

Supplementary material to:  
Chemistry-climate interactions of aerosol nitrate from  
lightning

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# 1 LNO<sub>x</sub> emissions

## 1.1 ARG - preindustrial

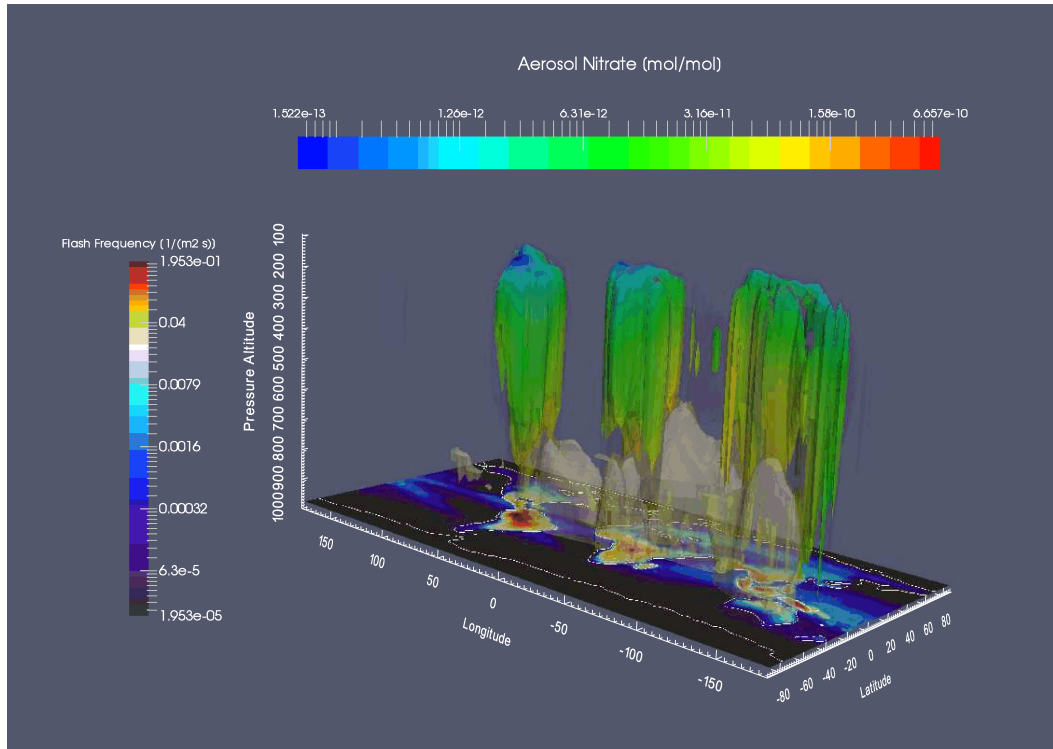


Figure 1: 3D visualisation of LNO<sub>x</sub> emissions (coloured isosurface of  $1 \cdot 10^{-16}$  kg/(m<sup>3</sup>s) and darker shaded isosurface of  $3 \cdot 10^{-16}$  kg/(m<sup>3</sup>s)) and the total aerosol nitrate mixing ratios (grey isosurface of 0.1 ppb<sub>v</sub>). Additionally, the mean flash rate in 1/s 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

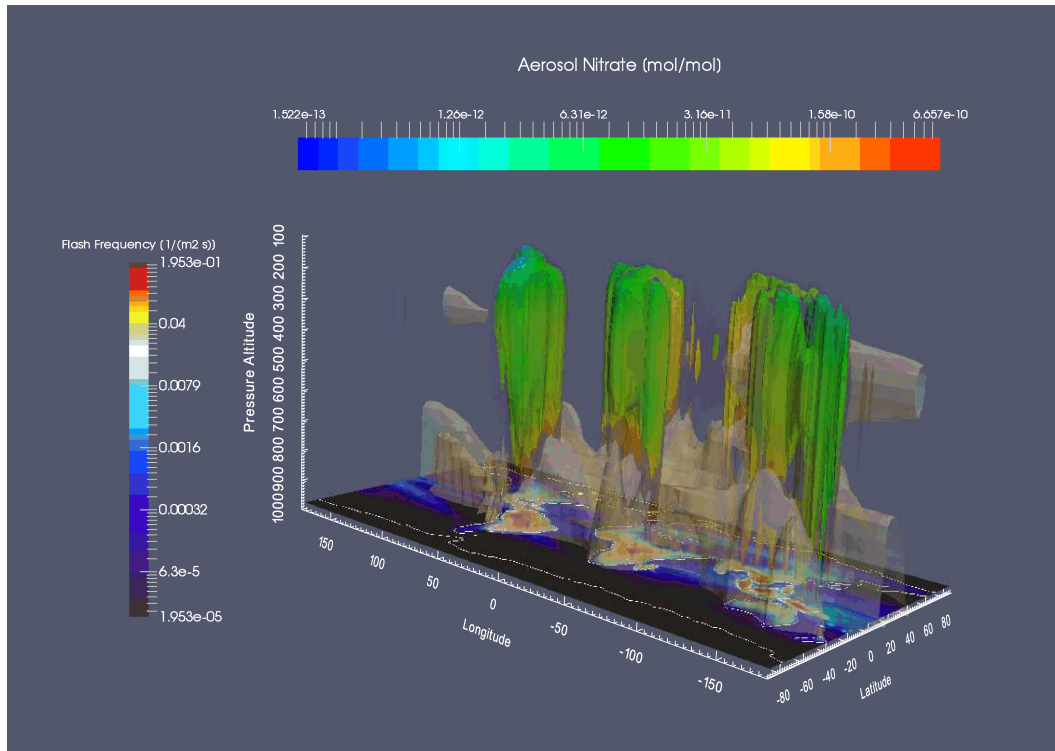


Figure 2: As Fig. 1, but for present day conditions with the KK model configuration.

### 1.3 KK - preindustrial

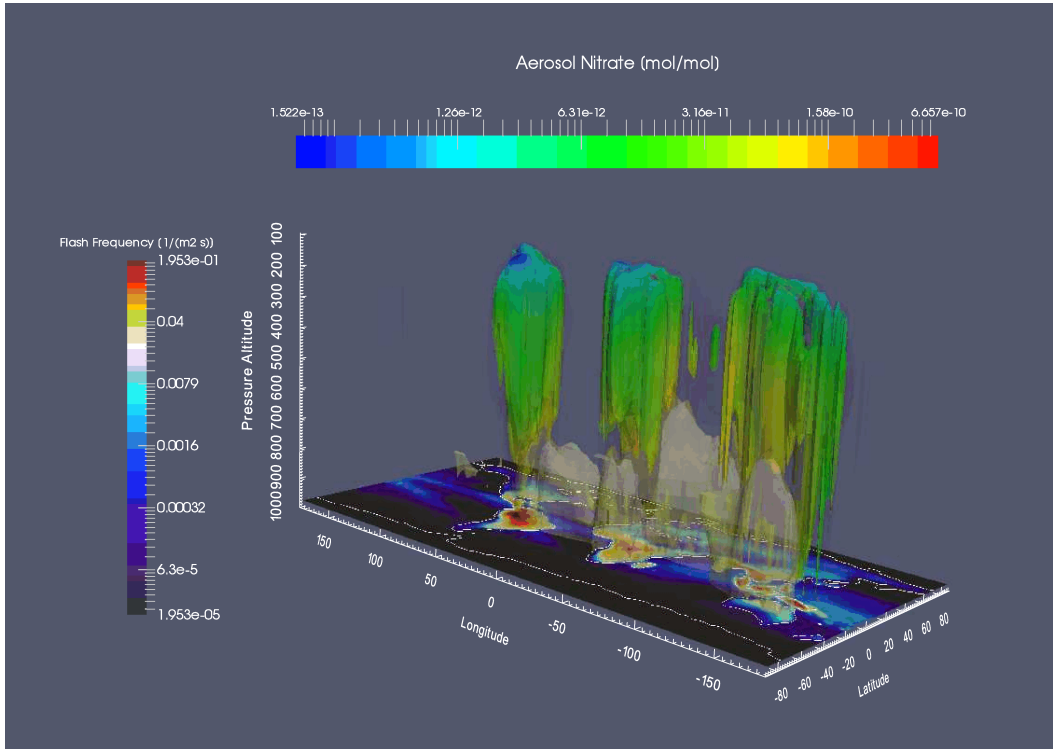


Figure 3: As Fig. 1, but for preindustrial conditions with the KK model configuration.

## 2 Nitrate distributions

### 2.1 Budget tables

Table 1: Tropospheric budget in the kappa-koehler simulation of the important highly oxidized nitrogen species, i.e. gaseous  $\text{HNO}_3$ , gaseous  $\text{N}_2\text{O}_5$ , aerosol  $\text{NO}_3^-$  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).

	$\text{HNO}_3$	$\text{N}_2\text{O}_5$	$\text{NO}_3^-$	Total N(V+)
Present day				
Absolute values (with $\text{LNO}_x$ emissions):				
Tropospheric Column burden	407	8.1	143	557
UT Column burden	177	5.1	27.5	209
Absolute differences due to $\text{LNO}_x$ 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 $\text{LNO}_x$ emissions):				
Tropospheric Column burden	237	4.5	67.8	310
UT Column burden	142	4.1	14.9	161
Absolute differences due to $\text{LNO}_x$ emissions:				
Tropospheric Column burden	157	3.8	27.8	189
UT Column burden	109	3.5	8.3	121
Relative differences in (%) due to $\text{LNO}_x$ emissions:				
Tropospheric Column burden	66.2	82.6	41.0	61.0
UT Column burden	76.7	86.0	55.4	74.9

Table 2: Loss processes for the dominant N(V+) compounds for the present day and preindustrial ARG simulations including and excluding the effect of LNO<sub>x</sub> 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 <sub>3</sub> )	Dry deposition (aerosol NO <sub>3</sub> <sup>-</sup> )	Wet deposition	Sedi- mentation
Present day				
Absolute values (with LNO <sub>x</sub> ):	6.03	1.13	25.48	8.24
Absolute change due to LNO <sub>x</sub> :	0.13	0.08	4.44	0.70
Relative change due to LNO <sub>x</sub> :	2.09	7.37	17.43	8.54
Preindustrial conditions				
Absolute values (with LNO <sub>x</sub> ):	1.227	0.27	8.90	2.45
Absolute change due to LNO <sub>x</sub> :	0.20	0.08	4.46	0.78
Relative change due to LNO <sub>x</sub> :	16.54	29.78	50.07	31.63

Table 3: Loss processes for the dominant N(V+) compounds for the present day and preindustrial kappa-coehler simulations including and excluding the effect of LNO<sub>x</sub> 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 <sub>3</sub> )	Dry deposition (aerosol NO <sub>3</sub> <sup>-</sup> )	Wet deposition	Sedi- mentation
Present day				
Absolute values (with LNO <sub>x</sub> ):	6.23	1.22	25.48	8.27
Absolute change due to LNO <sub>x</sub> :	0.27	0.17	4.48	0.71
Relative change due to LNO <sub>x</sub> :	4.35	14.08	17.58	8.64
Preindustrial conditions				
Absolute values (with LNO <sub>x</sub> ):	1.23	0.27	8.90	2.46
Absolute change due to LNO <sub>x</sub> :	0.20	0.08	4.47	0.79
Relative change due to LNO <sub>x</sub> :	16.22	29.89	50.15	32.10

## 2.2 Geographical distributions

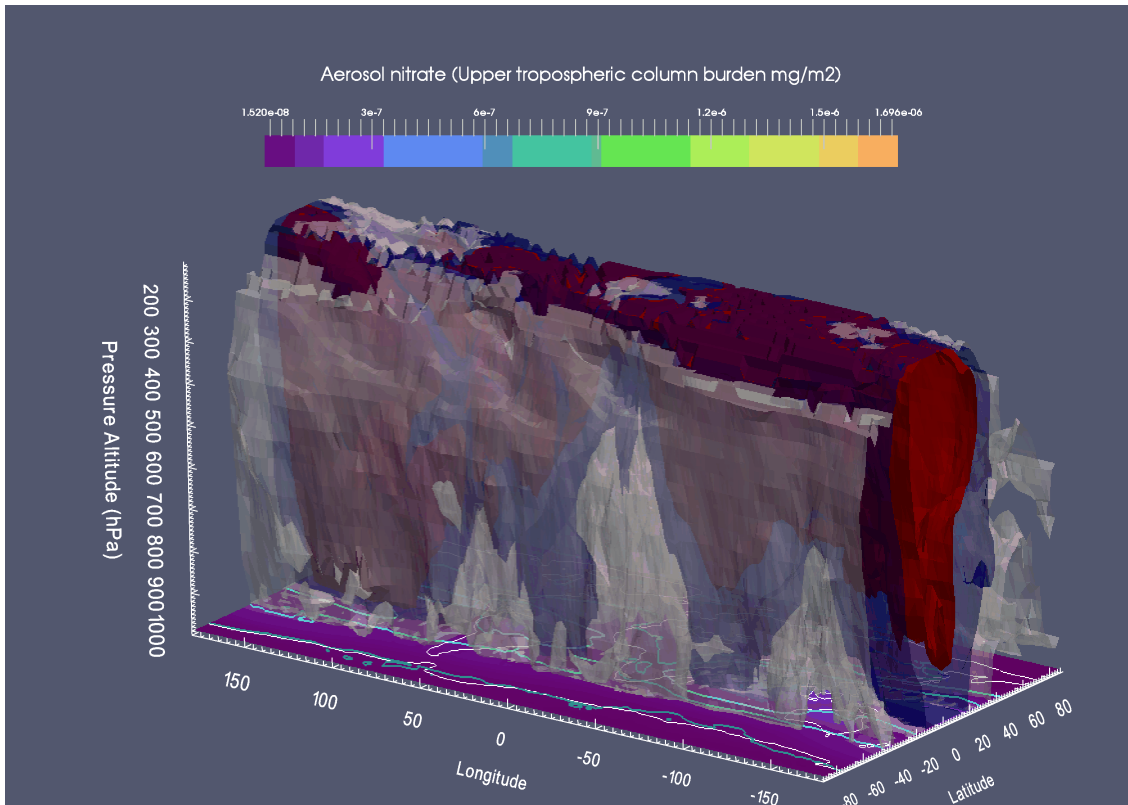


Figure 4: 3D visualisation of the relative differences in tropospheric aerosol nitrate mixing ratios between the simulations with and without LNO<sub>x</sub> emissions to the simulation including LNO<sub>x</sub> emissions. The white isosurface depicts a relative difference of 30%, the blue isosurface of 45%, and the red isosurface of 60%. Additionally, the upper tropospheric aerosol nitrate column burden (in mg/m<sup>2</sup>) 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.

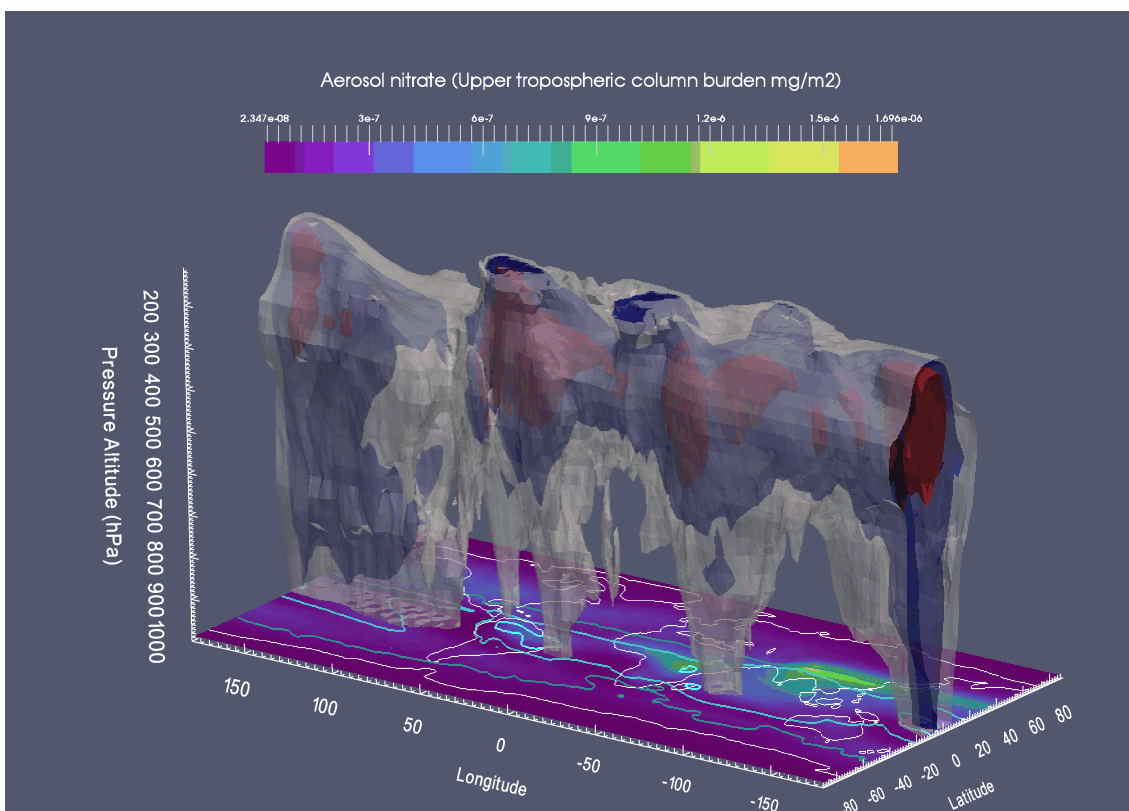


Figure 5: As Fig. 4, but with the KK configuration for present day conditions.



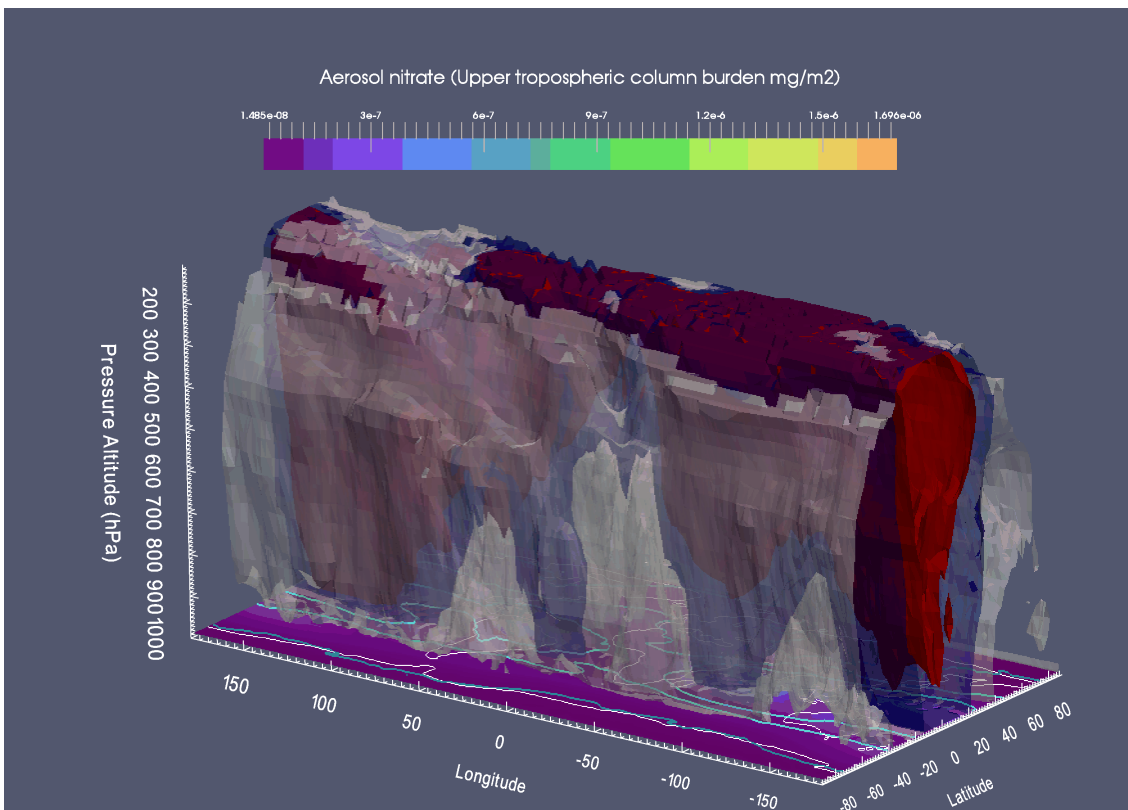


Figure 6: As Fig. 4, but with the KK configuration for preindustrial conditions.

### 3 Size distributions

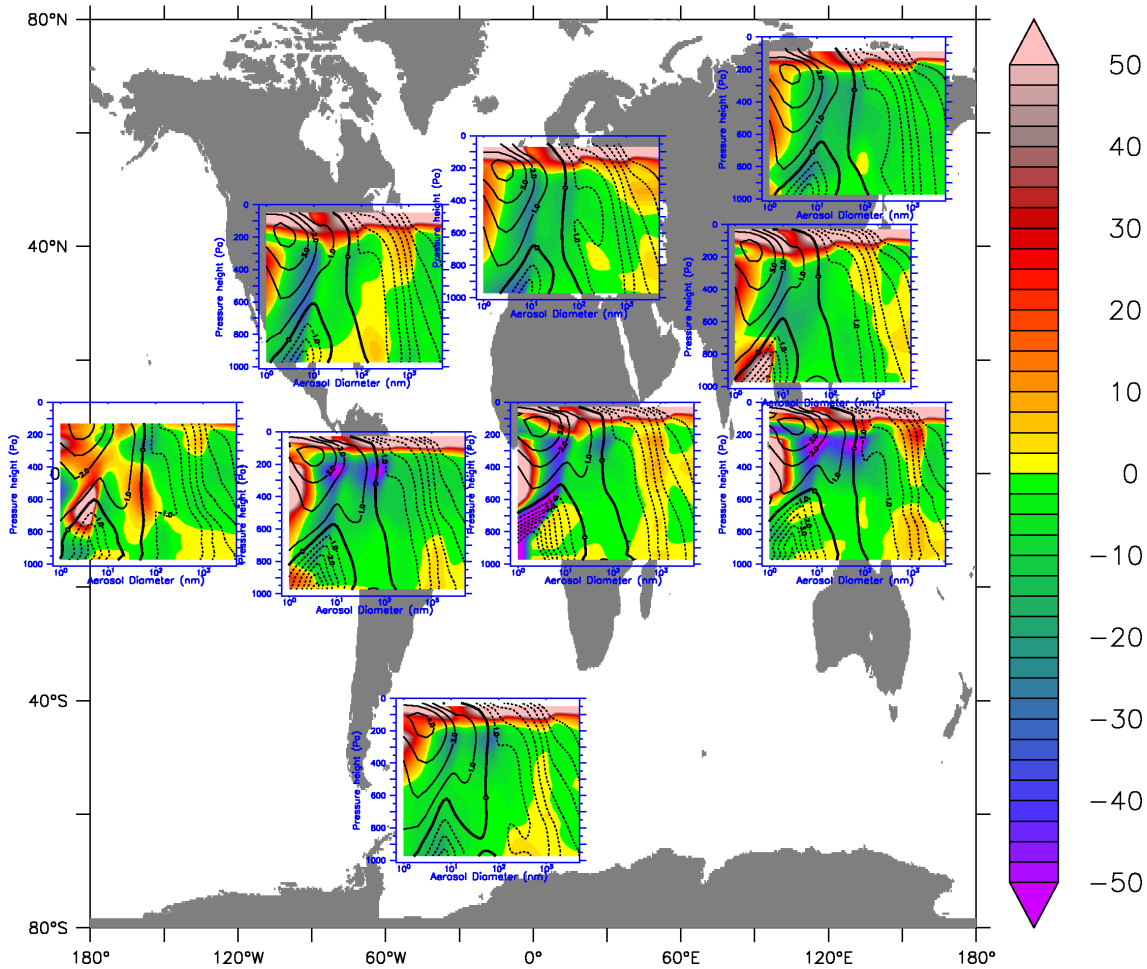


Figure 7: Map of vertically resolved percentage differences (including LNO<sub>x</sub> emissions as the reference case) in the aerosol size distributions as spatial and regional average (for the respective regions). Overlaid are the contours of the absolute values of the size distributions as calculated from the spatial and temporal mean in particles/cm<sup>3</sup>. The figure depicts the preindustrial conditions for the ARG configuration.

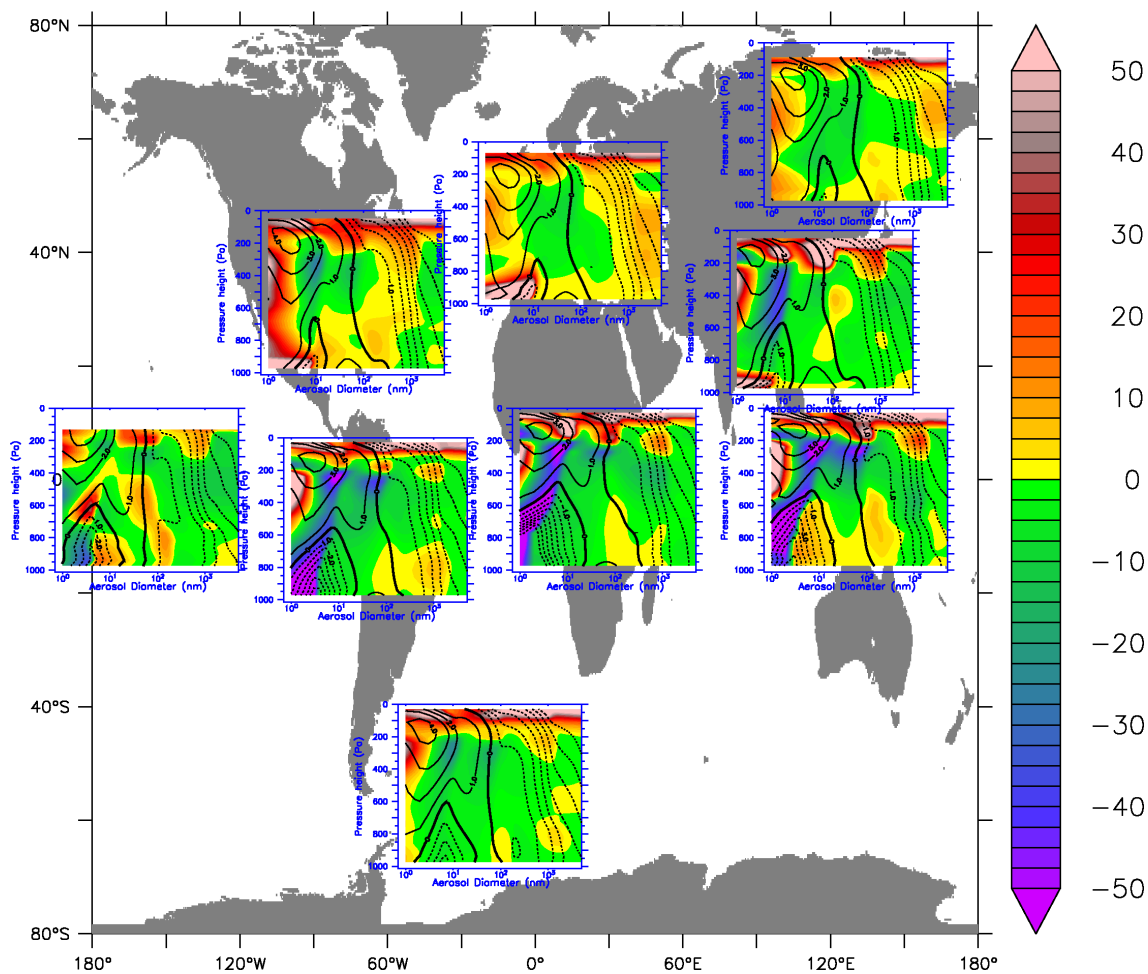


Figure 8: As Fig. 7, but for present day conditions with the KK model configuration.

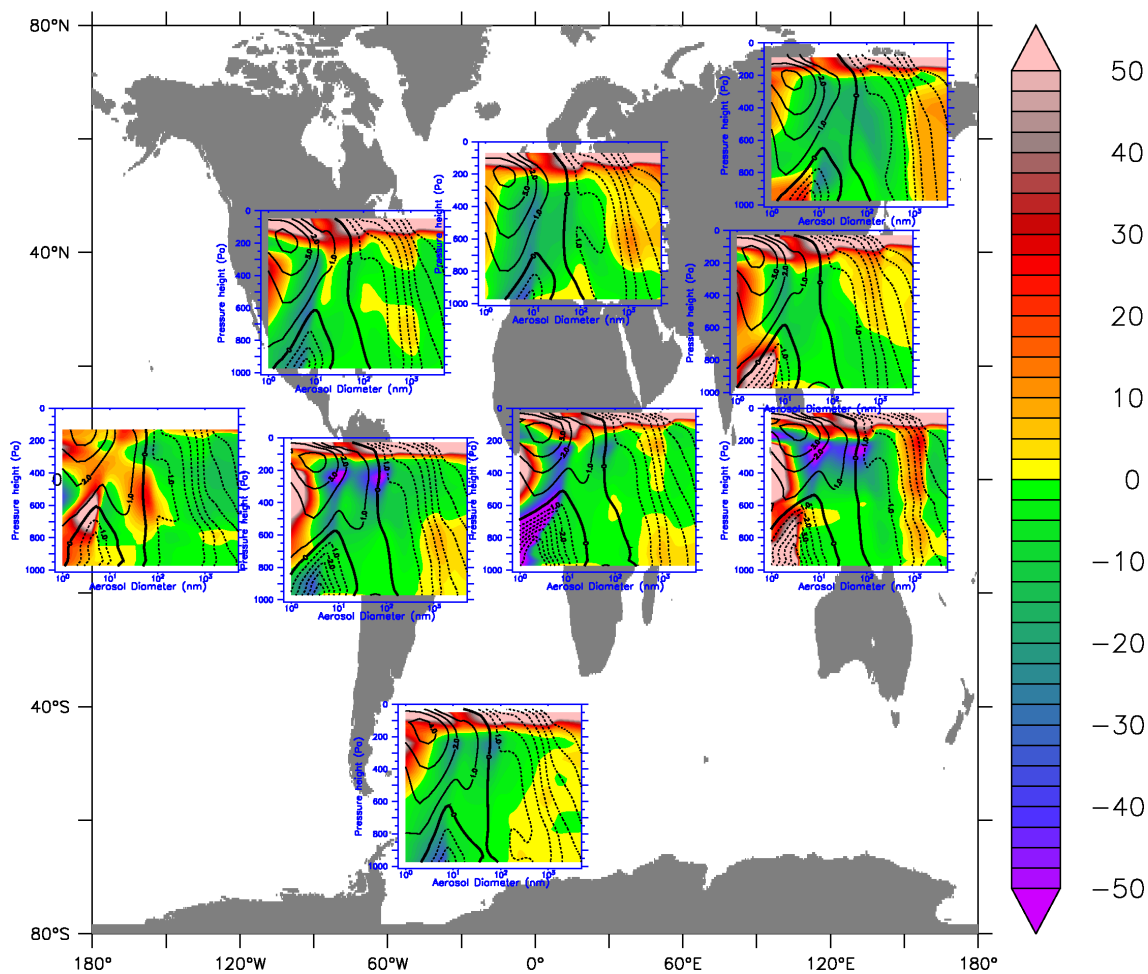


Figure 9: As Fig. 7, but for preindustrial conditions with the KK model configuration.

### 3.1 Growth Factor

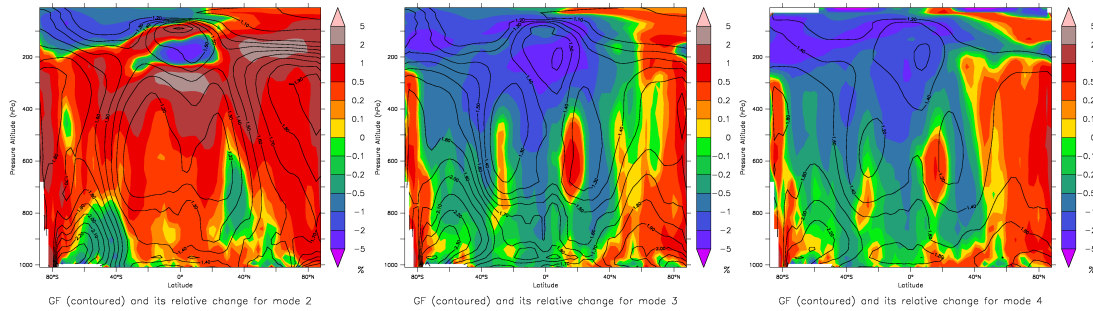


Figure 10: Zonal mean of the change in growth factor (colours) due to  $\text{LNO}_x$  emissions and absolute value of the growth factor (wet diameter / dry diameter) for the aitenken (left), accumulation (center) and coarse (right) mode. The figure depicts the ARG configuration for present day conditions.

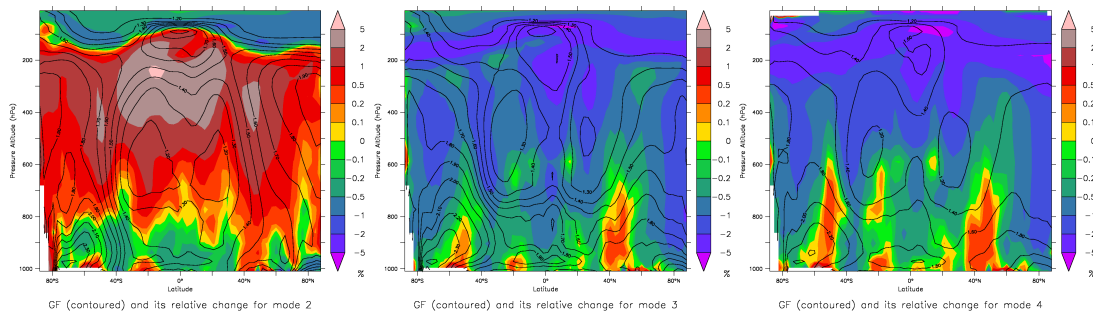


Figure 11: As Fig. 10, but for preindustrial conditions.

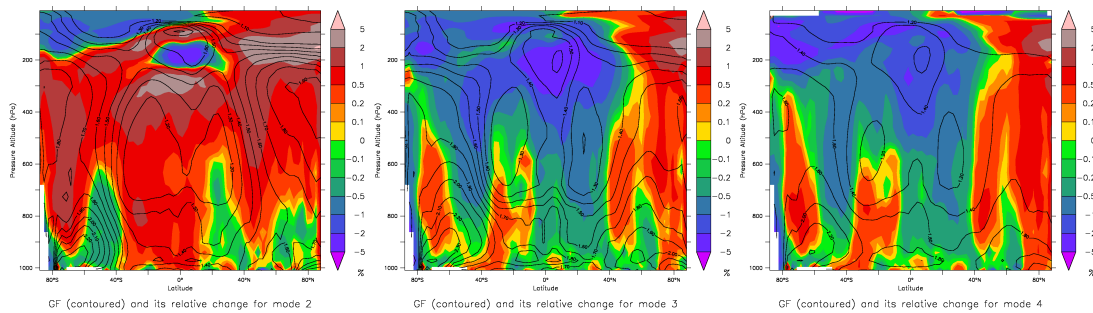


Figure 12: As Fig. 10, but with the KK configuration.

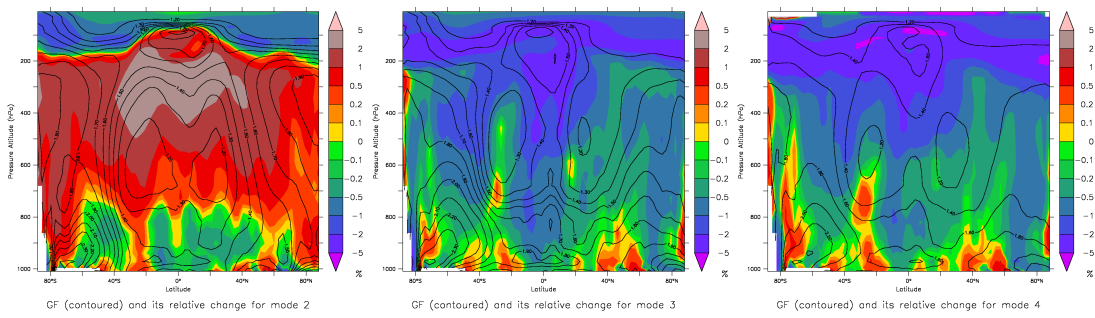


Figure 13: As Fig. 11, but with the KK configuration.

## 4 Aerosol optical properties

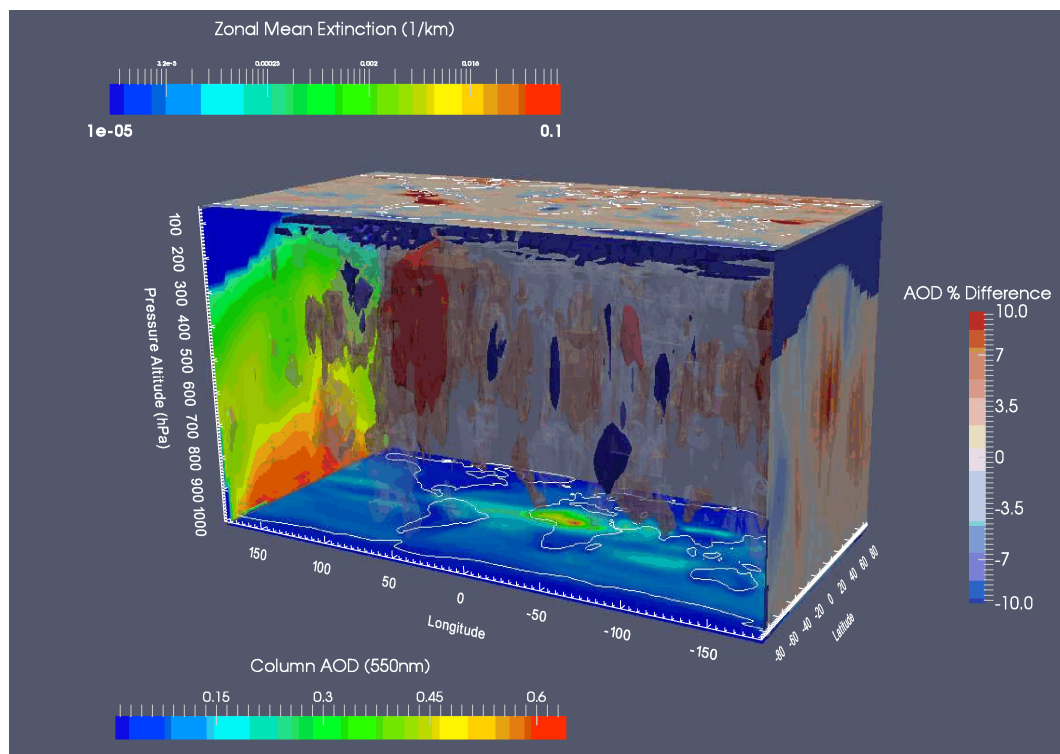


Figure 14: 3d Visualisation of aerosol extinction and the influence of  $\text{LNO}_x$  emissions. The floor shows a map of the vertically integrated column AOD (at 550 nm) when lightning emissions are included. The ceiling depicts the relative differences of the integrated column AOD between the simulation with lightning emissions minus the simulation without lightning  $\text{LNO}_x$  for the ARG configuration, but preindustrial conditions. The back panel displays the zonal average aerosol extinction (in  $1/\text{km}$  at 550 nm) of the full simulations (Please, note the logarithmic scale.). Additionally, the front panel depicts again relative percentage differences due  $\text{NO}_x$  emissions from lightning. The 3D isosurfaces in the center of the box represent the +10% (pale red) and +20% (dark red) of the enhanced extinction due to active  $\text{LNO}_x$  emissions, whereas the -10% (pale blue) and -20% (dark blue) isosurfaces mark regions, in which the emissions result in a reduction of the extinction.

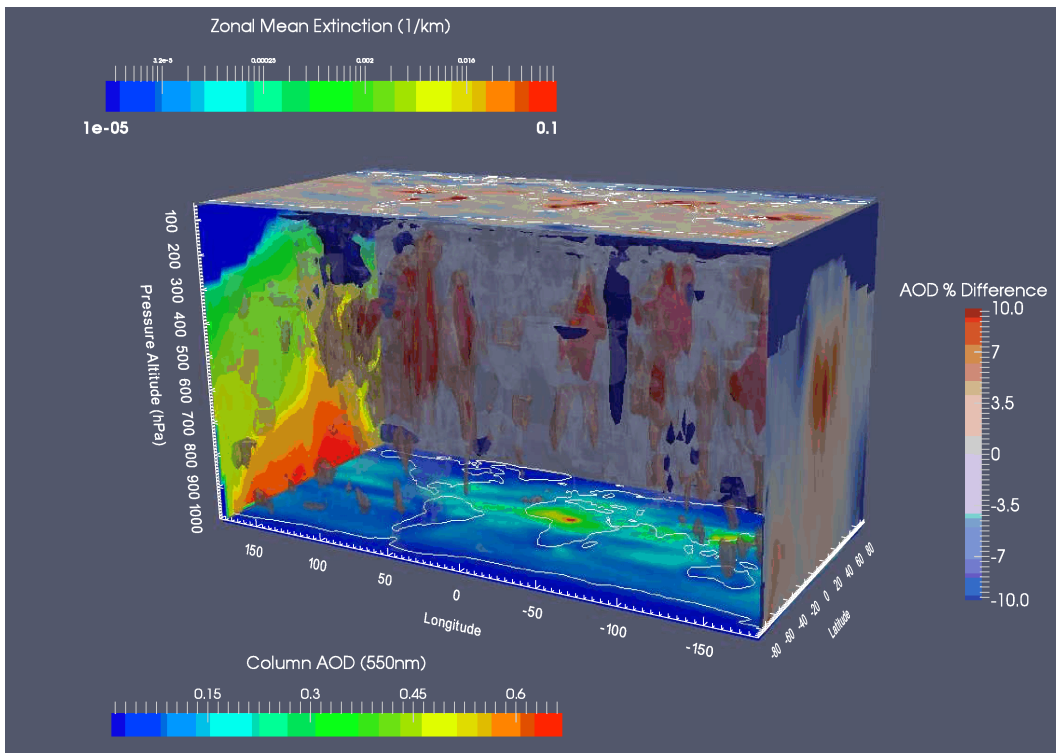


Figure 15: As Fig. 14, but for the KK configuration and present day conditions.

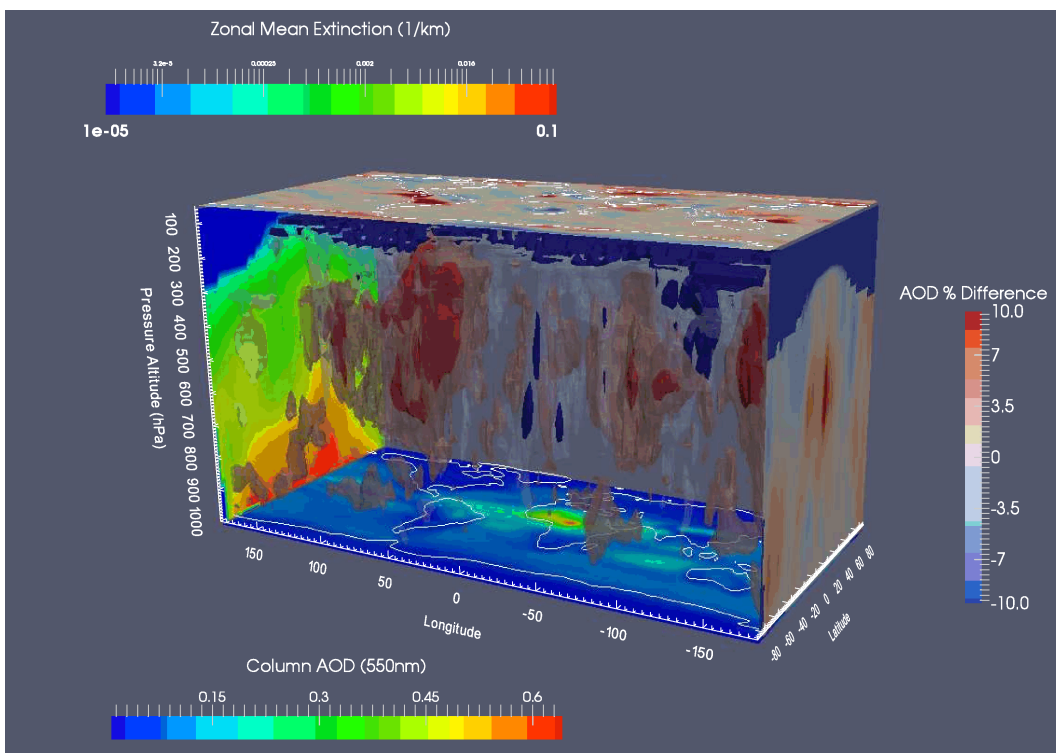


Figure 16: As Fig. 14, but for the KK configuration and preindustrial conditions.



## 5 Aerosol cloud interactions

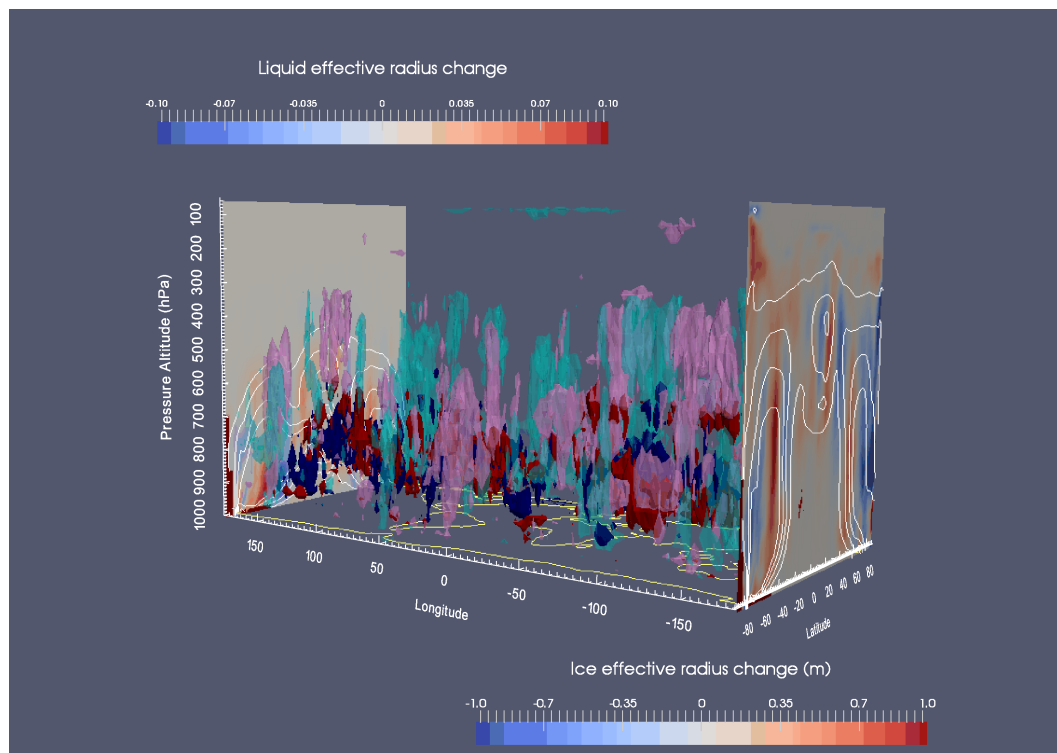


Figure 17: Visualisation of changes in the effective radius of liquid water droplets and ice crystals. The back panel displays the zonal average liquid droplet effect radius (white contours) and the absolute changes due to the LNO<sub>x</sub> emissions. The front panel depicts similarly the ice crystal effective size (white contours) and the absolute change due to the lightning emissions. Additionally, the isosurfaces represent the regions for substantial 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 to the LNO<sub>x</sub> emissions. The mean values for the preindustrial conditions in the ARG configuration are shown here.

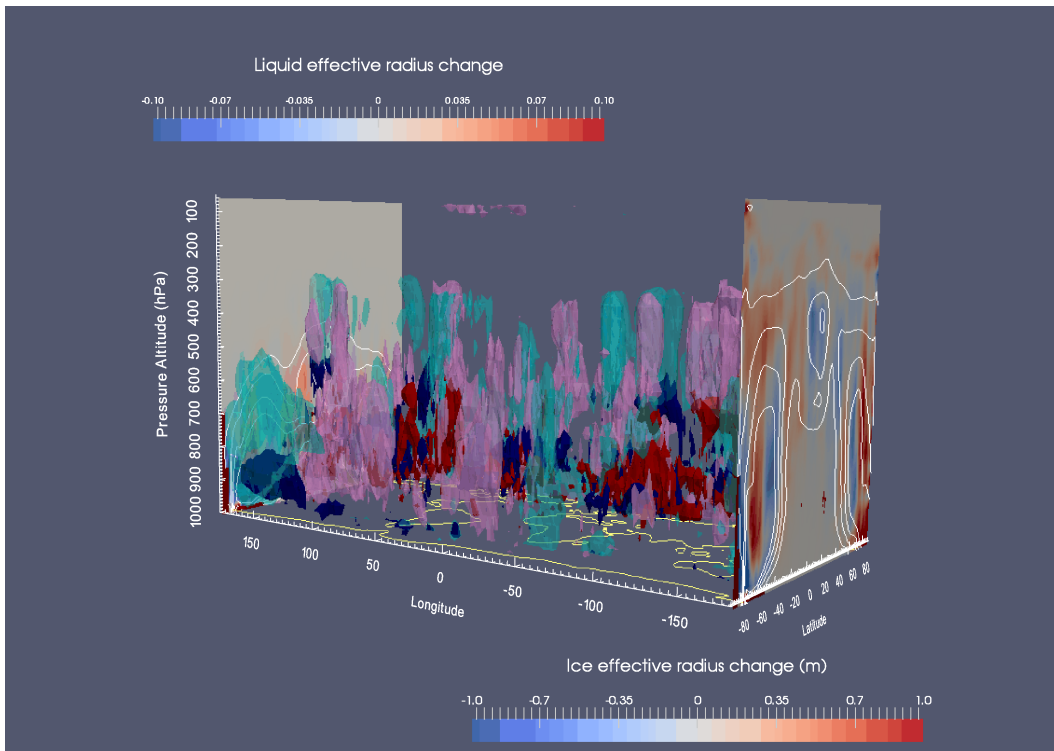


Figure 18: As Fig. 17, but for present day conditions in the KK configuration.

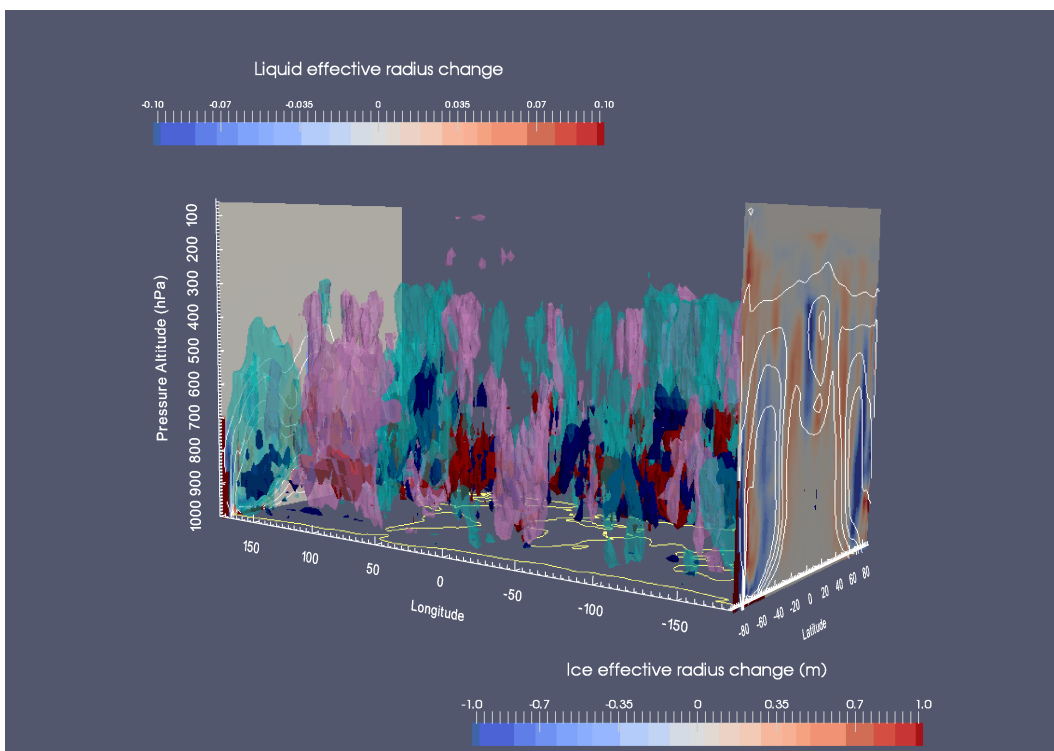


Figure 19: As Fig. 17, but for preindustrial conditions in the KK configuration.

## 6 Radiative fluxes

### 6.1 Anthropogenic aerosol effect

#### 6.1.1 ARG configuration

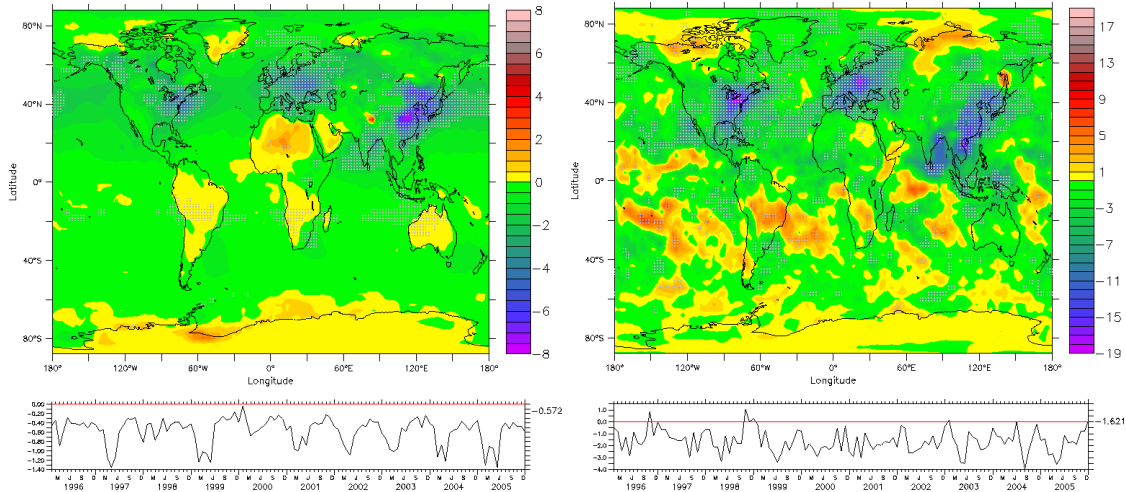


Figure 20: Decadal mean of the anthropogenic aerosol effect in shortwave radiation at the top of the atmosphere in the ARG configuration. The left panel depicts clear sky conditions, the right panel all sky conditions. Hatched regions represent areas with a statistical significance (90% confidence level of a two sided t-test, based on the annual mean fluxes).

#### 6.1.2 KK configuration

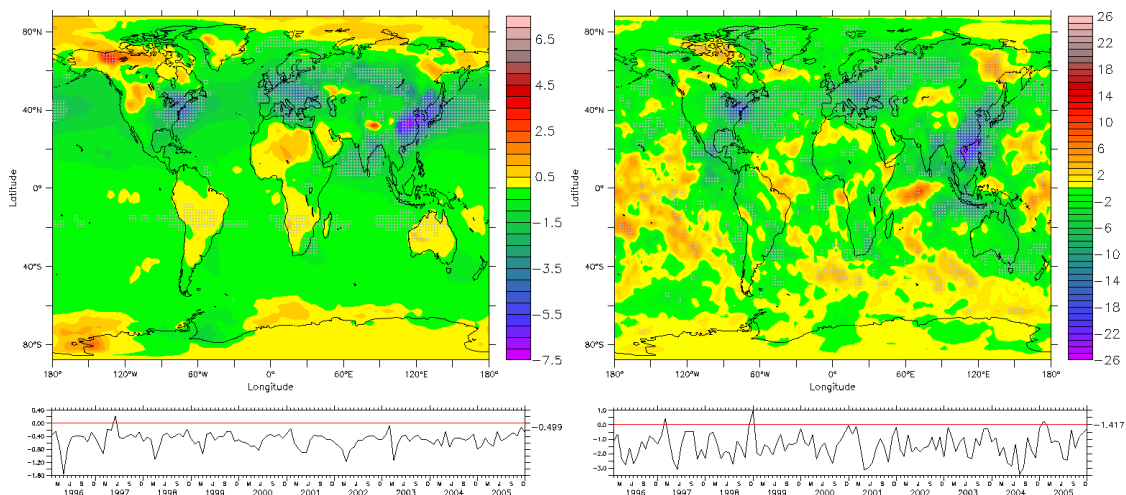


Figure 21: As Fig. 20, but for the KK configuration.

## 6.2 Lightning $\text{NO}_x$ based radiative flux disturbances

### 6.2.1 ARG - preindustrial

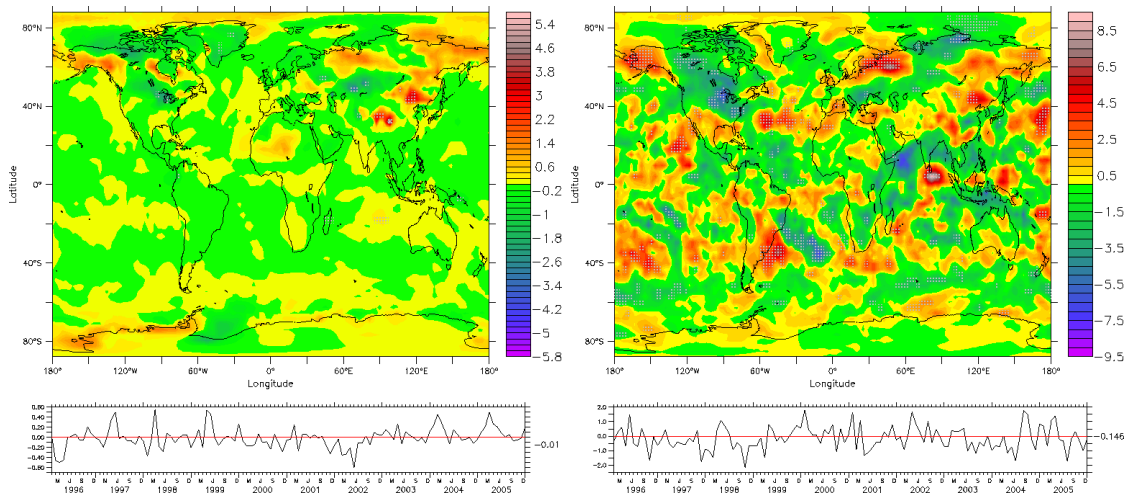


Figure 22: Decadal mean of the shortwave radiative flux disturbance in the ARG configuration at the top of the atmosphere for preindustrial conditions. The left panel depicts clear sky conditions, the right panel all sky conditions. Hatched regions represent areas with a statistical significance (90% confidence level of a two sided t-test, based on the annual mean fluxes).

### 6.2.2 KK - present day

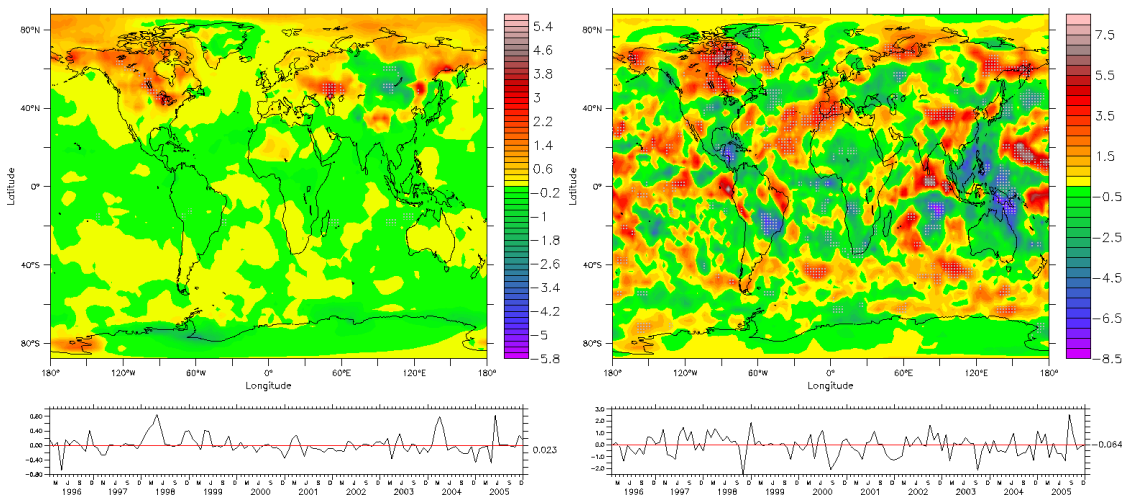


Figure 23: As Fig. 22, but for the KK configuration and present day conditions.

### 6.2.3 KK - preindustrial

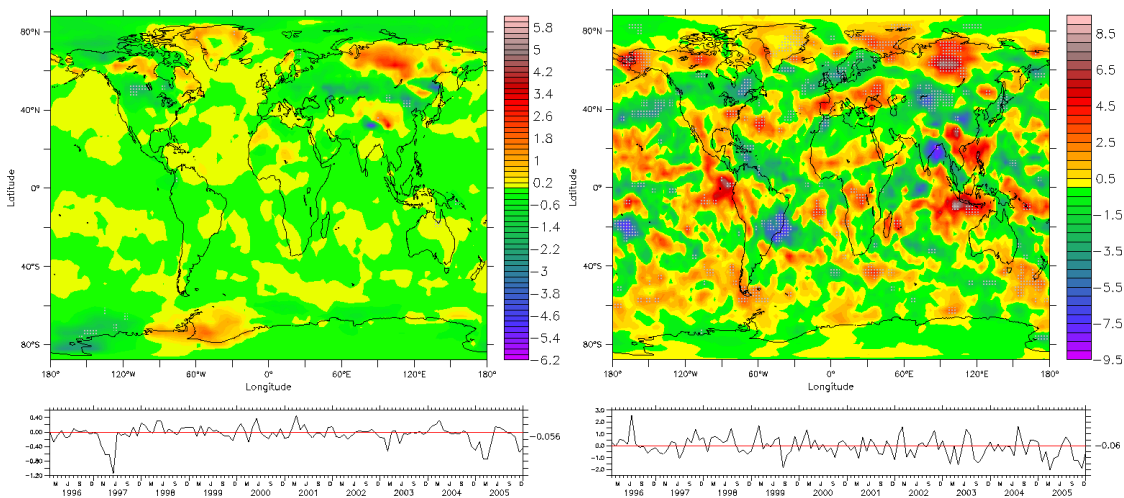


Figure 24: As Fig. 22, but for the KK configuration and preindustrial conditions.