

Supplementary information to “Significant underestimation of radiative forcing by aerosol–cloud interactions derived from satellite-based methods”

Hailing Jia^{1,2,3}, Xiaoyan Ma^{1*}, Fangqun Yu^{2*} & Johannes Quaas³

¹Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disasters, and Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, School of Atmospheric Physics, Nanjing University of Information Science & Technology, Nanjing, China

²Atmospheric Sciences Research Center, University at Albany, Albany, NY, USA

³ Institute for Meteorology, Universität Leipzig, Leipzig, Germany

Supplementary Table 1. Modeled and satellite-based RF_{aci} ($W\ m^{-2}$) for different regions, respectively.

Region	Modeled RF_{aci} ($W\ m^{-2}$)	Satellite-based RF_{aci} ($W\ m^{-2}$)			
		Aero_Cld_Modis	Aero_Cld	Cld	All_Cld
Global	-0.79	-0.36	-0.38	-0.75	-0.59
NH	-1	-0.57	-0.62	-1.21	-0.98
SH	-0.57	-0.15	-0.14	-0.28	-0.19
Land	-1.23	-0.24	-0.33	-1.22	-0.77
Ocean	-0.64	-0.4	-0.4	-0.59	-0.53
NPO	-0.94	-1.54	-1.67	-2.22	-2.17
NAM	-1.71	-0.33	-0.49	-2.12	-1.4
NAO	-1.45	-0.59	-0.59	-1.38	-1.05
EUR	-2.54	-0.7	-0.9	-1.86	-1.31
ASI	-2.22	-0.4	-0.58	-1.74	-1.3
TPO	-0.39	-0.22	-0.17	-0.21	-0.22
TAO	-0.51	-0.32	-0.31	-0.45	-0.42
AFR	-0.35	-0.12	-0.1	-0.61	-0.26
TIO	-0.15	-0.14	-0.09	-0.28	-0.14
SPO	-0.69	-0.11	-0.1	-0.09	-0.07
SAM	-0.6	-0.01	-0.07	-0.94	-0.4
SAO	-0.64	-0.17	-0.19	-0.2	-0.2
SIO	-0.57	-0.2	-0.18	-0.19	-0.19
OCE	-0.54	-0.04	-0.12	-0.22	-0.16

Supplementary Table 2. The list of the parameters, sources, and their corresponding temporal-spatial resolutions applied in present study.

Source	Time period	Temporal-spatial resolution	Parameters
CERES-SSF Edition 4A ¹	Jan 2002–Dec 2018	Daily, approximately 20×20 km ²	Surface type Surface-type percent coverage Solar zenith angle Shortwave TOA flux–upward Cloud fraction (<i>f</i>) ^a Cloud phase Cloud optical depth @3.7 μm (τ_c) Cloud effective radius @3.7 μm (r_e) Cloud liquid water path (<i>L</i>)
MODIS C61 ²	Jan 2002–Dec 2018	Daily, 1° × 1°	AOD @550 nm ^b
POLDER-3 ³	Mar 2005–Oct 2013	Daily, 1° × 1°	AOD & AOD _f @565 nm
MERRA-2 ⁴	Jan 2002–Dec 2018	Hourly, 0.5°×0.625°	AOD @550 nm Sulfate AOD@550 nm Black carbon AOD@550 nm Organic carbon AOD@550 nm Sea salt AOD@550 nm Angström exponent ^c (470-870 nm) Sulfate column mass density (SO4)
MACv2 ⁵		Monthly, 1° × 1°	Anthropogenic fractions of AOD _f from AeroCom phase1 ⁶ and phase2 ⁷ models

^a *f* is derived by clear area percent coverage.

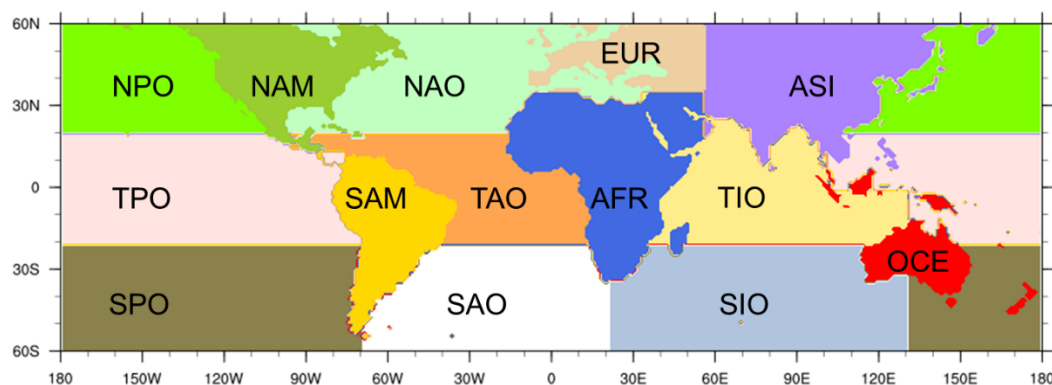
^b MODIS AOD is from the Dark Target and Deep Blue merged aerosol product.

^c Angström exponent is used to calculate aerosol index (AI = AOD×Angström exponent)

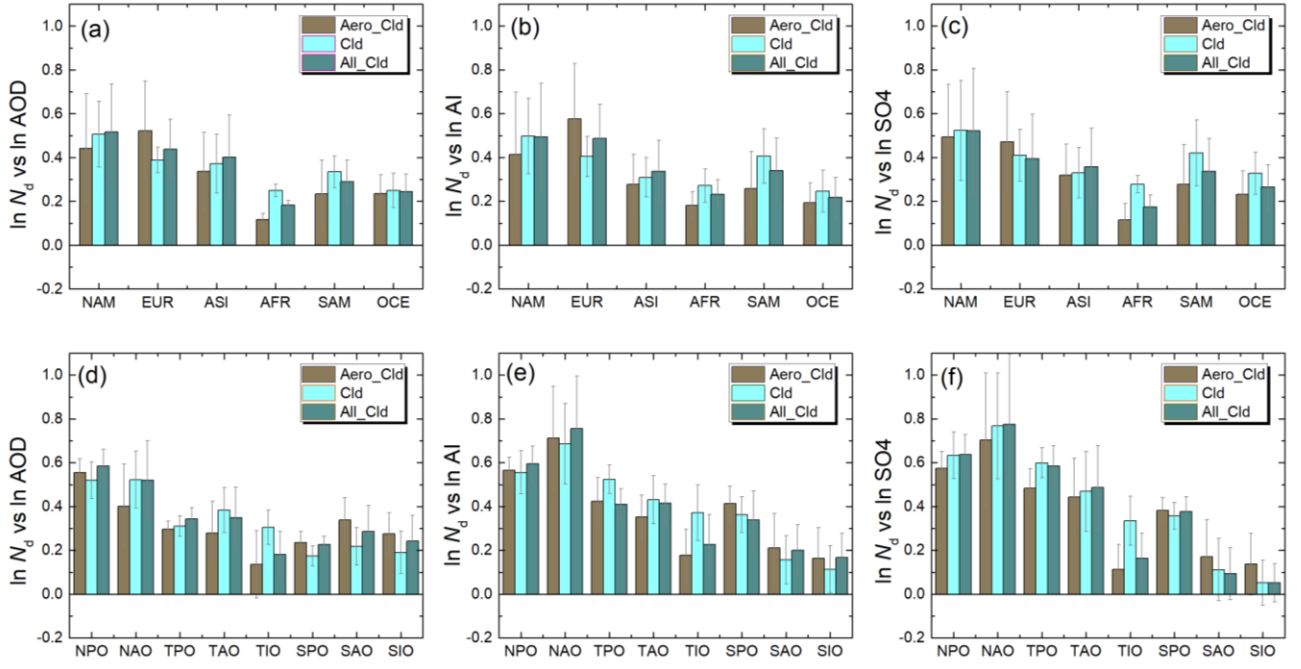
Supplementary Table 3. Global (60°S to 60°N) annual mean MERRA-2 daily-based and POLDER-3 monthly-based RF_{aci} (in $W m^{-2}$) estimated with different anthropogenic fractions, respectively. The corresponding global annual averages of anthropogenic fractions of AOD_f are also shown. The associated spatial distributions are given in Supplementary Figure 9.

Source	Anthropogenic fractions of AOD_f	MERRA-2 daily-based RF_{aci}	POLDER-3 monthly-based RF_{aci}
AeroCom 1	0.49	-1.68	-1.67
AeroCom 2	0.37	-1.07	-1.08
GC-APM	0.40	-1.41	-1.42
GC-APM-SS30	0.32	-1.09	-1.02

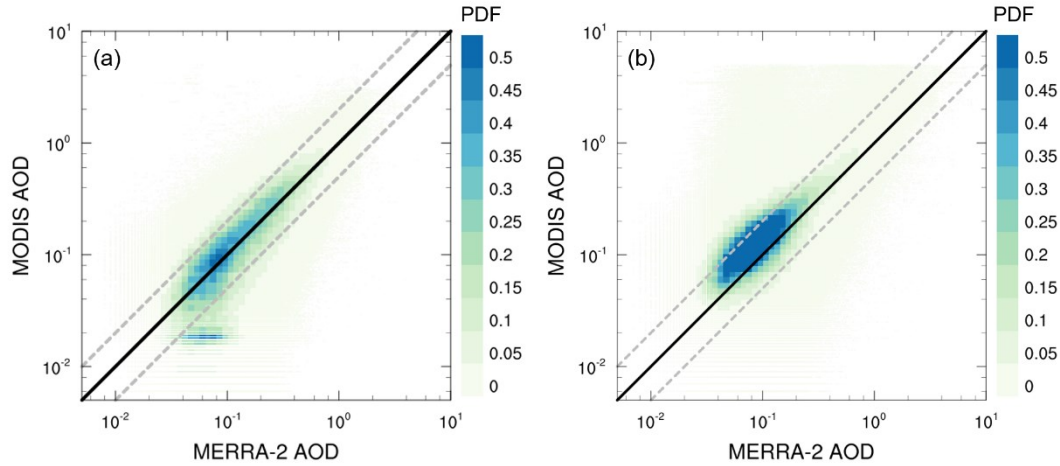
*Anthropogenic fractions of AOD_f are acquired from the simulations of (1) AeroCom phase1 models⁶ with the preindustrial year 1750 as reference (AeroCom 1), (2) AeroCom phase2 models⁷ with the preindustrial year 1850 as reference (AeroCom 2), (3) GEOS-Chem-APM⁸ with AOD_f defined as the sum of AODs of sulfate, black carbon and organic aerosol (GC-APM), and (4) GEOS-Chem-APM with AOD_f defined as the sum of AODs sulfate, black carbon and organic aerosol and 30 % sea salt aerosol (GC-APM-SS30), respectively.



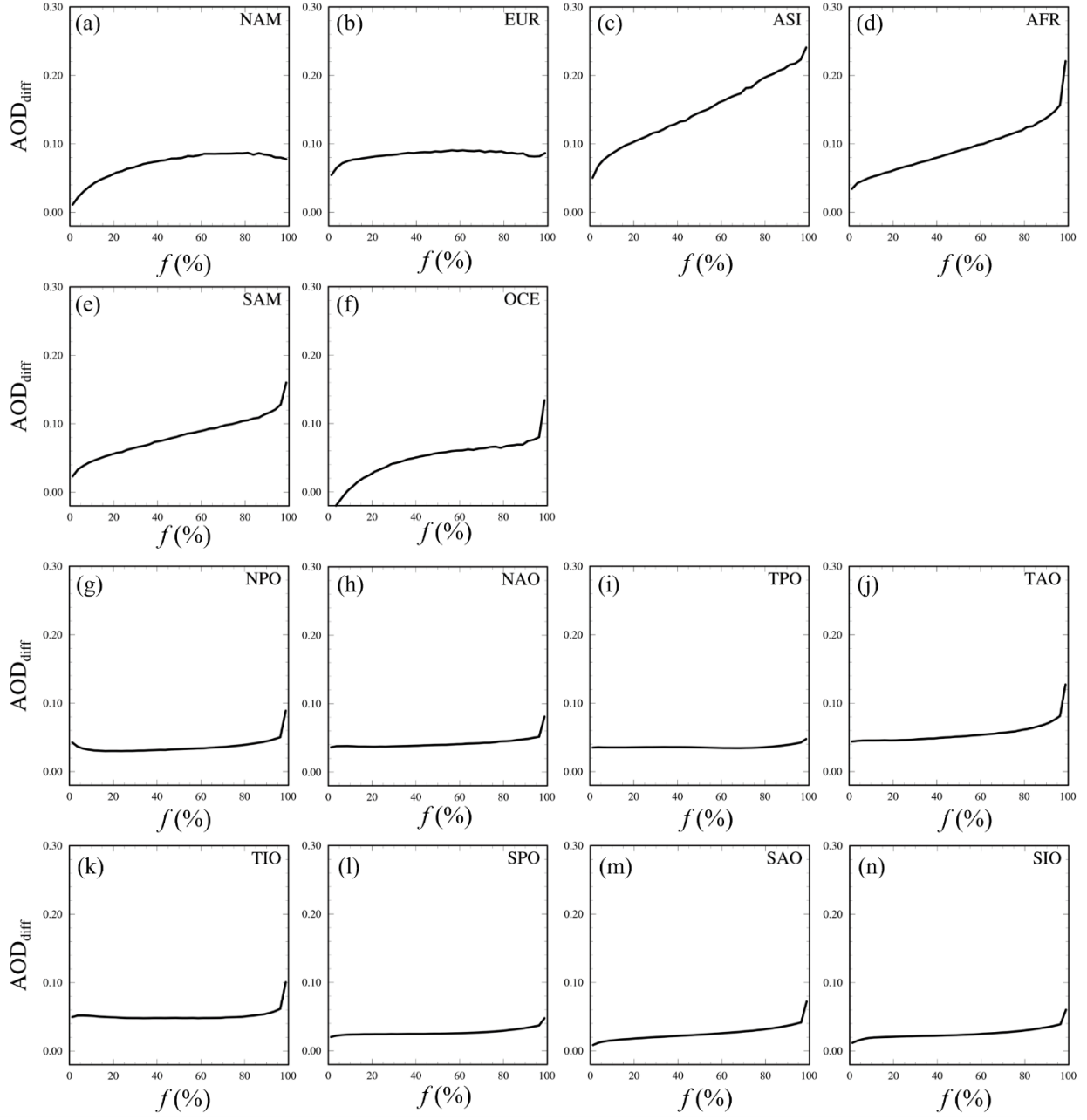
Supplementary Figure 1. The fourteen different regions used in this study. NPO: North Pacific Ocean; NAM: North America; NAO: North Atlantic Ocean; EUR: Europe; ASI: Asia; TPO: Tropical Pacific Ocean; TAO: Tropical Atlantic Ocean; AFR: Africa; TIO: Tropical Indian Ocean; SPO: South Pacific Ocean; SAM: South America; SAO: South Atlantic Ocean; SIO: South Indian Ocean; and OCE: Australia.



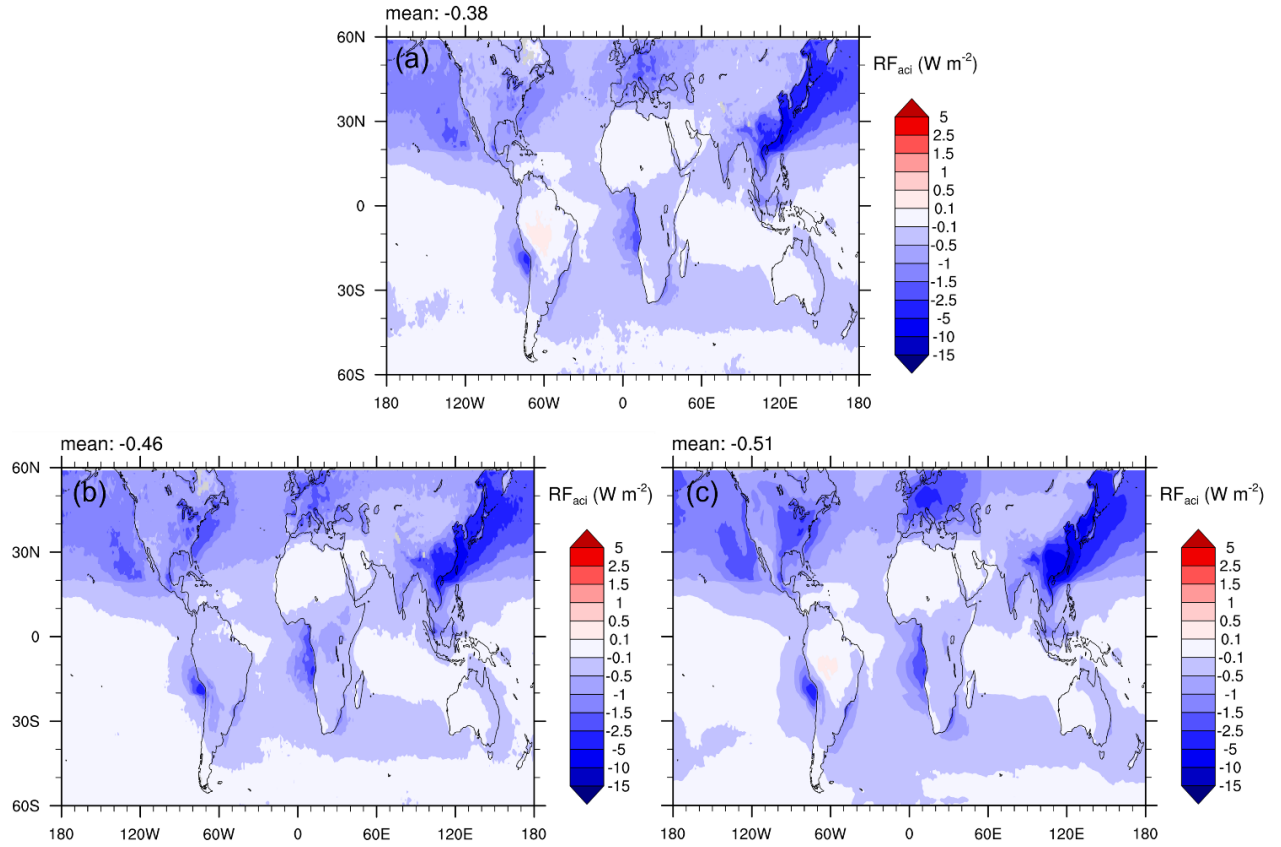
Supplementary Figure 2. Annual averaged slopes of the linear regressions between the logarithm of cloud droplet number concentration (N_d) and those of cloud condensation nuclei (CCN) proxies, including aerosol optical depth (AOD), aerosol index (AI), and sulfate column mass concentration (SO₄). Shown are the slopes of **a, d** $\ln N_d$ versus $\ln$ AOD, **b, e** $\ln N_d$ versus $\ln$ AI, and **c, f** $\ln N_d$ versus $\ln$ SO₄ for the regions over land (top) and ocean (bottom). Annual averaged slope here is the average of the monthly slopes. The standard deviation of the inter-monthly variability of the regression slopes is shown as an error bar.



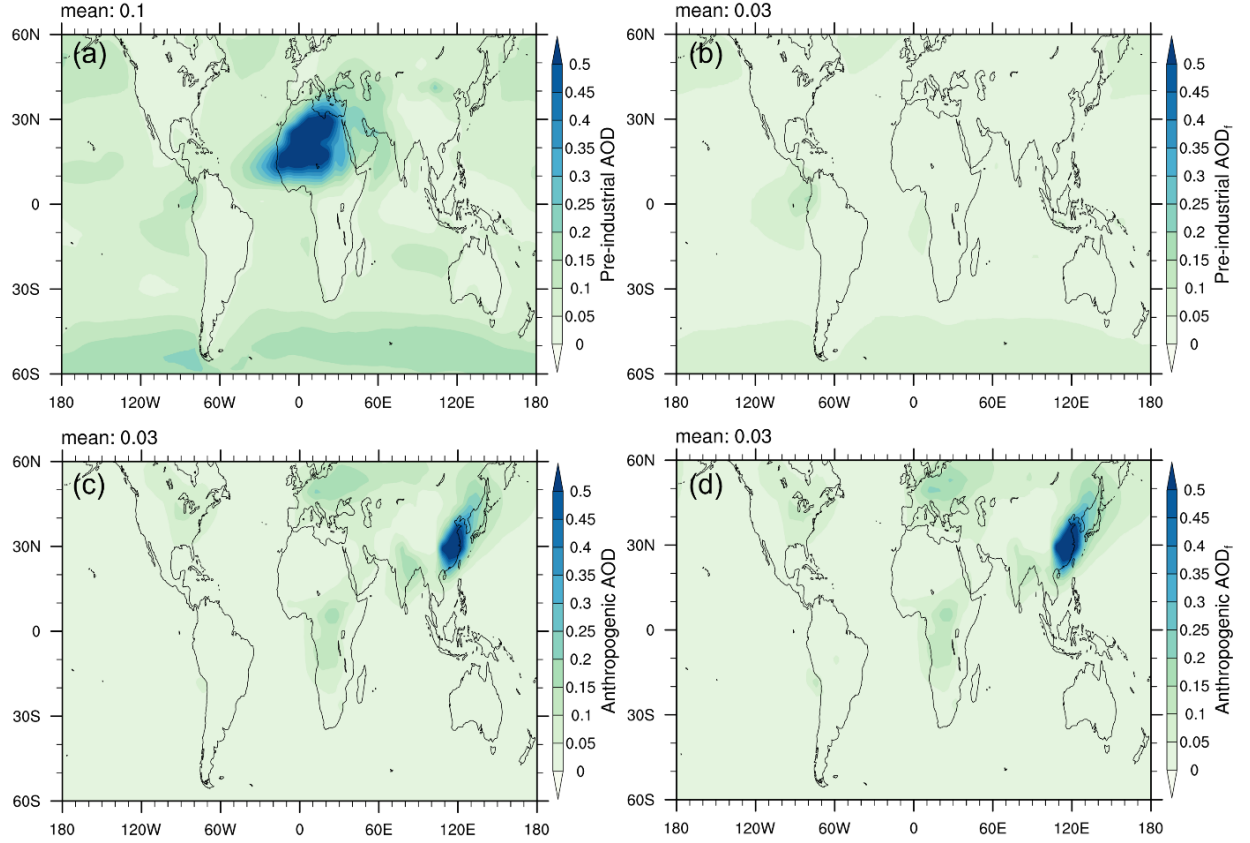
Supplementary Figure 3. Comparison of MERRA-2 and MODIS AOD for the year of 2010. **a** and **b** are for clear-sky and cloudy conditions, respectively. Color is coded by joint probability distribution function (PDF). The black line indicates the 1:1 line, and the gray dashed ones represent the lines of $y = 2x$ and $y = 0.5x$, respectively.



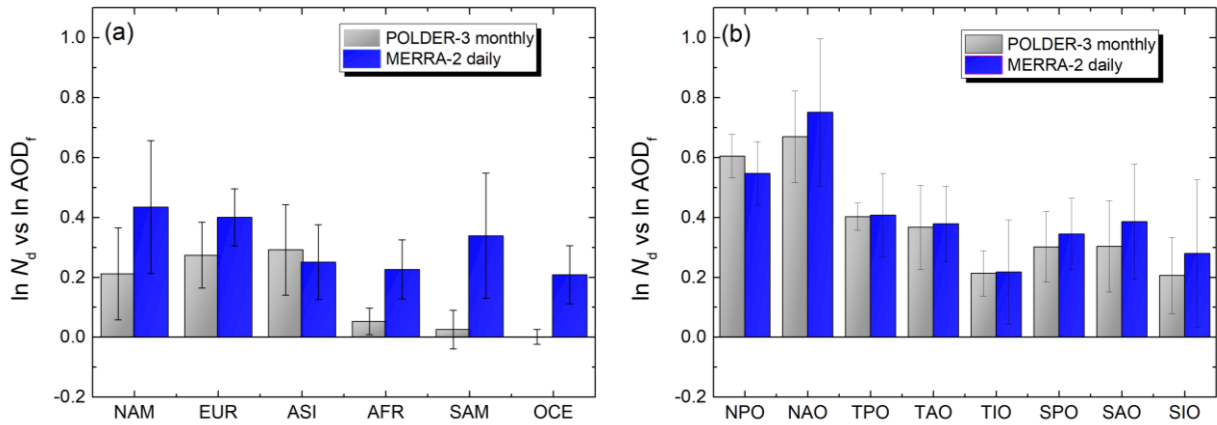
Supplementary Figure 4. The difference between MODIS AOD and MERRA-2 AOD as function of cloud fraction (f). a-n are for the fourteen regions as defined in Supplementary Figure 1.



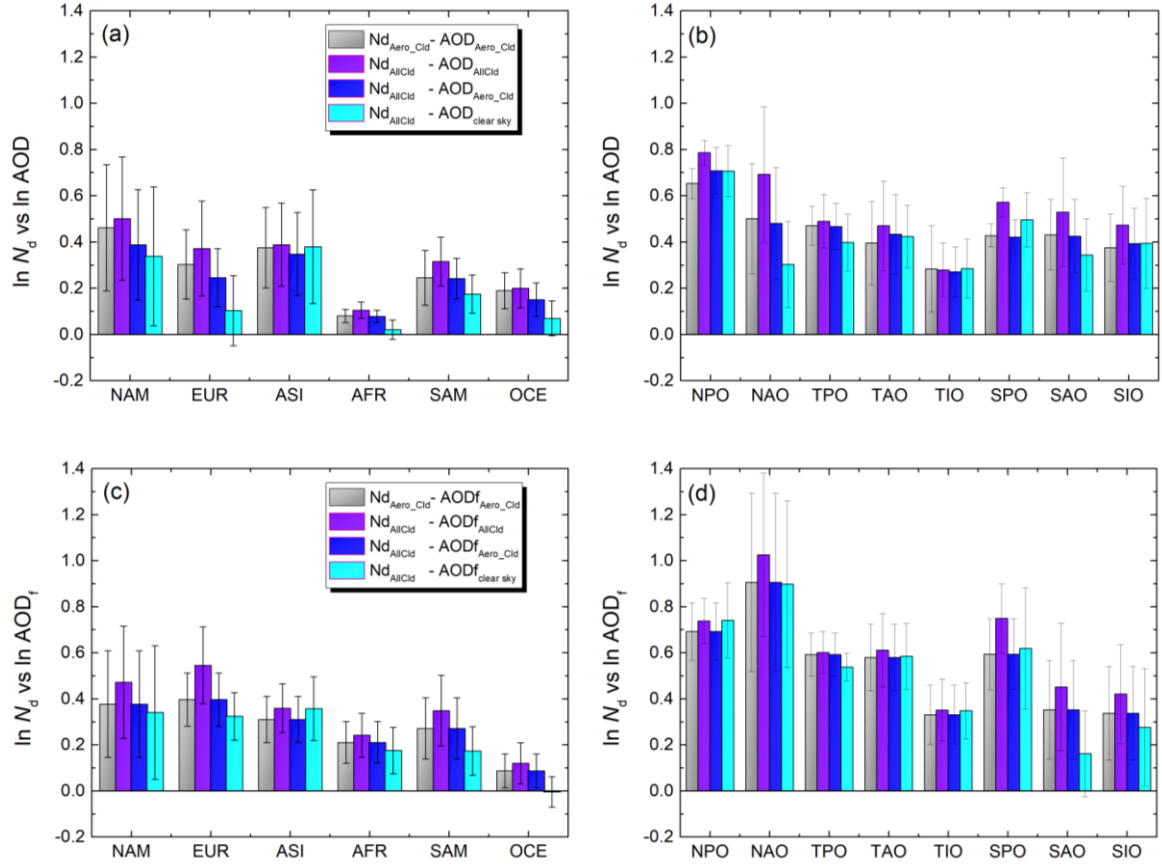
Supplementary Figure 5. Annual mean first indirect forcing (RF_{aci}) for three different scenarios. a The scenario that MODIS aerosol and cloud retrievals are simultaneously successful (Aero_Cld), **b** same cloud samples as Aero_Cld but using the regression coefficients for the scenario including all ambient clouds (Aero_Cld_R), which is designed to evaluate the effect of changed regression coefficients, and **c** using same regression coefficients as Aero_Cld but including all ambient clouds when computing RF_{aci} (Aero_Cld_C), which is designed to quantify the impact of changed cloud fraction. Aerosol optical depth used in all cases are from MERRA-2 reanalysis data.



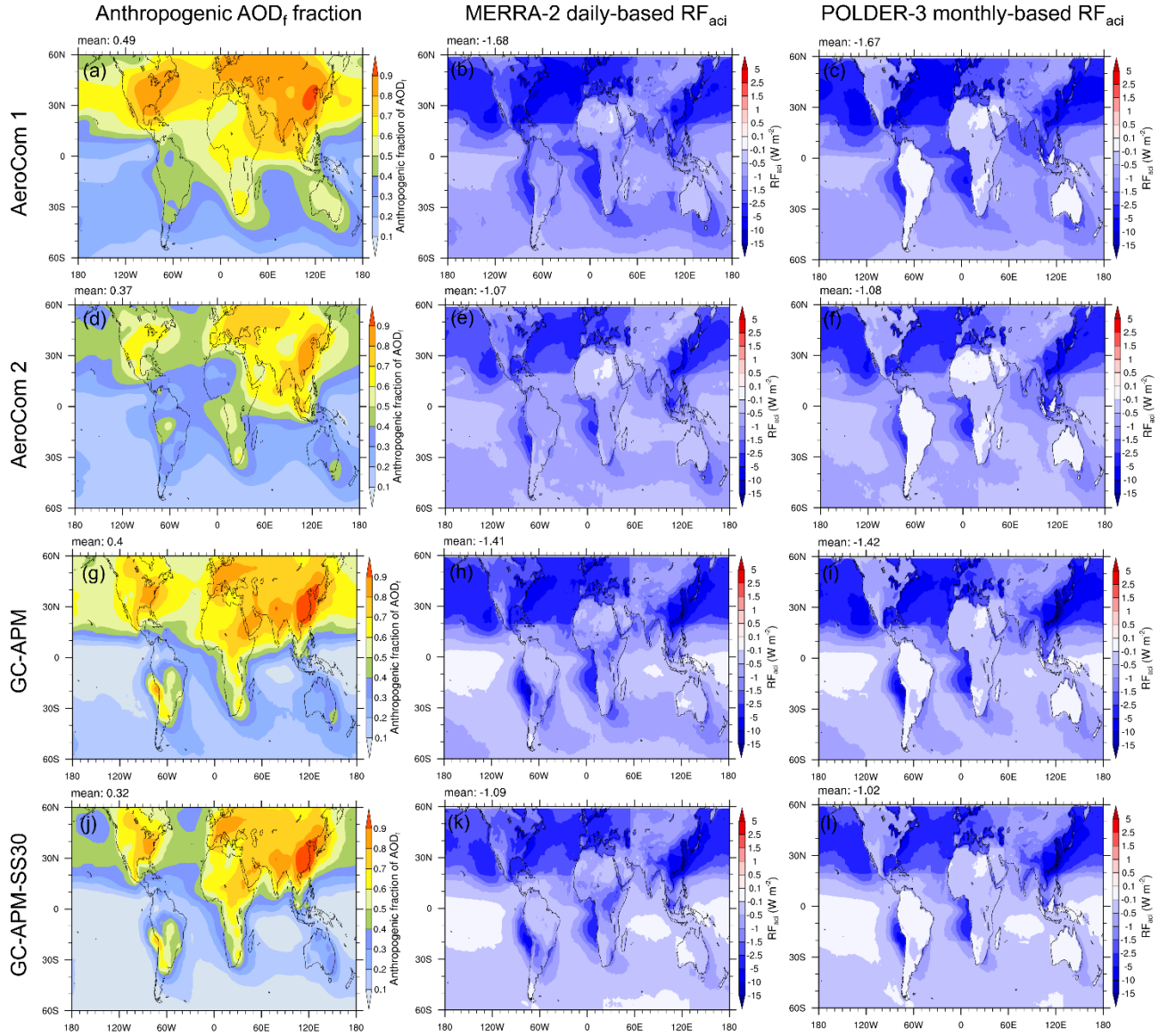
Supplementary Figure 6. Annual (2010) averaged pre-industrial and anthropogenic aerosol optical depth (AOD) and fine mode AOD (AOD_f). Shown are pre-industrial **a** AOD and **b** AOD_f , and **c** anthropogenic AOD and **d** AOD_f from the GEOS-Chem-APM simulations.



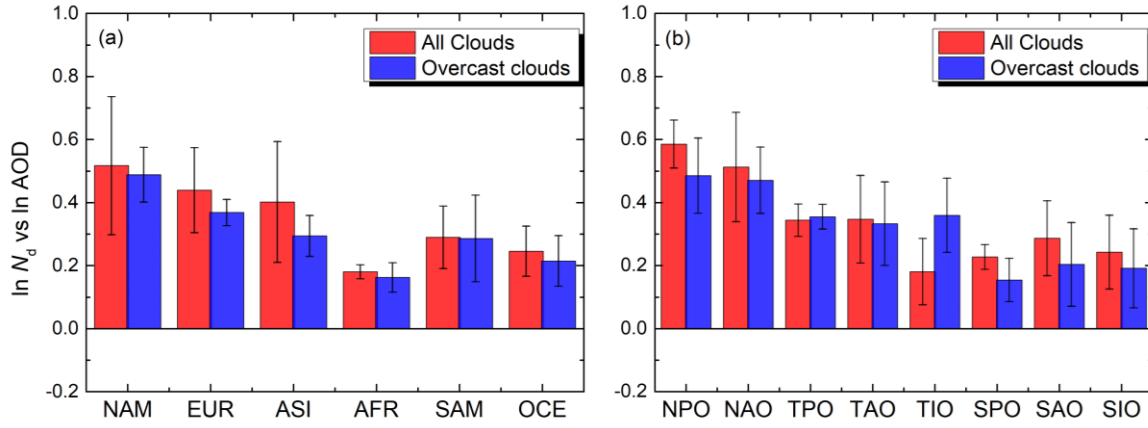
Supplementary Figure 7. Annually averaged slopes of the linear regressions between the logarithm of cloud droplet number concentration (N_d) and that of fine mode aerosol optical depth (AOD_f) for the fourteen regions. Grey and blue bars represent the slopes based on POLDER-3 monthly statistics and MERRA-2 daily (for the scenario including all ambient clouds, i.e., All_Cld) statistics over **a** continental and **b** oceanic regions, respectively. Both cases can sidestep the influence of sampling biases. The annually averaged slope here is the average of the monthly slopes. The standard deviation of the inter-monthly variability of the regression slopes is shown as an error bar.



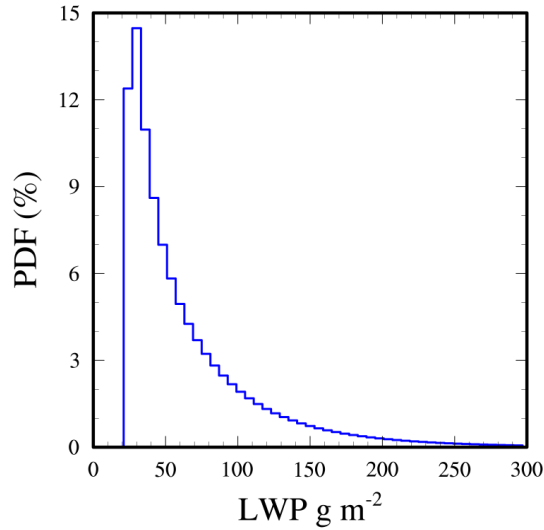
Supplementary Figure 8. Annually averaged slopes of the linear regressions between the logarithm of cloud droplet number concentration (N_d) and those of aerosol optical depth (AOD) and fine mode AOD (AOD_f) based on monthly aerosol-cloud associations. Shown are **a, b** $\ln N_d$ versus $\ln AOD$ and **c, d** $\ln N_d$ versus AOD_f for the fourteen regions. Here, AOD (AOD_f) is from MERRA-2 re-analysis data. Monthly N_d and AOD (AOD_f) statistics are subsampled from daily data according to four strategies as indicated by different subscripts (AllCld, AeroCld, clear sky). $N_{dAllCld}$ ($AOD_{AllCld} / AOD_{fAllCld}$) represents the monthly N_d (AOD / AOD_f) calculated from the data including all clouds. $N_{dAeroCld}$ ($AOD_{AeroCld} / AOD_{fAeroCld}$) represents the monthly N_d (AOD / AOD_f) calculated from the subset of data where aerosol and cloud retrievals are available simultaneously. Here the ‘clear sky’ case includes not only the AOD (AOD_f) with $f = 0$ but also the AOD (AOD_f) without successful N_d retrieval over entire $1^\circ \times 1^\circ$ grid box, which is used to represent the monthly AOD (AOD_f) that is irrelevant to monthly $N_{dAllCld}$. The annually averaged slope is the average of the monthly slopes. The standard deviation of the inter-monthly variability of the regression slopes is shown as an error bar.



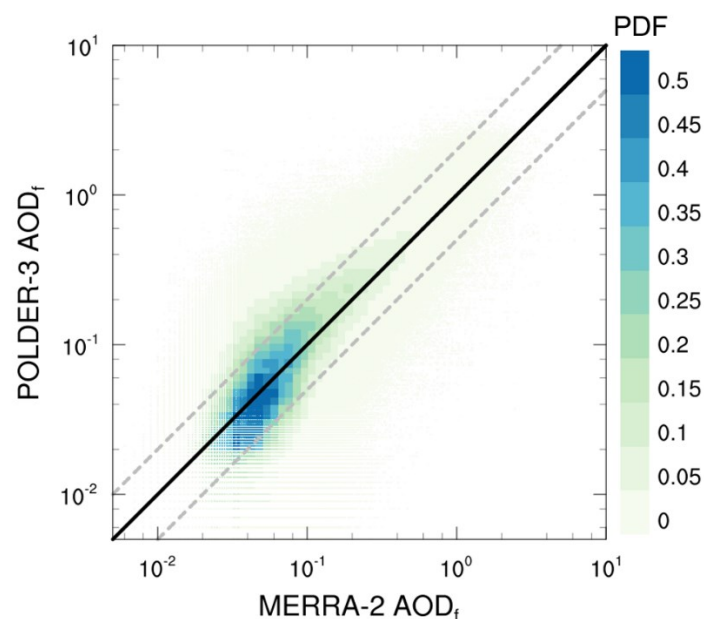
Supplementary Figure 9. Annual mean anthropogenic fractions and their associated first indirect forcing (RF_{aci}) based on daily and monthly aerosol-cloud associations, respectively. Shown on the left column are anthropogenic fractions of fine mode aerosol optical depth (AOD_f) acquired from the simulations of **a** AeroCom phase1 models with the preindustrial year 1750 as reference (AeroCom 1), **d** AeroCom phase2 models with the preindustrial year 1850 as reference (AeroCom 2), **g** GEOS-Chem-APM with AOD_f defined as the sum of AODs of sulfate, black carbon and organic aerosol (GC-APM), and **j** GEOS-Chem-APM with AOD_f defined as the sum of AODs sulfate, black carbon and organic aerosol and 30 % sea salt aerosol (GC-APM-SS30), respectively. **b, e, h, k** MERRA-2 daily-based and **c, f, i, l** POLDER-3 monthly-based RF_{aci} estimated with above-mentioned four anthropogenic AOD_f fractions are shown on the middle and right columns, respectively.



Supplementary Figure 10. Annually averaged slopes of the linear regressions between the logarithm of cloud droplet number concentration (N_d) and that of MERR-2 aerosol optical depth (AOD) for the fourteen regions. Red and blue bars represent the results based on all cloud samples and overcast clouds only, respectively. Here, the overcast pixel is defined as a pixel with $20 \times 20 \text{ km}^2$ resolved cloud fraction larger than 80 %. The annually averaged slope here is the average of the monthly slopes. The standard deviation of the inter-monthly variability of the regression slopes is shown as an error bar.



Supplementary Figure 11. Global probability density function of liquid water path (LWP) over the period 2002 – 2018.



Supplementary Figure 12. Comparison of MERRA-2 and POLDER-3 AOD_t for the year of 2010. Color is coded by the joint probability distribution function (PDF). The black line indicates the 1:1 line, and the gray dashed ones represent the lines of $y = 2x$ and $y = 0.5x$, respectively.

Supplementary References

1. Wielicki, B. A. et al. Clouds and the Earth's radiant energy system (CERES): An Earth observing system experiment. *B. Am. Meteorol. Soc.* **77**, 853–868 (1996).
2. Levy, R.C. et al. The Collection 6 MODIS aerosol products over land and ocean. *Atmos. Meas. Tech.* **6**, 2989–3034 (2013).
3. Dubovik, O. et al. GRASP: a versatile algorithm for characterizing the atmosphere. *SPIE Newsroom*, **25** (2014).
4. Randles, C. A. et al. The MERRA-2 aerosol reanalysis, 1980 onward. Part I: System description and data assimilation evaluation. *J. Climate*. **30**, 6823–6850 (2017).
5. Kinne, S. The MACv2 aerosol climatology. *Tellus B* **71**, 1–21 (2019).
6. Kinne, S. et al. An AeroCom initial assessment optical properties in aerosol component modules of global models. *Atmos. Chem. Phys.* **6**, 1815–1834 (2006).
7. Myhre, G. et al. Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations. *Atmos. Chem. Phys.* **13**, 1853–1877 (2013).
8. Yu, F. & Luo, G. Simulation of particle size distribution with a global aerosol model: Contribution of nucleation to aerosol and CCN number concentrations. *Atmos. Chem. Phys.* **9**, 7691–7710 (2009).