We thank all referees for giving valuable comments. We respond to each specific comment below. The comments and questions from referees are in italic font.

Anonymous Referee #1

I found the resubmitted paper much easier to read, and several of my comments were addressed in sufficient detail. The intro, method description, etc are quite good now. I still feel like the conclusions are not terribly definitive and there are questions raised by the results that are hard to answer, but the authors seem to do a pretty thorough job in the analysis and making use of available data.

There is obviously a significant decline in emissions in September 2014, and this is attributed to remaining controls leftover from the Games. I don't understand why the emissions would go back up in October 2014. Is there an explanation for this?

Actually the main reason for detecting the emission decrease apparently in September is a time lag typical for Kalman Filters in combination with: the fact that the Olympic games took place at the very last two weeks of August. The satellite measurements that measure directly the NO_2 columns without time lag show the reduction in August.

The more permanent measures (traffic-related) resulting from the YOG are indeed not seen in October, which means that they are likely to be a small fraction of the total emissions. According to the statistical information used for bottom-up inventories, traffic is less than 10 % of the total emissions.

We removed the sentence in page 16 line 6:

“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.”

We clarified this in the text

In page 14 line 22, we add:

“ […] previous years. **The more permanent measures (traffic-related) resulting from the YOG affect a small fraction of the total emissions.** In November […]S ”

We add these explanations on Page 16 line18 :

“[...] the emission reduction in this case seems to have a delay of one month. **The shaded area in Figure 10 represents the error on the derived emissions without taking into account the error introduced by the Kalman Filter time lag. 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 our case, this is partly a consequence of […]”

Specific comments:

In many cases, the authors use the expressions “with XX%”. Usually the correct English phrasing is “by XX%”.

Thank you for pointing out this problem. We change this grammar mistake in the paper.

Page 3, line 32: Same comment as my previous review: Change “concentration” to “column”.

We change “concentration” to “column” here.

We seem to have overlooked this comment at the previous review, but we have changed it this time.

Page 5, line 25: I still think this partitioning of industry sector needs justification, other than gut feeling

In general we selected the SNAP sector which was closest in description to the MEIC sector. Only in the case of the industry sector of MEIC we found that 3 important SNAP sectors are part of the industrial sector. Because of the lack of information about the industrial sectors in China, we more or less evenly distributed the industry sectors over the 3 SNAP sectors.

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Page 8, line 19: How is this concluded if you had no observations before? How do you get a 37% increase over zero observations? Also, on page 16, line 12 you blame lack of observations. It is still not clear to me how many observations you have. One, ten, hundreds? This is significant for your conclusions.

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.

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

For clarification, we change the sentence into :

“The number of observations over the whole domain increases by about 37 % on average.”

Page 9, line 5: You don't have to include this in the final paper, but curious if you tried emailing sources in China? Surely someone must know where this monitor is located!

We send emails to the website where we get the data. But we didn’t get any answers.

Page 10, line 23: Usually "noise" is used to discuss instrument measurement noise. I think you mean transient influences here.

We change the word "noise from" to "transient influences on" here.

Page 14, line 1: Again, this statement discards data because it does not meet a hypothesis which hasn't even been found to be correct yet. Reword.

Yes. We agree. We rephrased the text:

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

into:

"We also calculate the monthly average using all measurements and we still see **a high variability in the time series. Because of the high variability in the ground data and its low representativity for the whole city of Nanjing, we discarded this data set in our analysis**."

Page 15, line 18: If the NOx emissions are so dependent on air conditioning, might be helpful to include a sentence on the temperatures during summer 2013/2014 to convince reader this can be disentangled from the patterns you see. Was September 2014 particularly cold for example?

September 2014 was not cold and the temperature was not deviating much from the climatological values. We add a sentence on Page 15 line 28:

"[…]provincial scale (e.g. Zhang et al., 2009b). **The monthly average temperatures from May to September are above 20 degree. The monthly temperatures in 2014 were not deviating much from the climatological values.**"

Page 17, line 18: I'm not very familiar with Kalman filter data assimilation. Why don't you get larger error bars if there are few observations to assimilate in August 2014?

Indeed the error bars are larger in the YOG period. However, we show the error of the monthly mean emission data, which is lower because of many observations earlier in August.

Anonymous Referee #2

Thank you to the authors for carefully addressing all reviewer comments. However, I have to insist that they weaken their conclusions regarding emission reductions being caused by local legislation.

Specifically,

- p.1/l.25 should say something like "probably indicates that ..."

We prefer not to add the word here. We think this word weakens our conclusion too much. The main purpose of this study is to quantify the emission reduction caused by the air quality regulations during the Youth Olympic Games similar to the previous major international events in China. Most former studies were based on the NO_2 satellite or ground observations without considering the weather effects and concluded that emission reduction was caused by air pollution control measures. Different from these studies, our method not only take NO_2 columns into account but also the meteorological conditions and transport. We explained that the time lag in our result is inherent to the use of Kalman filter. This is at least enough to draw the conclusion that the emission reduction detected in this study is due to air quality measures.

- p.14/l.20 has to be weakened; the current statement "Due to the effect of the continuous air quality regulations" is not backed by the authors' analysis. Maybe, a "probably" could be inserted at the beginning of this sentence.

We agree. We insert a “possibly” at the beginning of this sentence.

- p.16/l.19 again states in certainty a causal relation between policy measures and NOx emission reductions. This has to be weakened, perhaps by adding "possibly": ", possibly showing that the air quality measures ..."

We prefer not to add the word “possibly” here. We think it is a very plausible conclusion that such strict air quality measures results in NO_x reductions. Starting from this a priori information, our paper discusses the quantification of the emission reduction.

Furthermore, the following issues have to be addressed before accepting this manuscript for publication in ACP:

- p.8/l.31 The authors should copy the exact formula/table they use to convert AQI to NOx from their reference so that this is available to non-Chinese community. This could be done in a supplement.

We have added the following conversion table and formula to the supplementary material:

The Air Quality Index (AQI) with its corresponding NO_2 concentration breakpoint.

Air Quality Index (I)	NO_2 hourly mean (ug m^{-3}) (C)
0	0
50	100
100	200
150	700
200	1200
300	2340
400	3090
500	3840

The formula to calculate the NO_2 concentration from the AQI is according the piecewise linear function:

$$C = \frac{I - I_{low}}{I_{high} - I_{low}} \left(C_{high} - C_{low}\right) + C_{low}$$

C is the NO_2 concentration; *I* is the AQI; C_{low} is the concentration breakpoint is lower than *C*; C_{high} is the concentration breakpoint is higher than *C*; *I* is the index breakpoint corresponding to C_{low}; I_{high} is the index breakpoint corresponding to C_{high}.

- p.10/l.22 The authors insist that using a 8month average of in-situ measurements allows them to judge the quality of the CTM simulations, in spite of studies they cite themselves. However, while using a long temporal average might average out any random fluctuations, but the spatial misrepresentation of a single point in-situ measurement compared to the large model grid cell cannot be corrected for using temporal averaging.

We argue in the manuscript that 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. Knowing that there still is a spatial misrepresentation we only draw conclusions about the diurnal cycle of the models compared to the ground-based observations.

- The authors should give the details for the injection height for each of the emission sectors, possibly in a supplement (see p.9/l.11 of the authors' response to the reviewer comments).

The injection height for each of the emission sectors described here is used for DECSO v3a (CHIMERE v2006). However, in DECSO v3b, which is used for the analysis, those injection heights are no longer used because they are defined within the model CHIMERE v.2013 itself. Admittedly, we forgot to mention this in the paper and we added a sentence on page 9 line 21:

"[…] In addition, the chemical reaction rates are updated and a new transport scheme is used in the new CHIMERE model. **The new CHIMERE model includes the emission injection height profile for different emission sectors. […]"**

- Given the authors' response to reviewer comments, p. 9/l. 29ff, doesn't this mean that DECSO favors emission increases over emission reductions? Maybe this could be added to p.7/l.2 of the revised manuscript.

No, DECSO favors emissions related to observations with low observational, representation and propagated emission errors. This varies from observation to observation, thus no general conclusions can be made for this.

- In p.21/l.3 of the authors' response, they write of "a large range of media, newspaper and internet sources". It would be good if all these sources could be properly cited, perhaps in a supplementary document

We are not able to give a complete list of references since this is a kind of common knowledge for the Chinese community, who follow the news from various sources (TV, radio, newspapers, blogs, etc.). We have not archived these sources. The language of these sources is often Chinese the therefore we have given a summary in Table 1. For both these reasons we did not think it useful to give specific references.

However, for your information we give a few links as example:

http://news.163.com/14/0801/13/A2IL9GP900014SEH.html

http://sports.people.com.cn/n/2014/0808/c383364-25429256.html

http://www.chinadaily.com.cn/china/2014-07/09/content_17680294.htm

- In p.23/l.16 of the authors' response, the authors say that their Kalman filter technique used by DECSO does not consider a time lag in the emission errors. This has to be explicitly mentioned in the manuscript in the discussion of Figure 10.
We add these explanations on Page 16 line18 :

"[...] the emission reduction in this case seems to have a delay of one month. **The shaded area represents the error on the derived emissions without taking into account the error**

introduced by the Kalman Filter time lag. 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 our case, this is partly a consequence of […]"

We also add one sentences on page 17 line 20:

"[…], which is typical for the Kalman filter approach used in DECSO. **Although the strong point of a Kalman Filter is its detailed error analysis, this time lag is not incorporated in its error formalism. In future research we intend to reduce this time lag by using a Smoothing Kalman Filter technique.**"

- In several places in the authors' response to the reviewer comments, they explain that the NOx emission reductions come from continuous AQ regulation which remains in effect also after the end of the YOG. However, in Table 1, only traffic-related emission control measures are marked as "permanent". Does this mean that the observed reduction comes mostly from the transportation/traffic sector? The authors should discuss this in the manuscript.

Actually the main reason for detecting the emission decrease apparently in September is a time lag typical for Kalman Filters in combination with: the fact that the Olympic games took place at the very last two weeks of August. The satellite measurements that measure directly the NO_2 columns without time lag show the reduction in August.

The more permanent measures (traffic-related) resulting from the YOG are indeed not seen in October, which means that they are likely to be a small fraction of the total emissions. According to the statistical information used for bottom-up inventories, traffic is less than 10 % of the total emissions.

We clarified this in the text

On page 15 line 15, we add:

" […] the industrial activities. **The more permanent measures (traffic-related) resulting from the YOG affect a small fraction of the total emissions.** Zhang et al. (2009) showed [..] "

Anonymous Referee #3

As raised by other reviewers as well, the 1-month lag in derived emission reduction is a main concern of the paper. The authors give fairly reasonable explanations in the response, including but not limited to the intrinsic problem of the Kalman filter. These expalanations should be included in the manuscript, for clarification.

We add these explanations on Page 16 line18 :

"[...] the emission reduction in this case seems to have a delay of one month. **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 our case, this is partly a consequence of […]"

We also add one sentences on page 17 line 20:

"[…], which is typical for the Kalman filter approach used in DECSO. **Although the strong point of a Kalman Filter is its detailed error analysis, this time lag is not incorporated in its formalism. In future research we intend to reduce this time lag by using a Smoothing Kalman Filter technique.**"

The Spring Festival is an important aspect of the current manuscript. It is good to compare with previous works (e.g., Lin and McElroy, 2011) that have also based on satellite measuremnts to show strong effects of Spring Festival on NO2 pollution and emissions.

Ref: Lin, J.-T. , and McElroy, M. B.: Detection from space of a reduction in anthropogenic emissions of nitrogen oxides during the Chinese economic downturn, Atmospheric Chemistry and Physics, 11, 8171-8188, doi:10.5194/acp-11-8171-2011, 2011

Thanks for providing the reference to us. We include the comparison between our result with this reference. We add the following sentences on Page 16 line 3:

"**Lin and McElroy (2011) also showed that the Spring Festival causes a reduction of about 10% on NO_x due to the decrease of thermal power generation based on the analysis of several satellite observations.**"

NO_x emission estimates during the 2014 Youth Olympic Games in Nanjing

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Abstract

The Nanjing Government 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 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 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~~–~~ 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 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 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 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 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 columns over East China ~~has been~~was 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~~ by 70% between 1995 and 2006 and Lamsal et al. (2011) found that anthropogenic NO_x emissions increased 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.

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 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 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~~ short-lived species, Martin et al. (2003) used the ratio of the simulated to the observed ~~concentration~~ column 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 of the emissions over the sectors (see Table 2).

As mentioned by ~~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. 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. 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. 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.

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

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

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. 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~~ by less than 20% and without introducing biases. Yet this effect is compensated by the advantage that more data becomes available. The number of observations over the whole domain increases ~~with~~ by about 37 ~~%.~~% ~~over the whole domain~~on average.

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 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 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 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 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. The new CHIMERE model includes the emission injection height profile for different emission sectors. 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 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°x0.25° 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 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~~ transient influences on the in-situ measurements, we 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 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

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 drop to zero in one day and then slowly increase again to the previous emission levels in the following days. 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, which are calculated as the quadratic sum of model and observation errors (Figure 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 these 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 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).

Figure 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 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 high aerosol concentrations are 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 the 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 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 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_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 difference in the seasonal average of the NO_2 column concentration 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. 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 Figure 8), 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 conclude that the in-situ measurements are not representative for the whole city of Nanjing.~~ We also calculate the monthly average using all measurements and we still see a high variability in the time series. Because of the high variability in the ground data and its low representativity for the whole city of Nanjing, we discarded this data set in our analysis."

Figure 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, it is negligible compared to the SD of the natural variability. It is clear that the NO_2 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 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 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. Possibly d~~D~~ue to the effect of the continuous air quality regulations for the YOG and afterwards, the NO_2 columns of the following months are also lower than for previous years. The more permanent measures (traffic-related) resulting from the YOG affect a small fraction of the total emissions. In November, the local government took similar air quality regulations for the first National Memorial ceremony held on 13th December, 2014. That might explain the lower NO_2 columns of the last two 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 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 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 column in August from 2005 to 2012, the NO_2 column of August in 2014 is decreased ~~with~~ by 32% in Nanjing. However, this significant decrease can be caused by the rainy weather during that month. Thus, 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_x emissions have a different seasonal cycle compared to the NO_2 columns of satellite observations in Nanjing. The months with high emissions are June and July while the highest NO_2 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 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). The monthly average temperatures from June to

September are above 20 degree. The month temperatures in 2014 were not deviating much from the climatological values.

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 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. Lin and McElroy (2011) also showed that the Spring Festival causes a reduction of about 10% on NO_x due to the decrease of thermal power generation based on the analysis of several satellite observations. 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. The shaded area in Figure 10 represents the error on the derived emissions without taking into account the error introduced by the Kalman Filter time lag. 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 our case, ~~T~~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 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 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 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 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 have really affected the NO_2 concentrations.

The results of our improved emission estimate algorithm DECSO show that NO_x emissions decreased ~~with~~ by 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. Although the strong point of Kalman Filter is its detailed error analysis, this time lag is not incorporated in its error formalism. In future research we intend to reduce this time lag by using a Smoothing Kalman Filter technique.

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 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 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 seasonality 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 a larger electricity production of power plants in summer causing resulting in higher NO_x emissions. Tu et al. (2007) studied the air pollutants in

Nanjing and also found high NO_2 columns in winter but concluded that the high NO_2 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 winter are caused by slower chemical processes and shallow boundary layers contributing to accumulation of NO_x. The table ~~of~~ in Wang et al. (2012) of 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 in the emission estimates, 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 by at least 25 percent. This, together with favorable meteorological conditions, was responsible for ~~the~~ a 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 still 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.

Table 2. Estimated redistribution 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	-	-	-	-	-

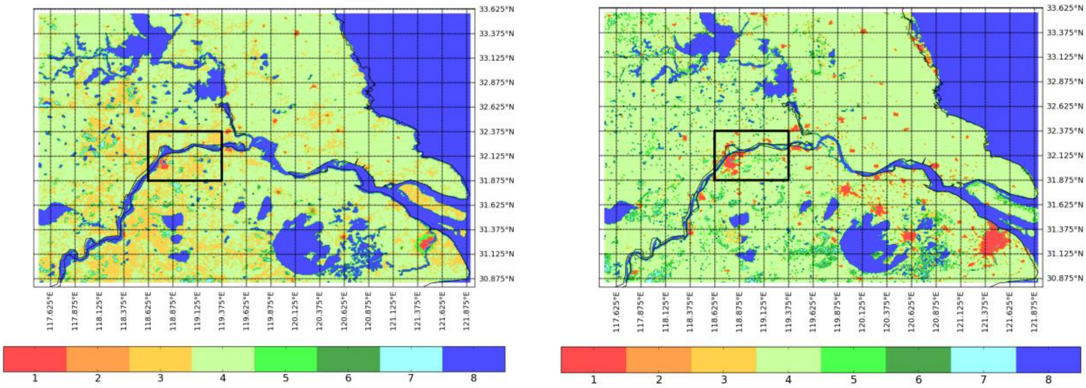


Figure 1. Land use over the Jiangsu Province from Global Land Cover Facility (1994) (left) and the GlobCover Land Cover (2009) (right) and as used in CHIMERE v2006 and CHIMERE v2013. The 8 categories are: 1. Urban, 2. Barren land, 3. Grassland, 4. Agricultural land, 5. Shrubs, 6. Needleleaf forest, 7.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

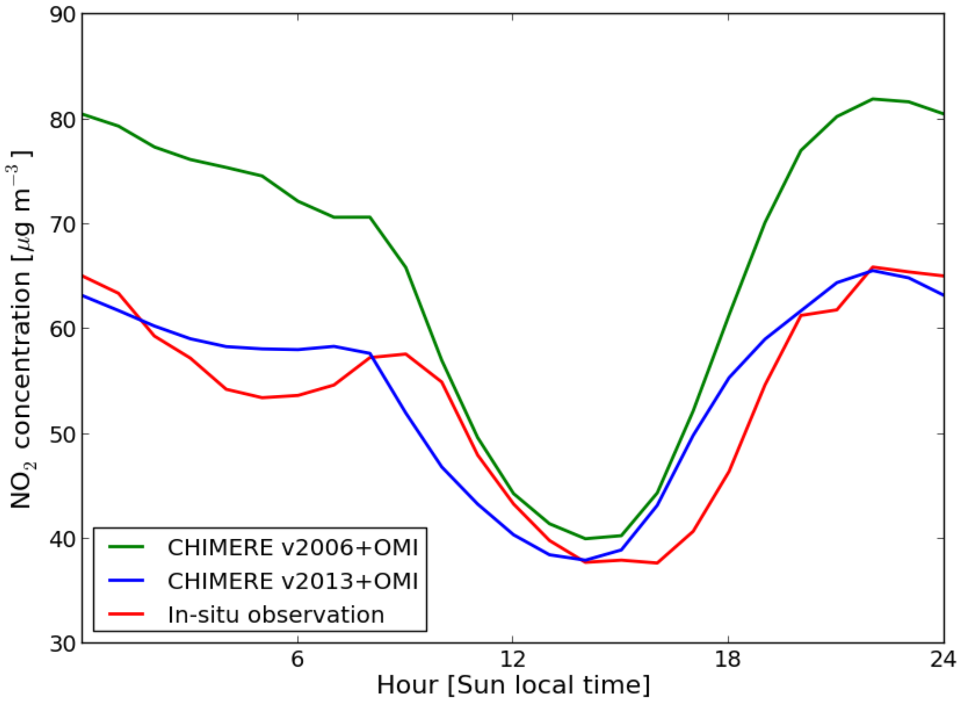


Figure 2. The diurnal cycle in Nanjing from January to August 2014 according in-situ observations, OMI-assimilated CHIMERE v2013 and CHIMERE v2006.

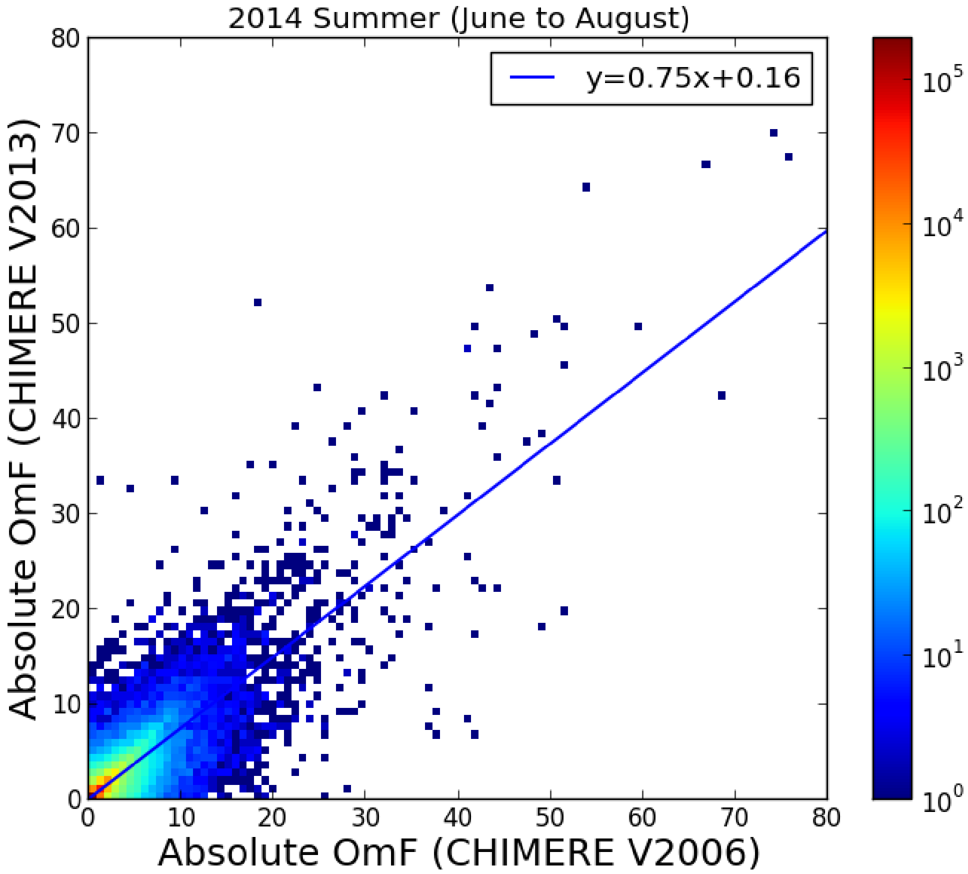


Figure 3. The comparison of the absolute OmF (10^{15} molecules/cm^2) of CHIMERE v2006 and CHIMERE v2013 for the whole East Asian domain from June to August . The colorbar represents the frequency of satellite observations for that specific value of OmF.

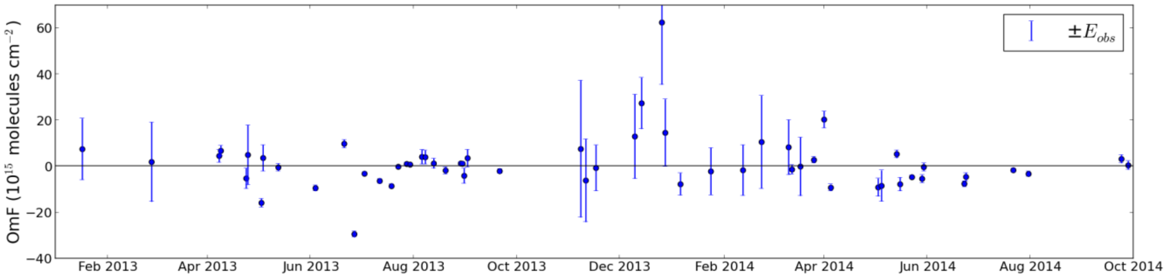


Figure 4. The time series of the OmF from January 2013 to September 2014 for the single grid cell over the center of Nanjing. The error bar is the root mean square error of observations (E_{obs}).

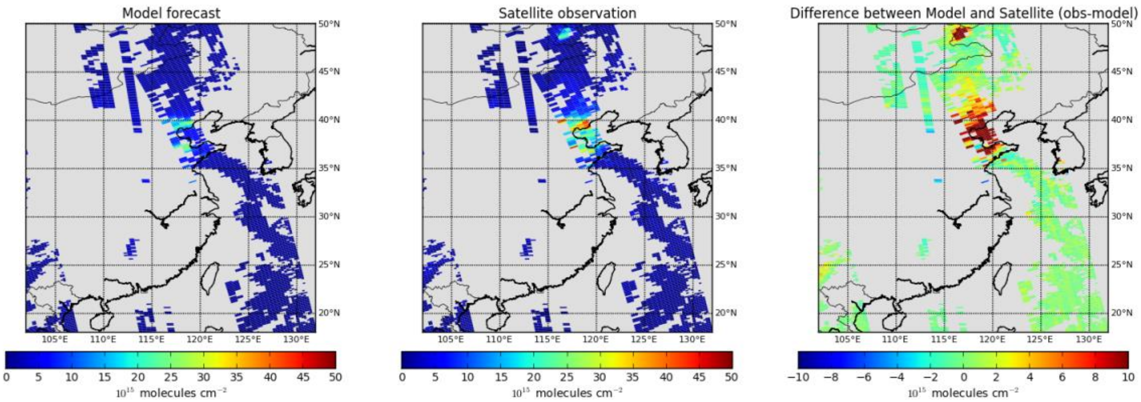


Figure 5. The comparison of the CHIMERE v2013 forecast (left) with OMI satellite observations (middle) on 6 May 2013. The right plot shows the difference between observations and forecast (OmF).

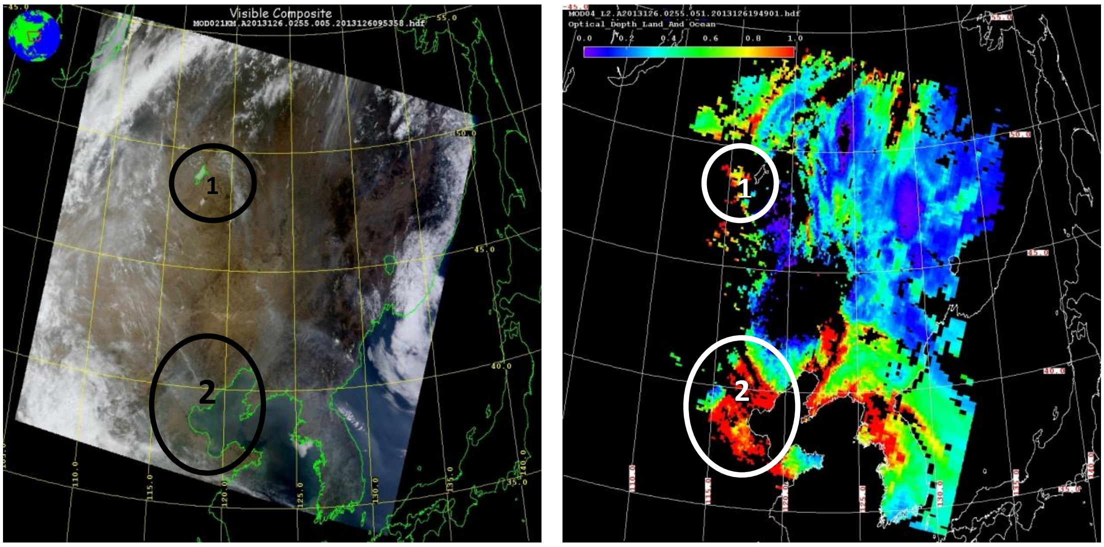
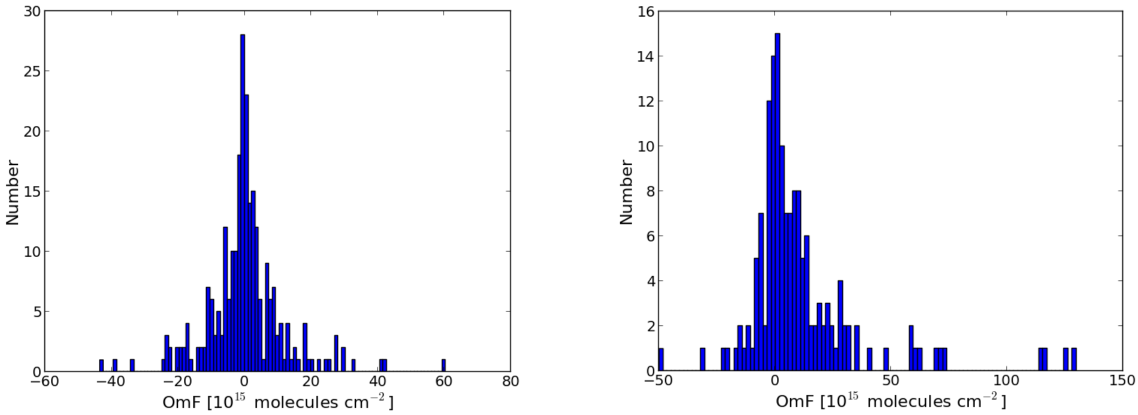


Figure 6. The RGB image (left) and Aerosol Optical Depth (right) from MODIS on 6 May 2013. Circle 1 and circle 2 represent the Hulunbuir sand land and the Bohai Bay respectively. (The figures are from https://ladsweb.nascom.nasa.gov/browse_images/granule_browser.html)



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

Figure 7. 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.

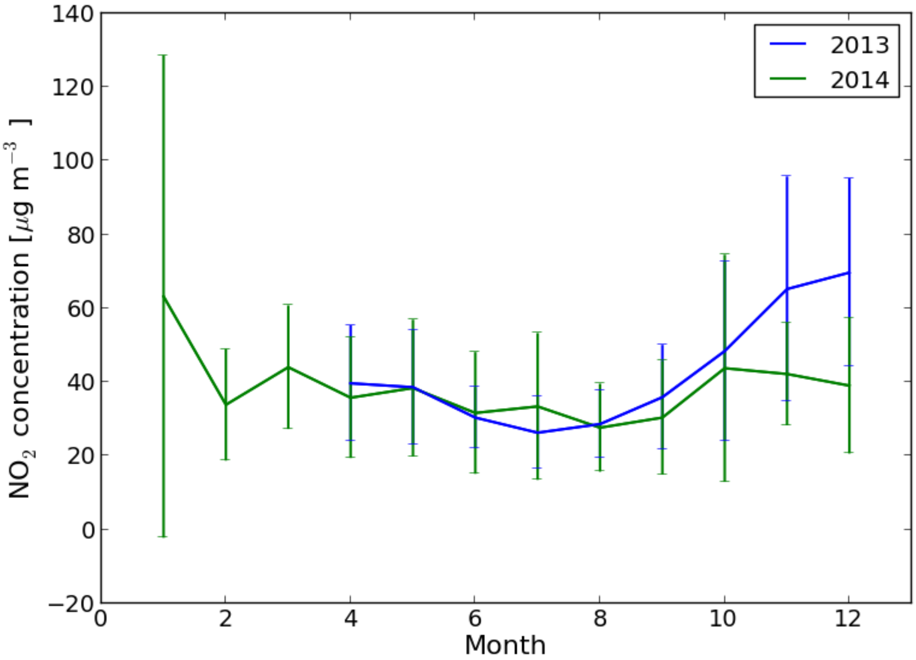


Figure 8. The monthly averaged in-situ NO_2 concentration at 13 local time in Nanjing for 2013 and 2014. The bar is the standard deviation (natural variability) of the observations for each month (derived from the daily data on www.aqicn.org).

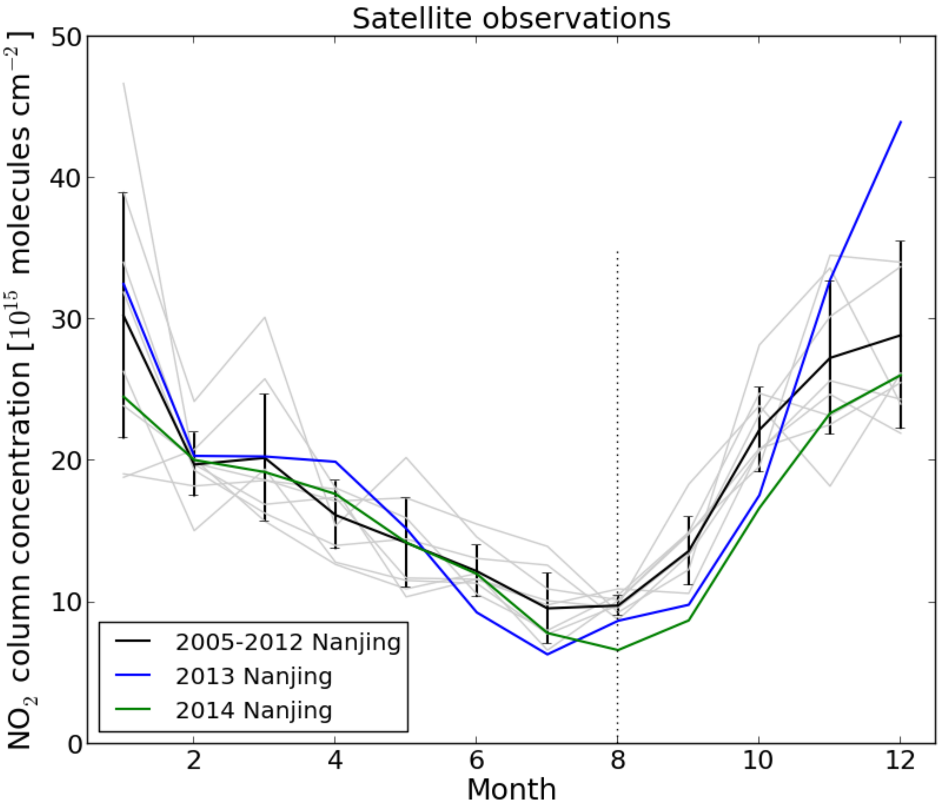


Figure 9. The monthly averages of OMI satellite observations of tropospheric NO_2 concentrations. The solid lines are the measurements over the Nanjing area. The grey lines are the monthly averages for each year from 2005 to 2012 to indicate the annual variability. The black lines show the average value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

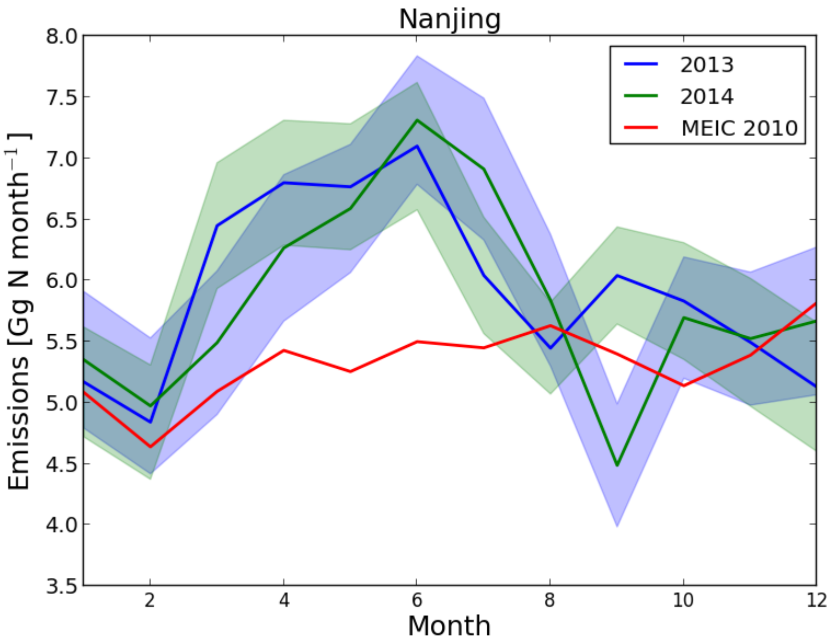


Figure 10. The monthly NO_x emission estimates by DECSO in Nanjing for 2013 (blue line) and 2014 (green line) and the monthly NO_x emission of the MEIC inventory of 2010 (red line). The shade areas show the error of the mean NO_x emission estimates from DECSO.