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 #2

This manuscript reports the results of a long-term monitoring campaign measuring ammonia using passive samplers at several sites in NE Colorado. This dataset is unique in terms of its duration, and the numbers of sites in a region of high spatial variability. In addition, a full annual record of time-integrated, vertically resolved measurements from a 300 m tower in the region are presented. The observations are compared to a new NH3 product from IASI, and to the output of a regional chemical transport model. Overall, I think this is a high quality manuscript within the scope of ACP, and it should be published after addressing the following points. Minor comments:

Section 2.2.1 – Please clarify whether Q was calculated for every hour over the sampling interval and then averaged, or was the average T and P for the interval used to calculate Q?

[Response]:

The following sentence has been added after "(http://www.radiello.com/english/nh3_en.htm):"

"Each diffusional flow rate (Q_{NH3}) was calculated for the averaged T and P for each interval sampling period."

Section 2.3 – Is it possible to give some measure of the minimum detectable level in the column from the satellite? How often is a 'successful' retrieval achieved? This becomes relevant for the discussion in Section 3.3

[Response]:

When retrieving NH_3 in the infrared (IR), the detection limit depends on the thermal contrast (TC in K), defined as the temperature difference between the surface and the air at the surface. In Figure 1, we present the detection limit (molec/cm²) as a function of TC: between -5 and 0 K, IASI can be considered as blind. Above 0 K, the detection limit is decreasing when the thermal contrast is increasing. Below -5 K, the detection limit is decreasing when the TC is becoming more negative. As shown in Figure 2, Western and Central United States is an area characterized by favorable thermal conditions for IR remote sensing with a TC of around 10K in summer (here 2015). At 5, 10 and 15 K, the detection limit at one sigma is respectively 6.3×10^{15}, 3.3×10^{15}, 2×10^{15} molec/cm².

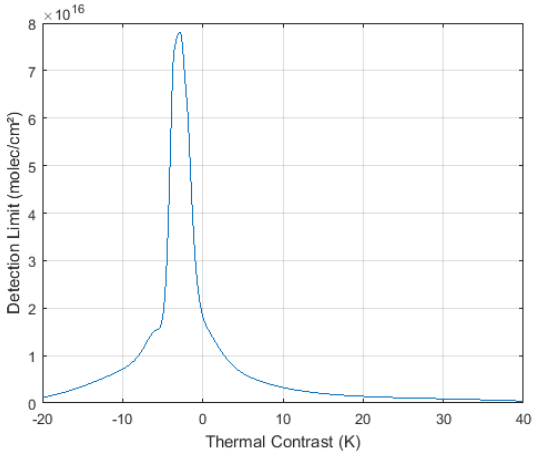


Figure 1: Detection limit (molec/cm²) of IASI NH_3 as a function of the thermal contrast (K).

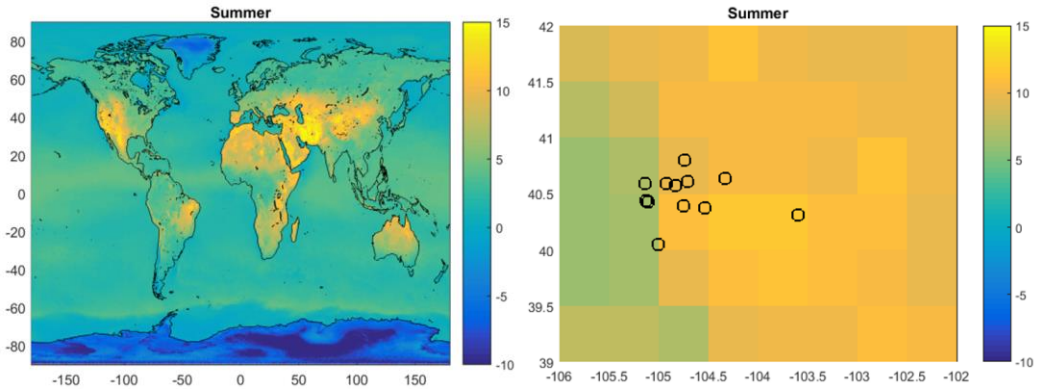


Figure 2: Mean thermal contrast (K) distribution for daytime measurements in summer 2015 (left: global; right: area of interest with the ground-based monitoring sites marked as black circle).

We added at the end of Section 2.3: "The IASI sensitivity to NH_3 is dependent on the thermal contrast (TC), defined as the temperature difference between the surface and the air at the surface. With a TC of 5, 10 and 15 K, the detection limit at one sigma is respectively 6.3×10^{15}, 3.3×10^{15} and 2×10^{15} molec/cm². In Northern Colorado, the TC during the summer period for the morning overpass of IASI is around 10 K."

Section 3.2 – The authors examine the relationship between ammonia concentration and temperature at individual heights and find minimal correlation at the lowest height, possibly due to the offsetting influences of emission rate and mixing. What about integrating the column up to 300 m and comparing the partial column integral to temperature? This would help to (partially) separating dilution and emission.

[Response]:

We appreciate the reviewer's suggestion and looked at the integrated column NH_3 up to 300 m. This partial column ammonia showed essentially the same correlation with temperature (slightly lower, actually) than did the NH_3 concentration at 300 m. Because there was no evidence that this approach performed better at separating emission from dilution effects, we propose not to make any changes to this section of the manuscript.

Section 3.3 More information should be provided about what proportion of days have successful retrievals that contribute to the satellite-derived column quantity. Are the rejected values likely to be lower, and does this result in a bias for the 'average' value reported? The left and right axes for Figure 7 seem to have been chosen to emphasize the 'agreement' between the two quantities. To some extent, that's fair but it would be more honest to at least have the zeros aligned on each side. Also, no effort is made to relate the column integral from BAO to the satellite column. This seems like a missed opportunity.

[Response]:

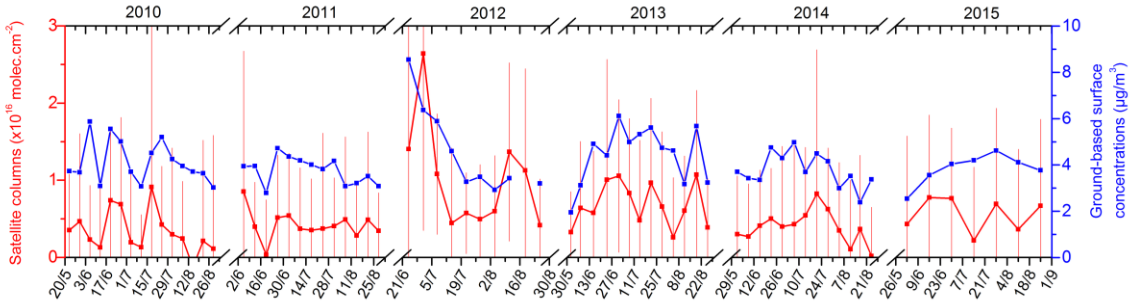
As stated at lines 445 and 446, we have used the IASI observations acquired on the same days as the ground-based measurements. For the four summers (2012-2015) considered in the domain [39-42°N; 106-102°W], complete Level 2 information (T profile, P profile, etc.) was provided for 31966 pixels (on 43125) from the morning overpass while 28125 were characterized as cloud-free observations (cloud coverage below 25%) and an NH_3 column was successfully retrieved for 98.9% of them, resulting in 27815 IASI-NH_3

observations. The post-filtering consisted of considering only IASI observations with a relative error below 100% or an absolute error below 5×10^{15} molec/cm^2, which was the case for 27713 pixels. This combined filtering using relative and absolute thresholds on the error avoids introducing a bias when averaging. In addition, 98.5% of the IASI cloud free observations were taken into account to produce Figure 6 (which is a zoom on the area [39.75-41°N; 105.5-103.5°W]).

We propose to change lines 447-450 to: "Only IASI observations with a relative error below 100% or an absolute error below 5×10^{15} molec/cm^2 were used for comparison in the latitude range from 39°N to 42°N and longitude range from 102°W to 106°W. This combined filtering using relative and absolute thresholds on the error avoids introducing a bias when averaging and results in considering 98.5 % of the IASI cloud-free morning observations for this area."

The time-series at the FC_W site (Figure 7) has been chosen to compare with IASI observations as it highlights the NH_3 transported from the High Park Fire in 2012, which was identified as an NH_3 source transported to FC_W site thanks to the satellite observations. In addition, the BAO site was only sampled *in situ* in summer 2015 (while the FC_W site was sampled during 6 summer periods). We provide below an updated Figure 7 with the zeros aligned. Because we used two separate axes, we now clearly identify in the caption that the units are different (we changed "(red)" to "(red, $\times10^{16}$ molec/cm^2)" at line 913 and "(blue)" to "(blue, µg/m^3)" at line 914).

However, we think that the first version of the figure is more illustrative of the qualitative agreement between satellite and ground-based measurements and would like to keep it as it was. If strongly preferred by the reviewer, we agree to put the version below in the final publication.



Section 3.4 The CAMx model description should be in Section 2. How are the livestock emissions used in the model different than what is estimated earlier in the paper?

[Response]:

Please refer to our previous comments (Anonymous Referee #1) for line 523. We agree with the reviewer and move the following paragraph to section 2 as "**2.4 Ammonia Modeling**"

"Chemical transport models are valuable tools for evaluating how various processes influence ambient air quality and pollutant deposition. They can be especially helpful in designing effective source control strategies for air quality improvement. Unfortunately, current models frequently have difficulties accurately simulating spatial concentrations of NH_3 (Battye et al., 2016;Adelman et al., 2015). 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). In order to examine some of these issues, NH3 measurements from this study are compared to modeled concentrations from the Comprehensive Air Quality Model with extensions (CAMx, http://www.camx.com/files/camxusersguide_v6-20.pdf). CAMx, a photochemical model that simulates the emissions, transport, chemistry and removal of chemical species in the atmosphere, is one of U.S. EPA's recommended regional chemical transport models and is frequently used for air quality analysis (EPA, 2007, 2011). The 2011 modelled period presented here (version base_2011a), including inputs representing emissions and meteorology, was developed for the Western Air Quality Data Warehouse (IWDW-WAQS, 2015); details on modeling protocol and model performance are available on the IWDW website (http://views.cira.colostate.edu/tsdw/)..."

Technical comments:

Line 67 – 'ag' should be 'agriculture'

[Response]:

Done

Lines 81-84 – are these values based on an inventory? More information would be useful beyond just a reference to the report.

[Response]:

The following sentence has been changed: "In 2002, NH_3 emissions from the Front Range were estimated to be 10288 tons/year from livestock and 5183 tons/year from fertilizer application…" The sentence now reads: "According to the 2002 Front Range NH_3 emission inventory, NH_3 emissions from the

Front Range were 10288 tons/year from livestock and 5183 tons/year from fertilizer application…"

Line 86 – 'showed that both' should be 'showed that together'

[Response]:

Done

Lines 384-385 – Isn't it really the duration of the integration period rather than the 'long time between successive passive measurements' that precludes this determination?

[Response]:

The following sentence has been changed: "The long time between successive passive measurements (1-2 weeks) in this study precludes a meaningful determination of surface removal rates based on the observed concentration gradient." This sentence now reads: "The long time duration of the integration period (1-2 weeks) in this study precludes a meaningful determination of surface removal rates based on the observed concentration gradient."

Line 935 – need to fix grammar

[Response]:

The following sentence has been changed: "Even though a one year measurement were conduct at BAO site from 12/13/2011 to 01/09/2013, the summer (06/19/2012-08/30/2012) average of NH_3 concentration were reported in Figure 1 to compare the NH_3 concentrations at other sites."

This sentence now reads: “Even though one full year of measurements was conducted at the BAO site (from 12/13/2011 to 01/09/2013), only the summer average NH_3 concentration (06/19/2012 to 08/30/2012) was reported in Figure 1 to compare with the NH_3 concentrations at other sites.”

Figure 1 – Is there much value in having a Google Earth image in the background for this figure? I just find it makes it harder to see the symbols.

[Response]:

With the Google Earth image in the background, the reader can also generally have an idea of the distribution of the agricultural area on the Front Range. If strongly preferred by the reviewer, we agree to remove the Google Earth image.

Figure 6 – I suggest making the outlines of the symbols for each site darker/thicker so that they stand out from the background color.

[Response]:

We agree with the reviewer and provide an updated Figure 6 at higher resolution with the edge of each symbol thicker. Figure 8 has been merged with Figure 6 based on the suggestion from the other referee.

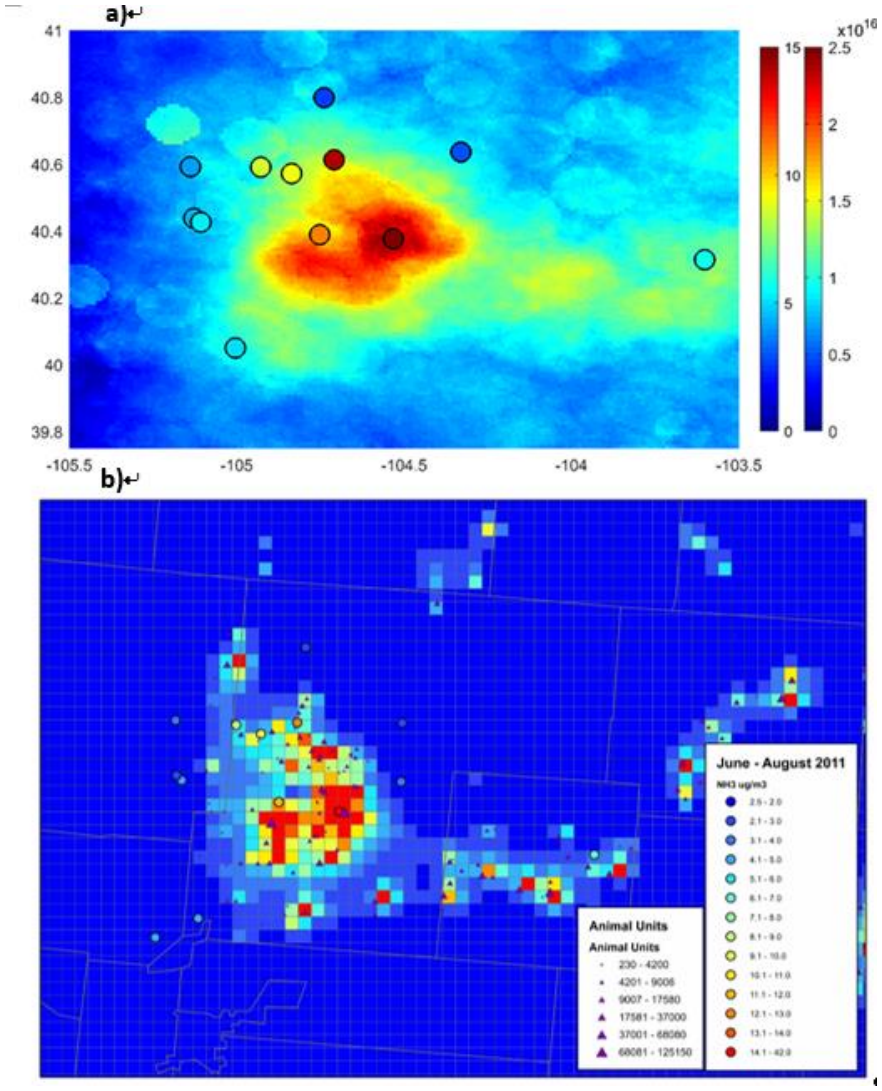


Fig. 6. Comparison of surface NH_3 concentrations with IASI satellite retrievals and CAMx model simulations. a) Radiello passive sampler surface NH_3 concentrations ($\mu g/m^3$, left color bar) plotted on top of IASI-NH_3 satellite column retrievals (molec/cm^2, right color bar), both averaged for the summers of four years (2012-2015). The BAO site was only sampled in situ in the summer of 2012. b) Comparison of measured and modeled NH_3 concentrations in the summer of 2011. The circles correspond to concentrations measured; these are superimposed on the CAMx modeled NH_3 concentration field. Animal units were indicated by the triangles.