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Indirect radiative forcing by ion-mediated nucleation of aerosol

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Abstract

A clear understanding of particle formation mechanisms is critical for assessing aerosol indirect radiative forcing and associated climate feedback processes. Recent studies reveal the importance of ion-mediated nucleation (IMN) in generating new particles and cloud condensation nuclei (CCN) in the atmosphere. Here we implement for the first time a physically-based treatment of IMN into the Community Atmosphere Model version 5. Our simulations show that, compared to globally averaged results based on binary homogeneous nucleation (BHN), the presence of ionization (i.e., IMN) halves H_2SO_4 column burden, but increases the column integrated nucleation rate by around one order of magnitude, total particle number burden by a factor of ~3, CCN burden by ~10 % (at 0.2 % supersaturation) to 65 % (at 1.0 % supersaturation), and cloud droplet number burden by ~18 %. Compared to BHN, IMN increases cloud liquid water path by 7.5 %, decreases precipitation by 1.1 %, and increases total cloud cover by 1.9 %. This leads to an increase of total shortwave cloud radiative forcing (SWCF) by 3.67 $W m^{-2}$ (more negative) and longwave cloud forcing by 1.78 $W m^{-2}$ (more positive), with large spatial variations. The effect of ionization on SWCF derived from this study (3.67 $W m^{-2}$) is a factor of ~3 higher that of a previous study (1.15 $W m^{-2}$) based on a different ion nucleation scheme and climate model. The large sensitivity of cloud forcing to nucleation process again calls for improving representation of secondary particle formation processes and aerosol-cloud interactions in climate models.

1 Introduction

Aerosol particles formed in the atmosphere influence climate indirectly by acting as cloud condensation nuclei (CCN) that affect cloud properties and precipitation (Twomey, 1977; Albrecht, 1989). The aerosol indirect radiative forcing (IRF) is a major source of uncertainty in interpreting climate change over the past century and projecting future change. New particle formation has been well recognized to be an important

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source of CCN in the atmosphere (Pierce and Adams, 2007; Spracklen et al., 2008; Makkonen et al., 2009; Wang and Penner, 2009; Yu and Luo, 2009; Kazil et al., 2010). Global climate simulations indicate that the aerosol IRF is sensitive to parameterizations of nucleation processes (Wang and Penner, 2009; Kazil et al., 2010). Wang and Penner (2009) showed that the first IRF of anthropogenic aerosols (forcing due to changes in droplet number and size but not liquid water content) ranges from −1.22 to $-2.03\,\mathrm{W\,m^{-2}}$ for six different combinations of binary homogeneous nucleation (BHN), empirical parameterization of boundary layer nucleation, and parameterization of primary sulfate emission to represent sub-grid scale nucleation. Kazil et al. (2010) investigated the impact of the individual aerosol nucleation mechanisms (neutral and charged nucleation of sulfuric acid throughout the troposphere, and cluster activation limited to the forested boundary layer) on the Earth's energy balance, and showed that the change in the net top of atmosphere shortwave radiative flux associated with nucleation is around $-2.55\,\mathrm{W\,m^{-2}}$. These previous studies highlight the importance of a clear understanding of atmospheric particle nucleation processes and proper representation of these processes in the climate models.

There still exist large uncertainties in nucleation mechanisms, despite significant progress achieved over the past several decades (e.g. Zhang et al., 2012). One of these uncertainties is the role of air ions generated by galactic cosmic rays and radioactive materials in the nucleation process (Yu and Turco, 2000; Lovejoy et al., 2004). While the relative contribution of ion-mediated nucleation (IMN) versus neutral nucleation has been controversial in the past years (Kulmala et al., 2007; Yu and Turco, 2008), recent detailed case studies (Yu and Turco, 2011) and laboratory measurements (Enghoff et al., 2011; Kirkby et al., 2011) clearly show a significant role of ionization in promoting nucleation. In particular, Yu and Turco (2011) demonstrated that the state-of-the-art multi-instrument field measurements taken in a boreal forest appear to strongly support the dominance of IMN mechanism, which is further supported by the most recent cluster mass spectrometer measurements at the site showing the absence of small neutral clusters (Jokinen et al., 2012). It should be pointed out that empirical parameterizations

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of boundary layer nucleation derived from the boreal forest nucleation measurements have been widely used to represent a yet-to-be-identified mechanism of new particle formation in global models (e.g. Wang and Penner, 2009; Kazil et al., 2010) but these parameterizations could in fact be a simplified fitting to the IMN process (Yu and Turco, 2011).

Considering the unequivocal evidence of the IMN process in producing atmospheric particles, we seek to assess the effect of ionization on new particle formation, CCN abundance, cloud properties, and cloud radiative forcing in this study by incorporating the IMN mechanism into the Community Atmosphere Model version 5 (CAM5), the atmospheric component of the Community Earth System Model version 1 (CESM1). Our strategy is to compare results with and without the effect of ionization on new particle formation. The kinetically self-consistent IMN is suitable for this purpose as it fully and consistently reduces to binary homogeneous nucleation (BHN) when the ionization rate is zero (Yu, 2010a). The remaining sections of this paper are organized as follows: model description and set-up are given in Sect. 2. Section 3 provides a detailed analysis of simulation results. Conclusions are given in Sect. 4.

2 Model and simulation description

The model we employed for this study is CAM5 with a modal aerosol module (MAM) using three log-normal modes (Aitken, accumulation, and coarse) to represent aerosols (MAM3). Detailed information about CAM5-MAM can be found in Eaton (2011) and Liu et al. (2012). Here we give a brief summary of key features of the model relevant to the present work. In MAM3, Aitken mode species include sulfate, secondary organic aerosol (SOA), and sea salt; accumulation mode species include sulfate, SOA, black carbon (BC), primary organic matter (POM), sea salt, and dust; coarse mode species include sea salt, dust, and sulfate. All species within a mode are assumed to be internally mixed. The model explicitly treats aerosol transport, primary emissions, aerosol nucleation (binary homogeneous and empirical boundary layer involving H_2SO_4 vapor),

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condensation of trace gases (H_2SO_4 and semi-volatile organics) on existing aerosol particles, coagulation (Aitken and accumulation modes), dry and wet deposition, and activation into stratiform cloud droplets and re-suspension (Liu et al., 2012). The nucleated particles are added to the Aitken mode, with coagulation loss as they grow from critical cluster size to Aitken mode size taken into account following the parameterization of Kerminen and Kulmala (2002). In stratiform clouds, aerosol activation is treated consistently with droplet nucleation which is parameterized in terms of updraft velocity and the properties of all of the aerosol modes (Abdul-Razzak and Ghan, 2000). The present CAM5 does not consider microphysical interaction between aerosol and convective condensate, and thus aerosol effects on cumulus are not simulated although convective precipitation scavenges aerosols (i.e., wet scavenging) (Liu et al., 2012). The radiative transfer scheme used in CAM5 is the Rapid Radiative Transfer Model for GCMs (RRTMG), a broadband k-distribution radiation model developed for application to GCMs (e.g. Iacono et al., 2008).

In this study, we implement for the first time the IMN mechanism (Yu, 2010a) in CAM5.1. As mentioned earlier, the physically-based IMN is supported by field measurements and consistently reduces to BHN when the ionization rate is set to zero, enabling us to distinguish the effect of ionization. We run the conventional CAM5.1-MAM3 at $1.9^\circ \times 2.5^\circ$ horizontal resolution with 30 vertical levels and a time step of 30 min, with prescribed sea surface temperature and sea ice. Two separate simulations have been carried out under present-day climate and present-day emissions (PDPD): one with IMN and the other based on BHN (i.e., without ionization). Neither simulation uses nucleation schemes (binary homogeneous and empirical boundary layer nucleation) contained in the original version of CAM5.1. The global ionization rates due to cosmic rays are calculated based on the schemes given in Usoskin and Kovaltsov (2006) and the contribution of radioactive materials from soil to ionization rates is parameterized based on the profiles given in Reiter (1992).

We use the same database and schemes for aerosol and precursor emissions (for year 2000) as described in Liu et al. (2012) and Wang et al. (2011), except that, in

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order to clearly assess the effect of nucleation, the fraction of anthropogenic sulfur emitted as primary sulfate (used to represent sub-grid nucleation process) has been set to zero. Many previous global aerosol modeling studies have assumed some fraction (0–5 %) of anthropogenic sulfur emitted directly as sulfate particles to account for the new particle formation in sub-grid SO_2 plumes (Luo and Yu, 2011; and references therein). However, to assume a constant fraction of sulfur emitted directly as particles (with an assumed percentage partitioning into Aitken and accumulation modes) may lead to large uncertainty in the simulated spatiotemporal distribution of particle number concentrations, owning to the strong dependence of sub-grid nucleation on many environmental parameters (especially OH concentration, temperature, surface area of pre-existing particles, etc.) (Yu, 2010b). In addition, to treat sub-grid nucleation as primary particle emission leads to the underestimation of the contribution of nucleation processes to global aerosol number abundance (Luo and Yu, 2011). We run the CAM5.1-MAM3 for 6 yr (2000–2005) for two cases (i.e, with and without effect of ionization on nucleation) and the results averaged over the last five simulation years (2001–2005) for each case are presented in this paper.

3 Results

Table 1 summarizes the globally averaged results of key variables for both BHN and IMN cases, as well as the differences for the two cases showing the impacts of ionization. To account for the differences in various altitudes, we vertically integrate all the 3-D variables to simplify the comparisons. It is clear from Table 1 that ionization has a significant effect on H_2SO_4 vapor concentrations (hereafter [H_2SO_4]), nucleation rates, concentrations of aerosol and CCN, cloud properties, precipitation, shortwave cloud forcing (SWCF), and longwave cloud forcing (LWCF). The corresponding spatial distributions of selected parameters are presented in Figs. 1–3 and Fig. S1 in Supplement.

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H_2SO_4 vapor from both anthropogenic (fossil fuels, etc.) and natural (DMS, volcano, etc.) sources is known to play an important role in forming and growing new particles. H_2SO_4 column burdens are high in the source and associated outflow regions (Fig. 1a, b), with highest values exceeding $6 \times 10^{16}\,m^{-2}$. H_2SO_4 vapor in the atmosphere is produced in-situ via photochemistry from anthropogenic and natural SO_2, and thus have strong diurnal variations. Because of ion-dipole interaction, IMN occurs at [H_2SO_4] (or H_2SO_4 supersaturation ratio) lower than that needed for BHN to occur. As shown in Table 1 and Fig. 1a, b, H_2SO_4 column burden is about halved at the presence of ionization. This is mainly a result of overall higher nucleation rate (Fig. 1c, d) and particle number concentration (Fig. 1e, f) and hence condensation sink when the effect of ions on nucleation is considered. The column integrated rate of IMN (J_{IMN}, Fig. 1c and also Table 1) is about one order of magnitude higher than that of BHN, despite lower average [H_2SO_4] in the IMN case. In the tropic and sub-tropic regions, areas of high nucleation generally co-locate with areas of high [H_2SO_4]. In the polar regions, nucleation is substantial over Antarctica but insignificant over Arctic regions, as a result of relatively higher [H_2SO_4] and lower temperature over Antarctica. It should be noted that BHN can still occur even in the presence of ionization but its rates are much smaller than those shown in Fig. 1d because IMN lowers [H_2SO_4] and BHN rates decrease sharply with decreasing [H_2SO_4]. Because of $\sim$ one order of magnitude higher overall nucleation rates with IMN, the total burden of condensation nuclei (CN, calculated as the total aerosol number over all sizes/modes) for IMN cases is about tripled when compared to BHN only cases (Table 1, also Fig. 1e, f). The relatively lower enhancement in CN (a factor of $\sim$3) compared to that of nucleation rate (a factor of $\sim$10) in the presence of ionization is mainly a result of coagulation, which is enhanced in the IMN case because lower [H_2SO_4] results in slower growth from cluster to Aitken size. Since the emissions of primary particles are the same for both IMN and BHN cases, we can see from Fig. 1e, f that IMN is a dominant source of atmospheric particles (in term of number abundance) almost everywhere except in several regions (South and East Asia, and parts of Africa and South America) where primary anthropogenic and

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biomass burning emissions are also significant. More information about the effects of ionization on [H_2SO_4], nucleation rates, and CN at different altitudes can be found in the Supplement.

In the atmosphere, the fraction of CN that can activate and produce cloud droplets depends on particle size distribution and composition as well as the water supersaturations (S) attained in clouds (up to ~1 % for convective clouds and ~0.2 % for stratus clouds). Based on the predicted 5-yr average vertically integrated CCN concentrations (or CCN burdens) at S = 1 % (CCN1.0), and S = 0.2 % (CCN0.2) as well as cloud droplet number (CDN) concentrations given in Table 1, ionization enhances CCN1.0 by 64.5 %, CCN0.2 by 9.3 %, and CDN by ~18.3 %. As expected, enhanced nucleation has a stronger effect on the concentrations of smaller particles (and hence CCN at higher S). In Table 1 we also give vertically integrated CCN1.0 and CCN0.2 burden within two layers in the lower troposphere: boundary layer (BL, surface – 900 hPa) and lower free troposphere (LFT 900–650 hPa). We can see that a large fraction of CCN resides in the lower troposphere (below ~650 hPa) and the effect of ionization is larger in LFT than in BL, as a result of primary particle emission in BL and higher growth rates of nucleated particles in the lower troposphere.

Through the aerosol indirect effects (Twomey, 1977; Albrecht, 1989), the changes of CCN and CDN concentrations lead to the modification of cloud liquid water path (LWP), cloud ice water path (IWP), precipitation, and total cloud cover (CLDTOT). Compared to BHN, IMN increases LWP by 7.5 %, decreases precipitation by 1.1 %, and increases CLDTOT by 1.9 % (Table 1). These percentage changes are globally averaged values and, because of various feedbacks, there exist large spatial variations of such changes. Figure 2 shows the horizontal distribution of CLDTOT and precipitation based on IMN and the differences of these two variables between IMN and BHN cases. The CLDTOT derived from MODIS and precipitation from the Global Precipitation Climatology Project (GPCP) for the same 5-yr periods are also shown for comparison. As seen in Fig. 2, the model simulations reasonably capture the spatial distribution of observed CLDTOT and precipitation. It is clear that the effect of nucleation on CLDTOT and precipitation

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is spatially inhomogeneous and can be both positive and negative depending on locations. The largest increase in CLDTOT associated with IMN appears to occur in the tropical and Arctic regions reaching 4–8 %.

While CAM5 captures the global distributions of total cloud cover quite well (within a few percentages for globally averaged values), there exist substantial differences between the predicted and observed global mean LWP and precipitation (~10 %) (Table 1). CAM5 appears to under-predict LWP and over-predict precipitation. It should be noted that there exist large uncertainties in the observed LWP; estimates from different satellites can differ by up to ~45 % or more (e.g. O'Dell et al., 2008; Seethala and Horváth, 2010). In contrast, the uncertainty in precipitation data, derived from the GPCP through a merged analysis that incorporates precipitation estimates from low-orbit satellite microwave data, geosynchronous-orbit satellite infrared data, and surface rain gauge observations, is expected to relatively smaller (~116 %) (Adler et al., 2003). It is conceivable that the under-prediction of LWP is a result of precipitation over-prediction which occurs largely over the tropical regions (see Fig. 2d, e). As mentioned earlier, the present CAM5 does not consider indirect effect of aerosols on convective clouds which dominate precipitations. Further research is needed to understand the interaction of aerosols with convective clouds and improve the representation of such interaction in CAM5.

Due to the large role of clouds in Earth's climate (total SWCF and LWCF in the order of $\sim -50\,\mathrm{W\,m^{-2}}$ and $25\,\mathrm{W\,m^{-2}}$, respectively, see Table 1 and Fig. 3), a small change in cloud properties can have substantial impacts on Earth's energy balance. Compared to the case without ionization (i.e. BHN), IMN induced changes in CN and CCN concentrations and thus on the LWP, precipitation, and CLDTOT increase the total SWCF by $\sim 3.67\,\mathrm{W\,m^{-2}}$ (more negative) and LWCF by $1.78\,\mathrm{W\,m^{-2}}$ (more positive). The effect of ionization on net cloud forcing is $-1.9\,\mathrm{W\,m^{-2}}$. A close look at the horizontal distributions (Fig. 3) reveals large spatial variations ranging from $\sim -20\,\mathrm{W\,m^{-2}}$ to $+10\,\mathrm{W\,m^{-2}}$ for ΔSWCF, and $\sim -5\,\mathrm{W\,m^{-2}}$ to $+20\,\mathrm{W\,m^{-2}}$ for ΔLWCF. ΔSWCF (ΔLWCF) is negative (positive) over most part of oceans but is positive (negative) over some part of continents.

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The large sensitivity of SWCF to CCN and secondary particle formation highlights the importance of reducing uncertainty in predicting key processes controlling CCN abundance in the troposphere.

Kazil et al. (2010) investigated the globally averaged annual mean contributions of the individual nucleation processes to changes in net top-of-atmosphere shortwave radiation and showed that the contribution of charged H_2SO_4/H_2O nucleation is $-1.15\,W\,m^{-2}$. This value is much smaller than the ΔSWCF value of $-3.67\,W\,m^{-2}$ derived from this study. One possible reason for the difference is that the IMN used in this study (Yu, 2010a) is different from the ion-induced nucleation (IIN) (Lovejoy et al., 2004; Kaizl and Lovejoy, 2007) used in Kazil et al. (2010). Previous comparisons (Yu and Turco, 2008; Yu et al., 2010) indicate that IIN rates based on the model of Lovejoy et al. are generally several orders of magnitude lower than the IMN rates and appears to under-predict the new particle formation rate in the troposphere. Another possible factor is that aerosol indirect effects in CAM5 are quite strong (Wang et al., 2011), which affects the magnitude of the IMN induced effects found in our study. Such a large difference in the impact of nucleation schemes on cloud forcing once again calls for a reduction of the uncertainty in modeling particle formation and growth processes as well as aerosol-cloud interactions in climate models.

4 Summary and discussion

Nucleation is widely known as an important source of atmospheric particles which are important to the Earth's climate through aerosol-cloud-precipitation-climate interactions. Recent detailed analysis of field studies and laboratory measurements clearly show significant impact of ionization in promoting nucleation. In the present work, based on the simulations of CAM5 with physics-based IMN mechanism incorporated, we show that ionization has a significant effect on H_2SO_4 vapor concentrations, nucleation rates, concentrations of aerosol and CCN, cloud properties, precipitation, and cloud forcing. Compared to the modeling results based on binary homogeneous

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nucleation (i.e., BHN case), the presence of ionization (i.e., IMN) increases the total SWCF by ~3.67 $W m^{-2}$ (more negative) and LWCF by 1.78 $W m^{-2}$ (more positive) The effect of ionization on SWCF derived from this study (3.67 $W m^{-2}$) is a factor of ~3 higher that of a previous study (1.15 $W m^{-2}$) based on a different ion nucleation scheme and climate model (Kazil et al., 2010). The large sensitivity of cloud forcing to nucleation process highlights the importance of reducing uncertainty in nucleation mechanisms and improving representation of aerosol-cloud-climate interaction processes in climate models. It should be pointed out that the present study on the impact of ionization is based on homogeneous and ion-mediated binary H_2SO_4-H_2O nucleation. Species other than H_2SO_4 and H_2O (such as ammonia, amines, organics) may influence both BHN and IMN and thus the net impact of ionization. Further research is needed to develop sound ternary homogeneous and ion-mediated nucleation theories and assess their effects on atmospheric particle formation and aerosol indirect radiative forcing.

The significant role of ionization in modifying global aerosol formation, CCN abundance, cloud properties, and cloud radiative forcing may provide an important physical mechanism linking climate change to various processes affecting atmospheric ionization (such as solar variations, Earth's magnetic field change, nuclear activities, etc.). Several previous modeling studies (Pierce and Adams, 2009; Kazil et al., 2012) suggest small impacts of solar variations induced modulation of galactic cosmic ray (GCR) flux on aerosols and clouds. To quantify the magnitude of this indirect solar forcing based on CAM5 will be the subject of future research.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/12/17347/2012/acpd-12-17347-2012-supplement.pdf.

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Table 1. Globally averaged sulfuric acid vapor concentration ([H_2SO_4]), nucleation rate (J), concentrations of condensation nuclei (CN) and cloud CN at water supersaturation ratio of 1.0 % and 0.2 % (CCN1.0, CCN0.2), cloud droplet number concentration (CDN), liquid water path (LWP), ice water path (IWP), precipitation (PRECT), total cloud cover (CLDTOT), shortwave cloud forcing (SWCF), and longwave cloud forcing (LWCF) for both BHN and IMN cases, as well as the differences for the two cases showing the impacts of ionization. To account for the differences in various altitudes, we vertically integrate all the 3-D variables to simplify the comparisons. For CCN1.0 and CCN0.2, we also give vertically integrated burden within two layers in the lower troposphere: boundary layer (BL, surface – 900 hPa) and middle layer (LFT, 900–650 hPa). The observed values of LWP, PRECP, CLDTOT, SWCF, and LWCF from different measurements are also given for comparisons.

		BHN	IMN	observations	IMN and BHN difference	
					Absolute	percentage
X		X_{BHN}	X_{IMN}		X_{IMN}-X_{BHN}	$X_{IMN}/X_{BHN}-1$
Column [H_2SO4]	(10^{16} # m^{-2})	2.80	1.46			−47.8 %
Column J	(10^{9} # m^{-2} s^{-1})	0.12	1.26			948.7 %
Column CN	(10^{10} # m^{-2})	219.90	618.27			181.2 %
Column CCN1.0	(10^{10} # m^{-2})	129.62	213.22			64.5 %
CCN1.0_BL	(10^{10} # m^{-2})	43.16	55.22			27.9 %
CCN1.0_LFT	(10^{10} # m^{-2})	40.12	67.02			67.0 %
Column CCN0.2	(10^{10} # m^{-2})	55.01	60.15			9.3 %
CCN0.2_BL	(10^{10} # m^{-2})	21.51	23.4			8.8 %
CCN0.2_LFT	(10^{10} # m^{-2})	19.49	21.73			11.5 %
Column CDN	(10^{9} # m^{-2})	13.08	15.46			18.3 %
LWP	(g m^{-2})	43.81	47.76	50 to 87[a]		9.0 %
IWP	(g m^{-2})	17.32	17.95			3.6 %
PRECT	(mm day^{-1})	3.00	2.97	2.67[b]		−1.1 %
CLDTOT		63.83	65.04	65.4[c], 66.7[d]		1.9 %
SWCF	(W m^{-2})	−50.80	−54.48	−46 to −53[e]	−3.68	
LWCF	(W m^{-2})	23.41	25.19	27 to 31[e]	1.78	

[a] Liquid water path is derived from SSM/I (for the years 1987–1994, Ferraro et al., 1996) and ISCCP for the year 1987 (Han et al., 1994). SSM/I data are restricted to oceans.
[b] Precipitation rate is taken from the Global Precipitation Climatology Project (GPCP) for the years 2001–2005 (Adler et al., 2003) (http://www.gewex.org/gpcpdata).
[c] Total cloud cover for 2001–2005 based on MODIS data.
[d] Total cloud cover for 2001–2005 based on ISCCP data.
[e] SWCF, LWCF are from ERBE for the years 1985–1989 (Kiehl and Trenberth, 1997) and CERES for the years 2000–2005 (Loeb et al., 2009).

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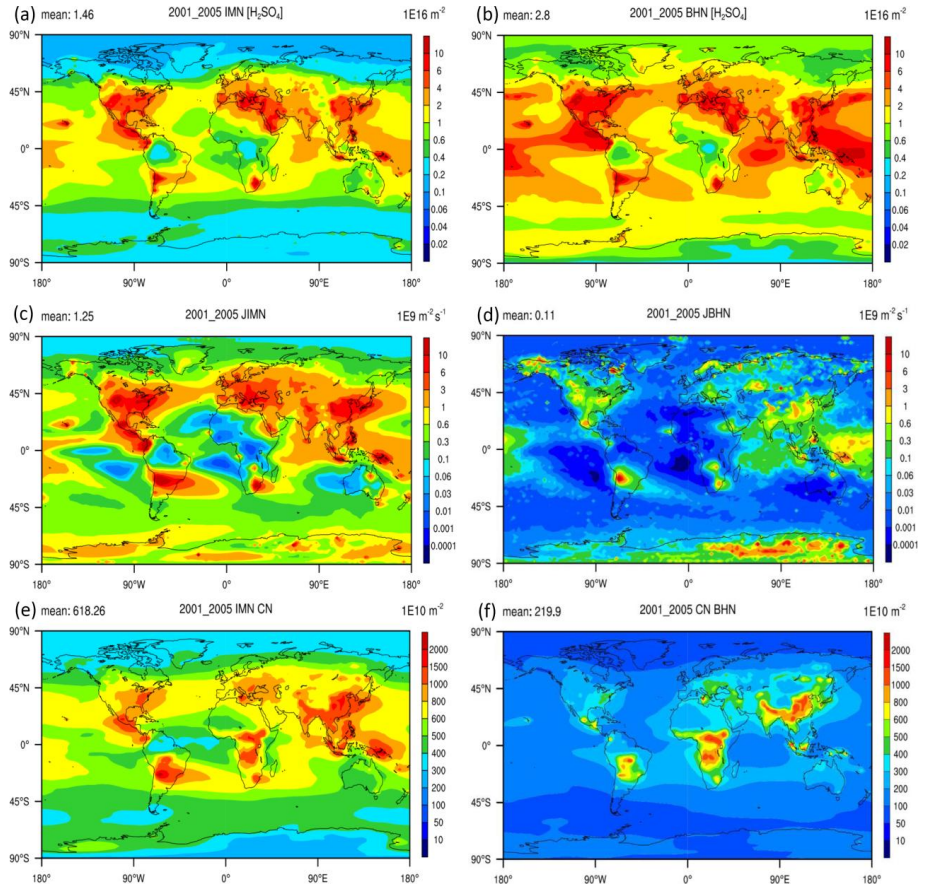

Fig. 1. Annual mean column burdens of H_2SO_4 vapor, column integrated nucleation rate (J), and total condensation nuclei (CN) number burden based on IMN **(a, c, e)** and BHN **(b, d, f)**.

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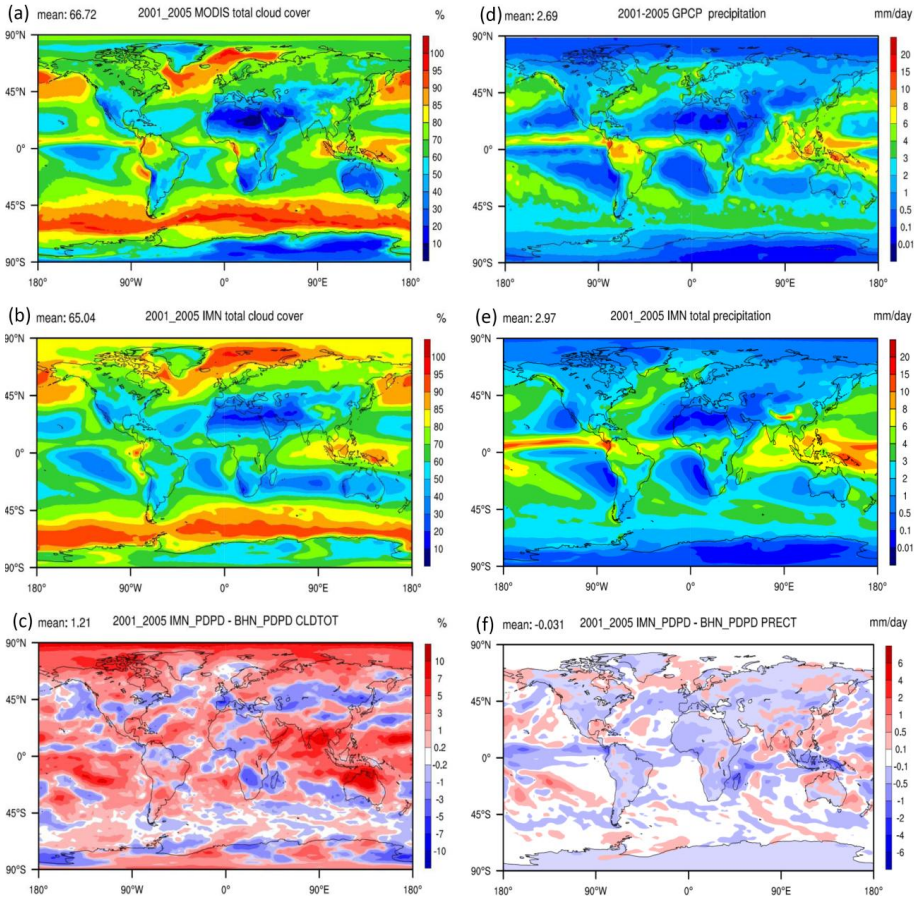

Fig. 2. Horizontal distribution of total cloud cover (CLDTOT) and precipitation rate based on IMN and the corresponding differences between IMN and BHN cases (IMN – BHN). The total CLDTOT derived from MODIS and precipitation from the Global Precipitation Climatology Project (GPCP) for the same 5-yr periods are also shown for comparison.

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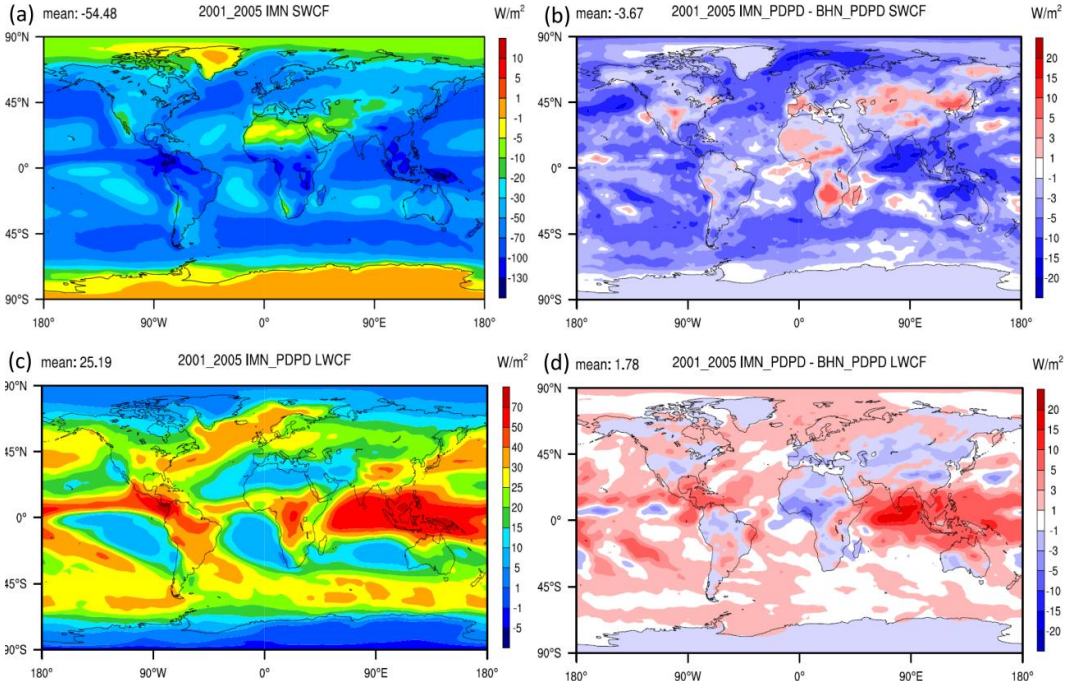

Fig. 3. Horizontal distribution of total SWCF and LWCF based on IMN and the corresponding differences between IMN and BHN cases.

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