Effect of model errors in ambient air humidity on the aerosol optical depth obtained via aerosol hygroscopicity in eastern China in the Atmospheric Chemistry and Climate Model Intercomparison Project datasets

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ABSTRACT

This analysis of the multi-model aerosol optical depth (AOD) in eastern China using the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) datasets shows that the global models underestimate the AOD by 33% and 44% in southern and northern China, respectively, and decrease the relative humidity (RH) of the air in the surface layer to 71%–80%, which is less than the RH of 77%–92% in reanalysis meteorological datasets. This indicates that the low biases in the RH partially account for the errors in the AOD. The AOD is recalculated based on the model aerosol concentrations and the reanalysis humidity data. Improving the mean value of the RH increases the multi-model annual mean AOD by 45% in southern China and by 33% in June–August in northern China. This method of improving the AOD is successful in most of the ACCMIP models, but it is unlikely to be successful in GISS-E2-R, in which the plot of its AOD efficiency against RH strongly deviates from the rest of the models. The effect of the improvement in the modeled RH on the AOD depends on the concentration of aerosols. The shape error in the frequency distribution of the RH is likely to be more important than the error in the mean value of the RH, but this requires further research.

1. Introduction

According to the multi-model results from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), the model biases in the aerosol optical depth (AOD) range from −30% to 20%. Most of these biases are lower than those in observational AOD datasets from satellites because of the absence of nitrates or secondary organic aerosols (SOAs) in some of the models (Shindell et al. 2013) and the low biases in the sulfate concentrations in winter and in the concentration of organic aerosols throughout the year (Chang et al. 2018). The concentration biases are relevant to the uncertainties in inventories of anthropogenic emissions (Chang et al. 2015; Fan et al. 2018) and the limitations of models in describing the aerosol chemistry of winter urban haze in northern China (Chen et al. 2016; Huang et al. 2014; Zheng et al. 2015).

In addition to aerosol concentrations, meteorological conditions also influence the AOD. Eastern China is a typical monsoon region and the AOD is higher in summer than in winter because the humid summer air increases the size of particulates via the uptake of water (Bian et al. 2014; Liu et al. 2011; Li et al. 2013; Zheng et al. 2017; Zhao et al. 2018; Zhu et al. 2019). In winter, the uptake of water by aerosols plays an important part in the growth of secondary aerosols on haze days (Chen et al. 2016; Wang et al. 2014; Zheng et al. 2015). The hygroscopicity of sulfate, the main anthropogenic aerosol, can be simulated using the variant parametric formulas of Köhler theory (Chin et al. 2002; Fitzgerald 1975; Ghan and Zaveri 2007; Tang and Munkelwitz 1994; Zhang et al. 2014). The particulate hygroscopic growth factor (i.e., the ratio of the wet particulate radius to the dry particulate radius) is a quadratic function of the relative humidity (RH) of the ambient air based on
chamber studies (Fitzgerald 1975; Tang and Munkelwitz 1994). The particulate size dramatically increases under conditions of high RH (e.g., RH >80%). It is difficult to precisely reproduce the Asian monsoon using climate models (Spärber et al. 2013) and the bias in the RH partially accounts for the biases in modeling the hygroscopicity of aerosols and the AOD.

This study assesses the influence of the ambient RH on the AOD in eastern China in the ACCMIP models from the perspective of the hygroscopicity of aerosols. The effects of the RH on the AOD reported here are not comprehensive because the effect of the RH on aerosol chemistry is not concerned because of the limitations of the ACCMIP dataset, but it is meaningful to illustrate the adverse impacts of the model bias in the RH on the simulation of the AOD.

2. Data and methods

The analysis is based on aerosol simulations from nine ACCMIP models for the year 2000 downloaded from the British Atmospheric Data Center (http://badc.nerc.ac.uk). The nine models are all two-way coupled chemistry-climate global models, except for CICERO-OsloCTM2, which is a chemistry transport model. Most of the models simulate the mass of aerosols using a bulk approach in which the size distributions of aerosols are prescribed. GISS-E2-R-TOMAS couples Two-Moment Aerosols Sectional (TOMAS) aerosol microphysics to simulate the aerosol number and mass size distributions. NCAR-CAM5.1 uses three internally mixed log-normal modes to represent the size distributions of aerosols. Aerosol indirect effects are represented in CICERO-OsloCTM2, GFDL-AM3, GISS-E2-R, GISS-TOMAS, HadGEM2, MIROC-CHEM, and NCAR-CAM5.1. More details of the models have been reported in the ACCMIP overview paper of Lamarque et al. (2013). The ACCMIP datasets contain the multi-model mean aerosol concentration. The high summer aerosol concentration is determined in mainland China (Figure 1), with annual biases of −44% and −33% in North and South China, respectively. The individual model errors in the two subregions range from −64% to 2%. The multi-model mean AOD shows seasonal variation, in agreement with the MODIS data (Figure 2). The high summer AOD in northern China is due to the large hygroscopic growth of hydrophilic aerosols during the southerly
wet monsoon (Li et al. 2016). The low model AOD from March to May is due to the low dust AOD, which is a common model error for uncertain dust emissions (Pu and Ginoux 2018). In winter, the low AOD is due to the lack of nitrates and SOAs in some of the models (Shindell et al. 2013). Additionally, the model chemistry is unable to reproduce the strong growth of sulfate during haze days in urban regions (Chen et al. 2016; Huang et al. 2014; Zheng et al. 2015), which leads to strong low-AOD in northern China even though the RH bias there is not as high as that in southern China.

The models underestimate the RH in the surface layer. According to the reanalysis dataset, the surface-layer RH ranges from 77%–92% throughout the year in southern China and 81% in July–August in northern China. Correspondingly, the models produce a lower RH of 71%–80% in the surface layer in southern China, and 71% in northern China, and extend the low bias into the upper layers.

The adverse effects of the RH errors to the AOD can be recognized by the hygroscopicities of sulfates and nitrates. Organic aerosols make up another important portion of the AOD, but its AOD error is a result of the low bias in the concentration of organic aerosols caused by the limitations of the organic chemistry in the models (Tsigaridis et al. 2014). Apart from GISS-E2-R and GISS-E2-R-TOMAS, the other models predict sulfate hygroscopicity using a variant parametric formula from Köhler theory, which are reproduced here in a FORTRAN program and the reproduced Köhler curves are shown in Figure 3(a). The two GISS models handle the hygroscopicity of aerosols in different ways. GISS-E2-R

Figure 1. Annual surface-layer RH (%) and AOD in the multi-model mean values (except CESM-CAM-superfast) for 2000 and their differences with GEOS4 reanalysis data and MODIS data for 2000–04. The frames in (c, f) mark the ranges of northern (31°–41°N, 112°–122°E) and southern (21°–30°N, 108°–122°E) China.

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parameterizes the wet aerosol size to reproduce precisely the extinction efficiency and asymmetry parameters of the solute aerosols at a laboratory wavelength of 633 nm (Schmidt et al. 2006). GISS-E2-R-TOMAS simulates the uptake of water by sulfate using a polynomial fit based on the results of the thermodynamic equilibrium model ISORROPIA (Lee, Adams, and Shindell 2015). It is hard to reproduce the hygroscopic growth curves in the two GISS models because of a lack of published data.

Figure 2. Annual cycles of the AOD and the surface-layer RH (%).

Figure 3. (a) Hygroscopic growth factors (GWF) of surface particulates plotted against RH in the ACCMIP models. (b) Probability distribution functions (PDF) of the RH in the surface layer in the reanalysis dataset at 0600 and 1200 GMT June–August 2000–04. (c) PDF of sulfate AOD corresponding to the RH in panel (b). The sulfate AOD is calculated for a dry particulate with a normal distribution of 0.05 ± 2.0 µm, a dry particulate density of 1.769 g cm$^{-3}$, and a column burden of 41 mg m$^{-2}$. 
Figure 3(a) shows the reproduced hygroscopic growth factor for sulfate, which is the ratio of the wet particulate radius to the dry radius as a function of the ambient RH value in the ACCMIP models. The variant formulas give roughly equivalent quadratic growth curves at high RH values, showing a growth factor of 1.4–1.5 for 70%–80% RH and 1.5–2 for 80%–95% RH. The variant implementations of Köhler theory are therefore unlikely to yield a substantial difference in the AOD for sulfate. The models underestimate the RH of the surface layer beyond 70%, which is the crucial RH value for accelerated hygroscopic growth of sulfate.

3.2. Improvements in AOD as a result of changes in the mean RH

Figure 2 shows that the baseline AOD is lower than the MODIS AOD for the similar causes to the original model results (e.g., low dust in spring and low anthropogenic secondary aerosols in winter), and thus the annual cycle in the baseline resembles the original model mean result. The baseline AOD is lower than the original model AOD because the monthly model results smooth the high concentrations. In southern China, as improving the mean value of the RH in the EXP2 experiment from the models (the multi-model mean value is 76%) to the reanalysis data (84%), the multi-model mean AOD in EXP2 is improved in each month, with an annual increase of 45% (Figure 2(c)). In northern China, the model RH is often less than 70% (Figure 2(b)) and the annual RH approaches the value in the reanalysis dataset. As a result, the increase in the AOD in EXP2 in northern China is only noticeable (33%) from June to August (Figure 2(a)).

The effectiveness of the RH correction is dependent on two factors. First, the high aerosol concentrations yield a strong improvement in the AOD. Figure 4 shows the difference in the surface-layer RH and the total AOD between the BASELINE and EXP2 results and the ratio of the differences, which denotes the AOD anomaly yielded by one unit change in the RH value. A high ratio is related to the abundance of sulfates. For example, the two GISS models show almost similar increases in the RH, but GISS-E2-R-TOMAS shows a greater increase in the AOD at high sulfate concentrations. The nitrate AOD in GISS-E2-R shows only a small response to the RH improvement because the nitrates in GISS-E2-R are more abundant in winter (Chang et al. 2018) when the RH is low. GFDL-AM3.1 and NCAR-CAM5.1 show higher and lower improving ratios, respectively, consistent with their sulfate burdens. In northern China, NCAR-CAM3.5 shows a slight positive change in the RH and a noticeable increase in the AOD. This model shows opposite changes in the RH between June and July–August. With the accelerating growth in particle size under high RH conditions, the increase in the AOD in July–August overwhelms the effect of the RH reduction in June.

Another factor affecting the response of the AOD is relevant to the RH frequency distribution. To confirm this, an idealized sulfate AOD is calculated for the multi-model mean sulfate column burden of 41 mg m⁻² in southern China under the 6-h RH in the same area at 0600 GMT June–August 2000–04. Figure 3(b,c) show that when the mean value of the RH is intentionally decreased from 73% to 63%, the corresponding mean value of the AOD decreases from 0.44 to 0.38, which is a decrease of 14%. AOD values larger than 0.5 are not observed.

In addition to the RH mean value, the shape of the RH distribution affects the AOD. For example, as a result of the low air temperature at night, the RH at 1200 GMT has a high mean value and a strong negative skewness (more extremely high values) than that at 0600 GMT (Figure 3(b)). The mean RH increases from 73% at 0600 GMT to 81% at 1200 GMT. Correspondingly, AOD values larger than 0.5 are more abundant at 1200 GMT (Figure 3(c)), with an increase in the mean AOD of 25%, higher than the change in the AOD (14%) due to the almost equivalent change in the RH mean value alone.

Urban haze often accompanies conditions of high humidity during winter, in which active heterogeneous reactions favor the strong growth of secondary particulates (Chen et al. 2016; Huang et al. 2014; Wang et al. 2014; Zheng et al. 2015). The radiative feedback of aerosols increases the RH of the surface layer (Gao et al. 2015), which tends to yield a higher instantaneous RH in extremely high tails. Therefore, the models underestimate the intensity of haze, weaken the radiative feedback of aerosols to the ambient RH, and presumably lower the AOD relevant to hygroscopic growth. Unfortunately, further evaluation of the shape of the RH distribution is difficult based on monthly results. It is suggested that if the models can be developed to give better aerosol concentrations on haze days, then the effect of improving the RH on the AOD results could be more pronounced.

3.3. Sulfate AOD efficiencies

Because hydrophilic particulates have equal opportunities to grow as the RH increases, the growth curve for a single particulate is reflected in the sulfate AOD efficiency — that is, the ratio of the sulfate
AOD to the sulfate column burden. Figure 5 shows the sulfate AOD efficiency plotted against the mean ambient RH values below 850 hPa in June–August in the original model results. Apart from GISS-E2-R, the models show a positive relationship, which suggests the sulfate AOD efficiency will possibly increase if the RH is improved in these models. In southern China, the increase in the AOD efficiency is slower than that predicted by Köhler theory because the strong precipitation in this region scavenges aerosols.

GISS-E2-R-TOMAS shows a sulfate AOD efficiency in agreement with the models that use bulk size approaches and the parametric formulas of Köhler theory. GISS-E2-R simulates a much higher AOD efficiency than the other models. The two GISS models are based on the same atmospheric model framework and have comparable monthly mean RH values. The large difference between their AOD efficiencies is likely relevant to the shapes of the RH distributions and the responses of the planetary boundary layer (PBL) to aerosol radiative forcings (a lower PBL height traps more aerosols in the

Figure 5. Sulfate AOD efficiency (the ratio of the sulfate AOD to the sulfate column burden) plotted against the mean RH below 850 hPa in June–August. The dashed lines represent the reanalysis RH values.
humid boundary layers). These models are two-way coupled chemistry–climate models. Aerosol radiative feedback may amplify the differences in the shape of the RH distributions and meanwhile decrease the wind speed near the surface (Yang et al. 2016; Yang et al. 2018), resulting in the strong differences in the AOD efficiency.

4. Summary
The ACCMIP models underestimate the ambient RH in the east of mainland China. The dry air biases decrease the hygroscopic growth of hydrophilic particles, which partly accounts for the low bias in the AOD. The difference in the monthly mean RH between the models and the reanalysis data changes the multi-model annual mean AOD by 45% in southern China and 33% in June and August in northern China. Most of the models agree that increasing the ambient RH increases the AOD efficiency of sulfate in a similar way to the Köhler curve for the hygroscopicity of single sulfate particulate. GISS-E2-R deviates from the other models and predicts the highest sulfate AOD efficiency, which could be due to its shape of RH distribution and PBL feedback to aerosol forcings being different from the other models. The AOD in this model cannot be improved by improving the mean value of the RH.

The study is limited by the monthly timescale of the model results, which prevents further discussion on the impacts of the shape error in the frequency distribution of the RH. The shape of the RH distribution is likely more effective than the mean value in altering the AOD because the aerosol radiative effect increases the concentration of secondary aerosols and the ambient RH on haze days. The hygroscopicity of organic aerosols is not discussed here because the concentration of SOAs is strongly underestimated in the model chemistry. The role of RH in the simulation of the AOD may be more pronounced as the heterogeneous reactions and SOA chemistry are improved in these models. Because a high AOD indicates a strong aerosol radiative effect, the propagation of error between the aerosol chemistry and meteorological conditions in two-way coupled chemistry–atmosphere models is an important topic for research on the effects of aerosols on climate in China.

Disclosure statement
No potential conflict of interest was reported by the author.

Funding
This work was jointly supported by the National Key Research and Development Program of China [grant number 2016YFE0201400] and the Basic Research Program of the State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences [grant number 7-082999].

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