Reviewer 1

Review of "Rapid growth in nitrogen dioxide pollution over Western China, 2005–2013" by Cui et al. In this paper, a detailed look is taken on NO_2 growth rates in Western China based on OMI NO_2 data. A wavelet analysis is performed on the background corrected gridded time series, linear trends are computed on the long-term trend component and compared to a nested GEOS-Chem run using scaled emissions, and the results are discussed in the context of economic and legislative development in China. The paper is clearly structured, well written, and contains many interesting results and discussions. The topic of NO_x emission trends and their impact on the atmosphere is of large interest in particular for rapidly developing regions such as China, the Western part of China not having received much attention in the past, and this study fits well into the scope of ACP. However, I do have concerns about several aspects of the study as listed below. I therefore recommend this paper for publication in ACP only if these comments have been addressed in a satisfactory way.

We thank the reviewer for comments, which have been incorporated into the revised manuscript.

Major comments

1. I'm confused by the description of the background correction:

• The authors claim that they use the seasonality of NO_2 to remove the natural contributions and provide a map of seasonality in Fig, 2.a in the manuscript. However, I cannot see where the seasonality information is then used with the possible exception of motivating the choice of the 1E15 molec cm-2 threshold applied to identify polluted pixels. It's also not clear where the 1E15 threshold is actually being used – I have the impression that the removal of nonanthropogenic contributions is done by simply subtracting monthly averages derived over certain background regions.

We have re-structured Sect. 3 to better show 1) how we find human-dominated locations, and 2) how we subtract the background values.

Indeed, the seasonality analysis is used to determine that locations with NO_2 exceeding 1×10^{15} cm^{-2} are dominated by anthropogenic emissions. We select these locations, and then subtract their NO_2 values by certain background values. The method to finding the background values is the same as Russell et al. (2012) for the United States.

• In this context it is not clear to me if all the values in the background regions shown in Fig. 1 are used or if they have been further filtered by the NO_2 threshold value.

We have clarified how we remove the background values in the new Sect. 3.2. All the values in the background regions are used. Over the background regions in the west,

NO_2 values at all grid cells are smaller than 1 x 10^{15} cm^{-2}.

The background values for YRD and PRD exceed 1 x 10^{15} cm^{-2}, as they may contain certain anthropogenic influence (e.g., due to horizontal transport from polluted regions). However, these background values are 6-7 times smaller than NO_2 over their corresponding polluted regions. Thus the effect of residual anthropogenic influence on our trend calculations is small.

• The amount of NO_2 in the background regions is stated to be small. However, from Table 1 it is clear that the background values are in several cases on the order of 30

Table 1 shows that the background values are 0.4-0.5 x 10^{15} cm^{-2} over the west and 0.7-1.2 x 10^{15} cm^{-2} over the east. These numbers are all much smaller than NO_2 over their corresponding polluted areas. Table 1 does not show any values in the order of 30.

• It is also not obvious that it makes sense to subtract the NO_2 columns from background regions, as soil and lightning emissions have specific regional patterns and cannot simply be assumed to be homogeneous over the large areas discussed here. In particular I would expect lower NO_x soil emissions in urban areas than in the rural background regions used.

We have added a detailed discussion of this aspect in the end of the new Sect. 3.2:

"Note that the chosen "background" values may not fully represent the actual natural contributions to the targeted human-dominant areas. For example, soil emissions may vary in space due to differences in temperature, radiation, land cover and land use type, and other climatic factors. Lightning emissions of NO_x may have spatial dependence as well. The "background" regions may not be totally free from anthropogenic influences, as certain amount of NO_x in the polluted areas may be oxidized to produce peroxyacyl nitrates (PANs), which can be transported to "background" areas and converted back to NO_x. For these reasons, our choice of background values is relatively rough. Nevertheless, unless the actual natural contributions differ substantially from the chosen values, which we do not expect to occur on a provincial average, the resulting effect on our trend calculations should be small, because the chosen background values are smaller than NO_2 over their corresponding polluted areas by a factor of 2–13 (Table 1). Future work is needed to fully separate the anthropogenic from natural NO_2 for individual locations."

Table R1 compares the trends with and without subtracting the "background" values. The two methods lead to similar results. In general, trends (%/yr relative to 2005) are enhanced when the "background" values are removed, especially for the northwestern provinces. This information is summarized in Sect. 4.2.1.

Table R1. Trends of OMI NO_2 VCDs over 2005-2013 (% yr^{-1}, relative to 2005).

	Region	Trend with "background" removed	Trend without removing "background"
Northwest	Gansu	7.5	7.0
	Inner Mongolia	10.2	9.0
	Ningxia	12.3	10.8
	Qinghai	11.2	9.6
	Shaanxi	10.5	9.7
	Xinjiang	15.1	12.2
Southwest	Chongqing	7.8	7.0
	Guangxi	4.0	3.9
	Guizhou	6.9	5.9
	Sichuan	6.1	5.5
	Yunnan	4.2	4.0
Region	West	8.6	7.9
	Northwest	11.3	9.8
	Southwest	5.9	5.2
	BTH	5.3	5.2
	YRD	4.1	4.0
	PRD	-3.3	-2.4

2. I'm not convinced by the usefulness of the wavelet analysis applied to the data prior to the trend determination. First of all, there seems to be a subjective element in the choice of "The approximate signal A5" used as representation of the long-term trend. I think the authors need to

• show how their results derived using a wavelet analysis compare to results from standard trend models used in previous work and explain why their approach is to be preferred

As explained in the new Sect. 3.3:

"Due in part to the short lifetime of NO_x, the tropospheric NO_2 VCDs respond quickly to emission changes at various temporal scales, from a general growth along with socioeconomic development to short-term perturbations such as the Chinese New Year holidays and the economic recession (Lin and McElroy, 2011;Lin et al., 2013). Also, uncertainties and sampling biases in the satellite data may introduce additional noises in the NO_2 monthly time series. If not separated, these short-term variability and noises may affect linear trend calculations.

Here we conducted discrete wavelet transform (DWT) (Daubechies, 1992;Partal and Küçük, 2006) to distinguish temporal variability of NO_2 at multiple scales. The wavelet transform is a useful tool for diagnosing the multi-scale and non-stationary processes over finite space and time periods, with the advantage of localization in the time and frequency domain (Echer, 2004;Percival and Walden, 2006), suitable for our analysis of NO_2 trends and variability. We chose Meyer orthogonal discrete wavelets as the wavelet functions which have been used to study ozone column, NDVI and land-cover changes (Abry, 1997;Echer, 2004;Freitas and Shimabukuro, 2008;Martínez and Gilabert, 2009). Different from the approaches adopted by previous NO_2 studies (e.g., (van der A et al., 2006)), our wavelet analysis does not require prior assumptions about seasonality and other temporal scales. As shown in Sect. 3.1, the magnitude of NO_2 seasonality is associated with the amount of annual mean NO_2 and anthropogenic sources, and this information is easily captured by the wavelet analysis here."

Table R2 compares the linear trends calculated based on the *A5* component against the trends based on the original time series. It is clear that the two methods produce similar trends. However, as the wavelet transform removes small-scale variability and noises, we believe the A5-based trends are more robust in general. We have added this information in the new Sect. 4.2.1.

Table R2. Trends of OMI NO_2 VCDs over 2005-2013.

	Region	A5-based trend (% yr^{-1}, relative to 2005)	Trend based on the original time series (% yr^{-1}, relative to 2005)
Northwest	Gansu	7.5	7.4
	Inner Mongolia	10.2	12.1
	Ningxia	12.3	13.4
	Qinghai	11.2	11.2
	Shaanxi	10.5	10.7
	Xinjiang	15.1	14.7
Southwest	Chongqing	7.8	7.5
	Guangxi	4.0	4.2
	Guizhou	6.9	7.2
	Sichuan	6.1	5.5
	Yunnan	4.2	3.5
Region	West	8.6	8.7
	Northwest	11.3	11.7
	Southwest	5.9	5.6
	BTH	5.3	6.4
	YRD	4.1	4.4
	PRD	-3.3	-2.9

• explain in more detail how the wavelet analysis was performed and why they think A5 is a good representation of the long-term trend, how they can identify D3 as seasonality and why they can be sure that details of the wavelet analysis do not impact on the trend determination

In wavelet transform for multi-scale analysis, the original time series is decomposed to several components of various temporal scales through an iterative multi-layer process, with an "approximation" signal and a "detail" signal for each layer of decomposition. In the first layer of decomposition, $f(t) = A_1 + D_1$. Then, $A_1 = A_2 + D_2$ and $f(t) = A_2 + D_2 + D_1$. And so on. The iteration stops when the period of the approximation time series is longer than the length of the dataset (e.g., 116 months in this study). At level 5, the period of the *A5* time series is longer than the length of the dataset (116 months). This criterion is typically used in investigating the long-term trend of a time series (Echer, 2004;Chen et al., 2014). In this study, we find the iteration stops at level 5 for any given province. We have added this information in the new Sect. 3.3.

Figure R1 shows the wavelet decomposition results for a grid cell in Xi'an, Shaanxi Province, with both approximation and detail components in each layer of decomposition. This grid cell is also illustrated in Fig. 3 of the main text. Figure R1 shows that the approximation component *A5* is the long-term signal, and its period is longer than the length of our dataset (116 months). Thus the iteration stops here.

Table R2 compares the linear trends calculated based on the *A5* component against the trends based on the original time series. It is clear that the two methods produce similar trends. However, as the wavelet transform removes small-scale variability and noises, we believe the *A5*-based trends are more robust in general. We have added this information in the new Sect. 4.2.1.

D3 is indicative of the seasonality. It has a period of about 12 months, and its phase is in line with the seasonal pattern of the original time series. Nevertheless, we have decided to remove the discussion of D3 (seasonality), as the main focus of this study is NO2 trends.

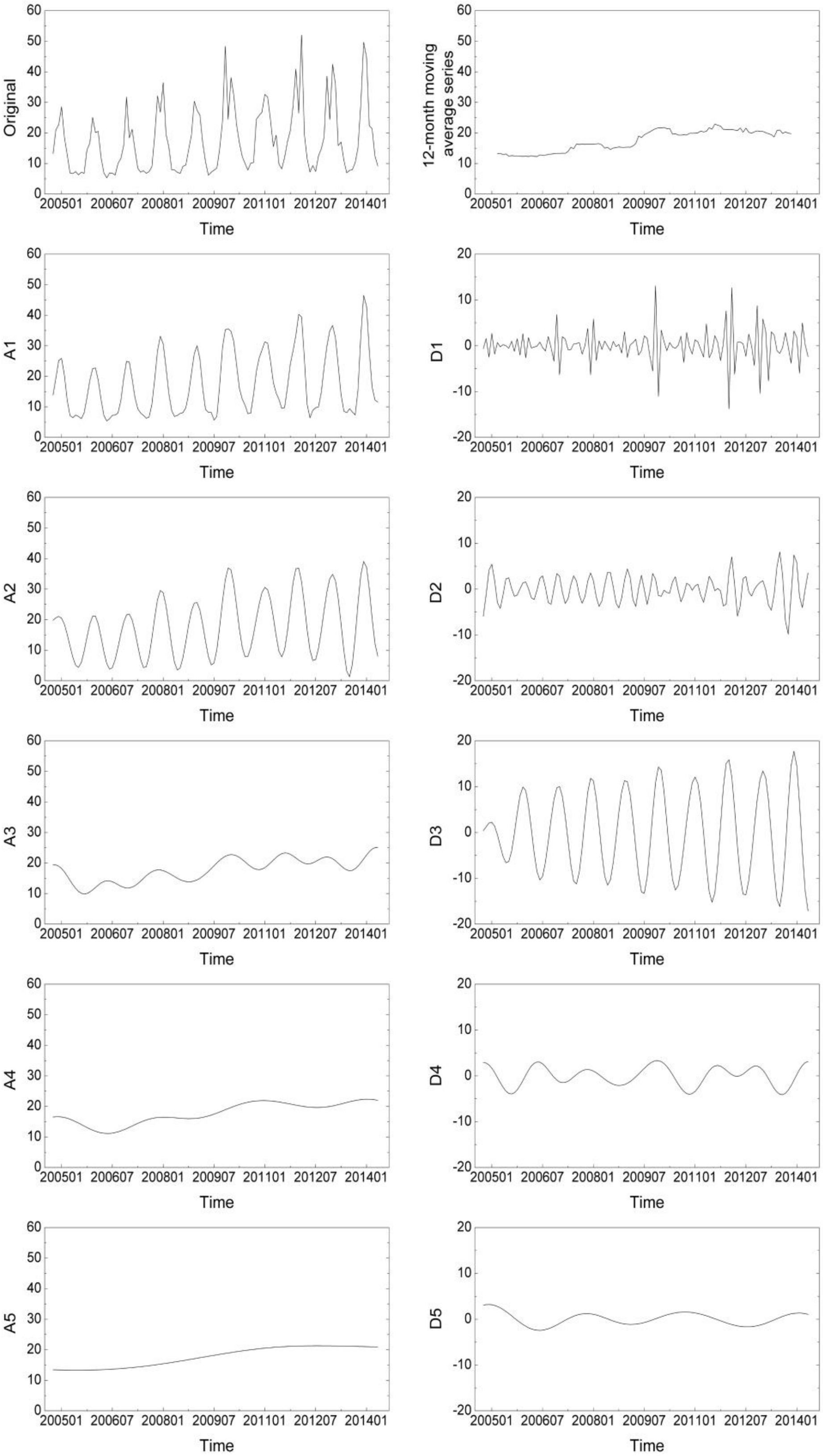

Figure R1. Wavelet analysis results for the OMI NO_2 time series at a grid cell in Xi'an, Shaanxi (34.5 °N, 108.9 °E). The top left panel shows the original monthly time series, and the top right panel displays the 12-month moving average time series.

3. In several places, the argument is made that the good agreement between model and data indicates that the trends observed are anthropogenic. While I'm convinced that the trends are anthropogenic, I don't see how the approach taken can prove that. As the emissions used are only available for one year, the authors scale the inventory by using the relative change of the OMI columns. To me it appears evident that such a procedure will lead to broadly consistent model and satellite trends (ignoring non-linearity effects) and I wonder what really can be learned from this exercise. In this context it is not clear

- what the spatial resolution of the MEIC inventory is

- how the scaling with OMI data was done – was this on a 0.25 x 0.25 degrees grid?

The spatial resolution of MEIC used in the simulation is 0.667 x 0.5 degree, according to the model resolution. The scaling with OMI data is done at the same resolution, after regridding the 0.25 x 0.25 degree satellite data to 0.667 x 0.5 degree. This information has been added in the revised Sect. 2.2.

We agree that scaling model anthropogenic emissions based on OMI NO_2 trends will lead to model NO_2 trends broadly consistent with OMI trends, if natural sources and meteorological conditions are not changed drastically. Therefore, in the new Sect. 4.2.1, we have added an additional model sensitivity simulation and associated discussion to confirm that anthropogenic emissions are the dominant factor of OMI NO_2 trends:

"To further confirm that anthropogenic emissions are the main driver of the observed NO_2 trends, we conducted an additional model simulation for 2012 where anthropogenic emissions are fixed at the 2005 levels (while natural emissions and meteorology correspond to the 2012 levels). We contrasted the model NO_2 change from 2005 to 2012 in this case to the standard case that has included year-specific anthropogenic emissions. Table 3 shows that inclusion of anthropogenic emission changes from 2005 to 2012 leads to large changes in model NO_2, and keeping anthropogenic emissions unchanged leads to much reduced changes in NO_2. The NO_2 growth reduces from 85.8% to 6.9% averaged over the northwestern provinces and from 46.8% to -6.3% over Southwestern China."

Minor comments:

Page 34916, line 12: to evaluating pollution => evaluating pollution

Modified.

Page 34918, line 10: are referred to => are described in

Modified.

Page 34918, line 15: As the AVK is given on satellite pixel basis, it is not clear how they are transferred to the model grid – please give more detail

In the revised Sect. 2.2, we have added: "Following our previous work (Lin et al., 2010;Lin, 2012), we regridded the pixel-specific AK to the 0.25° × 0.25° grid."

Page 34918, line 22: inventory => inventories

Modified.

Page 34921, line 24: DTW => DWT

Modified.

Page 34924, line 10: of other => other

Modified.

Page 34925, line 8: in all days => on all days

Modified.

Page 34925, line 21: but with a reduction in summer => but reduces it in summer

Modified.

Page 34926, line 22: Fig. 7 => Fig. 6

Modified.

Page 34928, line 22: growth rate of what?

Modified as " The average NO_2 growth rate".

Figure 2: NO_2 columns, not concentrations

Modified.

Figure 2: grid cell is sorted => grid cells are sorted

Modified.

Figure 4: As the topic of this paper is Western China, please add a scatter plot for the points of the study region

Figure R2 shows the scatterplot with linear regression for model vs. OMI NO_2 over Western China in 2012. We have updated the text and Table 2 to show the regression results for all years over Western China. Overall, the model-OMI consistency is very high (R^2 = 0.68–0.76 over 2005–2012).

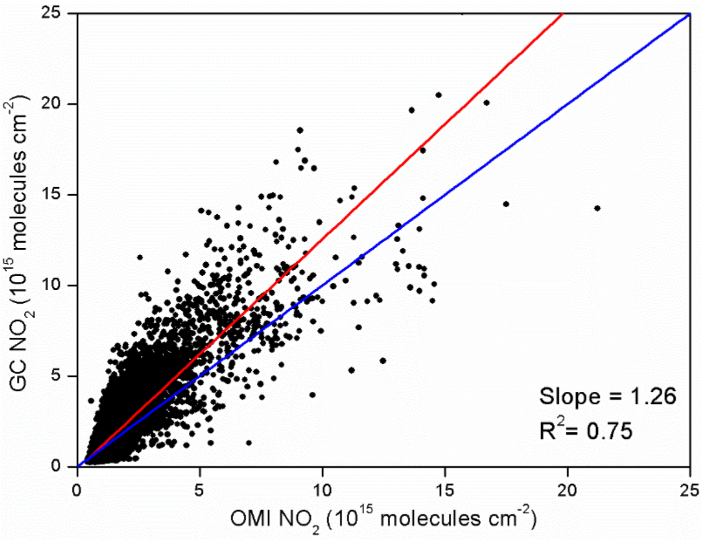

Figure R2. The scatterplot with linear regression for model vs. OMI NO_2 over Western China in 2012. The red line indicates the linear fit and the blue line is the 1:1 line.

Figure 4 and Figure 5: Colour scale difficult to read for colour blind readers

We have tried several color scales and have found the chosen one most readable for most readers.

Figure 5: subtracted by its => subtracted by their

Modified.

Figure 7: As stated above, I'm not convinced that this is the seasonality in the sense that for a given year, it reflects the seasonal change in NO_2 column. For example, the amplitude for Shaanxi increases by more than a factor of 2 during 2005 which appears unrealistic to me.

We have decided to remove the discussion on D3 (seasonality), as the main focus of this study is NO2 trends.

Reviewer 2

This manuscript, titled "Rapid growth in nitrogen dioxide pollution over Western China,

2005-2013" by Cui et al. is an interesting work, analyzing the recent NO_x emission trend over Western China using OMI observations. The paper is clearly written, except for a few noted word choices, and is well-suited for publication to ACP. However, there are several concerns that should be addressed carefully before being accepted for publication.

We thank the reviewer for comments, which have been incorporated into the revised manuscript.

Major comments:

1. The reliability of the wavelet decomposition analysis. This method is highlighted for being independent of prior assumptions. But the decomposition number is determined by the authors. How is the decomposition number selected? Is there any criteria? Will the estimated trend change if the decomposition number changes?

Please see our response to Reviewer 1 (major comment 2) for details. In particular, the decomposition is done through an iterative process, which stops when the period of the last approximation component (A5 in this study) is longer than the length of the dataset (116 months here). This criterion is typically used in investigating the long-term trend of a time series. Also, the A5-based trends are consistent with the linear trends calculated based on the original time series.

2. The reliability of subtracting "background". As far as I understand, the results will not change significantly without subtracting the background. If so, why bother?

We have re-structured Sect. 3 to better clarify why and how we treat the "background" values. Please see the new Sect. 3.2 for more detailed discussion of "background" values. In particular, the new Sect. 3.2 states that "To obtain the sole anthropogenic NO_2, we further subtracted all NO_2 VCDs by certain "background" values representing the natural influences. Removing the "background" influences is meaningful for Western China where the NO_2 VCDs are currently not at an extremely high level (see Sect. 4.2.1)."

As shown in our response to Reviewer 1 (major comment 1), Table R1 compares the trends with and without subtracting the "background" values. The two methods lead to similar results. In general, trends (%/yr relative to 2005) are enhanced when the "background" values are removed, especially for the northwestern provinces. This information is summarized in Sect. 4.2.1.

3. 34918, L6: OMI NO_2 is used to scale base-year emissions and further drives model simulations. What's the uncertainty of this assumptions? Will it be the major contributor to the agreement between OMI observations and model simulations?

As already discussed in the original manuscript, uncertainties may rise from the nonlinear relation between emissions and VCDs. We agree that scaling model anthropogenic emissions based on OMI NO_2 trends will lead to model NO_2 trends broadly consistent with OMI trends, if natural sources and meteorological conditions are not changed drastically. Therefore, in the new Sect. 4.2.1, we have added an additional model sensitivity simulation and associated discussion to confirm that anthropogenic emissions are the dominant factor of OMI NO_2 trends:

"To further confirm that anthropogenic emissions are the main driver of the observed NO_2 trends, we conducted an additional model simulation for 2012 where anthropogenic emissions are fixed at the 2005 levels (while natural emissions and meteorology correspond to the 2012 levels). We contrasted the model NO_2 change from 2005 to 2012 in this case to the standard case that has included year-specific anthropogenic emissions. Table 3 shows that inclusion of anthropogenic emission changes from 2005 to 2012 leads to large changes in model NO_2, and keeping anthropogenic emissions unchanged leads to much reduced changes in NO_2. The NO_2 growth reduces from 85.8% to 6.9% averaged over the northwestern provinces and from 46.8% to -6.3% over Southwestern China."

Specific comments:

1. 34914, L12: Consider different word use than "provincial regions".

Modified as "provincial-level regions".

2. 34916, L5: Please cite some literatures associated with emission inventories directly.

We have added a citation.

3. 34917, L9: Please check "30%+0.7*10^15".

Modified as "On a regional and monthly mean basis, the overall error of retrieved VCDs is about 30% (a relative error) plus 0.7×10^{15} molecules cm^{-2} (an absolute error)".

4. 34920, L9: The conclusion is similar with that in van der A et al. (2006). Some discussion about his work is recommended. In addition, Fig 2a is not quite straightforward. Please consider a new form.

The discussion and reference is added. We have further improved the explanations of Fig. 2a.

5. 34929, L18: What does “Qianghai province” refer to?

Modified as “Qinghai Province”.

6. Figure 4: Please add the meaning of the red and blue lines in the scatterplot.

Modified. In the scatterplot, the red line represents a linear fit, and the blue line is the 1:1 line.

7. Figure 6: The font size is too small to read.

Modified.