

## **Response to Referee #1**

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This study reviews recent literatures on estimating reactive nitrogen (Nr) deposition using the satellite retrievals of NO<sub>2</sub> and NH<sub>3</sub>, proposes a framework of using satellite data to estimate Nr deposition, and suggests a few research challenges. The topic of nitrogen deposition is important, and the compilation of recent literatures on reactive nitrogen deposition is useful to the research community. However, the manuscript mainly gives general descriptions of the previous results but lacks critical analysis and synthesis. The uncertainties in satellite measurements and chemical transport models, which are key to estimating Nr deposition based on satellite column measurements, are not addressed in detail. Overall, the scientific values of this work could be enhanced by more in-depth discussion of the advancement, challenges, and directions for future research.

The authors appreciate the valuable suggestions given by Referee #1 for improving the overall quality of the manuscript. In this document, we describe how we addressed the reviewer's comments. Detailed responses to each comment are given below (in blue).

### **Specific comments:**

1. The authors highlight the advantages of satellite-based method compared to ground-based monitoring and ACTM simulation method. But there are significant uncertainties of satellite column measurements, especially for NH<sub>3</sub>. In addition, the satellite-based method strongly depends on the ACTM simulation. What are the key uncertainties of the ACTM related to deposition estimates? How do the uncertainties in satellite measurements and ACTM affect satellite-based estimation? What are recommendations to reduce these uncertainties?

Yes, the uncertainties mainly came from the satellite retrievals and ACTM simulation.

We did not aim to improve the accuracy of the satellite observations or the ACTM themselves, but to combine their advantages to gain surface  $N_r$  concentrations with better performance with the ground-based measurements. We have added the following text for more clarifications in the discussion:

“For the dry deposition, the uncertainty mainly came from the satellite-derived estimates using the modeled vertical profiles. The uncertainty of vertical profiles modeled by CTM mainly resulted from the chemical and transport mechanisms. We recommend using the Gaussian function to determine the height of surface  $NO_2$  and  $NH_3$  concentrations that best matched with the ground-based measurements. There may exist systematic biases by simply using the relationship of  $NO_2$  columns and surface concentration to estimate satellite surface  $NO_2$  concentrations.”

2. The authors propose a framework for combining satellite data, ground-based monitoring and ACTM (Figure 1). But it is not clear if it is a new idea. It seems that the approach has already been used in previous studies as indicated in the literatures shown in sections after Figure 1.

Yes, it's a new framework proposed in this study. Previous studies mainly focused on the methods to estimate surface  $NO_2$  concentrations, while **Fig. 1** shows the general approach for estimating all  $N_r$  species on both concentration and deposition.

3. The title contains “ $N_r$  concentration and deposition”, but the introduction part and the framework only mention “deposition”. In my opinion, the estimation of  $N_r$  concentrations is just a part of the estimation of  $N_r$  depositions. There are many other studies which have offered more in-depth discussions of column concentrations of  $NO_2$  and  $NH_3$ . I am not saying that concentrations cannot be shown but suggest framing the paper with a clearer focus on deposition.

Thanks for your suggestion. But, we think the introduction is appropriate since the estimation of  $N_r$  concentrations is just a part of the estimation of dry  $N_r$  depositions. The title included both the “ $N_r$  concentration” and “deposition” because we reviewed on the methods of estimating both surface  $N_r$  concentration and  $N_r$  deposition.

4. Line 193-195: Why may this method lead to an underestimation of surface  $NO_2$  concentration? In your proposed framework, the similar method has been used to estimate the surface  $NO_2$  concentration. Why is there no large underestimation in your validation? While you use the Gaussian function to fit the vertical concentration profile, but for the surface layer, you still use the ACTM derived the relationship between the  $NO_2$  column and surface  $NO_2$  concentration.

No, the methods in this study were different from the previous studies. We did not simply use the relationship between the  $NO_2$  column and surface  $NO_2$  concentration from the CTM. As presented in the main text, we can estimate surface  $NO_2$  concentration at any height by using the Gaussian function. We used the surface  $NO_2$  concentration at a certain height which best matched with the ground-based measurements.

5. Line 405-409: The derived  $NO_2$  columns from these satellites are quite different. Can you give some suggestions to the readers about which satellite data to use? Why do you choose OMI  $NO_2$  in your estimation? What are the results if you use other satellite data?

The readers can use any satellite data combining the Gaussian function to estimate surface  $NO_2$  concentrations. They can use surface  $NO_2$  concentrations at a certain height which best matched with the ground-based measurements. The key is not selecting which satellite data we should use, but determining which height of surface  $NO_2$  concentrations that better matched with the ground-based measurements by

Gaussian function.

6. Line 550-552: Can the similar method in equation 9 and 10 be used to estimate wet reduced Nr depositions? What are the different challenges for the estimations of wet reduced Nr depositions, compared with oxidized Nr?

Yes, the methods were the same for estimating both oxidized and reduced  $N_r$  deposition. We did not identify big difference in the estimations of wet oxidized and reduced  $N_r$  depositions.

7. Section 5: For the trend estimation of Nr concentrations and depositions, have you conducted ACTM simulation for each year? The changes in emission and meteorology can significantly affect the Nr vertical profile and Nr species ratio, which are important in your satellite-based estimation.

Yes, we did. Please note that the simulated profile function has a general rule, which can be well simulated by Gaussian function for any year (for our case during 2005-2016). Thus, there is no need to simulate the vertical profile of  $NO_2$  and  $NH_3$  for each year.

8. Line 567-569: This statement needs to be modified. As mentioned above, the satellite-based method strongly depends on the ACTM simulation. The uncertainties in emission inventories and other parts of ACTM can also significantly affect the vertical distribution of pollutants and the ratios of  $NO_2$  and other Nr species (e.g.  $HNO_3$ ,  $NH_4^+$ ).

No, the emission inventories should not affect the vertical profiles shapes using Gaussian function, but the transport and chemical mechanism in the CTM may affect the accuracy of the vertical profile distribution. We mean that the satellite-based methods did not need to rely on the accuracy of the statistical emission data.

9. Line 697: Are there any previous studies using a mechanism method to estimate Nr

deposition?

As far as we know, previous studies using satellite NO<sub>2</sub> and NH<sub>3</sub> column to estimate wet N<sub>r</sub> deposition were through a statistical way, and no studies were done from a mechanism perspective.

**Minor comments:**

1. The authors should give the definition of reactive nitrogen (Nr). “Nr (such as NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>)” is mentioned in line 48, and “Nr (NO<sub>x</sub> and NH<sub>3</sub>)” is mentioned in line 59. This is confusing.

We have added the following text for clarifications:

“Nr refers to the general term of N-containing substances in atmosphere, plants, soils and fertilizers that are not combined with carbon”.

2. Line 57, change “mineral energy” to “fossil energy”.

We have revised it as suggested.

3. Line 83, add “and” between the two words “accurate quantitative”.

We have revised it as suggested.

4. Line 145-146: “Tian et al.” should be “Tan et al. (2018)”.

We have revised it as suggested.

5. Line 170: “Cheng et al. (Cheng et al., 2013)” should be “Cheng et al. (2013)”.

Please check the citation format throughout the manuscript.

We have checked the citation format throughout the manuscript as suggested.

6. Line 170-171: This sentence is not easy to understand. Please revise it.

We have revised it as follows:

“This method used the simple linear model and did not consider the vertical profiles of NO<sub>2</sub> (Cheng et al., 2013)”

7. Line 198-200: The study of Larkin et al., 2017 should be put in the previous

paragraph discussing the method using the satellite data and statistical model. I think that the authors are discussing the method using the satellite data and ACTM-derived relationship in this paragraph.

No, Larkin et al. (2017) were also based on the satellite data and ACTM-derived relationship similar to Geddes et al. (2016), and it should be there.

8. Line 225-232: This information based on Jia et al. (2016) has been mentioned in line 176-184. They are repetitive.

We have removed it to avoid repetitive.