

Referee #2

We are grateful to the reviewer for the time and energy in providing helpful comments and guidance that have improved the manuscript. In this document, we describe how we have addressed the reviewer's comments. Detailed responses to each comment are given below (in blue).

This manuscript showed interesting results on the temporal evolution of NO_x and NH_3 over China. By comparing the data resulting from inventories of REAS and EDGAR, the authors found that NH_3 and NO_x continually increased over China during 1980-2010. Furthermore, based on previous satellite observations and an atmospheric chemistry transport model (MOZART-4), they also found that NO_2 over China increased from 2005 to 2011 and then decreased significantly from 2011 to 2015. Finally the authors discussed the plausible reasons including control policies of Chinese government to the emission trends of reactive nitrogen. Overall the topic of the study is sound and the manuscript was written well. However, I have the following concerns to be addressed before recommending it for publication in Atmos. Chem. Phys.

Major comments:

1. In line 168 of page 8, the authors filtered the DOMINO product with an absolute error below 10^{15} molecules cm^{-2} . However the NO_2 vertical column densities (VCDs) error depend on the net values of NO_2 VCDs. Therefore the filter may arbitrarily exclude the high NO_2 VCD values. The authors should evaluate the influence of absolute errors on the final emission results and show it in current study.

We used the DOMINO NO_2 product developed by Boersma et al. (2011). The fundamental algorithm of the retrieved NO_2 columns are the residual of subtracting two large numbers (the total slant column, and stratospheric slant NO_2 column). Because high NO_2 columns with high absolute errors as well as negative (or zero) NO_2 columns are statistically meaningful, they should not be discarded, as described

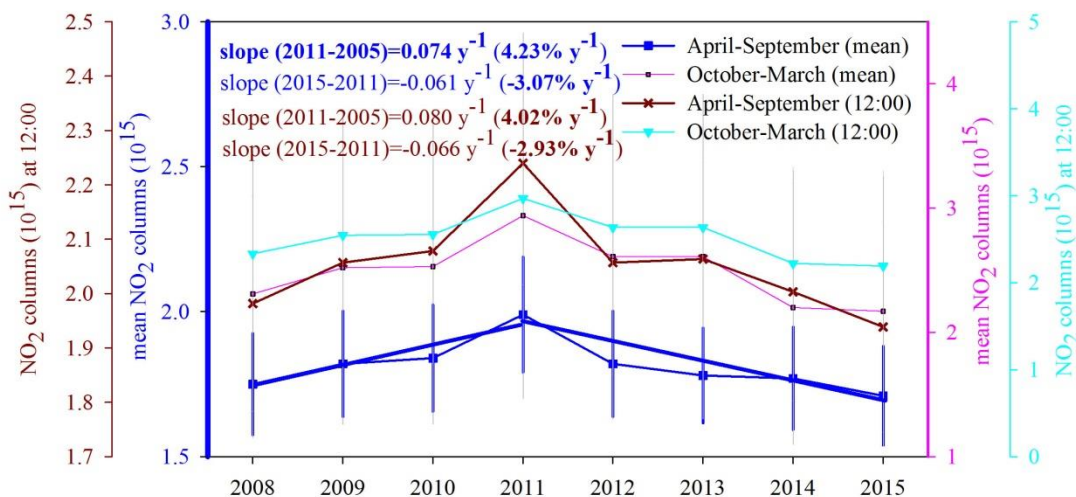
in the user manual (<http://www.temis>). In this revision, we did not filter the DOMINO product to perform the analysis.

2. The authors compared the emission data of NO₂ and NH₃ from satellite observations to that from Mozart-4 model simulations. But the authors did not explain whether the satellite overpass time has been considered during the comparison or not. The OMI satellite only gives the NO₂ data at about 1:30 pm of local time. The same time could also be used for the extraction of NO₂ data from Mozart-4 model. Whether this will influence the output results and conclusions of current study? This point should be clarified more.

Thank you very much for this good suggestion. In this revision, we have added the temporal trend analysis of NO₂ and NH₃ columns at 12:00 from MOZART to compare with that gained from satellite (OMI 1:45 P.M. local time) as shown in **Fig. 5, since the MOZART outputs vary over six hours** (00, 06, 12 and 18 h).

We gained very similar results between OMI NO₂ (13:45 P.M.) and MOZART NO₂ at 12:00 with an increase rate of 4.02% y⁻¹ vs 4.23% y⁻¹ before 2011 and a decrease rate of -2.93% y⁻¹ (OMI) vs -3.07% y⁻¹ (MOZART) between 2011 and 2015 (Fig. 5). In general, we found an agreement on the NO₂ temporal trend between MOZART (12:00) and OMI (13:45) (refer to **Paragraph 3 in Sect. 3.3**).

(a) MOZART NO₂



(b) MOZART NH₃

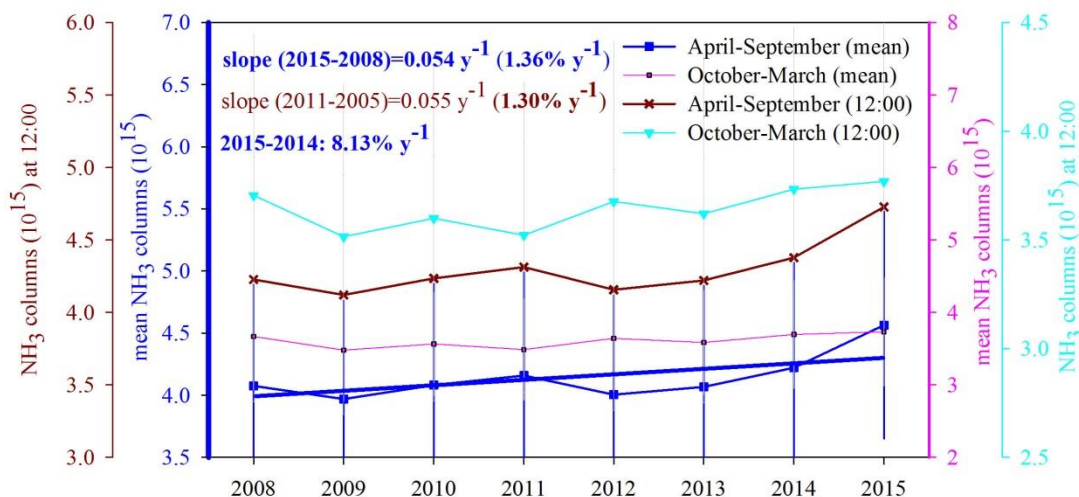


Fig. 5. Time series of MOZART NO₂ and NH₃ columns over China during average warm months (April-September) and cold months (October-March) from 2008 to 2015. The mean columns were calculated by averaging the columns at 00, 6, 12 and 18 h. The associated mean error for each period is presented here as error bars.

3. The MOZART-4 model contained 12 bulk aerosol compounds, 39 photolysis, 85 gas species as well as 157 gas-phase reactions. However, the authors did not discuss the influence of NO_x and NH₃ sink on their emission values at all while elucidating the data from MOZART-4. Although the authors have discussed the potential impacts of emission regulation or energy efficiency enhancement relevant government control policies on the NO_x and NH₃ emissions, they are encouraged to show their insight on the correlations of atmospheric process of NO_x and NH₃ with their final emission values.

Thank you very much for this good suggestion. NH₃ is the most abundant alkaline gas in the

troposphere and is important for its ability to neutralize acidic components such as sulfuric acid (H_2SO_4) and nitric acid (HNO_3) which form, respectively, by oxidation of emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x). Reactions of HNO_3 and H_2SO_4 with NH_3 generally form submicron ammonium nitrate (NH_4NO_3) and ammoniated sulfate (NH_4HSO_4 , $(\text{NH}_4)_2\text{SO}_4$, or other forms) particles. High temperatures also promote dissociation of NH_4NO_3 back to gaseous NH_3 and HNO_3 . Therefore, the temporal trends of NH_3 and NO_2 should have an interactive impact between each other.

We have discussed the potential correlations of atmospheric process of NO_x and NH_3 on the impact of the temporal trends in the following text added in **Paragraph 4 in Sect. 3.3** :

"In MOZART-4, the alkaline gaseous NH_3 and the acidic gaseous NO_2 (the precursor for HNO_3) and SO_2 are very important precursors for bulk NH_4NO_3 and $(\text{NH}_4)_2\text{SO}_4$ particles, which form the primary system of gas-particle partitioning ($\text{NH}_3\text{-NH}_4^+\text{-NO}_x\text{-NO}_3^-\text{-SO}_2\text{-SO}_4^{2-}$). The chemical shifts between particulate NH_4NO_3 and gaseous NH_3 and NO_x are correlated with the abundance of NH_3 and NO_x and meteorological factors. The decreased abundance of NO_x between 2011 and 2015 may also contribute to an increase in the NH_3 abundance in the gas stage resulting from decreased conversion to particulate NH_4NO_3 "

4. In section 3.1, the authors showed the emission data result from REAS and EDGAR, but they did not give convincing reasons for the different results of $0.24 \text{ kg N ha}^{-1} \text{ y}^{-2}$ from EDGAR and $0.17 \text{ kg N ha}^{-1} \text{ y}^{-2}$ from REAS. The authors should supply plausible explanations (e.g. induced by methodological difference of data compiling or meteorological factors etc.) to this. In addition, the authors thought $0.24 \text{ kg N ha}^{-1} \text{ y}^{-2}$ from EDGAR was much higher than $0.17 \text{ kg N ha}^{-1} \text{ y}^{-2}$ from REAS in lines 221-222 of page 11. However, they thought $0.33 \text{ kg N ha}^{-1} \text{ y}^{-2}$ was close to $0.24 \text{ kg N ha}^{-1} \text{ y}^{-2}$ in lines 231-232 of the same page. This is logically wrong. They need to correct it and also the relevant discussions.

We have added the possible reasons for the discrepancy between REAS and EDGAR as the following text in Sect. 3.1:

"The discrepancy in the magnitude of NH_3 increase rate from REAS and EDGAR ($0.24 \text{ kg N ha}^{-1} \text{ y}^{-2}$ vs $0.17 \text{ kg N ha}^{-1} \text{ y}^{-2}$) in China since 1980 may come from the different emission factors considered for estimating NH_3 emissions. The EDGAR v4.3.1 NH_3 emissions were calculated based on sectors of agriculture, shipping, waste solid and wastewater, energy for buildings, process emissions during production and application, power industry, oil refineries, transformation industry, combustion for manufacturing, road transportation, railways, pipelines and off-road transport, while the REAS v1.1 NH_3 emissions focused mostly on the agriculture source (i.e., manure management of livestock and fertilizer application) (Crippa et al., 2015; Ohara et al., 2007). Moreover, the fundamental methodology of estimating the REAS v1.1 NH_3 emissions did not consider the seasonal agricultural variations compared with that of EDGAR v4.3.1 NH_3 emissions (Kurokawa et al., 2013), and the removal efficiency (as a key element used to estimate NH_3 emissions) in REAS v1.1 was also reported to be much higher than that in EDGAR v4.3.1 (Kurokawa et al., 2013)."

In addition, we have rewritten the sentences, which were logically wrong as the reviewer pointed out, by the following text at **Paragraph 3 in Sect. 3.1**:

"A previous study (Liu et al., 2013) summarized published data on the national anthropogenic NH_3 and NO_x emissions with multi-periods in China (Wang et al., 2009; Wang et al., 1997; Streets et al., 2003; Klimont et al., 2001; Sun and Wang, 1997; Olivier et al., 1998; FRCGC, 2007), and also analyzed the temporal pattern of NH_3 emissions. Their results showed that the NH_3 emissions had increased at an annual average rate of 0.32 Tg N y^{-2} (about $0.33 \text{ kg N ha}^{-1} \text{ y}^{-2}$). The increase rate of NH_3 emissions ($0.33 \text{ kg N ha}^{-1} \text{ y}^{-2}$) by Liu et al. (2013) was double that in REAS ($0.17 \text{ kg N ha}^{-1} \text{ y}^{-2}$), implying the

NH₃ increase rate in China is still an open question, and should be further studied ".

5. In lines 311-315 of page 15, the whole daily coverage over China cannot be achieved also due to the row anomaly effect. This effect may cause half of the satellite pixels to be unusable. The discussions here should be rearranged.

Thank you very much for this good suggestion. We have added the description of row anomaly effect and rearranged this discussion by the following text:

"For daily NO₂, the spatial coverage gained by OMI were influenced by cloud radiance fractions, surface albedo, solar zenith angles, row anomaly and so on (Russell et al., 2011;De Smedt et al., 2015).

"Row anomaly" issue resulting from the OMI instrumental problem had an impact on approximately half of the rows undergoing unpredictable patterns in cross-track directions relying on latitudes and seasons and prevented obtaining convincing daily product with continuous coverage (Boersma et al., 2011;Boersma et al., 2016).".

6. Lines 99-101: the authors are encouraged to expand introduction on the method for converting satellite data to NH₃ column. Only a reference citation is not convenient for readers to follow up the work in a straight way.

We have expanded the introduction on the method of converting satellite data to NH₃ column by adding the following text:

"The retrieval algorithm of obtaining the IASI NH₃ total columns was based on the method in Whitburn et al. (2016). Two main steps were performed to derive the NH₃ columns from the satellite observations. First, deriving the spectral hyperspectral range index (HRI) based on each IASI observations (Walker et al., 2011;Van Damme et al., 2014). Second, converting HRI to NH₃ columns based on a constructed neural network with input parameters including vertical NH₃ profile, satellite

viewing angle, surface temperature and so on (Whitburn et al., 2016)".

Minor comments:

7. Line 102: the words of 'provides' and 'potential' should be changed to 'provide' and 'possibility'.

We have changed it as suggested.

8. Line 104: the description of 'emission data are also very important tools' is confusing, and there is no logic comparability with 'satellite observations' in the front dialogue, so I suggest to remove the 'tools' or modify the front dialogue properly.

We have removed the "tools".

9. Line 110: change 'resolutions' to 'resolution'.

We have changed it as suggested.

10. Line 170: change 'the manuscript' to 'previous work'.

We have changed it as suggested.

11. Line 130: change 'denotes' to 'denote'.

We have changed it as suggested.

12. Line 228-29: Similar information of the first dialogue here has been shown in lines 221-222, so there is no necessary to show it twice.

We have removed Line 228-229 to avoid repetition.

13. Line 229-230: the description of 'Liu et al. (2013) conducted that emissions of national anthropogenic NH₃ and NO_x summarized from published data during 1980-2010' is confusing and should be rearranged.

We have rewritten these sentences by the following text:

"A previous study (Liu et al., 2013) summarized published data on the national anthropogenic NH₃ and

NO_x emissions with multi-periods in China (Wang et al., 2009; Wang et al., 1997; Streets et al., 2003; Klimont et al., 2001; Sun and Wang, 1997; Olivier et al., 1998; FRCGC, 2007), and also analyzed the temporal pattern of NH₃ emissions. Their results showed that the NH₃ emissions had increased at an annual average rate of 0.32 Tg N y⁻² (about 0.33 kg N ha⁻¹ y⁻²). The increase rate of NH₃ emissions (0.33 kg N ha⁻¹ y⁻²) by Liu et al. (2013) was double that in REAS (0.17 kg N ha⁻¹ y⁻²), implying that the NH₃ increase rate in China is still an open question, and should be further studied in future work."

14. Figure 1: add error bars to panel b please

Figure 1 shows a descriptive statistic of observation numbers by year, and we do not have error bars.

Other corrections

Removed original Fig. 6.

Since the information on the increase rate (%) between 2014 and 2015 from MOZART and IASI has been added in Fig. 3 and Fig. 5 in this revision, we have removed the original Fig. 6 to avoid duplication.

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