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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Rapid growth in nitrogen dioxide pollution over Western China, 2005–2013" *by* Y.-Z. Cui et al.

Y.-Z. Cui et al.

linjt@pku.edu.cn

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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 NO2 growth rates in Western China based on OMI NO2 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 NOx 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 NO2 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 NO2 exceeding 1 x 1015 cm-2 are dominated by anthropogenic emissions. We select these locations, and then subtract their NO2 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 NO2 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, NO2 values at all grid cells are smaller than 1 x 1015 cm-2. The background values for YRD and PRD exceed 1 x 1015 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 NO2 over their corresponding polluted regions. Thus the effect of residual anthropogenic influence on our trend calculations is small.

The amount of NO2 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 1015 cm-2 over the west and 0.7-1.2 x 1015 cm-2 over the east. These numbers are all much smaller than NO2 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 NO2 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 NOx 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 NOx may have spatial dependence as well. The "background" regions may not be totally free from anthropogenic influences, as certain amount of NOx in the polluted areas may be oxidized to produce peroxyacyl nitrates (PANs), which can be transported to "background" areas and converted back to NOx. 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 NO2 over their corresponding polluted areas by a factor of 2–13 (Table 1). Future work is needed to fully separate the anthropogenic from natural NO2 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.

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 NOx, the tropospheric NO2 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 NO2 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 NO2 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 NO2 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 et al., 2007;Freitas and Shimabukuro, 2008).

Different from the approaches adopted by previous NO2 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 NO2 seasonality is associated with the amount of annual mean NO2 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.

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,ãĂŰ AãĂŮ_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.

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 NO2 trends will lead to model NO2 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 NO2 trends: “To further confirm that anthropogenic emissions are the main driver of the observed NO2 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 NO2 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 NO2, and keeping anthropogenic emissions unchanged leads to much reduced changes in NO2. The NO2 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 $0.25^\circ \times 0.25^\circ$ 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 NO2 growth rate”.

Figure 2: NO2 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 NO2 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 ($R2 = 0.68–0.76$ over 2005–2012).

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 NO2 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.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 34913, 2015.

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

Fig. 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

Fig. 2.

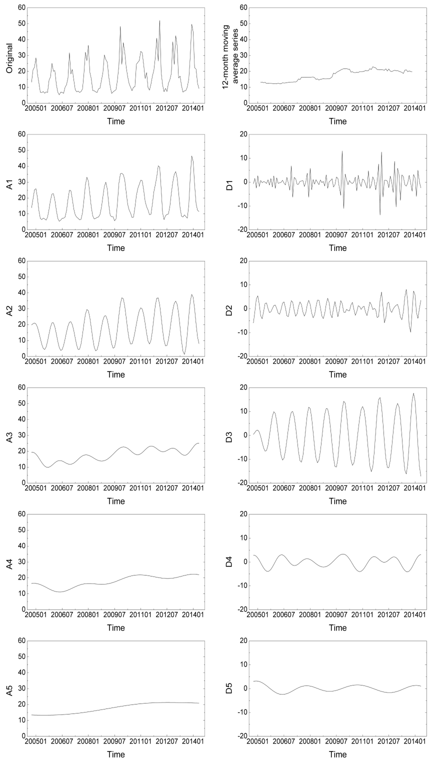

Fig. 3. Figure R1. Wavelet analysis results for the OMI NO2 time series at a grid cell in Xi'an, Shaanxi (34.5°N, 108.9°E). The top left panel shows the original monthly time series, and the top right panel d

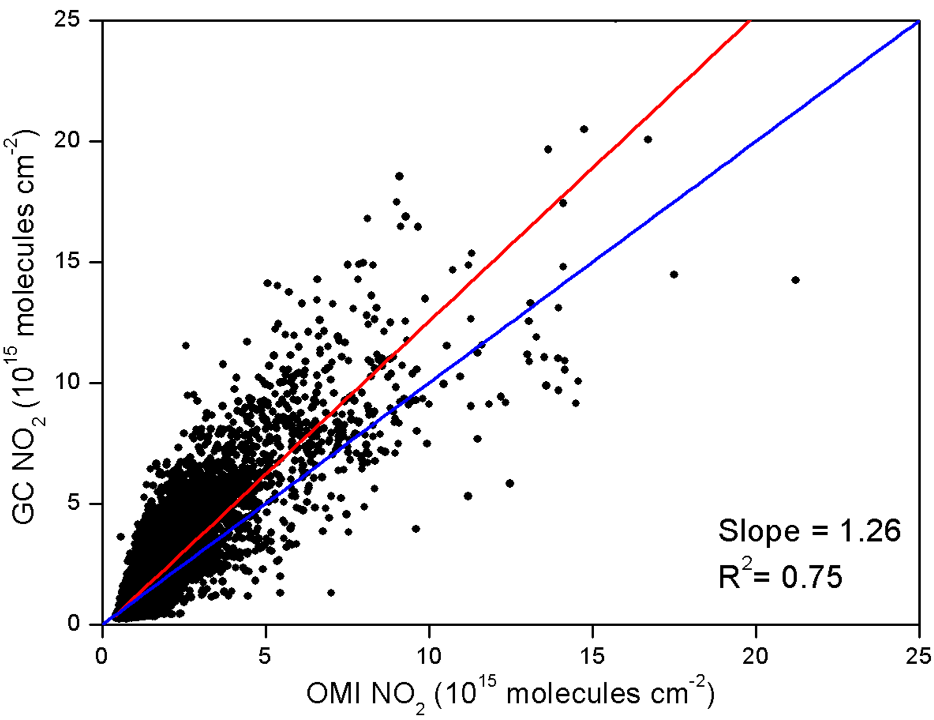

Fig. 4. Figure R2. The scatterplot with linear regression for model vs. OMI NO2 over Western China in 2012. The red line indicates the linear fit and the blue line is the 1:1 line.