

## Review of “Bridging the spatial gaps of the Ammonia Monitoring Network using satellite ammonia measurements”

### Summary

Multiple observations, both ground based and from satellites, strongly suggest that NH<sub>3</sub> concentrations are increasing; these increases will significantly impact ecosystems, air quality and human health. This paper presents the results of an important study that demonstrates the validity of using NH<sub>3</sub> data from IASI instruments to extend the limited information available from surface monitoring networks. This is an important capability, since surface networks are in general sparse, or in many regions, non-existent.

The authors compare IASI NH<sub>3</sub> columns with the two week surface means from the North American AMoN network and show that the IASI and AMoN data are mostly well correlated, as long as there is good temporal coverage by IASI during the two week AMoN measurement period and only IASI data within 25 km of the AMoN site are used. They then calculate trends from both datasets and show that they are comparable. Having established that IASI and AMoN data provide similar trends, they use the IASI data to calculate trends across the CONUS region, both annually and seasonally. They find that NH<sub>3</sub> is increasing faster than 10%·yr<sup>-1</sup> in the eastern U.S. and Midwest in the spring and in the western U.S. in the summer. Trends in NH<sub>3</sub> “hotspots” (e.g., central Iowa), and urban areas (e.g. New York), which are mostly far from any AMoN site, are also shown to be positive and significant.

The paper is very well organized and the results are certainly important. There are some sections that could be more clearly written, as I have detailed below. I recommend the paper be published with minor revisions.

### Presentation issues

**Section 2.2:** the authors state that they used AMoN data for all sites except UT01. This is true for the correlation analysis, but not the trend analysis, which used a much more limited set, as is explained later. This should be made clear in section 2.3.2.

**Section 2.3.1:** were the maps really created using the data for entire 2008-2018 period? Based on the discussion in section 4 it seems that they were created for each month of each year, which would be the only way to generate Figure 5. Or maybe multiple versions of the maps were created and used for different estimates? Please make this clear.

**Section 2.3.2:** the authors state that they use the Mann-Kendall test and the Theil-Sen slope estimator for trend analyses. This is a good approach, but never again do they mention either technique. The reader is left to assume that the Theil-Sen slope estimator is used in the trend analysis section, and it’s not clear where the Mann-Kendall test is used at all, since all the correlations are presented as Pearson coefficients. If the Mann-Kendall test and the Theil-Sen

slope estimator are only used in the trend section, please make this clear, and then provide a few sentences demonstrating how they are applied.

**Tables 2 and 3:** it seems that the first row captions have been switched between Tables 2 and 3. And please define pair. Does it mean all the IASI pixels co-located with an AMoN site during one two week period? It would be better to say: AMoN-IASI pair. Or add the following on line 238: ... for comparison, establishing an AMoN-IASI pair.

### **Section 3.3**

Please provide the correlations between IASI and AMoN for all four seasons and not just winter.

Please clarify how Figure 4 was obtained. The text suggest that IASI data were oversampled over the entire 2008-2018 period. I don't see how correlations can be calculated, since such oversampled would have no temporal information. Was the oversampling done on a month by month basis? This point is key for understanding how the results in section 4 were obtained.

I suggest a scatter plot of the AMoN median values and the Pearson coefficient. This would reinforce the conclusions in the paragraph starting at line 360.

The section in the last paragraph in section 3.3 starting at line 377 (However, ...) jumps ahead to discuss results in the next section. I think the authors should move this text to the next section, and use the plot I suggested above here to make the points stated at the beginning of this paragraph.

### **Section 4**

Please provide more detail on the calculation of the trends: were the trends calculated on each 2 km grid box, how long were the averaging time periods, how were the IASI trends averaged to compare with AMoN trends, how were the IASI data averaged to provide regional, hotspot and CONUS trends. This information is critical for understanding the results discussed in section 4.1 and 4.2. For example, it's hard to understand how the CONUS trend can be 3.9%/yr (line 428), 8.0%/yr (line 436) and 6.8%/yr (line 528). Either there is an error or these three values are calculated differently, but it's not clear what the difference is.

**Figure 5:** It would be interesting to show this figure for a few more sites; this would provide the reader with an idea of the variability in the slope and correlation; the sites mentioned at the end of section 3 (CA67 and CA 83) should be shown and could be discussed here.

**Figure 6:** are the IASI trends calculated in each 2 km grid box?

What is the spatial extent of each "hotspot" box?

## Minor revisions

Line 17: The limited number of NH<sub>3</sub> observations hinders ...

Line 19: ...networks are few and sparse across most of the globe,

Line 34: (2008-2018), suggesting the NH<sub>3</sub> will become a greater contributor to nitrogen deposition. NH<sub>3</sub> trends at AMoN sites are correlated with IASI NH<sub>3</sub> trends ( $r=0.6$ ), and show similar spatial patterns, with the highest increases in the Midwest and eastern U.S

Line 37: respectively. NH<sub>3</sub> hotspots (defined as regions where the IASI NH<sub>3</sub> column is larger than the 95th percentile of 11-year CONUS map,  $6.7 \text{ \AA} \sim 1015 \text{ molec/cm}^2$ ), are also experiencing increasing concentrations over time, with a median of NH<sub>3</sub> trend of  $4.7\% \cdot \text{yr}^{-1}$ .

Line 42: The increases in NH<sub>3</sub> ...

Line 43: areas, and therefore should be carefully monitored and studied.

Line 52: NH<sub>4</sub>NO<sub>3</sub>

Line 55: emissions are decreasing under new pollution controls

Line 58: in sensitive ecosystems.

Line 79: Move definition of IMPROVE to this line.

Line 85: hotspot regions

Line 94: (S-NPP) and on JPSS-1 and JPSS-2

Line 113: validation efforts were carried out in specific seasons

Line 126: in the atmosphere is limited

Line 146: in more detail.

Line 167: on bi-weekly/seasonal

Line 169: We avoided converting column NH<sub>3</sub> into surface concentrations

Line 178: sounder deployed on board

Line 183: The latest version is a reanalysis dataset that uses the European ....

Line 185: Because these meteorological data are coherent in time, the reanalyzed NH<sub>3</sub> dataset is the most appropriate one to study trends.

Line 200: imagery, we determined

Line 214: this algorithm weights IASI measurements by their uncertainties, which include varying sensitivities to thermal contrast, as described

Line 218: The sentence : “For each season, we were able to achieve sufficiently overlapped IASI pixels through calculating the sum of the unnormalized spatial response function (SRF) of the oversampling results” is not clear. Please clarify: a bit more detail might be helpful.

Line 229: Unlike simple linear regressions,

Line 236: For the initial analysis, we first used the simplest method for comparing ...

Line 237: for each each AMoN site, we average all IASI observations within a given radius of the site during the AMoN sampling time frame (2 weeks)

Line 239: between the two datasets

Line 250: and the number of IASI pixels

Line 254: or in regions

Figure 1: label each plot with the AMoN site location

Line 267: the 2-week AMoN integration period, (using a 25 km spatial window), could affect the results.

Line 276: as representative as

Line 281: for most days

Line 285: To this end, we explore the dependence of the correlation between IASI and AMoN on the IASI data temporal coverage of the 2-week sampling period and total number of IASI pixels within the 2-week AMoN sampling period, using the 25 km spatial window.

Line 288: The impact of temporal coverage and the number of IASI pixels within the sampling period are

Line 314: For each AMoN site, we repeated the two different sampling strategies 100 times

Line 361: Temporal averaging and regridding approaches, such as tessellation oversampling and physical oversampling, are common methods used to achieve higher spatial resolution

Line 364: neglect the interannual variability and calculate the multi-year averaged IASI NH<sub>3</sub> concentrations, both annual and seasonal, using the 25 km

Line 366: coverage and numbers of IASI pixels increase

Line 393: The methodology and comparison results in section 3 demonstrate that IASI NH<sub>3</sub> can be used to verify and augment regional NH<sub>3</sub> trends over the last decade. Here we will compare IASI NH<sub>3</sub> trends with the AMoN observed NH<sub>3</sub> trends in the CONUS region over the last decade.

Line 398: from Indianapolis

Figure 5: shouldn't the caption read "2008-2018 trends in monthly averaged NH<sub>3</sub>"?

Line 417: Remove the sentence starting with "The absolute" as this information is presented in the next paragraph.

Line 433: Analyzing NH<sub>3</sub> hotspots,

Line 434: indicating that the regions with the largest emissions are also seeing concentrations increasing with time.

Line 436: smaller than the trend in the CONUS median

Line 437: higher than the trend in the CONUS median

Line 438: define column-areal weighting

Line 441: see the smallest changes.

Line 456: in the eastern US

Line 457: western US and the Northeast

Line 487: add some text like: (Wang et al., 2021), **a gap that IASI data can fill.**

Line 488: the cumulative distribution of the CONUS population as a function of the distance from an AMoN site.

Line 493: mobile sources and trends in population centers.

Line 502: in total account for more than seventy million people

Figure 9: Cumulative distribution of CONUS population as a function of distance from an AMoN site.

Line 517: The temporal coverage of the IASI data during the two week AMoN sampling period is the controlling factor of the correlation between the IASI and AMoN measurements, presumably because of the large day-to-day variability of NH<sub>3</sub>.

Line 521: the IASI NH<sub>3</sub> product

Line 522: shown the unique role

Line 530: as well as similar spatial patterns.

Line 531: show the largest increases in the Midwest and eastern U.S., with a moderate correlation between the IASI and AMoN trends for the entire CONUS .

Line 535: deposition in most regions in the U.S.;

Line 536: (2016), which will have adverse impacts

Line 547: communities with limited resources

Line 540: (cropland dominated) and in summer in the western U.S. (feedlot dominated), highlighting the impacts

Line 543: for characterizing NH<sub>3</sub> magnitude