Figure S1: Timeseries of available data from operational GEOS systems coloured based on the configuration of GOCART
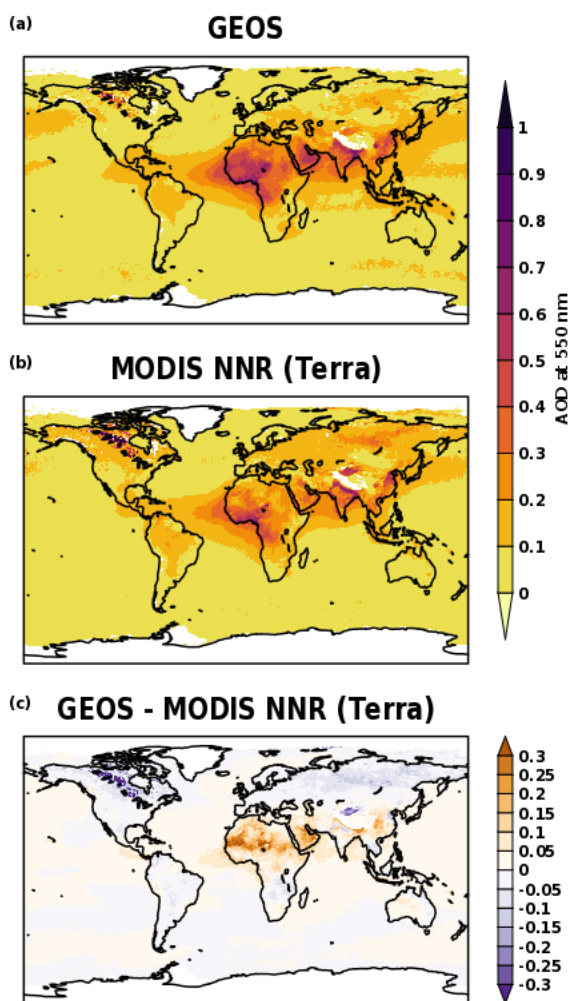
Figure S2: Average AOD at 550 for the period of January 2016 through December 2019 in the (a) GEOS GOCART2G benchmark simulation, (b) MODIS NNR observational product from Terra, and (c) the difference between the model and observations.
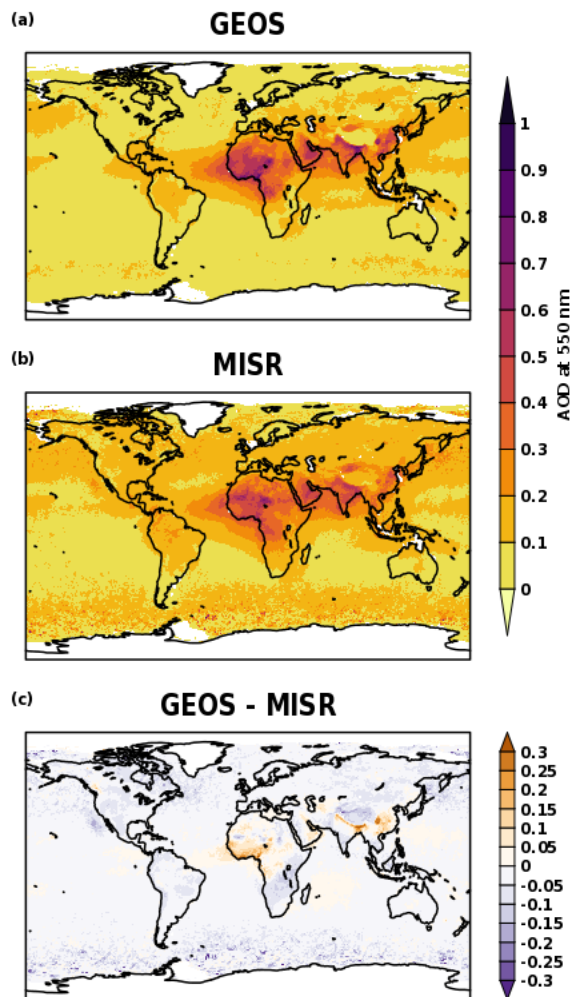
Figure S3: Average AOD at 550 for the period of January 2016 through December 2019 in the (a) GEOS GOCART2G benchmark simulation, (b) MISR, and (c) the difference between the model and observations.
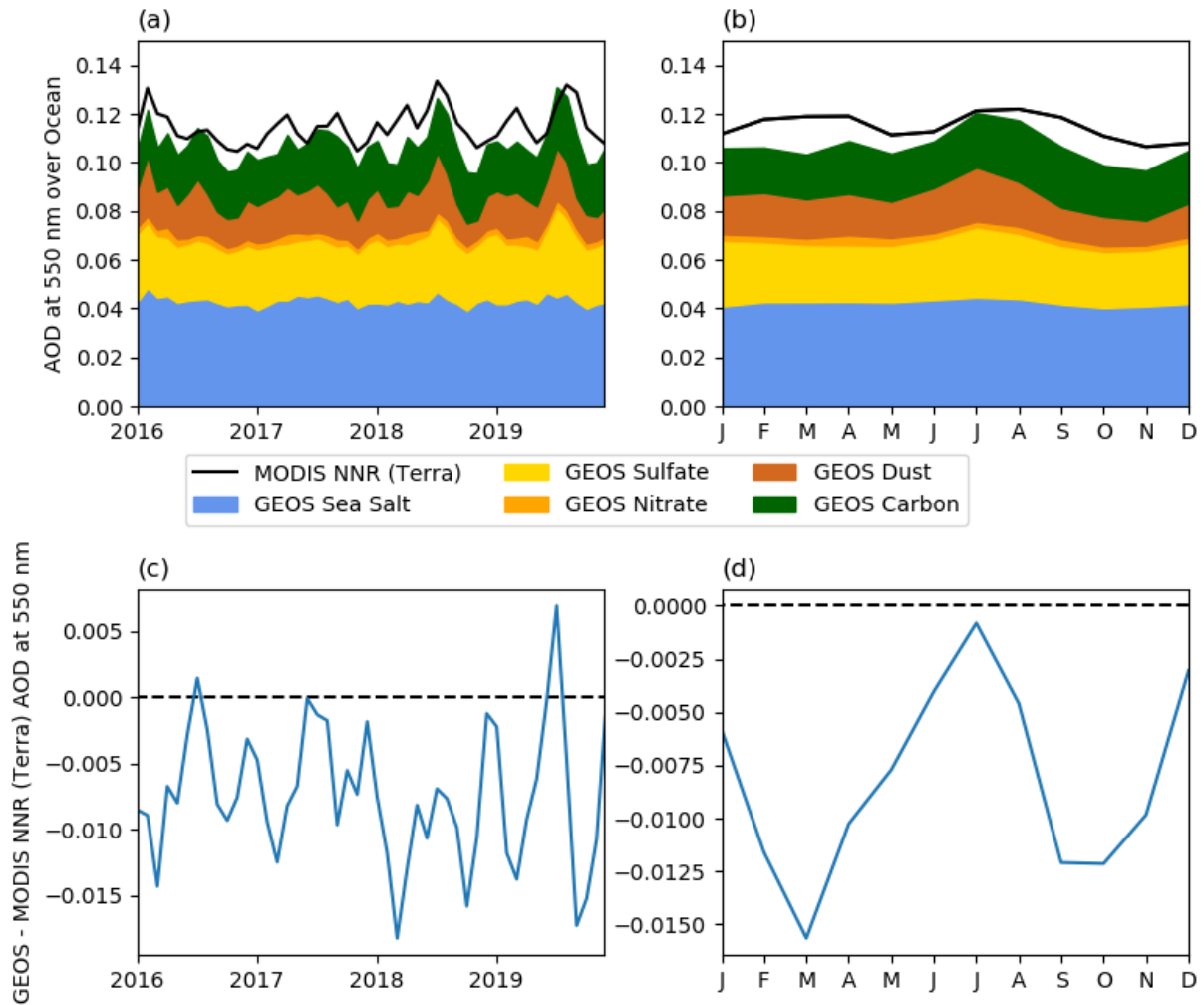
Figure S4: Timeseries of ocean area-averaged (a) monthly mean AOD from the Terra MODIS NNR observational product and the speciated AOD from the GEOS GOCART2G benchmark simulation, (b) mean seasonal cycle, and the difference between the model and observations for the (c) monthly mean AOD and (d) seasonal cycle of AOD.
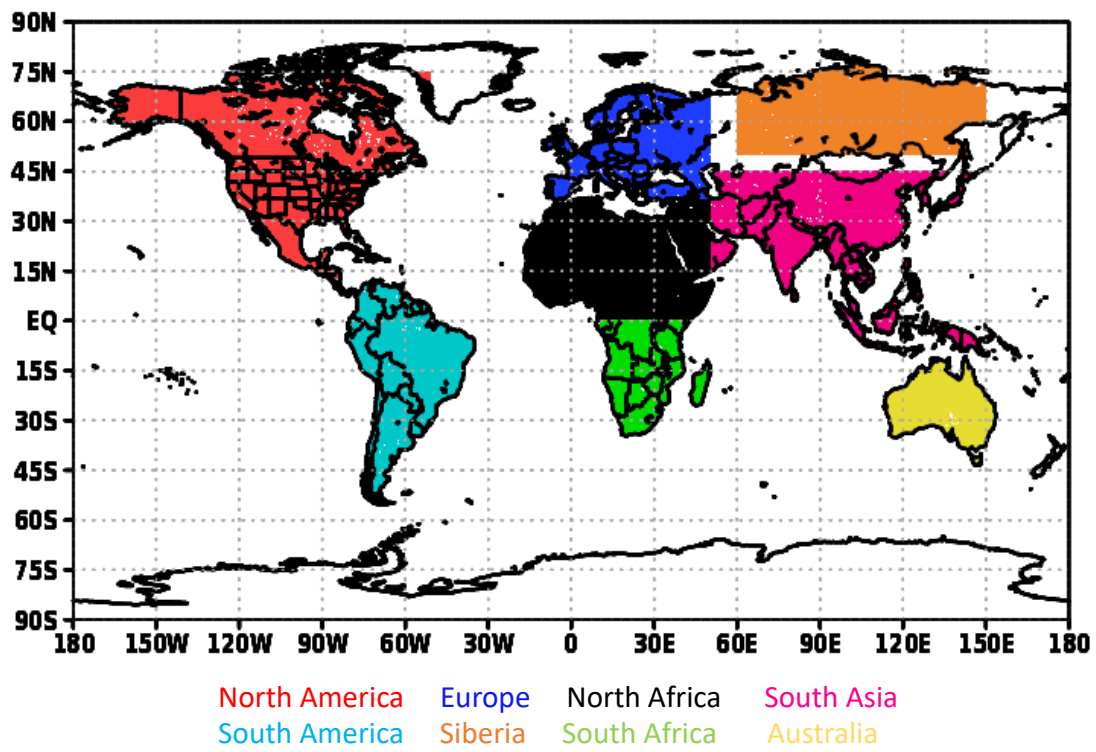
Figure S5: Map showing the area averaged regions for the AOD timeseries.
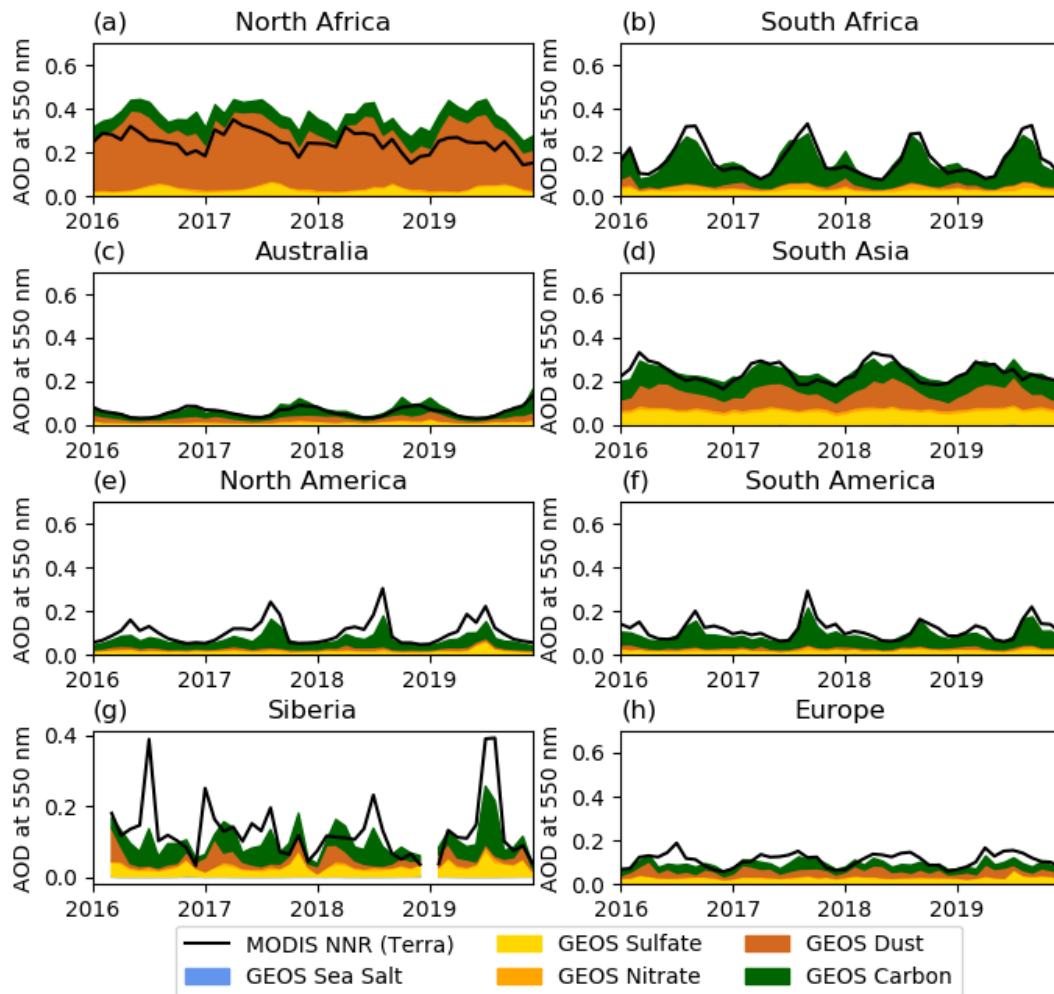
Figure S6: Timeseries of area-averaged monthly mean AOD from the Terra MODIS NNR observational product and the speciated AOD from the GEOS GOCART2G benchmark simulation over (a) North Africa, (b) South Africa, (c) Australia, (d) South Asia, (e) North America, (f) South America, (g) Siberia, and (h) Europe.
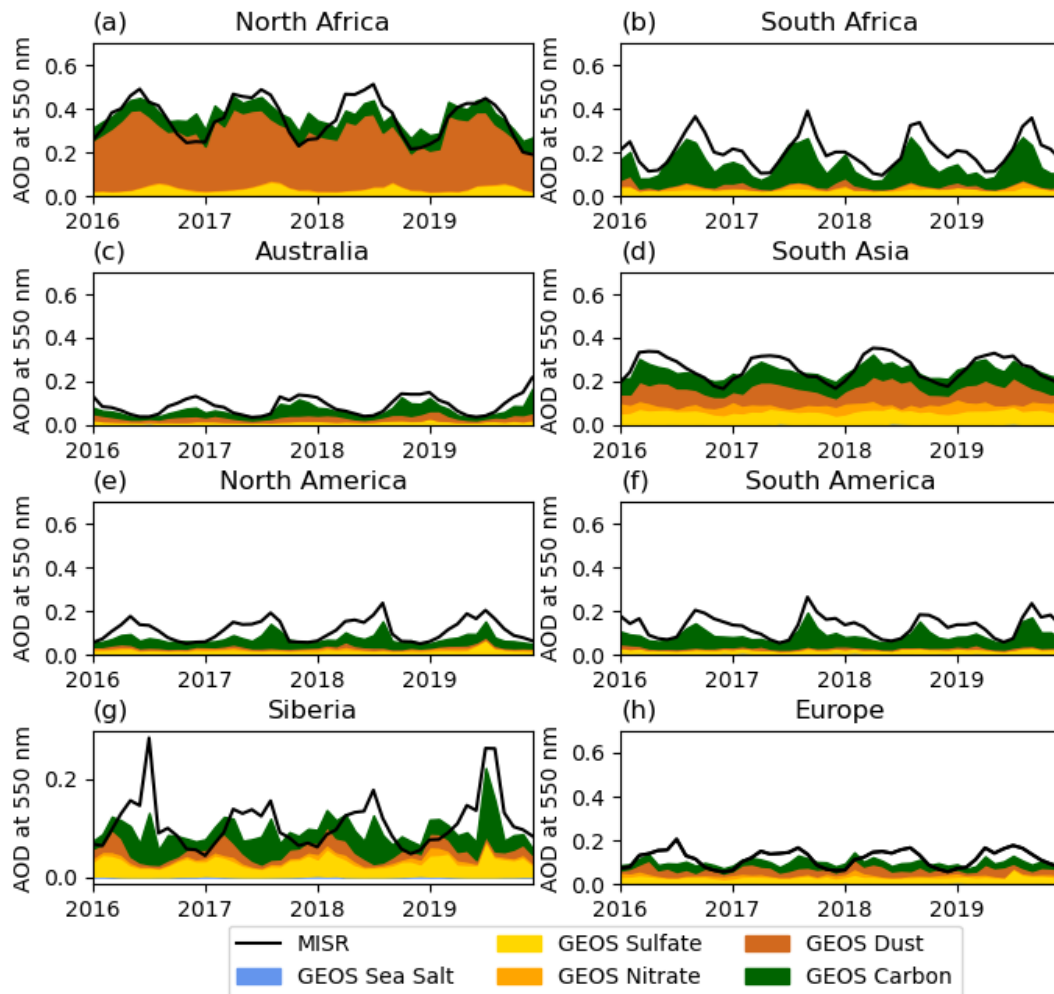
Figure S7: Timeseries of area-averaged monthly mean AOD from MISR and the speciated AOD from the GEOS GOCART2G benchmark simulation over (a) North Africa, (b) South Africa, (c) Australia, (d) South Asia, (e) North America, (f) South America, (g) Siberia, and (h) Europe.
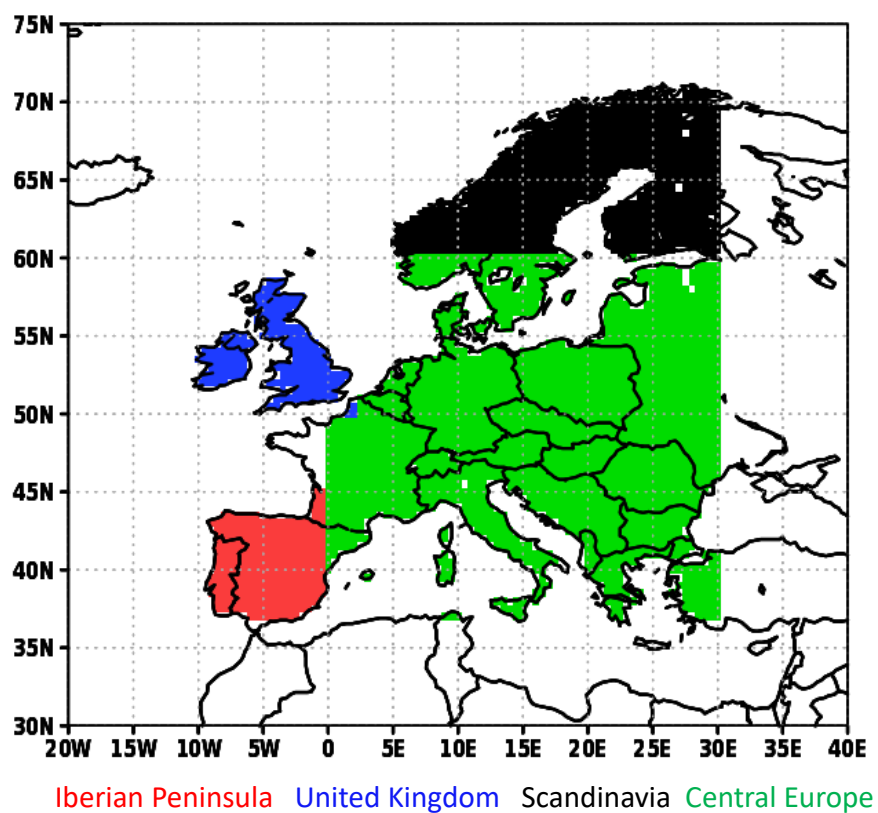
Figure S8: Map of regions used for the area averaged AOD over Europe
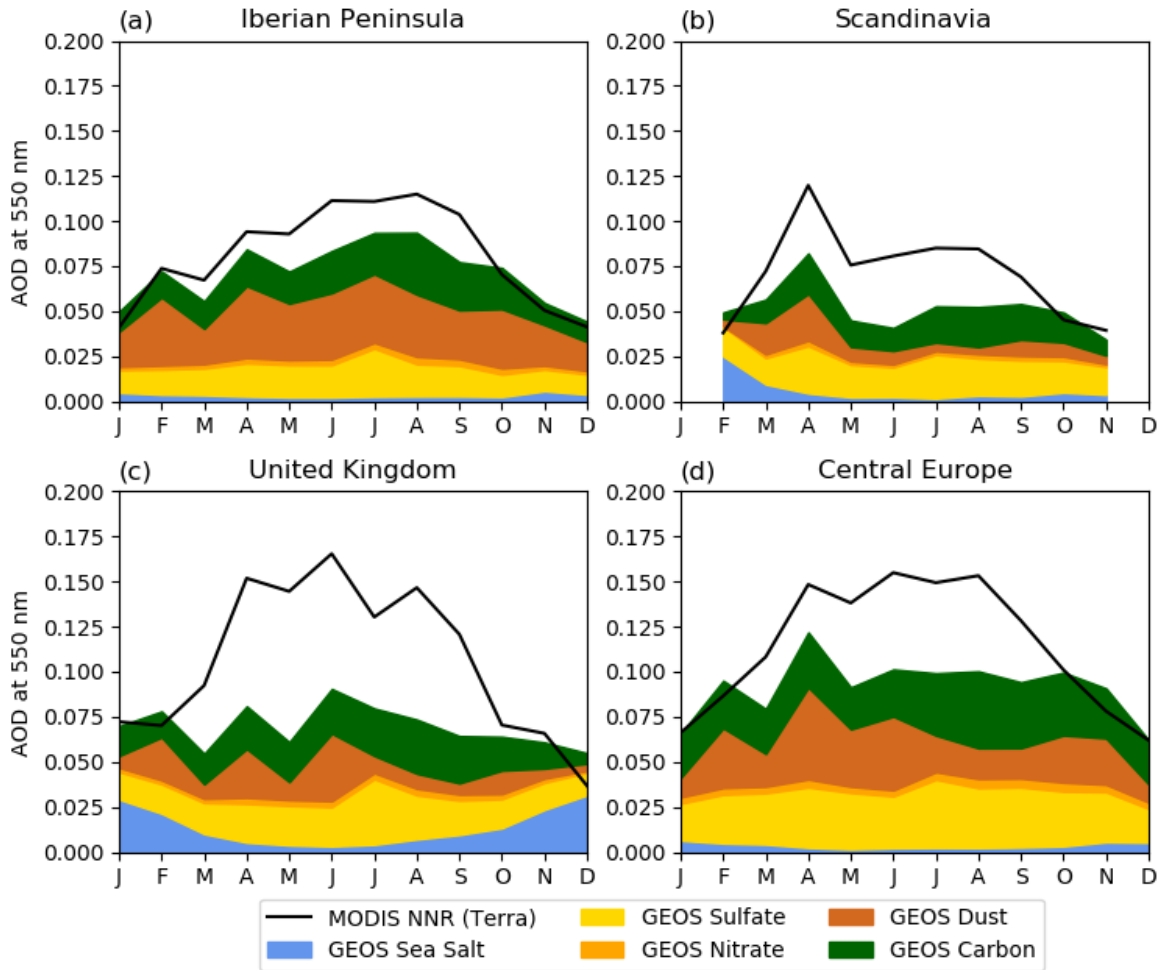
Figure S9: Timeseries of area-averaged monthly mean AOD from the Terra MODIS NNR observational product and the speciated AOD from the GEOS GOCART2G benchmark simulation over (a) the Iberian Peninsula, (b) Scandinavia, (c) the United Kingdom, and (d) central Europe.
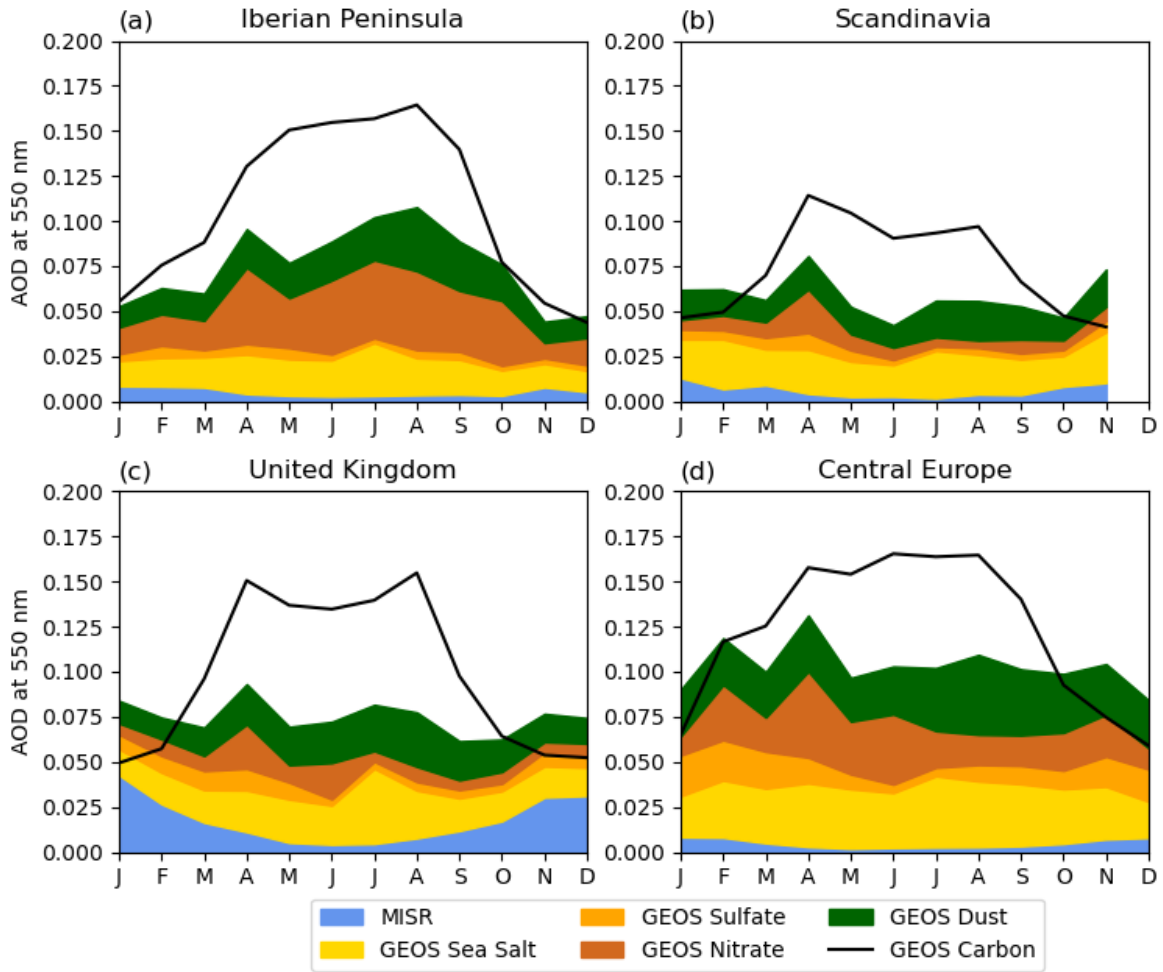
Figure S10: Timeseries of area-averaged monthly mean AOD at 550 nm from the Terra MODIS NNR observational product and the speciated AOD from the GEOS GOCART2G benchmark simulation over (a) the Iberian Peninsula, (b) Scandinavia, (c) the United Kingdom, and (d) central Europe.
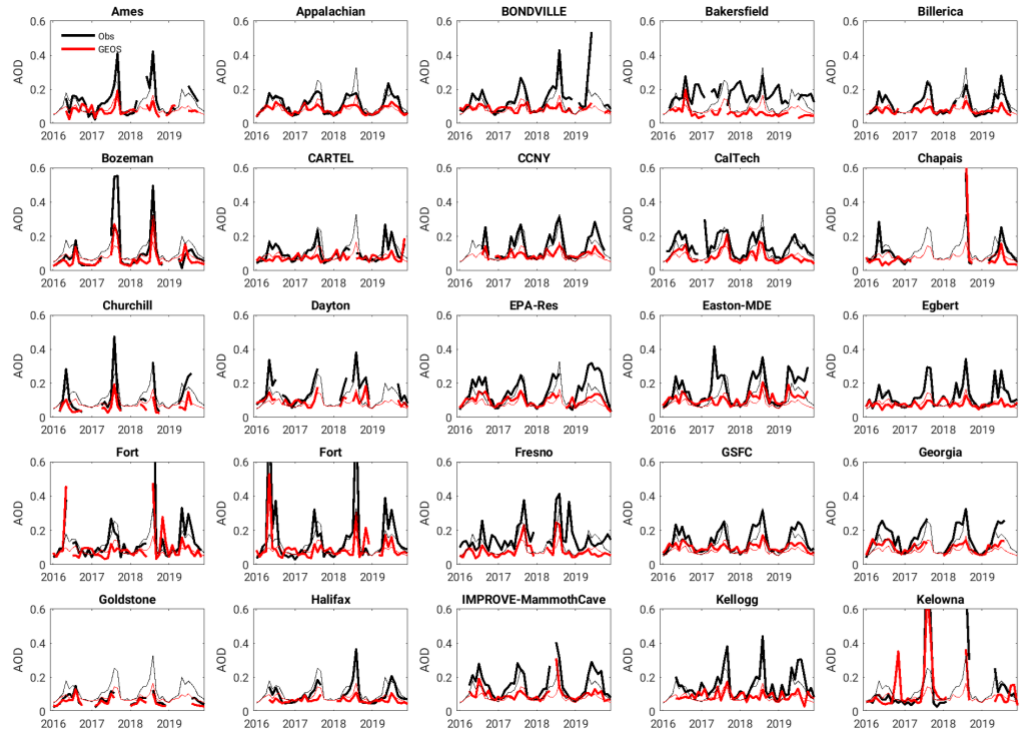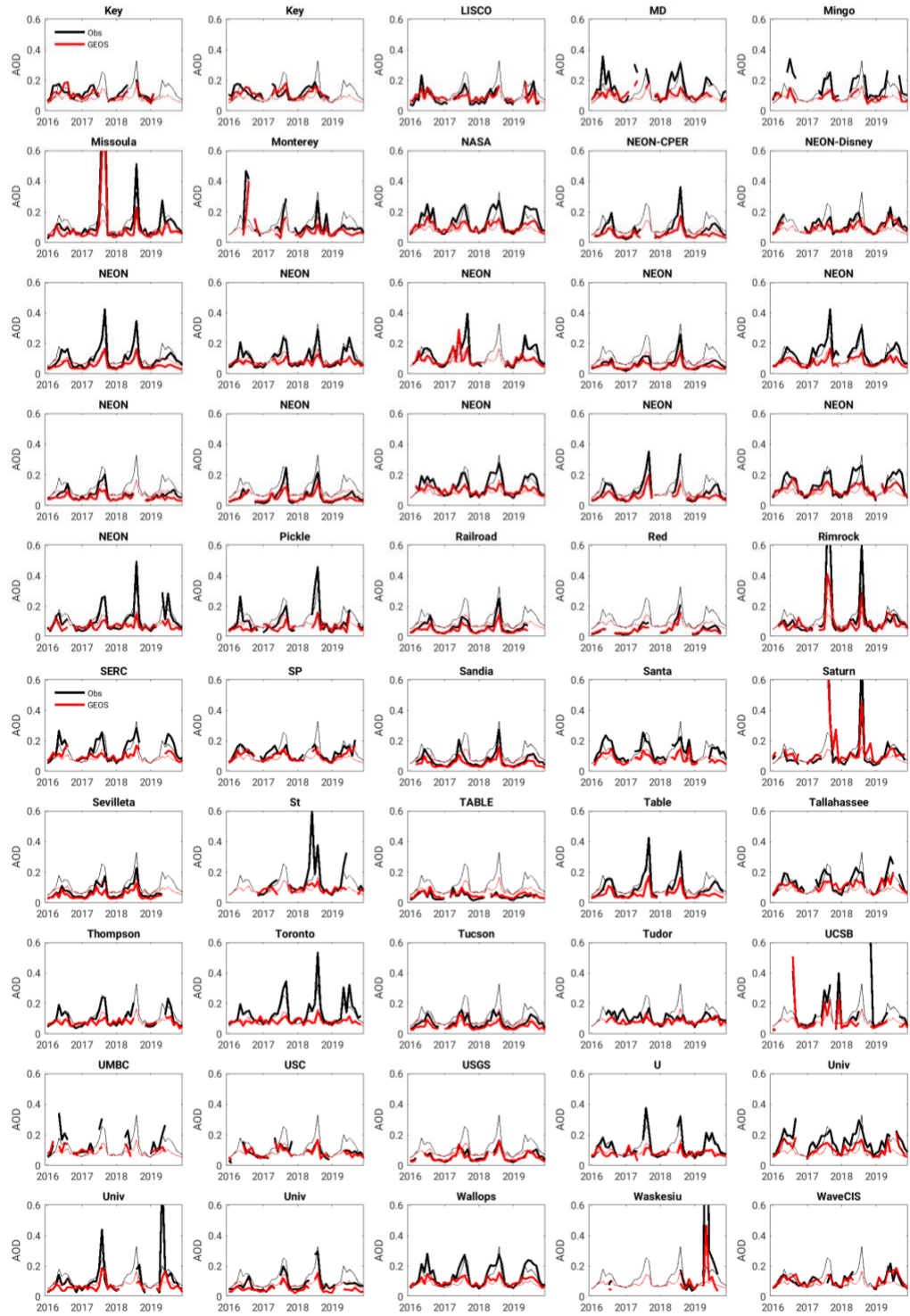
Figure S11: Time series of AOD at 550 nm from GEOS and Aeronet sites across the United States and Canada (bold lines). Thin lines indicate the mean for all sites.
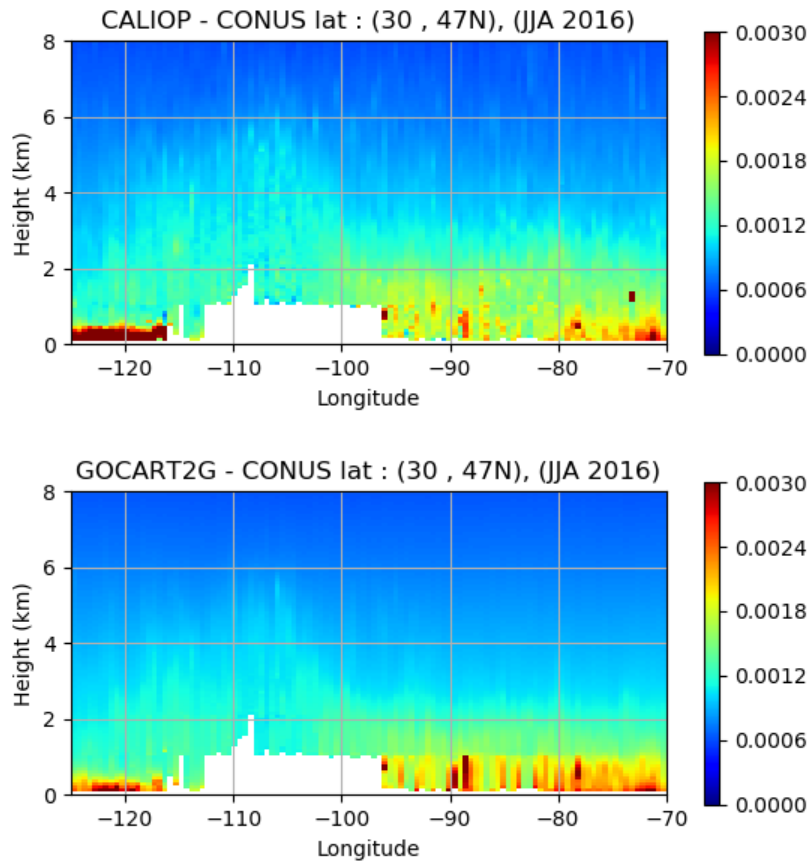
Figure S12: Regional 3-month average (JJA 2016) of CALIOP attenuated backscatter coefficient (km^{-1}sr^{-1}) at 532nm over the continental United States (30ºN-47ºN, 120ºW-70ºW) on the top. On the bottom, GEOS GOCART2G attenuated backscatter coefficient sampled on the CALIPSO track for the same period.
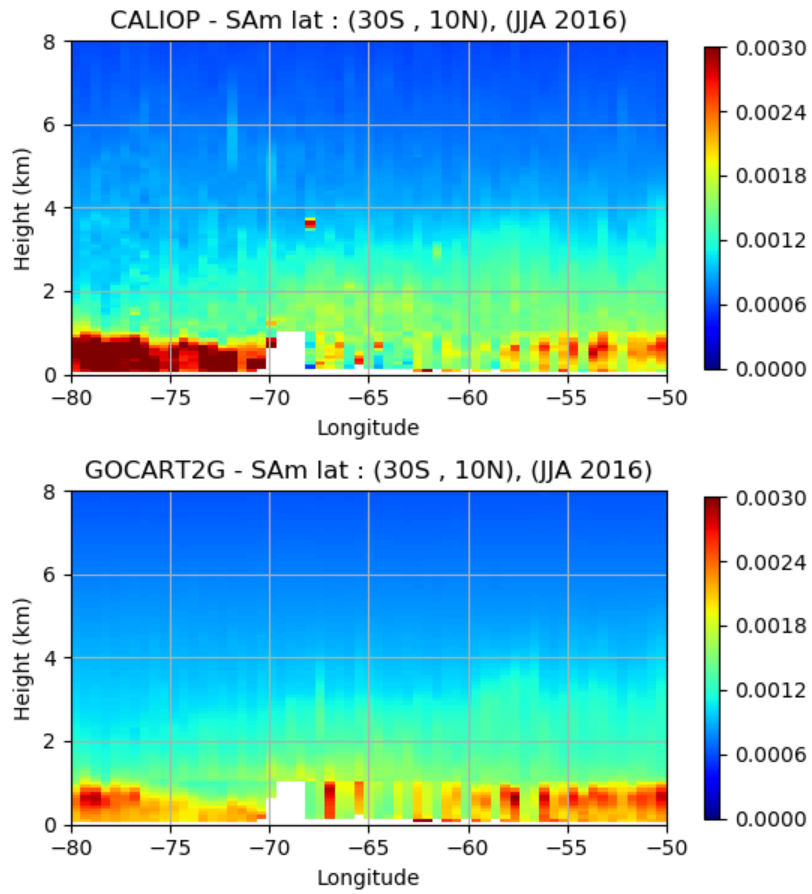
Figure S13: Same as figure S12 but over South America (30ºS-10ºN, 60ºE-20ºE).
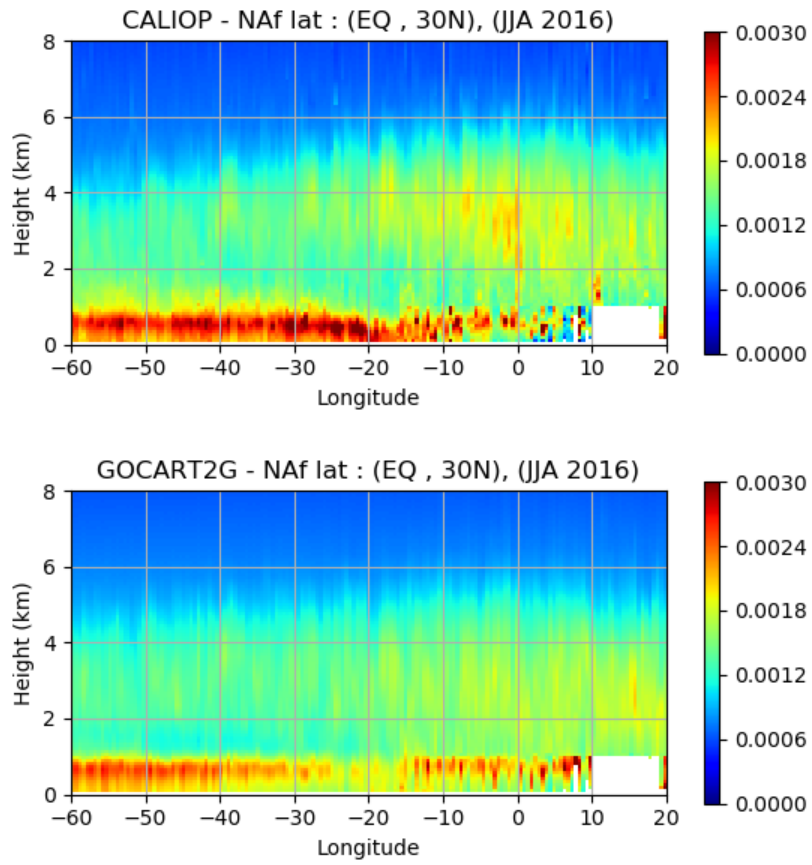
Figure S14: Same as figure S12 but over northern Africa (Eq-30ºN, 60ºE-20ºE).
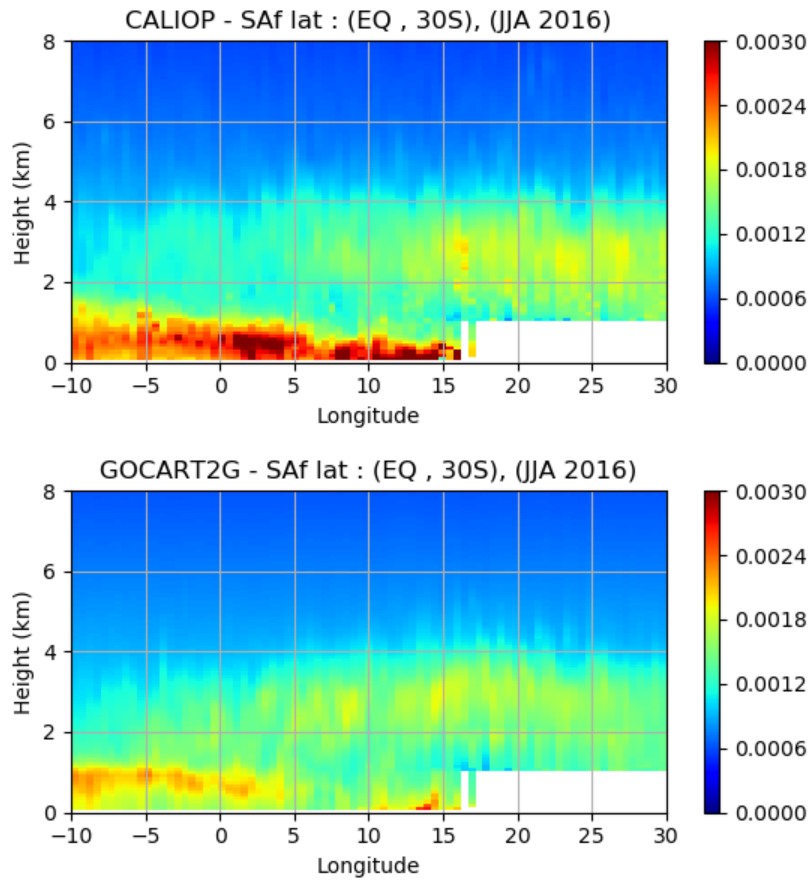
Figure S15: Same as figure S12 but over southern Africa (30°S-Eq, 10°W-30°E).