

We thank the referee #1 for giving valuable comments. We respond to each specific comment below. The comments and questions from referee #1 are in italic font.

General comment:

My only general concern might be that the results could be somewhat overstated. The last two figures (9 and 10) of the paper show the main results. The in situ results are discarded earlier as they don't match the hypothesis, and Figure 9 does not clearly indicate the satellite results show a decrease in NO₂ during the period of the YOG, so the authors need Figure 10 (NO_x emissions estimates) to prove their point and quantify a 25% reduction in NO_x from the emissions controls. However, from both figures 9 and 10, it's clear there is quite a lot of year-to-year variability. In fact, both 2013 and 2014 in that figure are consistently lower for the summer/fall period than the average (almost looks like a downward trend over previous years?). Also, the only significant reduction shows up in September 2014 (after the games period) due to the lack of observations during August 2014. I realize there is probably an issue of computational power, but an indication of the variability of emissions in Figure 10 from other years (2005-2012) or in the text could be useful. I believe these concerns could be assuaged with some more caveats in the text and the abstract, and I would recommend publication in ACP.

We appreciate the valuable comments from referee #1. Although we still stand behind our conclusions, we weaken our conclusions somewhat in this paper.

We changed the text at page 6353, line 8-10 into:

“This reduction is probably caused by the more permanent air quality regulations taken by...”

And line13:

“This is partly a consequence of the use of monthly means, while the regulations became active at the end of August. It is also a consequence of the lack of.....”

We changed our conclusion given in line 20 on the same page 6353: “We conclude that the NO_x emission reduction detected by DECSO **for the YOG period and afterwards** was at least 25%, showing that the air quality regulations taken by the local government were effective.”

We can see the decrease of the NO₂ concentration during the YOG (August) in figure 9. But we do not know if the decrease is caused by a reduction of NO_x emission during this period or by the rainy weather. That's why we need NO_x emission estimates to check if the air quality regulations applied by the local government were effective.

The NO₂ concentrations are lower in 2013 and 2014 compared to the average of 2005-2012 because there were important events in both years (the Asian Games in 2013 and the YOG in 2014).

Because of the issue of heavy computational demand, we haven't calculate the NO_x emissions from 2005 to 2012 in Figure 10.

Specific Comments:

Thanks for correcting the mistakes of English writing. We revise them in the text. We answer each specific question below. The questions/comments from referee #1 are in italic font.

6338,23: This sentence is a bit vague, maybe give example.

We think we already give an example in the text. “For example, the Nanjing smog episode in December 2013 led to a strong increase in NO₂ concentrations without an increase in NO_x emissions.” We are not sure what kind of example the referee means.

6341,1-4: Martin et al used observed columns, not concentrations for top-down emissions.

We agree with the referee. We change the word “concentration” to “columns”.

6343,4: Is there a reference for the “industry” partitioning?

We don’t have a reference. We estimate the factor table for the situation of China to the best of our knowledge.

The caption of Table 2 is changed to “Table 2. The estimated redistribution of MEIC sectors over SNAP 97 sectors”

The sentence in line 3 page 6343 is changed to “[...] in the CHIMERE model, we estimate the redistribution of the emissions over the sectors (see Table 2).”

6343,7: What is the source of the climatological profiles?

The climatology was compiled from a 2003-2008 run of the global chemistry transport model TM5. We use the same method as described in Mijling and van der A (2012). So we didn’t give detailed information for this issue.

We add the reference on Page 6343 line 5 and mention the source of the climatological profiles: “As mentioned in the paper of Mijling and van der A (2012), to compare CHIMERE simulations with satellite observations, we extend the modelled vertical profiles from 500hPa to the tropopause by adding a climatological partial column, which is from an average of a 2003-2008 run of the global chemistry transport model TM5.”

6344,20: Lin et al. 2014 is missing from reference list.

Thank you for checking the reference and giving another good study to cite. We also noticed that it is missing in the references just after the paper was accepted for ACPD. We add the reference and also cite the paper of Leitaó et al. 2011.

6345, Section 2.3: *This is an interesting way to back out an “in situ NO₂”, which seems hard to find for China, particularly for non-Chinese speakers. Can you give a reference for the Technical Regulation manual? Also, is it complicated to do this? I think it would be useful here to briefly describe this process. Is the AQI a direct function of the NO₂ amount, or is it a single value that includes contributions from the other species (O₃, SO₂, PM). How do you back out the NO₂ amount? Also, you mention errors. Since you use the data to “validate” improvements in your model in Figure 1, but then ignore the data for the analysis of the YOG emissions, I think it would be instructive to give some numerical examples of the uncertainties, maybe by referencing other papers that use in situ data.*

The aqicn.org team publishes the hourly Air Quality Index (AQI) of specific air pollutants, such as NO₂, SO₂, and particulate matter (PM10 and PM2.5). Thus, we calculate NO₂ concentration by converting the AQI of NO₂. To make it clear for readers: We replace the words “different air pollutants” by “specific air pollutants” on line 16 page 6345

We add the link of Technical Regulation on Ambient Air Quality Index in China in line 18 on page 6345:

<http://kjs.mep.gov.cn/hjbhzbz/bzwb/dqhjbh/jcgfffbz/201203/W020120410332725219541.pdf>

We couldn't find any information of the uncertainties of the in-situ observation. We ignore the data for analysis of the concentration during the YOG due to its large daily and monthly variability. The location of the in-situ observation is in the center of Nanjing. As we mentioned on Page 6347, “in urban areas the local sources have transient influences on in-situ observations”. We agree with the conclusion of Blond et al. (2007) and therefore we don't use the daily and hourly in-situ observations to validate the model results. However, we think 8 month of in-situ measurements have enough statistics for validation of the diurnal cycle.

6346, Section 3.1: *Figure 1 shows hardly any bias, but this seems like it might be more coincidence than anything since as you noted earlier, the errors in the in situ data could be large, and also why would that in situ data be representative of the 0.25x0.25 model grid but you ignore the data later on for looking at emissions changes in August 2014 as non-representative of the area? Again, I think it is important to emphasize the uncertainties in the in situ data, even if they match the model well.*

We explained in the last comments.

We add the explanation in line 16:

“Blond et al. (2007) concluded [...] In spite of this, by using the 8-month average of the diurnal cycle to reduce the noise from the in-situ measurements. We see some improvements for these averaged NO₂ concentrations in CHIMERE v2013.”

6348,21: *I'm surprised these might not be systematically biased one way. Can you elaborate on the causes?*

In the text on page 6349 we have added: “The effect of high aerosol concentrations on the NO₂ retrieval is non-linear and depends strongly on both the type of aerosol and its concentration. Also the height of the aerosol layer and the presence of clouds play a role. (Lin et al., 2014, Leitao et al., 2010)”

6350,6: *I'm confused about what you mean by "removed in a single day"*

We re-elaborate the sentence: "At these dates the derived NO_x emissions drop to zero in one day and then slowly increase again to the previous emission levels in the following days."

6350,22: *13:30LT is the overpass time at the equator. What is the typical overpass at Nanjing? Is it closest to 13:00 LT in Nanjing? Since you only look at in situ data by itself, and not in combination with OMI data, why not use all 24-hour data to look for reductions in NO₂? (Conversely, if you did plan to use the in situ data in combination with OMI, you would want to consider using only data in coincidence with OMI overpasses to avoid day-to-day sampling issues, or use a CTM vs. observations scaling factor to correct for OMI sampling.)*

We have checked the typical overpass time at Nanjing for OMI. The average overpass time is 13:30 local time. We add this to the text.

We have made a monthly average plot by using all 24-hour data to look for reductions in NO₂, but we didn't see any reduction. The standard deviation of monthly data by using all 24 hour data is very large, because the NO₂ concentration is very high during night time. We try to compare the monthly average of in-situ observations with OMI data and that's why we use the data of 13LT. However, we still see a high variability in the monthly averaged data, indicating that the data are strongly affected by highly variable local sources (e.g. local traffic) and weather.

For a thorough validation of the OMI observations, the referee gave some good suggestions. However, we think the quality of ground data is not good enough to justify such a validation effort.

6351,3: *This statement implies you know the answer before there is any data to support your hypothesis. Reword this statement.*

Thank you for this comment. We revise the sentence to:

"Therefore, we conclude that the in-situ measurements are not representative for the whole city of Nanjing."

6351,13: *Not clear what is meant by a small trend. Do you mean upwards, downwards, 2013+2014 lower than others, etc...? Expand on this statement.*

There is a small increasing trend of the NO₂ column from 2005 to 2011 in Nanjing.

We change the sentence to:

"Although a small increasing trend from 2005 to 2011 is visible in the satellite data, it is negligible compared to the SD of the natural variability."

6351,20: *Why are concentrations lower for the following months? You discuss the timeline of regulations in Table 1, but nothing past August 31. The lifetime of NO_x isn't such that concentrations would stay low after August. Were regulations kept in place? Elaborate here.*

Several measures taken by government were continued, especially related to NO₂. In Table 1 we have underlined the regulations with a permanent character. Also some less well documented technical improvements have been implemented. At the end of page 6339 we added:

“In addition, several technical improvements have been implemented to reduce pollution from heavy industry and power plants.”

Table 1. Air quality regulations taken by the Nanjing authorities in the year of YOG2014. The period is the start time of different regulations. The underline regulations are effective after the YOG.

| Period | Regulations |
|--|--|
| 1 st May - 30 th June | The local government started to shut down the coal-burning factories |
| 1 st July - 15 th July | All coal-burning factories have been shut down |
| 16 th July - 31 st July | The work on one third of construction sites was stopped. The parking fees in downtown increased sevenfold. |
| 1 st August – 15 th August | The work on 2000 construction sites was stopped. Heavy-industry factories reduced manufacturing by 20 percent. <u>Vehicles with high emissions were banned from the city.</u> Open space barbecue restaurants were closed. <u>900 electric buses and 500 taxis have been put into operation.</u> |
| 16 th August-31 st August | The work at all construction sites was put on hold |

We change the sentence line 20-22 page 6352 to “Due to the effect of the continuous air quality regulations during the YOG and afterwards, the NO₂ concentrations of the following months are also lower than for previous years.”

6353,13: You attribute the high values in August 2014 vs September to cloudy weather and lack of observations. I think it would be instructive to mention here how many OMI observations you actually get for each month. Also, the errors in Figure 10 are fairly consistent month-to-month. I'm not sure exactly how the assimilation works, but wouldn't one expect the errors to be larger for months that have very little observational data, so that August 2014 would have large error, but September would have small error?

The DECSO algorithm is using all measurements in the neighborhood that have been transported to or from the Nanjing region. This most important feature of DECSO has now been emphasized more in the text.

Reductions in emissions at the end of August or the following months can appear with a time lag in the Kalman filter results (see e.g Brunner et al., 2012). This time lag is not fixed but depends

on the amount, interval, accuracy and distance of the observations and it is therefore difficult to quantify. In future research we intend to reduce this time lag by using a Smoothing Kalman Filter technique.

Due to the fact that we use monthly mean values and the Olympic Games took place at the end of the monthly period, the effect will be less obvious in August because of the first half of the month having normal NO_x emission levels. Since many regulations for NO_x had a more permanent character the emission reduction is better visible in September.

And line13 is changed into:

“This is partly a consequence of the use of monthly means, while the regulations became active at the end of August. It is also a consequence of the lack of.....”

At the end of line 5 page 6352, we add:

“The emission estimates use not only satellite observations in the location of the YOG but use all observations over China that are transported from and to Nanjing. Besides transport of air, the meteorological effect on the lifetime of NO_2 is taken into account.”

6354,1: The noise in the observations is not discussed earlier, they are just dismissed as not supporting the conclusions. Elaborate on the dismissal of in situ data earlier in paper, and “noise”.

The in-situ observation has a large hourly and daily variability

We change the sentence into:

“The in-situ observations have a large variability, even after averaging to a monthly means.”

6354,14: NO_2 is only deposited through dry deposition, not wet deposition.

The wet deposition of NO_3 increases due to the rainy weather. NO_3 is one of the reservoir gasses for NO_2 . When NO_3 decreases, it will increase the dry deposition of NO_2 .

We add this explanation in our paper in line14 page 6345:

“because changes in NO_2 concentrations can have more causes such as horizontal transport of NO_2 or increased wet deposition of the NO_2 reservoir gas NO_3 due to the rainy weather.”

6354,22: Again, mention how few observations you have during this period.

During the YOG, the observations over Nanjing are few. But there are observations over other areas near Nanjing. Our DECSO algorithm considers the transport of NO_x emissions. The observations in other places can also affect the emission in Nanjing. We have clarified this in the paper.

6356,9: Again, not clear that it really is reduced from Figure 9. Lots of other months in 2013 and 2014 look low as well.

The deviation of August 2014 from the average value for the years from 2005 to 2012 is three times larger than the standard deviation. This is different from all the other months. This is mentioned in the paper.

Figure 1: I'm confused about what this figure indicates. Is it pure CHIMERE modeled NO₂ or is it OMI-assimilated (as indicated in legend)? Note this in caption.

It is OMI-assimilated. We add this in caption: "Figure 2. The diurnal cycle in Nanjing from January to August 2014 according in-situ observations, OMI-assimilated CHIMERE v2013 and CHIMERE v2006."

Figure 5: It is difficult to see the land borders in panel (b). These figures would be easier to read if the data were plotted with the same limits and scale side-by-side.

We change this figure.

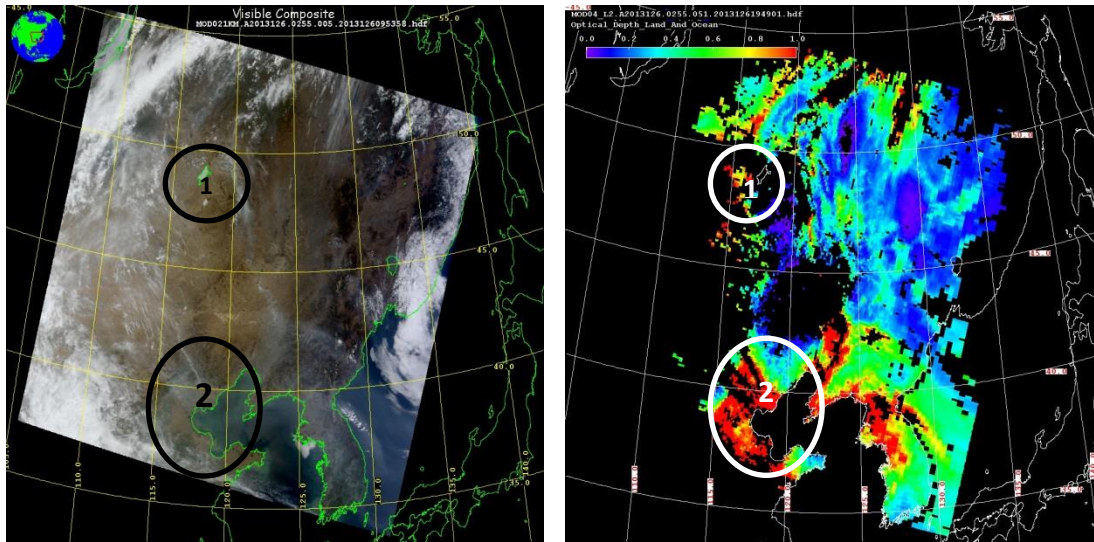


Figure 6. The RGB image (left) and Aerosol Optical Depth (right) from MODIS on 6 May 2013. Circle 1 and Circle 2 represent the Hulunbuir sand land and the Bohai Bay respectively. (The figures are from https://ladsweb.nascom.nasa.gov/browse_images/granule_browser.html)

Figure 8: This may seem picky, but there is no purple in the figure. "Inland water" all seems to show up as blue "ocean".

We also notice that there is no distinction between inland and sea water in the land use data.
We change this figure.

Figure 9 and 10: Line colors are reversed for 2013 and 2014 in Figure 9 and 10. It would be easier to read the figures if they were consistent between the two figures.

Yes, thank you for this comment. We make the colors consistent in these two figures.