

General response: We thank all reviewers for their useful suggestions and comments, which have been incorporated into the revised manuscript.

We added a detailed discussion in the introduction clarifying the reasoning for our choice of January 2005-2010 for purposes of quantifying emissions of NO<sub>x</sub> and impacts of the economic downturn and the Chinese New Year.

We re-organized Sect 5.2.2 to clarify the procedure deriving emissions of NO<sub>x</sub> in January for all years. We detailed the derivation of the methodology to calculate emissions of NO<sub>x</sub> employing satellite retrievals and model simulations. We strengthened the analysis of retrieved VCDs of NO<sub>2</sub> and subsequently derived emissions of NO<sub>x</sub> for all retrieval products. We added a detailed analysis of changes in meteorological conditions and impacts on VCDs of NO<sub>2</sub>. We strengthened the analysis of the impact of retrieval data availability on the calculation of monthly mean VCD of NO<sub>2</sub>, followed by an analysis for the importance of applying the AK to model results.

We moved the description of GEOS-Chem to a newly added section (Sect. 4), where we strengthened the analysis of model errors. We moved to Appendix A the analysis of model-retrieval differences in the magnitudes of VCDs for individual months.

We added Appendix B, putting together and strengthening the analysis of the possible effect of changing emissions of NO<sub>x</sub>, CO and NMVOC on the lifetime of NO<sub>x</sub> and its partitioning into NO and NO<sub>2</sub> under specified meteorological conditions.

Other revisions have been made also to accommodate individual comments from all reviewers.

At this point, we believe our manuscript is much improved and is ready for final publication in ACP.

## Reviewer 1

In this paper, the authors report on satellite observations of NO<sub>2</sub> over China showing the overall increasing trend reported earlier but interestingly also a significant reduction during the recent economic downturn. They use the GEOS-chem model to quantify the impact of meteorology and chemical changes on the NO<sub>2</sub> evolution and to derive NO<sub>x</sub> emissions from the satellite data. The results of both the NO<sub>2</sub> column trends and the derived NO<sub>x</sub> emissions are compared to statistics on power generation and good consistency between reduced power generation and derived NO<sub>x</sub> emissions is reported. Part of the reduction in power generation and NO<sub>x</sub> emissions is also linked to the Chinese New Year holidays, and by applying different methods to estimate this effect, the authors conclude that the economic downturn is responsible for an emission reduction of about 10% while the CNY accounts for the second half of the observed reductions.

The paper is well organised and clearly written. The topic is relevant for ACP and the results (detection and attribution of a short-term reduction in NO<sub>x</sub> emissions in China from space based observations) are very interesting. However, there are several aspects of this study which I find problematic, and the authors need to clarify these points before the paper can be accepted for publication in ACP.

Response: We thank the reviewer for the useful suggestions, which have been incorporated the suggestions into the revised manuscript.

### Major Points

- The main part of the paper is based on an analysis of January values. This is motivated by the fact that non-anthropogenic NO<sub>x</sub> sources are small in winter. However, in my opinion this choice is quite unfortunate for several reasons: 1) winter observations have the poorest observation geometry and largest satellite uncertainties 2) there are retrieval problems with snow which is currently not treated properly in the analysis, 3) NO<sub>2</sub> life time is longest in winter and transport as well as the effect of diurnal variations in emissions and NO<sub>x</sub> losses have a large effect on the derived emissions, 4) data from one month are more noisy than averages over seasons or years, and 5) the interference of the changing data of the Chinese New Year unnecessarily complicates interpretation of the data. I think that the study would be more convincing if not only one month but two seasons (summer / winter) were analysed. A very first step in this direction would be the change of Fig. 2 as suggested below.

Response: We chose to quantify emissions of NO<sub>x</sub> in January completely based on considers for a larger signal of the economic downturn, a better relation between anthropogenic sources of NO<sub>x</sub> and VCDs of NO<sub>2</sub>, as well as an interesting timing of the Chinese New Year affecting also economic activities. We have added a much more detailed analysis, with a figure (Fig. 1), in the introduction to justify this choice.

We are aware of difficulties for retrievals in winter due to less optimal geometry and snow/ice on the ground, which have been discussed in the introduction. We also

understand that a month of data may be more susceptible to retrieval errors than data for a longer period. These are part of the reasons the TPG data are used to provide independent information to evaluate satellite-based results. Based on our finding, the satellite retrievals, particularly for the OMI product retrieved by KNMI, does a good job identifying the signal of the economic downturn, qualitatively and quantitatively. In addition, our previous studies also did not find significant problems in deriving emissions of NO<sub>x</sub> by choosing a month of retrieval products rather than a longer period (Lin et al., 2010b; Lin and McElroy, 2010).

Horizontal transport due to the longer lifetime of NO<sub>x</sub> is not a significant factor in this study, since we focus the analysis in Northern East China as a whole (with large area) so that the effect of transport inside the domain is not important (Lin and McElroy, 2010). The longer lifetime does affect to some extent the analysis for individual locations, as specified in the revised manuscript. This issue, however, is not particularly important as the lifetime of NO<sub>x</sub> is still relatively short in winter (i.e., 10-20 hours) not to allow for the pollutant to be transported too far from its source and, more importantly, as most of our analysis is done for the domain as a whole.

The diurnal variation of emissions is not an important issue in winter, as the longer lifetime of NO<sub>x</sub> significantly reduces its dependence on the timing of emissions (van Noije et al., 2006; see reference in the end). In summer, on the other hand, the lifetime of NO<sub>x</sub> is shorter, so the time a specific amount of NO<sub>x</sub> is emitted into the air becomes more important for its tropospheric abundance. This gives another reason for us not to include summer in quantifying emissions of NO<sub>x</sub> for purposes of analyzing the effect of the economic downturn.

The diurnal variation of NO<sub>x</sub> loss is not an issue here, as it is accounted for in CTM simulations for comparisons with satellite retrievals.

The occurrence of the Chinese New Year is another point of interest, rather than an ‘interference’, of our study. The coincident occurrence of the CNY in January 2009 in addition to the economic downturn makes the analysis of emission reduction more interesting, adding to our motivation to conduct the analysis for January. In addition, the CNY added another opportunity to test the capability of the retrievals in detecting short-term reductions in emissions. We have made these points clearly in the revised manuscript.

We have considered an analysis also for summer, July in particular. We then decided not to include it for several reasons. The economic downturn occurred mainly during late 2008 – mid 2009. In other words, the signal of the downturn is very small (if at all) for summer months, so it is much harder to detect from space. Second, a large portion of NO<sub>x</sub> is emitted from soil/lightning in summer, furthering complicating the analysis for the economic downturn. Third, in summer, the summer monsoon brings a lot of moisture to Northern East China, leading to more cloudy and/or precipitating days. As a result, the data coverage is much lower than that in winter, as shown in the figure below (Fig. R1). In addition, changes in diurnal cycles of emissions across the years may introduce interference in the analysis of emission trend for summer, due to the shorter lifetime of NO<sub>x</sub> (see above).

- For the separation of emission changes from other effects as well as for the inversion of emissions, the study relies on results from the GEOS-chem model. However, as stated several times in the paper and evident from Fig. 3, the model does not do a good job in reproducing observed NO<sub>2</sub> columns over China. This would be even more evident if the model fields had been added in Fig. 4 (which I would recommend). Please explain why one should trust a model that is off by more than a factor of 3 in the tropospheric columns.

Response: The model-retrieval differences in the magnitude of NO<sub>2</sub> VCDs are caused not only by model errors but also by retrieval errors, both of which were discussed extensively in the original manuscript. The analyses are organized and strengthened in Sect. 3 and Sect. 4 of the revised manuscript.

It is well known that retrievals tend to overestimate VCDs of NO<sub>2</sub>, particularly for KNMI products (by up to 40%). Relative to OMI\_NASA, the modeled VCDs are only 24% lower than the retrieved values in January 2006 when INTEx-B emission dataset (as representative of 2006) are used as model inputs. Scaling down the KNMI retrievals by a factor of 1.4 would lead to a model-retrieval difference of only ~ 30% for January 2006. The remaining differences are caused by model errors (including those in emission inputs) as well as retrieval errors that have not been fully taken into account. The model-retrieval differences are larger in January of later years because emission inputs for the CTM are unchanged (for purposes of evaluating the effect of non-emission factors on VCDs of NO<sub>2</sub>) while the actual emissions grew significantly over these years (except for January 2009). These points are organized and strengthened in Appendix A in the revised manuscript.

Despite potentially large retrieval errors in the magnitude of VCDs of NO<sub>2</sub> for individual months, the satellite products can be used to analyze trends of NO<sub>2</sub> VCDs and thus trends of emissions of NO<sub>x</sub>, as found by our study as well as many previous studies cited in our paper.

The main focus of our analysis is the effect of the economic downturn and the CNY for Northern East China as a whole. In addition, the magnitude of model-retrieval difference is relatively consistent across the domain. Therefore, we decided it is not necessary to include figures for spatial distributions of modeled VCDs.

- In several places in the manuscript it is stated, that the larger changes in winter are due to the longer lifetime in NO<sub>x</sub>. While this is true for absolute columns, I don't see why this should affect the relative changes discussed in the paper. Similarly, I don't understand why the fact that in winter "the magnitude of NO<sub>2</sub> VCD is most sensitive to changes in emissions" should have any impact on the relative changes. In my opinion, the difference between winter and summer trends is related to emission changes and to changes in NO<sub>x</sub> lifetime resulting of nonlinearities in NO<sub>x</sub> chemistry at these large NO<sub>x</sub> concentrations (see also next point). Please comment.

Response: In discussing the interannual trend of 12-month mean VCDs of NO<sub>2</sub>,

the statement that ‘the magnitude of VCDs of NO<sub>2</sub> increased most significantly in winter due to the longer lifetime of NO<sub>x</sub>’ was ambiguous. What we meant is that the significant seasonality of VCDs (values being 3-7 times larger in winter than in summer) results in a contribution to the 12-month mean value for a relative change of VCDs in winter larger than the contribution of the same amount of relative change in summer. Anyhow, this statement has been decided as unnecessary and was removed in the revised manuscript.

I don’t understand why the fact that in winter “the magnitude of NO<sub>2</sub> VCD is most sensitive to changes in emissions” should have any impact on the relative changes. – The comment comes from the place where we compared the impact of the economic downturn on 12-month mean VCD of NO<sub>2</sub> and the impact on 12-month TPG. First, by ‘changes in emissions’ we meant changes in anthropogenic emissions. In winter, tropospheric NO<sub>x</sub> come almost purely from anthropogenic emissions, at least for Northern East China. In summer, emissions from soil and lightning contribute to a large portion of NO<sub>x</sub> sources. Therefore, for a given amount of relative change in anthropogenic emissions, the relative change of the abundance of NO<sub>x</sub> in the troposphere will be larger in winter than in summer, given that other emission sources are held constant. Second, and more importantly, the reasons for the VCD-TPG difference were given for the timing of the downturn and the seasonality of VCDs and TPG. Based on the reviewer’s comment, we have decided the reasoning in the original manuscript was not written clearly enough, thus a much clearer analysis has been conducted in the revised manuscript to eliminate the ambiguity.

- Results from a GEOS-chem sensitivity study are reported where NO<sub>x</sub> emissions have been changed by 47% and “insignificant” non-linearities were detected. This is in contradiction to the results reported by Stavrou et al., 2008 who found quite large seasonal dependent changes in NO<sub>2</sub> lifetime as result of the changing NO<sub>x</sub> emissions in China. Please comment.

Response: The effect of changing NO<sub>x</sub> emissions is not just on the lifetime of NO<sub>x</sub> but also on the partitioning of NO<sub>x</sub> into NO and NO<sub>2</sub>. Stavrou et al. (2008) found a 10% increase in the lifetime of NO<sub>x</sub> in Northern East China due to a 70-100% increase in emissions of NO<sub>x</sub> from 1997 to 2006. Such nonlinear effect, however, is offset by a reduced NO<sub>2</sub>: NO ratio, leading to largely unchanged relation between emissions of NO<sub>x</sub> and VCDs of NO<sub>2</sub> (Martin et al., 2006). Martin et al. (2006) did a comprehensive test on this issue, with conclusions consistent with our findings.

We have put together and strengthened in Appendix B in the revised manuscript the analysis of the effect of changing emissions of NO<sub>x</sub>, CO, and NMVOC on the nitrogen chemistry under specified meteorological conditions.

- I find the lower panel in Fig. 2 very misleading for two reasons: 1) As the average is taken over the preceding 12 months, the timing of the observed reduction is shifted relative to the real changes 2) The minimum in NO<sub>2</sub> columns appears to be in summer 2009, the maximum in summer 2008. Comparison to the upper panel shows,

that differences between these two summers were quite small, and in fact, values in summer 2009 were higher, not lower than in summer 2008. I'd suggest to change this figure by either using annual averages from Jan – Dec or by computing time-series averages for each month and showing relative deviation from these values.

Response: The way the data was presented was to better identify the impact of the economic downturn. In particular, we would like to show the exact start of the economic downturn (i.e., the figure clearly and accurately shows that 12-month TPG starts to decline in October 2010, consistent with Fig.1 of the revised manuscript describing year-on-year changes of monthly TPG). Such way of presenting the data was chosen also in our previous study analyzing particulate air pollution in China (Lin et al., 2010a).

In clarifying the analysis of interannual variations of VCDs and TPG (in the revised manuscript Sect. 5), we changed the word 'annual' to '12-month mean' in order NOT to imply that the value only represents the periods starting from January and ending in December. At the beginning of Sect. 5, we clearly defined the meaning of '12-month mean', and stated that a '12-month mean' can be applied to any consecutive 12-month periods, e.g., Oct 2008 – Sep 2009.

The year-on-year growth rate of monthly TPG for each month is shown in the newly added Fig. 1 for purposes of analyzing the timing and intensity of the economic downturn.

- In section 5.2.2 the emissions derived from the different retrievals are compared. There are significant differences between the values, and it is concluded that “Overall changes in emissions of NO<sub>x</sub> derived from OMI-KNMI seem to best capture changes in emissions inferred from changes in TPG. Therefore OMI-KNMI is used in the following section...” I think this argument is flawed as it basically implies that out of the 4 data sets available, only the one that fits best to expectations is going to be used. There might be good reasons for using OMI-KNMI only, but the way the choice is justified is very unfortunate.

Response: We have detailed the analysis for results relative to each retrieval product in the revised manuscript. As was stated very clearly in the original manuscript (and strengthened in the revised manuscript), the independent results based on the TPG data were used to evaluate results derived from individual satellite retrievals for year-on-year changes in emissions of NO<sub>x</sub> in January. OMI\_KNMI showed up to best detect this particular event of emission change. Results from OMI\_KNMI were evaluated further in distinguishing the signal in January 2009 of the economic downturn from the effect of the CNY. In short, OMI\_KNMI was found to best capture the signal of the economic downturn by an objective analysis rather than subjective 'expectation' as the reviewer appeared to suggest.

Note that the present analysis cannot be used in any way to conclude that in general OMI\_KNMI is superior to other retrieval products. This kind of general conclusion should only be reached, if possible, when a systemic analysis is conducted

based on data in multi years, multi seasons and multi events from multi aspects, with causes of the retrieval differences identified quantitatively. This, however, is out of the scope of the present study.

- Numbers are given for deduced changes in emissions and attribution to economic downturn and CNY. However, in the absence of any estimates of the uncertainties of the values given, this is not very useful. Please add your estimates of the uncertainties of your results.

Response: Results from TPG and from four retrievals (OMI\_KNMI in particular) were analyzed side by side, providing a semi quantitative estimate of uncertainty in quantifying changes in emissions of NO<sub>x</sub> due to the economic downturn and the CNY. In addition, three relatively independent approaches were used to distinguish the effect of the downturn from that of the CNY, and consistency among them gave us further confidence on the accuracy of the emission estimate. It is very difficult, however, to pinpoint the exact uncertainty in our estimate due to many assumptions and likely errors (involved in the derivation of emission changes) which cannot be quantified accurately at the moment. Therefore we decide that it is not possible at the moment to assign a number to quantify the uncertainty in a very useful and meaningful sense.

#### **Minor Points**

- in Table 1, the spectral window given for SCIAMACHY is not correct

Response: Corrected.

- Please note that in the OMI product, the surface albedo data used changed on February 17, 2009. How does this affect your trend analysis?

Response: The point was added in Sect. 3. Its effect is 5% for retrieved VCDs in winter (Boersma et al., 2011), rather small as compared to the increase of 39% from January 2009 to January 2010.

- In section 4.2.1 you mention that in summer, NO<sub>2</sub> is located higher in the atmosphere where satellite observations are more sensitive which “tends to increase the retrieved NO<sub>2</sub> VC”. However, this effect should be corrected by the change in modelled NO<sub>2</sub> vertical distribution, so with the exception of the OMI-NASA product, this argument doesn’t hold.

Response: The discussion here is done for the seasonal variation of VCDs in the retrieval products, which are affected by the sensitivity of satellite sensors to the height of NO<sub>2</sub>.

- Please use the same scale in all upper panels in Fig. 3 to facilitate direct

comparison.

Response: We have separated and enlarged the figures for OMI, GOME-2 and SCIAMACHY retrievals in the revised manuscript (the new Figs. 4, 8, 9).

The scales are set differently for individual retrievals in order to better show year-on-year changes in VCDs relative to each product.

- The interference of the CNY which is discussed in the text in detail should also be taken up in the abstract

Response: The analysis for the CNY was shown in the abstract.

## Reviewer 2

The manuscript of Lin and McElroy describes retrievals of satellite data over China and their use to estimate emission data. This is a well established technique, as the long citation list shows. Anyhow, they use satellite data to get further detailed information on the effect of the economic downturn in China in 2008. The authors showed, that the decrease of emissions was partly overlay be the decrease due to the Chinese Spring Festival.

The first part of the manuscript is well written, the second parts needs major revisions. The authors use data of five different satellites. In the second part of the article the reader gets the impression the authors got lost in the amount of data. It was extremely difficult to follow their descriptions, especially in section 5.2.2.

Response: We thank the reviewer for the comments and suggestions. In accordance, we have restructured the manuscript, especially Sect. 5.2.2, to better present our analysis for the general reader. (see the general response)

The red line is missing in the second part. I strongly recommend to reorder the manuscript to avoid a mixing of technical details with results. Start with technical information on retrieval, model description, and the estimate of emission data. You may separate the impact of the economic downturn from influences of CNY on emissions and VRD. Clarify the double impact early in your paper.

Response: As stated above and in the general response, we have restructured the manuscript, taking the suggestion of the reviewer. Now we believe the analysis is much clearer.

The curves in the different figures are described in details, explanations of differences are mainly missing, as well as the interpretation of the data. Add a paragraph on the impact of meteorological parameters and how they are used for the calculation of emission data. You have to show some results of Rm. Anyhow, the results differ a lot from measured VCDs. Following your explanation, you have used an old model version. I strongly recommended to repeat the simulation with an updated model. At least indicate an possible impact on your results.

Response: (see the general response) In the revised manuscript, we have detailed the analysis of retrieved and modeled VCDs of NO<sub>2</sub> and subsequently derived emissions of NO<sub>x</sub> relative to each satellite product. We have analyzed changes in meteorological conditions and impacts on VCDs of NO<sub>2</sub>. We have analyzed model as well as retrieval errors in details.

The model-retrieval differences in the magnitude of NO<sub>2</sub> VCDs are caused not only by model errors but also by retrieval errors, both of which were discussed extensively in the original manuscript. The analyses are organized and strengthened in

Sect. 3 and Sect. 4 of the revised manuscript.

It is well known that retrievals tend to overestimate VCDs of NO<sub>2</sub>, particularly for KNMI products (by up to 40%). Relative to OMI\_NASA, the modeled VCDs are only 24% lower than the retrieved values in January 2006 when INTEx-B emission dataset (as representative of 2006) are used as model inputs. Scaling down the KNMI retrievals by a factor of 1.4 would lead to a model-retrieval difference of only ~ 30% for January 2006. The remaining differences are caused by model errors (including those in emission inputs) as well as retrieval errors that have not been fully taken into account. The model-retrieval differences are larger in January of later years because emission inputs for the CTM are unchanged (for purposes of evaluating the effect of non-emission factors on VCDs of NO<sub>2</sub>) while the actual emissions grew significantly over these years (except for January 2009). These points are organized and strengthened in Appendix A in the revised manuscript.

Despite potentially large retrieval errors in the magnitude of VCDs of NO<sub>2</sub> for individual months, the satellite products can be used to analyze trends of NO<sub>2</sub> VCDs and thus trends of emissions of NO<sub>x</sub>, as found by our study as well as many previous studies cited in our paper.

VCDs of NO<sub>2</sub> in the troposphere is determined by the lifetime of NO<sub>x</sub> and its partitioning into NO and NO<sub>2</sub>. To date we have not found any updates of the model modules that could significantly change the simulation of the nitrogen chemistry.

Sect. 4.2.1

Page 202, line 13: Maximum in winter: A long discussion is given in Zhang et al (2007), which is not cited in this context.

Response: added.

Sect. 4.2.2

Fig 2b: What is the reason for the time shift in max VCDs and TPG in 2008?

Response: The seasonality of TPG differs significantly from that of VCDs, for which variations in meteorological conditions play a major role. In addition, NO<sub>x</sub> is emitted not just from anthropogenic emissions (that are highly related to TPG) but also from soil and lightning, particularly in summer. Furthermore, satellite products are subject to retrieval errors and data coverage (particularly for SCIAMACHY). Variations in meteorological conditions and natural sources of NO<sub>x</sub>, together with uncertainties in satellite data, lead to the maximum of retrieved VCDs a little earlier than that of TPG and differing a little from each other as well.

Sect. 5.1

P 203, L 23-25: Numbers for decrease and increase in TPG differ from page 200, line 18-20. It is the 3rd repeat of these numbers. You may skip line 22-25 P203.

Response: the numbers in P203, L23-25 are for data in January only, differing

from the numbers in P200, L18-20 presenting 12-month means.

P204, L 8: The early date of CNY in 2009 is used to divide downturn by CNY and economics. This could be mentioned here.

Response: The discussion of the CNY, including the dates of the associated holiday, has been moved to the introduction in the revised manuscript.

#### Sect 5.2.1

Split the model description line 2 to 19

Why are identified errors still in the used model code? Why the updated N2O5 uptake rate is not used? To repeat the simulation in order to get better comparable results would strengthen the paper.

Response: The model description and error analysis have been moved to the newly added Sect. 4. Possible errors of the CTM are discussed in the uptake rate of N2O5 on aerosol surfaces, together with many other possible sources of model errors. At the moment, information is not sufficient to systematically improve the parameterization of N2O5 hydrolysis. For example, the studies of Bertram et al. (2009) and Brown et al. (2009) differ substantially in the importance for the uptake of N<sub>2</sub>O<sub>5</sub> of environmental parameters such as relative humidity and aerosol compositions. This point has been added in the revised manuscript.

#### Sect. 5.2.2.

This section remains unclear to me, even after reading it more than once. Sort your arguments and differ between retrieval data, model results and emission data. This section has to be rewritten, explanations to be included, not only descriptions of up and downs in the curves. Fig 3 is crowded and not readable in the small size given by ACPD. Split curves. Clarify how you calculate the emissions, e.g include a figure. At the moment, the function of the model remains unclear and the results are not used to explain your data. Table with description of simulations? Table with description of emissions?

Response: We have restructured the manuscript, Sect. 5.2.2 in particular, to clarify the analysis for emissions of NO<sub>x</sub> derived from space. (see general response) The issues raised here are addressed accordingly.

P 205, L 27: Why decrease from 2005 to 2006?

Response: This was a result of changing meteorological conditions (lower PBLH and higher air temperature in particular). The effect of variations in meteorological conditions on VCDs of NO<sub>2</sub> has been detailed in the revised manuscript (Sect. 6.2.1 and 6.2.2).

P 206, L 21 Show results of rm in a figure.

Response: Rm was shown as the red lines (in the original Figs. 3a,b and in the new Figs. 4, 8, and 9).

P 207, L 1: Change of ??? Interannually changes? Changes in Jan data?

Response: In the whole section, data for January are analyzed, as was stated clearly in the end of the introduction and at the beginning of Sect. 5 of the original manuscript. We have made this more clearly in the revised manuscript.

P 207, L 3-4: Changes reflect met. conditions? Blue line shows change in met condition. Red line is strongly influenced by the amount of valid retrieval data. Rm equal to red or blue line? Why differences between blue and green line? What does this mean for retrieval results? Please, give an explanation for the maximum values in 2008 without increasing emissions (Sciamachy).

Response: All the lines are explained and analyzed in detail in the revised manuscript (Sect. 6.2.1 and 6.2.2). The impacts of changing meteorological conditions are analyzed also for individual retrieval products, not just SCIAMACHY.

P 207, L 18-26: Move the paragraph

Response: We have put together and strengthened in Appendix B of the revised manuscript the analysis of the impacts of changes in emissions of NO<sub>x</sub>, CO and NMVOC on the nitrogen chemistry.

Figures:

Increase the font size

Response: Figures are revised as necessary.

Fig. 3:

It is almost impossible to see anything.

Separate model results, sat. data and emission data.

Increase the figure, split the curves, e.g. 1. to 4 and 5 to 8

Legend is not linked to the text. What means e.g. 'Emis w.r.t. 4 1'?

Better legend which describes the lines!

Skip GOME-2 results? Then you can enlarge your single figures.

Response: Figures are separated and enlarged for OMI, GOME-2 and SCIAMACHY retrievals (the new Figs. 4, 8, 9). Legends are revised.

Fig. 5:

Year 2009 is missing

Unit?

Difficult to divide light blue and grey.

Caption: '... See text...' add section here.

Very large values over the ocean. Why?

Response: Panels for January 2009 are skipped to save space, as the values are uniformly unity.

Data presented are ratios relative to January 2009, so there are no units.

Light blue and grey differ clearly on my monitor.

Section name added.

Large values over the ocean because ratios of relatively small values (with larger noise) are presented in the figures.

### Reviewer 3

#### General comments:

This is a thorough and well-written study that investigates trends in NO<sub>x</sub> emissions over northeastern China in the 2004-2010 timeframe. The authors have intelligently used the rhythm of the Chinese New Year to separate the effects of emission reductions because of the CNY holidays and those because of the economic crisis that happened to coincide in January-February 2009. They had to come up with this method, because they focused on the January months from 2005 to 2010. Whereas this choice is likely to give some advantages such as long NO<sub>2</sub> lifetimes, and probably a small role for non-linear chemistry, there are also some downsides to choosing these months only. These difficulties concern both the satellite retrievals (more likely to be affected by snow and clouds in winter than in summer) as well as inverse modelling (more susceptible to transport of NO<sub>x</sub> towards and from the study region). Nevertheless I think the numbers reported by the authors on changes in NO<sub>x</sub> emissions over northeastern China are plausible, although using inversions for some other months (July for example) would increase the robustness of this study. The authors are clearly up-to-date with the quality and issues with the state-of-science of OMI NO<sub>2</sub> retrievals, and their study also contributes to transparency on different retrieval philosophies. The topic is appropriate for ACP, but I think the paper still needs some major revisions.

Response: We thank the reviewer for useful comments and suggestions, which have been addressed carefully one by one.

#### Major issues:

On page 196, l21-23, January is discussed as the month of choice for year-to-year analysis. It is stated that in January "...the effect of economic activities on VCDs of NO<sub>2</sub> is most significant". It seems the authors mean to say here that the lifetime on NO<sub>2</sub> is generally longest in this month, and that therefore the effect of increasing emissions can be better observed than in a summer month when the NO<sub>2</sub> signal is weaker. Picking January has other advantages as well as drawbacks. The advantages that I think the authors are exploiting are: slower chemistry compared to summer, and less sensitivity to meteorology. But the drawbacks are a reduction in the number of samples that could have been achieved in summer, because of a larger probability of clouds and snow in winter. Besides, the solar zenith angles are generally higher in winter, leading to less optimal retrieval conditions. I think the authors need to at least discuss these issues when explaining their choices, and probably extending their analysis and also include July months from 2005 to 2010 would be a true test of the robustness of the methods and results presented here.

Response: We chose to quantify emissions of NO<sub>x</sub> in January completely based on considers for a larger signal of the economic downturn, a better relation between anthropogenic sources of NO<sub>x</sub> and VCDs of NO<sub>2</sub>, as well as an interesting timing of

the Chinese New Year affecting also economic activities. We have added a much more detailed analysis, with a figure (Fig. 1), in the introduction to justify this choice.

We are aware of difficulties for retrievals in winter due to less optimal geometry and snow/ice on the ground, which have been discussed in the introduction of the revised manuscript. We also understand that a month of data may be more susceptible to retrieval errors than data for a longer period. These are part of the reasons the TPG data are used to provide independent information to evaluate satellite-based results. Based on our finding, the satellite retrievals, particularly for the OMI product retrieved by KNMI, does a good job identifying the signal of the economic downturn, qualitatively and quantitatively. In addition, our previous studies also did not find significant problems in deriving emissions of NO<sub>x</sub> by choosing a month of retrieval products rather than a longer period (Lin et al., 2010b; Lin and McElroy, 2010).

We have considered an analysis also for summer, July in particular. We then decided not to include it for several considerations. The economic downturn occurred mainly during late 2008 – mid 2009. In other words, the signal of the downturn is very small (if at all) for summer months, so it is much harder to detect from space. Second, a large portion of NO<sub>x</sub> is emitted from soil/lightning in summer, furthering complicating the analysis for the economic downturn. Third, in summer, the summer monsoon brings a lot of moisture to Northern East China, leading to more cloudy and/or precipitating days. As a result, the data coverage in summer is much lower than that in winter, as shown in the figure below (Fig. R1).

P196, l22-23: '...January from 2005 to 2010 when the effect of economic activities on VCDs of NO<sub>2</sub> is most significant.' Are the authors implicitly suggesting that the effect of economic activities is strongest because the lifetime of NO<sub>2</sub> is longest in January? If so, please clarify (like the authors do in 4.2.1).

Response: What we meant was that the economic downturn was most intense in January 2009, and, by comparing emissions of NO<sub>x</sub> derived for all January months, the effect of the downturn on emissions could be detected most easily from space without significant interference from natural sources of NO<sub>x</sub> and retrieval errors. Anyhow, we have detailed in the introduction of the revised manuscript the analysis of the choice of January 2005-2010 in quantifying year-on-year changes in emissions of NO<sub>x</sub> and impacts of the economic downturn.

P203, l5: 'The reductions (in VCDs) were about twice as large as the reduction in annual TPG'. This is an interesting statement, in the context of the abstract stating that the 49% increase in total power generation apparently only leads to a 30% increase in NO<sub>2</sub> VCDs. Can the authors provide some more information about why changes in TPG would lead to a response in NO<sub>2</sub> that is approximately half as strong? Did only TPG change, and were other sources constant?

Response: The reductions in VCDs due to the economic downturn were larger than that for TPG. This was a result of 1) the timing of the economic downturn (most

intense in the 2008-2009 winter) and 2) differences in seasonal variability between TPG (small seasonality) and retrieved VCDs (values in winter are 3-7 times larger than those in summer). We have revised the manuscript to deliver an extensive explanation (see Sect. 5.2.2).

Prior to the downturn, retrieved VCDs of NO<sub>2</sub> increased at a rate lower than the increase of TPG. This was because anthropogenic emission activities (represented by TPG) grew consistently across the seasons (i.e., without strong dependence of the growth rate on seasons) while emission factors were reduced also relatively consistently across the seasons. In addition, natural emissions of NO<sub>x</sub> may not grow during the years prior to the downturn.

P207, l18-26: Although I credit the authors for undertaking the sensitivity test described here, changes in NO<sub>x</sub> emissions are not the only reason for nonlinear chemistry effects leading to nonlinearities in the NO<sub>x</sub> emissions: NO<sub>2</sub> column ratios. The other important factor could be changes in VOC and CO emissions that also have significant effects on oxidant chemistry, especially in VOC-limited regions like northeastern China. Therefore, I think that the statement that changes in emissions of NO<sub>x</sub> ... were affected insignificantly by the nonlinear photochemistry lacks basis. Probably these changes, similar to changes in NO<sub>x</sub> emissions, are more important for non-linear chemistry in summer than in winter, but they should not be discarded here.

Response: The effect of changing emissions of NO<sub>x</sub>, CO and NMVOC, under specified meteorological conditions, on the nitrogen chemistry is organized and detailed in Appendix B of the revised manuscript. In short, our statement holds about the relation between emissions of NO<sub>x</sub> and VCDs of NO<sub>2</sub> being not affected significantly by emissions of NO<sub>x</sub>, CO, NMVOC for the range of emissions relevant for the present analysis.

Since 17 February 2009, OMI KNMI retrievals use the OMI Lambert Equivalent Reflectance (LER) as input for the air mass factor calculations. Prior to that date the LER used was the TOMS/GOME set. This change leads to reductions in tropospheric NO<sub>2</sub> of 5% over eastern China, as shown in various international presentations, and also presented in a draft manuscript publicly accessible through [www.temis.nl](http://www.temis.nl) <<http://www.temis.nl>> . The changes for July are smaller. I think the authors need to be aware of these results, since it seems that their analysis, at least for January 2010, would be affected by this change. In any case they need to account for it in their study.

Response: We thank the reviewer for the information. The change of surface reflectance dataset has been discussed in the revised manuscript. Nonetheless, the 5% change in retrieved VCDs of NO<sub>2</sub> due to changing surface reflectance datasets is relatively small compared to the increase of about 39% from January 2009 to January 2010.

Specific comments:

Title: since most of the results indicate an overall increase in NO<sub>x</sub> emissions in the 2004-2010 timeframe, with only temporary reductions in 2009, it seems odd to focus in the title on this temporary reduction in emissions.

Response: The increase in NO<sub>x</sub> emissions over the past decade have been addressed in previous studies. Thus it is not a very new finding compared to results for the effect of the downturn. The general increase is analyzed here for purposes of contrasting the emission reduction due to the downturn.

P194, l8: I'm not familiar with the names of Chinese regions, but I think 'northeastern China' is the correct way to indicate this region rather than 'North Eastern China'.

Response: 'northeastern China' represents the far northeastern part of China, and is not a correct name for the domain in the present analysis.

P194, l21: The rapid increase occurred mainly during the last decade, compared to the more modest economic growth between 1980-2000, if I'm not mistaken?

Response: The growth of emissions of NO<sub>x</sub> was already very rapid in the 1990s (Zhang et al., 2007).

P195, l13: the name should be spelled Jaegl é

Response: Corrected.

P195, l29: it would be appropriate to also cite van der A et al. [2006] here who were the first to apply a sophisticated trend analysis model (that accounts for the seasonality and provides detailed information on the statistical significance of the trend) to the remote sensing data over China.

Response: The reference is added.

P202, l10-114: I think the authors need to clarify the presumed mechanism here, i.e. that OMI observations at 13:40 hrs are more sensitive to the OH+NO<sub>2</sub> reaction leading to NO<sub>x</sub> loss than the early morning retrievals from GOME-2 and SCIAMACHY.

Response: The mechanism has been clarified.

P203, l8-10: 'Thus ... VCD of NO<sub>2</sub>'. This sentence is unclear. Could the authors please clarify what they mean?

Response: As stated above, this part of the manuscript has been revised to clarify the reasoning why the effect of the economic downturn was larger on retrieved VCDs than on TPG.

P207, 15-7: in Figure 3, the effect of applying the averaging kernel looks small to me. Could the authors quantify the differences between NO<sub>2</sub> columns with and without applying the kernel?

Response: The differences are about  $0.4-0.8 \times 10^{15}$  molec./cm<sup>2</sup> (or 6-12%) across the January months averaged over Northern East China, as clearly shown in the original Fig. 3c and the new Fig. 4a.

P207, 19-11: could the authors please clarify this sentence?

Response: What we meant was that monthly mean values calculated from model results in all days differ from the values calculated from model results sampled based on days with valid retrieval data. Anyhow, we have detailed the analysis of the impact of data coverage in the revised manuscript. (see general response)

P208, 117: typo 'SCIMACHY' should be 'SCIAMACHY'.

Response: Corrected.

P209, 11-2: I would suggest to add to this sentence that the effect of CNY is a reduction of 12%.

Response: Changed.

P209, 110-14: this part is vague. I suggest the authors rephrase it to make it more clear what they exactly did here.

Response: the paragraph has been revised.

P210, 113-15: I suggest that in addition to 'prior to the downturn', the authors also provide the exact period for which the 27-33% reduction holds.

Response: The time has been specified.

# References:

van Noije, T. P. C., Eskes, H. J., Dentener, F. J., Stevenson, D. S., Ellingsen, K., Schultz, M. G., Wild, O., Amann, M., Atherton, C. S., Bergmann, D. J., Bey, I., Boersma, K. F., Butler, T., Cofala, J., Drevet, J., Fiore, A. M., Gauss, M., Hauglustaine, D. A., Horowitz, L. W., Isaksen, I. S. A., Krol, M. C., Lamarque, J. F., Lawrence, M. G., Martin, R. V., Montanaro, V., Muller, J. F., Pitari, G., Prather, M. J., Pyle, J. A., Richter, A., Rodriguez, J. M., Savage, N. H., Strahan, S. E., Sudo, K., Szopa, S., and van Roozendaal, M.: Multi-model ensemble simulations of tropospheric NO<sub>2</sub> compared with GOME retrievals for the year 2000, *Atmos. Chem. Phys.*, 6, 2943-2979, 2006.

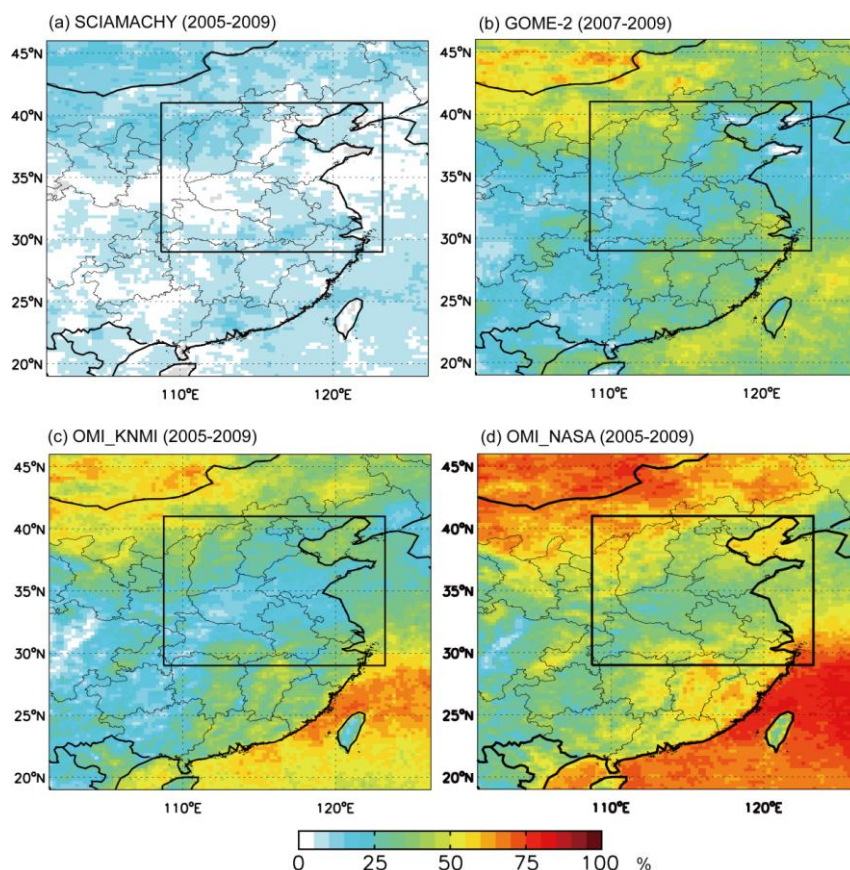


Figure R1. The percentage of days in July with valid data averaged over 2005 – 2010 in the four retrieval products. OMI\_NASA has more coverage than OMI\_KNMI because of weaker criteria for cloud screening. Also shown is boundary specifications (black lines) of Northern East China analyzed in this study. The data coverage here is much lower than that in January (Fig. 2)