Referee #1

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 presents an overview of the temporal characteristics of various datasets relevant to NOx and NH$_3$ abundance over China. The authors discuss trends in emission inventories (EDGAR and REAS), trends in satellite NO$_2$ and NH$_3$ columns (from OMI and IASI respectively), and trends in MOZART-4 model output for the region. Decreasing NO$_2$ since 2011 suggests that China's 12th Five Year Plan has resulted in successful emission reductions. On the other hand, the lack of a significant trend in NH$_3$ points to the growing importance of controlling and monitoring reduced nitrogen. The authors are to be commended for compiling and exploring multiple datasets in deter mining patterns in reactive nitrogen over China. However, I have some general comments about the overall scientific significance and scientific quality. I look forward to hearing from the authors in this discussion phase.

Major comments:

(1) While the analysis of IASI NH$_3$ columns focusing on China might be somewhat new, I find the analysis of OMI NO$_2$ that is presented in this manuscript lacking in novelty or insight. In particular, I would refer the authors to de Foy et al. (2016) and to Liu et al. (2016). Both of these studies use OMI NO$_2$ observations from 2005-2015 to discuss long-term trends and the 2011 peak in NO$_2$ over China in detail. In my opinion, the observations made by the authors of this present manuscript have not added new insight into this discussion (and in fact treat the analysis with less rigor, as I will discuss below). In its current state, I am concerned that this manuscript does not represent a substantial enough contribution. I encourage the authors to refer to the above references and explicitly address what new

insight is gained from their analysis.

The new insights gained from this study are for Ammonia (NH_3) as well as the potential interactive impact between NO_2 and NH_3. The temporal trend analysis of NH_3 columns over China in the present work is relatively new, and to date studies focusing on the NH_3 trends based on the IASI observations over China are still few.

Although there have been several studies regarding the temporal trends of NO_2 columns over China including Foy et al. (2016) and Liu et al. (2016), their analysis did not show the discussion on the possible interactions between NO_2 and NH_3. NH_3 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 the 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, $(NH_4)_2SO_4$, 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.

An increase in NH_3 columns in recent years may also be due to decreased NH_3 removal leading to a larger fraction remaining in a gaseous state for a long time rather than changing to the condensed phase, which can be attributed to continuous decreased acidic gases over China including the NO_2 and SO_2 under strong control policy in 12-th FYP. This can largely decrease the fraction of the chemical conversion to $(NH_4)_2SO_4$ and NH_4NO_3 in the atmosphere (Paragraph 2 in Sect. 3.2.1).

In addition, we used different methods than Foy et al. (2016) and Liu et al. (2016). We adopted the method of Russel et al. (2012) (concentrating on the US in the original paper) to quantify the change of NO_2 columns over China with focusing on the temporal analysis of warm months due to the relatively

low uncertainty compared to cold months.

Technical Comment:

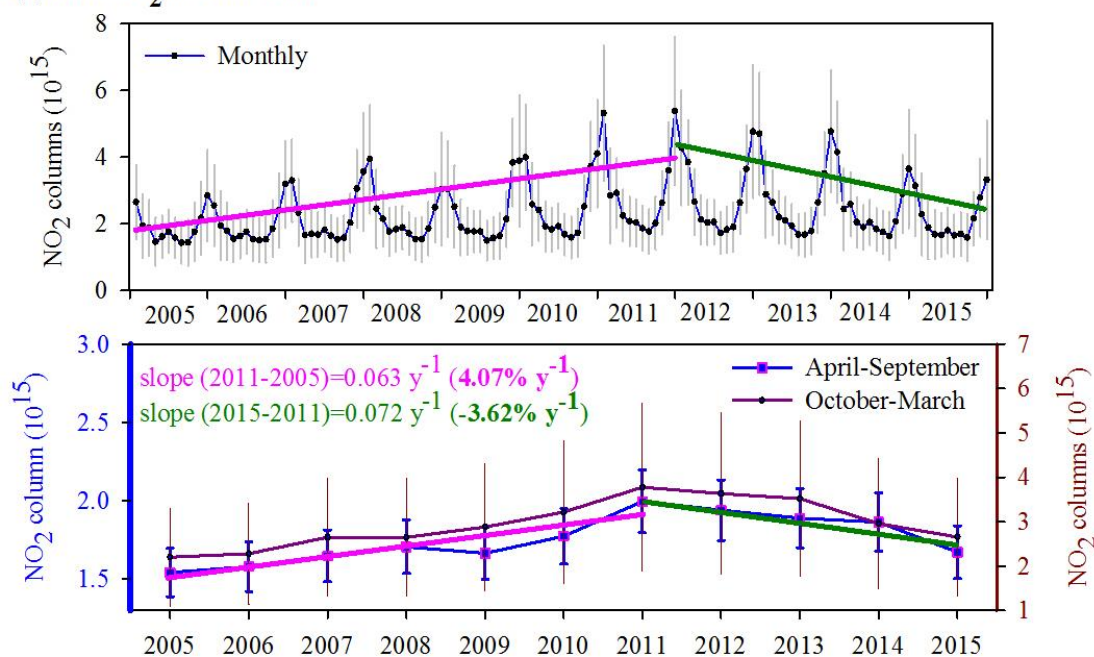(1) In determining the trend in NO_2, the authors have calculated a linear fit to the monthly average data. I find this approach to be problematic, since the trend seems to be influenced strongly by an increasing seasonal amplitude. In my opinion, the authors need to remove (or account for) the seasonality before calculating a long-term trend. Specifically, the winter monthly means seem to be driving most of the increase in their linear fit – but these values have the highest uncertainty (borne out by the larger magnitude of the error bars compared to summer months). Accounting for seasonality in determining trends in NO_2 is common practice. This can be accomplished, for example, by fitting the seasonal amplitude separately (e.g. Lamsal et al. (2015)), or by calculating trends in seasonal averages (e.g. Russel et al. (2012)).

Here, we respond to the Technical Comment before other major comments.

We agree with the reviewer that considering seasonality in determining trends of NO_2 is important. We have carefully reviewed all the given references including Foy et al. (2016) and Liu et al. (2016) in Major comments (1) as well as the given references of Lamsal et al. (2015) and Russell et al. (2012) in the Technical Comment.

We adopted the method of Russel et al. (2012) as suggested by the reviewer. In this method, averages were computed for both cold months (October-March) and warm months (April-September). We concentrated more on the temporal analysis of warm months due to the relatively low uncertainty compared to cold months. We have added related explanations and introduction text at Paragraph 1 in Sect. 3.2.1.

(a) OMI NO$_2$ at 13:45 P.M.

slope (2011-2005)=0.063 y^{-1} (**4.07% y^{-1}**)
slope (2015-2011)=0.072 y^{-1} (**-3.62% y^{-1}**)

(b) IASI NH$_3$ at 9:30 A.M.

slope (2015-2008)=0.338 y^{-1} (6.80% y^{-1})
slope (2014-2008)=0.118 y^{-1} (**2.37% y^{-1}**)
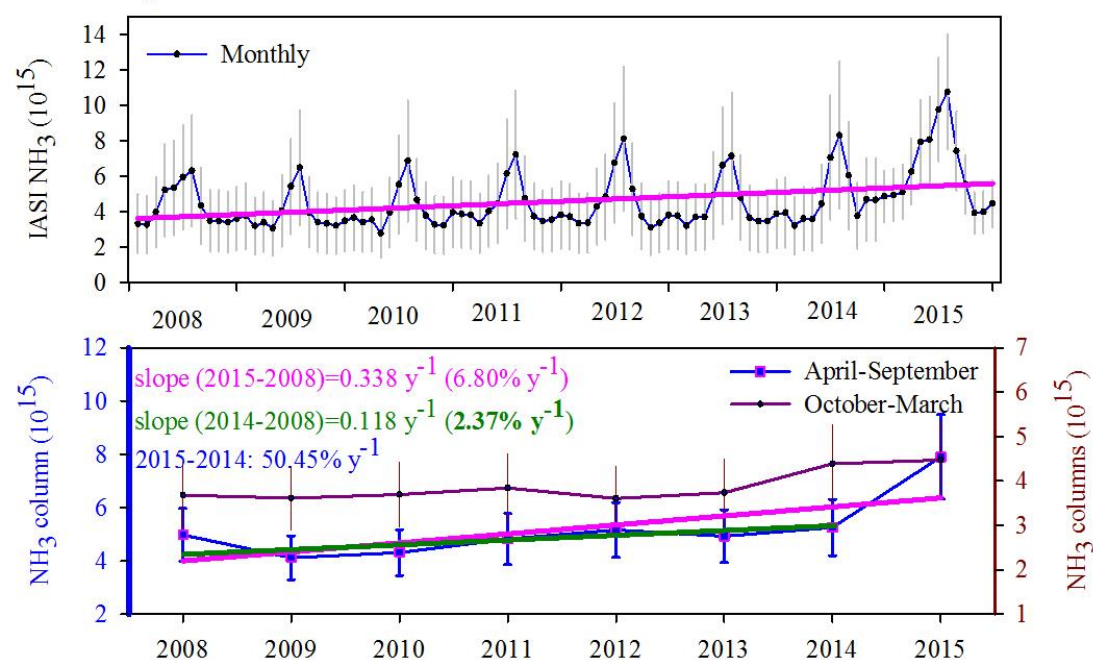2015-2014: 50.45% y^{-1}

Fig. 3. Time series of average OMI NO$_2$ and IASI NH$_3$ columns over China during warm months (April-September) and cold months (October-March). The time period of NO$_2$ columns was from 2005 to 2015, while the timespan of NH$_3$ columns was from 2008 to 2015 over China. The associated mean error for each period is presented here as error bars.

(2) The inclusion of model results has added very little insight to the analysis. The MOZART model is

driven by the EDGAR emissions to begin with (which are discussed in more detail separately). For

both NO_2 and NH_3, I would expect the relationship between emissions and tropospheric columns to be pretty strong, so it's not clear what is expected to be learned by comparing trends in EDGAR emissions with trends in model output based on EDGAR emissions. Moreover, there is no analysis or discussion of the NO_2 model output at all, so why has this output been included in the figures? The authors must expand on or address why the model output has been included, and demonstrate clearly what insight is gained.

Satellite NO_2 and NH_3 columns were observed at overpass time as an instantaneous point in a day (at 9:30 A.M. for IASI NH_3 and at 1:45 P.M. for OMI NO_2 local time). These instantaneous satellite observations may not be representative for the temporal trend analysis over China (refer to Paragraph 1 in Sect. 3.3). We calculated the monthly mean NO_2 and NH_3 columns from MOZART varying 6 hours every day (00, 06, 12, 18 h) in order to: (1) calculate the temporal trend of mean NO_2 and NH_3 columns (averaged at 00, 06, 12, 18 h) rather than the instantaneous values; (2) compare the temporal trend analysis of NO_2 from MOZART at 12 h with that gained from satellite at the overpass time (OMI 1:45 P.M. local time) as well as for NH_3.

In general, we found an agreement on the NO_2 temporal trend between MOZART (12:00) and OMI (13:45), while we found a lower increase rate from MOZART (12:00) than from IASI (9.30 A.M.). We have expanded the analysis and discussion of the NO_2 as well as NH_3 from MOZART at Paragraph 2 and 3 in Sect. 3.3. Please refer to them.
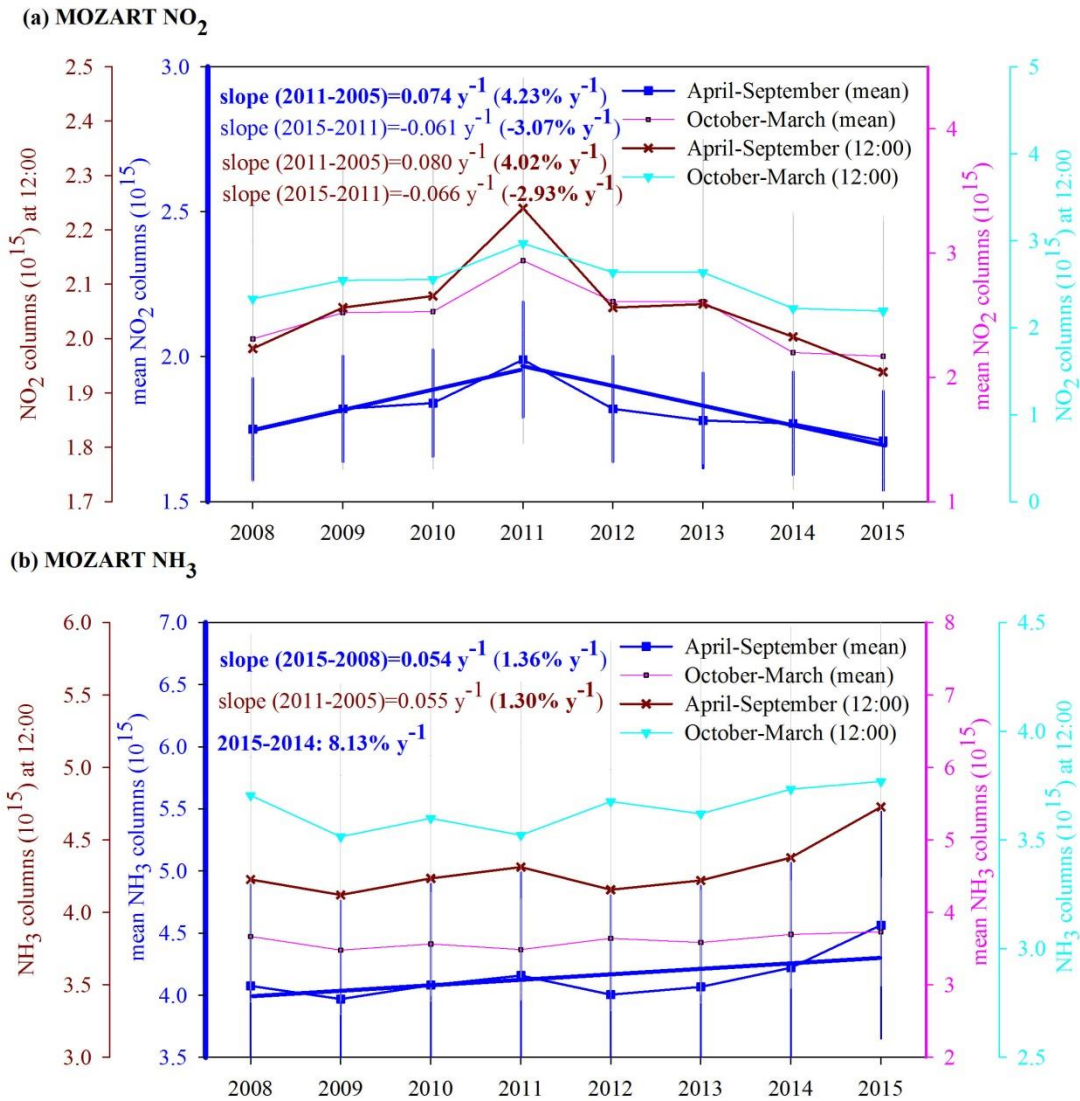
(a) MOZART NO$_2$

slope (2011-2005)=0.074 y^{-1} (4.23% y^{-1})
slope (2015-2011)=-0.061 y^{-1} (-3.07% y^{-1})
slope (2011-2005)=0.080 y^{-1} (4.02% y^{-1})
slope (2015-2011)=-0.066 y^{-1} (-2.93% y^{-1})

- April-September (mean)
- October-March (mean)
- April-September (12:00)
- October-March (12:00)

(b) MOZART NH$_3$

slope (2015-2008)=0.054 y^{-1} (1.36% y^{-1})
slope (2011-2005)=0.055 y^{-1} (1.30% y^{-1})
2015-2014: 8.13% y^{-1}

- April-September (mean)
- October-March (mean)
- April-September (12:00)
- October-March (12:00)

Fig. 5. Time series of MOZART NO$_2$ and NH$_3$ 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 bar.

(3) The authors conclude the discussion section with implications for estimating long term reactive nitrogen deposition. The discussion about uncertainty and challenges in estimating dry and wet deposition seems to be out of place in this manuscript. Of course, there is an obvious connection between emissions, atmospheric abundance, and deposition - but this manuscript does not bring up the question of deposition until this final section, so it appears as a digression. While I agree with the conclusion made by the authors (that more long-term data sets are needed), I feel their discussion has not presented any new concepts based specifically on the results presented in this manuscript. The

connection between their analysis and insight into nitrogen deposition should be made stronger throughout the manuscript. Specifically, what has been gained from the analysis?

We agree with you that the connection between the trend analysis and insight into nitrogen deposition should be made stronger. The current paper describes the temporal characteristics of atmospheric NH_3 and NO_2 over China based on multiple datasets including emission data, satellite observations and atmospheric transport modeling results since 1980. We believe the multiple datasets used in the current work have implications for estimating long-term reactive nitrogen (Nr) deposition datasets, and Sect. 3.4 describes this point and the future work will be done soon. We have changed the title from "3.4. Implications for estimating long-term Nr deposition datasets" to "3.4. Implications for estimating long-term Nr deposition datasets and recommendations for future work". To make this point more clear, we have added the following text for more clarification:

"Satellite observation provides a new perspective for estimating Nr depositions regionally. For example, to improve the modeling performance in dry gaseous NO_2 depositions from GEOS-Chem (Goddard Earth Observing System chemical transport model), Nowlan et al. (2014) applied the OMI NO_2 columns to calibrate the simulated ground NO_2 concentrations, and then estimated the deposition between 2005 and 2007. Our previous work focusing on the dry particulate NO_3^- deposition over China was also based on the OMI NO_2 columns, MOZART simulations and monitored-based sources (Liu et al., 2017b). Geddes et al. (2017) also used the satellite NO_2 columns from GOME, GOME-2 and SCIAMACHY instruments to calibrate the NO_x emissions in GEOS-Chem to estimate the NO_x depositions since 1996. The simulations combining the satellite measurements and CTM model to derive Nr depositions (Geddes and Martin, 2017;Nowlan et al., 2014) in recent years will provide relatively accurate datasets (certainly need to be validated and modified by ground measurements)."

"Despite progress in satellite techniques in recent decades (for NO_2 since 1997 by GOME and for NH_3 since 2008 by IASI), we can hardly tracked studies concerning Nr depositions before 1997 based on satellite observations. Thus, with the help of emissions data such as REAS and EDGAR, we can derive long-term Nr depositions, especially before 1997. Long-term emissions data such as REAS and EDGAR will also provide a valuable dataset to expand the modeling Nr depositions in recent years. In order to derive the Nr depositions from the emission data, the atmospheric chemistry transport models (CTMs) are frequently used through modeling the wet (simplified as the product of scavenging efficiency and precipitation amount) and dry processes (simplified as the inferential method by multiplying the deposition velocity and gaseous or particulate concentrations). However, we still lack a comprehensive dataset of gridded long-term Nr depositions including both the dry (NO_2, HNO_3, particulate NO_3^-, NH_3 and particulate NH_4^+) and wet (NH_4^+ and NO_3^- in precipitation) processes over China, which will be addressed in future work".

"Another gap is that, all the above mentioned studies focused on the NO_x depositions and did not derive the NH_y (NH_3 plus NH_4^+) depositions over China. Our recent work (Liu et al., 2017a) using IASI NH_3 columns combining the vertical profiles from MOZART benefits our understanding of the ground NH_3 concentrations over China, and the satellite-derived ground NH_3 concentrations were generally in accord with the national measurements from NNDMN. To date, there are still no reports of using the satellite NH_3 columns to derive the temporal and regional NH_y depositions over China, which dominated the total Nr depositions (NO_x plus NH_y) (Liu et al., 2016;Liu et al., 2013). The gaps of modeling NH_y depositions by applying the satellite observations combining the CTMs simulations require more efforts and further research".

We herein list some important works regarding Nr depositions using satellite, CTMs and emissions as

well as cited them in the main text:

(1) Liu, L., Zhang, X., Zhang, Y., Xu, W., Liu, X., Zhang, X., Feng, J., Chen, X., Zhang, Y., Lu, X., Wang, S., Zhang, W., and Zhao, L.: Dry Particulate Nitrate Deposition in China, Environmental Science & Technology, 10.1021/acs.est.7b00898, 2017. **Our recent work focused on the dry NO_3^- deposition based on OMI NO_2, MOZART simulations and monitor-based sources**.

(2) Liu, L., Zhang, X., Xu, W., Liu, X., Lu, X., Wang, S., Zhang, W., and Zhao, L.: Ground Ammonia Concentrations over China Derived from Satellite and Atmospheric Transport Modeling, Remote Sensing, 9, 467, 2017. **Our recent work focused on ground NH_3 concentrations based on IASI NH_3 and MOZART simulations, and we can gain dry NH_3 depositions combining the deposition velocity**.

(3) Zhang, X., Lu, X., Liu, L., Chen, D., Zhang, X., Liu, X., Zhang, Y.: Dry gaseous NO_2 deposition inferred from Ozone Monitoring Instrument NO_2 columns and atmospheric chemistry transport model over China, Journal of Geophysical Research-Atmosphere, 2017 (submitted). **Our recent work focused on the gaseous NO_2 depositions based on OMI NO_2 and MOZART simulations**.

(4) Geddes, J. A., and Martin, R. V.: Global deposition of total reactive nitrogen oxides from 1996 to 2014 constrained with satellite observations of NO2 columns, Atmos. Chem. Phys. Discuss., 2017, 1-44, 10.5194/acp-2016-1100, 2017. **Geddes's recent work focused on the total NO_x depositions globally based on the GOME, GOME-2 and SCIAMACHY NO_2 and GEOS-Chem.**

(5) Nowlan, C., Martin, R., Philip, S., Lamsal, L., Krotkov, N., Marais, E., Wang, S., and Zhang, Q.: Global dry deposition of nitrogen dioxide and sulfur dioxide inferred from space‐based measurements, Global Biogeochemical Cycles, 28, 1025-1043, 2014. **Nowlan's previous work focused on the gaseous NO_2 depositions globally based on the OMI NO_2 and GEOS-Chem.**

Specific comments:

line 88: The authors use of the term "widely" warrants more than two examples in the citation.

We added 4 new references in this line: Castellanos et al., 2015, Lamsal et al., 2015, Liu et al., 2016 and Foy et al., 2016.

line 110: "is believed to have the highest spatial resolution". Surely this statement can be confirmed instead of believed.

We have changed "is believed to have the highest spatial resolution" to "has the highest spatial resolution".

line 117: I suggest the authors replace the expression "multivariate", since this term usually implies something different (i.e. modeling). May I suggest the authors use "multiple datasets" throughout the manuscript, instead of "multivariate".

We have changed "multivariate data" to "multiple datasets" throughout the manuscript.

lines 151-153: Repeating the thresholds for error consideration is redundant here.

We have removed the repetition in these lines.

line 202: Please also include the spatial resolution of the model simulation.

We have added it as suggested.

line 223: "their thread values both positive". Please clarify this sentence.

We have changed "their thread values both positive" to "their thread values of 0.24 kg N ha^{-1} y^{-2} (EDGAR) vs 0.17 kg N ha^{-1} y^{-2} (REAS) both reflected a continuous increasing trend (in this regard they are consistent)".

line 232-233: I think the closer agreement with one other estimate does not necessarily mean the EDGAR estimate is "more reasonable". Please qualify.

The original discussion in line 232-233 was logically wrong, and we are now aware of that. Reviewer 2 also commented, "the authors thought 0.24 kg N ha^{-1} y^{-2} from EDGAR was much higher than 0.17 kg N ha^{-1} y^{-2} from REAS in lines 221-222 of page 11. However, they thought 0.33 kg N ha^{-1} y^{-2} was close to 0.24 kg N ha^{-1} y^{-2} in lines 231-232 of the same page. This is logically wrong. They need to correct it and also the relevant discussions."

In this revision, we have rewritten the sentences as the following text in the third paragraph 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 kg N ha^{-1} y^{-2}). The increase rate of NH$_3$ 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 NH$_3$ increase rate in China is still an open question, and should be further studied in future work.".

line 255 (and elsewhere): The use of the expression "no big changes" does not have much scientific meaning. May I suggest "no significant changes" followed by the results of some statistical test?

We have changed "no big changes" to "0.118×10^{15} molec. cm^{-2} y^{-1} (2.37% y^{-1}) in warm months".

line 256: The slope in NH3 of 0.025 x 10^15 is actually twice the slope of NO2 (0.011 x 10^15), so can the authors clarify why the slope in NH3 is not determined to be important or large? Should they clarify that they are speaking in relative terms to the atmospheric concentrations? What are the trends in %/year for NO2 compared to NH3?

Yes, we refer to the percent increase rate rather than the absolute increase rate. We have added the

following text for explanations as well as the percent increase rate (% y^{-1}) by the following text:

"The percent increase rate for NH_3 by year (2.37% y^{-1}) from 2008 to 2014 is lower than that for NO_2 (4.07% y^{-1}) from 2005 to 2011, although the absolute NH_3 increase rate of 0.118×10^{15} molec. cm^{-2} y^{-1} from 2008 to 2014 was higher than that of 0.063×10^{15} molec. cm^{-2} y^{-1} for NO_2 from 2005 to 2011.".

line 305-306: Can the author confirm these numbers are coming from the reference in the preceding sentence (Wang et al. 2012)?

No, these numbers come from the reference (Xia et al., 2016), and we have added the reference.

line 311: Can the authors explain why it would be better to calculate trends based on daily data? This would be unusual.

This sentence has been rewritten and clarified by the following text:

"It is difficult to gain whole coverage based on the daily data over China for both IASI NH_3 and OMI NO_2. For daily NO_2, 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).".

line 350, 351, and 353: Are the authors referring to the panels in Figure 6, not Figure 5?

Yes, we have changed it.

line 359: "...this is the conclusion we really concerned." Please clarify this sentence.

We referred to the sentence "At the current state, we can, at least, draw a conclusion that the NH_3 columns over China indeed increased in 2015 both from IASI and MOZART, but a debate or

inconsistency exists on the increase rate of the NH_3 columns in 2015". We have marked this sentence in red and removed "this is the conclusion we really concerned".

line 360: "... the following discussion in this paragraph was all hypothetical". Are the authors referring to the next two sentences? This isn't much of a discussion.

Yes, we refer to the sentence: "For IASI NH_3 columns, the sharp increase in 2015 over China may be an artifact, which may be due to an update of the input data."

line 373: "in high level". I suggest replacing this expression with something more clear.

We have changed "in high level" to "in high level with an average of 1.87 molec. cm^{-2} y^{-1} compared with that (1.65 molec. cm^{-2} y^{-1}) during 2005-2010".

line 401: "no big variations". Again, I suggest replacing this statement with something more scientifically/statistically clear.

We have changed "no big variations" to "the percent increase rate of 2.37% y^{-1}".

References:

de Foy et al. (2016), Scientific Reports, http://dx.doi.org/10.1038/srep35912

Liu et al. (2016), Environmental Research Letters, http://dx.doi.org/10.1088/1748-9326/11/11/114002

Lamsal et al. (2015), Atmospheric Environment, http://dx.doi.org/10.1016/j.atmosenv.2015.03.055

Russell et al. (2012), Atmospheric Chemistry and Physics, http://dx.doi.org/10.5194/acp-12-12197-2012

We have reviewed and added all the suggested references.

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 original Fig. 6 to avoid duplication.

References

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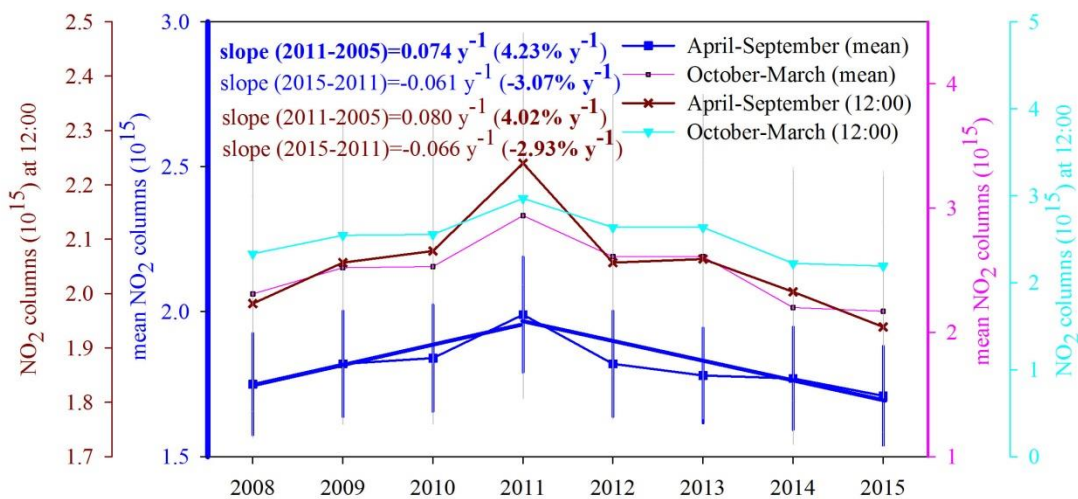
in the user manual (http://www.temis). In this revision, we did not filter the DOMINO product to perform the analysis.

2. The authors compared the emission data of NO_2 and NH_3 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 NO_2 data at about 1:30 pm of local time. The same time could also be used for the extraction of NO_2 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 NO_2 and NH_3 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 NO_2 (13:45 P.M.) and MOZART NO_2 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 NO_2 temporal trend between MOZART (12:00) and OMI (13:45) (refer to **Paragraph 3 in Sect. 3.3**).
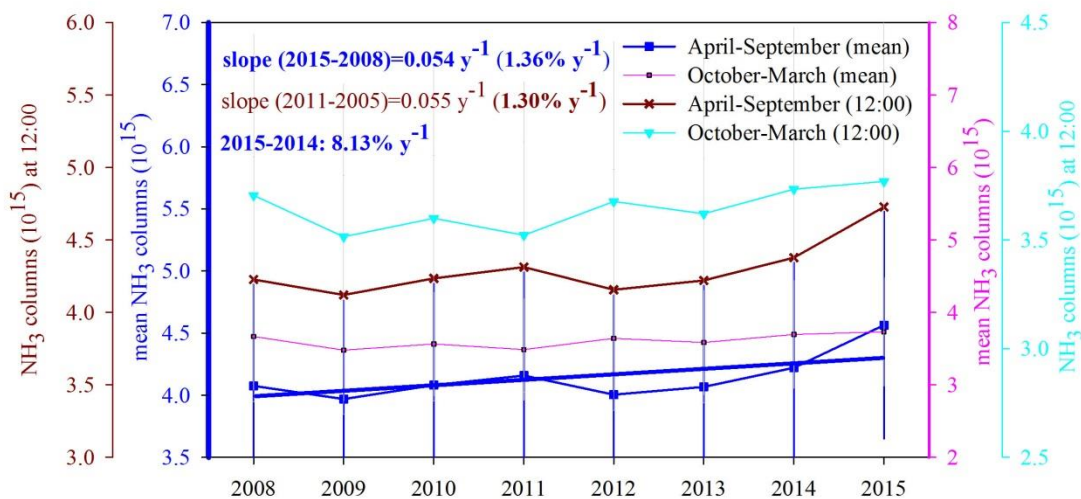
Fig. 5. Time series of MOZART NO$_2$ and NH$_3$ columns over China during average warm months (April-September) and cold months (October-March) from 2008 to 2015. The mean columns were calculated by averaging the columns at 00, 6, 12 and 18 h. The associated mean error for each period is presented here 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 NH$_3$ 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 NH$_3$ emissions, they are encouraged to show their insight on the correlations of atmospheric process of NO$_x$ and NH$_3$ with their final emission values.

Thank you very much for this good suggestion. NH$_3$ 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, $(NH_4)_2SO_4$, 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 $(NH_4)_2SO_4$ particles, which form the primary system of gas-particle partitioning (NH_3-NH_4^+-NO_x-NO_3^--SO_2-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 kg N ha^{-1} y^{-2} from EDGAR and 0.17 kg N ha^{-1} 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 kg N ha^{-1} y^{-2} from EDGAR was much higher than 0.17 kg N ha^{-1} y^{-2} from REAS in lines 221-222 of page 11. However, they thought 0.33 kg N ha^{-1} y^{-2} was close to 0.24 kg N ha^{-1} 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 kg N ha^{-1} y^{-2} vs 0.17 kg N ha^{-1} 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 kg N ha^{-1} y^{-2}). The increase rate of NH_3 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 the

NH$_3$ 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 NO$_2$, 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 NH$_3$ 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 NH$_3$ column by adding the following text:

"The retrieval algorithm of obtaining the IASI NH$_3$ total columns was based on the method in Whitburn et al. (2016). Two main steps were performed to derive the NH$_3$ 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 NH$_3$ columns based on a constructed neural network with input parameters including vertical NH$_3$ profile, satellite

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 NOx 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 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 kg N ha^{-1} y^{-2}). The increase rate of NH$_3$ 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 NH$_3$ 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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Temporal characteristics of atmospheric ammonia and nitrogen dioxide over China based on emission data, satellite observations and atmospheric transport modeling since 1980

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Abstract

China is experiencing intense air pollution caused in large part by anthropogenic emissions of reactive nitrogen (Nr). Atmospheric ammonia (NH_3) and nitrogen dioxide (NO_2) are the most important precursors for Nr compounds (including N_2O_5, HNO_3, HONO and particulate NO_3^- and NH_4^+) in the atmosphere. Understanding the changes of NH_3 and NO_2 has important implications for the regulation of anthropogenic Nr emissions, and is a requirement for assessing the consequence of environmental impacts. We conducted the temporal trend analysis of atmospheric NH_3 and NO_2 on a national scale

since 1980 based on emission data (during 1980-2010), satellite observations (for NH_3 since 2008 and for NO_2 since 2005) and atmospheric chemistry transport modeling (during 2008-2015).

Based on the emission data, during 1980-2010, both significant continuous increasing trend of NH_3 and NO_x were observed from REAS (Regional Emission inventory in Asia, for NH_3 0.17 kg N ha^{-1} y^{-2} and for NO_x 0.16 kg N ha^{-1} y^{-2}) and EDGAR (Emissions Database for Global Atmospheric Research, for NH_3 0.24 kg N ha^{-1} y^{-2} and for NO_x 0.17 kg N ha^{-1} y^{-2}) over China. Based on the satellite data and atmospheric chemistry transport modeling named as the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), the NO_2 columns over China increased significantly from 2005 to 2011 and then decreased significantly from 2011 to 2015; the satellite-retrieved NH_3 columns from 2008 to 2014 increased at a rate of 2.37% y^{-1}. The decrease in NO_2 columns since 2011 may result from more stringent strategies taken to control NO_x emissions during the 12th Five-Year-Plan, while no control policy focused on NH_3 emissions. Our findings provided an overall insight on the temporal trends of both NO_2 and NH_3 since 1980 based on emission data, satellite observations and atmospheric transport modeling. These findings can provide a scientific background for policy-makers that are attempting to control atmospheric pollution in China. Moreover, the multiple datasets used in this study have implications for estimating long-term Nr deposition datasets to assess its impact on soil, forest, water and greenhouse balance.

Keywords: trends, seasonal cycle, ammonia

1. Introduction

Reactive nitrogen (Nr) emissions have increased significantly in China due to anthropogenic activities such as increased combustion of fossil fuels, over-fertilization and high stocking rates of farm animals (Canfield et al., 2010;Galloway et al., 2008;Liu et al., 2013). Elevated Nr in the environment has led to

a series of effects on climate change and ecosystems, e.g. biodiversity loss, stratospheric ozone depletion, air pollution, freshwater eutrophication, the potential alteration of global temperature, drinking water contamination, dead zones in coastal ecosystems and grassland seed bank depletion (Basto et al., 2015;Lan et al., 2015;Shi et al., 2015). Atmospheric reactive N emissions are dominated by nitrogen oxides ($NO_x = NO + NO_2$) and ammonia (NH_3) (Li et al., 2016a;Galloway et al., 2004). Atmospheric NO_2 and NH_3 are the most important precursors for Nr compounds including N_2O_5, HNO_3, HONO and particulate NO_3^- and NH_4^+ in the atmosphere (Xu et al., 2015;Pan et al., 2012). Therefore, an understanding of both the spatial and temporal patterns of NO_2 and NH_3 is essential for evaluating N-enriched environmental effects, and can provide the scientific background for N pollution mitigation.

To investigate the spatial and temporal variations of atmospheric NO_2 and NH_3, ground measurements are acknowledged to be an effective way in monitoring the accurate concentrations of NO_2 and NH_3 (Xu et al., 2015;Pan et al., 2012;Meng et al., 2010). Ground measurements of NO_2 concentrations in China, including about 500 stations in 74 cities, have been monitored and reported to the public since January 2013 (Xie et al., 2015). By the end of 2013, this network was extended with hourly NO_2 concentrations from more than 850 stations in 161 cities. However, there are fewer NH_3 measurements across China than NO_2 measurements. The China Agricultural University has organized a Nationwide Nitrogen Deposition Monitoring Network (NNDMN) since 2010, consisting of 43 monitoring sites covering urban, rural (cropland) and background (coastal, forest and grassland) areas across China (Xu et al., 2015;Liu et al., 2011). Xu et al. (2015) reported the ground NH_3 concentrations throughout China for the first time, providing great potential to understand the ground NH_3 concentrations on a national scale. Other networks include (1) the Chinese Ecosystem Research Network (CERN) which was

established in 1988, including 40 field stations (Fu et al., 2010). However, to our knowledge, there are

no detailed reports about ground NH_3 concentrations from CERN on a national scale. (2) Four Chinese

cities (Xiamen, Xi-An, Chongqing and Zhuhai) have joined the Acid Deposition Monitoring Network

in East Asia (EANET) since 1999. However, only one site (Hongwen, Xiamen) in EANET measured

the ground NH_3 concentrations and that data is not continuous. Finally, ground NH_3 concentrations at

ten sites in Northern China from 2007 to 2010 have been reported by Pan et al. (2013). All of the above

ground measurements provide the potential to understand NH_3 and NO_2 concentrations on a regional

scale. However, there is limited information on the spatial and temporal variations of NH_3 and NO_2 in

the atmosphere across China. This is due to the limited observation sites and monitoring period, as well

as given the uneven distribution of the monitoring sites. Importantly, atmospheric NH_3 and NO_2

monitoring based on ground-based local sites may have limited spatial representativeness of the

regional scale as both NH_3 and NO_2 are highly variable in time and space (Clarisse et al., 2009;Wichink

Kruit et al., 2012;Boersma et al., 2007).

In order to complement ground-based measurements, satellite observation of NH_3 and NO_2 is a

welcome addition for analyzing the recent trends of NH_3 and NO_2 in the atmosphere. Satellite remote

sensing offers an opportunity to monitor atmospheric NH_3 and NO_2 with high temporal and spatial

resolutions (Warner et al., 2017;Li et al., 2016b). NO_2 was measured by multiple space-based

instruments including the Global Ozone Monitoring Experiment (GOME), SCanning Imaging

Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY), Ozone Monitoring

Instrument (OMI) and Global Ozone Monitoring Experiment-2 (GOME-2). The OMI NO_2 provides the

best horizontal resolution (13×24 km^2) among instruments in its class and near-global daily coverage

(Levelt et al., 2007). OMI observations have been widely applied in environmental-related studies and

for the support of emission control policy (Russell et al., 2012;Zhao and Wang, 2009;Castellanos et al., 2015;Lamsal et al., 2015;Liu et al., 2016a;Foy et al., 2016). First measurements of NH_3 from space were reported over Beijing and San Diego areas with the Tropospheric Emission Spectrometer (TES) (Beer et al., 2008) and in fire plumes in Greece with the Infrared Atmospheric Sounding Interferometer (IASI) (Coheur et al., 2009). The first global map of NH_3 was created from IASI measurements by correlating the observed brightness temperature differences to NH_3 columns using the averaged datasets in 2008 (Clarisse et al., 2009). Shortly after that, many studies focused on developing techniques to gain more reliable NH_3 columns (Whitburn et al., 2016a;Van Damme et al., 2014b), validating the retrieved NH_3 columns using the ground measurements (Van Damme et al., 2014a;Dammers et al., 2016) and comparing the data with the results of the atmospheric chemistry transport models (Van Damme et al., 2014c;Whitburn et al., 2016a), and the estimated NH_3 columns obtained from Fourier transform infrared spectroscopy (FTIR) (Dammers et al., 2016). The retrieval algorithm of obtaining IASI NH_3 columns was based on the method described in Whitburn et al. (2016). Two main steps were performed to derive the NH_3 columns from the satellite measurements. First, derive the spectral hyperspectral range index (HRI) based on each IASI observations (Walker et al., 2011;Van Damme et al., 2014b). Second, convert HRI to NH_3 columns based on a constructed neural network with input parameters including vertical NH_3 profile, satellite viewing angel, surface temperature and so on (Whitburn et al., 2016a). The progresses made on the satellite techniques provide possibility for understanding both the spatial and temporal variations of NH_3 and NO_2 in the atmosphere.

In addition to satellite observations, the emission data are also very important for investigating the temporal trends of NH_3 and NO_2 such as the IIASA inventory (Cofala et al., 2007), EDGAR (Emission

Database for Global Atmospheric Research, RAINS-Asia (Regional Air Pollution Information and Simulation) and Asia REAS (Regional Emission inventory in Asia). REAS is considered as the first inventory by integrating historical, current and future emissions data for Asia based on a consistent methodology (Ohara et al., 2007), and EDGAR is the global emission data with 0.1 by 0.1 grid, which has the highest spatial resolution among different datasets mentioned above. Thus, REAS and EDGAR are used to analyze the historical trends of NH_3 and NO_2 during 1980-2010 in this study. Based on the EDGAR emission data, a widely used atmospheric transport model named as the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4) was also used to model the temporal trend of NH_3 and NO_2 columns during 2008-2015 in comparison with the temporal trends of NH_3 and NO_2 columns measured by satellite instruments.

We aim at getting an overall insight on the temporal trends of both NO_2 and NH_3 since 1980 based on the multiple datasets including the emission data, satellite observations and atmospheric transport modeling. We herein show the Chinese national trend of REAS and EDGAR NH_3 and NO_x emission data during 1980-2010, satellite-retrieved NH_3 during 2008-2015 and NO_2 columns (2005-2015), and atmospheric transport chemistry modeling NH_3 and NO_2 columns (2008-2015). It should be noted here that the satellite NH_3 columns were retrieved from the IASI, and can only be obtained since 2008. It is beneficial to analyze the temporal variations of both NH_3 and NO_2, hence providing a scientific basis for policy makers to reduce N-enriched environmental pollution in China.

2. Materials and methods

2.1. NH_3 and NO_2 Emissions

We examined the emission inventory dataset for Asia REAS (Regional Emission inventory in Asia) with $0.5\,° \times 0.5\,°$ resolution for the period 1980-2010, and analyzed the temporal trends of NO_x and NH_3

over China. REAS v1.1 is believed to be the first inventory of integrating past, present and future dataset in Asia based on a consistent methodology. The REAS datasets have been validated by several emissions, and denote agreement with the recent growth status in Chinese emissions (Ohara et al., 2007). We also collected NO_x and NH_3 emission data from EDGAR (Emissions Database for Global Atmospheric Research) v4.3.1, which was developed by the Netherlands Environmental Assessment Agency and European Commission Joint Research Centre (Jgj et al., 2002). The EDGAR emissions are calculated on the basis of a point emissions inventory conducted by the International Energy Agency. EDGAR also has a long time period 1980-2010 with the highest spatial resolution globally (0.1 °×0.1 °) (http://edgar.jrc.ec.europa.eu/overview.php?v=431).

2.2. Satellite observations

IASI is a passive remote-sensing instrument operating in nadir mode and measures the infrared radiation emitted by the Earth's surface and the atmosphere (Clarisse et al., 2009). It covers the entire globe twice a day, crossing the equator at a mean solar local time of 9:30 A.M. and P.M. and has an elliptical footprint of 12 by 12 km up to 20 by 39 km depending on the satellite-viewing angle. In this study we use daytime satellite observations as these are more sensitive to NH_3 and are associated with a large positive thermal contrast and a significant amount of NH_3 (Van Damme et al., 2014b;Whitburn et al., 2016a). The availability of measurements is mainly driven by the cloud coverage as only observations with cloud coverage lower than 25% are processed to be a good compromise between the number of data kept for the analysis and the bias due to the effect of clouds. As the amount of daily data is not always sufficient to obtain meaningful distributions (due to cloud cover or the availability of the temperature profiles from the EUMETSAT operational processing chain) (Van Damme et al., 2014b), it is more appropriate to consider monthly or yearly averages for this trend analysis. We

consider IASI observations with a relative error below 100% or an absolute error below 5×10^{15} molec. cm^{-2} for analysis over China. For the error, the filtering depends on the use of the data. Doing this, low columns typical for background conditions with a large relative error but a small absolute error are also taken into account. For other applications, such as comparing with ground measurements, we would recommend to use a threshold of 75% or even 100% relative error. We gained the data upon request from the Atmospheric Spectroscopy Group at Université Libre De Bruxelles (http://www.ulb.ac.be/cpm/atmosphere.html). This data can be gridded on 0.1 °latitude×0.1 °longitude (Dammers et al., 2016), 0.25 °latitude×0.25 °longitude (Whitburn et al., 2016a) and 0.5 °latitude×0.5 °longitude (Whitburn et al., 2016b) or even coarser resolutions depending on the usage of the data. For IASI NH_3, we firstly divided China into 0.5 ° latitude×0.5 ° longitude grid. For each grid cell, we calculated the monthly arithmetic mean by averaging the daily values with observations points within the grid cell. Similarly, we calculated the annual arithmetic mean by averaging the daily values with observations points within the grid cell over the whole year.

The NO_2 columns are obtained from the OMI instrument on NASA's EOS Aura satellite globally everyday. We used the generated products by the project "Derivation of Ozone Monitoring Instrument tropospheric NO_2 in near-real time" (DOMINO) to analyze the temporal trends of NO_2 columns over China. In DOMINO products, only the observations with a cloud radiance fraction below 0.5 were processed for analysis. The retrieval algorithm is described in detail in the previous work (Boersma et al., 2007) and recent updates can be found in the DOMINO Product Specification Document (http://www.temis.nl/docs/OMI_NO2_HE5_1.0.2.pdf). We used tropospheric NO_2 retrievals from the DOMINO algorithm v2.0. The retrieval quality of NO_2 products is strongly dependent on different aspects of air mass factors, such as radiative transfer calculations, terrain heights and surface albedo.

The OMI v2.0 data were mainly improved by more realistic atmospheric profile parameters, and include more surface albedo and surface pressure reference points than before (Boersma et al., 2011;Boersma et al., 2016). The DOMINO NO$_2$ datasets are available from http://www.temis.nl/airpollution/no2.html. We should state in particular that we used directly the DOMINO v2.0 products of monthly means from 2005 to 2015 over China for the trend analysis. The DOMINO NO$_2$ columns were gridded at a resolution of 0.125 °latitude×0.125 °longitude grid globally, which has been widely used for scientific applications (Ma et al., 2013;Ialongo et al., 2016;Castellanos et al., 2015).

To illustrate measurement availability, we presented here some measurement statistics. A total number of cloud-free daytime observations as characterized by the operational IASI processor by year were retrieved in China during 2008-2015 for NH$_3$ (Fig. 1b). We retrieved more observation numbers after 2010 than those during 2008-2009. In 2010, the update of the improved air temperature profiles, cloud properties products and cloud detection, which are important for calculating the thermal contrast, increased the quality of retrieval (Van Damme et al., 2014b;Van Damme et al., 2014c). In September 2014, there was another update of the air temperature profiles, cloud properties products and cloud detection for calculating the thermal contrast. For the updates of the IASI-NH$_3$ data, you can refer to Van Damme et al. (2014b), Van Damme et al. (2014c) and Whitburn et al. (2016). The monthly observation numbers are also presented in Fig. 1a, showing that spring (Mar, Apr and May), summer (Jun, Jul and Aug), autumn (Sep, Oct and Nov) and winter (Dec, Jan and Feb) months represent 29% , 26%, 23% and 21%, respectively. Compared with large variations of observation numbers for NH$_3$, the observation numbers for NO$_2$ varied less by year; winter season had the least, while other seasons varied little.

2.3. Atmospheric transport chemistry model

Atmospheric transport chemistry model is also of central importance in modeling the tropospheric NO_2 and NH_3. We applied a widely used atmospheric global atmospheric transport chemistry model named as the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4) to simulate the tropospheric NO_2 and NH_3 columns during 2008-2015 in accordance with the time period of IASI NH_3 measurements.

The MOZART-4 model is driven by the meteorological data from the NASA Goddard Earth Observing System Model, Version 5 (GEOS-5) at a resolution of $1.9°$ latitude $\times 2.5°$ longitude spatially. The emission data applied for driving the simulations are based on the updated EDGAR emission inventories. 12 bulk aerosol compounds, 39 photolysis, 85 gas species as well as 157 gas-phase reactions were integrated in MOZART-4. The chemical mechanism on N compounds including the NO_2, NH_3 and aerosols are detailedly integrated to MOZART-4, which is considered to be suitable for tropospheric chemical compositions (Emmons et al., 2010;Pfister et al., 2008;Sahu et al., 2013). The output data used in the current work are temporally varying six hours every day, which were upon request by Louisa Emmons at National Center for Atmospheric Research (NCAR). The monthly means of NO_2 and NH_3 columns were averaged by the daily data, and then used for the trend analysis over China. For more details about MOZART-4, the reader should refer to previous studies (Emmons et al., 2010;Brasseur et al., 1998;Beig and Singh, 2007).

3. Results and discussions

3.1. NH_3 and NO_2 emissions during 1980-2010

We conducted the temporal analysis of NH_3 and NO_x emissions since 1980 based on REAS and EDGAR. Both significant continuous increasing trends of NH_3 and NO_x were observed from REAS

(for NH_3 0.17 kg N ha^{-1} y^{-2} and for NO_x 0.16 kg N ha^{-1} y^{-2}) and EDGAR (for NH_3 0.24 kg N ha^{-1} y^{-2} and for NO_x 0.17 kg N ha^{-1} y^{-2}) over China (Fig. 2). We found a relatively consistent increase in NO_x emission from EDGAR and REAS over China, i.e. 0.17 kg N ha^{-1} y^{-2} vs 0.16 kg N ha^{-1} y^{-2}, but inconsistency in the magnitude of NH_3 emissions from EDGAR and REAS over China, i.e. 0.24 kg N ha^{-1} y^{-2} vs 0.17 kg N ha^{-1} y^{-2}. The increase rate in NH_3 emissions over China from EDGAR was much higher than that from REAS, indicating the magnitude of increase trend in NH_3 over China remains a debate, although their thread values of 0.24 kg N ha^{-1} y^{-2} (EDGAR) vs 0.17 kg N ha^{-1} y^{-2} (REAS) both reflected a continuous increasing trend (in this regard they are consistent). It implies that, at least, the NH_3 emissions are indeed increasing during 1980-2010. We also conducted a simple correlation analysis of the NH_3 (Fig. 2a) and NO_x (Fig. 2b) from REAS and EDGAR, showing agreement in the magnitude (slope=1.06) and temporal trend (R^2=0.96) for NO_x, but some inconsistency in the increase rate (slope=1.33) for NH_3.

The discrepancy in the magnitude of NH_3 increase rate from REAS and EDGAR (0.24 kg N ha^{-1} y^{-2} vs 0.17 kg N ha^{-1} y^{-2}) in China since 1980 may be caused by the different emission factors considered for estimating NH_3 emissions. The EDGAR v4.3.1 NH_3 emissions were calculated based on a variety of sectors including 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 mainly 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 on 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 to estimate NH_3 emissions) was also reported to be much higher in REAS v1.1 than in EDGAR v4.3.1 (Kurokawa et al., 2013).

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 kg N ha^{-1} y^{-2}). The increase rate of NH_3 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 NH_3 increase rate in China is still an open question, and should be further studied.

3.2. Satellite NH_3 and NO_2 over China in the recent decade

3.2.1. Temporal trends

We referred to the method of a previous study (Russell et al., 2012) to conduct the temporal trend analysis by calculating the average values during cold months (October-March) and warm months (April-September) respectively. We herein concentrated more on the temporal analysis of satellite observations during warm months because of the relatively lower uncertainty in comparison with that during cold months. Fig. 3 shows the temporal trend of NO_2 columns during warm and cold months between 2005 and 2015 as well as monthly average values. From satellite observations, the NO_2 columns over China increased with a slope of 0.063×10^{15} molec. cm^{-2} y^{-1} (4.07% y^{-1}) in warm months from 2005 to 2011 and then decreased with a slope of -0.072 molec. cm^{-2} in warm months (-3.62% y^{-1}) from 2011 to 2015 (Fig. 3). The decreasing trends were consistent with NO_x emissions since 2011 over China (decreasing from 24.04×10^6 ton in 2011 to 20.78×10^6 ton in 2014, China Statistical Yearbook, http://www.stats.gov.cn/). During the Chinese 11th Five-Year-Plan (FYP) period (2006-2010), Chinese

government undertook a series of strategies to increase energy efficiency and to reduce NO_x emissions, but NO_x emissions were not successfully restrained, which created a big challenge for improving air quality over the country (Xia et al., 2016). During the 12th FYP period (2011-2015), more stringent strategies were implemented to control NO_x emissions, including the application of selective catalytic/non-catalytic reduction (SCR/SNCR) systems in the power sector, staged implementation of tighter vehicle emission standards and a series of standards with aggressive emission limits for power, cement, and the iron and steel industries. These strategies are believed to have helped achieve national targets of NO_x emission abatement (Xia et al., 2016).

However, the satellite-retrieved NH_3 columns increased with a slope of 0.118×10^{15} molec. cm^{-2} y^{-1} (2.37% y^{-1}) in warm months from 2008 to 2014 (Fig. 3), but increase largely in 2015 (this will be discussed in Sect. 3.3 in comparison with MOZART-4 simulations in detail). The percent increase rate for NH_3 by year (2.37% y^{-1}) from 2008 to 2014 is lower than that for NO_2 (4.07% y^{-1}) from 2005 to 2011, although the absolute NH_3 increase rate of 0.118×10^{15} molec. cm^{-2} y^{-1} from 2008 to 2014 was higher than absolute NO_2 increase rate of 0.063×10^{15} molec. cm^{-2} y^{-1} from 2005 to 2011. An increase in NH_3 columns from IASI may be due to decreased NH_3 removal leading to a larger fraction maintaining in gaseous state for a long time rather than changing to the condensed phase. Specifically, NH_3 is considered as an important alkaline gas that is abundant in the atmosphere, and is able to neutralize acidic components including HNO_3 and H_2SO_4 through the oxidation of NO_x and SO_2, respectively (Li et al., 2014;Liu et al., 2011;Liu et al., 2017c;Xu et al., 2015). The decreased NH_3 removal to some degree can be attributed to continuous decreased acidic gases including the NO_2 and SO_2 over China under strong control policy in 12-th FYP, which can largely decrease the fraction of the chemical conversion to $(NH_4)_2SO_4$ and NH_4NO_3 in the atmosphere. Increasing trend in NH_3 columns may be

associated with continuous N fertilizer use for guaranteeing increase of crop productions (Erisman et al., 2008). Although there was no strong NH_3 emission control regulation, N fertilizer efficiency should be further improved over China. In 2015, the Ministry of Agriculture formally announced a "Zero Increase Action Plan" for national fertilizer use by 2020, which requires the annual increase in total fertilizer use will be less than 1% from 2015 to 2019, with no further increment from 2020 (Liu et al., 2015).

If the "Zero Increase Action Plan" for N fertilizer can be effective, future NH_3 emissions should be consistent with the current NH_3 emissions. In addition, due to strong emission control of NO_x, the NO_x emissions were believed to decrease significantly from 2011 to 2015. We can reasonably make two major conclusions. First, the atmospheric NO_2, as a key indicator of oxidized N compounds (NO_2, HNO_3 and NO_3^-), decreased since 2011, and will continue to decrease under the current policy. Second, the atmospheric NH_3, as a key indicator of reduced N (NH_3 and particulate NH_4^+), will slightly increase or stay at the current level in the future with the "Zero Increase Action Plan". Thus, due to a decreasing trend of oxidized N (NO_x-N), ammonia N (NH_x-N) should still dominate Nr deposition (oxidized N plus reduced N) in China, and is expected to play a more significant role in Nr deposition. Therefore, monitoring the reduced N on a regional scale is encouraged to assist in enacting effective measures to protect the environments and public health, with respect to air, soil and water quality.

3.2.2. Spatial pattern

High NH_3 columns were found in Beijing, Hebei, Henan, Shandong, Hubei and Jiangsu provinces and in Eastern Sichuan province (Fig. 4a), which were consistent with their high NH_3 emissions due to intensive fertilizer application and livestock (Huang et al., 2012). Guangdong, Guangxi, Hunan and Jiangxi provinces also showed high NH_3 columns, due to high volatilization from paddy fields in these

regions, with rice being the dominant crop and contributing the most emissions. High NH_3 columns in southern China are in agreement with the high percent paddy farmland area (Fig. S1a) and the high NH_3 columns in northern China are in agreement with the high percent dry farmland area (Fig. S1b). In addition, the NH_3 emissions from vehicles in urban areas could also contribute to the observed high NH_3 columns. For example, in Beijing, the contribution of vehicles equipped with catalytic converters, particularly since the introduction of three-way-catalysts, to non-agricultural NH_3 emissions has recently been considered and might be the most important factor influencing NH_3 concentrations in urban cities (Meng et al., 2011;Xu et al., 2017). In addition, Xinjiang province also emits remarkable NH_3 emissions related to sheep manure management (Huang et al., 2012;Kang et al., 2016;Zhou et al., 2015;Liu et al., 2017a). The lower NH_3 columns are located mostly in the Tibet Plateau area, where there is a minimal amount of arable land and low use of synthetic nitrogenous fertilizers.

NO_2 columns (Fig. 4b) show significantly higher values over vast areas covering North China, East China, and the Sichuan Basin. The NO_2 columns also show high values over the Pearl River Delta, the southern part of Northeast China, and some areas in Northwest China. High NO_2 columns are mostly distributed in populated areas (Fig. S2), where there is a mix of various anthropogenic NO_x sources, such as vehicles and industrial complexes (Wang et al., 2012;Xu et al., 2015;Meng et al., 2010). It should be noted that an enhanced emission intensity from transportation is confirmed since 2005, even with staged implementation of tightened emission standards for on-road vehicles (Wang et al., 2012). For example, NO_x emissions from transportation grew to 30% for the whole country in 2014, and the values reached 44%, 55%, and 33% for Beijing, Shanghai, and Guangdong, respectively (Xia et al., 2016). Therefore, transportation is believed to play an increasingly important role in regional NO_2 pollution, especially when emissions from stationary sources are gradually controlled through

increased penetration of selective catalytic/non-catalytic reduction (SCR/SNCR) systems.

3.2.3. Limitations of satellite observations

It is difficult to gain whole coverage over China based on the daily data for both IASI NH_3 and OMI NO_2. For daily NO_2, 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). For NH_3, the satellite instruments were strongly dependent on the meteorological conditions such as cloud fractions or the availability of the temperature profiles (Van Damme et al., 2014b;Boersma et al., 2011), and we cannot retrieve the whole coverage based on daily data over China. It will be beneficial to analyze a very local region with enough numbers of observations, but not appropriate to analyze such large coverage over China.

Facing this big challenge, we used the monthly data for the trend analysis over China. The uncertainty of DOMINO v2.0 NO_2 columns has been well documented in Boersma et al. (2011), and the relative error is reported lower than 20-30% in East Asian by an improved altitude-dependent air mass factor look-up table, a more realistic atmospheric profile, an increased number of reference vertical layers and advanced surface albedos (Boersma et al., 2011). The reader is strongly suggested to refer to Boersma et al. (2011) for more details on the uncertainty analysis.

The potential uncertainty of IASI NH_3 columns resulted from IASI observation instruments and retrieval algorithms. In this paper, the NH_3 datasets were generated based on the recent-updated robust and flexible NH_3 retrieval algorithms, which were designed to overcome some shortcomings of the

current algorithms (Whitburn et al., 2016a). The current algorithms were designed firstly to calculate the hyperspectral range index (HRI), a measure for the NH_3 signature strength in the spectrum, and then converted to IASI NH_3 columns by using the thermal contrast (TC) and lookup tables (LUT) of (HRI, TC) pair corresponding to NH_3 columns. The retrieval of HRIs is strongly dependent on the amount of NH_3 and the thermal state of the atmosphere (Whitburn et al., 2016a). The quality of the IASI NH_3 product has been validated by atmospheric chemistry transport models, ground-based and airborne measurements, and NH_3 total columns obtained with ground-based Fourier transform infrared spectroscopy (FTIR). A first validation of the IASI NH_3 using the LOTOS-EUROS model was conducted over Europe, indicating the respective consistency of IASI measurements and model simulations (Van Damme et al., 2014c). A first evaluation of IASI NH_3 dataset using ground-based measurements was made worldwide, presenting consistency with the available ground-based observations and denoting promising results for evaluation by using independent airborne data (Van Damme et al., 2014a). A first validation of of IASI NH_3 dataset using ground-based FTIR derived NH_3 total columns was evaluated, demonstrating a mean relative difference of −32.4±(56.3)%, a correlation r of 0.8 with a slope of 0.73 (Dammers et al., 2016).

3.3. Atmospheric chemistry transport model NO_2 and NH_3 columns since 2008

Satellite NO_2 and NH_3 columns were observed at overpass time as an instantaneous point in a day (at 9:30 A.M. for IASI NH_3 and at 1:45 P.M. for OMI NO_2 local time). These instantaneous satellite observations may not be representative for the temporal trend analysis over China. We further retrieved the monthly variations of NO_2 and NH_3 columns since 2008 from MOZART varying 6 hours every day (00, 06, 12, 18 h). We compared the temporal trend analysis of NO_2 from MOZART at 12 h with that gained from satellite at the overpass time (OMI 1:45 P.M. local time) as well as for NH_3.

Fig. 5 shows the NO_2 columns at 12:00 during warm and cold months between 2008 and 2015 from MOZART. The percent increase rate for NO_2 columns at 12:00 during warm months (April-September) between 2008 and 2011 was 4.02% y^{-1} (Fig. 5), which was comparable with that (4.23% y^{-1}) derived from OMI (Fig. 3). During 2011-2015, we found a slightly lower decrease rate (-2.93% y^{-1}) in NO_2 columns during warm months at 12:00 from MOZART (Fig. 5) than that (-3.62% y^{-1}) gained from OMI at 13:45 (Fig. 3). The temporal variations of NO_2 columns at 12:00 from MOZART were generally in accord with those from OMI at 13:45 P.M. local time. Fig. 5 also demonstrates the average NO_2 columns (averaged at 00, 06, 12 and 18 h) during warm and cold months between 2008 and 2015. We found a close increase rate at 12:00 (4.02%) with that averaged at 00, 06, 12 and 18 h (4.23%) before 2011, as well as a similar decrease rate at 12:00 (-2.93%) and the average (-3.07%), implying that the temporal trend analysis at 12:00 vs. that averaged at 00, 06, 12 and 18 h can be considered mostly consistent over China from MOZART.

For NH_3, we found the percent increase rate at 12:00 during warm months between 2008 and 2015 was 1.30% y^{-1} from MOZART (Fig. 5), which was lower than that (2.37% y^{-1}) from IASI during 2008-2014. The percent increase rate by daily average (at 00, 06, 12 and 18 h) during warm months between 2008 and 2015 was 1.36% y^{-1} from MOZART (Fig. 5). In 2015, we found a relatively large increase in NH_3 columns in China during warm months between 2014 and 2015 (50.45%) from IASI, while an increase from MOZART was about 8.13% between 2014 and 2015. 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 $(NH_4)_2SO_4$ particles, which form the primary system of gas-particle partitioning (NH_3-NH_4^+-NO_x-NO_3^--SO_2-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.

Large difference in the NH_3 increase rate in 2015 was found between IASI (50.45%) and MOZART (8.13%). This may be still an open question on this point, here we only show this two possibilities. We should clarify in particular we do not aim at validating which is right or wrong from IASI and MOZART (which may be beyond the discussion in this paper), but the NH_3 columns in 2015 indeed increased both from IASI and MOZART. At the current state, we can, at least, draw a conclusion that the NH_3 columns over China indeed increased in 2015 both from IASI and MOZART, but a debate or inconsistency exists on the increase rate of the NH_3 columns in 2015. We should state in particular again that the following discussion in this paragraph was all hypothetical and should be tested in the future work. For IASI NH_3 columns, the sharp increase in 2015 over China may be an artifact, which may be due to an update of the input data. Similar jumps in IASI NH_3 increase in 2015 can also be visible in the USA and European (Fig. 6), indicating that it may be necessary for a recalculation of the earlier input datasets used for calculating the IASI NH_3 columns since September, 2014.

3.4. Implications for estimating long-term Nr deposition datasets and recommendations for future work

We found both the NO_x and NH_3 over China increased continuously from 1980 to 2010 based on emissions data from REAS and EDGAR. In recent years, based on satellite observations, we found an increase of 2.37% y^{-1} in NH_3 columns during 2008-2014. We also found high-level NO_2 columns over China from 2005-2011 (4.07% y^{-1}) but a decrease from 2011 to 2015 (-3.62% y^{-1}). Despite the decline, the NO_2 columns during 2011-2015 were still in high level with an average of 1.87×10^{15} molec. cm^{-2} y^{-1} compared with that (1.65×10^{15} molec. cm^{-2} y^{-1}) during 2005-2010. Notably, these emissions

certainly lead to the deposition of atmospheric Nr in form of dry and wet processes into aquatic ecosystems and terrestrial, with implications affecting ecosystem and human health, biological diversity and greenhouse gas balances (Lu et al., 2016). Hence, it is very crucial to estimate Nr deposition with high spatiotemporal resolutions in order to drive ecological models such as the Denitrification-Decomposition (DNDC) model and Integrated BIosphere Simulator (IBIS), to assess its impact on soil, forest, water and greenhouse balance. Here, we call for a long-term dataset of Nr depositions both regionally and globally to investigate how the N emissions affect the environment. Challenge still exits in estimating both the dry (NO_2, HNO_3 particulate NO_3^-, NH_3 and particulate NH_4^+) and wet (NH_4^+ and NO_3^- in precipitation) depositions for a long-term dataset such as since 1980 or earlier possibly due to the complex scheme of N transformations and transportation or limited available data both from emissions, satellites and a limited number of ground measurements.

Satellite observations provide a new perspective of estimating Nr depositions regionally, and have been used to improve the estimation performance. For example, to improve the modeling performance in dry gaseous NO_2 depositions from GEOS-Chem (Goddard Earth Observing System chemical transport model), Nowlan et al. (2014) applied the OMI NO_2 columns to calibrate the simulated ground NO_2 concentrations, and then estimated the deposition between 2005 and 2007. Our previous work focusing on the dry particulate NO_3^- deposition over China was also based on the OMI NO_2 columns, MOZART simulations and monitored-based sources (Liu et al., 2017b). Geddes et al. (2017) used the satellite NO_2 columns from GOME, GOME-2 and SCIAMACHY instruments to calibrate the NO_x emissions in GEOS-Chem to estimate the NO_x depositions since 1996. The simulations combining the satellite measurements and CTM models to derive Nr depositions (Geddes and Martin, 2017;Nowlan et al., 2014) in recent years will provide relatively accurate datasets (certainly need to be validated and

modified by ground measurements).

Despite progress in satellite techniques in recent decades (for NO_2 since 1997 by GOME and for NH_3 since 2008 by IASI), we can hardly tracked studies concerning Nr depositions before 1997 based on satellite observations. Thus, with the help of emissions data such as REAS and EDGAR, we can derive long-term Nr depositions, especially before 1997. Long-term emissions data such as REAS and EDGAR will provide valuable dataset to expand the modeling Nr depositions in recent years. In order to derive the Nr depositions from the emission data, CTMs are frequently used through modeling the wet (simplified as the product of scavenging efficiency and precipitation amount) and dry process (simplified as the inferential method by multiplying the deposition velocity and gaseous or particulate concentrations). However, we still lack a comprehensive dataset of gridded long-term Nr depositions including both the dry (NO_2, HNO_3 particulate NO_3^-, NH_3 and particulate NH_4^+) and wet (NH_4^+ and NO_3^- in precipitation) processes over China, which will be addressed in future work.

Another gap is that, all the above mentioned studies focused on the NO_x depositions and did not derive the NH_y (NH_3 and NH_4^+) depositions over China. Our recent work (Liu et al., 2017a) using IASI NH_3 columns combining the vertical profiles from MOZART benefits our understanding of the ground NH_3 concentrations over China, and the satellite-derived ground NH_3 concentrations were generally in accord with the national measurements from NNDMN. To date, there are still no reports of using the satellite NH_3 columns to derive the temporal and regional NH_y depositions over China, which dominated the total Nr depositions (NO_x plus NH_y) (Liu et al., 2016b;Liu et al., 2013). The gaps of modeling NH_y depositions by applying the satellite observations combining the CTMs simulations require more efforts and further research.

4. Conclusion

Atmospheric ammonia (NH_3) and nitrogen dioxide (NO_2) play an important role in determining air quality, environmental degradation and climate change. The emission data, satellite observations and atmospheric transport modeling have great potential for understanding the temporal variations of atmospheric NH_3 and NO_2 on a regional scale, with high spatial and temporal resolutions. This study analyzed the characteristics of atmospheric NH_3 and NO_2 over China since 1980 based on the multiple datasets. The major findings were as follows:

1. Based on emission data, both significant continuous increasing trend of NH_3 and NO_x were observed from REAS (for NH_3 0.17 kg N ha^{-1} y^{-2} and for NO_x 0.16 kg N ha^{-1} y^{-2}) and EDGAR (for NH_3 0.24 kg N ha^{-1} y^{-2} and for NO_x 0.17 kg N ha^{-1} y^{-2}) over China during 1980-2010.

2. Based on the satellite observations, we found high-level NH_3 columns with the percent increase rate of 2.37% y^{-1} from 2008 to 2014. For NO_2, we found continuous high-level NO_2 columns over China from 2005-2011 but a decrease from 2011 to 2015 (still in high level). The decrease of NO_2 columns may result from more stringent strategies taken to control NO_x emissions during the 12th Five-Year-Plan, including successful application of SCR/SNCR systems in the power sector, tighter emission standards on vehicles and a series of standards with aggressive emission limits. Increasing trend of NH_3 columns may be due to continuous N fertilizer use for guaranteeing continuous increase of the crop productions. An increase in NH_3 columns may be due to decreased NH_3 removal leading to a larger fraction maintaining in gaseous state for a long time rather than changing to the condensed phase, which may be related with continuous decreased acidic gases including the NO_2 and SO_2 over China under strong control policy in 12-th FYP.

3. Based on MOZART simulations, the temporal variations of NO_2 columns at 12:00 from MOZART were generally in accord with those from OMI at 13:45 P.M. local time. We also found a close increase rate at 12:00 (4.02%) with that averaged at 00, 06, 12 and 18 h (4.23%) before 2011, as well as a similar decrease rate at 12:00 (-2.93%) and the average (-3.07%). For NH_3, we found a lower percent increase rate from MOZART (1.30% y^{-1}) than IASI (2.37% y^{-1}) between 2008 and 2014. Large difference in the NH_3 increase rate in 2015 was found between IASI (50.45%) and MOZART (8.13%).

4. The multiple datasets used in the current work have implications for estimating long-term Nr deposition datasets. The simulations combining the satellite measurements and CTM models to derive Nr depositions will provide relatively accurate datasets, and the REAS and EDGAR emissions have potential to expand the modeling Nr depositions to long-term datasets. In particular, modeling NH_y depositions by applying the satellite observations combining the CTMs simulations require more efforts and further research.

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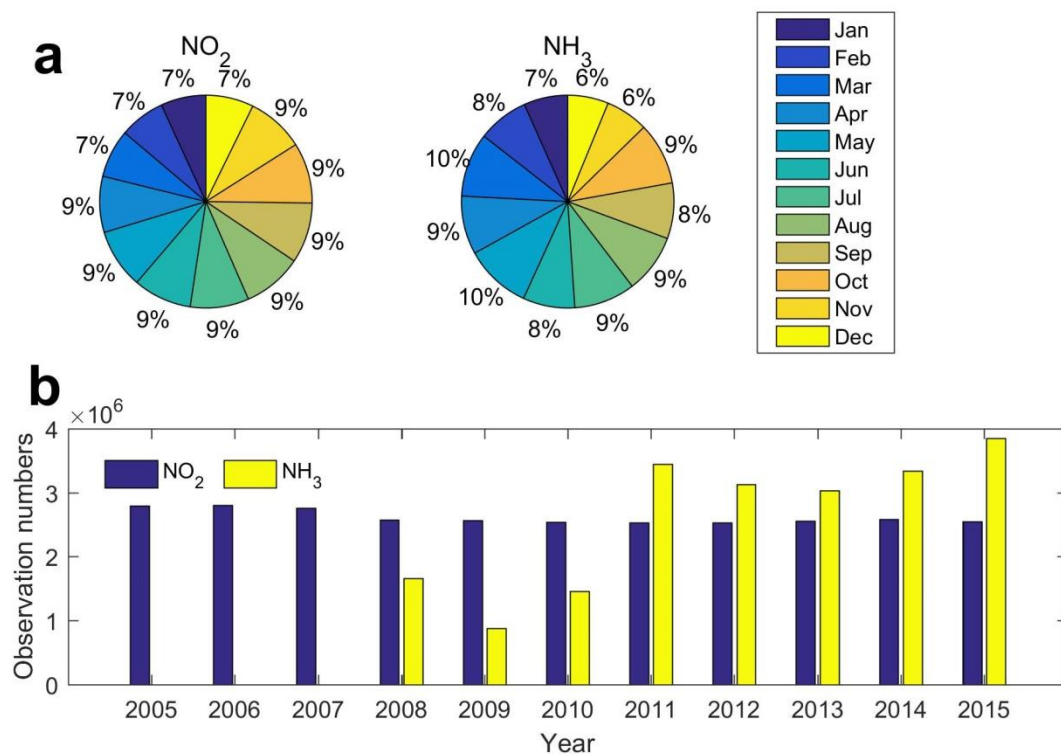http://dx.doi.org/10.1016/j.atmosenv.2015.01.069, 2015.

Figures

Fig. 1. The satellite-derived observation numbers for NO$_2$ and NH$_3$. (a) denotes the percentages of observations in each month in 2010 for NO$_2$ and in 2015 for NH$_3$ and (b) represents the total observation numbers for NO$_2$ and NH$_3$ over China. Notably, the NO$_2$ observation numbers were gained from DOMINO products with a cloud radiance fraction below 0.5, while the IASI observations with a relative error below 100% or an absolute error below 5×10^{15} molec. cm^{-2} were processed for analysis over China.
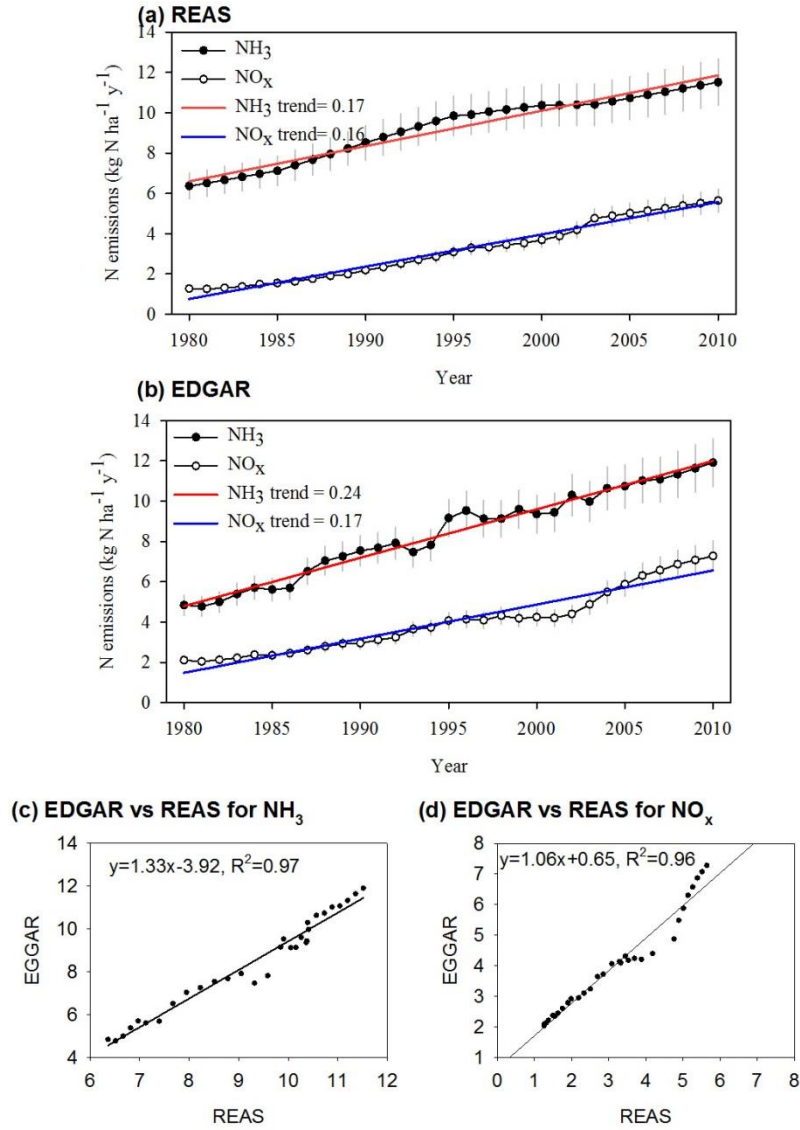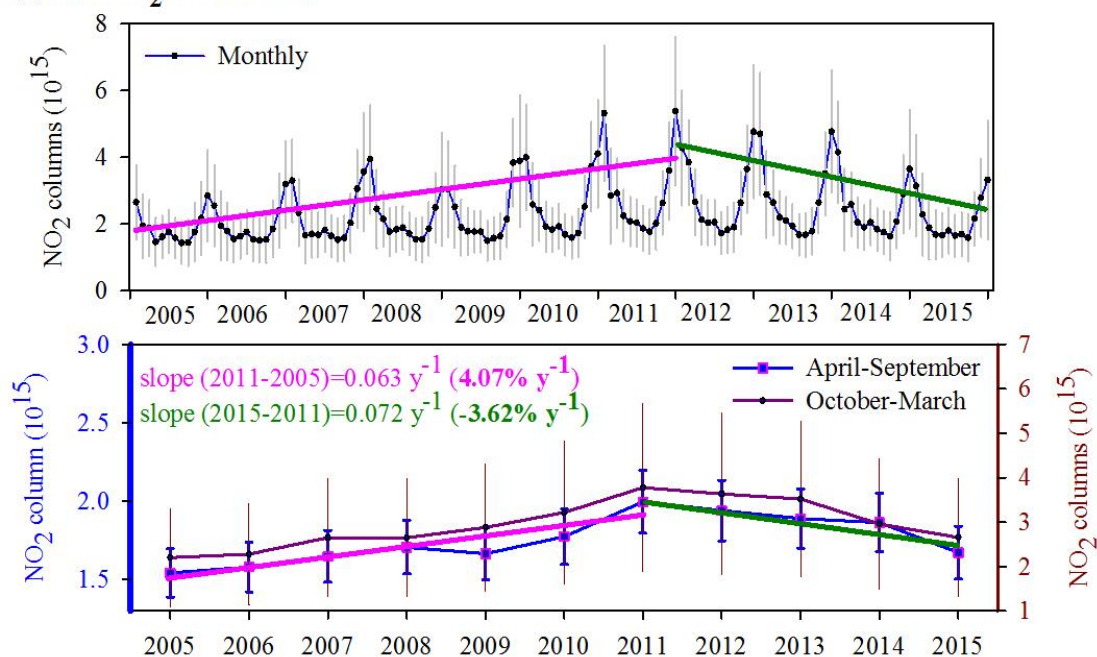
Fig. 2. The NO_2 and NH_3 emissions over China. (a) denotes the NO_2 and NH_3 emissions over China from 1980 to 2010 from REAS, (b) represents the NO_2 and NH_3 emissions over China from 1980 to 2010 from EDGAR, (c) demonstrates the relationship of NO_2 emissions over China from REAS and EDGAR and (d) shows the relationship of NH_3 emissions over China from REAS and EDGAR.
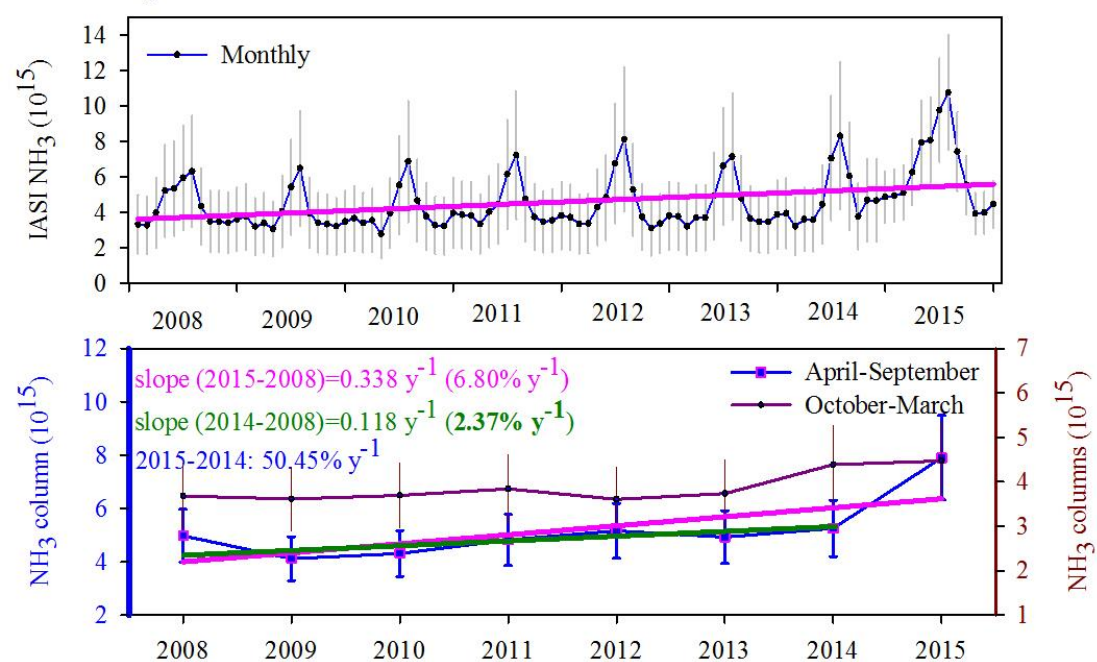
Fig. 3. Time series of average OMI NO$_2$ and IASI NH$_3$ columns over China during warm months (April-September) and cold months (October-March). The time period of NO$_2$ columns was from 2005 to 2015, while the timespan of NH$_3$ columns was from 2008 to 2015 over China. The associated mean error for each period is presented here as error bars.
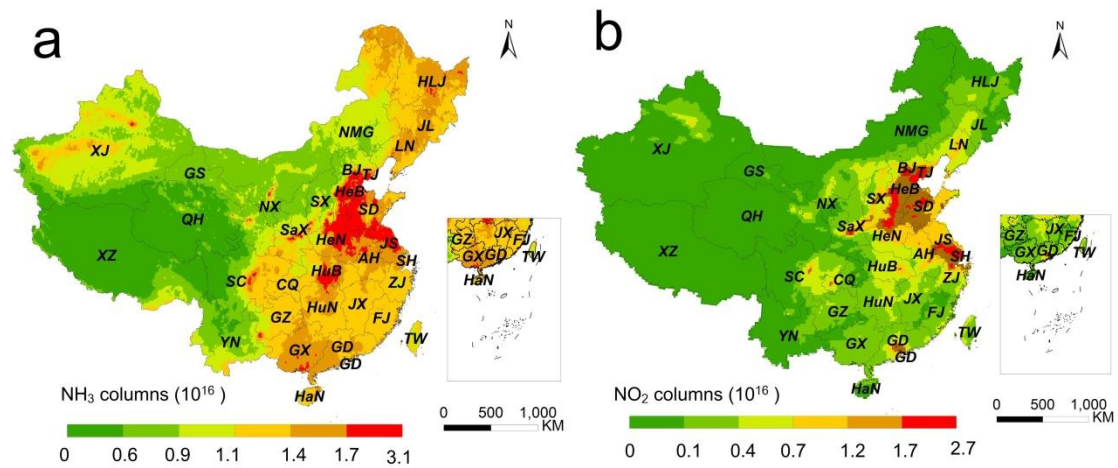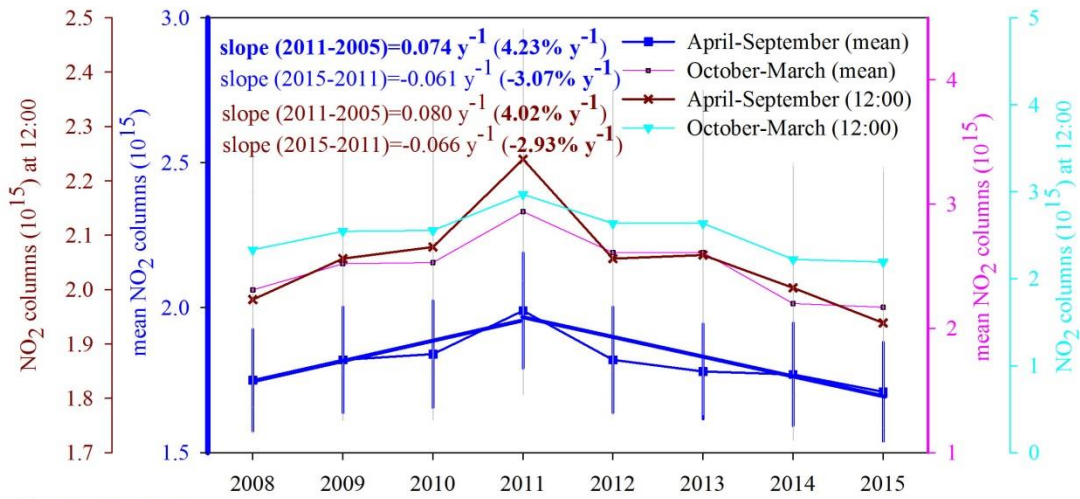
Fig. 4. Spatial distribution of the annual NH$_3$ (a) and NO$_2$ (b) columns (molecules cm^{-2} year^{-1}). The successfully full provincial names are Beijing (BJ), Tianjin (TJ), Hebei (HeB), Shandong (SD), Shanxi (SX), Henan (HeN), Shaanxi (SaX), Liaoning (LN), Jilin (JL), Heilongjiang (HLJ), Neimenggu (NMG), Gansu (GS), Ningxia (NX), Xinjiang (XJ), Shanghai (SH), Jiangsu (JS), Zhejiang (ZJ), Anhui (AH), Hubei (HuB), Hunan (HuN), Jiangxi (JX), Fujian (FJ), Guangdong (GD), Hainan (HaN), Yunnan (YN), Guizhou (GZ), Chongqing (CQ), Sichuan (SC), Guangxi (GX), Xizang (XZ) and Qinghai (QH).

(a) MOZART NO$_2$

slope (2011-2005)=0.074 y^{-1} (4.23% y^{-1})
slope (2015-2011)=-0.061 y^{-1} (-3.07% y^{-1})
slope (2011-2005)=0.080 y^{-1} (4.02% y^{-1})
slope (2015-2011)=-0.066 y^{-1} (-2.93% y^{-1})

April-September (mean)
October-March (mean)
April-September (12:00)
October-March (12:00)

(b) MOZART NH$_3$

slope (2015-2008)=0.054 y^{-1} (1.36% y^{-1})
slope (2011-2005)=0.055 y^{-1} (1.30% y^{-1})
2015-2014: 8.13% y^{-1}

April-September (mean)
October-March (mean)
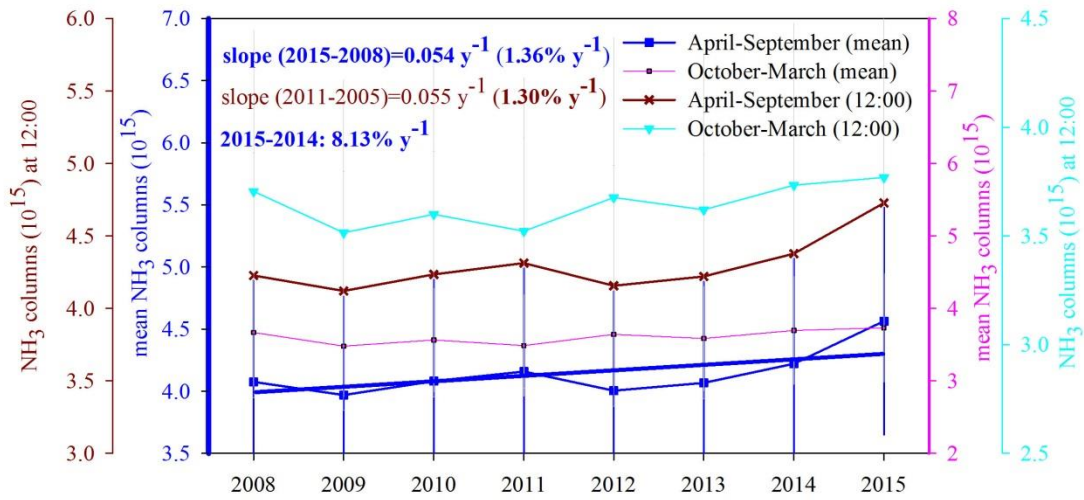April-September (12:00)
October-March (12:00)

Fig. 5. Time series of MOZART NO$_2$ and NH$_3$ 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.
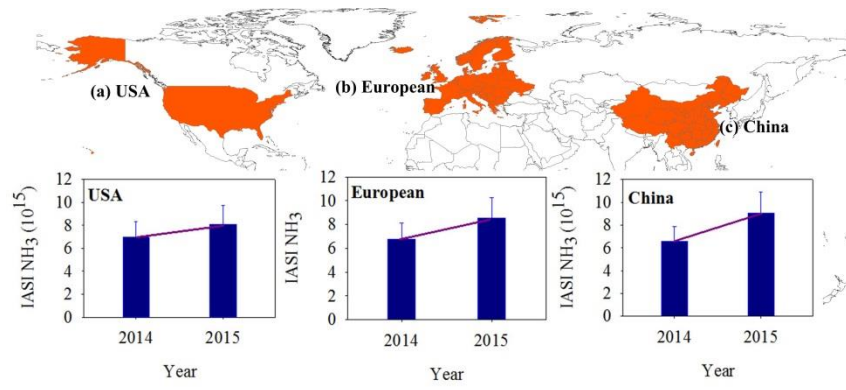
Fig. 6. IASI NH$_3$ columns in USA, European and China between 2014 and 2015.