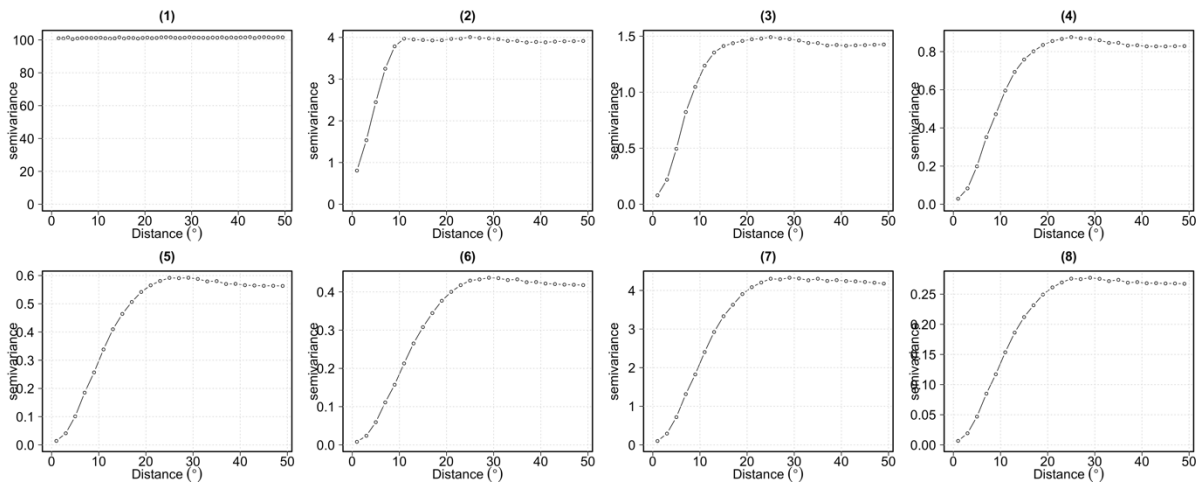
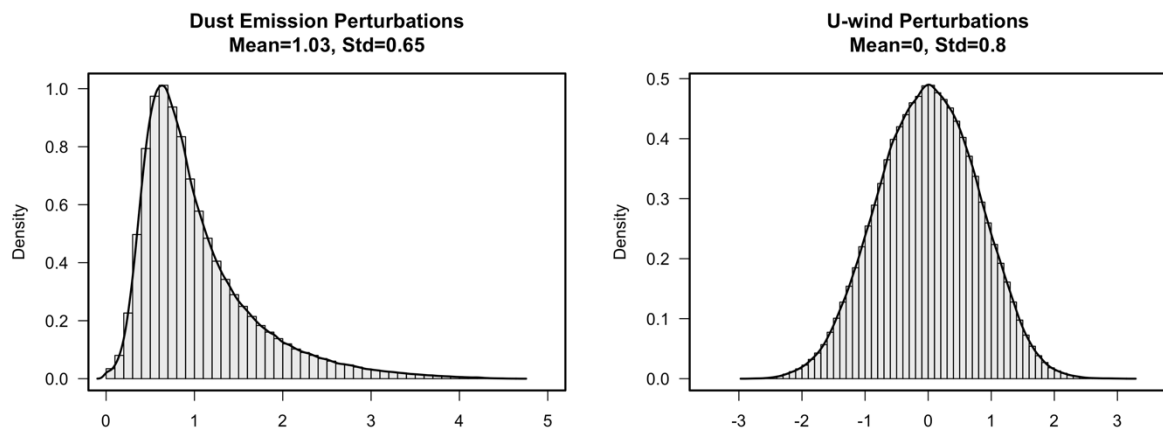
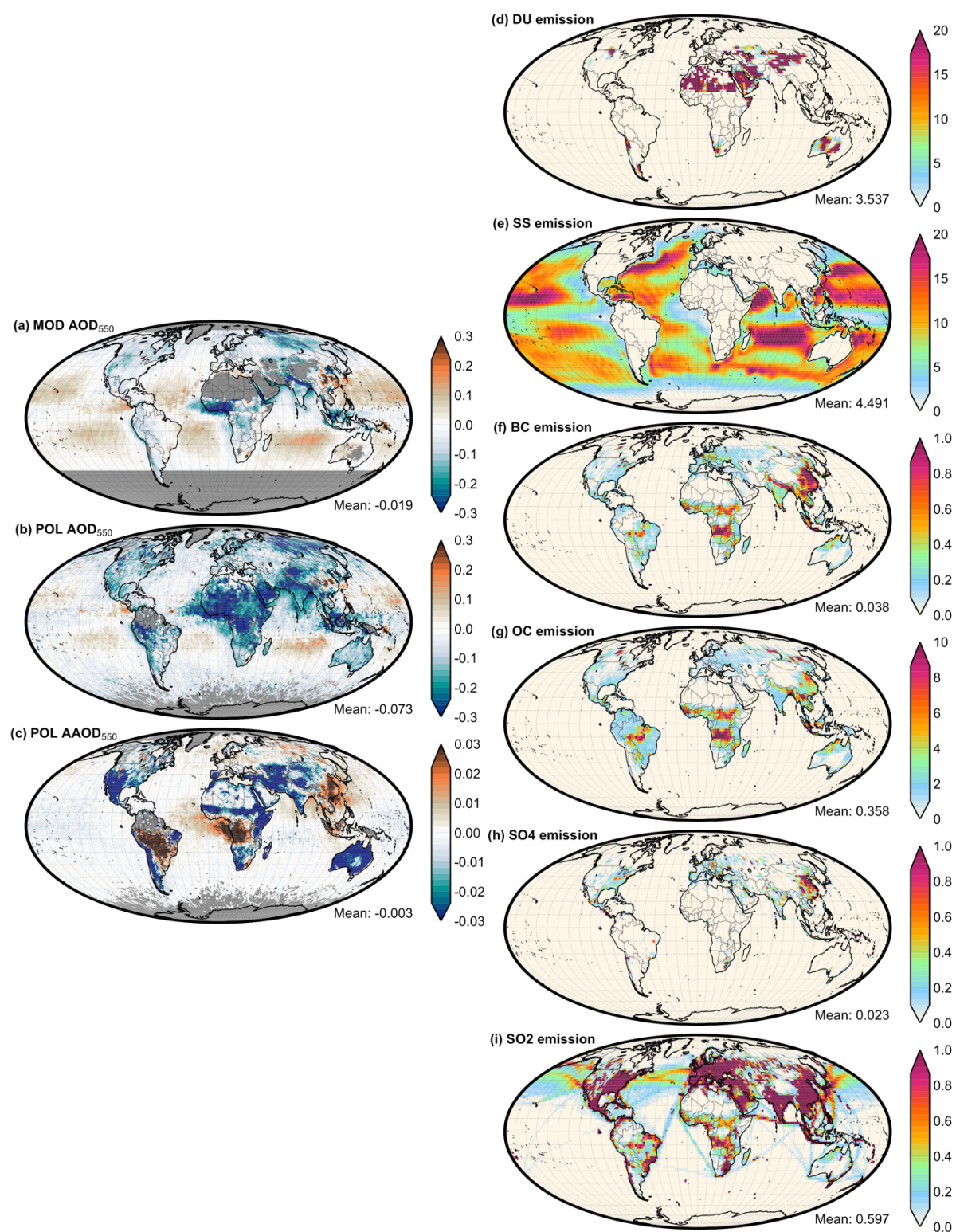
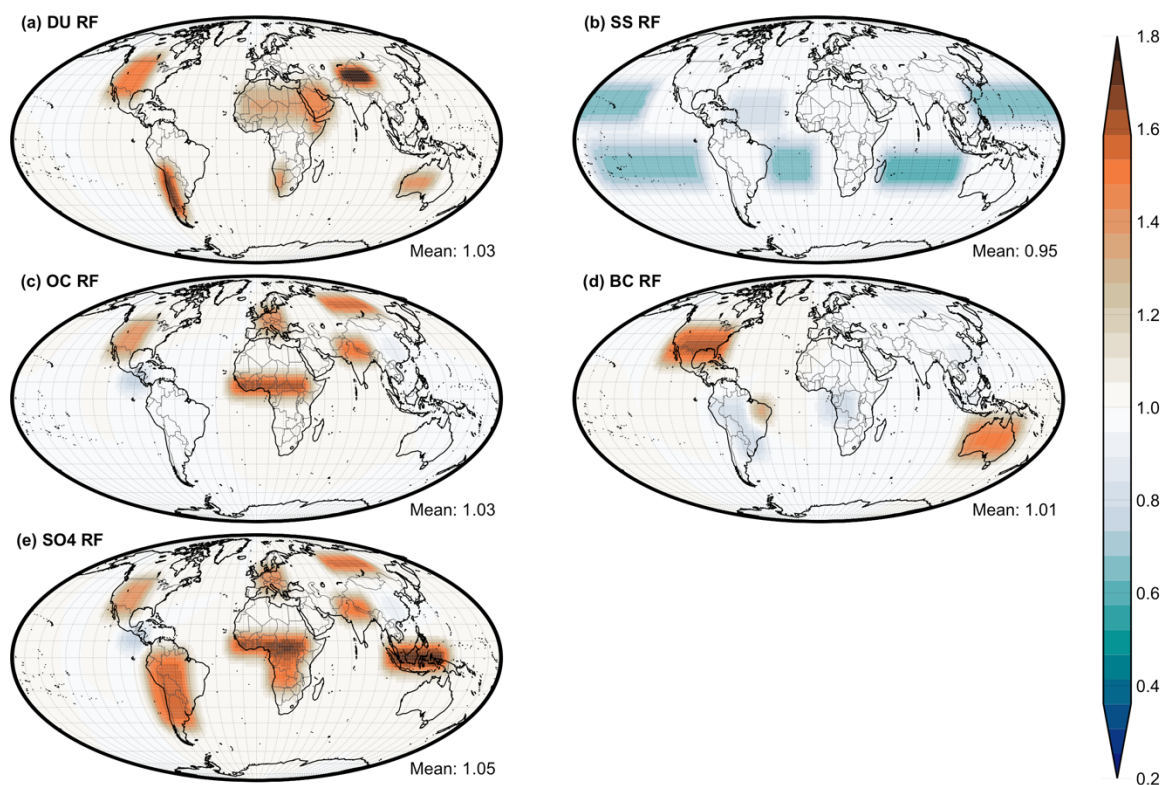
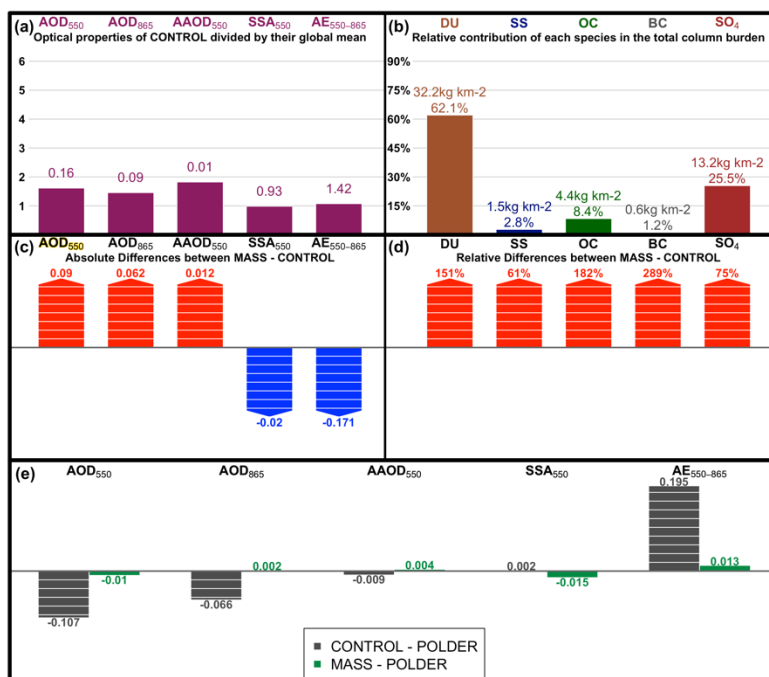
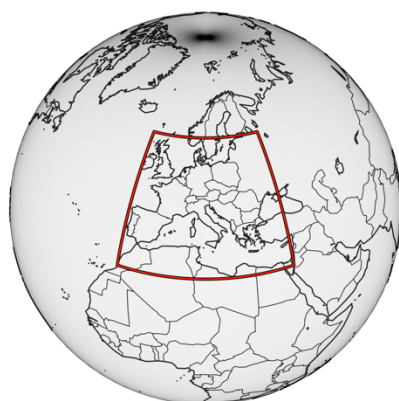
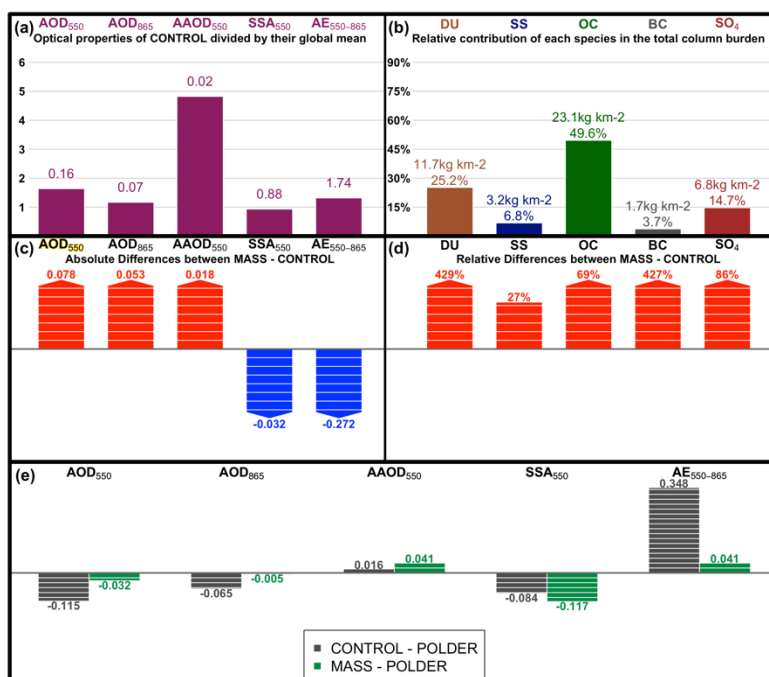
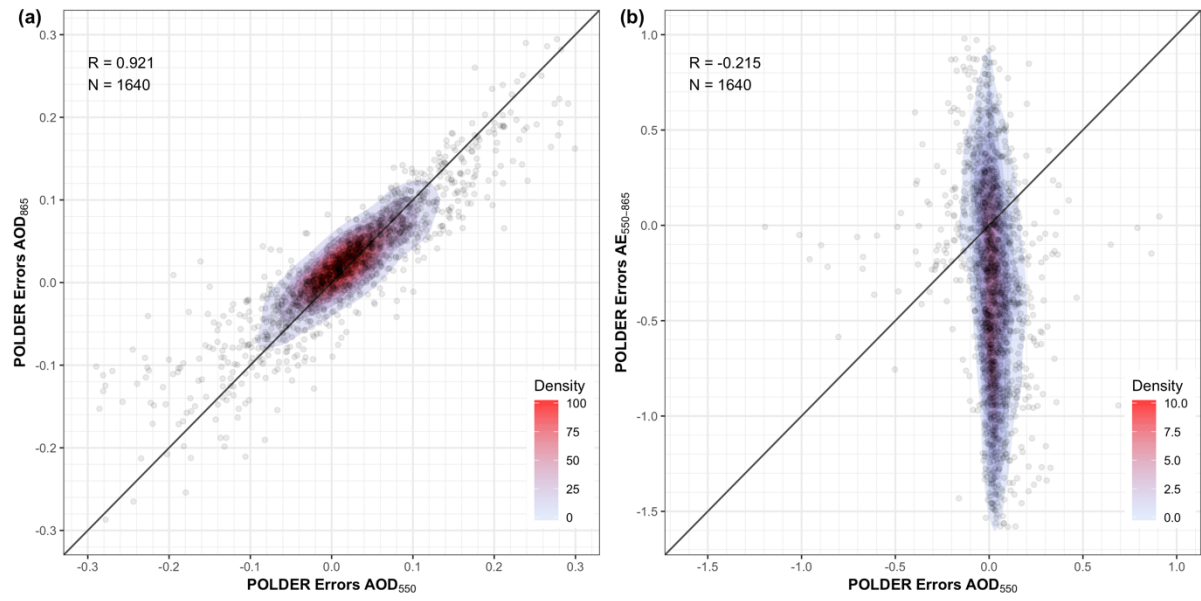
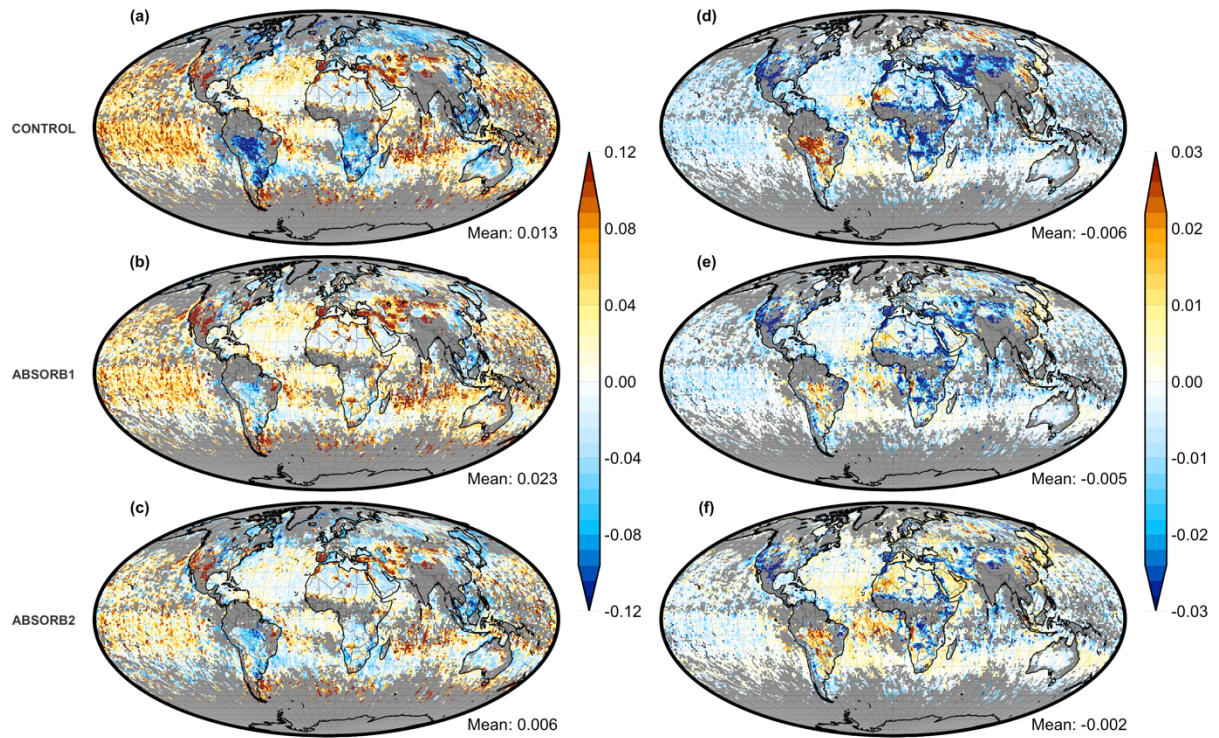
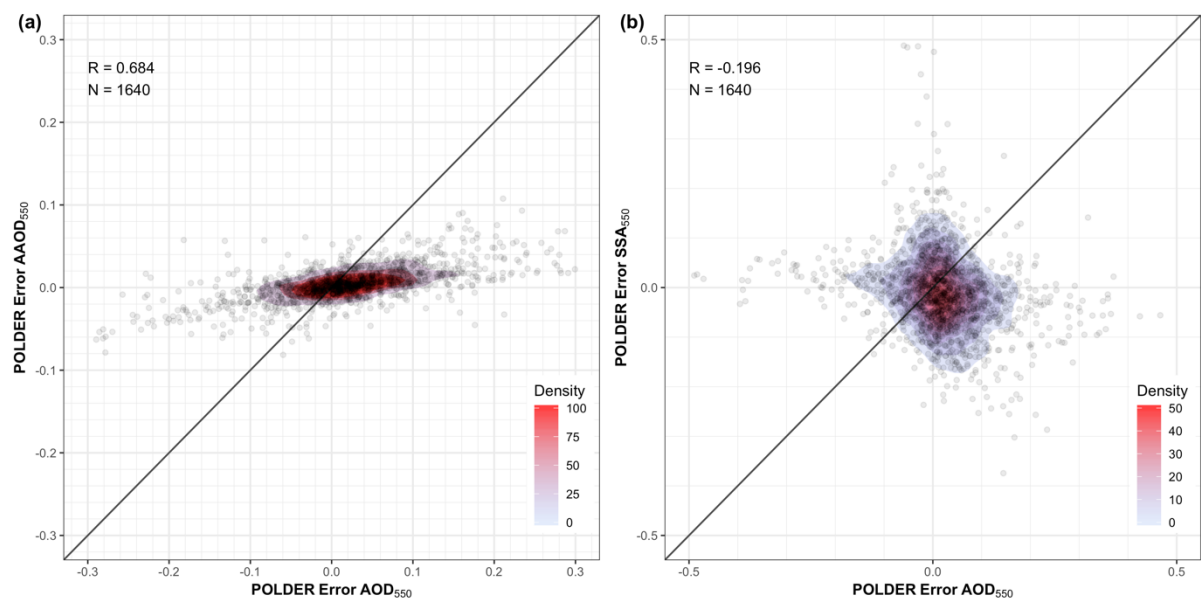
Supplementary

FigureS 1. Step by step the generation of the spatially correlated fields used to perturbe aerosol emission and wind. Each plot depicts the spatial field of one member in different stages.

FigureS 2. Variogram for the respective spatial fields shown in FigureS 1.

FigureS 3. Perturbations distribution for dust emission and U-component of the wind using all grid-box values.

FigureS 4. ECHAM-HAM minus (a) MODIS-DT AOD$_{550}$, (b) POLDER AOD$_{550}$ and POLDER AAOD$_{550}$ along with the emission fluxes (kg·km^{2}·day^{-1}) of all aerosol species in the model (d-i) for 2006.

FigureS 5. Emission Rescaling Factors (RF) used in the experiments for (a) DU, (b) SS, (c) OC, (d) BC, (e) SO_4 and SO_2.

FigureS 6. An overview of the changes due to POLDER AOD$_{550}$ assimilation over Europe. (a) Depicts the mean values of five aerosol optical properties and the column burden of five aerosol species as simulated in the experiment MASS. The percentage in the column burdens of each species specifies the relative contribution of each species in the total column burden. (b) Illustrates the changes caused in aerosol optical properties and mixing ratio due to the assimilation of AOD$_{550}$ (MASS – CONTROL). (c) Displays the aerosol optical properties bias in comparison to POLDER for the experiments CONTROL and MASS.

FigureS 7. An overview of the changes due to POLDER AOD$_{550}$ assimilation over South America. (a) Depicts the mean values of five aerosol optical properties and the column burden of five aerosol species as simulated in the experiment MASS. The percentage in the column burdens of each species specifies the relative contribution of each species in the total column burden. (b) Illustrates the changes caused in aerosol optical properties and mixing ratio due to the assimilation of AOD$_{550}$ (MASS – CONTROL). (c) Displays the aerosol optical properties bias in comparison to POLDER for the experiments CONTROL and MASS.

FigureS 8. POLDER errors estimated by comparing it with AERONET for (a) AOD_{550} against AOD_{865} and for (b) AOD_{550} against $AE_{550-865}$. R denotes Pearson Correlation.

FigureS 9. Differences between the experiments CONTROL (a,d), ABSORB1 (b,e) and ABSORB2 (c,f) and POLDER for $AAOD_{550}$ and SSA_{550}. All fields are spatiotemporally collocated to the available measurements of POLDER for the period 20th of July to 28th of August 2006. Grey-filled grid cells indicate the absence of any valid POLDER measurements for the study period.

FigureS 10. POLDER errors estimated by comparing it with AERONET for (a) AOD_{550} against $AAOD_{865}$ and for (b) AOD_{550} against SSA_{550}. R denotes Pearson Correlation.