Impact of anthropogenic absorbing aerosols on clouds and precipitation: A review of recent progresses*

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Impact of anthropogenic absorbing aerosols on clouds and precipitation: A review of recent progresses

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ABSTRACT

The climate impact of anthropogenic absorbing aerosols has attracted wide attentions recently. The unique forcing distribution of these aerosols displays, as instantaneous and in solar band, a significant heating to the atmosphere and a cooling in a close but smaller magnitude at the Earth's surface, leading to a positive net forcing to the Earth-atmosphere system, i.e., the forcing at the top of the atmosphere, which brings a warming tendency to the climate system. On the other hand, the atmospheric heating and surface cooling introduced by these aerosols have been demonstrated to be able to interact with dynamical processes in various scales to alter atmospheric circulation, and hence clouds and precipitation. Recent studies have suggested that the changes in precipitation caused by persistent forcing of anthropogenic absorbing aerosols through certain dynamical interactions, often appearing distant from the aerosol-laden regions, are likely more significant than those caused through aerosol–cloud microphysical connection confined locally to the aerosol concentrated areas. An active research field is forming to understand the changes in cloud and precipitation caused by anthropogenic absorbing aerosol through various dynamical linkages. This review discusses several recent findings regarding the effect of anthropogenic absorbing aerosols on cloud and precipitation, with an emphasis on works relate to the coupling between aerosol forcing and dynamical processes.

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1. Introduction

Absorbing aerosols in the atmosphere represent a unique particulate constituent. These aerosols can attenuate solar radiation through both scattering and absorption. Instantaneously, the latter effect leads to a significant heating to the atmosphere, and both effects cause a surface cooling. Besides exerting a residual positive radiative forcing at the top of the atmosphere that brings a warming tendency to the climate system, the atmospheric heating and surface cooling by these aerosols are also able to influence atmospheric thermal

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stability, surface heat exchange and evapotranspiration, and also circulation as well as formation and development of clouds and precipitation.

Anthropogenic absorbing aerosols are mainly from incomplete combustions of either fossil fuel or biofuel, resultant from various human activities from vehicle and industrial emissions to biomass burning events. These are soot particles containing the so-called black carbon (BC), a strong absorber of sunlight.

The context of anthropogenic absorbing aerosols, however, now extends beyond just black carbon, including for example certain organic matters that mostly contain humic acids as well as nitrated aromatics, polycyclic aromatic hydrocarbons, benzaldehydes, benzoic acids, aromatic polycarboxylic acids, and phenols, with absorption efficiency per mass that is though weaker than BC but non-negligible, and is often called brown carbon (Jacobson, 1999; Andreae and Gelencsér, 2006; Alexander et al., 2008). Another component of anthropogenic absorbing aerosols is the soil dust from land use, especially in emerging economies where large-scale construction projects often produce such pollutants throughout decades of continuing developments. Due to their potentially large mass abundance, both brown carbon and soil dust could represent a non-negligible contribution to the total particulate absorption. In addition, new single particle measurements (though still limited) have provided evidence for the existence of aerosol mixtures containing both absorbing and scattering constituents, for instance, a type that is consisted of pure scattering constituents such as sulfate coated on the surface of a black carbon core (e.g., Schwarz et al., 2008). Due to the absorbing core, the entire such mixed aerosol would behave like an absorbing aerosol. Comparing to natural absorbing aerosols, i.e., desert dust, current estimations suggest that anthropogenic absorbing aerosols dominate the particulate solar absorption in most regions globally except for those under influence of flows from deserts (Chung et al., 2005; Ramanathan et al., 2007; Wang et al., 2009a; Chin et al., 2009).

All these advanced knowledge have not just broadened the context of anthropogenic absorbing aerosols but also stimulated more in-depth research on aerosol mixing state and the life cycle of these mixed aerosols. Therefore, despite many existing unknowns regarding the soil dust from land use and also mixed absorbing aerosols, this review uses the term of anthropogenic absorbing aerosols instead of black carbon aerosols.

The climate effects of anthropogenic absorbing aerosols were first realized on their potential role in changing surface temperature. The positive forcing on the top of the atmosphere (TOA) seems suggesting that such aerosols might exert a warming effect to the climate system (e.g., Penner et al., 1998; Hansen et al., 1998; Myhre et al., 1998; Jacobson, 2000). This is based on the long established concept of equilibrium climate sensitivity, a ratio of equilibrium climate response measured by the global mean surface temperature change and the forcing at the TOA or the top of the tropopause, which has been found to be a nearly constant to many forcing agents (e.g., Manabe and Wetherald, 1967). However, due to the heterogeneous aerosol distribution both horizontally and vertically and the fact that the aerosol layer does not always locate close to the surface, the efficiency of the TOA direct forcing of anthropogenic absorbing aerosols in causing climatological surface temperature change clearly differs than that by the much more uniformly distributed and long-last greenhouse gasses including carbon dioxide, considering among others the planetary vertical and horizontal mixing scale and efficiency, and also adjustment due to long-wave radiative damping from the heated atmospheric layers (e.g., Penner et al., 2001). This has been demonstrated by the modeling studies based on equilibrium climate response derived from long-term model integrations using coupled climate models, suggesting that the ratio between the equilibrium climate sensitivity to black carbon and that to carbon dioxide is clearly lower than 1 (e.g., Wang, 2004; Roberts and Jones, 2004; Forster et al., 2007). There is proposal to adopt alternative quantities such as the adjusted forcing or effective forcing to replace the traditional radiative forcing in estimating the (equilibrium) climate response of aerosols, building upon an idea to allow certain fast feedbacks to disappear gradually and thus to give the way of equilibrium response to forcings that last longer (e.g., Lohmann et al., 2010). Considering the temporally varying horizontal and vertical distribution of anthropogenic absorbing aerosols and their large forcing gradient (e.g., Matsui and Pielke, 2006), both differing the cases of long-lived greenhouse gasses, such an effort is still up to thorough assessments from long integrations of coupled models under different forcing and feedback configurations. Rapid advancement in computational technology has provided us with much more computing powers to afford long-term climatological simulations. The climate response issue can be addressed much more practically than about four decades ago when the linear estimation based on instant radiative forcing was established. In-depth analyses of climate responses should be done with climate models with interactively predicted or even prescribed aerosol profiles to reveal many details including critical feedbacks along with the regional and global responses.

The climate impacts of anthropogenic absorbing aerosols on clouds and precipitation through their direct radiative forcing have been revealed recently in often centennial long equilibrium simulations using aerosol-climate models coupled with ocean component (e.g., Wang, 2004, 2007; Chung and Seinfeld, 2005). As a matter of fact, such a topic has become a new research focus now. It has also been extended to the coupling of aerosol forcing with dynamical processes in various scales to understand the potential changes caused by these aerosols to critical large-scale precipitation systems such as ITCZ and the monsoons. Increasing researches toward this direction reflect a graduate transition of the field from studying aerosol–cloud interaction, i.e., the radiative effects of aerosol through clouds, to aerosol–cloud-precipitation connection that includes hydrological besides radiative-thermodynamical effects, and from observing phenomenal changes to searching for in-depth understanding through the dynamical and physical reasons behind these changes.

It is inevitable that the climate response particularly in the context of clouds and precipitation needs to be examined with the assistant of numerical models, especially general circulation models or global climate models (GCMs), including aerosol processes. This review hence biases toward the global scale assessment and analyses using GCMs. The discussions are much more on the physical natures rather than using abstract definitions of various types of “forcings”, thus many commonly used terms that often lead to confusions are not

used extensively here. In addition, it is not the author's intention to produce a thorough literature review or survey, which is nearly impossible and also unnecessary for this dynamical field. Instead, only a few selected representative results are discussed. The review starts from a discussion of a few key factors that reflect the characteristics of anthropogenic absorbing aerosols and their climate forcings, along with description of recent efforts to represent these factors in the models. The research efforts and findings of the climate impacts of anthropogenic absorbing aerosols on clouds and precipitation will be discussed in the following sections, presenting respectively the aerosol effect through microphysical connection, local dynamical coupling, and global scale dynamical coupling.

2. Key factors determining the forcing of anthropogenic absorbing aerosols

The sunlight attenuation by aerosols through scattering and absorption exerts an external forcing to the atmosphere that is commonly referred to as the direct radiative forcing of aerosols. On the other hand, by providing existing surfaces to allow nucleation of cloud particles to occur, aerosols lead to a dominant role of heterogeneous nucleation in cloud formation in the atmosphere. The influences of anthropogenic aerosols on the formation of clouds as cloud condensation nuclei (CCN) or ice nuclei (IN) are also an additional effect above those by the natural aerosols. The resultant change in cloud radiation due to the activation of anthropogenic aerosols is commonly referred to as the first indirect radiative forcing of aerosols, or the albedo effect, while other resultant changes in cloud evolution including the formation of precipitation seem more adequate being referred to as responses than forcings [e.g., Ramaswamy et al., 2001].

Both the optical/radiative and microphysical effects of aerosols are determined to a large extent by the size and chemical composition of these particles. The extinction coefficient of the Mie scattering is a strong function of particle size (e.g., Seinfeld and Pandis, 1998). This is no exception for the absorbing aerosols. On the other hand, the chemical composition of aerosols defines the hygroscopicity of the particles and along with size it determines the activation probability of the particle when the thermodynamical condition is suitable (i.e., supersaturation to water or to ice; Pruppacher and Klett, 1997).

To estimate the direct or indirect radiative forcing of anthropogenic aerosols, it is desirable to have the information of size distribution for each aerosol constituents that presumably determining the chemical composition and hence the hygroscopicity. In an aerosol model, this can be done by building a size description of particles, assuming that aerosol constituents are all external mixtures, i.e., existing as themselves without mixing with each other. Therefore, one set of equations describing a single number concentration along with mass concentrations of all included aerosol constituents within a given size range will serve the purpose (Fig. 1a). Significant progress in representing size distribution in the global climate models has been made in recent years. For a better summary the reader is referred to Tao et al. (2012).

For absorbing aerosols, unfortunately, using size alone cannot well describe their climate effects. When exist as external mixture these particles are often poor CCN or IN. However, a coating of water-soluble matters such as inorganic acids on their surface, i.e., a core-shell structure, would change their hygroscopicity. Such core-shell mixture has been found in field measurement (Martins et al., 1998; Posfai et al., 1999; Naoe and Okada, 2001; Hara et al., 2003; Schwarz et al., 2008; Twohy et al., 2008; Moffet and Prather, 2009; Pratt and Prather, 2010) and is also known to significantly enhance the bulk absorption per mass of aerosols compared to external mixtures of black carbon (Ackerman and Toon, 1981; Horvath, 1993; Chýlek et al., 1995; Bond and Bergstrom, 2006; Kim et al., 2008; Khalizov et al., 2009). The mixing state of anthropogenic absorbing aerosols is thus a critical factor in determining their effects in both direct forcing and through microphysical connection or "indirect forcing". Therefore, the description of anthropogenic absorbing aerosols needs to be extended from size-dependent only to size- and mixing-state dependent (Fig. 1b). There are a few attempts already in recent years to include the mixing state as a new dimension to describe aerosols in the global climate models (e.g., Jacobson, 2006; Kim et al., 2008; Bauer et al., 2008). The model should include both external and internal mixtures of various constituents. The conversions between any two mixtures would only occur following certain microphysical or chemical relation. The preliminary results from these works have revealed a substantial difference between simulations with and without considering the mixing states. Note that there are many works in literature citing the internal mixing of aerosols in the models, however, most of these efforts are accomplished by assuming all the aerosol constituents within a given size increment in the model to be "internally mixed", i.e., without actually simulating the microphysical and chemical processes that make the conversion and growth of these mixtures possible. This is actually the approximation within a one-dimensional space of freedom (i.e., size; Fig. 1a) that attempts to mimic a two-dimensional space of freedom (i.e., size and mixing status; Fig. 1b).

The absorption strength of a core-shell mixture and the mass fraction of its absorbing core in total mass is nonlinearly correlated: a small fraction of 10–30% of BC would turn the entire core-shell particle to behave much like a pure BC particle in terms of absorption ratio in the total extinction (Ackerman and Toon, 1981; Chýlek et al., 1995; Bond et al., 2006; Kim et al., 2008). This suggests that if such mixing forms in a substantial quantity in the atmosphere, the forcing of the entire anthropogenic aerosol family to the climate system would shift to be much less negative, and in certain cases particularly over some regions could even become positive or near zero. This can be demonstrated by the following simplified calculation. At an atmospheric column, the aerosol optical depth, a critical dimensionless measure of aerosol solar extinction and thus radiative forcing, can be derived as:

$$\tau = \tau_S + \tau_A = \sigma_S M_S + \sigma_A M_A \quad (1)$$

Here τ represents the aerosol optical depth of aerosol, σ the mass extinction coefficient (m^2/g), M the column loading of mass (g/m^2), subscript S and A represent scattering and absorbing constituent, respectively.

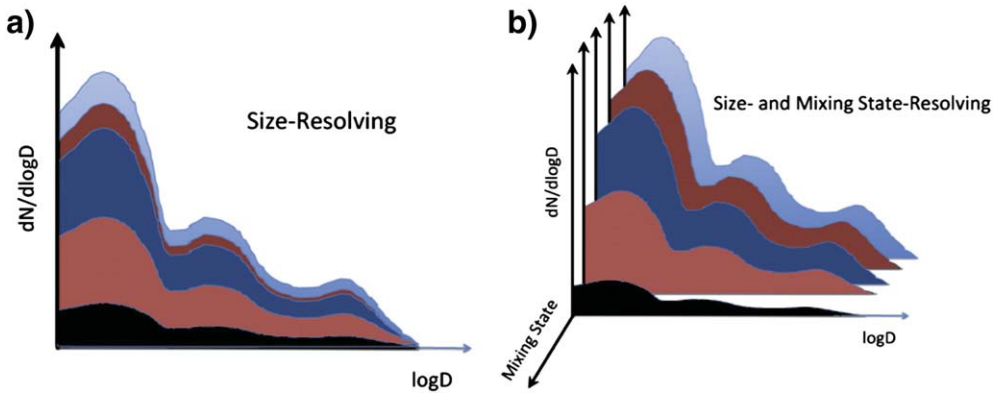

Fig. 1. A diagraph showing the aerosol model configuration based on either size only (a) or both size- and mixing-state (b). Different colors represent various types of aerosols in different mixing states.

Assuming now that the core-shell mixture is formed by taking a portion each from scattering and absorbing constituents, respectively, with a mass column loading of $\Delta M_S + \Delta M_A = \Delta M$, the change of total aerosol optical depth is thus:

$$\Delta\tau = \sigma_M \Delta M - \sigma_S \Delta M_S - \sigma_A \Delta M_A = \sigma_M \Delta M - \Delta\tau_S - \Delta\tau_A. \quad (2)$$

Now, for a given ΔM considering the absorbing core mass ratio $R = \Delta M_A / \Delta M$. The extinction coefficient σ_M and the single scattering albedo ω_M of the core-shell mixture are both nonlinear functions of R, and assuming the geometric mode size of 20 nm for the core-shell mixture at 500 nm wave band, they can be fitted based on Kim et al. (2008) as:

$$\sigma_M = -11.366R^2 + 23.956R - 2.9159 \quad (3a)$$

and

$$\omega_M = 0.2767R^{-0.225} \quad (3b)$$

here the fitting range of (3a) is $0.005 \le R \le 0.995$, and $0 \le \omega_M \le 1$.

For the same wave band, the mass extinction coefficient is given as 14.5 and 6.0 for absorbing and scattering components, respectively. The resultant changes in aerosol optical depth (AOD) and absorbing aerosol optical depth (AAOD) from an all external mixture case to an external plus core-shell mixture case are shown as varying highly nonlinearly with R in Fig. 2. Actually, when AAOD increases, the sign of TOA forcing of total anthropogenic aerosols could change from negative to positive regardless if AOD increases or not. This would even more likely to be the case when a bright surface such as cloud layer is beneath the aerosol layer. These all demonstrate that the direct radiative forcing of anthropogenic absorbing aerosols needs to be described with a consideration of mixing state. In fact, results of global aerosol-climate models using size- and mixing state-dependent aerosol model have demonstrated the difference introduced in direct radiative forcing of anthropogenic absorbing aerosols comparing to models with only external mixtures or well-mixed internal mixtures. For instance, Kim et al. (2008) found that climatologically the core-shell mixture of black carbon and sulfate along with the internal mixture of organic carbon (OC) and sulfate possess about 36%, 48%, and 32% of BC, OC, and sulfate mass in the atmosphere, respectively. The inclusion of these mixed aerosols in the model derives a much less negative TOA forcing of all carbonaceous and sulfate aerosol compounds compared to the cases excluding these. Of course the inclusion of such mixtures in the model also enhances the atmospheric heating by aerosols due to an increased absorbing particulate mass that should be largely counted as scattering matters otherwise.

3. Microphysical connection study

Black carbon aerosol is generally hydrophobic unless other matters contaminate its surface. Therefore, only after being coated by water-soluble matters such as sulfate can these aerosols affect the formation of clouds. Microphysical effects of anthropogenic absorbing aerosols on clouds and

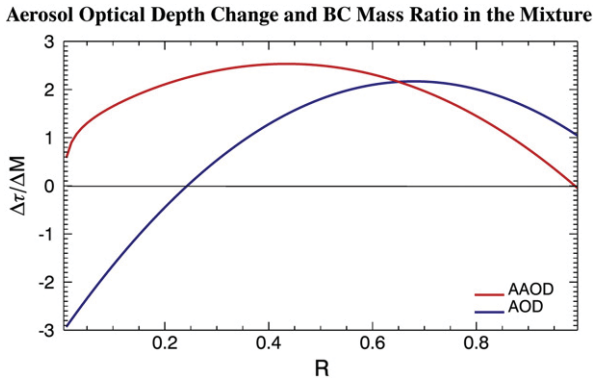

Fig. 2. Changes in aerosol optical depth and absorbing aerosol optical depth at 0.5 μm wavelength of anthropogenic aerosols due to the inclusion of core-shell mixture as function of the black carbon mass ratio in the mixture (R). $\Delta\tau$ is the optical depth difference between cases with and without the core-shell mixture in the model. ΔM is the total mass column loading of such mixture. Calculations are done based on Kim et al. (2008; Fig. 3) where all aerosol modes are assumed to have a log-normal size distribution, with the given geometric size as 0.02 μm for the core-shell BC-sulfate mixture, and 0.01, 0.02 and 0.05 μm for external mixtures of BC, OC, and sulfate, respectively.

precipitation are thus determined by the hygroscopicity of their shell matters besides the size of the particles.

There is no fundamental difference in enhancing or weakening of cloud radiative effects by hygroscopic absorbing aerosols than other aerosols through aerosol–cloud microphysical connection, that is by changing the number of CCN or IN first and then the cloud droplet number concentration (Koch et al., 2011). The outcome can be implied from general aerosol studies on so-called aerosol indirect forcing (e.g., Haywood and Boucher, 2000; Ramaswamy et al., 2001; Lohmann and Feichter, 2005). These microphysical effects could also extend to the formation and development of precipitation, possibly through suppressing the coalescence between cloud droplets to form precipitating drops (suppressing or delay onset of precipitation), or forming large quantity of small droplets and thus increasing the total condensed water content or the buoyance of the air parcel to speed up the uplifting and thus condensation growth (either enhancing or suppressing precipitation), with a potential consequence of forming a large amount of ice particles through freezing.

In estimating the impact of aerosols on precipitation it is important to consider dynamical feedbacks in different scales. In many cases, such feedbacks might be too strong to overshadow the signal initiated from changes in aerosols. The attribution of precipitation change to aerosol effect would thus become difficult particularly when the analysis is based on observations. Another character of microphysical effects of anthropogenic absorbing aerosols is that it is mainly confined to the aerosol-laden regions, i.e., a local effect.

It is worth indicating the progress toward introducing more advanced cloud microphysical algorithms such as the two-moment microphysics schemes for cloud droplets and ice crystals in the models (e.g., Lohmann et al., 1999, 2007; Ming et al., 2007; Morrison and Gettelman, 2008; Wang and Penner, 2010). These schemes connect aerosol and cloud processes with a size-dependent feature despite that many microphysical processes are still highly parameterized in these models. Aerosol-climate models equipped with these schemes would provide good frameworks to study the microphysics–dynamics feedbacks on global scale initiated by anthropogenic aerosols including absorbing aerosols. In addition, these schemes would still maintain certain computational efficiency of the global models in conducting ensemble and long integrations comparing to more sophisticated and computational demanding bin microphysics (cf. Tao et al., 2012).

There are a few unique characters associated with the microphysical effects of absorbing aerosols on clouds and precipitation that do not exist in the cases of other types of aerosols.

After activation or being captured through impaction scavenging by precipitating drops (a kinetic process where aerosol collides and then merges with a liquid or solid drop), the insoluble absorbing cores inside the cloud particles would likely stay intact. This forms a new core-shell structure of absorbing core with water shell and makes the cloud droplets more absorbing (Chýlek et al., 1984). This effect has been parameterized as a simple alteration to cloud optical properties based on the black carbon fraction inside cloud water content, and is found to cause a change in cloud albedo that is rather small globally while substantial over certain regions (Chuang et al., 2002; Li et al., 2011). Jacobson (2006) examined the global climate response to the absorption and scattering associated with BC cores within cloud and precipitation particles. This was achieved by allowing BC to enter cloud and precipitating drops through various microphysical and chemical processes, undergone other removing processes of BC by impaction scavenging, sedimentation, and dry deposition. Two methods of treating the optics of BC in size-resolving cloud liquid, ice, and graupel were compared: the core-shell approximation and the iterative dynamic effective medium approximation. Both methods led to a slightly increase of global surface temperature response. BC cloud absorption was also found to increase water vapor, decrease precipitation, and decrease cloud fraction.

Uncoated anthropogenic absorbing aerosols with lower hygroscopicity might have a good chance to survive the activation at cloud base, where the highest supersaturation in uplifted air parcel would likely occur and the aerosols with high hygroscopicity would be activated. Beyond this point, condensation growth of newly formed cloud droplets would thereafter lower the relative humidity to be just above or at saturation level so that low hygroscopic particles would have less chance to be activated. These aerosols would become interstitial aerosols above the cloud base and thus continually be transported into higher atmospheric layers (Engström et al., 2008). The aerosols escaped from activation at cloud base would have another opportunity to affect the formation of cloud particles, mostly in ice phase, by serving as ice nuclei in the upper atmosphere (cf. DeMott et al., 1999; Gorbunov et al., 2001; DeMott et al., 2010). This effect has been explored in a cloud-resolving model study (e.g., Ekman et al., 2007) with limited observational support. Ekman et al. (2007) found that by allowing interstitial BC aerosols to become IN, it would generally increase the total number of aerosols for heterogeneous nucleation in the upper tropospheric portion of a deep convective cloud (their model actually included prognostic aerosol number concentration considering both nucleation and impaction scavenging). This increase of IN could generate higher updraft velocities and a subsequent and rather short delay in the onset of heavy precipitation. The higher updraft velocities would also affect the homogeneous nucleation rate, prolong the precipitation event and result in, on average, a higher total precipitation amounts compared with the control case. However, this impact is found to be non-monotonic with IN increase, and also less effective comparing to the increase of CCN. Besides by the vertical transport, BC can enter the upper troposphere through direct emissions of aircrafts as well. Hendricks et al. (2005) estimated the potential effect of BC from this source on cirrus clouds, by allowing all aircraft emitted BC to become IN and actually form ice crystals in a global climate model, they found that aircraft emissions might account for 10–60% in total ice crystal concentration, thus are not a negligible factor.

Chen et al. (2010) have assessed the global scale effect of two hypothetical mitigations of black carbon aerosol by a decrease in particle number emissions, and thereby by a decrease in global CCN concentrations (a reverse “cloud albedo effect”): one with 50% reduction of primary black carbon/organic carbon mass and number emissions from fossil fuel combustion, and another with 50% reduction of

primary BC/OC mass and number emissions from all primary carbonaceous sources (fossil fuel, domestic biofuel, and biomass burning). They found a decrease in global cloud radiative forcing in responding to these two scenarios.

4. Local dynamical effects

Persistent existence of anthropogenic aerosols in the atmosphere will exert an external forcing to the climate system, and in many cases a perturbation to certain dynamical features. This would generate a coupling between aerosol forcing and perturbed dynamical features. Likely consequences include an impact on the formation of clouds and precipitation either locally or remotely. For the absorbing aerosol, the forcing to the atmosphere is implemented through both atmospheric heating and surface cooling.

One of the local effects of such a coupling between anthropogenic absorbing aerosols and dynamics is the perturbation of vertical thermodynamical profile through atmospheric heating by these aerosols. Depending on the vertical location of the aerosol layer this effect can stabilize or destabilize atmospheric layers ranging from the planetary boundary to the free troposphere (Hansen et al., 1997).

The optical/thermodynamical perturbation to the planetary boundary layer (PBL) in many conditions might not be strong enough to cause a long lasting change but daily variation of low clouds due to the solar diurnal cycle. Ackerman et al. (2000) used a large-eddy simulation (LES) model to demonstrate that during the winter monsoon season over the Indian Ocean, solar absorption by aerosols can reduce daytime cloud coverage by nearly half in an observed case of trade cumulus. Consequently, the reduction of cloudiness exerts a positive radiative forcing at the TOA that partially offsets the direct aerosol forcing and the conventional indirect forcings. However, several other factors including meteorological conditions such as dryness could cause the similar cloudless situation during the observation period. The authors admitted that a lack of clouds could also be largely due to the dry air from the Indian subcontinent, and the absorbing aerosol effect just served to diminish cloud cover even further.

Johnson et al. (2004) have also studied whether absorbing aerosols could cause a decrease in low marine cloud cover and liquid-water path (LWP) using a LES model with only direct radiative effects of absorbing aerosols. The study found that when absorbing aerosol layer locates within the boundary layer, it would cause a decrease in LWP and thus lead to a positive cloud forcing (due to the less cloudy condition) that is about three times larger than the aerosol direct forcing that initiated this effect. When the absorbing aerosol layer situates above the cloud, the LWP was found to increase and thus leads to a negative cloud forcing.

Interestingly, Feingold et al. (2005) found that if smoke aerosols exist close enough to the ground, the heating of these absorbing aerosols could make planetary boundary layer to become unstable and thus stimulate convection.

These studies generally propose the atmospheric heating rather than the surface cooling effect of anthropogenic absorbing aerosols to be the main ignition in the local aerosol-dynamics coupling. Such heating is hypothesized to decrease the relative humidity and to be less favorite to the formation of cloud should the microphysical connection is concerned, and to cause stabilization to further suppress the formation of cloud or, should aerosols situate close enough to the ground to destabilize the PBL, causing the cloud to form. In whichever case, the cloud radiative forcing change caused by the aerosol would easily exceed that of the direct forcing of aerosols itself.

Using satellite retrievals including cloud fraction, AOD, and cloud top pressure, Koren et al. (2008) suggest that the cloud invigoration would have been mostly done by aerosol through microphysical processes, especially when AOD is low (cf. Andreae et al., 2004). Only when AOD is high enough (assuming AAOD increases with AOD) to exert significant dynamical perturbation could the dynamical effect through aerosol absorption suppress cloud development (Fig. 3). This hypothesis has been supported by a recent study using both global model and MODIS retrievals by Ten Hoeve et al. (2012).

Note that the variability of clouds could be affected by many factors besides aerosol forcing. The issue still exists on attributing the variability of clouds to the local dynamical effect of aerosols at current stage.

When such concept of local dynamical effect is developed further to connect to large-scale dynamics, it would be expected that correlated changes in low clouds could occur in different regions but with opposite sign as indicated recently by Persad et al. (2012). The study offered a view into the coupling of atmospheric heating of anthropogenic absorbing aerosols and large-scale uplift and subsidence. The authors found that over the convective regions, for instance the West Pacific warm pool (WPWP; Fig. 4), heating in the free troposphere hinders the vertical development of deep cumulus clouds, resulting in the detrainment of more cloudy air into the large-scale environment and stronger cloud reflection. A different mechanism operates over the subsidence regions, for example the South Pacific subsidence region (SPSR; Fig. 4) where heating near the top of boundary layer causes a substantial reduction in low cloud amount by decreasing relative humidity and by lowering cloud top. Overall speaking, the study suggests the large reductions in low cloud amount over both uplifting and subsidence regions (Fig. 4). Again, the resulting cloud forcing significantly exceeds the initial direct forcing of aerosols. This study along with a few others advances current understanding of the effect of absorbing aerosol on low clouds, from considering the effect of aerosol heating on thermodynamical profile alone to including the dynamical adjustment on the height of PBL along with thermodynamical effects associated with large-scale vertical motion.

Snow albedo effect also reflects a coupling of absorbing aerosols with local dynamical processes. Several representative studies addressing this issue used global climate models, the net effect reported, however, was mostly global arithmetic average in radiative forcing. The effects on large-scale dynamics were not well defined. Jacobson (2004) explored the effect of black carbon on changing modeled spectral albedo and emissivity over snow and sea ice in a global climate model. BC was calculated to reduce snow and sea ice albedo by 0.4% in the global average and 1% in the Northern Hemisphere. A positive response in surface temperature change was found in the case of including such an effect in

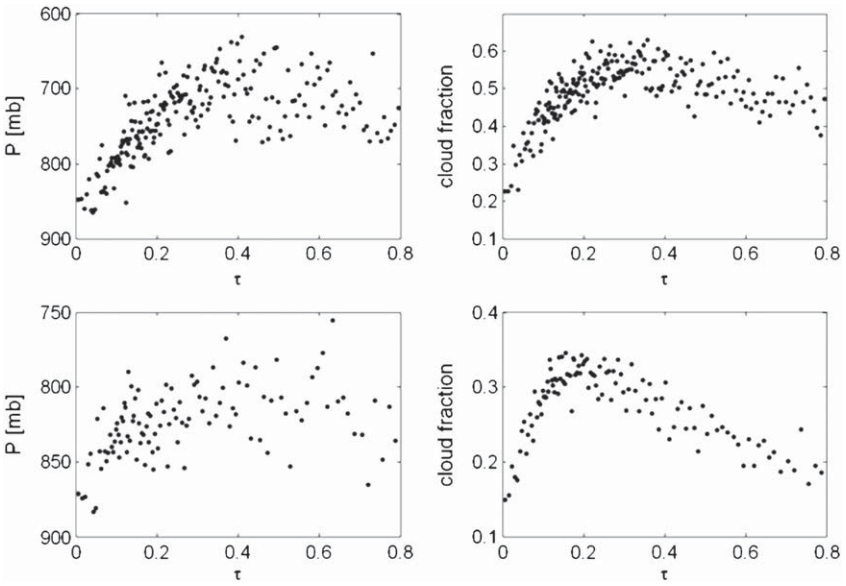

Fig. 3. Relationships between cloud properties and aerosol loading estimated using aerosol optical depth, τ. (Left panels), cloud top pressure P versus τ. Lower P may indicate taller convective clouds that reach to higher levels of the atmosphere. (Right panels) cloud fraction versus τ. The upper row shows all data and the lower row shows data restricted to a cloud fraction <0.5. From Koren et al. (2008). Copyright AAAS. Reprinted with permission.

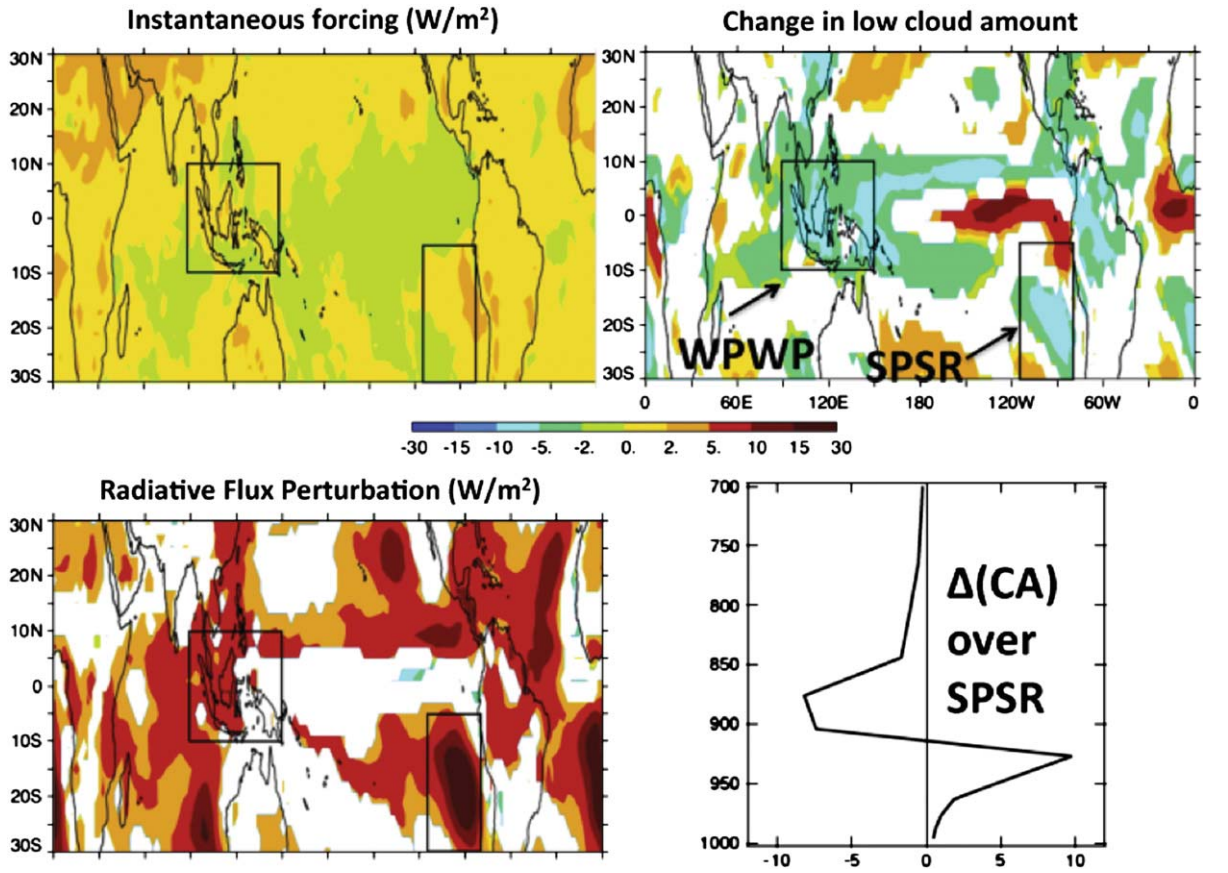

Fig. 4. Annual-mean (upper left) instantaneous forcing (W/m^2), (lower left) radiative flux perturbation (W/m^2), and (upper right) change in middle cloud amount (%) due to boundary layer BC. The rectangular boxes denote the West Pacific warm pool (WPWP) and South Pacific subsidence region (SPSR), respectively. Also shown is the vertical distribution of change in cloud amount over SPSR (lower right). From Persad et al. (2012). Copyright American Meteorological Society.

the model, implying that the warming due to soot absorption in snow and sea ice (in shortwave) might have offset reduced warming due to lower emission from darker snow and ice (in longwave).

Flanner et al (2007) also estimated climate forcing and response from black carbon in snow along with uncertainty in forcing estimates. They found that BC emissions are still the largest source of uncertainty in estimating BC snow effect, followed by snow aging. The rate of snow aging determines directly the snow reflectance and the magnitude of albedo change caused by BC. By the contrasting results of a strong and a weak boreal fire year, the researchers derived a net positive forcing caused by the BC snow effect with a majority (at least 80%) of contribution comes from anthropogenic absorbing aerosols. The result seems also suggesting that the BC/snow forcing is more effective than forcing by CO_2 in causing surface temperature change, consistent with earlier estimation by Hansen and Nazarenko (2004). Such an effect could occur in several climatologically sensitive regions including the polar area and the Tibetan Plateau. A recent study suggested that the heating brought by anthropogenic and natural absorbing aerosols built up against the foothill of Tibetan Plateau could exert a positive feedback mechanism to speed up snow melt (Lau et al., 2010). It seems a necessity to include the snow and ice albedo effect due to black carbon in future studies so that the coupling with dynamics and associated impacts on clouds and precipitation could be further examined.

The potential impact on cloud dynamics by interstitial anthropogenic absorbing aerosols along with the absorbing cores inside cloud droplets still needs to be carefully examined, perhaps first in process level with high resolution and then in global scale.

5. Large-scale dynamical effects

Besides local dynamical effect, persistent forcing of anthropogenic absorbing aerosols can also change large-scale atmospheric circulation and thus propagate the aerosol impact downstream to regions even distant to the aerosol-laden areas. Clouds and precipitation influenced by the associated dynamical system can be modified through a remote impact or teleconnection due to dynamical system changes rather than direct on-site aerosol effect (e.g., Wang, 2004). The key for such a "dynamical path" to work is whether the aerosol effect can form a persistent perturbation on certain dynamical features. Results have demonstrated that when such path is established, aerosol-induced perturbation in clouds and precipitation can be distributed more widely and has more significant strength in comparison to the local effects whether through microphysical path or local dynamical coupling.

From a global-mean viewpoint, the net positive TOA forcing by anthropogenic absorbing aerosols leads to a surface temperature increase and this would generally create a positive response in precipitation. Ming et al. (2010) demonstrated through GCM simulations that the atmospheric heating by absorbing aerosols could suppress global-mean precipitation and, more interestingly, outweigh the enhancing effect through surface warming, resulting in a net decrease in precipitation.

Nevertheless, the above argument does not imply simple patterns of regional cloud and precipitation changes caused by absorbing aerosols. In fact, global model simulations have repeatedly indicated that the global mean precipitation and cloud coverage change caused by absorbing aerosols are mostly insignificant, while the shift of regional patterns along with the magnitude of accompanying changes, however, is much stronger in comparison. For example, studies using different GCMs all indicate that the direct radiative forcing of black carbon can lead to a northward shift of precipitation in the inter-tropical convergence zone (ITCZ), especially over the Pacific Ocean (Wang, 2004; Roberts and Jones, 2004; Chung and Seinfeld, 2005). Ramaswamy and Chen (1997) and Rotstayn and Lohmann (2002) both found that the inclusion of aerosol indirect forcing in their models could force a similar shift in ITCZ precipitation but in opposite direction.

Research works aiming to understand the effects of aerosols on precipitation through coupling with large-scale dynamics have increased steadily. These efforts are often benefited from knowledge about connected dynamical systems themselves. For instance, aerosol forced shift of ITCZ convection and precipitation is consistent with the findings from the simulations studying the climate response to idealized hemispheric forcing (Broccoli et al., 2006; Kang et al., 2008). In some other cases, different interpretations to the associated dynamical features and sensitivity might bring contradicting opinions regarding aerosol effects. An excellent example is seen in recent works on the impact of anthropogenic absorbing aerosols on the Indian summer monsoon.

The impact of aerosols, particularly absorbing aerosols, on the Indian summer monsoon has become a recent research focus, represented by rapidly increasing publications in literature (Ramanathan et al., 2001; Chung et al., 2002; Ramanathan et al., 2005; Lau et al., 2006, 2008; Meehl et al., 2008; Randles and Ramaswamy, 2008; Wang et al., 2009b; Collier and Zhang, 2009; Krishnamurti et al., 2009; Manoj et al., 2010; Ganguly et al., 2012a, 2012b; Shindell et al., 2012; among others). All these analyses come to a conclusion that the forcing of anthropogenic absorbing aerosols over the Indian subcontinent particularly during the premonsoon season (normally May to June, corresponding to the start of heavy rainfall over land) is strong enough to cause significant changes in monsoonal rainfall. Averaged over the Indian land area, most studies suggest an aerosol-caused decrease in rainfall during the summer monsoon season and an increase during pre-monsoon and onset seasons. Analyses looking into detailed pattern change found the decrease of rainfall mostly occurs over most of the central and south Indian subcontinent while increasing in other regions such as northwestern India. Additionally, these aerosol-caused changes in the monsoonal circulation could further affect other dynamical features, e.g., tropical cyclone intensity over nearby Arabian Sea as suggested by a recent study of Evan et al. (2011).

Opinions to explain these results differ, however, because of the viewpoints emphasized on different driving factors of the monsoon system. Ramanathan et al. (2005) emphasized on the dimming (surface cooling) effects of anthropogenic absorbing aerosols and suggested that this effect could decrease the meridional sea surface temperature gradient, one of the driving force of the Indian summer monsoon, and

subsequently the surface evaporation. These two factors would reduce the monsoonal circulation and rainfall amount. Lau et al. (2006) proposed to consider the effect of heating by absorbing aerosols on the slope of the Tibetan Plateau, and suggested that such an effect could initiate a positive feedback by drawing water convergence from oceans first and then form condensation and thus further heat the air over these elevated places. Wang et al. (2009b) looked into the large-scale stability issue driving the monsoon system and indicated that anthropogenic absorbing aerosols, acting alone or together with scattering aerosols, can alter the meridional gradient of moist static energy (MSE) in the sub-cloud layer over the Indian subcontinent. This changes the large-scale atmospheric stability that drives the deep convection in particular during onset period and forces a north- and northwestward shift of convection and associated heavy rainfall, which is coincidently in general agreement with the observed monsoon precipitation changes in recent decades. As the new simulations include not only the direct but also indirect effects of aerosols, precipitation pattern change becomes even more complicated, for example, Ganguly et al. (2012a) suggest that the indirect effect brought mainly by water-soluble anthropogenic aerosols (mostly scattering in their configuration) would enhance the surface cooling effect through cloud radiation changes due to an increase in cloud droplet number concentration.

A critical factor regarding the aerosol–monsoon effect, i.e., the ocean response is still somewhat overlooked in current studies. As a matter of fact, some of the differences in precipitation pattern change caused by aerosols might have been introduced by different ocean configuration in modeling, some studies used fully coupled ocean GCM or slab ocean model, while others used prescribed sea surface temperature. The latter configuration definitely suppresses the oceanic response to atmospheric perturbation while different ocean models favor responses with different frequencies.

Such an issue has been addressed by a most recent work by Ganguly et al. (2012b) in a wider scope, where the authors compared the fast and slow responses of the South Asian monsoon system to anthropogenic aerosols. They have found that the slow response, mostly due to the SST changes caused by aerosol effect, would dominate the precipitation changing patterns over the Indian subcontinent land area south of 25°N. On the other hand, certain changing patterns including zonal and latitudinal asymmetric distributions of land precipitation changes are mostly determined by the relatively fast response of the monsoon system to the local radiative effect, cloud feature change, and land surface effect.

To matching geographically a given climate response with causal anthropogenic forcing is an interesting while still challenging topic because of the limited capability of current GCMs in revealing detailed regional response patterns (Shindell and Faluvegi, 2009). Nevertheless, among a limited number of such efforts, certain precipitation response to the direct radiative forcing of anthropogenic absorbing aerosols is found to depend more on the forcing location rather than absolute quantity, as demonstrated in Wang (2009). In that study emissions of black carbon from five continent-scale source regions were included separately in different GCM simulations coupled with mixed layer ocean model, in addition to a model run that included all the emissions from these continental scale sources. The derived zonal-mean precipitation changes from the long equilibrium integrations that each only containing the forcings of BC from one given region exhibit a nonlinear dependency on the emission quantity, where the arithmetic summation of these individual precipitation changes is much larger than the actual precipitation change derived from the run that included all the regional emissions (Fig. 5). Recently, Teng et al. (2012) have successfully captured the correlation of temperature change over North America with black carbon aerosol forcing over Asia in a set of simulations using coupled GCM with prescribed forcing. Further work is needed to connect given patterns in precipitation change with their corresponding forcings over different regions around the globe.

6. Summary and discussions

The research community of aerosol-climate interaction has witnessed a substantial progress in identifying the impacts of anthropogenic absorbing aerosols on clouds and precipitation. In fact, a new research field is gradually forming, aiming to understand the effect of anthropogenic absorbing aerosols on clouds and precipitation particularly through a coupling between persistent aerosol forcing and dynamical features in different scales, and to isolate the aerosol effects from other factors including the forcings of other aerosol constituents and from certain natural variabilities. Studies have demonstrated that by simply perturbing thermodynamical and dynamical features in different scales without even altering the microphysical structures of clouds on-site, anthropogenic absorbing aerosols could cause significant changes in clouds and precipitation of many critical systems including the ITCZ and the monsoons. The impacts are often appearing remote to the aerosol-laden regions.

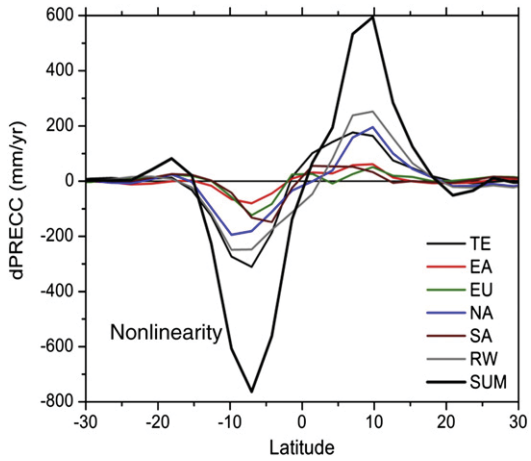

Fig. 5. Zonal means of changes in convective precipitation (mm/year) caused by BC, derived from various model run. Data shown are year 41–60 means. EA, EU, NA, SA, and RW denote the result of simulation that only includes BC emissions from East Asia, Europe, North America, South Asia, and rest of the world. TE denotes the result from the model run including emissions from all continents. SUM represents an arithmetic summation of the results of all regional emission runs, i.e. SUM = EA + EU + NA + SA + RW. The difference between SUM and TE thus reflects the nonlinearity of precipitation response to emission amounts. From Wang (2009).

An obvious benefit for the researches toward this new direction is the rich knowledge base of general circulation theory and tropical meteorology, among others. Lesson can be learned when an analogy is established between non-aerosol forcings in certain dynamical processes and aerosol forcings. Such examples including the progresses made by taking advantage from previous findings in teleconnection, ENSO, and hemispheric forcing-response to better understand the remote impact of absorbing aerosol on atmospheric circulation, and from quasi-equilibrium hypothesis to explain the impact of absorbing aerosols on the monsoon precipitation distribution.

In each aspect of the efforts from estimating emissions, field measurement and laboratory experiments, to modeling, uncertainties still exist. Many of such uncertainties might never be completely eliminated. Therefore, the critical step to move the field forward is to identify and then to narrow these uncertainties as much as possible while carrying out analyses that take these uncertainties into account.

It would serve the best purpose to conduct synergetic research to fill several critical gaps. One of these gaps is the lack of connection between the so-called bottom-up or process-based studies and the top-down or macro-constrained studies, especially toward estimating emissions or evaluating modeled aerosol properties in the atmosphere. A combination of the bottom-up and top-down approaches in a same framework could minimize potential problems caused by the incomplete survey rooted in the bottom-up approach, while improving the missing geographical distribution in the top-down approach due to the lack of well-covered observational data. As a matter of fact, various so-called inverse modeling efforts, or the top-down approaches, have been applied mainly to optimize aerosol emissions based on modeled and observed aerosol properties in the atmosphere (e.g., Collins et al., 2001; Zhang et al., 2005; Dubovik et al., 2008). However, such efforts are still limited by the availability of aerosol observational data, same as to the work in detection and attribution of temperature and precipitation changes by aerosols (e.g., Jones et al., 2011).

It could also add value from combining sophisticated process models and abstract or theoretical modeling to connect the findings in different scales (e.g., Koren and Feingold, 2011). Process based cloud-resolving and large-eddy simulation models with aerosol have been used to study detailed aerosol–cloud-precipitation in local scale (e.g., Feingold and Kreidenweis, 2000; Ekman et al., 2007). However, due to the lack of information of feedback from and to large-scale, the results from such studies have not been extensively used to improve large-scale models. With the assistance of abstract modeling, results from detailed studies could be possibly generalized for global models to use, or at least to test for a much less computational cost. In addition, certain sophisticated process models describing the processes in scales beyond the reach of current global climate models can be used to derive the so-called reduced form models or emulators (e.g., Tatang et al., 1997). These reduced-form models can be then used in global models as a parameterization for subgrid or fast processes or in uncertainty analysis with high computational efficiency. An example in aerosol-climate model application of such effort is Cohen et al. (2011), where the fast aerosol physical and chemical processing was parameterized based on an air-quality model and such derived reduced-form model was incorporated in a global aerosol-climate model.

Ultimately, the aerosol–cloud-precipitation connection and its climate effects need to be examined in the global frameworks. A task specifically in connecting aerosol-dynamical linkages is thus to study the modulations by various natural variabilities or dynamical features on aerosol-induced perturbations. Such knowledge is critical to understand the climate response to aerosol forcings, however, related effort is still rare.

Interstitial aerosols are likely to be absorbing due to their inferior surface property that makes them hardly to serve as good CCN or even IN. Such aerosols inside the cloud layer, aided by the absorbing cores inside liquid or solid cloud particles, could exert additional forcing to the atmosphere (e.g., Jacobson, 2012). However, whether such forcing could be compensated by cloud dynamical feedback still remains an open question. Process studies based on high-resolution models and in-situ measurements would have to lead the way to address this issue. In addition, the nucleation and impaction scavenging of anthropogenic absorbing aerosols needs to be treated more carefully considering their mixing state and shell property that varies with air humidity change. It is likely that these efforts would encounter the same issues in dealing with cloud radiation change by water-soluble aerosols as described in general literature (e.g., Rotstayn and Penner, 2001; Wang and Penner, 2009; Hoose et al., 2009; Koch et al., 2011). However, due to the relatively low number concentration, perhaps additional radiative effect from inside cloud droplets and the effects of interstitial particles particularly in the upper troposphere would be more important than the so-called indirect radiative forcing.

Much of our knowledge gained so far about the climate impacts on clouds and precipitation of anthropogenic absorbing aerosols is based on the equilibrium climate responses, i.e., from model simulations using constant forcing/emissions to force the GCMs reach equilibrium after a long integration. Future studies using transient simulations, built upon ensemble configuration to consider various uncertainties and aided by attribution tools, are also needed to provide such information in the format of time varying descriptions of probability of given climate responses. This is for the purpose to not just advance our science but also provide insights for relevant policy making procedures looking into different time frames (e.g., UNEP, 2012).

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