

Reply to Referee #1

We appreciate valuable comments. Our replies are as follows.

Referee #1:

Fig. 2: Please specify the targeted year.

Reply:

The target year is 2010 in BASE, and 2030 in BAU0 and PC1. The target year is added to the caption and within the figures.

Referee #1:

Figs. 3, 4, 5, 6, 7, 8, and Figs. S1, S2, S3: Please specify the year and scenario (it would be 2010 on BASE scenario).

Reply:

All the figures show simulated results in BASE for 2010. They are added to the captions.

Referee #1:

Fig. 4: Are observation date different from simulation date?

Reply:

The observation date at a few stations are different from 2010. The observation date are added within the figures.

Reply to Referee #3

We appreciate valuable comments. Our replies are as follows.

Referee #3:

Authors have modified the MS and they have added comparison with observations at some of the sites in South Asia. However, I feel that validation part of the model setup is still very weak, particularly, considering the future projection of ozone. Major flaw is simulation and comparison for a single year. Since ozone chemistry is highly complex and highly non-linear (with respect to growth in precursors), authors should make simulations for different years, say 1990 or 2000, and also compare with the available measurements. This will give confidence in using present setup for such studies. After comparing the observations for a set of two years (1990 and 2000 or 2000 and 2010), work on year 2030 would be more realistic. This argument would be clearer, after my concerns mentioned below.

Reply:

We definitely agree that a validation for different years could add confidence of this study. However, we need to estimate emissions for past years to validate simulations for different years. We utilized the best available information to estimate the emissions in 2010. It is much difficult to obtain information with the same quality for past years (e.g. 1990 and 2000). Therefore, it is necessary to set a lot of reliable assumptions for emission factors and activities for past years to compensate lack of reliable information. That is a challenging task and out of the scope of this study, whereas model performance for past years is largely affected by accuracy of emissions estimated for past years.

The purpose of this study is not predicting “realistic” tropospheric ozone concentration for future years. We developed three future scenarios, in which degrees of implementation of energy and environment strategies are different. The purpose of this study is to show potential effects of strategies assumed in scenarios to provide scientific understandings for considering effective strategies. BAU0 assumes no additional energy and environmental strategies. We believe BAU0 is not a realistic future pathway because the government should implement strategies at least by 2030 based on the information like the ones obtained in this study.

The sentence in the line 11 of the page 9520 is modified as follows.

“This paper describes prediction of tropospheric ozone over South and East Asia...”

The sentence in the line 23 of the page 9520 is modified as follows to clarify the purpose of this study.

“The purpose of this study is to evaluate effects of potential energy and environmental strategies implemented in China and India on tropospheric ozone over South and East Asia based on simulated results for three future scenarios.”

This issue is also mentioned in our reply to the later comments.

Referee #3:

It should be noted that simulated surface ozone is higher by 15-20 ppbv for 3-4 sites in India. Therefore, model results are already biased. Co-incidentally, increase in ozone (during 2030) is also about 20 ppbv for India.

Comparison of precursors like NO/NO₂/NO_x, CO, HC etc (from surface observations) are still missing.

Reply:

The horizontal resolution of the simulation in this study is 60 km x 60 km. It cannot inherently represent a horizontal variation below 60 km x 60 km. The target of this study is regional tropospheric ozone above this horizontal scale. Comparison of simulated values with observed ones affected by local sources should be avoided. EANET is an ideal network in which the monitoring stations are mainly located in remote areas. It is hard to find corresponding monitoring data in India. Nevertheless, the data obtained in the different year from 2010 and at the stations which are not located in the remote areas had to be included to show the model performance over India. Therefore, uncertainties caused by this limitation have to be taken into account.

The surface ozone is overestimated in Ahmedabad and Thumba throughout the year. The station in Ahmedabad is located in the urban area (Lal et al., 2000). The station in Thumba is located 10 km away from the city center (David and Nair, 2011). It appears that the observed values at the both stations are affected by local sources. MOZART-4 also overestimates the values at the both stations, implying a limitation to reproduce low values with the current horizontal resolution, not a bias of the simulation. Simulated ozone

around local sources is influenced by a horizontal resolution, and its temporal mean values tend to become lower with higher resolution (Chatani et al, 2011). Such a low value affected by local sources is not the target of this study. A lifetime of NO_x in the atmosphere is much shorter than ozone. It is not appropriate to compare observed NO_x values at respective surface stations with simulated values obtained in 60 km x 60 km meshes. It is difficult to find enough CO and HC monitoring data in India.

We mainly relied on the satellite data in this study. Although the satellite data contains uncertainties, its great advantage is comparisons between observed and simulated values which are both consistently averaged in the same individual meshes. It is suitable for validation of regional air quality simulations.

Following sentences are added in the line 2 of the page 9521 to mention a limitation due to the horizontal resolution.

“It must be noted that the horizontal resolution of the regional air quality simulations is 60 x 60 km. Therefore, it cannot inherently represent a horizontal variation below 60 km x 60 km. The target of this study is regional tropospheric ozone above this horizontal scale, which is not directly affected by local sources.”

Following sentences are added in the line 20 of the page 9526 to mention suitable observation data to validate simulations in this study.

“Although it should be avoided to use the observation data directly affected by local sources, some data had to be included due to limited data availability in the target region. We additionally relied on the satellite data. Its great advantage is comparisons between observed and simulated values which are both consistently averaged in the same individual meshes. It is suitable to validate the regional air quality simulations in this study.”

The sentences in the line 23 of the page 9526 are replaced by following sentences to show advantages of EANET.

“It is an ideal monitoring network to validate the simulations in this study because the monitoring sites are mainly located in remote areas. The ten monitoring sites classified as “Rural” and “Remote” were picked up to avoid sites affected by local sources (Network

Center for EANET, 2012). The monthly mean surface ozone concentration observed at these sites and the corresponding values simulated in the bottom layer are compared as shown in Fig. 3.”

A following sentence is added to the line 8 of the page 9528 to show a limitation of the data in India.

“Some of them are not located in remote areas.”

Following sentences are added as a new paragraph after the line 29 of the page 9528 to describe the overestimation at some sites in India.

“The surface ozone is overestimated in Ahmedabad and Thumba throughout the year. The station in Ahmedabad is located in the urban area (Lal et al., 2000). The station in Thumba is located 10 km away from the city center (David and Nair, 2011). It appears that the observed values at the both stations are affected by local sources. MOZART-4 also overestimates the values at the both stations, implying a limitation to reproduce low values with the current horizontal resolution, not a bias of the simulation. Simulated ozone around local sources is influenced by a horizontal resolution, and its temporal mean values tend to become lower with higher resolution (Chatani et al, 2011). Such a low value affected by local sources is not the target of this study.”

Referee #3:

Similar to the above, simulated tropospheric column ozone is also higher (probably more than double) when compared with satellite data, mainly in winter and spring (Fig 8). This can be better seen in the percentage differences (not shown). Therefore, increase in ozone for year 2030 should to carefully seen and percentage difference (between observations and simulations) should be kept in mind.

Reply:

The observed and simulated monthly column ozone in each region are shown in Fig. S3 in the supplementary material. The difference between observed and simulated monthly column ozone is 12 DU (37.9%) in January in Center (India) at the maximum. It actually falls far below double. Likewise, the difference between observed and simulated seasonal column ozone is 8.0 DU (24%) in DJF in Center (India). Ziemke et al. (2006) showed that the column ozone simulated by Global Modeling Initiative’s (GMI) Combined

Stratosphere-Troposphere Chemical Transport Model (COMBO CTM) is 5-10 DU higher than the value obtained from OMI/MLS in DJF in the corresponding region. The model performance in this study is comparable to them.

Following sentences are added to the line 27 of the page 9532 to describe overestimation in winter.

“The simulated values are higher than the observed ones over the mid latitude in winter. The difference between observed and simulated monthly column ozone is 12 DU (37.9%) in January in Center (India), and the difference between observed and simulated seasonal column ozone is 8.0 DU (24%) in DJF in the same region at the maximum. Ziemke et al. (2006) showed that the column ozone simulated by Global Modeling Initiative’s (GMI) Combined Stratosphere-Troposphere Chemical Transport Model (COMBO CTM) is 5-10 DU higher than the value obtained from OMI/MLS in DJF in the corresponding region. The model performance in this study is comparable to them.

Referee #3:

More detail on PC0 and PC1 is also important for studying the impact on ozone. Like, how much decrease in NO_x, VOC, etc is considered in these two scenarios. Then it would be better to compare ozone decrease in these two scenarios with increase in ozone from 2010 to 2030.

Reply:

We have published several papers describing PC0 and PC1 for China (Xing et al., 2011; Zhao et al., 2013a, 2013b, 2013c). The concept of PC0 and PC1 is similar for India. We will submit a separate paper describing PC0 and PC1 in details for India. The changes in NO_x and VOC in these two scenarios are shown in Table 1, and discussed in the Section 2.2.2.

Referee #3:

In case of BAU0, high ozone is seen along west coast of India and another hot spot around eastern India during winter. On the other hand, higher ozone are seen along south-east land mass of India and ozone levels are lesser along IGP region, one of the most polluted region. It is also interesting to see higher ozone along IGP region in Fig 8, but higher ozone along IGP is not very much clear in Fig 2.

Reply:

Fig. 2 shows surface ozone whereas Fig. 8 shows tropospheric column ozone. Differences between them may be caused by processes in the lower troposphere. A possible cause of less surface ozone in the polluted IGP region in winter is NO_x titration under less active photochemical reactions. It is more evident in northeastern China in which the decrease of surface ozone is significant as shown in Fig. 2 whereas the effect on tropospheric column ozone is not evident as shown in Fig. 8.

Referee #3:

Since surface ozone observations are available during different periods (from 1990s to present, though may not be continuous) at some of the sites in South and East Asia. Therefore, some discussion is also warranted on those observations and how do they compare with projected/demonstrated increase during 2030. I have also noted that there are several papers by this group on future projections. Since this MS is based on ozone, mere projected emissions scenarios and influence on ozone may not be sufficient.

Reply:

Surface ozone observation may be available during different periods at different locations. However, in order to evaluate model performance on changes in ozone due to changes in precursor emissions, it is important to use the data obtained at the same locations to avoid effects of spatially inhomogeneous distributions of emission sources and economic growths. In addition, ambient pollutant concentrations in a single year are affected by inter-annual variation in the meteorological fields. Its effects cannot be avoided in a set of two years (Chatani and Sudo, 2011). Therefore, long-term continuous simulations and corresponding observation data at the same locations are required to evaluate model performance on changes in ozone due to changes in precursor emissions. Such an observation data is hardly available in South Asia.

Kurokawa et al. (2009) conducted a simulation over East Asia for springtime during 1981-2005. Chatani and Sudo (2011) also conducted a simulation over East Asia for continuous ten years during 1996-2005. They have revealed that their simulations which is similar to the one of this study has a good performance on the long-term trends of surface ozone continuously observed at the same locations in Japanese network.

The section 3 of this manuscript is devoted to detailed descriptions of our validation of the model performance. We believe this manuscript does not describe mere projected

emissions scenarios and influence on ozone.

The sentences in the line 13 of the page 9520 are replaced by following sentences to show the experiences of regional simulations in South and East Asia.

“Several studies applied regional air quality simulations in East Asia. For example, Yamaji et al. (2008) conducted simulations to predict future surface ozone over East Asia. Kurokawa et al. (2009) and Chatani and Sudo (2011) showed good performances of the simulations to reproduce the trend of surface ozone over Japan for past years. Although applications in South Asia are limited, Kumar et al. (2012) described extensive validation of the regional simulations applied over South Asia. One of the outstanding features of this study is the domain covering China and India together, which are both key developing countries in Asia.”

Referee #3:

I strongly feel that simulation should be made for two sets of year for showing model's capability in demonstration of future projections. This is important for ozone. Somehow, present MS shows significant enhancement in ozone over South Asia, I afraid that it may not be realistic. To have our confidence, it is essential to compare with past observations. I also feel that present MS lacks in-depth scientific discussion and I strongly feel that this MS is not suitable for ACP.

Reply:

The tasks of this study are estimating emissions in the current year and three future scenarios, and simulating changes in tropospheric ozone in scenarios in the same meteorological field. They are independent of estimating emissions for past years and inter-annual variation in the meteorological fields as described in our previous replies. Moreover, in case that ozone chemistry is linear, changes in ozone due to changes in precursor emissions for past years should be consistent for future years. In fact, ozone chemistry is highly complex and highly non-linear as mentioned in the referee comment. Therefore, changes in ozone for past years are not necessarily consistent with changes for future years.

Based on the discussions above, we do not think that simulations for a set of two past years are mandatory tasks to make sure that this study is scientifically valid. On the other hand, we think simulations for multiple coming years are helpful to check if the trend of

tropospheric ozone obtained in this study follows the trend observed in the actual atmosphere.

Following sentences are added as a new paragraph after the line 7 of the page 9539 to mention an importance of simulation for future coming years as one of remaining issues.

“This study predicted potential future changes of tropospheric ozone due to changes in precursor emissions in the same meteorological field. It is desirable to check if the trend of tropospheric ozone for coming years is following the changes simulated in this study to evaluate the effects of existing strategies in the real atmosphere and to consider additional strategies. Long-term continuous monitoring of pollutant concentrations, periodical update of the emission inventory, and simulations for multiple coming years would be helpful.”

Reference

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