

Response to comments #2

We appreciate your constructive and positive comments. The comments and proposed corrections have been taken into account and helped to improve the paper. Each comment has been addressed as follows. There is an extensive discussion among the authors regarding how to revise the content. So the response is delayed, and we are sorry for this.

General comments

This paper presents ground based FTIR measurements at the Hefei site including error analysis of the NH₃ retrievals, vertical distribution, time series and seasonal trend analyses. More additional works such as comparisons of IASI data, relationship with surface CO, temperature, wind speed and direction, and back trajectories analysis are made in this paper. I believe this paper is suitable for publication to Atmos. Meas. Tech. after considering comments as below. As for dividing into two papers by another referee, if authors can prepare more analyses and discussions for AMT and another paper, I think it would be better. If not, One paper of AMT looks not bad.

Response: We didn't divide the paper into two papers after discussion among the authors

Specific comments

line 2

I feel "measured" is not fitted because of "measured from observations". Retrieved, derived, obtained etc. would be better.

Response: We replaced "measured" with "retrieved".

Line 32 and 402

If possible, could you provide error (one standard deviation) for 22.14 % yr⁻¹ annual increase rate?

Response: We replaced "annual increase rate" with "increase of the annual mean", as another reviewer think it is premature to report the annual trend. So we gave the annual mean and one standard deviation for the two years.

Line 38-39

"Further, high correlation of NH₃ columns with air temperature is obvious from their diurnal variation during the observation period."

"In addition, the clear correlation between NH₃ columns and air temperature in spring and autumn over Hefei, suggests that agriculture was indeed the main source of ammonia in spring and autumn."

I think a correlation coefficient NH₃ columns with air temperature should be provided, since NH₃ columns with CO concentration is described as R=0.77.

Response: We calculated the correlation coefficient between NH₃ columns with air

temperature from their diurnal variation, and the correlation coefficient is 0.39, showing a weak correlation between NH₃ and temperature from their diurnal variation. The NH₃ data measured after 14:00 PM local time are sparse as we collect the MIR and NIR solar spectra alternately every day, so the data in some time period lacks of representation for diurnal variation analysis. Therefore we removed the discussion about the correlation of NH₃ columns with air temperature from their diurnal variation, including Figure 8, in section 3.3 “Identification of emission sources of NH₃”.

Line 83-89

Authors had better add the GOSAT retrieval from TANSO-FTS TIR spectra as recent results.

Citation: Someya, Y., Imasu, R., Shiomi, K., and Saitoh, N.: Atmospheric ammonia retrieval from the TANSO-FTS/GOSAT thermal infrared sounder, *Atmos. Meas. Tech.*, 13, 309–321, <https://doi.org/10.5194/amt-13-309-2020>, 2020.

Response: We added the GOSAT retrieval from TANSO-FTS TIR spectra as recent results, and included the citation.

Line 99-102

“More recently, FTIR measurements have been shown to also provide total column and vertical profiles of ammonia at a high temporal resolution, and are now also used for validation of satellite NH₃ observations (Dammers, et al., 2015; Dammers, et al., 2016; Dammers, et al., 2017b).”

I recommend to adding a name of the satellite, that is, IASI.

Response: We added the name of the satellites, i. e. “IASI and CrIS”.

Line 226

Some of working in same field can understand “phase”, but for wider readers, a little explanation might be necessary.

Response: We added the description of “phase”, including two citations.

Line 250

“Many spectra ranging from 700 to 1350 cm⁻¹ are saturated in summer (due to high humidity), causing the retrieved NH₃ data to be sparsely sampled relative to those in other seasons.”

Are there else any better spectral windows for retrieval of NH₃ in summer season?

Response: We tested a few spectral windows for retrieval of NH₃ in summer season, and chose the two microwindows, according to the fitting RMS, DOFS and other output parameters.

Line 281

“The annual increasing rate of ammonia columns in Hefei estimated by our two-year FTIR measurements (22.14 % yr⁻¹) is much larger than the reported value by satellite observations over China. This is likely due to the different sampling years. The

increasing trend of NH₃ in Hefei is likely caused by either an increased fertilizer use, or increasing air temperature, or decreased sulfur emissions due to strict SO₂ control measures.”

Is it possible to verify the annual increasing rate of ammonia columns over Hefei using other satellite or model data?

Response: We used the IASI data from 2016 to 2018 over Hefei to calculate the annual increase of ammonia columns. The increase of NH₃ is 19.66% and 8.92% for the annual mean from 2016 to 2017, and from 2017 to 2018, respectively, for IASI-A data. It is 5.97% and 5.13% for the annual mean from 2016 to 2017, and from 2017 to 2018, respectively, for IASI-B data. The increase values are very different for IASI themselves. I don't know the reason. So I didn't use other satellite or model data to verify the annual increasing rate of ammonia columns over Hefei.

Line 291

“We remove the data with negative IASI-NH₃ columns due to large retrieval error.”

Negative values for the IASI-NH₃ columns are not physically meaning. I think large retrieval error is not fundamental reason.

Response: We corrected this sentence.

Line 303-311

There would be different results comparing of IASI A and IASI B data with the Hefei FTIR data, but they are within one standard deviation. Could you describe this reason by citing the literatures or technical reports from the IASI team? If impossible, a description that the difference is within one standard deviation would be there.

Response: The IASI team provided the new version of IASI data. We compared the IASI data of new version with FTIR data, so the comparison results are slightly different with the previous ones. We added the description that the difference is within one standard deviation.

Line 319-321

“So the relative differences between IASI total columns and our FTIR data and standard deviations of the differences are within the range of comparison results from other NDACC site data, and the correlation coefficients are comparable to that of other comparison results.”

In table 3 of Damers et al. (2017b) paper, the mean relative difference (MRD) at Wollongong site is only positive ($6.0 \pm (74.3)\%$) and other sites are negative. If readers know this, readers may get confused. More detail discussions and descriptions are necessary.

Response: We added discussions about the relative difference.

Line 338

“However, NH₃ columns show high correlation with CO concentrations in summer, as displayed in Figure 7(a).”

How is other seasons ?

Response: We added “NH₃ columns show negligible correlation with CO concentrations in other seasons.”

Line 345

“Meanwhile, NH₃ columns show weak correlation (R=0.47) with PM_{2.5} concentrations (Fig. 7(b)), meaning that NH₃ contributed to the formation of fine particulates significantly in summer.”

If so, a correlation with PM_{2.5} concentrations in other seasons might be higher than that in summer. Did authors check them?

Response: We calculated the correlation of NH₃ columns with PM_{2.5} concentrations, but there are negligible correlations in other seasons. Sorry the concentrations of ammonium(NH₄⁺) and sulfate-nitrate- ammonium(SNA) aerosols near our Hefei site are not available, it is difficult to assess the contribution of NH₃ to PM_{2.5}. Also, the purpose of the section is to find the possible emission sources of NH₃, so we removed the description about the relationship of NH₃ and PM_{2.5} in the paper.

Line 352

“High correlation of NH₃ columns with air temperature is obvious from their diurnal variation during the observation period, as seen in Figure 8. Our measurements are performed generally from 9:00 to 16:00 local time. The whole data are averaged per hour during the two years.” Considering discussions that follow, a plot in Figure 8 prepared for whole data (I understand all seasons) should be prepared for each seasons. Could authors explain a reason that NH₃ columns decreased from 11:30 to 13:30 in figure 8? If plots are prepared for each season, decreasing in spring and autumn might be appeared.

Response: We ever plotted Fig.8 for each seasons, but the variations of temperature and NH₃ columns seem unreasonable. Also, We calculated the correlation coefficient between NH₃ columns with air temperature from their diurnal variation, and the correlation coefficient is 0.39, showing a weak correlation between NH₃ and temperature from their diurnal variation. The NH₃ data measured after 14:00 PM local time are sparse as we collect the MIR and NIR solar spectra alternately every day, so the data in some time period lacks of representation for diurnal variation analysis. Therefore we removed the discussion about the correlation of NH₃ columns with air temperature from their diurnal variation, including Figure 8, in section 3.3 “Identification of emission sources of NH₃”.

Technical corrections

line 28-29

I fell there is a duplication. One idea is to remove “a measurement site in”.

Response: We removed “a measurement site in”.

Line 32

measurement-> measurements

Response: We corrected this word.

Line 35

Analyze -> analyzed?

Response: We corrected this word.

Line 143

Remove “retrieved”

Response: We removed this word.

Line 169

“recorded”-> “has been recording”

Response: We corrected this word.

Line 179

“vertical profile”->” vertical profiles”

Response: We corrected this phrase.

Line 204

“general”-> “generally”

Response: We corrected this word.

Line 235

“The seasonal averaged surface level of NH₃ decreased from 10.82 ppb in summer to 2.92 ppb in winter during 2017 and 2018, and the corresponding values are about 5.48 and 6.04 ppb in spring and autumn, respectively.”

Line 261

“The annual mean NH₃ column is 1.31×10^{16} and 1.60×10^{16} molec cm⁻², respectively, with an increase rate of about 22.14 %.”

If readers can know errors or standard deviation, they might be good.

Response: We added the standard deviation for these values.

Line 243, 244, 249 and other

“retrievals at the Hefei site” or “retrievals in the Hefei site.” A lot of inconsistency, “at the Hefei site” might to be good.

Response: We corrected this phrase throughout the paper.

Line 264

If authors use “practices” as a noun, I think “maybe” is adverb and there is no verb in this sentence.

Response: We corrected this sentence.

Line 269

“agriculture”-> agricultural area?

Response: We corrected this phrase.

Line 325

“tunnel studies”

Simple description for them is grateful.

Response: We added simple description for tunnel studies.

Line 330

“The Dongpu Reservoir air quality monitoring station (31.91°N, 117.16°E) is very close to our site, part of a National Ambient Air Quality Monitoring Network, which monitors and routinely publishes the concentrations of main gaseous pollutants, including CO, NO₂, PM_{2.5}, PM₁₀, SO₂, O₃ and Air Quality Index (AQI) etc.”

There is no citation for the data.

Response: We added a citation for the air quality data.

Line 434

“Future work” -> “Future works”

Response: We corrected this word.

Line 435

“to estimate regional”->“estimating regional” or “estimation of regional”

Response: We corrected this phrase.

Figure 1 caption

“The regional distributions of NH₃ columns (molec cm⁻²) from 2008-2018 IASI-A and 2013-2018 IASI-B morning overpasses of ANNI-NH₃-v3R data.”

Are they averaged values or overlaid ? Clarification would be necessary.

Response: They are averaged values. We clarified this.

Figure 4 (a)

Higher than 40 km should be removed for a color bar for the altitude and replot would be necessary. If Authors can do them, readers may understand which colors are altitudes for VMR averaging kernels. But I don't know it is useful.

Response: We replotted Figure 4, removed the part higher than 20km.

Figure 7 (b)

“ug”->“micro g” Micro is small Greek letter.

Response: we removed Fig. 7(b), as we removed the description about the relationship of NH₃ column with PM_{2.5} concentration.

Figure 10

Digits after the decimal point might be not necessary for NH₃ column. Also digits after the first decimal point might be not necessary for wind speed.

I feel wind speed would for radial axes and NH₃ columns for color bars would be better for better understanding relationship of NH₃ columns to wind direction and speed. If authors did not try, please try.

Response: We replotted Fig. 10, with wind speed for radial axes and NH₃ columns for color bars.

Figure S1.

Back trajectories colored with black are very cloudy, if possible could authors color them for each cluster?

Response: Sorry it is difficult to color back trajectories for each cluster. We colored them using black dashed lines to replace black solid lines, to reduce cloudy.

Figures S1 and S2

What is light blue curved lines? There is no description for them.

Response: The light blue curved lines are rivers in China. Some panels showed them, while others didn't show. We replotted Figure S1 and S2 (Now Fig. 11 and Fig.12), making the light blue curved lines disappear for consistency.