Supporting Information for

Anthropogenic contribution to cloud condensation nuclei and the first aerosol indirect climate effect

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1. Rapid Radiative Transfer Model for GCMs (RRTMG)

RRTMG is a broadband k-distribution radiation model [1-3] that has been widely used in community models (such as WRF, CAM5, etc.). The RRTMG for shortwave (SW) (used in this study) can calculate fluxes and heating rates over 14 contiguous shortwave bands (820-50000 cm\(^{-1}\), or 0.2-12.20 microns). The individual band ranges (in wavenumbers, cm\(^{-1}\)) are: 2600-3250, 3250-4000, 4000-4650, 4650-5150, 5150-6150, 6150-7700, 7700-8050, 8050-12850, 12850-16000, 16000-22650, 22650-29000, 29000-38000, 38000-50000, and 820-2600, with the last band coded out of sequence to preserve spectral continuity with the longwave bands. The optical properties of water clouds are calculated for each spectral band based on the scheme of Hu and Stamnes [4], where the optical depth, single-scattering albedo, and asymmetry parameter are parameterized as a function of cloud equivalent radius and LWC. The optical properties of ice clouds are calculated for each spectral band from the Fu scheme [5], which assumes the ice crystals are hexagonal and randomly-oriented in space and parameterizes the optical depth, single-scattering albedo, and asymmetry parameter as a function of the generalized effective size.
of the ice crystals and the ice water content (IWC). Detailed information of RRTMG can be found in the model webpage (http://rtweb.aer.com/rrtm_frame.html).

2. GEOS-Chem/APM

The GEOS-Chem model has been developed and used by many research groups around the world and contains a number of state-of-the-art modules treating various chemical and aerosol processes with up-to-date key emission inventories [6-23]. The model is driven by assimilated meteorology from the NASA Goddard Earth Observing System 5 (GEOS-5). Details of the model can be found at http://acmg.seas.harvard.edu/geos/index.html.

The computationally efficient APM model incorporated into GEOS-Chem by Yu and Luo [16] substantially enhances the capability of GEOS-Chem in predicting size-resolved particle properties. The model is optimized to accurately simulate secondary particle (SP, composed of sulfate, nitrate, ammonium, and SOA) formation and their growth to CCN sizes, with a higher size resolution for the size range of importance (1.2 – 120 nm: 30 bins, 10 additional bins for 120 nm -12 μm). Nucleation rates are predicted by the ion-mediated nucleation mechanism [24] which is supported by the state-of-the-art multi-instrument field measurements taken in a boreal forest [25] and gives overall good agreement with both absolute values and spatial distributions of particle number concentrations derived from land-, ship-, and aircraft- based measurements [26]. The condensation of low volatile secondary organic gases from successive oxidation [27] is taken into account. The present version of the APM employs 20 bins for sea salt to cover the dry diameter size range of 0.012 μm to 12 μm, and 15 bins for dust particles to cover size range of 0.03 μm to 50 μm. Because of the large differences in the median sizes of black carbon (BC) and primary organic carbon (POC) from fossil fuel combustion and biomass burning, we employ two
log-normal modes (one for fossil fuel and another for biomass burning) to represent hydrophobic BC and two other log-normal modes for hydrophilic BC. Similarly, 4 log-normal modes are used to represent hydrophobic and hydrophilic POC.

Recent implementations of the aerosol optical properties look up table [28] and radiation transfer (RF) model of the Canadian Center for Climate Modeling and Analysis (CCCma) [29] enable us to derive aerosol direct radiative forcing (DRF). A more recent comparison of DRF values (clear sky vs. all sky) based on GEOS-Chem/APM with those of other AeroCom models (discussion version of Myhre et al. [30]) indicates that CCCMa RF code may have underestimated the impacts of clouds on radiation. We found out that the underestimation is likely associated with the cloud overlapping assumption. The version of CCCMa RF code we integrated into GEOS-Chem does not contain the widely used McICA (Monte-Carlo Independent Column Approximation) scheme — a fast, flexible, approximate technique for computing radiative transfer in an inhomogeneous cloud field. For this study, we have incorporated RRTMG-SW, which contains the McICA scheme, into GEOS-Chem/APM.

GEOS-Chem/APM-RRTMG results have been included into the revised manuscript on radiative forcing of the direct aerosol effect from AeroCom Phase II simulations [30] and another revised manuscript on host model uncertainties in aerosol radiative forcing estimates [31]. GEOS-Chem/APM is also one of 12 global aerosol models participating in the AeroCom Phase II aerosol microphysical properties Intercomparison and evaluation. GEOS-Chem/APM performs well in these intercomparisons.

3. Precursor concentrations, nucleation rates, and particle number concentrations under pre-industry (PI) emission scenario.
Figure S1. Horizontal distributions of annual mean SO$_2$, H$_2$SO$_4$, J, CN10, CCN0.4, and CDN values in the lower troposphere under PI emission scenario.

Figure S1 shows the horizontal distributions of annual mean SO$_2$, H$_2$SO$_4$, J, CN10, CCN0.4, and CDN values in the lower troposphere under the PI emission scenario. The hot spots in Figures S1a and S1b are associated with non-eruptive volcanic emissions. [H$_2$SO$_4$] and
temperature are the two most important parameters for determining the new particle formation rate \( (J) \) [24]. While \([\text{H}_2\text{SO}_4]\) is high in the tropical LT, \( J \) is generally very small (Fig. S1c) due to the high temperatures are present. High \( J \) in the LT is generally confined to mid-latitude zones in both hemispheres, where \([\text{H}_2\text{SO}_4]\) is relatively high and temperature is relatively low. It should be noted that while \( J \) is small in the tropical LT, it is higher in the tropical upper troposphere [26]. Downward transport of particles formed above is a major source of particles in the tropical LT, consistent with observations [32]. In the LT under the PI emission scenario, CN10 ranges from \( \sim 150 \) – \( 2000 \) cm\(^{-3}\), CCN0.4 from \( \sim 30 \) – \( 500 \) cm\(^{-3}\), and CDN from \( \sim 70 \) – \( 500 \) cm\(^{-3}\).

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