

Anonymous Referee #3

The authors appreciate the reviewers very much for reviewing our manuscript and providing constructive comments. As suggested, we carefully revised the manuscript thoroughly according to the valuable advices, as well as proof-read the manuscript to minimize typographical, grammatical, and bibliographical errors. Our replies to the comments and our actions taken to revise the paper (in blue) are given below (the original comments are copied here). The figures added in the reply is represented by ‘Figure’, which is distinguished from ‘Fig.’ in the manuscript.

General comments:

1. This paper presents the atmospheric chemistry component of CAS-ESM, IAP-AACM, and compares the offline model results (driven by WRF) with various observational data worldwide. This is an important step towards improving the Earth system simulations by CAS/IAP, a key participant of IPCC assessments. Below are a few suggestions to improve the paper.

The model evaluation focuses on comparisons with measurements of surface concentrations of pollutants, particularly aerosol pollutants. Because this model is developed primarily for climate studies, evaluation of the tropospheric chemistry (in addition to surface air quality) will be very important. Specifically, It would be very useful to include/expand the evaluation of vertical profiles and tropospheric burdens against observations. There are many satellite data for ozone, NO₂, SO₂ and HCHO, and many vertical profile data (e.g., ATOM) for gaseous/aerosol species. Other important measures of tropospheric chemistry that can be discussed include the mean OH concentration and budgets, ozone budgets, methane lifetime, and MCF lifetime.

Reply: It is a good suggestion to include vertical comparison to improve the model evaluation work. We evaluated the tropospheric column concentration of NO₂ and O₃ with satellite data (GOME2A and OMI) and discussed the profile concentration of OH with other models, in the light of reviewer’s comments. The budget of ozone and CO are also elaluated in the manuscript.

Table 1 the budget of O₃ and CO compared with the other models.

Species	Process	IAP-AACM	
CO	Anthrop.	546.4	
	Emission (Tg yr ⁻¹)	Bio. burning	336.2
	Total 994	Biogenic	92.7
		Others	18.3
	Top condition inflow (Tg yr ⁻¹)	28	

	Chem pro (Tg yr ⁻¹)	1270
	Chem lss (Tg yr ⁻¹)	2292
	Dry dep (Tg yr ⁻¹)	0
	Burden (Tg)	327
	Lifetime (days)	52
	<hr/>	
	Top condition inflow (Tg yr ⁻¹)	473
	Chemical production (Tg yr ⁻¹)	3940
	Chemical loss (Tg yr ⁻¹)	3564
O ₃	Dry dep. (Tg yr ⁻¹)	849
	Burden (Tg)	370
	Lifetime (days)	30.6
	<hr/>	

O₃: The vertical tropospheric column (VTC) of O₃ is compared against satellite observation derived from OMI (shown in Figure 1). In the main board, the pattern of the seasonal cycle was covered by the model. In mainland of Northern Hemisphere, the higher O₃ VTC appears during June-July-August (JJA), while in Northern Hemisphere, it appears during September-October-November (SON), with a range of 40-60 DU. The model still keeps a high value (40-50 DU) in tropics during DJF, possibly due to the high concentration of CO emit from biomass burning. The O₃ VTC is significantly underestimated over ocean in middle-high latitudes, and the reasons need to be further studied.

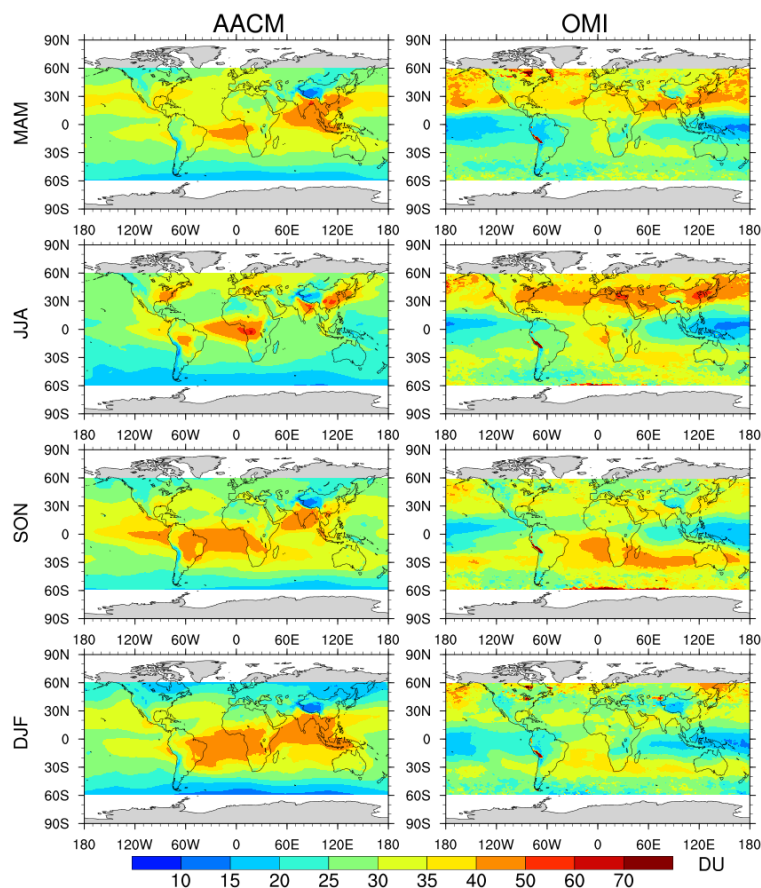


Figure 1 Seasonal mean column concentration of O₃ in IAP-AACM (left column) and OMI (right column). Seasons are defined as December-January-February (DJF), March-April-May (MAM), June-July-August (JJA), and September-October-November (SON). The unit is DU.

NO₂: The VTC of NO₂ is also compared against satellite observation derived from GOME2A (shown in Figure 2). The NO₂ VTC has a range of 20-150 ×10¹⁴ molecule cm⁻² in most source areas. By and large, IAP-AACM reproduced the magnitude in different regions. In addition, the model captured seasonal variations of NO₂ concentration in the vertical troposphere well. In anthropogenic source areas of Northern Hemisphere (e.g., North America, Europe, East Asia), the NO₂ VTC is higher in SON and December-January-February (DJF) while lower in JJA, caused by seasonal human activities such as fuel heating. The column concentration in South America and South Africa is higher during JJA, while it is higher in central Africa during DJF, due to the vegetation burning in dry season. Compared with GOME2A, IAP-AACM showed a larger column concentration over ocean. The overestimation is also reflected in the comparison of surface concentration. This is probably caused by insufficient oxidation to nitrate and a higher injection height in the emission which leads to a farther transportation distance. Generally, the distribution of NO₂ by the model is consistent with satellite observation, except some source areas (e.g., underestimation in Australia and South America, overestimation in East Asia), which suggests a bias of emission inventory.

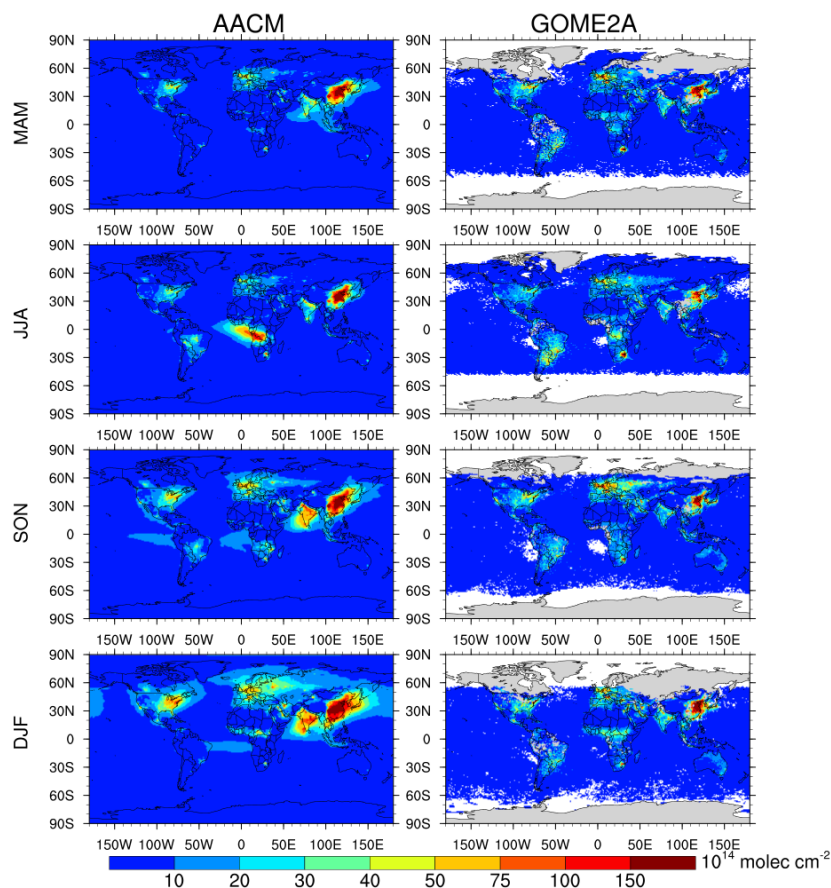


Figure 2 Seasonal mean column concentration of NO_2 in IAP-AACM (left column) and GOME-2A (right column). Seasons are defined as December-January-February (DJF), March-April-May (MAM), June-July-August (JJA), and September-October-November (SON). The unit is 10^{14} molecule cm^{-2} .

OH: Oxidation is the basic characteristic of atmospheric chemistry. As the most important oxidant in atmosphere, hydroxyl radical (OH) is one of the crucial species to simulate the general properties in CTMs. OH formation in troposphere is mainly due to O_3 photolysis with the reaction $\text{O}_3 + h\nu (\lambda \leq 320\text{nm}) + \text{H}_2\text{O} \rightarrow 2\text{OH} + \text{O}_2$. The tropospheric (200hpa to the surface) mean OH concentration of IAP-AACM is 13.0×10^5 molec cm^{-3} . It is a little higher than the mean OH concentration study ($11.1 \pm 1.6 \times 10^5$ molec cm^{-3}) from 16 ACCMIP models for 2000 by Naik et al. (2013). It potentially leads to strong atmospheric oxidation. The lower concentration of CO over oceans may be related to it. The zonal mean OH concentrations for January, April, July and October are shown in Figure 3. Like other chemistry models, OH concentration in the tropics keeps highest all the year round and decreases gradually from tropics to poles. This is due to the positive influence of solar radiation and water vapor concentration. The seasonal north-south oscillation of OH maximum area is also ascribed to the seasonal variation of these two factors. The mean OH inter-hemispheric (N/S) ratio of the model is 1.26, in accordance with the present-day multi-model mean ratio (1.28 ± 0.1) for 2000 (Naik et al., 2013). Vertically, the highest concentration is in the layer of 2-4 km above the tropics. In Northern Hemisphere, the highest OH concentration appears in

summer. Peak value of OH in July is located at around 30°N, in the sky above 2km. Generally, the range of OH concentration is similar with other models (e.g., TM5 (Huijnen et al., 2010), NMMB-MONARCH (Badia et al., 2017)), except a slightly higher peak concentration of 30-35 molec cm⁻³, compared with the other models above-mentioned (under 30 molec cm⁻³).

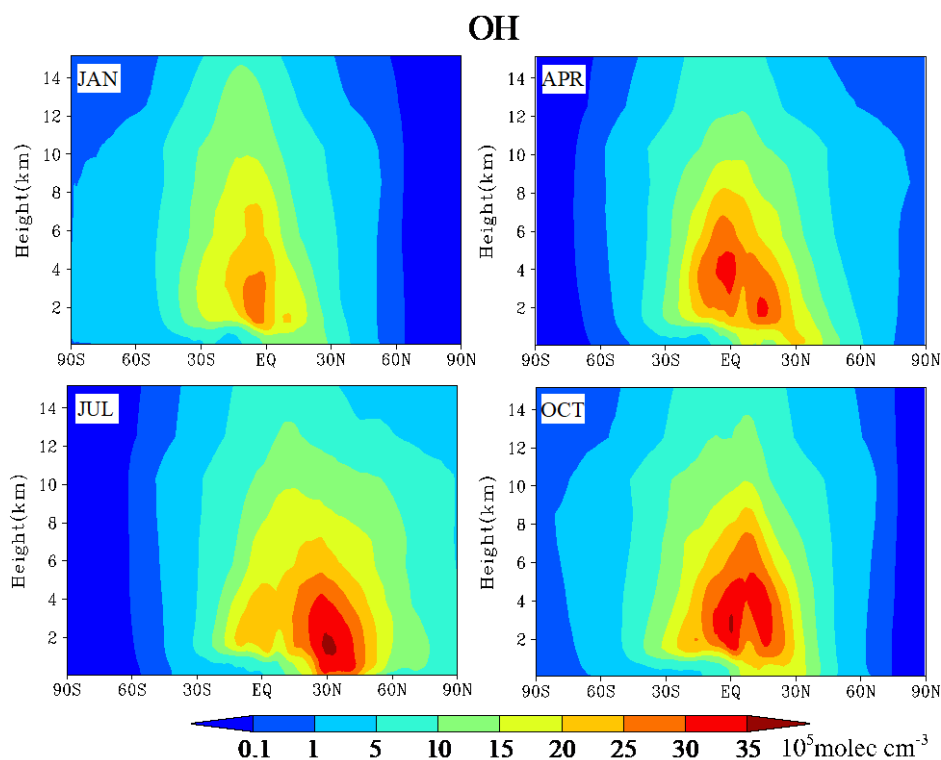


Figure 3 Zonal monthly mean concentration of OH for January, April, July and October by the IAP-AACM. The unit is 10⁵ molecule cm⁻³.

2. Measurement data often contain missing values and outliers and have different temporal resolutions from model simulations. Please specify how the measurement data are processed and how model results are sampled (temporally and spatially) according to measurements. In particular, satellite data contain large amounts of missing values. Near-surface NO₂ measurements are contaminated by other nitrogen species, and what would be the implications for model evaluation (especially when discussing the model bias).

Reply: The measurement datasets (except CNEMC) collected in this paper are monthly or annual results which have been processed by the observation workgroups. The hourly CNEMC observations are processed by data quality control. The corresponding simulation data compared with aforementioned observations are sampled at the same locations and altitudes as observations, with the model grid cells containing the observational sites. The simulation of seasonal cycle in different regions or cities are first sampled at the model grid cells containing the observational sites and then averaged within sub-regions. When compared with satellite data, the missing values of satellite data are kept and shown in the figures.

As shown in Figure 12 in the manuscript, model results for NO₂ concentration are a bit underestimated (NMB= -0.63). As the “NO₂” values reported by routine monitoring sites are NO₂^{*}, which partially includes HNO₃ and NO₃⁻, it is common to underestimate the observed “NO₂”. Thus the model’s overestimation should become worse. It reflects the shortcoming of multiphase processes in IAP-AACM. The overestimation of NO₂ and underestimation of nitrate in daytime of summer and autumn is related to the over decomposition of nitric acid at high temperature condition in the thermodynamic equilibrium module. Moreover, heterogeneous chemical reactions in the model should partly be responsible for the NO₂ overestimation in summer. Reactive heterogeneous uptake of gases may be crucial for the formation of secondary aerosols when the other oxidants (e.g. ozone, OH) are in low concentrations level (Jacob, 2000; Martin et al., 2003). The heterogeneous chemical module coupled in IAP-AACM has been tested in North China in winter (Li et al, 2018). The uptake of SO₂ by wet aerosols significantly enhanced sulfate formation under highly polluted conditions, contributing 50%-80% of total concentration of sulfate. The mechanism also reduced the overestimation of nitrate which is also appeared in other models. However, when it comes to the problem here, we checked the simulations excluded heterogeneous chemical processes and found a better performance of NO₂ in summer (shown in Figure 4). It implicates that a more comprehensive mechanism should be considered in model development.

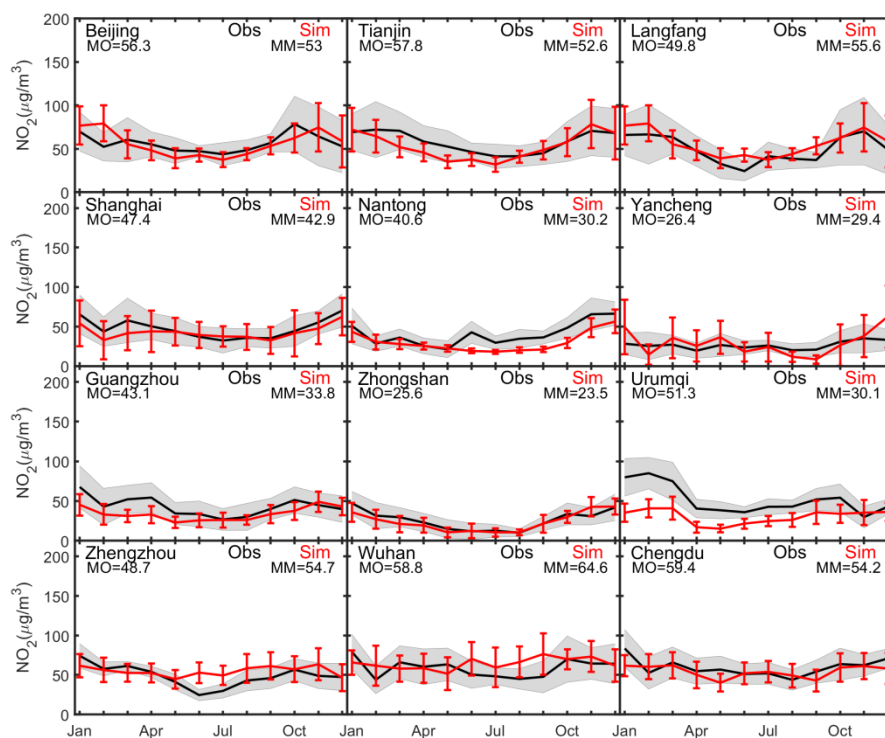


Figure 4 Seasonal cycle of NO₂ (µg m⁻³) simulated without heterogeneous chemical process over China. The black line and red line represent monthly mean concentration of city-averaged observation and simulation respectively. Gray shaded areas and red vertical bars show 1 standard deviation over the sites

for observations and for model results, respectively. MO and MM stand for annual mean concentration of observation and simulation respectively.

3. The resolution dependence discussed in Sect. 3.4 has also been studied in other recent works. It would be nice to refer to or compare against previous findings.

Reply: That's a good suggestion. High-resolution helps to improve CTMs performance, but it is limited by the scale applicable to the parameterization scheme of physical and chemical processes. Recently, sensitivity to horizontal grid resolution has been discussed in many regional model works. Wang et al. (2014) showed a better simulation of particles in North China with CMAQ when increasing the resolution from 36km to 12km. A study of PM_{2.5} health impact assessment with CMAQ by Jiang et al. (2018) found that model results at 12 km generally performed better and had substantially lower computational burden, compared to 4 km resolution. As a global nested model, we also want to evaluate the improvements or not due to higher horizontal resolution.

4. The spin-up time (one month) is too short for CO, ozone and other longer-lived species. This may explain part of the underestimate in CO. Please comment on the effect of spin-up time.

Reply: We agree that the spin-up time of one month is not enough for longer-lived species. It may lead to an underestimation of some trace gases such as CO. But in this study we used monthly mean concentration of CO, O₃ and NO₂ from MOZART-4 as the top boundary condition. It can offset the potential underestimation of CO and O₃ substantially. Furthermore, to verify the effect of shorter spin-up time here, we also run a case with spin-up time of one year. The annual mean result is very similar to the case of one month spin-up time as shown in Figure 5.

The underestimation of CO potentially reflects a difference in emissions. The natural sources of CO over ocean are included in the HTAP models whereas they are not considered in IAP-AACM. Besides, it may reflect differences in chemical transformation between models. As shown in Figure 3, the OH concentration is a bit higher in IAP-AACM than the other models. Due to the sink reaction of CO ($\text{CO} + \text{OH} \rightarrow \text{CO}_2 + \text{H}$), the CO loss will be faster in IAP-AACM.

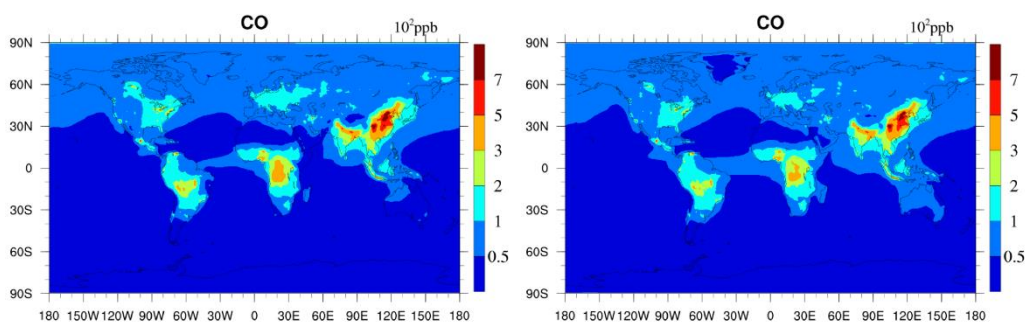


Figure 5 The annual mean surface concentration of CO. The left one is the surface distribution with one month spin-up time, the right one is with one year spin-up time.

5. There have been discussions in the literature on bug fixes in ISOROPIA II. Are these bugs and fixes relevant here?

Reply: No, it's not relevant here. The code bug only affects the forward (in which the concentration of both gas and aerosol of each species is fixed) stable state calculation. In IAP-AACM, we use reverse mode (in which the concentration of each species in the aerosol phase is fixed) to calculate.

6. Brief descriptions of WRFv3.3 would be very useful. The vertical resolution of WRF is different from that in IAP-AACM, so how is the conversion done?

Reply: The WRF version used in this study is a global version of WRFv3.3 (GWRF). It is an extension of mesoscale WRF that was developed for global weather research and forecasting applications. GWRF has more general choice of map projection (to include both conformal and nonconformal map projections). It includes specific boundary conditions and can be run as a traditional C-grid GCM by filtering. The specification of planetary constants, physics parameterizations and timing conventions are also improved to allow the model to be run as a global model. Thus, it has multiscale and nesting capabilities, blurring the distinction between global and mesoscale models and enabling investigation of coupling between processes on all scales. The model has been applied to simulation at various scales to Mars, and at global scales to Titan and Venus (Richardson et al., 2007).

Output of WRF is interpolated to keep accordance with IAP-AACM vertically. The information has been added in the revised manuscript.

7. Table 1 – do you extrapolate the emissions to 2014? If not, what would be implications for your model evaluation against measurements in 2014?

Reply: Yes, we extrapolate the emission of SO₂ to 2014. As a consequence of government control policy included in the twelfth Five-Year Plan (FYP), China has achieved an obvious decrease in air pollution in the past years, especially for SO₂. The FYP controls suppress SO₂ emissions in energy and industry sectors which is the major source of SO₂. Considering the cutting effect on SO₂ (China completed the emission reduction task of 12th FYP (2010~2015) ahead of schedule in 2014 with a reduction ratio reaching by 12.9%), we adjusted the total SO₂ emission for 2014 by a factor of 0.9 in China. For other species, the intensity of emission reduction is not so great like SO₂. The study by Zheng et al. (2018) showed that the dramatic reduction of emissions is mostly happened after 2013 for China's Clean Air Action implemented during 2013-2017. Relative change rates of China's anthropogenic emissions during 2010-2017 are estimated as follows: -62% for SO₂, -17% for NO_x, -27% for CO, -27% for BC and -35% for OC. And the emission mostly decreased during 2013-2017, by 59% for SO₂, 21% for NO_x, 23% for

CO, 28% for BC and 32% for OC. Compared to 2010, emissions of trace gas in 2014 decreased not significant except SO₂ (shown in Figure 6). So we only extrapolate the emission of SO₂. It will partly be responsible for the underestimation of some species (e.g., NO₂ in Fig. 13) in our simulation.

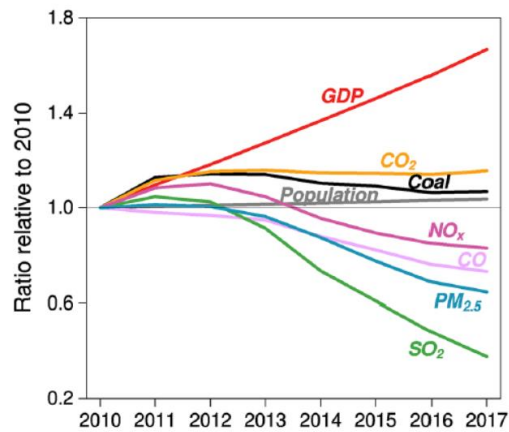


Figure 6 Emission trends and underlying social and economic factors from 2010 to 2017 by Zheng et al. (2018).

8. In the comparisons over China, only a few cities are selected, although there are CEMC measurements in other cities as well. Please explain the rationale for choosing these cities.

Reply: The cities selected are divided into six regions (North China, Pearl River Delta, Yangtze River Delta, Northwest China, Central China, Southwest China). The six regions not only represent the major geographical regions over China, but also include the regions with the most severe air pollution at present which means the focus regions of research.

Specific comments:

1. Abstract – please specify which part of the writing is for the evaluation of global model and which is for nested model. Also, please present the bias (in addition to R) of the model.

Reply: Some words have been added to the abstract to specify global and nested evaluation (see below). Also, normal mean biases are supplemented, too.

For global simulation, the 1-year simulation for 2014 shows that the IAP-AACM is within the range of other models, and well reproduces both spatial distribution and seasonal variation of trace gases and aerosols over major continents and oceans (mostly within the factor of two). The model well captures spatial variation for carbon monoxide but with a bit underestimation (normal mean bias (NMB) of -0.59~-0.23) especially over the ocean that also shown in other models, which suggests the need for more accurate emission rate of ocean source. For aerosols, the simulation of fine-mode particulate matter (PM_{2.5}) matches observation well and it has a better simulating ability on primary aerosols (NMB are

within ± 0.67) than secondary aerosols (NMB are greater than 1.0 in some regions). This calls for more investigation on aerosol chemistry. Furthermore, for nested regional simulation, IAP-AACM shows the superiority of global model, compared with regional model, on performing regional transportation for the nested simulation over East Asia. With regard to the city evaluation over China, the model reproduces variation of sulfur dioxide (SO₂), nitrogen dioxide (NO₂) and PM_{2.5} accurately in most cities, with correlation coefficients (R) above 0.5 and NMB within ± 0.5 .

2. L48-67 – the references are relatively old. Please use newer ones. Also, aerosols affect the cardiovascular diseases very significantly.

Reply: The citation of IPCC has been updated to the latest report. References for aerosols' health effect are also updated (see below).

Aerosols formatted from these precursor gases, together with aerosols from other sources, have a direct radiative forcing. By modifying cloud properties, the aerosols also have important indirect effects. As reported in the Fifth Assessment Report (AR5) of IPCC (Myhre et al., 2013), the radiative forcing of aerosols ranges from $-1.9 \sim -0.1 \text{ W m}^{-2}$, with the direct radiative forcing ranges from $-0.85 \sim 0.15 \text{ W m}^{-2}$. With better model performance and more robust observation network, AR5 achieved increasing confidence in the assessment compared with AR4 (Boucher et al., 2013), but the largest uncertainty to the total radiative forcing estimate is still aerosols. In addition, aerosols have adverse impacts on human health including respiratory diseases, cardiovascular risk and lung cancer, which has drawn increasing public attention (Burnett et al., 2014; Pope et al., 2011; Powell et al., 2015).

3. L71 – change “prediction” to “projection”

Reply: It has been corrected.

4. L87-88 – there have been model evaluation studies over China in recent years. Please refer to these studies.

Reply: Yes, there have been several model evaluation studies with observation in China. The description in the introduction has been updated.

5. L97 – remove “precise”. Every model has its limitations.

Reply: It has been modified in the revised manuscript.

6. L100 – change to “lateral (and upper) boundary conditions”

Reply: It has been modified in the revised manuscript.

7. L147 – specify the resolution

Reply: The high resolution is $0.25^{\circ} \times 0.25^{\circ}$, we have specified it in the revised manuscript.

8. L160 – do you mean “natural dust”?

Reply: Yes, it is.

9. L199 – do you mean the first layer center is 50 m?

Reply: Yes, we have specified the meaning.

10. Table 2 – please explain the meanings of these statistics and provide the units.

Reply: Captions and units are added in the revised manuscript.

11. L279 – why not just use the WDCGG data in 2014?

Reply: The dataset of WDCGG provides a large number of trace gases observations globally. But some sites are without invalid records in 2014. To get more data to evaluate the model over the world, we expanded the time range to ten years (2006-2015).

We have re-selected the observation data for 2014 to comparison with model results. Overall, the results have not changed much in terms of the evaluation of model's simulation capability. The simulation bias is reduced in some regions while it is increased in some other regions. The simulation of NO_2 performs better with the NMB of Asia and Europe closer to zero. The underestimation of CO in Antarctica disappeared due to the change of the observed value. There are some changes in the trend of the seasonal variation of O_3 in Northern Hemisphere. All the figures (as shown in Figure 7~ Figure 9) and tables related to these changes are updated in the manuscript, and the corresponding analysis is updated in the manuscript, too.

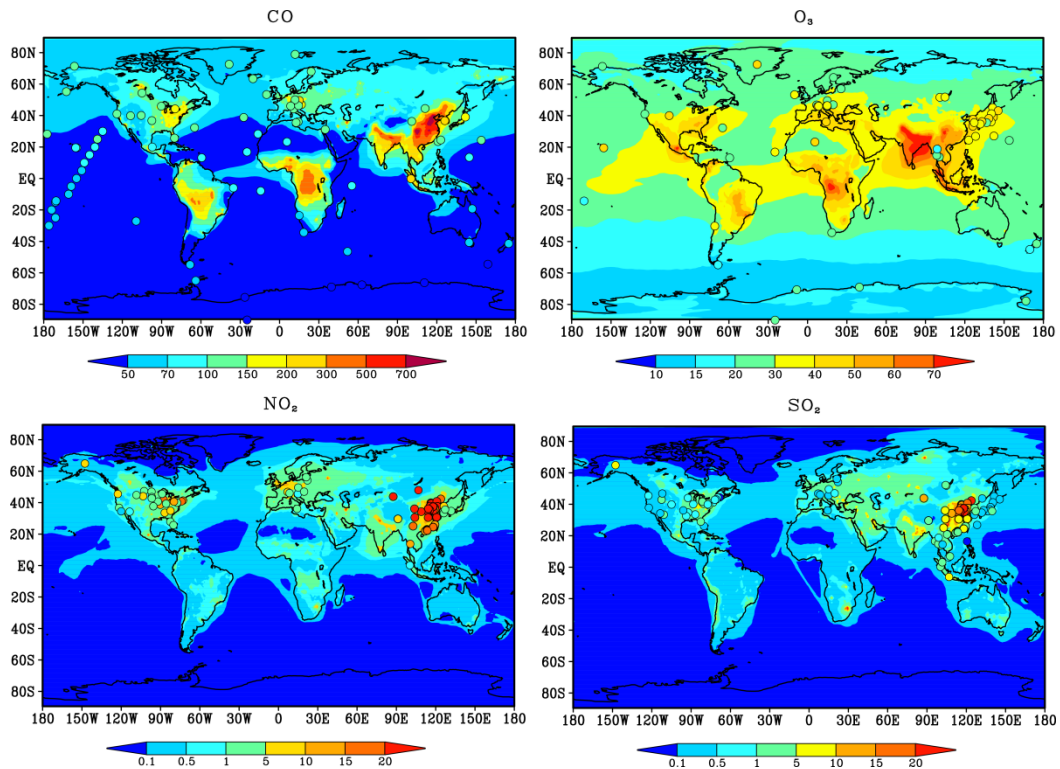


Figure 7 Annual mean concentration (ppb) of the surface layer in IAP-AACM. The circles represent site observations. The first row is CO and O₃, the bottom row is NO₂ and SO₂.

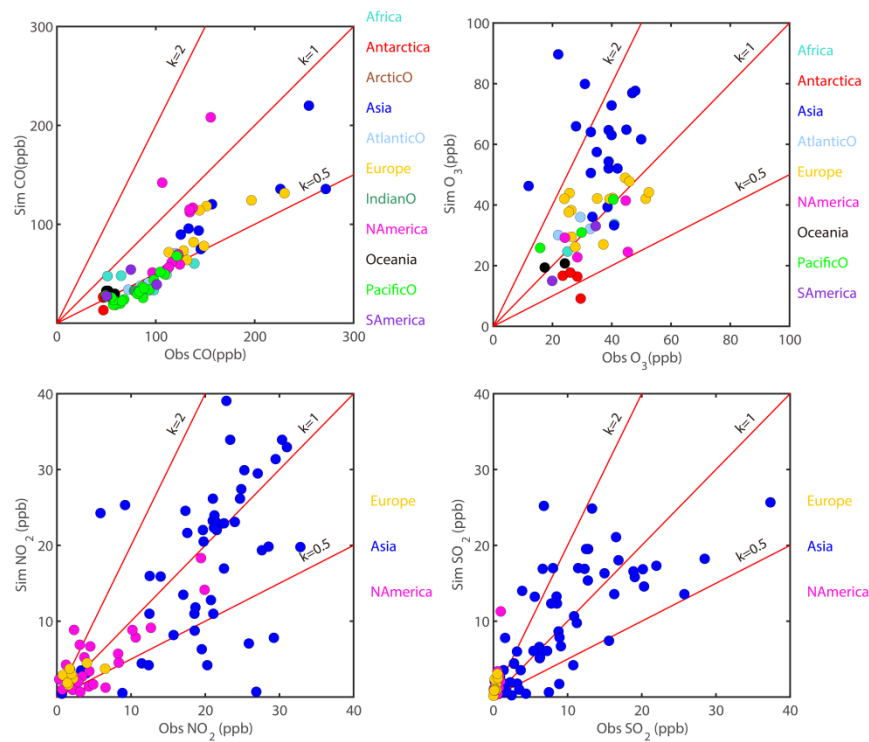


Figure 8 Scatter plots of annual mean concentrations (ppb) in Africa, Antarctica, Arctic Ocean (ArcticO), Asia, Atlantic Ocean (AtlanticO), Europe, Indian Ocean (IndianO), North America (NAmerica), South

America (SAmerica), Oceania and Pacific Ocean (PacificO). The abscissa shows the observation and the ordinate shows the simulation. The color of the points represents different regions. (a) ~ (d) show CO, O₃, NO₂ and SO₂ respectively.

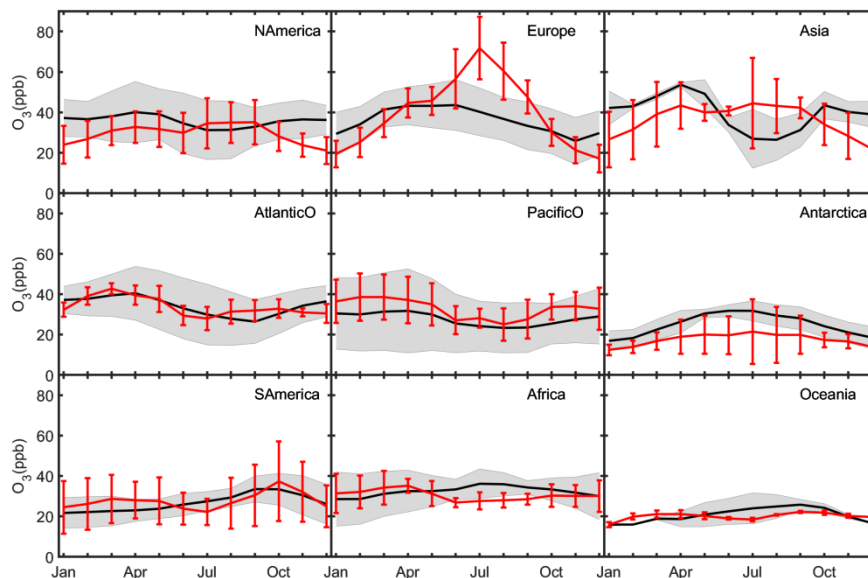


Figure 9 Mean seasonal variation of O₃ (ppb) over NAmerica, Europe, Asia, AtlanticO, PacificO, Antarctica, SAmerica, Africa and Oceania sites. Black lines and red lines represent the average of observations and simulations respectively. Gray shaded areas and red vertical bars show 1 standard deviation over the sites for observations and for model results respectively.

12. CO model evaluation – could you comment on the effect of spinup time and coarse model resolution?

Reply: We agree that the spin-up time of one month is not enough for longer-lived species. It may lead to an underestimation of some trace gases such as CO and O₃. But in this study we used monthly mean concentration of CO, O₃ and NO₂ from MOZART-4 as the top boundary condition. It can offset potential underestimation of CO and O₃ substantially. Furthermore, to verify the effect of the shorter spin-up time here, we also run a case with spin-up time of one year. The annual mean result is almost the same with the case of one month spin-up time as shown in Figure 5.

On one hand, the results of coarse-resolution models are often lower than those of high-resolution models due to the effect of gridded average on static emission sources. On the other hand, it's difficult to reproduce the atmospheric dynamics characteristics under complex underlying surface conditions for coarse resolution models. The coarse resolution of global models cannot represent local orographically driven flows or sharp gradients in mixing depths. It's unfavorable to simulate pollutant diffusion process. Furthermore, the inputs of meteorological fields with larger grids are also poorly represented.

13. L416 – the ozone seasonality is not very well captured in many regions. Also, this paragraph is too long.

Reply: Agree. We have reanalyzed the simulation of ozone in this part in the revised manuscript. The model showed poorly performance on the seasonal cycle of surface ozone in the NH land, with overestimation in Europe and EA in summer while underestimation in winter in NH land, as shown in Figure 9 (the comparison drawn with WDCGG observation only for 2014).

The surface O₃ are also underestimated in spring over NH land. In IAP-AACM, the stratospheric-tropospheric exchange and corresponding photochemistry are not considered. It will lead to a large negative bias in the simulating. To date it has become apparent that the measured annual cycle of ozone shows a distinct maximum during spring. The stratosphere-to-troposphere ozone transport event occurs widely across mid-latitudes in the NH (Monks et al., 2000; Akritidis et al., 2018). Since the magnitude and frequency of the transport through tropopause is still not clear. There are large uncertainties in simulating the flux. Some researches (Munzert et al, 1985; Austin and Follows, 1991) showed that the maximum in the stratosphere to troposphere flux occurs in late winter/spring. It may partly responsible for the underestimation of O₃ in winter, too.

The surface O₃ concentrations over East Asia (sites mainly located in Japan) are overestimated in summer and early autumn. The same pattern is also found in the multi-model inter-comparison of 21 HTAP models (Fiore et al., 2009). The simulations in island countries of EA are sensitive to the timing and extent of the Asian summer monsoon (Han et al., 2008). The positive model bias in this season may stem from inadequate representation of southwesterly inflow of clean marine air.

14. L452 – please specify the quantitative difference between GFED3 and GFED4.

Reply: GFED3 and GFED4 are both monthly burned area emission data gridded to 0.5°×0.5° and 0.25°×0.25°, respectively. Due to the impact of a reduction of combustion area and decreasing in fuel consumption, there is about a 20%~30% reduction of CO emissions in GFED4 compared to GFED3 in the tropical regions (central Africa, South America, and Southeast Asia) (Werf et al., 2017). The NO₂ emission of GFED4 may be also decreased due to the reduction of burned area. This specific difference has been added to the revised manuscript.

15. L485-499 – please comment on the effect of difference in time (2006 for measurements and 2014 for model simulation).

Reply: As the simulation used emissions of 2010 but the measurements are for 2006, there is a mismatch on emission scenario. There is a bit increasing (less than 0.1Tg) of BC and OC emissions from 2006 to 2010 in China (Lu et al., 2011; Fu et al., 2012). Besides, the meteorological conditions also play a role.

As the analysis of the CAWNET observation over China (Zhang et al., 2015), there is no significant changes happened in the proportion of chemical component of PM₁₀ from 2006 to 2013. For the annual average trends of carbonaceous shown in Figure 10, both Southwest China and North China experienced a

process of declining first and then rising due to the unfavorable weather conditions. Pearl River Delta showed a significant falling (about half). Yangtze River Delta had a slight decreasing. Generally, it is reasonable to infer that the distribution of BC and OC in most areas have changed a little from 2006 to 2014, except for the Pearl River Delta region.

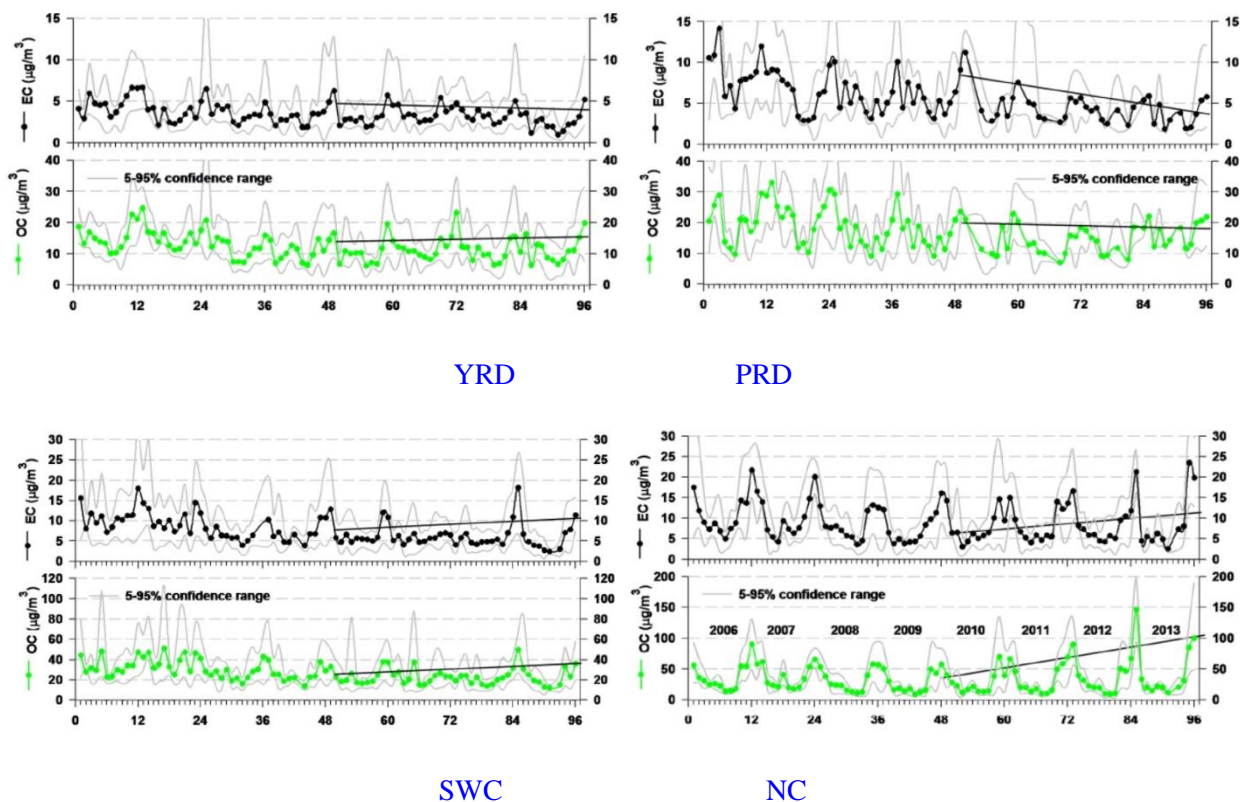


Figure 10 Monthly mean concentrations of OC and EC from 2006 to 2013 by Zhang et al., 2015. YRD, PRD, SWC and NC represents Yangtze River Delta, Pearl River Delta, Southwest China and North China, respectively.

16. L495 – BC depends on emissions and deposition processes.

Reply: Yes, it has been complemented.

17. L500 – please clarify which components are included in PM_{2.5}

Reply: The components of PM_{2.5} in Fig. 10 includes primary PM_{2.5}, BC, OC, SNA, SOA and also natural dust, this is supplemented in the revised manuscript.

18. L506 – please specify the version of MODIS AOD and how data are selected/sampled.

Reply: The product version is MYD04_L2-MODIS/Aqua Aerosol 5-Min L2 Swath 10km. It is available at the website: http://dx.doi.org/10.5067/MODIS/MYD04_L2.006. The product version and website is supplemented in the revised manuscript.

19. L512 – LAC or BC?

Reply: it should be BC here, it has been revised.

20. L522-531 – please consider to present the seasonality results in a line figure.

Reply: That's a good suggestion. A more detailed comparison of the global gridded average AOD on the seasonality variation is displayed in Figure 11. As the seasonality cycle is different in different regions, we not only showed the global average value, but also showed the gridded average value of Africa, South America and East Asia, which are major aerosol emission areas. Generally, the model captured seasonal variation in different regions, but there is a gap in the value between observation and simulation. The discrepancy in East Asia potentially stemmed from the inaccurate simulation of dust activities in spring, which is mainly due to the simulation of meteorological field (e.g., wind, precipitation).

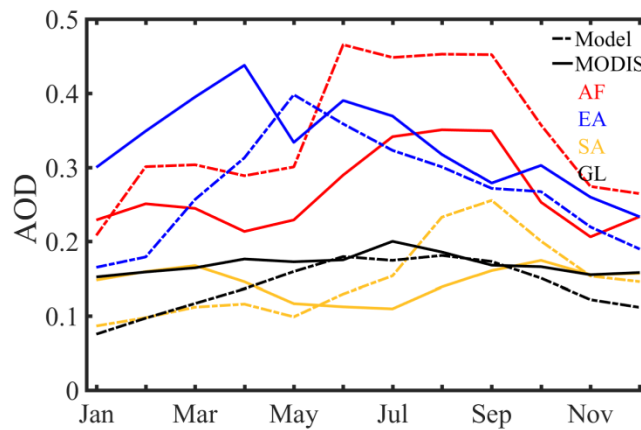


Figure 11 Gridded mean value of monthly averaged AOD for 2014, AF, EA, SA and GL represents Africa, East Asia, South America and global. Dash line and solid line represents model results and observation derived from MODIS, respectively.

21. L526 – In the model, DJF is not the season with the highest AOD over East China.

Reply: Yes, it's an incorrect expression here and we have deleted it. In fact, the highest AOD may not be in DJF, it often appears in MAM. This phenomenon is common in other model evaluation studies (e.g., GISS-TOMAS (Lee et al., 2010)). On one hand, China is frequently affected by dust in spring. On the other hand, AOD is an optical characteristic of aerosols for the whole vertical layer. It is not equivalent to aerosol mass concentration.

22. L548-558 – please be more quantitative.

Reply: To be more quantitative, we provided scatter plots of simulations in the nested domain in Figure 12. As shown in Figure 12, model results for SO_2 , PM_{10} and $\text{PM}_{2.5}$ are mostly within the factor of two with NMB within ± 0.52 , while NO_2 concentration are a bit underestimated (NMB= -0.63).

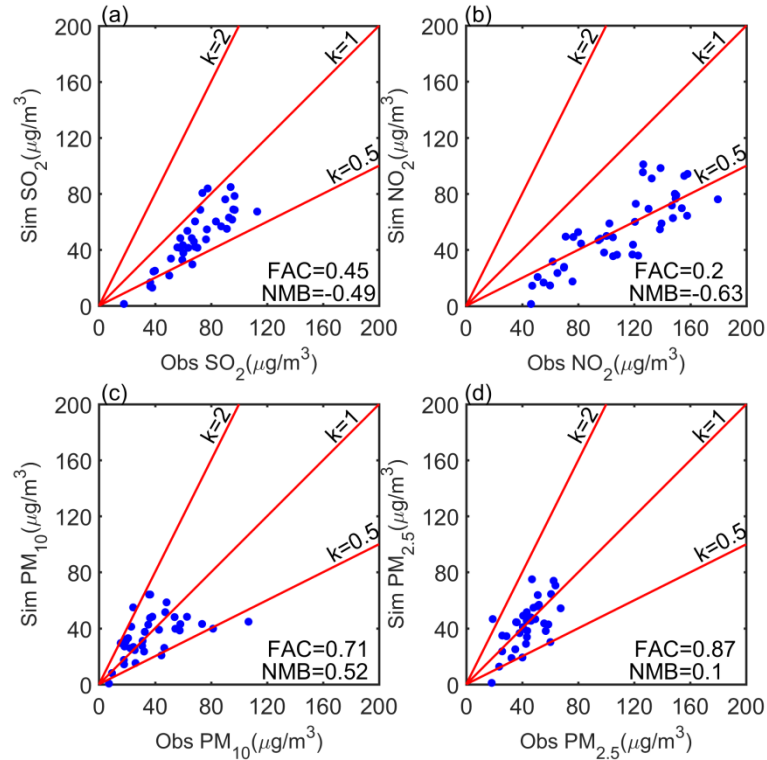


Figure 12 Scatter plots of annual mean concentrations ($\mu\text{g m}^{-3}$) in nested domain. (a)~(f) is SO₂, NO₂, PM₁₀ and PM_{2.5} respectively. The abscissa shows the observation and the ordinate shows the simulation.

23. L567-568 – please provide model versions.

Reply: the model versions are CMAQv4.7.1, WRF-Chemv3.9 respectively. This has been added in the revised manuscript.

24. L572 – do you mean “other regional models”?

Reply: It means the regional model. Here we compared the simulation of the nested domain in IAP-AACM with regional models of MICS-Asia.

25. L575-576 – what are the differences in emissions?

Reply: The differences of emissions between IAP-AACM and MICS-Asia models are natural sources. For anthropogenic source, IAP-AACM uses MIX inventory (incorporated into HTAP for Asia) as same as MICS-Asia models. For biogenic source, IAP-AACM uses MEGAN-MACC but models of MICS-Asia uses an earlier version of MEGANv2.04. For biomass burning source, IAP-AACM uses GFEDv4 but MICS-Asia models uses GFEDv3.

26. Fig. 14 – please specify the components in PM_{2.5}

Reply: The components of PM_{2.5} in Fig. 14 includes primary PM_{2.5}, BC, OC, SNA and SOA, this is supplement in the caption.

27. Table 6 – please specify which one is global model and which one is regional model. Also, please provide the mean values over these cities.

Reply: Do you mean Table 7 in the ACPD document, the statistics for 12 cities in global and nested domains? If so, all the results in this table are calculated with outputs from the global model IAP-AACM. The difference between D1 and D2 is the horizontal resolution. D1 represents domain 1 ($1^\circ \times 1^\circ$), D2 represents domain 2 ($0.33^\circ \times 0.33^\circ$). The statistics over these cities are supplemented in Table 7.

References

Akritidis, D., Katragkou, E., Zanis, P., et al. A deep stratosphere-to-troposphere ozone transport event over Europe simulated in CAMS global and regional forecast systems: analysis and evaluation. *Atmospheric Chemistry and Physics*, 18, 15515–15534, doi: 10.5194/acp-18-15515-2018, 2018.

Austin, J. F., Follows, M. J. The ozone record at Payerne: an assessment of the cross-tropopause flux. *Atmospheric Environment* 25A, 1873-1880, 1991.

Boucher, O., D. Randall, P. Artaxo, C. Bretherton, G. Feingold, P. Forster, V.-M. Kerminen, Y. Kondo, H. Liao, U. Lohmann, P. Rasch, S.K. Satheesh, S. Sherwood, B. Stevens and X.Y. Zhang, 2013: Clouds and Aerosols. In: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Burnett, R. T., Arden Pope, C., Ezzati, M., Olives, C., Lim, S. S., Mehta, S., Shin, H. H., Singh, G., Hubbell, B., Brauer, M., Ross, Anderson, H., Smith, K. R., Balmes, J. R., Bruce, N. G., Kan, H., Laden, F., Prüss-Ustün, A., Turner, M. C., Gapstur, S. M., Diver, W. R., and Cohen, A: An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure, *Environ. Health Persp.*, 122, 397, <https://doi.org/10.1289/ehp.1307049>, 2014.

Dai, Y. J., Zeng, X. B., Dickinson, R. E. , Baker, I. ,Bonan, G. B. & Bosilovich, M. G.: The common land model. *Bulletin of the American Meteorological Society*, 84(8), 1013-1023, 2015. doi:10.1175/BAMS-84-8-1013, 2015.

Falk, S., & Sinnhuber, B. M. Polar boundary layer bromine explosion and ozone depletion events in the chemistry-climate model EMAC v2.52: implementation and evaluation of airsnow algorithm. *Geoscientific Model Development*, 11(3), 1-15, <https://doi.org/10.5194/gmd-11-1115-2018>, 2018.

Fiore, A. M., Dentener, F. J., Wild, O., et al. Multimodel estimates of intercontinental source-receptor relationships for ozone pollution. *JOURNAL OF GEOPHYSICAL RESEARCH*, 114, D04301, doi:10.1029/2008JD010816, 2009, 2009.

Fu, T.-M., Cao, J. J., Zhang, X. Y., Lee, S. C., & Henze, D. K.: Carbonaceous aerosols in china: top-down constraints on primary sources and estimation of secondary contribution. *Atmospheric Chemistry and Physics*, 12(5), 2725-2746, doi:10.5194/acp-12-2725-2012, 2012.

Galmarini, S., Koffi, B., Solazzo, E., Keating, T., Hogrefe, C., & Schulz, M., et al. Technical note:

coordination and harmonization of the multi-scale, multi-model activities HTAP2, AQMEII3, and MICS-Asia3: simulations, emission inventories, boundary conditions, and model output formats. *Atmospheric Chemistry and Physics Discuss*, 17(2), 1543-1555, 2017.

Han, Z., et al. MICS-Asia II: Model intercomparison and evaluation of ozone and relevant species, *Atmos. Environ.*, 42, 3491 – 3509, doi:10.1016/j.atmosenv.2007.07.031, 2008.

Han, Z., Xie, Z., Wang, G., Zhang, R., & Tao, J. Modeling organic aerosols over east china using a volatility basis-set approach with aging mechanism in a regional air quality model. *Atmospheric Environment*, 124, 186-198, 2016.

Huijnen, V., Williams, J., van Weele, M., van Noije, T., Krol, M., Dentener, F., Segers, A., Houweling, S., Peters, W., de Laat, J., Boersma, F., Bergamaschi, P., van Velthoven, P., Le Sager, P., Eskes, H., Alkemade, F., Scheele, R., Nédélec, P., and Pätz, H.-W.: The global chemistry transport model TM5: description and evaluation of the tropospheric chemistry version 3.0, *Geosci. Model Dev.*, 3, 445–473, doi:10.5194/gmd-3-445-2010, 2010.

Jacob, D. J. . Heterogeneous chemistry and tropospheric ozone. *Atmos. Environ.* 34, 2131–2159, 2000.

Jiang, X. , & Yoo, E. H. The importance of spatial resolutions of community multiscale air quality (CMAQ) models on health impact assessment. *Science of The Total Environment*, 627, 1528-1543, doi: 10.1016/j.scitotenv.2018.01.228, 2018.

Lee, Y. H., & Adams, P. J. : Evaluation of aerosol distributions in the GISS-TOMAS global aerosol microphysics model with remote sensing observations. *Atmospheric Chemistry & Physics*, 10(5), 2129-2144, 2010.

Li, J., Chen, X., Wang, Z., Du, H., Yang, W., & Sun, Y., et al. : Radiative and heterogeneous chemical effects of aerosols on ozone and inorganic aerosols over East Asia, *Science of the Total Environment*, 622, 1327-1342, <https://doi.org/10.1016/j.scitotenv.2017.12.041>, 2018.

Li, Y., & Xu, Y.: Uptake and storage of anthropogenic CO₂ in the pacific ocean estimated using two modeling approaches. *Advances in Atmospheric Sciences*, 29(4), 795-809, 2012. doi: 10.1007/s00376-012-1170-4, 2012.

Jian, L., An, J., Yu, Q., Yong, C., Ying, L., & Tang, Y., et al. Local and distant source contributions to secondary organic aerosol in the Beijing urban area in summer. *Atmospheric Environment*, 124, 176-185, 2016.

Lu, Z, Zhang, Q, & Streets, D. G.: Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996–2010. *Atmospheric Chemistry and Physics*, 11(18), 9839-9864, 2011.

Martin, R.V., Jacob, D.J., Yantosca, R.M. . Global and regional decreases in tropospheric oxidants from photochemical effects of aerosols. *J. Geophys. Res.* 108 (D3):4097. <https://doi.org/10.1029/2002JD002622>, 2003.

Miyazaki, K. , & Bowman, K. Evaluation of ACCMIP ozone simulations and ozonesonde sampling biases using a satellite-based multi-constituent chemical reanalysis. *Atmospheric Chemistry and Physics*, 17(13), 8285-8312, 2017.

Monks, P. S. A review of the observations and origins of the spring ozone maximum. *Atmospheric*

Environment, 34(21), 3545-3561, 2000.

Munzert, K., Reiter, R., Kanter, H.-J., Potzl, K. Effect of stratospheric intrusions on the tropospheric ozone. In Proceedings of the Quad. Ozone Symposium, Halkidiki, Reidel, Dordrecht, pp. 735-739, 1985.

Myhre, G., D. Shindell, F.-M. Brón, W. Collins, J. Fuglestedt, J. Huang, D. Koch, J.-F. Lamarque, D. Lee, B. Mendoza, T. Nakajima, A. Robock, G. Stephens, T. Takemura and H. Zhang, 2013: Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Richardson, M. I., Toigo, A. D. and Newman, C. E: Planet WRF: A General Purpose, Local to Global Numerical Model for Planetary Atmosphere and Climate Dynamics, Journal of Geophysical Research, 112, E09001, doi:10.1029/2006JE002825, 2007.

Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J.-F., Lin, M., Prather, M. J., Young, P. J., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R., Eyring, V., Faluvegi, G., Folberth, G. A., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., van Noije, T. P. C., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R., Shindell, D. T., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., and Zeng, G.: Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmos. Chem. Phys., 13, 5277–5298, doi:10.5194/acp-13-5277-2013, 2013.

Pope III, C. A., Burnett, R. T., Turner, M. C., Cohen, A., Krewski, D., Jerrett, M. et al.: Lung cancer and cardiovascular disease mortality associated with ambient air pollution and cigarette smoke: shape of the exposure–response relationships, Environ. Health Persp., 119, 1616, <https://doi.org/10.1289/ehp.1103639>, 2011.

Powell, H., Krall, J. R., Wang, Y., Bell, M. L., and Peng, R. D.: Ambient coarse particulate matter and hospital admissions in the Medicare Cohort Air Pollution Study, 1999–2010, Environ. Health Persp., 123, 1152, <https://doi.org/10.1289/ehp.1408720>, 2015.

Simpson, W. R., von Glasow, R., Riedel, K., Anderson, P., Ariya, P., Bottenheim, J., Burrows, J., Carpenter, L. J., Frieß U., Goodsite, M. E., Heard, D., Hutterli, M., Jacobi, H.-W., Kaleschke, L., Neff, B., Plane, J., Platt, U., Richter, A., Roscoe, H., Sander, R., Shepson, P., Sodeau, J., Steffen, A., Wagner, T., and Wolff, E.: Halogens and their role in polar boundary-layer ozone depletion, Atmos. Chem. Phys., 7, 4375–4418, doi:10.5194/acp-7-4375-2007, 2007.

Xu, R. T., Tian, H. Q., Pan, S. F., Prior, S. A., Feng, Y. C., & Batchelor, W. D., et al.: Global ammonia emissions from synthetic nitrogen fertilizer applications in agricultural systems: Empirical and process-based estimates and uncertainty. Global Change Biology, 25, 314-325, doi: 10.1111/gcb.14499, 2019.

Wang, L. T., Jang, C., Zhang, Y., Wang, K., Zhang, Q., & Streets, D., et al.: Assessment of air quality benefits from national air pollution control policies in china. Part I: background, emission scenarios and evaluation of meteorological predictions. Atmospheric Environment, 44(28), 3449-3457, 2010.

Wang, L. T., Wei, Z., Yang, J., Zhang, Y., Zhang, F. F., & Su, J., et al. The 2013 severe haze over southern Hebei, China: model evaluation, source apportionment, and policy implications. Atmospheric Chemistry and

Physics, 14(11), 28395-28451, doi:10.5194/acp-14-3151-2014, 2014.

Werf, G. R. V. D., Randerson, J. T. , Giglio, L. , Leeuwen, T. T. V. , Chen, Y. , & Rogers, B. M. , et al. Global fire emissions estimates during 1997–2016. *Earth System Science Data*, 9(2), 697-720. <https://doi.org/10.5194/essd-9-697-2017>, 2017.