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 NO_2 *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 NO_2 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 NO_2 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 NO_2 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 Leitao et al. 2011.

6345, Section 2.3: This is an interesting way to back out an “in situ NO_2”, 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 NO_2 amount, or is it a single value that *includes contributions from the other species (O_3, SO_2, PM). How do you back out the NO_2 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 NO_2, SO_2, and particulate matter (PM10 and PM2.5). Thus, we calculate NO_2 concentration by converting the AQI of NO_2. To make it clear for readers: We replace the words "different air pollutants" by "specific air pollutants" on line16 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/hjbhbz/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 NO_2 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 NO_2 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 NO_2? (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 NO_2, but we didn't see any reduction. The standard deviation of monthly data by using all 24 hour data is very large, because the NO_2 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 NO_2 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 NO_2. 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**
1st May - 30th June	The local government started to shut down the coal-burning factories
1st July - 15th July	All coal-burning factories have been shut down
16th July - 31st July	The work on one third of construction sites was stopped. The parking fees in downtown increased sevenfold.
1st August – 15th August	The work on 2000 construction sites was stopped. Heavy-industry factories reduced manufacturing by 20 percent. Vehicles with high emissions were banned from the city. Open space barbecue restaurants were closed. 900 electric buses and 500 taxis have been put into operation.
16th August-31st 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 NO_2 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.

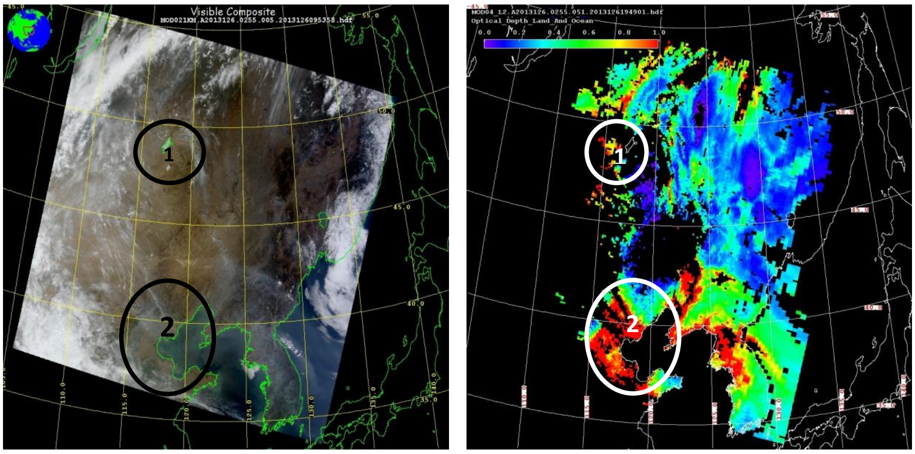

Figure 6. The RGB image (left) and Aerosol Optical Depth (right) from MODIS on 6 May 2013. Circle 1 and Circle 2 represent the Hulunbuir sand land and the Bohai Bay respectively. (The figures are from https://ladsweb.nascom.nasa.gov/browse_images/granule_browser.html)

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.

We thank the referee #2 for giving valuable comments. We think that the paper has been improved based on the suggestions from the referee. We respond to each specific comment below. The comments and questions from referee #2 are in italic font.

In the introduction, the authors cite van der A (2006) regarding the strong NO2 increase over China over the past two decades. I think it would be good scientific practice to acknowledge the earlier scientific literature on this subject (Irie et al., Richter et al. both from 2005).

We agree with this suggestion. We add the reference of Richter et al., 2005 in the introduction (Page 6339) by changing line 9-10 to "For instance, satellite measurements showed that NO_2 column concentrations increased about 50 % from 1996 to 2005 (Irie et al. 2005; Richter et al, 2005; van der A et al., 2006)."

On p. 6343, the authors describe that they extend the modelled NO_2 fields above the CHIMERE upper boundary at 500hPa using a climatological partial column. This gives rise to the following questions:

– How is this column derived? – How is the AK applied to this partial column?

We use the same method as Mijling and van der A (2012). So we didn't give detail information for these two questions. We add the reference on Page 6343 line 5: "As mentioned in the paper of Mijling and van der A (2012), to compare…"

Also on p. 6343, the authors describe an updated version of DECSO. It is not clear to the reader, why some parts of the description of the DECSO improvements are here, i.e., in section 2.1, and others are in the later chapter 3.

The description of the updated version in this part shows the difference of the default version we use in this study and the original version from the paper of Mijling and van der A (2012). We refer to this version as DECSO v3a. More detail information on the improvements leading to DECSO v3a will be published in another paper. However, we add some details in this paper to clarify this to the readers. During the research, we found some problems over the study domain East Asia. We solve the problems by replacing the CTM model and adding a satellite observation filter, This improved version is referred to as DECSO v3b.

On the same subject: The authors fail to give either details or reference regarding the sector-dependent injection height in the model.

We give more details about the sector-dependent emission injection height.

We change this paragraph on Page 6343.

"In this study, we used an updated version of DECSO, which is referred to as DECSO v3a. In particular, the calculation speed has been improved in this update. DECSO does not distinguish between biogenic emissions and the anthropogenic sectorial emissions. Emission differences are attributed to anthropogenic contribution only, i.e. the biogenic emissions are assumed to be modelled correctly by the CTM. Emission updates are distributed by ratio over the sectors (power, industry, transport, domestic) as described by the apriori emission inventory. If a grid cell is dominated by power plant emissions, however, emission updates are attributed to the power sector only. The locations of power plants are provided to the algorithm as additional a priori information. In DECSO v3a, the emission injection height has been made sector-dependent. Emissions are injected in the lowest three model layers of the CTM; each sector having its characteristic vertical emission distribution. For example, transport emissions are released at the surface, while power plant emissions are fully released in the third model layer corresponding at a typical smokestack height."

On the same subject: The authors fail to give either details or reference regarding the used backward trajectory calculations.

We add the following explanation in this part of the text.

"Trajectory calculations of the observed species are crucial in the determination of the source-receptor relations. The DECSO algorithm uses meteorological wind fields (the same as used in the CTM) to calculate how the content of a tropospheric column is advected over the model domain. Here, the injection heights are distributed according the modelled vertical NO_x distribution. In DECSO, the forward trajectory calculation is changed to a backward trajectory calculation, i.e. the source-receptor relations are calculated backward in time, based on the height distribution of NO_x modelled at satellite overpass time."

On the same subject: The authors' re-definition of the observation error Eobs seems arbitrary. Specifically,

– the authors fail to justify this error tuning; they simply describe the effects, but not why it should be a valid assumption to give more weight to larger columns.

Assuming the relative error of observation will be more or less equal. Then during the data assimilation process in DECSO, the error of high observations will be relatively high, thus the weight of this high observation is low. But the low NO_2 observation value with low error has more weight. This will favor the low observations and thus, the emission updates cannot easily capture new emission points or high emission episodes.

We add the reason after the description of the effects by tuning the satellite error on Page 6343 (line 21). "[…absolute error for low values (typically around 0.5 10^{15} molecules cm^{-2}). <u>In this way, DECSO captures better new emission points or high emission episodes."</u>

– the wording the error [. . .] is recalculated is misleading, as it suggests that Eobs is a physically meaningful error estimate. I suggest re-wording this along the lines of DESCO v3 uses tuned synthetic error estimates derived from the original satellite uncertainties via [. . .].

We agree this comment. The sentence on Page 6342 line 14-15 is changed as following:

"In DECSO v3a, tuned synthetic error estimates E_{obs} are used, derived from the original satellite observation via:"

– I fail to see how, using typical Csat of 5E15, f can be anything different than zero, which according to equation (1) just leads to a 50% reduction of the observational error

The C_{sat} is a normalized value from 10^{15}. We add the following explanation in the text (Page 6343 line 18).

"[…]the satellite observations. The unit in this formula is 10^{15} molecules cm^{-2}. The modified […]".

This aim of this formula is to keep the original observation error for low NO_2 observation values and half the original error for high NO_2 observation values. So the high observation values will have more weight during the data assimilation process.

On l. 27 on p. 6344, the authors should justify why they filter out the outmost 4 pixels on either side of the scan.

Considering this comment, we add the following explanation on line 27 Page6344.

"because the size of these pixels is 3 times larger than the model grid cell. After the filtering, the largest footprint is about 75x21 km^2".

In l. 1 on p. 6345, the authors should detail which surface albedo dataset they use for the filtering.

The surface albedo is given in the retrieval data product and this filter is suggested by the DOMINO Product Specification Document on www.temis.nl. We use this surface albedo criterion only to remove the observations over snow and ice.

We modify the sentence on line1 page 6345 into:

"To reduce the influence of bright surface scenes on the quality of the retrieval product, we use only observations having a surface albedo lower than 20% to remove observations over snow and ice (Product Specification Document of DOMINO v2 on www.temis.nl)."

In ll. 8–10 on p. 6345, the authors (quite inspecifically) describe that the selected data is still of sufficient quality; however, they should also discuss the potential influence / additional uncertainties this modified cloud filter criterion has on / adds to the satellite measurements.

We have checked the distribution of the selected satellite data, which remained unchanged. Indeed, the error on the monthly mean data changes. Therefore, we have replaced line 9-11 on page 6345 with:

"From our analysis of the satellite data we conclude that as a result of this new limit on the cloud fraction the error on the measurements increases with less than 20% and without introducing biases. Yet this effect is compensated by the advantage that more data becomes available. The number of observations increases with about 37 % over the whole domain"

On the same subject: The authors claim that the number of observations increases [by] about 37%, however, an increase of 37% on a total number of zero measurements (as given earlier) is still zero measurements.

The emission of Nanjing can be affected by the observations over the whole east Asian domain due to the transport. The transport process of NO_2 concentrations is considered in our DECSO algorithm. The 37% increase of measurements is over the whole domain. This is added in the paper.

In l. 18 on p. 6345, the authors should give the exact URL where the conversion table from the Technicsl Regulation on Ambient Air Quality Index in China is available.

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/hjbhbz/bzwb/dqhjbh/jcgfffbz/201203/W020120410332725219541.pdf

On the same subject: It is not entirely clear which in-situ NO_2 measurements the authors actually use. From what is described in the text, it seems that they do not use the original in-situ measurements but rather calculate the in-situ measurements from the AQI values. However, air quality indices are usually derived from a number of different air quality indicators; the aqicn.org website lists PM, O_3, NO_2, SO_2, CO. Mathematically, the calculation of the AQI is therefore a mapping from an $n-$dimensional state space to a $1-$dimensional value. Therefore, it is not clear how the authors can *derive the NO_2 concentrations which lead to a given AQI value from the AQI (and the mapping table) alone.*

The aqicn.org team publishes the hourly Air Quality Index (AQI) of specific air pollutants, such as NO_2, SO_2, and particulate matter (PM10 and PM2.5). Thus, we calculate the NO_2 concentration by converting the AQI of NO_2.

To clarify this we replace the words "different air pollutants" by "specific air pollutants" on line16 page 6345.

In l. 11 on p. 6346, the authors write that NO emissions cannot be negligible. This sounds incorrect to me; maybe the authors meant to write can be non-negligible?

Yes, we agree and we change the word "*negligible"* to "neglected".

The new sentence in line 11 on page 6346 is:

"The added biogenic emissions can affect the emissions estimated for rural areas as biogenic NO emissions in rural areas cannot be neglected in summertime."

In l. 19 on the same p. 6346, the authors write that land use may have large differences in 15 years. Why don't they just show the land use maps for the Nanjing region from both datasets? Also, the authors should reference their Fig.8 in this context.

Thanks for the valuable suggestion. We moved the Figure 8 to Figure 1 and add the new figure of the land use (GLCF 1994) used in CHIMERE 2006.

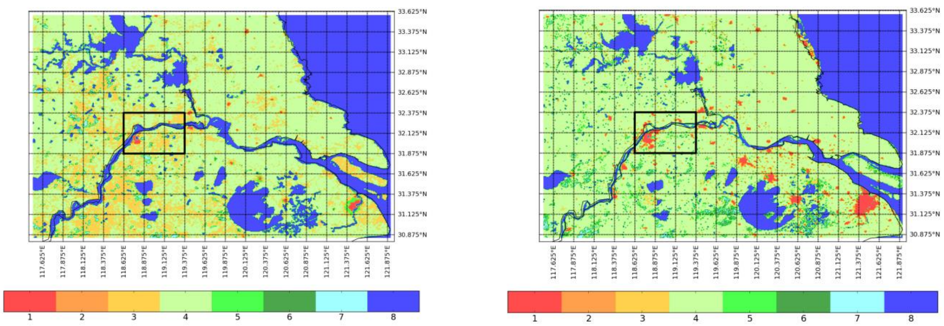

Figure 1. Land use over the Jiangsu Province from Global Land Cover Facility (1994) (left) and the GlobCover Land Cover (2009) (right) and as used in CHIMERE v2006 and

CHIMERE v2013. The 8 categories are: 1. Urban, 2. Barren land, 3. Grassland, 4. Agricultural land, 5. Shrubs, 6. Needleleaf forest, 7. Broadleaf forest, 8. Water. The solid rectangle (about 50 x 90 km^2) indicates the 6 grid cells that cover the Nanjing area.

In l. 22 on the same page 6346, the authors should write the differences between DECSO versions 3a and 3b. In the earlier chapter 2.1, the authors only talked about v3, and now there are two sub-versions, and the authors do not explicitly state their differences.

DECSO v3a uses the old model CHIMERE v2006 and DECSO v3b uses the new version CHIMERE v2013b. The DECSO algorithm we described in part 2.1 is actually DECSO v3a. We change the version name in that part to make it more clear.

In ll. 5f on p. 6347, the authors state that CHIMERE v2013 improves the NO_2 concentrations during night, while improvements during daytime are rather small. However, it is unclear with respect to which reference the concentrations improve. For nighttime, the authors have stated that CHIMERE v2013 gets rid of unrealistically low boundary layer heights which previously lead to unrealistically high NO_2 concentrations, but for the dayime, it is unclear which reference the authors refer to when they notice a small improvement, especially, since they did not point towards any deficiencies of CHIMERE v2006 during daytime.

To make it clear, we modify the sentence in line 5 page 6347 into:

"As expected, v2013 improves the surface concentration simulation at nighttime, while differences during daytime are rather small compared to the in-situ observations."

Again on the matter of improvements of CHIMERE v2013: In l. 16 the authors claim to see some improvements for averaged NO_2 concentrations; but given their own reference to Blond et al. (2007), and given that the surface concentrations used in the present study are unreliable (as previously stated by the authors), maybe the conclusion of noticable improvements is not justified well enough.

We think the 8 month in-situ measurement have enough statistics for the diurnal cycle. We agree with the conclusion of Blond et al. (2007) and that is why we don't use the daily and hourly in-situ observations to validate the model results.

We add 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 of the in-situ measurements, we see some improvements for these averaged NO_2 concentrations in CHIMERE v2013. "

In l. 25 on the same page 6347, the authors see indication of a better performance of CHIMERE v2013 in summertime, based on the OmF they calculate. However, this conclusion would only be valid if the satellite measurements were reflecting the true NO_2 concentrations in the atmosphere. Given that the NO_2 measurements are subject to measurement uncertainties, the statistical significance of the conclusion CHIMERE v2013 is better than CHIMERE v2006 depends on the magnitude of the measurement uncertainties. At least theoretically, it would be possible that the CHIMERE v2006 forecast were more accurate than the CHIMERE v2013 forecast; if now the NO_2 measurements were strongly biased towards the v2013 forecast, this could lead to the observed OmF values. Therefore, I do not see justification for the authors' conclusion that v2013 is better than v2006. I would appreciate at least a comment from the authors on this matter.

The comparison of CHIMERE v2006 and v2013 are done with the same set of NO_2 values. If, as the reviewer suggests the NO_2 observations are biased, the data assimilation will adapt the emissions (i.e. the free parameter) to remove these biases. This means that the analysis in the data assimilation will in general be close to the observations. In the next assimilation step the model error will grow and the OmF values can be used to judge the performance of the model over a single time step. In addition, the OmF distributions of both CHIMERE v2006 and V2013 are checked and did not show any biases. Since this is a common method for data assimilation schemes (see e.g. Data Assimilation: Making Sense of Observations, W. Lahoz, B. Khattatov, R. Menard(Eds), 2010, page 357), we prefer not to add an additional explanation to the text.

In lines 6 f. on p. 6348, the authors write [. . .] the NO_x emissions are almost entirely removed [. . .]. This statement is not understandable to a reader who is not familiar with the DECSO algorithm. I suggest the authors elaborate a bit on this statement so that readers not familiar with DECSO can understand it.

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."

In line 8 on the same page 6348, the authors say that the unrealistic emission updates are related to large OmF values. However, correlation is not causation, so I would appreciate if the authors could modify their statement from a relation to a concurrence.

We agree with this comment.

"These unrealistic emission updates concurred with extreme OmF values (lower than -5 or higher than 10 10^{15} molecules cm^{-2}) with relative small OmF variances[…]"

In line 13 on the same page 6348, the authors should give reference to the PM data set they used for this observation.

Following the reviewer’s suggestion, we add the reference of PM data set here.

“ […] the in-situ observations of PM10 from CNEMC (see section 2.3) show […]”

In line 17 on the same page 6348, the authors write about an underestimation of cloud fraction [. . .] from OMI. However, this statement is only admissible if it were already established that the MODIS cloud fraction is more correct / of higher quality than the OMI cloud fraction. As the authors do not give reference to any study allowing this conclusion, I believe it is not adequate to speak of an underestimation.

We agree with the referee that it is too premature to state that MODIS is giving better cloud fraction values under these circumstances, especially since the relation between aerosols and cloud fraction is rather complex. Therefore we have changed “The underestimation of cloud fraction” into “ “The deviating cloud fraction”.

In line 19 on the same page 6348, the authors should give proper reference to the cloud retrieval algorithm.

We have added the proper reference for the cloud algorithm product that is part of DOMINO2:

Acarreta, J. R., J. F. De Haan, and P. Stammes (2004), Cloud pressure retrieval using the O2–O2 absorption band at 477 nm, J. Geophys. Res., 109, D05204, doi:10.1029/2003JD003915.

Stammes, P., M. Sneep, J. F. de Haan, J. P. Veefkind, P. Wang, and P. F. Levelt (2008), Effective cloud fractions from the Ozone Monitoring Instrument: Theoretical framework and validation, J. Geophys. Res., 113, D16S38, doi:10.1029/2007JD008820.

In line 22 on the same page 6348, the authors give reference to Lin et al. 2014. However, in the reference list of the article, there is no such reference to be found; a proper study to cite in this context would be Leitao et al. 2011.

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 Leitao et al. 2011.

In line 24 on the same page 6348 the authors should remind the reader that the observational uncertainty they are talking about to actually refers to the redefined obsertational error E_{obs} from Eq. 1.

Thanks for the advice. We make a reference here to Equation 1.

In lines 3 f. on page 6349, the authors should explain why haze around the Bohai Bay [. . .] indicates that the high aerosol concentrations are near the surface.

We change the sentence to "The RGB image of MODIS shows haze around the Bohai Bay, which indicates that high aerosol concentrations are present in that area." The height of the aerosols is in fact not important, we just want to stress that high aerosol concentration may have affected the NO_2 retrieval products.

In line 12 on the same page 6349, the authors should explain why they chose the filter criterion they mention, and how they arrived at this criterion.

An OmF filter is a common method in data assimilation to filter out the outliers. In lines 13-27 on page 6349, we explained why we use this particular criterion.

In line 17 on the same page 6349, the authors should explain why the longer NO_2 lifetime can lead to larger OmF values in winter.

The lifetime of NO_2 is much longer in winter than in summer. Therefore, the NO_2 concentration is higher than in summer. Assuming the relative error of the observation is the same in winter and summer, this leads to larger OmF values in winter time. We add a short explanation in the paper.

In lines 13-19 on the same page 6349, the authors should discuss what it means for their conclusions that the distribution of the OmF in winter is clearly not Gaussian (see the heavy tail to the right in Fig. 6b), given that they explicitly state on line 7 of the same page 6349 that /In the data assimilation it is assumed that the OmF distribution is Gaussian.

In data assimilation, the OmF is assumed to be Gaussian distribution. The figure shows that the tails of distribution on both side do not follow the good Gaussian distribution. Therefore we use this OmF criterion to filter out those heavy tails. The distribution after applying the filter show a better Gaussian shape. In the paper, we add that the tails are not Gaussian and filtered.

In lines 9 f. on page 6350, the authors write that /when the pixel size of the satellite is twice that of the model grid cell, the updates of emissions in that grid [cell] will even be doubled/. This statement again is not understandable to a reader who is not familiar with DECSO, so the authors should explain why this is the case. As a side note, to a reader not familiar with DECSO, this sounds like a serious flaw of the DECSO algorithm, so I do see the necessity to explain.

This is probably not explained very well by us, since this statement is completely unrelated to the DECSO algorithm. It is simply that the back-of-the-envelope calculation is done for emissions and concentrations in a single grid cell. If the satellite observation measures an average concentration for twice this area, the total amount of NO_2 will be twice as high and therefore the emissions have to be twice as high to explain this amount of NO_2.

On second thought, we decided to remove this remark in the paper to avoid confusion for the reader.

In line 13 on page 6351, the authors write that they include this in the SD. This statement is not understandable. What is SD? Why do they chose to include the trend in the SD? What does this even mean?

It is the rule of ACP that Standard Deviation is SD and it is automatically changed. We saw that the small trend of NO_2 can be neglected compared to the standard deviation of the NO_2 concentration from year to year.

We replace the previous sentence with: "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."

In line 16 on the same page 6351, the authors speak of a small decrease in [. . .] February. However, I do not see any decrease in Fig. 9 in February.

Compared to the NO_2 concentration in Jan. and Mar., the NO_2 concentration in Feb. is lower than in these two months.

In line 19 on the same page 6351, the unit molec cm-2 cannot be correct with a number 6.6.

Yes, the referee is correct. We change it into 6.6 10^{15} molecules cm^{-2}.

In lines 20 f. on the same page 6351, I the authors write that consequently [. . .] NO_2 concentrations of the following months are also lower than in previous years. Given the short lifetime of NO_2 in the atmosphere, I do not understand the causal connection implied by the authors' use of the word consequently. It would be nice to hear the authors' interpretation of this: Were the pollution control measures prolonged by the authorities? Were they voluntarily continued by the population? Is this a mystery? Also, the authors should define the following months, given that they never clearly stated their study period.

Considering the comments of the referee, we remove the word "consequently". We change the previous sentence into "Due to the effect of the continuous air quality regulations during the YOG and afterwards, the NO_2 concentrations of the following months are also lower than for previous years."

Several measures taken by government were continued, especially related to NO_2. 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."

In line 27 on the same page 6351, the authors should cite previous studies showing that differences [. . .] can be attributed to the meteorological conditions.

We cite a previous study here.

Lin, W., Xu, X., Ge, B., and Liu, X.: Gaseous pollutants in Beijing urban area during the heating period 2007–2008: variability, sources, meteorological, and chemical impacts, Atmos. Chem. Phys., 11, 8157-8170, doi:10.5194/acp-11-8157-2011, 2011.

The authors have to modify the conclusion they give in lines 12-16 on page 6353. From my understanding, the lack of observations in the second half of August 2014 means that it is impossible to decide if the emission reductions shown by DECSO for September 2014 actually occurred in August or September 2014. While I agree that it is highly probable that the emission reductions did indeed occur in August as a consequence of the implemented pollution control strategies, from a scientific point of view, it is impossible to draw this conclusion without doubt. I believe it is absolutely *necessary to explicitly state this uncertainty of the time of emission reduction. Also, looking at Figure 3, it seems that there are no measurements in the Month of September, except for the last days of the month. What is the implication of this for the conclusions? And again, the statement cannot be understood by a reader unfamiliar with the DECSO algorithm, so the authors should add one or two sentences about this.*

We agree with the referee that although highly probable we are not sure the emission reductions did occur at the end of August. There are several reasons that makes it probable:

- Although in Figure 3 no measurements appear in September, this Figure shows only observations directly over the center of Nanjing, while 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 NOx emission levels. Since many regulations for NOx had a more permanent character the emission reduction is better visible in September.

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……..”

Also the conclusion given in line 20 on the same page 6353 has to be modified, as DECSO did not detect any emission reduction for the YOG period, as there were almost no measurements during the YOG period. The conclusion has to be softened.

We soften our conclusion in the revised text.

“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.”

In line 20 of page 6354, the authors should explain why the legislative measures which were only in place until 31 Aug 2014 (see Tab. 1) still effect NO_2 concentrations in the following month, given the short lifetime of tropospheric NO_2.

There are several regulations still effective after the YOG. We underline those regulations in table. 1

In line 27 on the same page 6354, the authors should explain how they arrive at a resolution of 50 x 90km; the spatial resolution of the model was not explicitly given before.

We mentioned the model resolution in Section 2.1 P6342 line 9. The model has a spatial resolution of 0.25x0.25 degree. The solid rectangle showed in figure 8 is about 50x90 km^2. We add this in the caption of the land use figure to make it more clear to the readers.

In lines 12-15 on page 6355 (or earlier, when introducing the concept of the OmF filter), the authors should elaborate why they chose an OmF filter and do not explicitly filter out scenes contaminated by high aerosol loads, using OMI or MODIS AOD measurements.

As shown by Lin et al. (2014) the relation between aerosols and cloud retrievals are complex and non-linear. It is therefore not straightforward to filter deviating NO_2 retrievals based on aerosol information. It depends on the type of aerosols and the concentration as shown by both Lin et al. (2014) and Leitao et al. (2010). The study of the effect of aerosols on NO2 retrievals and how to filter or to improve these retrievals is topic of future research. On the other hand, filtering of outliers in data assimilation by using an OmF filter criterion is not uncommon. The reason why we have not applied such a filter in previous versions of DECSO is already explained in the text. However, in this case we have applied a very cautious version of the OmF filter, which avoids building a complicated filter based on aerosol information of type, concentration and its interaction with clouds.

In the text on page 6349 we have added: "The effect of high aerosol concentrations on the NO_2 retrieval is non-linear and depends strongly on both the type of aerosol and its concentration (Lin et al., 2014, Leitao et al., 2010). It is therefore difficult to filter out outliers in the observed NO_2 based on aerosol data."

In line 24 on the same page 6355, the authors write about high electricity consumptions by power plants. They probably mean high electricity production of power plants.

Yes, the referee is correct. We modify it in the paper.

In Table 1 on page 6361, it is not clear if earlier regulations were complemented or replaced by later regulations. Also, the authors should give reference to the source of information.

The time in the table shows the start time for different regulations. Some of them will be effective after the YOG. We change the table in our paper to make it clear. We got the information in this table from a large range of media, newspaper and internet sources.

In the discussion of Table 2 on page 6362, the authors should clearly state on which grounds they decided on the redistribution factors, or they are arbitrary.

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). ”

In Figure 1 on page 6363, the authors should clarify if the x axis is sun local time or time zone local time.

Sun local time and time zone local time is the same in Nanjing. But we add sun local time in the figure to make it more clear.

In Figure 2 on page 6364, the colorbar does not have any units. What do the colors mean?

The colorbar represents the frequency of satellite observations for that specific value of OmF. We add this in the caption of Figure 2.

In the same Figure 2, it is unclear what is actually shown. Are these time averages of the whole period Jun-Aug? All individual satellite measurements? All individual model grid cells? What is the spatial domain used for this Figure?

They are all individual satellite measurements over the whole Asian domain from Jun. to Aug.

We change the caption of Figure 2 into:

“Figure 3. The comparison of the absolute OmF (10^{15} molecules/cm^2) of CHIMERE v2006 and CHIMERE v2013 for the whole East Asian domain from June to August . The color represents the frequency of satellite observations for that specific value of OmF. ”

For Figure 3 on page 6365, the authors should explain the meaning of the dotted horizontal lines. Also, it is not clear which spatial domain this Figure refers to. Finally, it is not clear what σ_{obs} refers to - is this the same as E_{obs} from Eq. 1?

We remove the dotted horizontal lines in Figure 3. The time series of the OmF is for the single grid cell over the center of Nanjing. We added this to the caption. The error bar is the root mean square error of observations (E_{obs}).

Here is the new caption of figure 3:

“The time series of the OmF from January 2013 to September 2014 for the single grid cell over the center of Nanjing. The error bar is the root mean square error of observations (E_{obs}). ”

Figure 5 on page 6367 lacks any proper reference to the source of the images

We mentioned it in the acknowledgement. But we also add the link to the figure caption.

https://ladsweb.nascom.nasa.gov/

In Figure 7 on page 6369, the authors should clarify if they mean sun local time or timezone local time. Also, given that the vertical bars are not errors but show the natural variability within one month, maybe the authors should not call the bars error bars.

We clarify the local time is sun local time. Indeed the vertical bars show the natural variability and are not error bars. We change the caption of this figure.

In Figure 8 on page 6370, it might be helpful to see a second subplot showing the effectively chosen land cover type for each grid cell, i.e., the scaled-down version of the same data.

CHIMERE model use the information of the percentage of different land use categories. Figure 1 (formerly Figure 8) shows the land use information used by CHIMERE.

In Figure 9 on page 6371, again the authors write error bars even though the vertical bars do not contain error information but show the natural variability for a single month. Also, SD should be spelled out as standard deviation.

We agree with the referee and change the caption. We wrote standard deviation. But the SD is a rule of ACP and automatically changed.

Figure 10 on page 6372 shows that there is a problem with the error of the mean NO_x emission estimate from DECSO (shaded areas): For August 2014, the minimum value of the shaded area is higher than the maximum value for September 2014, i.e., there is no overlap between the credible regions of the emission estimate for August and September 2014. However, the authors do draw the conclusion that the emission reductions seen in September actually happened in August already. I see a contradiction here: Either, the authors' conclusion is correct; in this case, the errors shown as shaded areas in Figure 10 are clearly too small. Or, the errors shown in Figure 10 are correct, but then the authors' conclusion would be merely speculation, as it would not be backed by the error estimates.

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. Although the strong point of a Kalman Filter is its detailed error analysis, this time lag is not incorporated in its formalism. Thus the error of DECSO does not account for the delay. There is no good procedure to calculate the error due to this time lag. In future research we intend to reduce this time lag by using a Smoothing Kalman Filter technique. We add more explanation in our paper.

We thank the referee #3 for giving valuable comments. We respond to each specific comment below. The comments and questions from referee #3 are in italic font.

One concern is the 1-month lag between derived emission reduction (in September) and the actual emission regulation (in August and prior). The lag has been attributed to the issue of Kalmar-based inversion and the lack of OMI data. While this may be the case, a more detailed analysis is needed. This is because the lifetime of NO_x *is short (several hours in summer), and thus there should not be any obvious lag (i.e., more than 1 day) between regulation and reduction. How many days are missing in August compared to other months (e.g., February, when there is no lag)? In addition, all NOx-relevant emission regulations have been implemented by Aug 15, therefore it is not clear why emission reduction is not shown in August and previous months. In particular, Fig. 10 shows that June-August 2014 have higher emissions than June- August 2013, in contrast to the emission regulation starting from May 2014.*

We understand the concern of the referee and have added more explanations to 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. Although the strong point of a Kalman Filter is its detailed error analysis, this time lag is not incorporated in it is formalism. In future research we intend to reduce this time lag by using a Smoothing Kalman Filter technique.

The question how many days are missing cannot be answered straightforwardly, because the DECSO algorithm is using all measurements in the neighborhood that has been transported to or from the Nanjing region. This most important feature of DECSO has been emphasized more in the text. In February no time lag appears because the Spring Festival is usually in the beginning of February, thus a time lag of several days will have no effect on the results.

The difference between 2013 and 2014 are all within the error bars of the emission estimates, so it is difficult to draw any conclusions from that. Due to the fact that we use monthly mean values and the Olympic Games took place at the end of the monthly period, its 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.

At the end of page 3 we added:

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

We changed the text at page 17, 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…….."

The authors make efforts to filter out aerosol-affected data. The criterion is based on NO2 comparison (OmF) rather than on the aerosol amount directly. There is a concern with this practical choice – whether a scene is filtered out or included depends on how "bad" the comparison is rather than depending on the underlined physical reasons (i.e., aerosols). As lots of "outliers" are filtered out, the criteria certainly affect the subsequent emission inversion. Is it possible to compare the chosen filter to an alternative filter where days with aerosols higher than some threshold are excluded? Are there high-aerosol days in the chosen days?

As shown by Lin et al. (2014) the relation between aerosols and cloud retrievals are complex and non-linear. It is therefore not straightforward to filter deviating NO2 retrievals based on aerosol information. It depends on the type of aerosols and the concentration as shown by both Lin et al. (2014) and Leitao et al. (2010). The study of the effect of aerosols on NO2 retrievals and how to filter or to improve these retrievals is topic of future research. On the other hand, filtering of outliers in data assimilation by using an OmF filter criterion is not uncommon. The reason why we have not applied such a filter in previous versions of DECSO is already explained in the text. However, in this case we have applied a very cautious version of the OmF filter, which avoids building a complicated filter based on aerosol information of type, concentration and its interaction with clouds.

In the text on page 6349 we have added: "The effect of high aerosol concentrations on the NO_2 retrieval is non-linear and depends strongly on both the type of aerosol and its concentration (Lin et al., 2014, Leitao et al., 2010). It is therefore difficult to filter out outliers in the observed NO_2 based on aerosol data."

How many data are available for each month?

There are about 1500 OMI observations used in DECSO each day for the whole domain of East Asia.

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.

The OmF filter is based on absolute value rather than percentage value. Considering the seasonality in NO_2, applying the filter means stronger filtering in summer and weaker filtering in winter. This will affect the derived emission seasonality. Please discuss.

We considered to use a percentage value during the research. However, it didn't work well when we used the percentage of the forecast or observation value. For example, if the forecast value is very high, the OmF absolute value could also be very high but the percentage can be small. It still causes the problem that 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. On the other hand, if the forecast or observation value is very low, the OmF absolute value are low enough and doesn't cause any problem in DECSO but the percentage can be higher than the criterion. In general, when we used a percentage value as a criterion, many data causing a wrong emission update were still used, while many data with good quality were filtered out.

We add “Not losing sensitivity to new emission sources is also the reason we do not choose a relative filter criterion” in the paper.

It looks like stronger filtering in summer and weaker filtering in winter. However, the amount of observations are much higher in summer than in winter. After using the same filter for summer and winter, there are still more observations used in summer than in winter.

The choice of asymmetric filtering could be better discussed. Line 24-27 of P6349 is not clear.

Assuming the relative error of observation will be more or less equal. Then during the data assimilation process in DECSO, the error of high observations will be relatively high, thus the weight of this high observation is low. But the low NO_2 observation value with low error has more weight. This will favor the low observations

We add the following sentences at the end of *Line 24-27 of P6349:*

“The observation with low error have more weight in the data assimilation process.”

A looser cloud screening is used to include more OMI pixels. CRF > 50% means less than half of TOA radiance comes from ‘clear-sky’ portion of the pixel. The looser criterion may increase the data noise, which is especially relevant to the daily-based emission inversion. Some relevant figures and quantitative analysis would be welcome.

We have checked the distribution of the selected satellite data, which remained unchanged. Indeed, the error on the monthly mean data changes. Therefore, we have replaced line 9-11 on page 6345 with:

“From our analysis of the satellite data we conclude that as a result of this new limit on the cloud fraction the error on the measurements increases with less than 20% and without introducing biases. Yet this effect is compensated by the advantage that more data becomes available. The number of observations increases with about 37 % over the whole domain”

On the same issue – the ‘back-of-the-envelop’ calculation should be cautiously interpreted, as the lifetime of NO_x could vary dramatically from one day to another, leading to large changes in NO_2 VCD even with the same emissions.

The purpose of back of the envelop calculation is to give readers an impression on the effect of the large OmF on emission estimates. We use seasonal average to reduce the effect of horizontal transportation. The lifetime of NO_x could vary from one day to another. We give the difference of the NO_2 concentration caused by different emissions on an average value. We add this to the paper.

The effect of Chinese Spring Festival has been studied by previous research on satellite NO_2, so as the effects of lifetime versus emission on the seasonality of NO_2. Please give more discussions on or comparisons with these relevant studies.

Wang et al. (2011) showed the monthly NO_2 ground observations of the three cities Beijing, Shanghai and Guangzhou in 2005. There is also a small decrease of NO_2 concentrations in February. But they didn't discuss this small decrease.

Most recent studies we find about the Chinese Spring Festival and NO_x emissions only focus on the emissions from fireworks (usually aerosols) during the Spring Festival and from traffic during the travelling before and after the Spring Festival.

In line 17 page 6351, we add:

"The monthly averages of NO_2 in-situ observations shown by Wang et al. (2011) for Beijing, Shanghai and Guangzhou in 2005 were also reduced by around 10% in February."

At the end of line 7 page 6353, we add:

"Ran et al. (2009) explained high NO_x concentrations in are caused by slower chemical processes and shallow boundary layers contributing to accumulation of NO_x. The table of Wang et al. (2012) annual and summer NO_x emissions from coal-fired power plants in 2005-2007 for different provinces in China showed that the NO_x emissions in Jiangsu Province in summer is higher than mean seasonal emissions."

The model seems to exclude lightning emissions. Lightning emissions are very important in summer, especially considering the much increased sensitivity of OMI to aloft NO2. In addition, lightning emissions vary significantly from one year to another, and a climatological adjustment does not affect the interannual variability. Please discuss the implications of the lightning treatment on the emission inversion here.

Opposite to regions more to the South in China the vertical column of NO_2 from lightning (max. $0.5*10^{15}$ molecules/cm^2) in the region of Nanjing is much less than the NO_2 from anthropogenic sources ($10\text{-}30*10^{15}$ molecules/cm^2) as can be seen in studies of e.g. Schumann and Huntrieser (2007), van der A et al. (2008), and Belmonte Rivas, et al., (2015). Thus 2% of the total column NO_2 can be attributed to lightning and variations in lightning activity will be much lower than 2%. Therefore, we have ignored the lightning contribution in this study, although in other regions this can have important consequences.

Is 'biogenic' emission of NO the same as soil NO_x? If not, does the model consider soil emissions that also peak in summer?

Yes. Biogenic emission of NO is soil emission. We clarified this in the revision.

How is the model error treated?

This is described in the paper of Mijling and van der A (2012) and we have added an additional reference to the paper.

The choice of Eobs in Eq. 1 needs more justification. Also, what is the unit of Csat, Eobs and Esat?

The C_{sat} is a normalized value from 10^{15}. We add the explanation in this part (Page 6343 line 18).

"[…] the satellite observations. The unit in this formula is 10^{15} molecules cm^{-2}. The modified […]".

This aim of this formula is to keep the original observation error for low NO_2 observation values and half the original error for high NO_2 observation values. So the high observation values will have more weight during the data assimilation process.

We add the reason after the description of the effects by tuning the satellite error on Page 6343 (line 21). "[…] absolute error for low values (typically around 0.5 10^{15} molecules cm^{-2}). Thus, DECSO can better capture new emission points or high emission episodes."

The paper cites H. Zhang et al. (2009) for explanation of the large emission seasonality. What is the quantitative result from Zhang et al. and is their result comparable to here?

They showed the trends of electricity consumption in Nanjing from 2000 to 2006. It kept increasing and has strong seasonality during these 7 years. The value of electricity consumption in summer is at least two times higher than in winter every year as shown in their figure. They studied the relation of temperature and electricity consumption and concluded there is a significant correlation between them. We can't quantify the effect on the amount of NO_x emissions from the value of electricity consumptions, since there are also other sources involved. We add "The value of electricity consumption in summer is at least two times higher than in winter every year and keeps increasing during those 7 years." in the text.

We thank the referee #4 for giving valuable comments. We respond to each specific comment below. The comments and questions from referee #4 are in italic font.

This paper by Ding et al. focuses on the estimation of NOx emissions during the 2014 Youth Olympic Games in Nanjing. They constrain daily NO2 column observations from OMI and simulations from the regional CHIMERE model to infer NOx emissions. I agree with one of the reviewers that the most significant results are presented in Figure 9 and 10, which I am most concerned with. I do not think this paper is suitable for publication in ACP unless substantial revisions are made.

I agree with most comments from Reviewer #2. I have few additional comments:

Although we do not know which specific comments the referee is referring to, the responses on those comments can be found in the answers to referee 2.

1) MEIC inventory as well as Zhang et al [2009] inventory suggests small monthly variation in emissions. Emission estimates from the DECSO algorithm is suggesting ~50% higher emissions in July than January. Small drop in February is explained by previous publications by Zhang et al., but the seasonal variation in Figure 10 looks unrealistic based on all existing bottom-up inventories over China. It is most likely coming from deficiency of the DECSO algorithm or the CTM they use. Satellite retrievals may have seasonal bias, yielding seasonal biases in NOx emissions. There are several factors that could lead to the biased inversion. Either exploring those factors or providing enough justification why bottom-up emission is wrong is necessary.

Unfortunately we couldn't infer from literature how the seasonal cycle in MEIC is constructed. We assume that it is similar as described in Zhang et al. (2009). According Zhang et al. (2009) the seasonal cycle is superimposed on the annual inventory. This seasonal cycle is based on monthly activities per province for industrial activities, while for residential emissions it is based on the method of Streets et al. (2003). This seasonal cycle for the residential sector is based on assumptions on the use of stove operation as function of temperature, and is derived for the whole of China.

The difference with our work is two-fold:

1) Our results are for 2013 and 2014, more than 10 years after the analysis of Streets et al. (2003), in which time a lot has changed in the Chinese society. For the city of Nanjing there is practically no coal used for heating, but on the other hand the use of air condition has increased strongly in the last 15 year.

2) The seasonal cycle was applied to the bottom-up inventory on a provincial level or even national level, while we are presenting results at a city-level (Nanjing). As shown in H. Zhang et al., (2009) electricity consumption is higher in summer than for winter for this specific city.

We would also like to add that the used NO_2 satellite data has been validated and show no significant seasonal cycle. (e.g. Boersma et al., 2011 and references therein) .

In the text we added the following discussion: "The difference with the seasonal cycle of MEIC might be attributed to the fact that our results are derived on city-level, while the seasonal cycle for bottom-up inventories are often derived on a national or provincial scale (e.g. Q. Zhang et al. , 2009)."

2) Use of OMI data: There might be some limitations in the understanding and use of OMI data. I think, DOMINO algorithm accounts for aerosol effect through not just cloud information as discussed in the paper but also surface reflectivity (OMI LER). Exclusion of scenes with high aerosols may remove polluted days since high aerosols may occur for days with high NO_2 pollution. Results from Lin et al., who use MODIS reflectivity and model aerosol, may not provide sufficient justification as the study did not examine the relationship between Kleipool LER with LER calculated from MODIS reflectivity and observed aerosol. Discussions on the application of averaging kernel are necessary since the idea here is to replace the TM4 profiles used in retrievals by DECSO profiles. Section title "Improvement of the satellite data" is misleading because this work does not improve any aspect of retrieval algorithm and satellite data product. Better title would have been "Data selection and pre-processing" or something similar. 70% cloud radiance fraction threshold is higher than many previous studies use. Since cloudy observations have larger errors, inversion is more error prone with higher threshold. Criterion for OmF is very subjective. Why choose the range [-5, 10] e15? Why not [-5, 5] e15? Why not relative value rather than absolute value? Why not percentile range? Is the selected range applicable to any region or just over China?

- Although some of our authors have a thorough understanding of the DOMINO algorithm, since they are involved in the development of DOMINO for many years, we think discussing the performance/flaws of the DOMINO algorithm and the related findings of the Lin et al. is out of scope of this paper and not relevant for our results. We only conclude, based on the data assimilation results, that a very small fraction of the DOMINO NO_2 data is unrealistic and that these observations always occur in the presence of high aerosol concentrations.

- We agree that the title is not entirely correct and we changed the title of the section into "Quality control of satellite data"

- Although a limit of 70 % for the cloud fraction on average increases the error of the measurements by less than 20%, this is compensated by the 37% increase in the number of observations.

- Although the exact limit to filter outliers in OmF values is always arbitrary, we give a detailed description why we choose these limit values (page 6349, line 13 - page 6350, line 18). The asymmetric limits are chosen because the OmF distribution is asymmetric in its tails, caused by the fact that there are no negative NO_2 observations, but there is virtually no limit to the maximum NO_2. To use a relative factor will filter a lot of data for the low NO_2 concentrations where we have found no problems with the observations. In the end, we have been deliberately on the cautious side with our choice of this criterion and less than 3% of the data is filtered over China. The limit is also applicable for other regions.

In our discussion we added on page 6349,

On line 11: "….new power plants. <u>Not losing sensitivity to new emission sources is also the reason we do not choose a relative filter criterion</u>. We select…."

On line 12: "We select a OmF filter criterion in the range of [-5, 10] x 10^{15} molecules cm^{-2} <u>based on our analysis discussed below</u>."

On line 14: "….. September 2014is Gaussian <u>except for its tails and 97% of ….</u>"

3) Use of surface data: I do not understand the logic of using surface data. There is a big unknown about the quality of the surface NO_2 data they use for validation of the model results. How the (comparison) exercise is going to be insightful if the accuracy of the data used in the analysis is unknown? In addition, the comparison of NO_2 at a surface site with model results at 0.25x0.25 is not really helpful.

This is exactly what we argue in the manuscript: the validation by comparison with surface data is of very limited value. Therefore, we use only hourly values averaged over a long time period of 8 months for the comparison. Since the surface data is available, we did not want to ignore this source of information for our model validation. For the sake of completeness we feel obliged to present our conclusions on the comparison with ground data.

4) Data analysis: Based on information presented in Table 2, it is more logical to focus the analysis for May-September period examining how each regulation was effective in reducing pollution level. From Figure 9, it is difficult to link the changes observed in the YOG period to regulations in place as the results are similar for 2013 and 2014. Authors state in introduction that derived emission is better to study the effectiveness of the air quality measures, but it is unclear to me how satellite-derived emission is better than satellite observations themselves as the model is not providing any additional information regarding regulations. In fact, one might introduce model errors in the inferred emissions. For the nature of the work presented in the paper, I do not see much advantage of the chosen approach.

As argued here and in previous papers (e.g. Mijling et al., 2009) this analysis based on satellite concentrations only is not sufficient, since meteorological conditions and transport of polluted air are strongly affecting the concentrations. This means that there is no linear relation between air quality regulations and NO_2 concentrations. On the other hand the relation between regulations and emissions is very direct and linear, which not only justifies our approach but it is also an <u>improvement</u> on the analysis of measured NO_2 concentrations.

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 taking transport into account the meteorological effect on the lifetime of NO_2 is taken into account.

Indeed, this is, as pointed out by the referee, shown by the fact that Figure 9 is less convincing that the results in Figure 10. Figure 9 shows only concentrations, while Figure 10 is showing the emissions taken into account transport and meteorological conditions leading to actually <u>smaller</u> errors on the results.

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 taking transport into account the meteorological effect on the lifetime of NO_2 is taken into account."

5) Several statements in the "Model improvement" section require citations. Please, use NO2 columns consistently instead of NO2 concentrations and NOx emissions instead of NO2 emissions.

The Chimere model is described in section 2.1 with references to Schmidt, 2001; Bessagnet et al., 2004; and Menut et al., 2013.

We have made the use of "NO_2 columns" and "NO_x emissions" more consistent throughout the text.

NO_x emission estimates during the 2014 Youth Olympic Games in Nanjing

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Abstract

The Nanjing Government ~~has taken~~applied temporary environmental regulations to guarantee good air quality during the Youth Olympic Games (YOG) in 2014. We study the effect of those regulations by applying the emission estimate algorithm DECSO (Daily Emission estimates Constrained by Satellite Observations) to measurements of the Ozone Monitoring Instrument (OMI). We improved DECSO by updating the chemical transport model CHIMERE from v2006 to v2013 and by adding an Observation minus Forecast (OmF) criterion to filter outlying satellite retrievals due to high aerosol concentrations. The comparison of model results with both ground and satellite observations indicates that CHIMERE v2013 is better performing than CHIMERE v2006. After filtering the satellite observations with high aerosol loads that were leading to large OmF values, unrealistic jumps in the emission estimates are removed. Despite the cloudy conditions during the YOG we could still see a decrease of tropospheric NO_2 column concentrations of about 32% in the OMI observations as compared to the average NO_2 ~~concentrations~~ colums from 2005 to 2012. The results of the improved DECSO algorithm for NO_x emissions show a reduction of at least 25% during the YOG period and afterwards. This indicates that air quality regulations taken by the local government ~~were successful~~have effect in reducing NO_x emissions. The

algorithm is also able to detect an emission reduction of 10% during the Chinese Spring Festival. This study demonstrates the capacity of the DECSO algorithm to capture the change of NO_x emissions on a monthly scale. We also show that the observed NO_2 columns ~~concentrations~~ and the derived emissions show different patterns that provide complimentary information. For example, the Nanjing smog episode in December 2013 led to a strong increase in NO_2 concentrations without an increase in NO_x emissions. Furthermore, DECSO gives us important information ~~of~~ on the non-trivial seasonal relation between NO_x emissions and NO_2 concentrations on a local scale.

1 Introduction

Reducing air pollution is one of the biggest environmental challenges currently in China. Nearly 75% of urban areas are regularly polluted in a way that ~~is~~ was considered unsuitable for their inhabitants in 2004 (Shao et al., 2006). In mega cities and their immediate vicinities, air pollutants exceed the Chinese Grade-II standard (80 μg m^{-3} for daily NO_2) on 10-30% in~~of~~ the days (Chan and Yao, 2008). Air pollution is directly related to the economic growth in China and its accompanying increase of energy consumption. In the last two decades, air pollutants persistently increased in China. For instance, satellite measurements showed that NO_2 column concentrations increased about ~~20% per year~~50 % from 1996 to 2005~~-~~ (Irie, 2005; Richter et al., 2005; van der A et al., 2006). By combining satellite observations with air quality models, Itahashi et al. (2014) showed that the strong increase of NO_2 ~~concentrations~~ columns over East China ~~is~~ has been caused by a doubling of NO_x (NO_x=NO+NO_2) emissions during 2000 to 2010. Zhang et al. (2007) found that NO_x emissions increased with 70% between 1995 and 2006 and Lamsal et al. (2011) found that anthropogenic NO_x emissions increased ~~with~~ 18.8% during the period 2006 to 2009.

Nanjing, the capital of Jiangsu Province, is a highly urbanized and industrialized city located in East China, in the northwest part of the Yangtze River Delta (YRD). By 2012, the area of Nanjing had a population of 8.2 million (Nanjing statistical Bureau, 2013). The YRD is one of the largest economic and most polluted regions in China. Tu et al. (2007) found that the largest fraction of air pollution by NO_x and SO_2 can be attributed to local sources in Nanjing. Li et al. (2011) concluded that air pollutant concentrations and visibility demanded urgent air pollution regulations in the YRD region. From 16th to 29th August 2014, the Youth Olympic Games (YOG) was held in Nanjing. To guarantee good air quality during the Games, the city government carried out temporary strict environmental regulations with 35 directives from May to August. Other cities in the YRD cooperated with Nanjing to ensure good air quality during the Games. The periods with the main regulations are shown in Table 1. ~~-~~In addition, several technical improvements have been implemented to reduce pollution from heavy industry and power plants.

~~Also for~~For previous major international events in China, local authorities have tried to comply with the air quality standards of the World Health Organization (WHO), which has a limit of 200 µg m^{-3} for hourly NO_2 concentrations. For each event, the local government imposed restrictions on heavy industry, construction and traffic. In 2008 the Beijing Municipal Government implemented a series of air pollution control measures for Beijing and surrounding cities to guarantee good air quality for the 29th Olympic Games. These control measures significantly reduced the emissions and concentrations of pollutants. Satellite data show the NO_2 column concentrations decreased ~~with~~ at least 40% compared to previous years (Mijling et al., 2009; Witte et al., 2009). Both bottom-up and top-down emission estimates show a decrease of about 40% in NO_x emissions (Wang et al., 2009; Wang et al., 2010, Mijling et al., 2013). During the 2010 World Expo in Shanghai the NO_2 ~~concentration~~ column was reduced by 8% from May to August according to an analysis of Hao et al. (2011) of space-based measurements compared to previous years. In November 2010 emission reduction measures introduced by the Guangzhou authorities also successfully improved air quality for the Asian Games. Wu et al. (2013) claimed a NO_x emission reduction of 43.5% based on mobile DOAS measurements. The emission reduction of NO_x based on model simulations was estimated to be about 40% (Liu et al., 2013).

However, to study the effectiveness of the air quality measures, it is not enough to look at the concentration measurements alone, as the reduction of air pollutants can also be affected by favorable meteorological conditions. Emissions need to be derived to better show the effect of temporary air quality regulations carried out for the Games. Up-to-date emission data is difficult to obtain, as most emission inventories are developed by a bottom-up approach based on statistics on source sector, land-use and sector specific emission factors.

The bottom-up approach introduces large uncertainties in the emission inventories. To improve emission inventories, a top-down approach can be used by estimating emissions from satellite observations (Streets et al., 2013). For constraining emissions of short lived species, Martin et al. (2003) used the ratio of the simulated to the observed concentration to scale a priori emissions. They used optimal estimation to weigh the a priori emission inventory with the top-down estimates, resulting in an a posteriori inventory with error estimates. This method assumes that the relationship between emissions and concentrations is not affected by transport. Non-linear and non-local relations between emission and concentration can be indirectly solved by applying the method iteratively (e.g. Zhao and Wang (2009)), although a posteriori error estimates are lost in this way. Kurokawa et al., (2009) and Stavrakou et al. (2008) used 4DVAR techniques to estimate emissions by applying an adjoint model of the chemistry transport model to calculate the sensitivities. Another popular data assimilation method is the Ensemble Kalman Filter (Evensen, 2003), which does not require an adjoint model and is relatively easy to implement. As an extension of the Kalman filter, it employs a Monte Carlo approach to represent the uncertainty of the model system with a large stochastic ensemble. Whenever the filter requires statistics such as mean and covariance, these are obtained from the sample statistics of the ensemble (Miyazaki et al., 2012).

To get fast updates for short lived air pollutants, Mijling and van der A (2012) designed a Daily Emission estimates Constrained by Satellite Observation (DECSO) algorithm. DECSO is an inversion method based on an extended Kalman filter. The algorithm only needs one forward model run of a chemical transport model (CTM) to calculate all local and non-local emission/concentration relations. It updates emissions by addition instead of scaling, enabling the detection of unaccounted emission sources.

In this study, we use the latest version of DECSO with OMI satellite data to study how the environmental regulations affect the NO_x emissions in Nanjing during the 2014 YOG. Detecting emission changes for Nanjing is challenging, as it is a smaller city than e.g. Beijing. In addition, Nanjing is in one of the most populated areas of China close to Shanghai with a population of about 24 million. Therefore we have introduced a few improvements in the DECSO algorithm to better resolve small scale emission changes in time and location. The improvements consist of an updated CTM and better filtering of erroneous satellite observations. The emission estimates will be based on the satellite observations of OMI, taking advantage of its high spatial resolution needed to resolve the changes in the Nanjing area. With this improved algorithm we will compare the NO_x emissions during the YOG with NO_x emissions of the previous year in the Yangtze Delta River.

2 Methods

2.1 Emission estimates

For the emission estimates of NO_x over China we use the DECSO algorithm (Mijling and van der A, 2012). It uses a CTM to simulate the NO_2 concentrations and daily satellite observations of NO_2 column concentrations to constrain NO_x emissions. The algorithm is based on an extended Kalman filter to get new emission estimates by optimizing NO_2 column concentrations of model and satellite observations. The inclusion of sensitivities of NO_2 column concentrations on the NO_x emissions in other locations is an essential part of DECSO. A terrain-following trajectory analysis is used in this calculation to describe the transport of NO_2 over the model domain for a time interval between two overpasses of the satellite instrument. This approach results in a fast algorithm suitable for daily estimates of NO_x emissions on a 0.25^o x 0.25^o resolution. A detailed description of DECSO v1 can be found in Mijling and van der A (2012).

The CTM used in DECSO is CHIMERE (Schmidt, 2001; Bessagnet et al., 2004; Menut et al., 2013). CHIMERE is implemented on a 0.25^o x 0.25^o spatial grid over East Asia from 18^o N to 50^o N and 102^o E to 132^o E. It contains 8 atmospheric layers up to 500 hPa. The meteorological input for CHIMERE is the operational meteorological forecast of the European Centre for Medium-Range Weather Forecasts (ECMWF) with a horizontal resolution of approximately 25x25 km^2. The Multi-resolution Emission Inventory for China (MEIC) (He, 2012) for 2010 gridded to a resolution of 0.25^o x 0.25^o, is used for the initial emissions in DECSO. Outside China, where no MEIC emissions are defined, the emission inventory of INTEX-B (Zhang et al., 2009b) is used. As the emission sector definition used in MEIC and INTEX-B does not match the 11 activity sectors according to the SNAP (Selected Nomenclature for Air Pollution) 97, which are internally used in the CHIMERE model, we estimate the redistribution ofe the emissions over the sectors according to(see Table 2).

As mentioned in the paper of Mijling and van der A (2012), tTo 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. The simulated NO_2 column concentrations on the model grid are redistributed to the satellite footprints. To enable direct comparison between simulated and observed tropospheric vertical column, the averaging kernel from the satellite retrieval is then applied to the modelled vertical profile.

In this study, we used an updated version of DECSO, which is referred to as DECSO v3a. ~~Especially~~ In particular, the calculation speed has been improved in this update. DECSO does not distinguish between biogenic emissions and the anthropogenic sectorial emissions. Emission differences are attributed to anthropogenic contribution only, i.e. the biogenic emissions are assumed to be modeled correctly by the CTM. Emission updates are distributed by ratio over the sectors (power, industry, transport, domestic) as described by the apriori emission inventory.~~Furthermore, the emission injection height has been made sector-dependent~~ If a grid cell is dominated by power plant emissions, however, emission updates are attributed to the power sector only. The locations of power plants are provided to the algorithm as additional a priori information. In DECSO v3a, the emission injection height has been made sector-dependent. Emissions are injected in the lowest three model layers of the CTM; each sector having its characteristic vertical emission distribution. For example, transport emissions are released at the surface, while power plant emissions are fully released in the third model layer corresponding at a typical smokestack height. -Trajectory calculations of the observed species are crucial in the determination of the source-receptor relations. The DECSO algorithm uses meteorological wind fields (the same as used in the CTM) to calculate how the content of a tropospheric column is advected over the model domain. Here, the injection heights is distributed according to the modeled vertical NO_x distribution. In DECSO, the forward trajectory calculation is changed to a backward trajectory calculation, i.e. the source-receptor relations are calculated backward in time, based on the height distribution of NO_x modelled at satellite overpass time.

~~and the forward trajectory calculation is changed to a backward calculation.~~ In DECSO v3a, ~~the~~ tuned synthetic error estimates E_{obs} ~~of~~ estimates are used, derived from the original ~~a~~ satellite observation via ~~is recalculated according to~~:

$$E_{obs} = f \cdot E_{sat} + (1-f) \cdot (0.5 \cdot E_{sat}),\ with\ f = e^{\left(-\frac{c_{sat}}{2}\right)} \quad (1)$$

where E_{sat} is the original observation error from the retrieval method and C_{sat} is the retrieved NO_2 column of the satellite observation. The unit in this formula is 10^{15} molecules cm^{-2}. The modified errors give more weight to satellite observations with high values during the assimilation by reducing their relative error while maintaining the dominating absolute error for low values (typically around 0.5 10^{15} molecules cm^{-2}). In this way, DECSO captures better new emission points or high emission episodes.

2.2 Satellite observations

In this study, satellite observations from the Dutch-Finnish Ozone Monitoring Instrument (OMI) on NASA's (National Aeronautics and Space Administration) Aura satellite (Levelt et al., 2006) are used in DECSO. The satellite was launched on 15 July 2004 into a sun-synchronous polar orbit at 705 km altitude. OMI is a nadir-viewing spectrometer measuring the atmosphere-backscattered solar light in the ultraviolet-visible (UV/VIS) range from 270 to 500 nm with a spectral resolution of about 0.5nm. The 114^{o} wide view of OMI results in a swath width of 2600 km, providing daily global coverage in about 14 orbits. The local overpass time is around 13:30 local time (LT). The pixel size of OMI is 24x13 km^2 at nadir and increases to about 150x28 km^2 at the end of the swath.

We use the tropospheric NO_2 vertical column concentrations retrieved with the Dutch OMI NO_2 retrieval (DOMINO) algorithm version 2 (Boersma et al., 2011). The dataset is available on the Tropospheric Emissions Monitoring Internet Service (TEMIS) portal (http://www.temis.nl). The DOMINO algorithm first obtains NO_2 slant columns from the OMI reflectance spectra by using Differential Optical Absorption Spectroscopy (DOAS). After separating the stratospheric and tropospheric contribution to the slant column, DOMINO converts the tropospheric slant column to a vertical column with the tropospheric air mass factor (AMF) (Boersma et al., 2007, 2011). DOMINO v2.0 mainly improves the NO_2 air mass factor by improved radiative transfer, surface albedo, terrain height, clouds and a priori vertical NO_2 profiles. The bias between DOMINO v2.0 and Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) ground observations at 5 locations is only -10±14% over China and Japan (Irie et al., 2012). The DOMINO algorithm does not explicitly account for the effect of aerosols on the solar radiation. Rather it is indirectly accounted for by the higher cloud fraction in aerosol contaminated scenes. However, Lin et al. (2014) concluded that especially in China the effects of aerosols and surface reflectance anisotropy have implications for retrievals of NO_2 from OMI and suggested that exclusion of high aerosol scenes supports better emission estimates at fine spatial and temporal scales.

Since 25 June 2007, OMI data has been affected by the so-called row anomaly, which deteriorates the spectral observations for particular viewing directions of OMI (Boersma et al., 2011; Kroon et al., 2011). 29 out of the 60 rows are affected by the row anomalies and no longer used after 1 January 2011. We also filter out the 4 pixels at either side of the swath,~~.~~ because the size of these pixels is 3 times larger than the model grid cell. ~~This makes~~After the filtering, the largest footprint is about 75x21 km^2. To reduce the influence of cloudy and bright surface scenes on the quality of the retrieval product, we use only observations having a surface albedo lower than 20% to remove observations over snow and ice (Product Specification Document of DOMINO v2 on www.temis.nl). The observations with clouds below 800 hPa are also filtered out as these retrievals are very sensitive to small differences in the NO_2 profile shape and the retrieved cloud height. Mijling and van der A (2012) filter out the observations with a cloud fraction higher than 20%. Based on this filtering, there are no tropospheric NO_2 satellite observations over Nanjing during the YOG due to the cloudy conditions at the overpass time of the satellite. Thus, to obtain more NO_2 satellite observations, we use observations with a cloud radiance fraction lower than 70% (comparable with a cloud fraction of about 30-35%) instead of the cloud fraction lower than 20%. From our analysis of the satellite data we conclude that as a result of this new limit on the cloud fraction the error on the measurements increases with less than 20% and without introducing biases. Yet this effect is compensated by the advantage that more data becomes available. The number of observations increases with about 37 %.~~After checking the monthly averages and the distribution of the satellite data, we find that the selected data is still of sufficient quality while the number of observations increases with about 37%~~over the whole domain.

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2.3 Ground-based observations

To validate the model results in Nanjing, we use available independent measurements from the national in-situ observation network, which are collected and maintained by the China National Environmental Monitoring Center (CNEMC). The aqicn.org team publishes the hourly Air Quality Index (AQI) of ~~different~~ specific air pollutants, such as NO_2, SO_2, and particulate matter (PM10 and PM2.5), on their website based on the measurements from CNEMC. The AQI is calculated by the conversion table from the Technical Regulation on Ambient Air Quality Index in China published by the Ministry of Environmental Protection (http://kjs.mep.gov.cn/hjbhbz/bzwb/dqhjbh/jcgfffbz/201203/W020120410332725219541.pdf). We use the same table to convert the AQI back to the surface concentration unit of μg m^{-3}. For this study, the NO_2 hourly ~~ground~~ in-situ measurements of Nanjing for the period of April 2013 to December 2014 are used. The location of these measurements is the Nanjing People's Government building, which is located in the center of Nanjing. Interpretation of the validation results is troubled by the absence of peripheral information of the ~~ground~~ in-situ measurements. For instance, the type of instrument is unknown and the exact location of the measurement such as the height or the distance to a local traffic road is unclear.

3 Improvements of DECSO

3.1 Model improvement

The performance of the CTM is important for the DECSO results. CHIMERE v2006 is an outdated model version which has been used in DECSO algorithm versions up to v3a. To improve the emission estimation results, we updated the CTM to CHIMERE v2013 (DECSO v3b).

The new model adds biogenic emissions of six species: isoprene, α-ioporene, α-pinene, β-pinene, limonene, ocimene and NO. These biogenic emissions are calculated by the model preprocessor using the MEGAN model and land use data (Menut et al., 2013). The added biogenic emissions can affect the emissions estimated for rural areas as biogenic NO emissions in rural areas cannot be ~~negligible~~ neglected in summertime. Compared to the old version of CHIMERE, the new model version includes a more advanced scheme for secondary organic aerosol chemistry. In addition, the chemical reaction rates are updated and a new transport scheme is used in the new CHIMERE model. For CHIMERE v2013 we use the same input data except for the land use data. We use land use data from the GlobCover Land Cover (GCLC version 2.3) database, which is updated for the year 2009, while the land use database included in CHIMERE v2006 is the Global Land Cover Facility (GLFC) giving the land use of 1994. As China is a fast developing country, the land use may have large differences in 15 years due to urbanization (see Figure 1). Thus, the updated land use database will positively affect the model simulations over China.

To assess the effect of the new CTM, we run DECSO v3a and DECSO v3b for the period January 2013 to August 2014. Figure ~~1~~ 2 shows the comparison of the average diurnal cycle of surface NO_2 concentrations from the two CHIMERE models with in-situ observations in Nanjing averaged for January to August 2014. We select the 0.25^ox0.25^o model grid cell that contains the in-situ measurement location. According to GCLC database, 70% of the grid cell is urban area. We see that the surface NO_2 concentration of CHIMERE v2013 during nighttime is closer to the observations than for CHIMERE v2006. Our earlier model evaluations of CHIMERE showed that the nocturnal surface NO_2 concentrations simulated by CHIMERE v2006 are usually too high in urban areas caused by unrealistically low boundary layer heights and too little vertical diffusion. In CHIMERE v2013, the boundary layer heights over urban areas are limited by a minimum boundary layer height. As expected, v2013 improves the surface concentration simulation at nighttime, while ~~improvements~~ differences during daytime are rather small compared to the in-situ observations. We calculate the bias and Root Mean Square Error (RMSE) between the model results and in-situ observations. The bias of CHIMERE v2013 is 3.7 μg m^{-3} which is 10 μg m^{-3} smaller than for CHIMERE v2006. The difference of RMSE between the two models is very small, the RMSE of CHIMERE v2013 is 28 μg m^{-3} and of CHIMERE v2006 is 31 μg m^{-3}. For the satellite overpass time, the bias improves from 4.4 to 1.8 μg m^{-3} while the RMSE remains the same. However, in urban areas the local sources have transient influences on in-situ observations. Blond et al. (2007) concluded that urban in-situ observations of NO_2 cannot be used for the validation of a CTM model with low spatial resolution because the representativeness of the in-situ measurement for the grid cell is very low. In spite of this, by using the 8-month average of the diurnal cycle to reduce the noise from the in-situ measurements, we ~~still~~ see some improvements for averaged NO_2 concentrations in CHIMERE v2013.

In order to get a more comprehensive validation of the model results, we compare the two CHIMERE models with OMI satellite observations. During the data assimilation of DECSO the daily "Observation minus Forecasts" (OmF) values have been stored. The OmF is a common measure for the forecasting capabilities of the model in the data assimilation. We compare the absolute OmF of both models for the summer (June to August) of 2014 in Figure ~~2~~3. In the Figure a linear regression is fitted through the data points that shows the absolute OmF of CHIMERE v2013 is lower than that of CHIMERE v2006 indicating a better performance of CHIMERE v2013 in summertime. However, the absolute OmF of two models is similar in wintertime. Since biogenic emissions are negligible in wintertime, this may point to an effect of the missing biogenic emissions in the older version of CHIMERE. Based on these comparisons we selected CHIMERE v2013 in DECSO v3b for NO_x emission estimates in this study.

3.2 Quality control of satellite data ~~Improvement of the satellite data~~

Earlier studies showed that the DOMINO v2 retrievals do not account enough for the effect of high aerosol concentrations on NO_2 columns (see section 2.2) and at the same time we know that high aerosol concentrations are a significant problem in most mega cities in China. When checking the time series of NO_x emissions over Nanjing for 2013 by DECSO v3b, we find some suspicious fluctuations at particular days. At these dates the derived NO_x emissions ~~are almost entirely removed~~drop to zero in one day and then slowly increase again to the previous emission levels in the following days. These unrealistic emission updates ~~are related~~ concurred with ~~to~~ extreme OmF values (lower than -5 or higher than 10 10^{15} molecules cm^{-2}) with relative small OmF variances, which are calculated as the quadratic sum of model and observation errors (Figure ~~3~~4). In the time period of our study there are 20 days with these extreme OmF values, 6 are positive and 14 are negative. All are having a significant impact on the NO_x emissions. For most of those 20 days, the in-situ observations of PM10 from CNEMC (see section 2.3) show high aerosol concentrations, which are above 100 μg m^{-3} in Nanjing. We also see a strong haze above Nanjing for all the 20 days from visual inspection of the MODIS RGB images. In addition, we noticed that the MODIS images show higher cloud fractions than the fractions retrieved from OMI observations. The ~~underestimation~~ deviating of cloud fraction information from the OMI satellite retrieval is probably due to the aerosol conditions, which are not taken into account in the cloud retrieval algorithm(Acarreta et al., 2004; Stammes et al., 2008). High aerosol concentrations can not only complexly affect the cloud fraction and cloud pressure retrieval but also directly affect the NO_2 retrieval and results in either over- or under- estimated NO_2 column concentrations (Lin et al., 2014).

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Figure ~~4~~ 5 shows an example of such an extreme case for East China on 6 May 2013 with high (positive) OmF values in combination with low observational uncertainties (Eq. 1). In the image we identify two areas with satellite observations that are at least 10 10^{15} molecules cm^{-2} higher than the model forecast. One is over the Hulunbuir sand land at the border of China and Mongolia, the other one is around the Bohai Bay. We compared the observations with the MODIS RGB and Aerosol Optical Depth (AOD) images on that day (Figure ~~5~~6). The MODIS AOD image shows high aerosol values around the Bohai Bay and over the Hulunbuir sand land. The RGB image of MODIS shows haze around the Bohai Bay, which indicates that ~~the~~ high aerosol concentrations are ~~near the surface~~presented in that area. However, the aerosol information is not used in the retrieval of the DOMINO NO_2 product leading to NO_2 observations that are strongly deviating from the model forecast.

The effect of high aerosol concentrations on the NO_2 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 (Leitão et al., 2010; Lin et al., 2014). It is therefore difficult to filter out outliers in the observed NO_2 based on aerosol data." In the data assimilation it is assumed that the OmF distribution is Gaussian and OmF can be used to filter outliers from the data. So far, no OmF outlier criterion has been used in DECSO. Our previous analysis, however, shows the need for the detection of outliers. A filter has to be implemented with care, to avoid that the algorithm becomes insensitive to new emission sources such as new power plants. Not losing sensitivity to new emission sources is also the reason we do not choose a relative filter criterion. We select an OmF filter criterion in the range of [-5, 10] 10^{15} molecules cm^{-2} based on our analysis discussed below..

The distribution of OmF of all pixels over our domain from January 2013 to September 2014 is Gaussian except for its tails and 97% of the OmF is in the interval of [-5, 10] 10^{15} molecules cm^{-2}. However, over highly polluted areas both satellite observations and model results have larger errors resulting in higher OmF values. In addition, the lifetime of NO_2 is much longer in winter than in summer. Therefore, the NO_2 column concentration is higher than in summer, which may lead to large OmF values in winter time. We choose 15 high polluted cities in China based on AQI and study the distribution of the OmF for the summer period (April to September, 2013) and the winter period (October, 2013 to March, 2014) (Figure ~~6~~7). As expected, the distribution of OmF is wider in winter than in summer. In summer 70% of the OmF values are in the interval of [-5, 10] 10^{15} molecules cm^{-2}, while in

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winter 50% of the OmF values are within [-5, 10] 10^{15} molecules cm^{-2}. We select an asymmetric interval ~~–~~because the assimilation is especially sensitive to very negative outliers in OmF caused by low observations (having small observational errors associated), as opposed to very positive outliers caused by high observations, which are associated with large observational errors. The observations with low error have more weight in the data assimilation process.

To figure out the effect of a large OmF on ~~NO_2~~ NO_x emission estimates, we compare a free run of CHIMERE v2013 with the MEIC inventory with a run with the DECSO v3b assimilation. During the summertime, the ~~mean~~ difference in the seasonal average of the NO_2 column concentration~~s~~ between these two runs is 4.8 10^{15} molecules/cm^2 in the Nanjing area (six grid cells). This column difference is caused by the NO_x emission difference of 9.2 10^{15} molecules cm^{-2} h^{-1}. From a simple back-of-the-envelope calculation we derive that a negative 5 10^{15} molecules cm^{-2} difference in NO_2 columns requires a 9.6 10^{15} molecules cm^{-2} h^{-1} emission change, which would mean that all NO_x emissions in Nanjing would be removed in a single day. This change in emission is comparable to the total emissions of 2 large-sized coal-fired power plants. ~~In addition, when the pixel size of the satellite is twice that of the model grid cell, the updates of emissions in that grid will even be doubled.~~ This shows that a change in OmF of 5 10^{15} molecules cm^{-2} is very unrealistic even in the most extreme cases. Therefore, this limit will be used as a criterion to filter outliers, which are in general caused by wrong NO_2 retrievals. To avoid the influence of the extreme OmF on emission estimates and still be able to monitor real emission changes, we filter out negative OmF values lower than 5 10^{15} molecules cm^{-2} and positive OmF values more than 10 10^{15} molecules cm^{-2} to be conservative. After applying the OmF filter criteria, we filter out 16% of the extreme OmF in the polluted cities and less than 3% in the whole domain. The large unrealistic jumps in emission disappear from the time series.

4 Emission analysis for the Nanjing Youth Olympic Games

First, we compare NO_2 monthly average concentrations in 2014 with previous years using in-situ and satellite observations. For the in-situ observations we select the monthly mean at 13:00 LT to be able to compare the results with the satellite observations whose overpass time is about 13:30 LT (see ~~Figure7~~Figure8), which is also the average overpass time in Nanjing. Compared to the year 2013 the in-situ measurements show no significant improvement in the surface NO_2 concentration at 13:00 LT for the period (May to August, 2014) when the government took air quality regulations for the YOG. However, we 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. We also calculate the monthly average using all measurements and we still see no improvements of the surface NO_2 concentration for the YOG period. Therefore, we ~~assume~~ conclude that the in-situ measurements are not representative for the whole city of Nanjing.

Figure ~~8~~ 1 shows the land-use over Jiangsu Province. The rectangle referred to as the Nanjing area, covers the whole of Nanjing including all industrial areas along Yangtze River. According to the MEIC sector distribution, the power plants in the selected area are dominating the NO_x emissions. To study the effects of the air quality regulations for the YOG on tropospheric NO_2 column concentrations, we compare the monthly averages of satellite observations over the Nanjing area for each year from 2005 to 2014 by regridding the observational data on the model grid over the area.

The satellite observations show that on average the NO_2 column concentrations are rather similar from year to year (Figure 9). Although a small increasing trend from 2005 to 2011 is visible in the satellite data, ~~we include this in the standard deviation~~it is negligible compared to the SD of the natural variability. It is clear that the NO_2 ~~concentrations~~ columns have a seasonal cycle that is lower in summer than in winter due to the seasonal change of the NO_2 lifetime (van der A et al., 2006). Note that the small decrease in ~~concentrations~~ columns in February might be caused by the reduced emissions during the Spring Festival (Zhang et al., 2009b). The monthly averages of NO_2 in-situ observations shown by Wang et al. (2011) for Beijing, Shanghai and Guangzhou in 2005 were also reduced by around 10% in February. We see that the NO_2 ~~concentration~~ column during the YOG period (August 2014) is on average only 6.6 10^{15} molecules cm^{-2}, which is the lowest value among the last 10 years and more than 3 standard deviations from the mean. ~~Consequently, D~~due to the effect of the continuous air quality regulations for the YOG and afterwards, the NO_2 ~~concentrations~~ columns of the following months are also lower than for previous years. In November, the local government took similar air quality regulations for the first National Memorial ceremony held on 13^{th} December, 2014. That might explain the lower NO_2 ~~concentrations~~ columns of the last two

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months of 2014 compared to those of 2013 and compared to the average of the last 8 years. However, it is still within the range of the standard deviation of NO_2 ~~concentrations~~ columns for the last 8 years. Differences from year to year can also be attributed to the meteorological conditions (Lin et al., 2011). Particularly in December 2013, NO_2 ~~concentrations~~ columns are very high. This episode is well known as a heavy smog period in Nanjing because stagnant air in the region accumulated anthropogenic pollution. Compared to the averaged NO_2 ~~concentration~~ column in August from 2005 to 2012, the NO_2 ~~concentration~~ column of August in 2014 is decreased with 32% in Nanjing. However, this significant decrease can be caused by the rainy weather during that month. Thus, ~~NO_2~~ NO_x emission estimates are needed to show if the air quality regulations were really effective. 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.

To compare the NO_x emissions in Nanjing in 2014, especially during the YOG, with the same period of the year 2013, we run DECSO v3b with the OmF criterion as described in Section 3.2 from October 2012 to December 2014, where the first three months are used as spin-up period. Figure 10 shows the monthly NO_x emissions in Nanjing for the year 2013 and 2014 estimated by this version of DECSO. For comparison the initial MEIC inventory is also plotted in the Figure. The ~~NO_2~~ NO_x emissions have a different seasonal cycle compared to the NO_2 ~~concentrations~~ columns of satellite observations in Nanjing. The months with high emissions are June and July while the highest NO_2 ~~concentrations~~ columns of the satellite observations appear in January and December. According to the sector distribution in the MEIC inventory, the emissions of power plants and industrial activities are the main sources in Nanjing. At least 50% of the total NO_x emissions are from power plants and 40% are from the industrial activities. Zhang et al. (2009) showed that the seasonal cycle of the electricity consumption in Nanjing for the ~~6~~ 7 years from 2000 to 2006 peaks in the summertime, ~~,~~ because the electricity consumption and power load are highly correlated with temperature in summer. The value of electricity consumption in summer is at least two times higher than in winter every year and keeps increasing during those 7 years. The seasonality of electricity consumption is caused by the increasing usage of air conditioning in the hot season, while there is no heating system used in winter time in Nanjing. The opposite cycles of column concentrations (Figure 9) and emissions (Figure 10) show that the high NO_2 concentrations in winter in Nanjing are mainly affected by the long lifetime of NO_x, while the seasonal cycle of

NO_x emissions is reversed as a result of the increased electricity consumption in summertime. The difference with the seasonal cycle of MEIC might be attributed to the fact that our results are derived on city-level, while the seasonal cycle for bottom-up inventories are often derived on a national or provincial scale (e.g. Zhang et al., 2009b)

We see a drop in NO_x emissions in February for both years calculated with DECSO, which is also visible in the MEIC inventory of 2010 (Figure 10). This jump is consistent with the decrease of NO_2 concentrations columns of the satellite observations in February compared to the neighboring months. Compared to the neighboring months, the NO_x emission reduction in February is about 10% in 2013 and 2014. This NO_x emission decrease was also noticed by Zhang et al. (2009b) in the INTEX-B inventory and likely to be caused by the reduced industrial activities during the Spring Festival.. Interestingly, we do not see an increase of NO_x emissions in the December 2013 smog period. This shows that the smog is caused by the meteorological conditions rather than increased emissions.

Figure 10 shows a large reduction of NO_x emissions in September, 2014. This reduction is probably caused by the more permanent air quality regulations taken by the local government to reduce air pollutants during the YOG period. The total NO_x emissions in September in Nanjing are 4.5 Gg N. Compared to the same time of the year 2013, the reduction is about 25%. However, the emission reduction in this case seems to have a delay of one month. 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 satellite observations due to the rainy (and therefore cloudy) weather in the second half of August 2014 when the YOG took place. For this these kind of conditions, DECSO only detects the full extent of the emission reduction in September. We also see a NO_x emission reduction of 10% in August, 2013, compared to the neighboring months. One likely reason for this reduction is that the Asian Youth Games were held during that time. The local government also took measures to ensure good air quality for that event but not as strict as for the YOG in 2014. 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.

5 Discussion and conclusions

In this study the effect of the air quality regulations of the local government during the YOG in Nanjing in 2014 has been quantified by analyzing observations on the ground and from the satellite. The focus in this study was on the reduced NO_2 concentrations and NO_x emissions. We compared NO_2 during the YOG period with previous years using the in-situ and the OMI satellite observations. The in-situ observations ~~are very noisy~~have a large variability, even after averaging on a monthly basis. This is probably caused by the variability of local sources and it indicates that these in-situ observations are not representative for the larger area of Nanjing. The in-situ data shows no significant decrease during the YOG period. Since we have no error estimates of the in-situ observations and very little information on the instrument and measurement techniques we discard the results of the in-situ observations in our conclusions.

For the view from space we limited ourselves to retrievals of tropospheric NO_2 from OMI, taking advantage of the high spatial resolution of OMI observations compared to similar instruments. The monthly OMI satellite observations showed a 32% decrease of the NO_2 column concentration during the YOG period in Nanjing compared to the average value for the last 10 years. However, the decrease of NO_2 ~~concentrations~~ columns observed by the satellite is not an objective measure to verify the impact of the air quality regulations taken by the local government, because changes in NO_2 ~~concentrations~~ columns 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. Furthermore, due to cloudy conditions, the August average of 2014 is based on few observations. Therefore, it is important to analyze the emissions to show if the air quality regulations ~~has~~ have really affected the NO_2 concentrations.

The results of our improved emission estimate algorithm DECSO show that NO_x emissions decreased with at least 25% in September 2014, which shows that the air quality regulations were effective during the YOG period and that only a small part of the reduced NO_2 column concentrations were caused by the weather conditions. However, the reduction has one month delay in our results. This is because satellite observations were scarce in the Nanjing area during the YOG (16 to 29 August) causing the DECSO algorithm to converge slower to the new emissions, which is typical for the Kalman filter approach used in DECSO.

We were able to see the emission reduction of NO_x in the selected 6 grid cells representative for the Nanjing area. That means that DECSO at least is able to estimate NO_x emissions on a spatial resolution of about 50 x 90 km^2. If we apply the same analysis on single grid cells the results are noisier because the footprint of the OMI covers on average a larger area than a single grid cell. To achieve emission estimates in a smaller area, either satellite observations with a higher spatial resolution are required, or longer time periods should be considered.

The quality of our emission estimates is highly related to the quality of the model and the satellite observations. We improved the DECSO algorithm by using a new version of the CTM: CHIMERE v2013 instead of CHIMERE v2006. The comparison of OmF between two models showed that CHIMERE v2013 has a better performance in summertime. Good quality of satellite observations is also essential for emission estimates. The DOMINO retrieval algorithm does not properly account for the effects of high aerosol concentrations, which are common in China, on the retrieved NO_2 columns. In case of high aerosol concentrations, the difference of the model simulations and the retrievals is very large, which leads to wrong updates of NO_x emission in DECSO. To improve the satellite observations we have set an OmF criterion to filter out erroneous observations and to avoid unrealistic NO_x emission updates. We set the limitation to the range -5 to 10 10^{15} molecules cm^{-2} for the OmF. With this filter criterion, the unrealistic updates of NO_x emissions are mostly prevented. We will further ~~analysis~~ analyze the impact of high aerosol concentrations on the retrieved NO_2 columns in future research.

Furthermore, we observed an opposite seasonal cycle of NO_x emissions compared to the NO_2 ~~concentrations~~ columns observed by OMI satellite. The seasonal cycle of NO_x emissions is not the same for the whole China domain since the different climate in the North and the South of China leads to a different variability of energy consumption during the year. In Nanjing, as in most parts of Southern China, people use air conditioning in summer and do not use heating systems in winter. This leads to larger electricity ~~consumptions~~ production of power plants in summer causing higher NO_x emissions. Tu et al. (2007) studied the air pollutants in Nanjing and also found high NO_2 ~~concentrations~~ columns in winter but concluded that the high NO_2 ~~concentrations~~ columns were caused by high NO_x emissions in winter, while our emission estimates show the opposite. Wang et al. (2007) analyzed the seasonality of NO_x emissions based on GOME satellite observations for the regions north and south of Yangtze River, defined as north and south China. Their results of south China showed the same seasonal cycle of NO_2 columns but a very weak seasonality of NO_x emissions and they also concluded that the NO_x lifetime mainly determines the NO_2 columns. Ran et al. (2009) explained high NO_x concentrations in are caused by slower chemical processes and shallow boundary layers contributing to accumulation of NO_x. The table of Wang et al. (2012) annual and summer NO_x emissions from coal-fired power plants in 2005-2007 for different provinces in China showed that the NO_x emissions in Jiangsu Province in summer is higher than mean seasonal emissions.

In conclusion, in the emission estimates we not only found a reversed seasonal cycle peaking in summertime, but also indications for reduced emissions during the Spring Festival, the Asian Youth Games in 2013 and the YOG 2014. Based on our emission estimates the air quality regulation during the YOG 2014 and afterwards reduced the NO_x emissions ~~with~~ by at least 25 percent. This, together with favorable meteorological conditions, was responsible for the decrease of 32% in NO_2 column concentrations observed from space. For the case of the YOG, our results can help the local government to identify the impact of their air quality regulations on reducing NO_x emissions.

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

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Table 2. Estimated r~~R~~edistribution of MEIC sectors over SNAP 97 ~~-~~sectors

MEIC sectors / **SNAP 97 sectors**	Power	Industry	Transport	Residential	Agriculture
Combustion in energy and transformation industries	1	-	-	-	-
Non-industrial combustion plants	-	-	-	1	-
Combustion in manufacturing industry	-	0.3	-	-	-
Production process	-	0.3	-	-	-
Extraction and distribution of fossil fuels and geothermal energy	-	0.4	-	-	-
Solvent and other product use	-	-	-	-	-
Road transport	-	-	1	-	-
Other mobile sources and machinery	-	-	-	-	-
Waste treatment and disposal	-	-	-	-	-
Agriculture	-	-	-	-	1
Other source and sinks	-	-	-	-	-

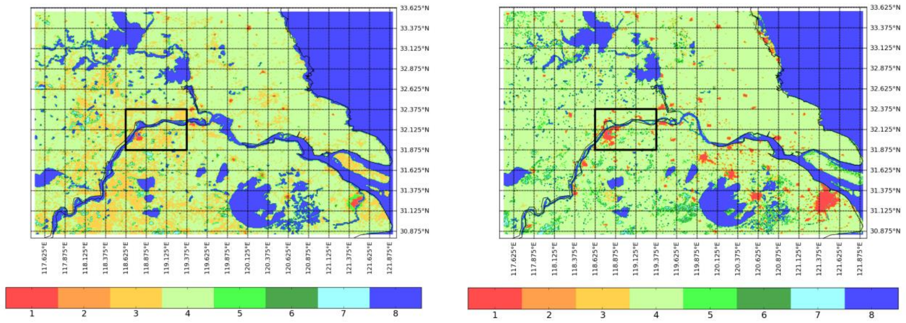


Figure 1. Land use over the Jiangsu Province from Global Land Cover Facility (1994) (left) and the GlobCover Land Cover (2009) (right) and as used in CHIMERE v2006 and CHIMERE v2013. The 8 categories are: 1. Urban, 2. Barren land, 3. Grassland, 4. Agricultural land, 5. Shrubs, 6. Needleleaf forest, 7. Broadleaf forest, 8. Water. The solid rectangle (about 50 x 90 km^2) indicates the 6 grid cells that cover the Nanjing area.

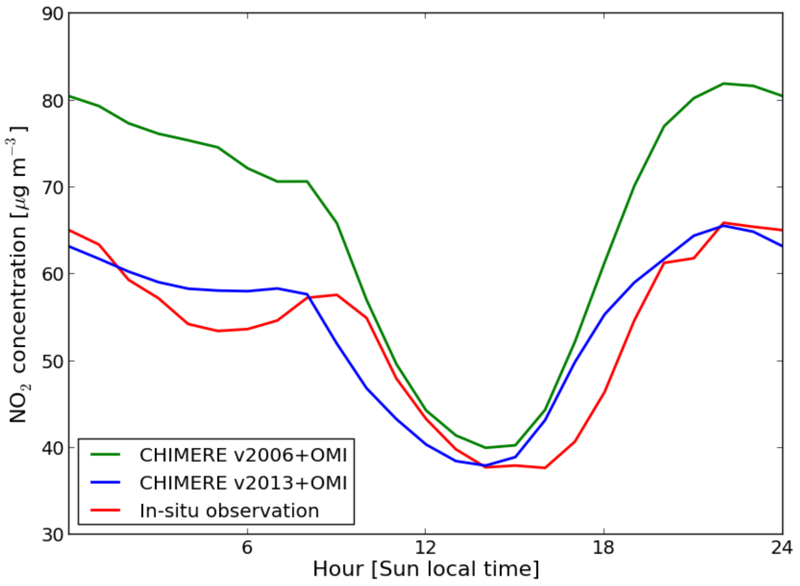


Figure ~~1~~ 2. The diurnal cycle in Nanjing from January to August 2014 according in-situ observations, OMI-assimilated CHIMERE v2013 and CHIMERE v2006.

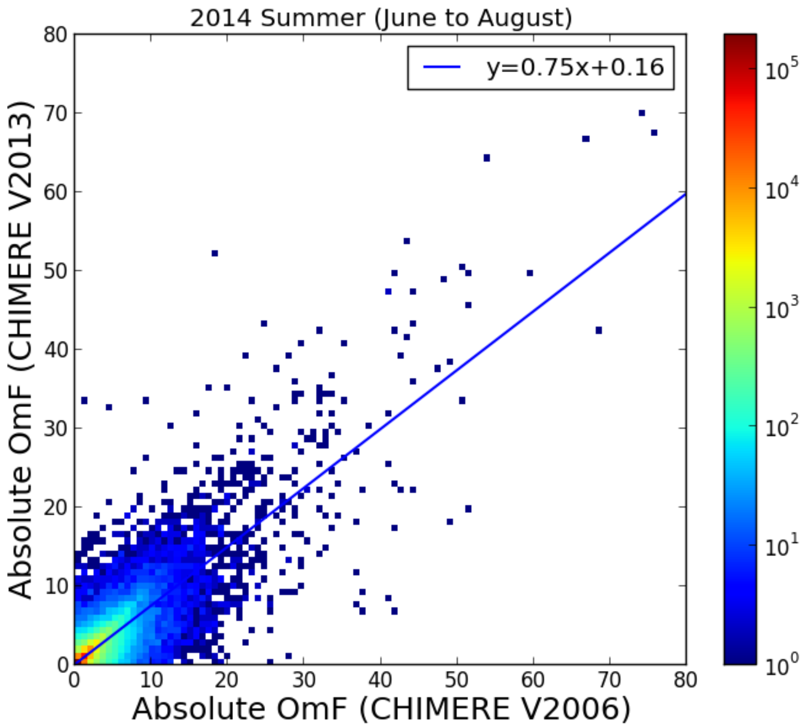


Figure ~~2~~3. The comparison of the absolute OmF (10^{15} molecules/cm^2) of CHIMERE v2006 and CHIMERE v2013 for the whole East Asian domain from June to August . The colorbar represents the frequency of satellite observations for that specific value of OmF.

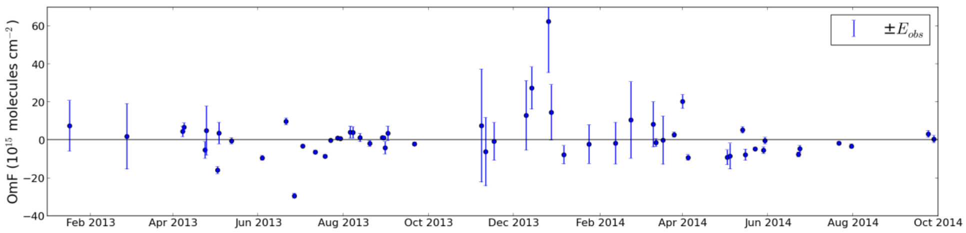


Figure 34. The time series of the OmF from January 2013 to September 2014 for the single grid cell over the center of Nanjing. The error bar is the root mean square error of observations ($\sigma_{obs}E_{obs}$).

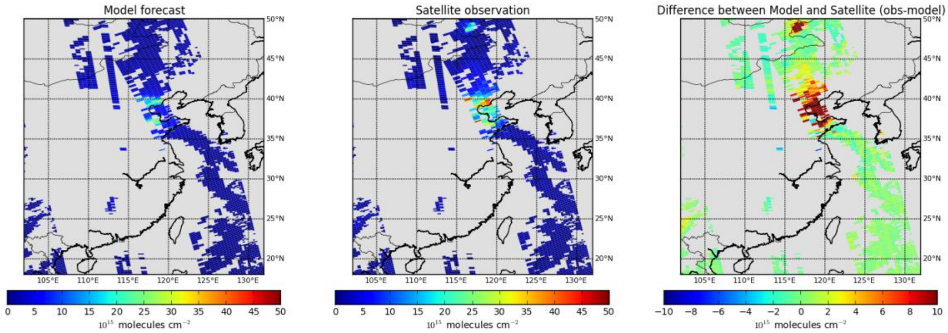


Figure ~~4~~5. The comparison of the CHIMERE v2013 forecast (left) with OMI satellite observations (middle) on 6 May 2013. The right plot shows the difference between observations and forecast (OmF).

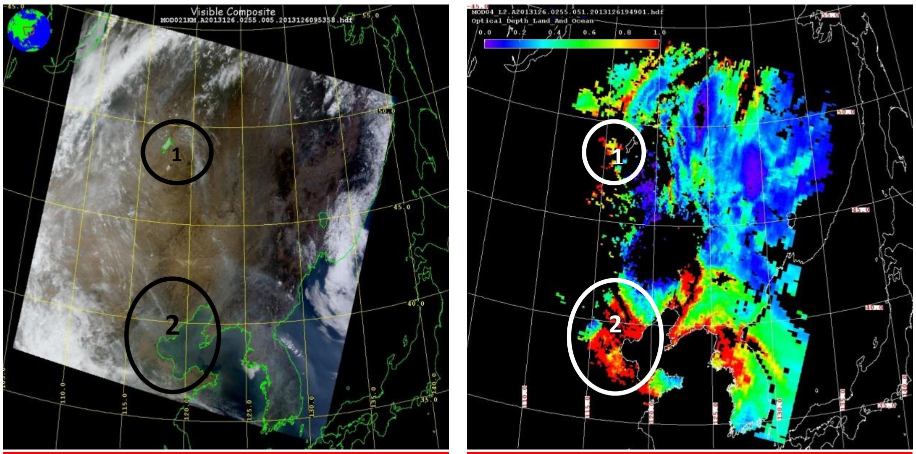
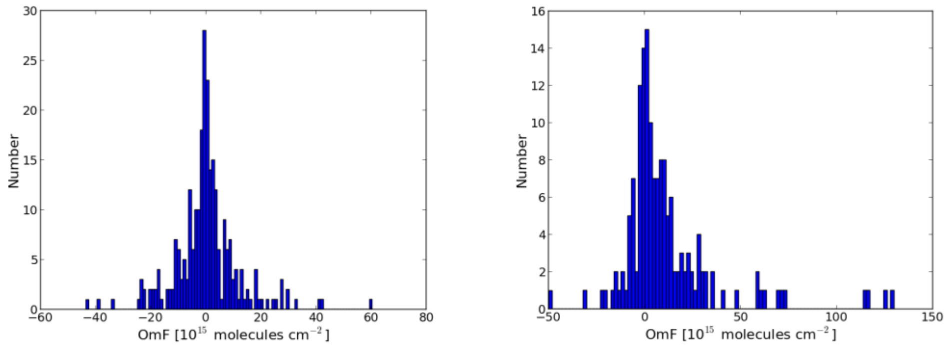


Figure ~~5~~6. The RGB image (left) and Aerosol Optical Depth (right) from MODIS on 6 May 2013. Circle 1 and circle 2 represent the Hulunbuir sand land and the Bohai Bay respectively. (The figures are from https://ladsweb.nascom.nasa.gov/browse_images/granule_browser.html)



a. April to September 2013 b. October 2013 to March 2014

Figure 7~~6~~. The distribution of the OmF values over 15 polluted cities in summer (a) and in winter (b). The 15 polluted cities are Baoding, Beijing, Chengdu, Harbin, Hohhot, Guangzhou, Jinan, Shanghai, Shenyang, Shijiazhuang, Tianjin, Wuhan, Xi'an, Xingtai and Zhengzhou.

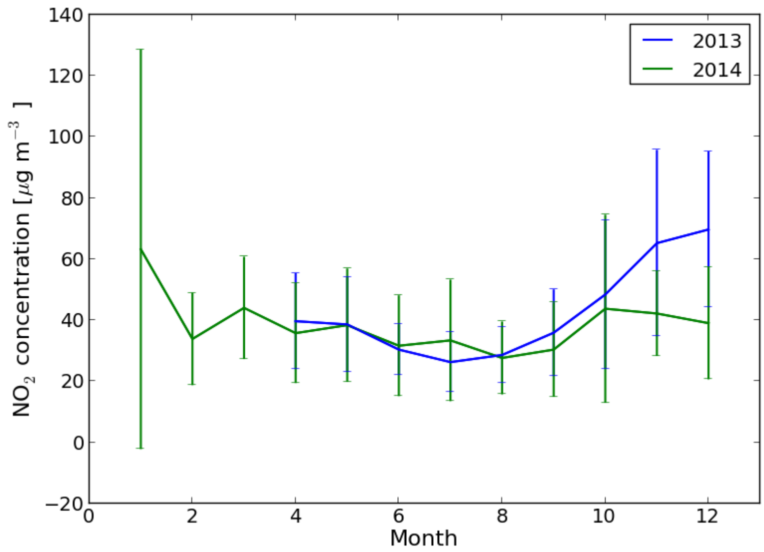


Figure ~~7~~8. The monthly averaged in-situ NO_2 concentration at 13 local time in Nanjing for 2013 and 2014. The ~~error~~ bar is the standard deviation (natural variability) of the observations for each month (derived from the daily data on www.aqicn.org).

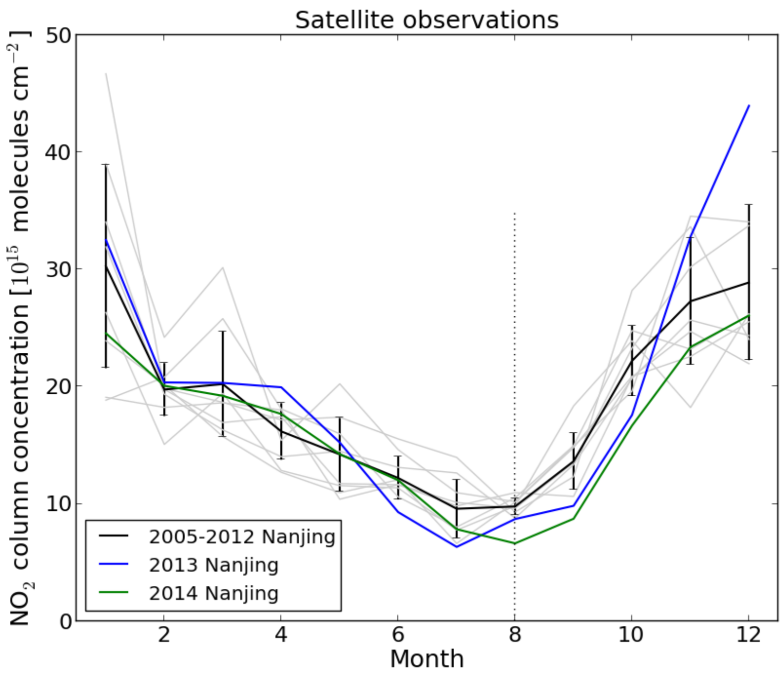


Figure 9. The monthly averages of OMI satellite observations of tropospheric NO_2 concentrations. The solid lines are the measurements over the Nanjing area. The grey lines are the monthly averages for each year from 2005 to 2012 to indicate the annual variability. The black lines show the average value for the years from 2005 to 2012. The ~~error~~bars are the standard deviations of monthly NO_2 observations from 2005 to 2012.

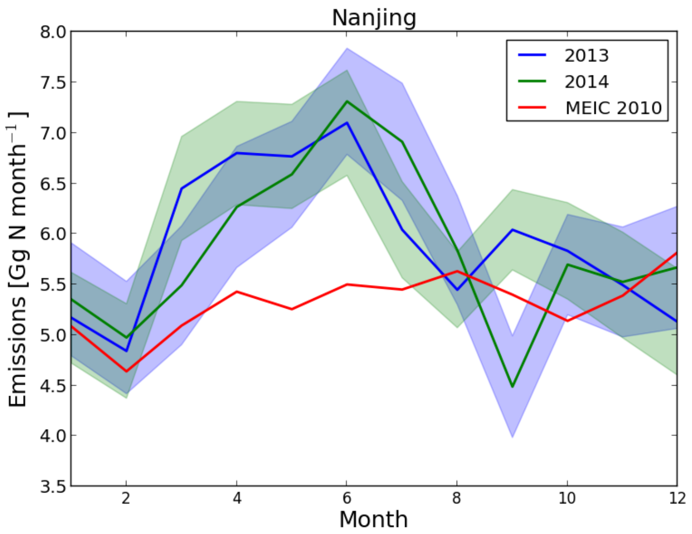


Figure 10. The monthly NO_x emission estimates by DECSO in Nanjing for 2013 (blue line) and 2014 (green line) and the monthly NO_x emission of the MEIC inventory of 2010 (red line). The shade areas show the error of the mean NO_x emission estimates from DECSO.