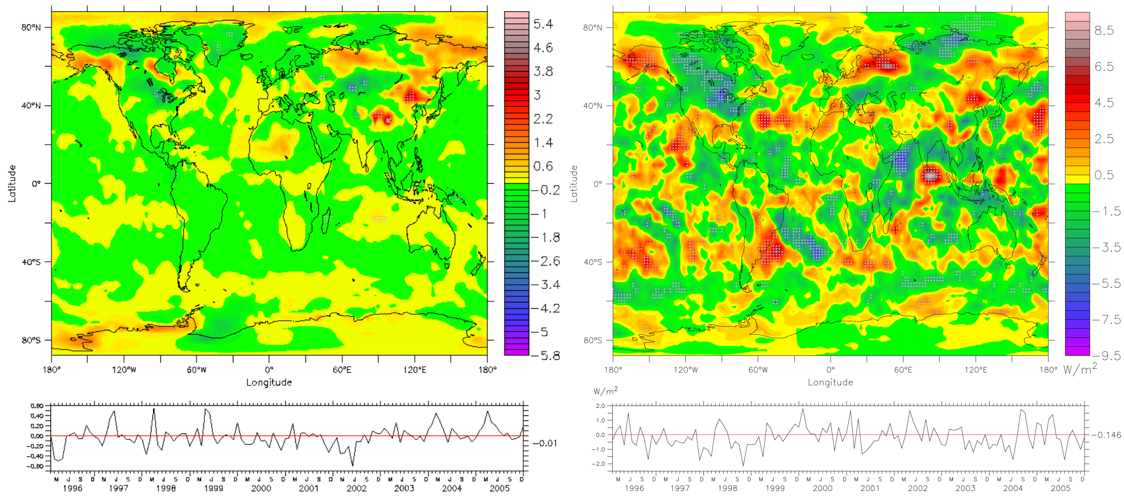

Figure 28: Decadal mean of the shortwave radiative flux disturbance in the ARG configuration at the top of the atmosphere for preindustrial conditions. The left panel depicts clear sky conditions, the right panel all sky conditions. Hatched regions represent areas with a statistical significance (90% confidence level of a two sided t-test, based on the annual mean fluxes).

6.2.2 KK - present day

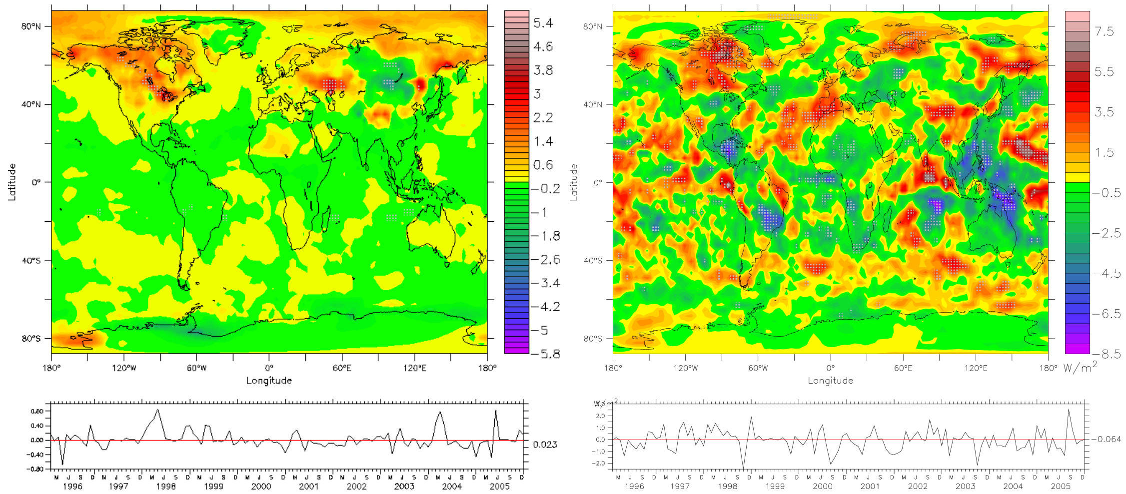

Figure S29: As Fig. 28, but for the KK configuration and present day conditions.

6.2.3 KK - preindustrial

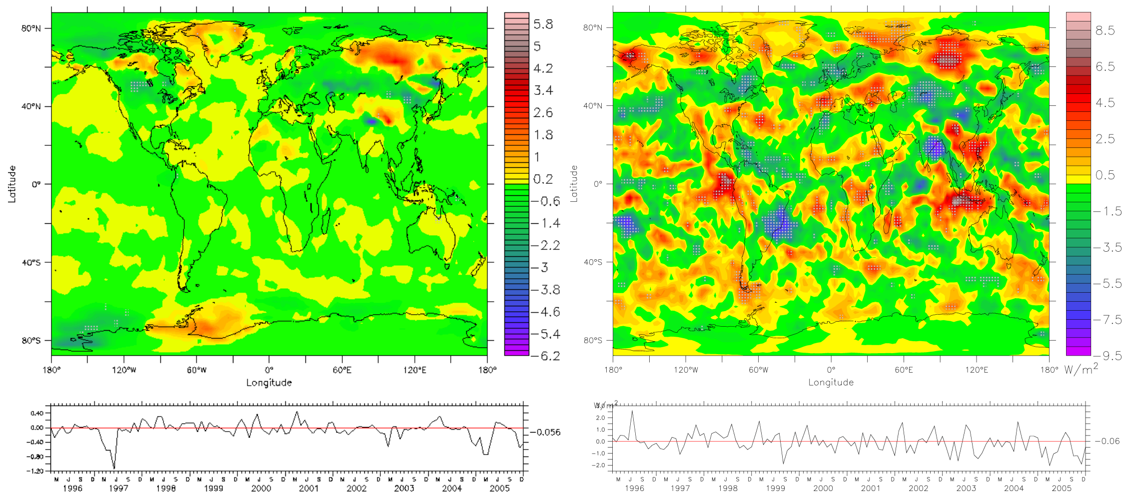

Figure S30: As Fig. 28, but for the KK configuration and preindustrial conditions.