Thank you very much for the comments and suggestions for this paper. All the references have been checked and should now be in the required format.

We are grateful to referee for very thoughtful and detailed comments that helped to improve the manuscript. Below we provide a point-by-point response to the reviewers' comments and how we have addressed them in the revised manuscript (in blue).

Anonymous Referee #1

The manuscript by Li et al. reports measurements of NH_3 concentrations in northeastern Colorado, and compares them to remote sensing retrievals and model estimates.

The combination of measurements, including surface and tower, with the satellite and model analysis is fairly unique, and is quite valuable for learning more about the accuracy of each. The paper is generally clear and well written, although there are some parts where more references could be provided and the introduction could use more content regarding remote sensing and modeling, given their importance to the value of this work. My comments below touch on these points, as well as a few others, which constitute minor revisions.

Comments:

55-56: Please split up these so that they are associated with the specific impacts being discussed, rather than all placed at the end of the sentence

such that it's not clear which paper is about which impact.

[Response]:

The references have been split up as shown below.

"$PM_{2.5}$ has been linked to adverse effects on human health (Davidson et al., 2005; Schwartz and Neas, 2000; Lelieveld et al., 2015) and regional visibility (Park et al., 2006), and also impacts climate via direct and indirect changes in radiative forcing (Langridge et al., 2012; Parry et al., 2007)."

70-72: That seems rather obvious. What is the contribution of NH_3, relative to oxidized sources, to Nr deposition more broadly, not just near farms?

[Response]:

This sentence was added after "…nitrogen deposition to sensitive ecosystems": "Li et al. (2016) analyzed wet and dry deposition of reactive nitrogen across the U.S. and found that reduced nitrogen, derived from ammonia emissions, now constitutes the majority of inorganic nitrogen deposition in most regions."

In response to a later reviewer comment, we also added the following sentence concerning other, non-agricultural sources of ammonia:

"Besides the dominant contributions from agricultural sources, ambient NH_3 also originates from other sources such as vehicles with three-way catalysts (Shelef and Gandhi, 1974;Chang et al., 2016). Biomass burning (such as wildfires) is another important source of NH_3 (Benedict et al., 2017): in the 2014 U.S. NEI, wildfires make up nearly 4.3% of national NH_3 emissions."

74-75: Which of these references are government regulators? I don't think any of them. You might instead specifically cite voluntary NH₃ control programs in the US, or actual government control programs in other parts of the world.

[Response]:

"Agricultural NH₃ emissions have become one of the most significant air pollution problems in recent years and have attracted growing concern from environmental scientists and government regulators (Aneja et al., 2006; Pan et al., 2012; Bauer et al., 2016)"

has been changed to

"Agricultural NH₃ emissions have become one of the most prominent air pollution problems in recent years and have attracted growing concerns (Aneja et al., 2006; Pan et al., 2012; Bauer et al., 2016). Within the U.S., efforts to routinely monitor ammonia concentrations have been growing via the Ammonia Monitoring Network (Amon: http://nadp.sws.uiuc.edu/AMoN/sites/data), ammonia can now be considered as a precursor to PM₂.₅ in the state implementation planning process for meeting the national ambient air quality standards, and voluntary reductions in ammonia emissions have been prioritized as part of efforts to reduce reactive nitrogen deposition in Rocky Mountain National Park (http://www.rmwarningsystem.com/ReducingAmmoniaEmissions.aspx)."

85-88: Please provide references for the RoMANS studies. Also, several other studies also examined source of Nr in RMNP, such as: Benedict et al., AE,

2013; Malm et al., JAWM, 2013; Thompson et al., JGR, 2015; Lee et al., ACP, 2016.

[Response]:

A link to the official website for the RoMANS studies and several references related to Nr in RMNP (Beem et al., EP, 2010; Benedict et al., AE, 2013; Malm et al., JAWM; 2013, Thompson et al., JGR, 2015; and Malm et al., AE; 2016) have been added.

111 -115: Can some background be provided on comparison of remote sensing of NH_3 to in situ measurements? Surely this isn't the first study to perform such a comparison, for IASI or other instruments; or if it is, this aspect should be more prominently featured. Additionally, has the vertical profile of NH_3 been studied before, with tower measurements, remote sensing, or aircraft-based instruments? I find some text on this later in the manuscript, but sees like for a major component of the work it should be included in the introduction.

[Response]:

The following paragraph has been added to the manuscript after "…a cause of NH_3 concentration under-estimation in the west".

"Van Damme et al. (2015) used measured NH3 data from the U.S., China, Africa, and Europe (ground-based and airborne observations) and compared these data with IASI-NH_3 columns. During the DISCOVER-AQ campaign, Sun et al. (2015) also compared in situ observations (airborne and vehicle-based) with Tropospheric Emission Spectrometer (TES) NH_3 columns. Both comparisons demonstrated fair agreement between in situ measurements

and satellite total columns, indicating that NH_3 data from in situ measurements and satellite retrievals are reliable. The discrepancy between model predictions and observations of NH_3 concentrations suggests that variability in the spatial and/or temporal distribution of NH_3 is not captured by current emissions inventories or model inputs, and additional understanding of atmospheric NH_3 distributions, for example, with height above ground level, is needed. Vertical NH_3 profiles have previously been reported from airborne studies such as CalNex (Nowak et al., 2012;Schiferl et al., 2014), the DISCOVER-AQ campaign (Sun et al., 2015;Müller et al., 2014), and from measurements made at the Canadian oil sands (Shephard et al., 2015). These studies have found strong variation of NH_3 concentration above ground, but do not provide a sufficient basis to characterize the general vertical distribution of NH_3 with limited sampling periods.

111 - 115: Same goes for CAMx âA˘T what have previous studies found with regards to CAMx model estimates of NH_3, or is this work the first study to compare CAMx estimates with in situ NH_3 measurements?

[Response]:

The following paragraph has been added to the manuscript after the satellite observation discussion referred to in the last comment:

"Several model performance evaluations (MPEs) have found model predictions of NH_3 concentrations in the western U.S. to be low (Rodriguez et al., 2011; Thompson et al., 2015; Battye et al., 2016). Rodriguez et al. (2011) and Thompson et al. (2015) utilized the Comprehensive Air quality Model with extensions (CAMx); Battye et al. (2016), meanwhile, ran a different photochemical model (CMAQ), and utilized emissions inventories generated

with less focus on the precise spatial positioning of agricultural sector emissions in the Inter-Mountain West. Evaluation of NH_3 concentration prediction performance in larger scale models has suggested that uncertainty in emissions inventories is a cause of NH_3 concentration under-estimation in the west (Zhu et al., 2013;Heald et al., 2012).."

202: Can the authors briefly comment on how close these stations are to the NH_3 measurement sites?

[Response]:

The following paragraph has been added to the manuscript after "…weather stations (http://www.coagmet.com/) (Table S1)":

"The distance between the NH_3 measurement site and the nearby meteorological stations referenced in the paper were from 0.1 km (KSY01 to KY) to 68.1 km (BRG01 to BH), with an average value of 16.5 km."

284: Could the authors be more specific about the improvements? Not sure if I see the value of this statement otherwise.

[Response]:

The following sentence has been revised: "The new NN-based method inherits the advantages of the LUT-based HRI method whilst providing several significant improvements (Whitburn et al., 2016)."

This sentence now reads: "The new NN-based method inherits the advantages of the LUT-based HRI method whilst providing several significant improvements such as: (1) better sensitivity at low concentrations due to the large variation in temperature, pressure and humidity vertical profiles in the

retrieval; (2) a reduction of the reported positive bias of LUT retrieval at low concentrations; (3) the possible consideration of NH_3 vertical profile information from third party sources; and (4) a full uncertainty characterization of the retrieved column variables (Whitburn et al., 2016)."

296: Are the emission factors constant in time and season? How reasonable is this?

[Response]:

In some estimation methods, the emission factors do vary in time and season. Since the animal population given in each feedlot is just an annual value, we decided to use the constant emission factors reported by U.S. EPA (2004) and Todd et al. (2013) to estimate the general spatial NH_3 emissions patterns from livestock.

311: It's interesting that this is the first place in the paper that non agricultural source of NH_3 are mentioned. Biomass burning then shows up on line 368. Should some background on these other types of sources be included in the introduction?

[Response]:

As mentioned in an earlier response, we have added mention of non-ag sources to the revised introduction. Further, the following paragraph has been added:

"Biomass burning (such as wildfires) is another important source of NH_3 (Benedict et al., 2017): in the 2014 U.S. NEI, wildfires make up nearly 4.3% of national NH3 emissions."

307: Specify over what time period the word "average" refers to here.

[Response]:

"from 2010 to 2015," has been added after "The lowest average ambient NH_3 concentrations …".

296: It appears the authors are generating their own emissions estimates through a combination of local livestock information from CDPHE with emissions factors from EPA. How different then are their estimates of livestock NH_3 emissions in this area compared to other NH_3 emission inventories assembled by research groups or government organizations?

[Response]:

We used a simple calculation to estimate NH_3 emissions from livestock. The point here is simply to illustrate the spatial pattern of large livestock NH_3 sources. It is not intended to correctly predict the overall magnitude of this source category on a less refined spatial scale, the typical focus of other emission inventories.

313-316: The analysis here is a bit weak. The overall mean NH_3 at GC is 25% higher than at FC_W and 17% higher than at LD. Why are these insignificant differences? Given the variances in the measurements, couldn't one calculate whether these differences are significant with a t-test? My guess is the difference is systematic, not random, given that the reported value at GC is the highest of these 3 in every subperiod proved in Table 2. From a physical perspective, also would there be a particular time and or season when one

would expect fertilization of the golf lawn to have a big impact, and thus should analysis of the impact of this local source be sought within a narrower winder? I guess after reading the rest of this paragraph, where differences in other locations are up to x15, I understand better why such small differences here may have been glossed over, but still. . .

[Response]:

We agree with the referee on this, although our comments were really intended in the context of the much larger spatial variations observed across the full network. The following paragraph has been added after "…not a major, regional NH$_3$ source.":

"However, the NH$_3$ concentrations at the GC were modestly higher (17% on average) than NH$_3$ sampled at the LD site during each summer measurement campaign (Table 2), suggesting that the contributions from fertilization of the golf lawn cannot be neglected."

324 - 341: I recognize that not much data is available for trend analysis, but for those sites where trends were estimated to be significant, the authors did little here to comment on the nature of these trends, which was a bit disappointing. For example, is the decrease in BH possibly related to any of the management practices mentioned later?

[Response]:

The decreasing trend in BH site might be related to changes in nearby best management practices or animal populations, but we do not have adequate information to test this hypothesis and prefer not to speculate.

343: Can some reference be provided for these?

[Response]:

The website of best management practices for reducing ammonia emissions from Crop Production and animal production systems (http://www.rmwarningsystem.com/ReducingAmmoniaEmissions.aspx) were added in this paragraph.

370: There was also a paper from the TES group on correlation of NH_3 and CO from biomass burning as observed by TES.

[Response]:

The paper of Luo et al., 2015: *"Satellite observations of tropospheric ammonia and carbon monoxide: Global distributions, regional correlations and comparisons to model simulations"* has been added to the list of references.

376: There have been aircraft measurements from CalNeX, discover AQ, and over Canadian oil sands. These are probably different in terms of the extent and resolution of the analysis of vertical distributions. Still, it seems some specific references to earlier work is warranted.

[Response]:

We agree with the referee. Information about these aircraft measurements has been added to the introduction. Please refer to our response for Lines 111 -115.

We also changed the following sentence: "While surface measurements of NH$_3$ concentrations remain uncommon, measurements of vertical profiles of NH$_3$ concentrations above the surface are extremely rare."

This now reads:

"While surface measurements of NH$_3$ concentrations remain uncommon, measurements of vertical profiles of NH$_3$ concentrations above the surface are more rare, with the exception of a small number of aircraft measurements over limited time frames as mentioned in the introduction."

Fig 3: Why don't we see the impact of the High Park Fire in the time series of any other sites besides FC_W? What do the authors posit is the explanation for the peaks in GY in 2011?

[Response]:

The following paragraph has been added after "…wildfire (Prenni et al., 2012;Benedict et al., 2017).":

"The FC_W was site was the closest site to the High Park Fire and normally has relatively low ambient NH$_3$ concentration. The NH$_3$ emitted from the High Park Fire may also have reached other, more distant sites downwind; however, enhanced NH$_3$ concentrations at these sites from other nearby sources and the greater dilution of the smoke plume as it travels further downwind make it difficult to identify any impacts of the wildfire at these locations."

477: Provide some references.

[Response]:

Two references have been added as below:

"Unfortunately, current models frequently have difficulties accurately simulating spatial concentrations of NH_3 (Adelman et al., 2015; Battye et al., 2016)."

478-480: Provide some references.

[Response]:

Three references have been added as below:

"In addition to the typical model difficulties in accurately simulating transport, NH_3 emissions are not well constrained (Zhu et al., 2013) and the parameterization of NH_3 deposition is challenging (Bash et al., 2013; Pleim et al., 2013)."

498-503: Please include a brief summary of the model performance.

[Response]:

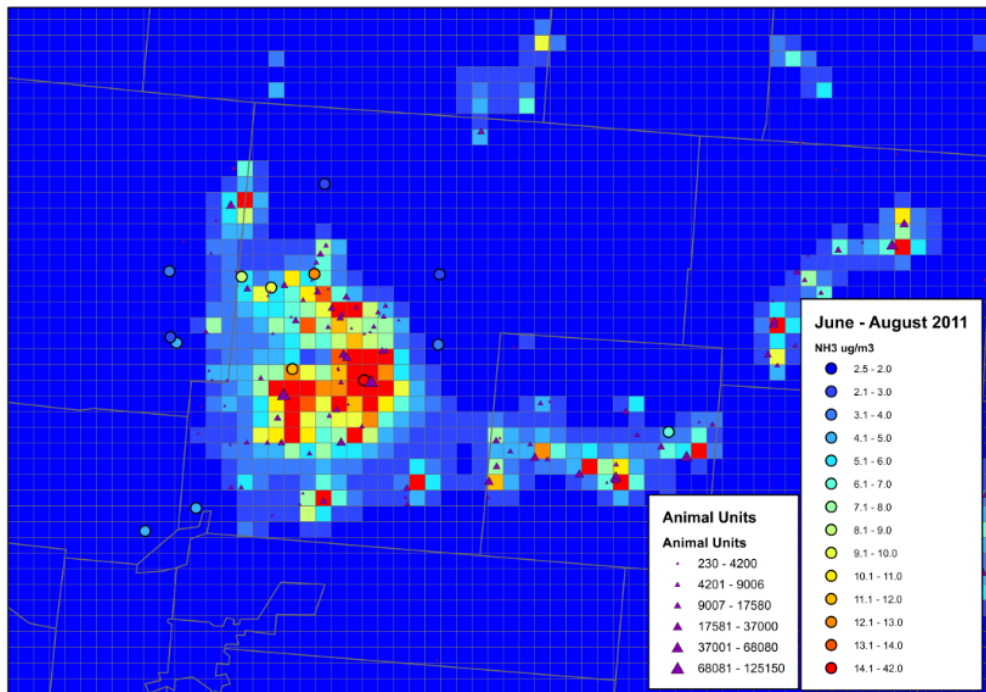
At the end of this paragraph, we added a brief summary of the model performance as below:

"Model performance was evaluated by the Intermountain West Data Warehouse team (Adelman et al., 2015). The model met performance standards as recommended by the U.S. EPA for regulatory photochemical modeling purposes (https://www3.epa.gov/scram001/guidance/guide/Draft_O3-PM-RH_Modeling_Guidance-2014.pdf). In general, model performance statistics for ambient concentrations of ozone and many individual species of fine particles fell within the recommended ranges. However, concentrations of

organic and elemental carbon (two particulate matter species) are over-predicted by the model and performance criteria falls outside the recommended range. Additionally, modeled particulate NO_3^- concentrations are over-predicted in the winter, and under-predicted in the summer in most locations. Model performance with respect to NH_3 can be best evaluated using the measurement data presented in this report."

Fig 8: The color scale here is a bit odd, and the gradation rather coarse, given that only one site exhibits concentrations in excess of 20 ug/m³. Could it be changed to show more of the variability at lower concentration levels?

[Response]:

Fig. 8 has been updated as below:

Line 523: Can this be explained a bit more? Did Battye 2016 find model concentrations much lower than the CAMx concentrations found here, or did they find observed NH$_3$ concentrations much higher than those reported here? Did the two studies use similar emission inventories?

[Response]:

The Battye et al., 2016 photochemical modeling effort uses the 2005 NEI projected to 2011 while our modeling efforts use the 2011 NEI. Projection methods can differ and there is no way to compare their projections to our inventories without their data. However, as an added improvement over the typical NEI treatment, our inventories were distributed spatially in Colorado such that emissions from CAFOs were allocated to the grid cell that the CAFO was located in (NEI spatial allocation for agriculture is typically done with county-level resolution, meaning that emissions are distributed homogeneous in the county in which the CAFO is located). The photochemical models used were different (they used CMAQ, we used CAMx). They did not use the CMAQ model with bi-directional flux capabilities. CAMx does not have bi-directional flux capabilities. Given that many of the measurements were the same, it would seem that the major difference between the model comparisons of the two studies is because of the inventories and models used.

Fig 6 and 8: I realize that a quantitative comparison between the CAMx estimates and the IASI columns are not possible, given the lack of an averaging kernel provided for the latter (so please don't try it); still, qualitative comparison might be useful. Even if the authors could remake Fig

8 to be on the same scale as Fig 6, and put them sideby-side, it would help show that some the features (regional max's) in the IASI data appear to correlate well with the locations of emissions and concentrations shown in Fig 8.

[Response]:

Fig.8 has been updated and merged together with Figure 6 as the new figure 6

Editorial:

19-22: This is a very long first sentence; consider breaking it up into two or more smaller ones.

[Response]:

This sentence has been changed:

"Concentrated agricultural activities and animal feeding operations in the northeastern plains of Colorado represent an important source of atmospheric ammonia (NH_3) that contributes to regional fine particle formation and to nitrogen deposition to sensitive ecosystems in Rocky Mountain National Park (RMNP) located ~80 km to the west."

This now reads:

"Concentrated agricultural activities and animal feeding operations in the northeastern plains of Colorado represent an important source of atmospheric ammonia (NH_3). The NH_3 from these sources contributes to regional fine particle formation and to nitrogen deposition to sensitive ecosystems in Rocky Mountain National Park (RMNP), located ~80 km to the west."

26: missing comma before 'with'

[Response]:

Done

40: the phrase "regional performance of each" is a bit awkward and unclear

abstract: seems a bit long, in general âAˇT can it be made a bit more concise? ˇ

[Response]:

The phrase "…providing insight into the regional performance of each…" was deleted for clarity.

In order to condense the abstract, "Seasonal changes in the steepness of the vertical concentration gradient were observed, with the sharpest gradients in cooler seasons when thermal inversions restricted vertical mixing of surface-based emissions." was removed from the abstract.

67: I'm not sure that "ag" is an appropriate abbreviation for agriculture in a manuscript text.

[Response]:

Changed

91: comma after 2010

[Response]:

Done

115: missing comma

[Response]:

Done

118: missing comma

[Response]:

Done

325: remove extra comma Fig 2 caption: clarify what is being regressed (concentration vs time, presumably)

[Response]:

Done. "The inter-annual variation of" has been add in the front of "average summertime NH_3 concentrations…"

367: It's still 2016 as I write this, so how are you citing a paper from 2017?

[Response]:

The paper was published in the first issue of Jan. 2017, and the citation was final before the end of 2016.