ENVIRONMENTAL RESEARCH LETTERS

LETTER • OPEN ACCESS

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

To cite this article: Fangqun Yu et al 2013 Environ. Res. Lett. 8 024029

View the article online for updates and enhancements.

You may also like

- The Influence of Cloud Condensation Nucleus Coagulation on the Venus Cloud Structure

Kevin McGouldrick and Erika L. Barth

- Effect of solar variations on particle formation and cloud condensation nuclei Fanggun Yu and Gan Luo
- <u>Connecting hygroscopic growth at high</u> <u>humidities to cloud activation for different</u> <u>particle types</u>

H Wex, F Stratmann, T Hennig et al.



This content was downloaded from IP address 3.142.197.198 on 03/05/2024 at 19:27

Environ. Res. Lett. 8 (2013) 024029 (8pp)

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

Fangqun Yu, Xiaoyan Ma and Gan Luo

Atmospheric Sciences Research Center, State University of New York at Albany, 251 Fuller Road, Albany, NY 12203, USA

E-mail: fyu@albany.edu

Received 19 March 2013 Accepted for publication 9 May 2013 Published 29 May 2013 Online at stacks.iop.org/ERL/8/024029

Abstract

Atmospheric particles influence the climate indirectly by acting as cloud condensation nuclei (CCN). The first aerosol indirect radiative forcing (FAIRF) constitutes the largest uncertainty among the radiative forcings quantified by the latest IPCC report (IPCC2007) and is a major source of uncertainty in predicting climate change. Here, we investigate the anthropogenic contribution to CCN and associated FAIRF using a state-of-the-art global chemical transport and aerosol model (GEOS-Chem/APM) that contains a number of advanced features (including sectional particle microphysics, online comprehensive chemistry, consideration of all major aerosol species, online aerosol-cloud-radiation calculation, and usage of more accurate assimilated meteorology). The model captures the absolute values and spatial distributions of CCN concentrations measured in situ around the globe. We show that anthropogenic emissions increase the global mean CCN in the lower troposphere by \sim 60–80% and cloud droplet number concentration by \sim 40%. The global mean FAIRF based on GEOS-Chem/APM is -0.75 W m⁻², close to the median values of both IPCC2007 and post-IPCC2007 studies. To the best of our knowledge, this is the first time that a global sectional aerosol model with full online chemistry and considering all major aerosol species (including nitrate, ammonium, and second organic aerosols) has been used used to calculate FAIRF.

Keywords: cloud condensation nuclei, aerosol indirect radiative forcing, global aerosol model, climate change, IPCC

S Online supplementary data available from stacks.iop.org/ERL/8/024029/mmedia

1. Introduction

Cloud droplets form onto pre-existing atmospheric particles or cloud condensation nuclei (CCN). For a given meteorological condition, an increase in CCN associated with anthropogenic emissions leads to an increase in both the number of cloud

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI. droplets and the scattering of solar radiation [1]. This is known as the Twomey effect or the first aerosol indirect radiative forcing (FAIRF). According to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC) published in 2007 [2] (named thereafter IPCC2007), the global annual mean FAIRF is -0.7 W m^{-2} with a 5–95% confidence range from $-1.8 \text{ to} -0.3 \text{ W m}^{-2}$ and a low level of scientific understanding. The spread among model-calculated FAIRF values constitutes the largest uncertainty among the quantified radiative forcings [2]. FAIRF values based on post-IPCC2007 studies still vary significantly [3–10]. There also exist large differences between model-derived and observation-constrained FAIRF values and the exact source of the difference is still under debate [6, 11–13]. In order to confidently interpret past and project future climate change, the reduction of FAIRF uncertainty or diversity is essential.

As pointed out in IPCC2007, a major source of FAIRF uncertainty is poor knowledge of the number concentration and spatial distribution of pre-industrial (PI) and present-day (PD) aerosols. Most (if not all) IPCC2007 and post-IPCC2007 FAIRF studies are based on global models with simplified chemistry and aerosol microphysics. For example, most of these studies simulated the sulfur cycle chemistry only and used off-line monthly mean concentrations of key oxidants. The aerosol size distributions, which are important for CCN calculation and aerosol indirect forcing, are either prescribed or predicted with a simplified modal approach. Among the 20 previous FAIRF studies listed in the table 2.7 of IPCC2007, at least 15 of them employed the prescribed (or bulk) aerosol scheme that assumes globally uniform size distributions. Bellouin et al [8] showed substantial differences in the aerosol forcings simulated by the bulk and modal schemes, and they pointed out that the bulk approach lacked the necessary sophistication to provide realistic aerosol input for aerosol-cloud-radiation calculations. More post-IPCC2007 FAIRF studies are based on the modal scheme which employs one or more log-normal modes to represent aerosols of various types. While the modal approach is more sophisticated compared to the bulk scheme, the assumption of fixed mode widths (or standard deviations), ranges and size distribution shapes can result in large biases in simulated particle size distributions and CCN concentrations [14–16]. Global size-resolved sectional particle microphysics models, aiming to simulate more accurately the particle size distributions, have been developed [15-20]. A comparison of modal and sectional aerosol schemes within the same global models showed that the sectional approach better reproduces observed size distributions at CCN sizes, and the parameters in the modal scheme need to be adjusted to achieve better agreement with the observations [15, 16]. Nevertheless, due to the computational cost, most FAIRF studies are still based on the bulk or modal aerosol scheme.

Yu and Luo [20] integrated a computationally efficient advanced particle microphysics (APM) model into a global chemical transport model (CTM) GEOS-Chem [21], allowing one to explicitly and more accurately simulate global size-resolved sectional particle microphysics at an affordable computational cost. The GEOS-Chem model, driven by assimilated GEOS-5 meteorology, contains a number of state-of-the-art modules treating comprehensive chemical and aerosol processes [21–29]. Here we expand the capability of GEOS-Chem/APM by (1) coupling it with the widely used rapid radiative transfer (RT) model for GCMs (RRTMG) [30] and (2) relating the APM-predicted particle size distributions to the cloud droplet number concentrations and therefore, the droplet radius and cloud albedo [31]. The resulting model is then employed to study the contribution of anthropogenic emissions to CCN and the first aerosol indirect climate effect. The GEOS-Chem/APM-RRTMG contains a number

of advanced features that address some limitations of the aforementioned previous FAIRF studies, which should help to reduce the uncertainty of FAIRF and improve its level of scientific understanding.

2. Models and simulations

To study aerosol impacts on cloud albedo and solar radiation, we have fully integrated RRTMG for shortwave (SW) into GEOS-Chem/APM (see supporting information, available at stacks.iop.org/ERL/8/024029/mmedia, for details of the two models). The GEOS-Chem/APM model explicitly simulate the formation and their growth of secondary particle (composed of sulfate, nitrate, ammonium, and secondary organic aerosol) as well as the coating of secondary species on primary particles (black carbon, primary organic carbon, dust, and sea salt particles) [20]. RRTMG-SW is called as a column model for online RT calculation at every chemistry time step. The variables inputted to RRTMG include solar zenith angle, surface temperature and albedo, along with the following profiles: (1) pressure, (2) temperature, (3) mixing ratios of CO, O_3 and water vapor, (4) aerosol extinction coefficient, single scattering albedo and asymmetry parameter at fourteen shortwave bands, (5) cloud cover and water content, and (6) effective radius of liquid cloud droplets. Default mixing ratios are assumed for CO₂, N₂O, CH₄, and O₂. The effects of atmospheric particles on ice formation are currently insufficiently understood and, similar to IPCC2007 studies, not considered in our FAIRF calculations. Items 1, 2, 5, and surface temperature are from GEOS-5 assimilated meteorology. Items 3 and solar zenith angle are from GEOS-Chem. Item 4 is calculated from APM-predicted aerosol properties [32]. The surface albedo dataset is taken from MODIS satellite retrievals [33]. The calculation of cloud droplet effective radius (item 6), key to FAIRF, is described below.

Effective radius of cloud droplets depends on water content and the number concentrations of particles activated as CCN. Besides particle size distributions and compositions, cloud scale vertical velocity is also important for aerosol activation. The large scale vertical velocity provided by assimilated GEOS-5 meteorology does not resolve sub-grid variations that determine cloud droplet nucleation [34]. In our study, the sub-grid scale variability of vertical velocity is prescribed by a normal probability distribution function (PDF) [35], with the large scale vertical velocity as its mean value and a standard deviation of 0.5 m s^{-1} which was derived from measured updrafts in stratiform clouds [36]. The PDF approach to approximate the sub-grid scale variability has been employed in several previous studies [37, 38], whose results have been used in IPCC2007. The cloud droplet number (CDN) concentration is calculated from the APM-predicted aerosol size distributions using a Köhlerbased activation parameterization. This parameterization combines the treatment of multiple aerosol types and a sectional representation of size to deal with arbitrary aerosol mixing states and size distributions [31]. This mechanistic parameterization relating the aerosol properties and CDN takes advantage of the detailed size, composition, and mixing state resolved particle properties predicted by the GEOS-Chem/APM. The CDN concentration is then used to calculate the cloud droplet effective radius needed by RRTMG with the formula given in [39]. It should be noted that different treatments of aerosol–cloud interaction processes can cause substantial variations in the derived FAIRF values [38, 39], which is out of the scope of the present study.

IPCC has defined the FAIRF as the influence of increased aerosol concentration (due to anthropogenic emissions) on the cloud optical properties and top of the atmosphere solar fluxes while holding the cloud fraction and liquid water content (i.e., in an entirely diagnostic manner where feedback mechanisms do not occur). GEOS-Chem, as a CTM driven by assimilated meteorology, is perfectly suitable for FAIRF calculation. To obtain the IPCC-defined FAIRF, we ran two simulations (10/2005–12/2006, first three months as spin up) driven by the same assimilated GEOS-5 meteorology fields: one with the present-day (PD) emissions (corresponding to year 2006) and the other with the pre-industrial (PI) emissions as specified in AeroCom II [40]. Eruptive volcano emission is turned off in both simulations. The annual mean results presented below are based on v8-03-02 of GEOS-Chem with APM and RRTMG integrated. The horizontal resolution is $2^{\circ} \times 2.5^{\circ}$ and there are 47 vertical layers in the model (surface to 0.01 hpa). The time step is 10 min for transport, and 30 min for chemistry, size-resolved aerosol microphysics, and RT calculation.

3. Results

Table 1 summarizes the globally and annually averaged SO_2 and H_2SO_4 vapor concentrations ([SO_2] and [H_2SO_4]), nucleation rate (J), concentrations of condensation nuclei larger than 10 nm (CN10), CCN at water supersaturation of 0.8% (CCN0.8), 0.4% (CCN0.4), and 0.2% (CCN0.2), and cloud droplet number (CDN) in the lower troposphere (averaged within lowest 18 model layers, surface to \sim 3 km) under both pre-industry (PI) and present-day (PD) emissions, as well as the corresponding changes from PI to PD. The global mean values of key variables related to aerosols and FAIRF for both PI and PD cases, as well as the PI-PD changes showing the impacts of anthropogenic emissions (including various aerosol precursors and primary carbonaceous particles). To account for the differences in various altitudes and focus on the lower troposphere (LT), where warm clouds form, we average all the 3D variables in the lowest 18 model layers (from surface to \sim 3 km) to simplify the comparisons. Figure 1 shows the horizontal distributions of [SO₂], [H₂SO₄], J, CN10, CCN0.4, and CDN changes from PI to PD (the corresponding absolute values for the PI case are given in figure S1 in the supporting information, available at stacks.iop.org/ERL/8/ 024029/mmedia). CN, CCN, and CDN include contributions from both primary and secondary particles. We use the Köhler theory to calculate CCN at given supersaturation ratios from the APM-predicted aerosol size distributions. Thus CCN concentrations depend on both sizes and compositions. It is

Table 1. Globally and annually averaged SO₂ and H₂SO₄ vapor concentrations ([SO₂] and [H₂SO₄]), nucleation rate (J), concentrations of condensation nuclei larger than 10 nm (CN10), CCN at water supersaturation of 0.8% (CCN0.8), 0.4% (CCN0.4), and 0.2% (CCN0.2), and cloud droplet number (CDN) in the lower troposphere (averaged within lowest 18 model layers, surface to \sim 3 km) under both pre-industry (PI) and present-day (PD) emissions, as well as the corresponding changes from PI to PD.

	PI	PD	$PI \rightarrow PD$ change (%)
[SO ₂] (ppt)	66.4	256.0	285
$[H_2SO_4]$ (ppt)	0.077	0.123	60
$J (\# cm^{-3} s^{-1})$	0.009	0.021	118
$CN10 \ (\# \ cm^{-3})$	385.4	569.4	45
$CCN0.8 \ (\# \ cm^{-3})$	197.6	323.8	61
$CCN0.4 \ (\# \ cm^{-3})$	128.4	224.5	75
$CCN0.2 \ (\# \ cm^{-3})$	86.7	158.2	82
$CDN \ (\# \ cm^{-3})$	159.9	228.0	41

clear from table 1 and figure 1. that anthropogenic emissions have a significant effect on globally averaged [SO₂], [H₂SO₄], J, concentrations of aerosols, CCN and CDN in the LT.

H₂SO₄ vapor, produced via oxidation of SO₂ from both anthropogenic and natural (dimethyl sulfide, volcano, etc) sources, is known to play an important role in forming and growing new particles that dominate particle number concentrations in most parts of the globe [20]. Without anthropogenic emissions (i.e., PI case), global mean LT [SO₂] and $[H_2SO_4]$ are respectively 66.4 ppt and 0.077 ppt (table 1). Anthropogenic emissions significantly increase [SO₂] over major continents, by a factor of 3-30 over large areas and above 100 over parts of China and India (figure 1(a)). It is interesting to note that [SO₂] slightly decreases over remote oceans in the southern hemisphere (SH) and over Antarctica, likely as a result of changes in atmospheric oxidation capacity and chemistry associated with anthropogenic emissions. Compared to the global mean LT [SO₂] increase of 285%, global mean LT [H₂SO₄] enhancement is only \sim 60%, with most of enhancement confined to anthropogenic source regions (figure 1(b)). In addition to the decrease of $[H_2SO_4]$ over SH high latitude regions as a result of lower [SO₂], [H₂SO₄] also decreases over the high latitude regions in the northern hemisphere (NH). This decrease, away from source regions, and the relatively weaker enhancement of [H₂SO₄] over source regions are largely a result of the condensation sink increase associated with anthropogenic emissions. The spatial distribution of J changes in the LT due to anthropogenic emissions (figure 1(c)) is similar to that of $[H_2SO_4]$ (figure 1(b)), which is expected as both PI and PD cases use the same meteorology fields. Anthropogenic emissions enhance the global mean J in the LT by 118%, with most of the increase over major SO₂ source regions (figure 1(c)). J decreases substantially in the middle and high latitude regions away from anthropogenic SO₂ sources, which is caused mainly by the decrease of $[H_2SO_4]$ (figure 1(b)) and increase of total particle surface areas in these regions.

In the atmosphere, the fraction of particles that can act as CCN and produce cloud droplets depends on particle number size distribution and the water supersaturation ratio



Figure 1. Impacts of anthropogenic emissions on annual mean SO_2 , H_2SO_4 , J, CN10, CCN0.4, and CDN in the low troposphere (averaged within lowest 18 model layers, surface to ~ 3 km). PD/PI ratios are given for all parameters except J as PD/PI ratios for J can be very big in the tropical region due to negligible J values in the region for both cases. Letters and numbers marked on panel (e) are the locations of sites where surface-based CCN0.4 measurements (for periods lasting a few weeks to more than a year) used for comparison in figure 2.

attained in clouds (up to ~0.8% for convective clouds and ~0.2% for stratus clouds). The main factors controlling the particle number size distribution include emission, nucleation, growth, coagulation, wet scavenging and transport. As we can see from table 1, anthropogenic emissions increase global mean LT CN10, CCN0.8, CCN0.4, CCN0.2, and CDN by 45%, 61%, 75%, 82%, and 41%, respectively. The relative increase of CDN is smaller (compared to those of CCNs) is a result of a larger fraction of CCN getting activated under the clean PI emission scenario. Most of the increases are over major continents and immediate outflow regions, with CN10, CCN0.4, and CDN enhanced by a factor of ~3–10 near where major anthropogenic emissions are located (figure 1). CN10 decreases substantially in the regions far away from

anthropogenic emissions (figure 1(d)) due to the reduction in the nucleation rates (figure 1(c)). In contrast, CCN0.4 and CDN increase in most parts of the remote regions except over some tropical areas. The difference is due to transport as well as the dynamic interactions among particles and precursors.

The accuracy of CCN simulations is critical for robust FAIRF estimations. Figure 2 compares the simulated CCN0.4 concentration in the lower boundary layer ([CCN0.4]_{LBL}) with observed values at 29 surface sites around the globe (locations are marked on figure 1(e) by the letters and numbers). It should be noted that the predicted [CCN0.4]_{LBL} for the PI case (letters and numbers in red) cannot be directly compared to PD observations, but they are given in figure 2 to illustrate the effect of anthropogenic emissions. It is clear



Figure 2. Comparison of simulated CCN0.4 concentration in the lower boundary layer ([CCN0.4]_{LBL}) for the PD case (letters and numbers in black) with observed values at 29 surface sites marked on figure 1(e). The CCN0.4 data include (1) those compiled by Andreae [41], (2) additional data points from recent publications as well as archival observations as described in Yu *et al* [42], and (3) three new data points (sites 1–3) from Ward and Cotton [43], Irwin *et al* [44], and Laakso *et al* [45]. To illustrate the effect of anthropogenic emissions, corresponding [CCN0.4]_{LBL} for the PI case are also given (letters and numbers in red). Model values correspond to the months when the observations were made.

from figure 2 that the GEOS-Chem/APM simulations for the PD case reproduce the absolute values as well as the horizontal variations of observed CCN0.4, with an overall correlation coefficient of 0.93 and a normalized mean bias (NMB) of 1.1%. The good agreement indicates that the model might have captured the key chemical and physical processes controlling global CCN abundance and distributions. This gives us more confidence on the robustness of predicted CCN for the PI case. As we can see from figure 2 (also figure 1(e), noting one for LBL and the other for LT), anthropogenic emissions significantly increase [CCN0.4]_{LBL} at the sites near the SO₂ source regions, up to a factor of 10. The enhancement is relatively small over remote regions where natural sources contribute significantly to CCN abundance.

Through the first aerosol indirect effects (1), the changes of CCN and CDN concentrations modify the cloud droplet effective radius, albedo, and radiative forcing. Figure 3 gives the horizontal distribution of FAIRF, calculated as the difference at the top of atmosphere SW cloud forcing between PD and PI cases. Anthropogenic aerosols have a negative first indirect effect in almost all the regions around the globe except in limited areas near the equator. The spatial distribution of FAIRF is similar to those of CCN and CDN changes (figures 1(e)–(f)), reaching above -2.5 W m⁻² over the southeastern Pacific Ocean near Chile and Peru and some regions in eastern Asia, Europe, and northeastern America. The global mean FAIRF is -0.75 W m⁻², which is close to the IPCC2007 median value (-0.7 W m⁻²) of about 20 modeling results.



Figure 3. Horizontal distribution of FAIRF, calculated as the difference in shortwave top of atmosphere cloud forcing between PD and PI cases.



Figure 4. A comparison of FAIRF based on GEOS-Chem/APM with those from various studies published after IPCC2007 as well as the summarized values and ranges from Third [46] and Fourth [2] IPCC Assessment Reports.

Figure 4 compares FAIRF based on GEOS-Chem/APM with those from various studies published after IPCC2007 as well as the median values and ranges from the Third and Fourth IPCC Assessment Reports. Table 2 summarizes some key model information for those post-IPCC2007 FAIRF studies that we can find in the literature. Published studies of total aerosol indirect radiative forcing that do not give FAIRF values (e.g., [47]) and those studies focusing on indirect forcing of carbonaceous combustion aerosol only [48] are not included in table 2. For several post-IPCC2007 studies that presented multiple values [4, 5, 10], either baseline case values or averaged values are given. Different FAIRF values based on different approaches, such as different meteorology fields [10], online versus off-line aerosol [9], and bulk versus modal approaches [8], are treated as separate studies because these approaches are quite different and have been used in other FAIRF studies.

Environ. Res. Lett. 8 (2013) 024029

		×				
			Aerosols ^a		FAIRF	
Study	Model type ^b	Chemistry	Size	Species	Approach	Value
Lohmann et al [3]	ECHAM5-HAM	Sulfur cycle, with off-line monthly mean oxidants	Double-moment modal	SO ₄ , OC, BC, SS, D	Online	-0.7
Rotstayn and Liu [4]	CSIRO	Sulfur cycle, with off-line monthly mean oxidants	Bulk	SO ₄ , OC, BC, SS, D	Online	-0.38
Wang and Penner [5]; Penner <i>et al</i> [6]	IMPACT driven by CAM3 met	Sulfur cycle, with off-line monthly mean oxidants	Modal (two modes for sulfate)	SO ₄ , OC, BC, SS, D	Off- line	-1.65 and -1.69
Zhou <i>et al</i> [10] II b II b m	IMPACT driven by CAM5 met	Sulfur cycle, with off-line monthly mean oxidants	Modal (3 modes for sulfate)	SO ₄ , OC, BC, SS, D	Off- line	−1.74 to −1.77
	IMPACT driven by GFDL AM3 met	incan oxidants				-1.26 to -1.44
Bauer and Menon [7]	GISS-modelE	Sulfur cycle, with off-line oxidants and nitric acid concentrations	Modal (2 moment)	SO ₄ , OC, BC, SS, D, NIT, NH ₄	Online	-0.17
Deandreis <i>et al</i> [9] LMDZ cc with INC LMDZ w off-line a	LMDZ coupled with INCA	Sulfur cycle (DMS, SO ₂ , OH, NO ₃ , H ₂ O ₂ ,O ₃)	Bulk	SO_4	Online	-0.36
	LMDZ with off-line aerosols	seneme			Off- line	-0.39
Bellouin <i>et al</i> [8]	HadGEM	Sulfur cycle, oxidants from an interactive chemistry model	Bulk Modal	SO4, NH4, OC, BC, SS, SOA	Online	-1.48 -1.17
Yu <i>et al</i> (this study)	GEOS- Chem/APM (driven by GEOS-5 assimilated met)	Full online chemistry (SOx–NOx–Ox– hydrocarbon–SOA)	SP 40 bins, SS 20 bins, D 15 bins, BC and OC 2 modes	SO4, NH4, NIT, SOA, SS, D, BC, OC	Online	-0.75

^a SO₄: sulfate; SS: sea salt; D: mineral dust; BC: black carbon; OC: organic carbon; N: nitrate; NH₄: ammonium; SOA: secondary organic aerosol; SP: secondary particles (containing SO₄, NH₄, NIT, and SOA).

^b AGCM: atmospheric general circulation model; CSIRO: Commonwealth Scientific and Industrial Research Organization; CAM: NCAR community atmosphere model; IMPACT: integrated massively parallel atmospheric chemical transport model; GFDL: Geophysical Fluid Dynamics Laboratory; GISS: NASA Goddard Institute for Space Studies; LMDZ: Laboratorie de Meteorologie Dynamique general circulation model; INCA: interaction with chemistry and aerosols; HadGEM: the Earth system model of the UKMet Office Hadley Centre.

As we can see from figure 4, large diversity of FAIRF values still exists among the post-IPCC2007 studies, ranging from ~ -1.76 to -0.17 W m⁻². Unfortunately, the distribution is far from normal, with only two (out of eleven) studies giving FAIRF close to the median values of ~ -0.75 W m⁻². Five studies show relatively large negative forcing (range: -1.76 to -1.17 W m⁻²; mean: -1.48 W m⁻²) while four other studies show much weaker forcing (range: -0.39 to -0.17 W m⁻²; mean: -0.33 W m⁻²). It should be noted that three of five studies with large negative forcing are based on the same global aerosol model, IMPACTS, while the other two are based on HadCEM. Many factors (including

those listed in table 2) may contribute to the large FAIRF diversity due to the differences in the treatments of chemistry, aerosol microphysics, aerosol-cloud-radiation interactions, and meteorology in various studies.

4. Discussion

Compared to other global models used for IPCC2007 and post-IPC2007 FAIRF studies (see table 2, also table 2.7 in IPCC2007), GEOS-Chem/APM has a number of advanced features including: (1) online full (SOx-HOx-NOx-VOC-ozone) chemistry instead of simplified sulfur cycle chemistry using off-line monthly mean oxidants; (2) consideration of important aerosol components (nitrate, ammonium, and SOA) that are not included in most of previous studies; (3) highly accurate representation (40 bins with 30 bins in the size range of 1.2-120 nm) of secondary particles that are known to dominate particle number concentrations over most parts of the globe; (4) the usage of more accurate assimilated meteorology (GEOS-5) that helps to reduce uncertainties associated with meteorology; (5) online radiative transfer (RT) calculation to ensure that the spatial and temporal variations of aerosols are consistent with meteorology fields (compared to the studies using monthly mean aerosol fields for off-line RT calculation). These advanced features put GEOS-Chem/APM as one of the most sophisticated and rigorous global models currently available for studying FAIRF. Actually, this is the first time (to our knowledge) that a global size-resolved (sectional) aerosol model with comprehensive online chemistry and considering all major aerosol species (including nitrate, ammonium, and second organic aerosols) is used to calculate FAIRF.

FAIRF remains to be a major source of uncertainty in studying climate change. It is important to identify the possible reasons for the large diversity in FAIRF values derived from different studies/models and quantify the uncertainties associated with the treatments of various processes related to chemistry, aerosol microphysics, cloud activation and physics, radiation, and meteorology. One main source of the large diversity in post-IPCC2007 FAIRF studies is associated with the difference in the predicted PD/PI aerosol properties, especially CCN number concentrations that are critical for FAIRF. Large differences in the PD/PI CCN predictions are expected because of very different consideration of chemistry, aerosol sizes and species and microphysical processes in various models (table 2). The level of complexity of GEOS-Chem/APM in treating chemistry-aerosol processes compared to other models (table 2) and its ability in reproducing observed CCN concentrations around the globe (figure 2) is expected to substantially reduce the uncertainty in the predicted impacts of anthropogenic emissions on key aerosol properties and associated aerosol climate effect. For example, GEOS-Chem/APM simulations indicate that compared to the pre-industrial time, the concentrations of key precursor gases, nucleation rates, and particle number concentrations decreased in some pristine areas (figure 1). Such a decrease is likely as a result of the changes in atmospheric oxidation capacity and chemistry and condensation sink associated with anthropogenic emissions, highlighting the importance of coupling between full online chemistry and comprehensive aerosol microphysics.

Additional sources of the large diversity in FAIRF values include the representation of aerosol-cloud interactions (sub-grid updraft velocities, aerosol activation, etc) and the uncertainties in the cloud fields (3D distributions of cloud fraction and water content, etc). It should be pointed out that, unlike wind, temperature, and water vapor in the re-analysis product (i.e., assimilated meteorology), cloud properties are often not adequately constrained. The GEOS cloud water simulation has been evaluated by Li *et al* [49] and low cloud cover has been compared to ISCCP data [50]. There exist substantial differences between simulated and observed cloud properties but the cloud water values derived from different satellites [49] also differ significantly. The performance of GEOS in simulating cloud properties is comparable to many other widely use climate models [49]. It remains to be studied how the uncertainties in cloud simulations and aerosol–cloud interaction parameterizations may impact the calculated FAIRF.

Acknowledgments

This work is supported by NASA under grant NNX11AQ72G, and NSF under grant 0942106. The GEOS-Chem model is managed by the Atmospheric Chemistry Modeling Group at Harvard University with support from NASA's Atmospheric Chemistry Modeling and Analysis Program. The shortwave RRTMG module integrated with GEOS-Chem in this study is downloaded from the website of Atmospheric and Environmental Research (http://rtweb.aer. com/). The authors thank Andrea Molod for information about the GEOS cloud products.

References

- Twomey S 1977 The influence of pollution on the shortwave albedo of clouds J. Atmos. Sci. 34 1149–52
- [2] Forster P et al 2007 Changes in atmospheric constituents and in radiative forcing Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change ed S Solomon et al (Cambridge: Cambridge University Press)
- [3] Lohmann U et al 2007 Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM Atmos. Chem. Phys. 7 3425–46
- [4] Rotstayn L D and Liu Y 2009 Cloud droplet spectral dispersion and the indirect aerosol effect: comparison of two treatments in a GCM *Geophys. Res. Lett.* 36 L10801
- [5] Wang M and Penner J E 2009 Aerosol indirect forcing in a global model with particle nucleation *Atmos. Chem. Phys.* 9 239–60
- [6] Penner J E, Xu L and Wang M 2011 Satellite methods underestimate indirect climate forcing by aerosols *Proc. Natl Acad. Sci. USA* 108 13404–8
- [7] Bauer S E and Menon S 2012 Aerosol direct, indirect, semidirect, and surface albedo effects from sector contributions based on the IPCC AR5 emissions for preindustrial and present-day conditions *J. Geophys. Res.* 117 D01206
- [8] Bellouin N *et al* 2012 Impact of the modal aerosol scheme GLOMAP-mode on aerosol forcing in the Hadley Centre Global Environmental Model *Atmos. Chem. Phys. Discuss.* 12 21437–79
- [9] Deandreis C, Balkanski Y, Dufresne J L and Cozic A 2012 Radiative forcing estimates of sulfate aerosol in coupled climate-chemistry models with emphasis on the role of the temporal variability *Atmos. Chem. Phys.* 12 5583–602
- [10] Zhou C, Penner J E, Ming Y and Huang X L 2012 Aerosol forcing based on CAM5 and AM3 meteorological fields *Atmos. Chem. Phys.* **12** 9629–52
- [11] Penner J E, Zhou C and Xu L 2012 Consistent estimates from satellites and models for the first aerosol indirect forcing *Geophys. Res. Lett.* 39 L13810
- [12] Quaas J, Boucher O, Bellouin N and Kinne S 2011 Which of satellite- or model-based estimates is closer to reality for

aerosol indirect forcing? *Proc. Natl Acad. Sci. USA* **108** E1099

- [13] Bellouin N, Quaas J, Morcrette J J and Boucher O 2012 Estimates of aerosol radiative forcing from the MACC re-analysis Atmos. Chem. Phys. Discuss. 12 20073–111
- [14] Weisenstein D K, Penner J E, Herzog M and Liu X 2007 Global 2-D intercomparison of sectional and modal aerosol modules Atmos. Chem. Phys. 7 2339–55
- [15] Bergman T et al 2011 Evaluation of the sectional aerosol microphysics module SALSA implementation in ECHAM5-HAM aerosol-climate model Geosci. Model. Dev. Discuss. 4 3623–90
- [16] Mann G W *et al* 2012 Intercomparison of modal and sectional aerosol microphysics representations within the same 3D global chemical transport model *Atmos. Chem. Phys.* 12 4449–76
- [17] Jacobson M Z 2001 GATOR-GCMM: a global through urban scale air pollution and weather forecast model. 1. Model design and treatment of subgrid soil, vegetation, roads, rooftops, water, sea ice, and snow J. Geophys. Res. 106 5385–402
- [18] Adams P J and Seinfeld J H 2002 Predicting global aerosol size distributions in general circulation models J. Geophys. Res. 107 4370
- [19] Ma X and von Salzen K 2006 The dynamics of the sulphate size distribution on a global scale J. Geophys. Res.
 111 D08206
- [20] Yu F and Luo G 2009 Simulation of particle size distribution with a global aerosol model: contribution of nucleation to aerosol and CCN number concentrations *Atmos. Chem. Phys.* **9** 7691–710
- [21] Bey I et al 2001 Global modeling of tropospheric chemistry with assimilated meteorology: model description and evaluation J. Geophys. Res. 106 23073–96
- [22] Martin R V, Jacob D J, Yantosca R M, Chin M and Ginoux P 2003 Global and regional decreases in tropospheric oxidants from photochemical effects of aerosols *J. Geophys. Res.* 108 4097
- [23] Park R J, Jacob D J, Field B D, Yantosca R M and Chin M 2004 Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: implications for policy J. Geophys. Res. 109 D15204
- [24] Evans M J and Jacob D J 2005 Impact of new laboratory studies of N₂O₅ hydrolysis on global model budgets of tropospheric nitrogen oxides, ozone, and OH *Geophys. Res. Lett.* **32** L09813
- [25] Liao H, Henze D K, Seinfeld J H, Wu S and Mickley L J 2007 Biogenic secondary organic aerosol over the United States: comparison of climatological simulations with observations J. Geophys. Res. 112 D06201
- [26] Henze D K, Seinfeld J H, Ng N L, Kroll J H, Fu T-M, Jacob D J and Heald C L 2008 Global modeling of secondary organic aerosol formation from aromatic hydrocarbons: high- versus low-yield pathways Atmos. Chem. Phys. 8 2405–20
- [27] van der Werf G R *et al* 2012 Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009) *Atmos. Chem. Phys.* 10 11707–35
- [28] Fisher J A et al 2011 Sources, distribution, and acidity of sulfate-ammonium aerosol in the Arctic in winter-spring Atmos. Environ. 45 7301–18
- [29] Murray L T, Jacob D J, Logan J A, Hudman R C and Koshak W J 2012 Optimized regional and interannual variability of lightning in a global chemical transport model constrained by LIS/OTD satellite data J. Geophys. Res. 117 D20307
- [30] Iacono M J et al 2008 Radiative forcing by long-lived greenhouse gases: calculations with the AER radiative transfer models J. Geophys. Res.-Atmos. 113 D13103
- [31] Abdul-Razzak H and Ghan S J 2002 A parameterization of aerosol activation—3. Sectional representation J. Geophys. Res. 107 4026

- [32] Yu F, Luo G and Ma X 2012 Regional and global modeling of aerosol optical properties with a size, composition, and mixing state resolved particle microphysics model *Atmos. Chem. Phys.* **12** 5719–36
- [33] Ma X and Yu F 2012 Effect of spectral-dependent surface albedo on Saharan dust directradiative forcing *Geophys. Res. Lett.* **39** L09808
- [34] Feingold G and Heymsfield A 1992 Parameterizations of condensational growth of droplets for use in general circulation models J. Atmos. Sci. 49 2325–42
- [35] Chuang C C, Penner J E, Taylor K E, Grossman A S and Walton J J 1997 An assessment of the radiative effects of anthropogenic sulfate *J. Geophys. Res.* **102** 3761–78
 [36] Paluch I R and Lenschow D 1992 Comment on
- [36] Paluch I R and Lenschow D 1992 Comment on 'measurements of aitken nuclei and cloud condensation nuclei in the marine atmosphere and their relation to the DMS–cloud–climate hypothesis' by Hegg DA, Radke LF, Hobbs PV J. Geophys. Res. 97 7657–8
- [37] Chuang C C et al 2002 Cloud susceptibility and the first aerosol indirect forcing: sensitivity to black carbon and aerosol concentrations J. Geophys. Res. 107 4564
- [38] Chen Y and Penner J E 2005 Uncertainty analysis of the first indirect aerosol effect Atmos. Chem. Phys. 5 2935–48
- [39] Rotstayn L D and Liu Y G 2003 Sensitivity of the first indirect aerosol effect to an increase of cloud droplet spectral dispersion with droplet number concentration *J. Clim.* 16 3476–81
- [40] Myhre G et al 2013 Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations Atmos. Chem. Phys. 13 1853–77
- [41] Andreae M O 2009 Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions *Atmos. Chem. Phys.* 9 543–56
- [42] Yu F, Luo G, Turco R P, Ogren J and Yantosca R 2012 Decreasing particle number concentrations in a warming atmosphere and implications Atmos. Chem. Phys. 12 2399–408
- [43] Ward D S and Cotton W R 2011 Cold and transition season cloud condensation nuclei measurements in western Colorado Atmos. Chem. Phys. 11 4303–17
- [44] Irwin M, Robinson N, Allan J D, Coe H and McFiggans G 2011 Size-resolved aerosol water uptake and cloud condensation nuclei measurements as measured above a Southeast Asian rainforest during OP3 Atmos. Chem. Phys. 11 11157–74
- [45] Laakso L et al 2012 Boundary layer nucleation as a source of new CCN in savannah environment Atmos. Chem. Phys. Discuss. 12 8503–31
- [46] Penner J E et al 2001 Aerosols, their direct and indirect effects Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change ed J T Houghton et al (Cambridge: Cambridge University Press) pp 289–348
- [47] Ghan S J *et al* 2012 Toward a minimal representation of aerosols in climate models: comparative decomposition of aerosol direct, semi-direct and indirect radiative forcing *J. Clim.* 25 6461–76
- [48] Spracklen D V, Carslaw K S, Pöschl U, Rap A and Forster P M 2011 Global cloud condensation nuclei influenced by carbonaceous combustion aerosol Atmos. Chem. Phys. 11 9067–87
- [49] Li J-L F, Waliser D, Woods C, Teixeira J, Bacmeister J, Chern J, Shen B-W, Tompkins A, Tao W-K and Köhler M 2008 Comparisons of satellites liquid water estimates to ECMWF and GMAO analyses, 20th century IPCC AR4 climate simulations, and GCM simulations *Geophys. Res. Lett.* 35 L19710
- [50] Molod A, Takacs L, Suarez M, Bacmeister J, Song I-S and Eichmann A 2012 The GEOS-5 atmospheric general circulation model: mean climate and development from MERRA to Fortuna NASA Technical Report Series on Global Modeling and Data Assimilation vol 28 (NASA TM-2012-104606/Vol 28) p 117