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Validation of NH_3 observations from AIRS and CrIS against aircraft measurements from DISCOVER-AQ and a surface network in the Magic Valley

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Abstract.

Ammonia is a significant precursor of $PM_{2.5}$ particles and thus contributes to poor air quality in many regions. Furthermore, ammonia concentrations are rising due to the increase of large scale, intensive agricultural activities, which are often accompanied by greater use of fertilizers and concentrated animal feedlots. Ammonia is highly reactive, and thus highly variable and difficult to measure. Satellite based instruments, such as the Atmospheric Infrared Sounder (AIRS), and the Cross-Track Infrared Sounder (CrIS) sensors, have been shown to provide much greater temporal and spatial coverage of ammonia distribution and variability than is possible with in situ networks or aircraft campaigns, but the validation of these data is limited.

Here we evaluate ammonia retrievals from AIRS and CrIS against ammonia measurements from aircraft in the California Central Valley and in the Colorado Front Range. The satellite datasets were small and in California were obtained under difficult conditions. We show that the surface values of the retrieved profiles are biased very low in California and slightly high in Colorado, and that the bias appears to be primarily due to smoothing error. We also compare three years of CrIS ammonia against an in situ network in the Magic Valley in Idaho We show that CrIS ammonia

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captures both the seasonal signal and the spatial variability in the Magic Valley, though it is biased low here also. In summary, analysis adds to the validation record but also points to the need for more validation under different conditions.

1. Introduction

Ammonia (NH_3) is one of the most common forms of reactive nitrogen and the primary alkaline gas in the atmosphere. Intended and unintended releases of NH_3 into the environment over the last century have significantly altered the natural nitrogen cycle (Erisman et al., 2008), so that the current emission levels of ammonia are about four times higher than in previous centuries (Battye et al., 2017). The main sources of NH_3 are agricultural emissions, namely from livestock raising and fertilizer application (EDGAR-Emission Database for Global Atmospheric Research, 2014), which account for 80% of all emissions globally (Sutton et al., 2013; Behera et al., 2013). There are also some locally or seasonally significant sources NH_3, the most notable being biomass burning events, which can generate large amounts of NH_3 (Coheur et al., 2009; Whitburn et al., 2015, 2016). In urban areas automobiles with three-way catalytic converters (Sun et al., 2017) can be a major source of NH_3. Nowak et al. (2012) estimate that in the Los Angeles basin cars contribute as much as 50% of the total NH_3 emissions.

Ammonia is the dominant base in the atmosphere, and it plays a significant role in the formation of fine particulate matter ($PM_{2.5}$) (e.g., Aneja et al., 2003), which can penetrate deep into the lungs and severely impact the respiratory and circulatory systems (Pope et al., 2009). Paulot and Jacob (2014) have shown that the costs associated with the health impacts of NH_3 associated with food production for export in the US offset half the revenue from these exports. Long-term exposure to ambient $PM_{2.5}$ is the leading environmental risk factor for premature mortality worldwide, leading to an estimated 2.5–3.4 million premature deaths annually (Cohen et al.,

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2017), 20% of which is estimated to stem from NH_3 emissions (Lelieveld et al., 2015). Ammonia is not yet a criteria pollutant in the US (its emissions are regulated by the European Union (EU) and it is a criteria pollutant in Canada), but the EPA has published established regulations (http://www.nsrlaw.com/single-post/2017/06/19/EPA-NSR-Chief-Outlines-NSR-Changes-at-2017-AWMA-Conference) mandating that every state must set area specific significant emission rates (SERs) for NH_3. Since the emissions of NH_3 are a key factor in the formation of $PM_{2.5}$, reducing emissions can be an effective path to reduce air pollution (Liu et al., 2021).

Given the rapid growth of industrial-scale agriculture (e.g., increase in egg, milk and meat consumption), especially in Asia (e.g., Xu et al., 2016), NH_3 emissions are projected to increase greatly over the next few decades in many parts of the world. The reduction of NO_x emissions due to more stringent controls will reduce the contribution of NO_x to the deposition of reactive nitrogen, but Paulot et al. (2013) suggest that an increase in NH_3 emissions will likely compensate for this reduction. NH_3 and its derivatives are also quickly deposited in the ecosystems, increasing their eutrophication and reducing biodiversity (Erisman et al., 2008).

There is thus growing recognition that NH_3 is an important pollutant, and that it will likely play a greater role in air quality and ecosystem health over the next decades, due to both the essential role NH_3 plays in feeding the world's population, and to the fact that the atmospheric emission potential of NH_3 is directly linked to increasing temperatures (Skjøth et al., 2013; Sutton et al., 2013). However, in situ measurements remain a challenge. NH_3 is easy to detect, but it is hard to measure accurately. There are many in situ techniques used to detect atmospheric NH_3 with varying time resolution and precision, but the main issue affecting precision is the inlet rather than the instrument. NH_3 is sticky, and so it is challenging to get it into a given instrument quantitatively and quickly (Roscioli et al., 2016), (Pollack et al., 2019). This feature is critical for

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characterizing the abundance of NH_3 in the background atmosphere, for making measurements of NH_3 fluxes, and deploying instruments on aircraft. New open-path sensors avoid this issue, but they cannot be deployed in many situations. Consequently, the emissions of NH_3 outside of a limited set of well-instrumented locations remain poorly constrained, reducing the accuracy with which models can represent concentrations and variability. The high spatial and temporal variability of NH_3 exacerbates the lack of continuous, spatially well sampled data over extensive regions. This also contributes to bottom-up inventories often underestimating emissions due to scaling difficulties (Nowak et al., 2012).

Satellite data, even though they come with their own uncertainties, provide by virtue of their spatial and temporal density, another option for quantifying these emissions. Currently there are multiple NH_3 datasets, with varying data record lengths and spatial coverage, obtained from the following instruments: the three Infrared Atmospheric Sounding Interferometer (IASI) instruments flying in a 9:30 am orbit, the Greenhouse Gases Observing Satellite (GOSAT) in a 1:30 pm orbit, along with the Tropospheric Emission Spectrometer (TES), the Atmospheric Infrared Sounder (AIRS), and the three Cross-Track Infrared Sounder (CrIS) instruments, all flying in a 1:30 pm orbit. The data obtained from these instruments has had numerous applications. Multiple papers (Van Damme et al., 2015; Shephard et al., 2011; Shephard and Cady-Pereira, 2015; Warner et al.; 2016; Shephard et al., 2020, Wang et al., 2021) have shown that NH_3 measurements from infrared sensors capture NH_3 hotspots, such as the Indo-Gangetic plain, eastern China and the American Midwest, as well as the expected regional seasonal variability and fire activity. Warner et al. (2017) used retrievals from AIRS to show definite positive trends in NH_3 concentrations over the US, the EU and China, which the authors ascribe to declines in SO_2 and NO_2 emissions in all three regions due to more stringent controls. Van

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Damme et al. (2018) used nearly a decade of IASI data to show that the emissions listed in the EDGAR (EDGAR, 2016) inventory for large source regions were wrong by as much as a factor of three; furthermore, emissions from smaller sources were often underestimated by an order of magnitude. Dammers et al. (2019) found similar results using CrIS and IASI NH_3 data. Zhu et al. (2013) have demonstrated that Tropospheric Emission Spectrometer (TES) NH_3 data over North America in the 2006–2009 period, though relatively sparse, could be used in an inverse modeling framework to constrain emissions sufficiently to improve agreement between GEOS-Chem output and surface measurements from the National Atmospheric Deposition Program (NADP) Ammonia Monitoring Network (AMoN) network. Using NH_3 measurements from CrIS and NO_2 measurements from TROPOMI, Cao et al. (2022) demonstrated that NH_3 emissions decreased substantially over downtown Los Angeles during the 2019 March COVID-19 lockdown; this result is in agreement with the conclusion from Nowak et al. (2012) that in urban areas traffic can be a major source of NH_3 and consequently greatly increase exposure to $PM_{2.5}$.

Yet in spite of the increasing use of NH_3 data from space-based instruments, validation of these data remains rather limited. Sun et al. (2015) compared a small set of NH_3 total columns from the TES instrument against columns derived from surface and aircraft measurements during the NASA Deriving Information on Surface conditions from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ) California 2013 campaign, and found small differences (less than 6%) and high correlation (R=0.82); however, note that TES, which is no longer operational, had much higher spectral resolution (0.06 cm^{-1}) and thus greater sensitivity to surface NH_3 and less interference from water vapor than the infrared sensors (AIRS, CrIS, IASI) currently providing data for NH_3 retrievals. Shephard et al. (2015) compared TES profiles against aircraft measurements taken during the 2013 Joint Canada–Alberta

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Implementation Plan for Oil Sands Monitoring (JOSM) campaign and showed that the TES profiles were unbiased. Warner et al. (2016) compared four NH_3 retrievals from AIRS against aircraft profiles obtained during DISCOVER-AQ California and found good qualitative agreement. Dammers et al. (2017) compared 218 IASI and CrIS total columns and CrIS profiles against corresponding data from ground based Fourier Transform Infrared (FTIR) observations at seven FTIR sites in the Network for the Detection of Atmospheric Composition Change (NDACC): overall the FTIR and CrIS data were well correlated (R=0.77) and mainly unbiased. Most of these FTIR stations in this analysis were located in high source regions, which somewhat limits the applicability of these results to the regions of greatest interest to the air quality community.

Van Damme et al. (2015) carried out what is likely the most extensive evaluation of NH_3 measured from space, comparing IASI NH_3 against data from six different monitoring networks in North America, Europe, Africa and China and from the California Research at the Nexus of Air Quality and Climate Change (CalNex) campaign in California. Most of the data from the surface networks were provided on bi-weekly or monthly scales: when IASI columns were converted to surface concentrations and averaged over the corresponding time period, they showed qualitative agreement in space and time with the surface data: the correlations in general were not high, though still significant. Recently Guo et al. (2021) (hereafter Guo2021) compared NH_3 columns from IASI with integrated profiles obtained from aircraft data during the Colorado 2014 DISCOVER-AQ campaign: the IASI columns were unbiased and significantly correlated. However, Guo2021 point out that the instruments currently used to measure NH_3 from aircraft have large uncertainties due to limited accuracy and slow response to changing NH_3 concentrations.

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To a varying degree all the studies above cite the same factors that complicate the validation of satellite NH_3 products:

- Sub-pixel inhomogeneity due to the high spatial-temporal variability of NH_3 driven by its short lifetime; thus the point data from an in situ instrument will only be partially correlated with the pixel scale data obtained from a satellite instrument
- Different measurement time scales (weeks or days vs instantaneous), especially for surface networks
- High instrument noise
- Validation results are strongly influenced by local atmospheric conditions and the vertical distribution of NH_3, which highlights the need for further validation campaigns under diverse conditions

Our objective is to add to the validation record at the single pixel scale with retrievals from L1B radiances from both the AIRS and CrIS instruments. The retrieved profiles here are obtained with the MUlti-SpEctra, MUlti-SpEcies, MUlti-Sensors (MUSES) (Fu et al., 2013, 2016, 2018) algorithm. which provides profiles, total columns and uncertainty estimates, all of which can also be evaluated against in situ data. AIRS and CrIS NH_3 will be compared against aircraft data from the P-3B aircraft flown during DISCOVER-AQ campaigns in California and Colorado. Warner et al. (2016) also compared AIRS with DISCOVER-AQ, but their retrievals used cloud cleared radiances and covered nine AIRS pixels (~45 km footprints at nadir). Using single pixel radiances provides several advantages over "cloud cleared radiances": the propagation of uncertainties from the radiances is simpler (see section 2.2) and the retrieved information is obtained on smaller scales, which is important for NH_3 (see section 5). This will be the first comparison of single pixel NH_3 profiles from either AIRS or CrIS against aircraft data. While aircraft campaigns can provide snapshots in time, they do not provide sustained measurements

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over strong point sources. In order to test the capability of MUSES NH_3 to capture temporal and spatial variability over an extended period, surface level CrIS NH_3 concentrations will also be evaluated against three years of data from a small monitoring network in the Magic Valley in Idaho. Section 2 will briefly review the NH_3 retrieval algorithm, section 3 will give an overview of the instruments, section 4 will present the analysis of the DISCOVER-AQ data and section 5 will follow with the analysis of the Magic Valley data; finally, section 6 will summarize our conclusions and discuss future work.

2. MUSES NH_3 Retrieval Algorithm

The first nadir retrievals of NH_3, obtained using a prototype retrieval with data from the TES instrument (Beer et al., 2008), exploited the NH_3 ν_2 vibrational band between 960 and 970 cm^{-1} to demonstrate that TES could measure the variability in NH_3 along a transect in China. Shephard et al. (2011) implemented a full optimal estimation (OE) approach (Rodgers, 2000), which sought to reduce the difference between the measured CrIS radiances in ν_2 band and the calculated radiances from a radiative transfer model (Moncet et al., 2008). Before the OE algorithm was run an a priori profile was chosen from one of three possible profiles, representing background, moderate and enhanced NH_3 concentrations. These profiles were derived by binning global distributions of NH_3 (Shephard et al., 2011) from the chemical transport model GEOS-Chem (Zhu et al., 2013). The profile is selected by applying an online/offline brightness temperature (BT) difference test centered around the 967 cm^{-1} line. The OE algorithm is then run as a refinement step, in which the a priori and the initial guess profiles are identical except for the background profile, for which the moderate profile is chosen as the initial guess, in order provide Jacobians with some sensitivity.

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The algorithm developed for the TES NH_3 retrievals has since been adapted with minor changes for CrIS (Shephard and Cady-Pereira, 2015; Shephard et al., 2020) and AIRS (this paper). The spectral retrieval window and the frequencies for the online/offline BT test were slightly modified for the CrIS and AIRS spectral resolutions, and a preliminary retrieval step to adjust the surface temperature and emissivity was introduced. This algorithm forms the core of the NH_3 component of the MUSES software used here and also of the CrIS Fast Physical Retrieval code, whose product has been used in a number of previous studies (e.g., Shephard and Cady-Pereira, 2015; Shephard et al., 2020; Dammers et al. (2017); Cao et al. (2022), Marais et al. (2021)). The two products have much in common (the same spectral microwindows, a priori selection, constraint matrices and forward model), but obtain temperature and water profiles and surface properties from different sources and use different software to carry out the optimal estimation. Preliminary comparisons have shown excellent agreement between the two algorithms, but a full comparison is beyond the scope of this paper, as the objective here is the validation of the MUSES AIRS and CrIS NH_3 retrievals.

AIRS and CrIS NH_3 retrievals were both obtained by applying the MUSES algorithm to AIRS or CrIS L1B single footprint radiances. The MUSES algorithm is an end-to-end optimal estimation process that provides a complete characterization of the parameters involved in the radiative transfer processes in the infrared region, using a multi-step approach. Before the NH_3 retrieval step is reached, the atmosphere has been well characterized by the previous retrieval steps: temperature and water vapor profiles, surface properties and cloud absorption are known and can be accounted for in the NH_3 retrieval, significantly reducing errors from radiatively interfering species. Since cloud optical depth is retrieved, cloud clearing algorithms (Susskind et al., 2003) are not needed and retrievals can be performed on every pixel, or field of view (FOV), rather

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than on the nine pixel field of regard (FOR). This allows for retrievals from AIRS with a 15 km rather than 45 km minimum footprint, which was the resolution for the earlier NH_3 retrievals from AIRS obtained by Warner et al. (2016) using cloud cleared radiances.

MUSES uses the Optimal Spectral Sampling (OSS) model (Moncet et al., 2008) as its forward model; OSS is a fast and accurate radiative transfer method designed specifically for the modeling of radiances measured by sounding radiometers in the infrared, although it is applicable throughout the microwave, visible, and ultraviolet regions.

Since the retrieval is non-linear, an a priori constraint is used for estimating the true state (Bowman et al., 2006). If the estimated (retrieved) state is close to the actual state, then the estimated state can be expressed in terms of the actual state through the linear retrieval (Rodgers, 2000):

$$\hat{\boldsymbol{x}} = \boldsymbol{x}_a + \mathbf{A}(\boldsymbol{x} - \boldsymbol{x}_a) + \mathbf{G}\boldsymbol{n} + \mathbf{G}\mathbf{K}_b(\boldsymbol{b} - \boldsymbol{b}_a) \quad (1)$$

where $\hat{x}$, x_a, and x are the retrieved, a priori, and the "true" state vectors respectively, **G** is the gain matrix, $\boldsymbol{b}$ is the vector that contains parameters not retrieved in the current step and $\boldsymbol{b_a}$ the a priori values for these parameters if they are retrieved in another step.

The averaging kernel, **A**, describes the sensitivity of the retrieval to the true state:

$$\mathbf{A} = \frac{\partial \hat{\boldsymbol{x}}}{\partial \boldsymbol{x}} = (\mathbf{K}^T\mathbf{S}_n^{-1}\mathbf{K} + \mathbf{S}_a^{-1})^{-1}\mathbf{K}^T\mathbf{S}_n^{-1}\mathbf{K} = \mathbf{G}\mathbf{K} \quad (2)$$

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where S_n is the instrument noise covariance matrix, and S_a is the a priori covariance matrix for the retrieval. The Jacobian, **K**, is the sensitivity of the forward model radiances to the true state vector, $\boldsymbol{K = \partial L / \partial x}$ .The rows of **A** are functions with a finite width corresponding to the vertical resolution of the retrieved parameter. The sum of each row of **A** provides a measure of retrieval information that comes from the measurement (Rodgers, 2000) at the corresponding altitude, provided that the retrieval is quasi-linear. The trace of the averaging kernel matrix gives the number of degrees of freedom for signal (DOFS) from the retrieval.

The total a posteriori error covariance matrix S_x for a given retrieved parameter $\hat{x}$ is given by:

$$\boldsymbol{S_x = (A - I)S_a(A - I)^T + GS_nG^T + GK_bS_b(GK_b)^T} \quad (3)$$

where $\mathbf{S_b}$ is the expected covariance of the non-retrieved parameters. The total error (or uncertainty) for a retrieved profile is expressed as the sum of: i) the smoothing errors (first term on the right-hand-side), i.e. the uncertainty due to unresolved fine structure in the profile; ii) the measurement errors (second term) originating from random noise in the spectrum; and iii) the systematic errors (last term) due to uncertainties in the forward model parameters not retrieved in the NH_3 step, some of which are constant and some of which change from retrieval-to-retrieval (Worden et al., 2006). For the current CrIS NH_3 algorithm this last term is not calculated and is not included in the total error estimate. By providing the expected error covariance and the averaging kernels, this approach facilitates the use of the retrieved profiles in inverse modeling efforts, since both terms are used to weight the information coming from the retrieval. The error covariance gives users an uncertainty estimate for each retrieved profile, which can be utilized to screen the data or be included in a statistical analysis. Furthermore, the estimated uncertainty

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derived from the error covariance can be compared to measured uncertainties, obtained by calculating the spread of the differences between satellite and in situ data, as will be shown in sections 4.1 and 4.2; this analysis can indicate if there are error terms missing from the optimal estimation formulation.

Rodgers and Connor (2003) presented a method for comparing satellite profiles of trace gases with limited vertical resolution with in situ profiles obtained on a much finer grid. This approach is often described as "applying the instrument operator" or "applying the averaging kernel". It attempts to estimate how the space based instrument would "see" an in situ profile by applying the equation below to the in situ data:

$$\mathbf{X} = \mathbf{X}_{apriori} + \mathbf{A} \left(\mathbf{X}_{aircraft} - \mathbf{X}_{apriori} \right) \quad (4)$$

The estimated profile **X** has been smoothed by the operator, simulating the smoothing due to the coarser resolution of the satellite observation. When X is compared to the satellite observations it is assumed that the smoothing error has been accounted for and can be ignored, which is not the case when satellite observations are compared directly to measured profiles; the remaining errors will be due to instrument noise and temporal and spatial sampling differences; the latter can be especially large for NH_3, due to its large variability, as was discussed above. While we present comparisons using the Rodgers and Connor approach, we will also show simple differences between the aircraft and satellite data; there are many end-users who will want to use the data as is in their own analysis and will want to know the corresponding uncertainties.

A note on applying the operator: AIRS and CrIS profiles extend to the top of the atmosphere, while the aircraft profiles used here rarely go above 700 hPa in California and 500 hPa in Colorado. We have therefore truncated the averaging kernels to these altitude ranges.

3. Data

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3.1 Aircraft Data

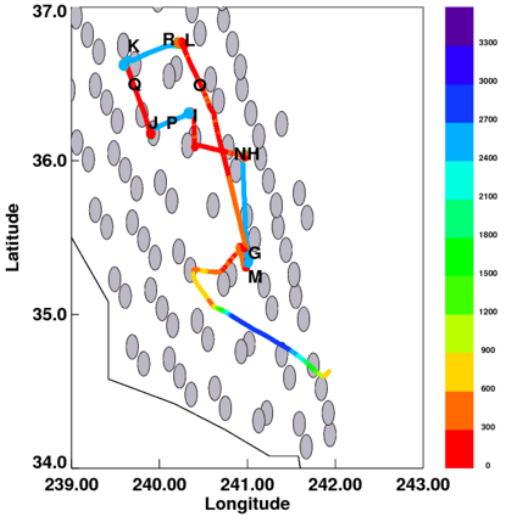

Figure 1: Sample aircraft track during DISCOVER-AQ in the Central Valley, CA; colors indicate altitude, letters locations of aircraft spirals; CrIS pixels are shown as grey ellipses

Vertical profiles of NH_3 were measured by the PTR-MS instrument (Muller et al. 2014) deployed aboard the NASA P3B aircraft during the NASA DISCOVER-AQ (Crawford and Pickering, 2014) campaigns in California in 2013 and Colorado in 2014. These campaigns were designed to validate collocated satellite observations of atmospheric pollutants over four regions in the United States: (Maryland/Washington D.C, California, Texas and Colorado), but NH_3 was only measured in California and Colorado. The P3B flight pattern was specifically designed for satellite validation: the aircraft flew repeated upward and downward spirals, typically 5 km wide connected by transects. An example trajectory, overlaid on the locations of CrIS NH_3 retrievals, is shown in Figure 1: the aircraft altitude is indicated by the colors, and the locations of the spirals are marked with letters. The PTR-MS instrument samples the atmosphere at 1Hz and the data were binned over 100 m to reduce noise. The estimated instrument uncertainty is 35% (Müller et al., 2014). Note that PTR-MS NH_3 data were a side product of the PTR-MS measurements during DISCOVER-AQ, which were designed to obtain data on volatile organic compounds (VOCs); this resulted in a higher detection limit and a slower response; the latter drawback leads to larger uncertainties when the NH_3 amounts are changing rapidly, as is the case when the aircraft is

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leaving the boundary layer on upward spirals and entering the boundary layer on downward spirals.

3.2 Ground Data

The USDA-ARS Northwest Irrigation and Soils Research Laboratory established a regional NH_3 monitoring network in the Magic Valley region of south-central Idaho, USA utilizing the NADP AMoN network technology and protocols, but with much greater spatial density.The network measured ambient NH_3 concentrations along two transects of the Magic Valley (North-South) and West-East) utilizing passive diffusive NH_3 samplers collected on a bi-weekly basis from February of 2018 through December of 2020. The objective of the project was to determine the spatial variability of ambient NH_3 concentrations across the region, which is dominated by agricultural production and high-density dairy operations, to better understand the potential for NH_3 transport within and downwind of the region.

3.3 Satellite data

3.3.1 AIRS single pixel NH_3 retrievals

AIRS is a nadir-viewing, scanning thermal infrared (TIR) spectrometer launched on board the Aqua satellite on May 4, 2002, into a sun synchronous polar orbit at an altitude of 705 km with a 1:30 am local solar time (LST) Equator in the ascending node and 1:30 pm (LST) in the descending node (Aumann et al., 2003). The daytime overpass is an ideal time for NH_3 retrievals, as thermal contrast is high and emissions are usually peaking, driven by higher temperatures. AIRS measures the thermal radiance between 3–12 microns with a spectral resolution of ~ 0.75 cm^{-1} and a noise level of ~ 0.15K at 270 K (Zavyalov et al., 2013) in the 970 cm^{-1} NH_3 absorption window. A single

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AIRS FOV has a circular footprint with ~15 km diameter at nadir and the AIRS swath width is ~1650, km which enables near global coverage twice daily.

3.3.2 CrIS single pixel NH_3 retrievals

CrIS is a Fourier Transform Infrared Radiometer (FTIR) launched on the Suomi National Polar Orbiter Preparatory (SNPP) platform in October 2011 and on the Joint Polar Satellite System (JPSS-1) in November 2017, into sun-synchronous orbits (824 km altitude) with the same LST crossing times as AIRS; CrIS is a cross track scanning instrument with a 2200 km swath width (+/- 50 degrees) with 14 km circular pixels (at nadir), a spectral resolution of 0.625 cm^{-1} and low spectral noise (~0.04 K at 270K) (Zavyalov et al., 2013) in the NH_3 spectral window. CrIS also provides twice daily global coverage. Note that in this paper only CrIS data from the SNPP platform have been used. There is a data gap in the SNPP-CrIS record between March and June 2019, corresponding to a malfunction in the electronics during that time.

4. DISCOVER-AQ Analysis

The DISCOVER-AQ campaigns in California and Colorado provide the most comprehensive set of in situ NH_3 profile data (as opposed to retrievals from FTIR instruments) available. Though limited to short (two month) periods in two strong source regions, these datasets can be used to demonstrate the strengths and limitations of satellite data; additionally, they allow for the evaluation of the accuracy of the retrieval estimated error, as calculated from Equation 3. During each campaign the aircraft flew multiple up and down spirals. The satellite profiles were co-located with aircraft profiles taken within one hour and 15 km of the satellite retrievals, following the approach described in Guo2021. This co-location criterion is much stricter than is usual for satellite validation (see Hegarty et al., 2022 for an example with CO retrievals from AIRS) but is necessary given the variability in NH_3 (Guo2021). In consequence, each CrIS or

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365 AIRS profile was compared with data from at most two spirals. Retrievals were checked for quality by examining the root mean square (RMS) of the residuals and the retrieved cloud optical depth. Comparing the aircraft and satellite profiles requires regridding the aircraft data to the satellite vertical grid. This was accomplished by defining layers centered around each AIRS/CrIS level, then finding the median value of the aircraft profile in each layer. The lowest layer
370 extended from the surface to the mid-point between the surface and the first AIRS/CrIS level above the surface.

While both AIRS and CrIS have the same equatorial crossing time (~ 1:30pm local solar time), this does not imply that both instruments are observing the same location at the same time at the same angle. For example, on January 22, 2013, CrIS flew almost directly over the Central Valley
375 around 2100 UTC, while AIRS had its closest observation 15 degrees to the west around 2200 UTC, near the edge of its swath. Therefore, the set of aircraft profiles co-located with each instrument is not identical.

4.1 California

DISCOVER-AQ in California took place during January and February of 2013. The Central
380 Valley is one of the strongest NH_3 source regions in North America (e.g., Clarisse et al., 2009, Shephard et al., 2020), and this was reflected in the aircraft data, which registered near surface amounts as high as 100 ppbv (Figure 2, upper left panel). However, the observing conditions were not ideal for satellite infrared retrievals, as there were thermal inversions over the entire period (Figure 2, upper right). Inversions lead to increased uncertainties in the retrieval, as they
385 effectively create an emission layer above the surface, i.e., a layer that is warmer than the surface and therefore emits more than it absorbs. Inversions also limit the vertical extent of the boundary layer, with consequently lower NH_3 concentrations at altitudes where the retrieval has greater

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sensitivity. Nevertheless, evaluating the AIRS and CrIS NH_3 profiles against the aircraft data is still a useful exercise, as the combination of inversions and strong sources is not a unique occurrence, and this analysis demonstrates the limitations of retrievals under these conditions. However, when averages over long periods and/or broad regions are desired, it would be reasonable to exclude cases with inversions.

43 AIRS and 58 CrIS profiles met the co-location criteria described above. When compared directly against the aircraft profiles (Figure 3, top panels), both instruments showed large negative biases near the surface, as low as -80 ppbv for AIRS and -100 ppbv for CrIS, while the average bias at this level was ~-38 ppbv for AIRS and ~44 ppbv for CrIS, with a spread of ~ 24 ppbv and 38 ppbv, respectively (Table I). This large negative bias is likely due to a combination of sub-pixel heterogeneity (Sun et al., 2015; Kille et al., 2019), the inherent difficulties of carrying out retrievals over thermal inversions, and smoothing errors. Clouds are accounted for in the MUSES retrieval (Kulawik et al., 2006) and thus cloudy conditions should not significantly impact the NH_3 retrievals. Both the bias and the spread drop significantly with increasing altitude, as do the measured concentrations; while the biases decrease, they become positive and are not insignificant at 825 hPa (~30% at 825 hPa), and the spread is quite large (~100%) suggesting that at this altitude some of retrievals are overestimating the NH_3 concentrations with respect to the aircraft. The mean CrIS value at the surface (~16 ppbv) is 60% greater than the AIRS value (~10 ppbv); the lower CrIS noise level (0.04) vs AIRS (0.15) could allow for greater sensitivity to surface changes, in spite of the inversions.

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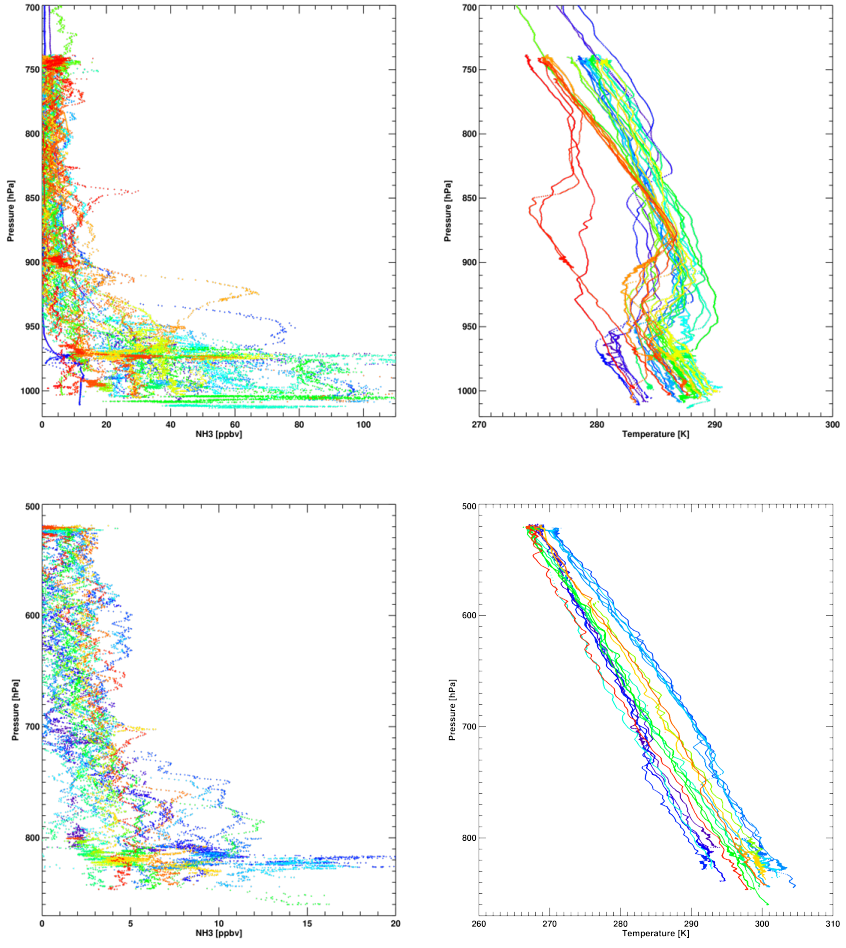

410 *Figure 2: NH_3 profiles (left) obtained from aircraft during DISCOVER-AQ in California (top) and Colorado (bottom); only profiles co-located with CrIS data are shown; corresponding temperature profiles (right)*

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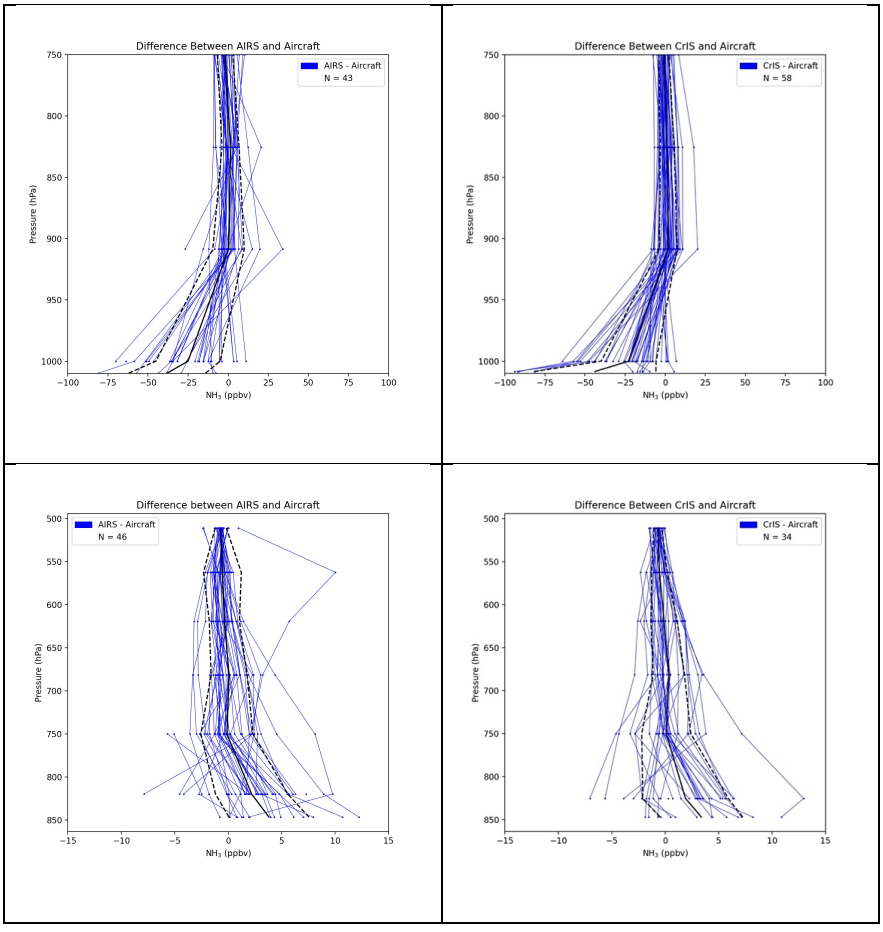

Figure 3: Difference between AIRS and aircraft NH_3 profiles (left) and CrIS and aircraft (right) during DISCOVER-AQ in California (top) and Colorado (bottom); solid line indicates mean bias and dashed lines standard deviations.

The sum of the rows of the averaging kernels (SRAK) (Figure 4, top two panels), which provides an estimate of the retrieved information at each level originating from the measurement rather than from the a priori, shows for both AIRS and CrIS that while the information from the data peaks just below 700 hPa, the data also significantly contribute to the retrieved surface values. The maximum SRAK (~1.2-1.3) is similar for both sensors, as is the SRAK at the surface (~0.6).

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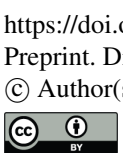

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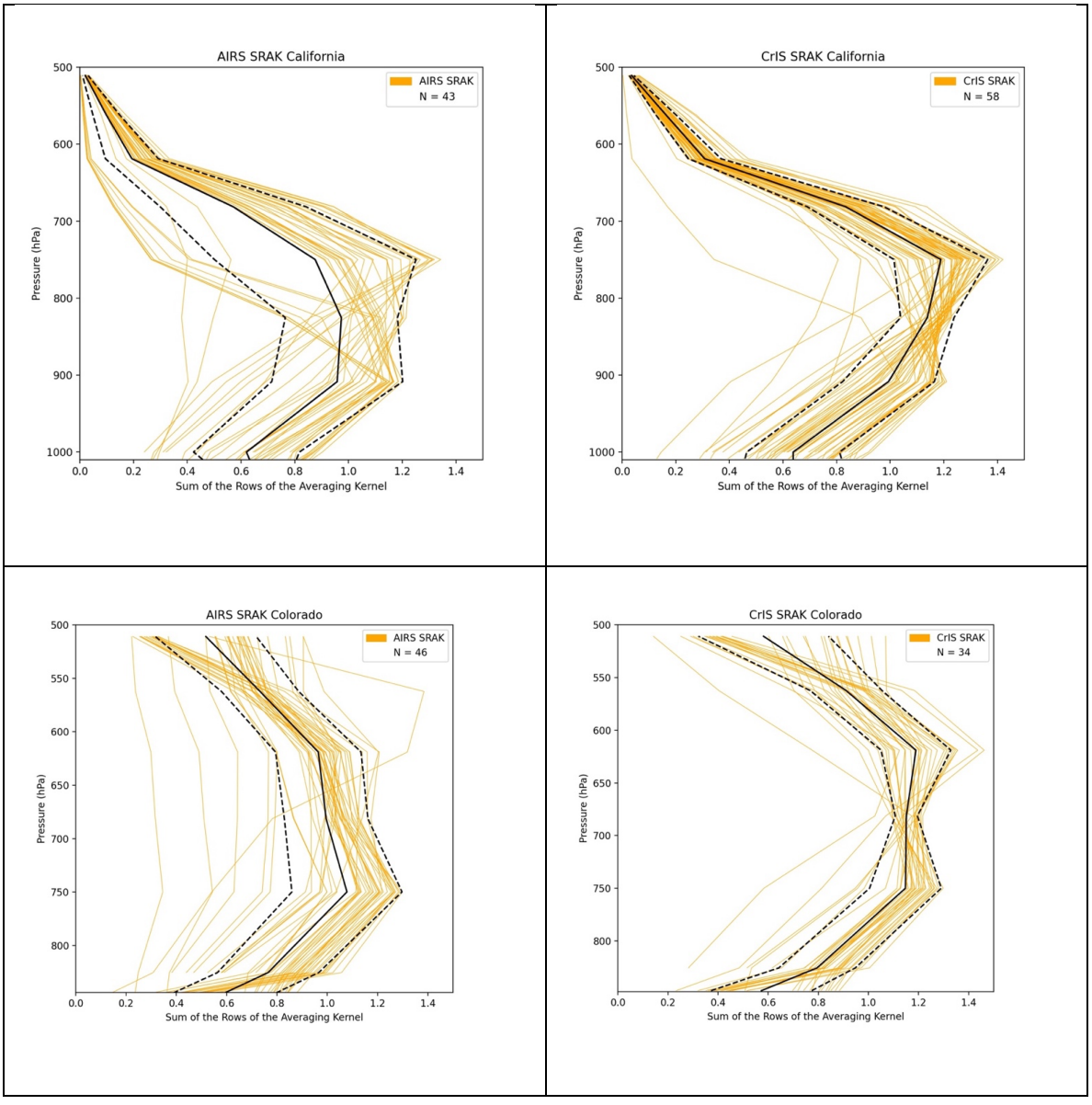

Figure 4: Sum of the row of the averaging kernels (SRAK) for AIRS (left panels) and CrIS (ritght panesls);California (upper panels) and Colorado (lower panels)

However, there is much greater variability in the AIRS SRAK, possibly due to higher instrument noise. Applying the instrument operators following equation 4 (Figure 5 (top panel), red and black curves) reduces the bias at all levels to close to or less than 1.0 ppbv (Table 1, top section)

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and dramatically reduces the spread at lower levels. These results indicate that the retrievals do not have significant systematic biases and that the bias in the direct comparisons originates from the a priori. If instead the a priori profiles are compared with the aircraft data, we see a large

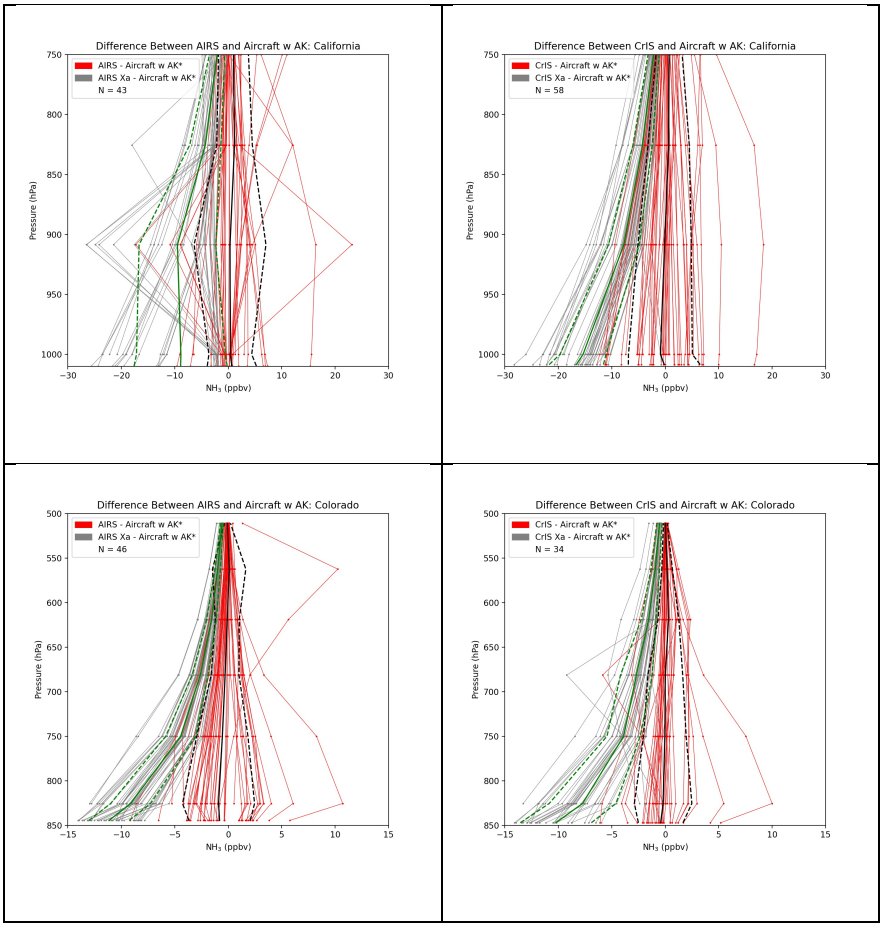

Figure 5: Difference between AIRS and aircraft NH3 profiles (left) and CrIS and aircraft (right) during DISCOVER-AQ in California (top) and Colorado (bottom); the averaging kernel has been applied to the aircraft data; red lines show differences between the retrieved profiles and the aircraft data, black solid lines indicate mean bias and black dashed lines standard deviations; grey lines show differences between the a priori profiles and the aircraft, green solid lines indicate mean bias and green dashed lines standard deviations.

negative bias near the surface (~-9 ppbv for AIRS and ~-18 ppbv for CrIS). It has been argued that the a priori selection for these retrievals already uses all the information available in the

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radiance data; this result demonstrates that the retrieval process does add significant information and reduces the a priori error.

The average estimated uncertainties from equation 3 (Figure 6, top panels, blue curves) are significantly lower than the measured uncertainties (the fractional standard deviation derived from the standard deviations of the differences in the "Satellite-Aircraft with AK" column in Table 1) (Figure 6, top panels, black curves) for both AIRS and CrIS, indicating that some error

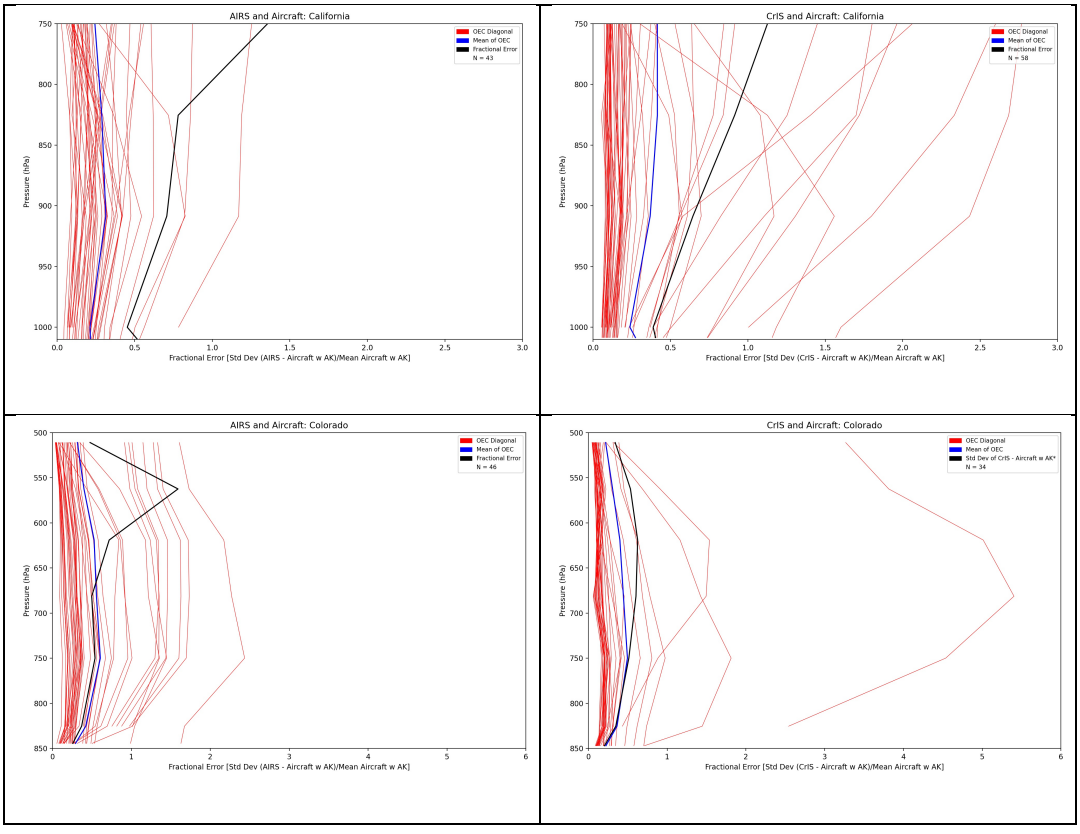

Figure 6: Fractional standard deviation between AIRS (left) and CrIS (right) and aircraft profiles with the averaging kernel applied (black) during DISCOVER-AQ in California (top panels) and Colorado (bottom panels); estimated uncertainty (red) and mean of estimated uncertainty (blue).

sources are not accounted for in the optimal estimation process. The most likely source is the sampling difference: AIRS and CrIS sampled three dimensional columns, 15 to 50 km wide,

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450 while aircraft instruments sampled in two dimensions, vertically and along a spiral line with a 5 km width. Systematic retrieval errors, such as errors in water vapor, may also contribute. These fractional differences increase with altitude, but this is likely due to the rapid decrease in NH_3 above the mixed layer, a case of large differences in small numbers. The measured uncertainties range from 5% to 50% in California. These large values point to the need for averaging over time

455 or space to obtain less noisy information.

Table 1: Statistical analysis of the DISCOVER-AQ data from AIRS, CrIS and the aircraft over California in 2013 and Colorado in 2014.

California												
	Profile				Satellite-Aircraft: no AK				Satellite-Aircraft: with AK			
	Mean		Std Dev		Bias		Std Dev		Bias		Std Dev	
Pressure	AIRS	CrIS	AIRS	CrIS	AIRS	CrIS	AIRS	CrIS	AIRS	CrIS	AIRS	CrIS
hPa	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv
1008.486	9.69	16.211	10.785	6.027	-38.03	-43.799	23.972	37.988	0.684	-0.171	4.593	6.716
1000	9.342	14.443	9.875	5.44	-25.049	-22.746	19.995	17.106	0.396	-0.89	4.003	5.973
908.514	9.693	7.63	8.634	4.101	0.238	1.956	9.81	5.582	0.344	-0.112	6.714	4.99
825.402	5.742	4.883	5.401	3.372	1.471	1.266	5.327	4.345	1.186	0.653	3.328	3.854
749.893	3.083	2.958	3.47	2.311	-1.96	-0.801	4.815	3.101	0.992	0.738	2.802	2.442
Colorado												
	Profile				Satellite-Aircraft: no AK				Satellite-Aircraft: with AK			
	Mean		Std Dev		Bias		Std Dev		Bias		Std Dev	
Pressure	AIRS	CrIS	AIRS	CrIS	AIRS	CrIS	AIRS	CrIS	AIRS	CrIS	AIRS	CrIS
hPa	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv	ppbv
844.469	10.341	9.805	3.415	3.801	3.817	3.387	3.741	3.871	-0.803	-0.42	2.868	2.085
825.402	8.214	7.529	3.064	3.595	2.19	1.902	3.377	4.006	-0.902	-0.193	3.368	2.687
749.893	3.889	3.852	2.173	2.101	-0.12	0.102	2.431	2.292	-0.543	-0.031	2.392	1.994
681.291	2.324	2.703	1.143	1.207	0.084	0.338	1.684	1.465	-0.288	0.012	1.3	1.626
618.966	1.474	1.858	0.943	0.974	-0.356	-0.102	1.41	1.228	-0.083	0.304	1.119	0.972
562.342	1.07	1.104	1.444	0.548	-0.538	-0.535	1.764	0.724	0.091	0.179	1.557	0.494
510.898	0.521	0.552	0.231	0.212	-0.695	-0.687	0.523	0.374	-0.065	0.016	0.279	0.182

Since much of the work on validating NH_3 from space-based infrared sensors has been done

460 using IASI data, which provide total columns rather than profiles (e.g., van Damme et al., 2015; Dammers et al., 2017; Guo2021), here we also compare AIRS and CrIS total columns with aircraft total columns calculated using the mixed layer height approach described by Guo2021.

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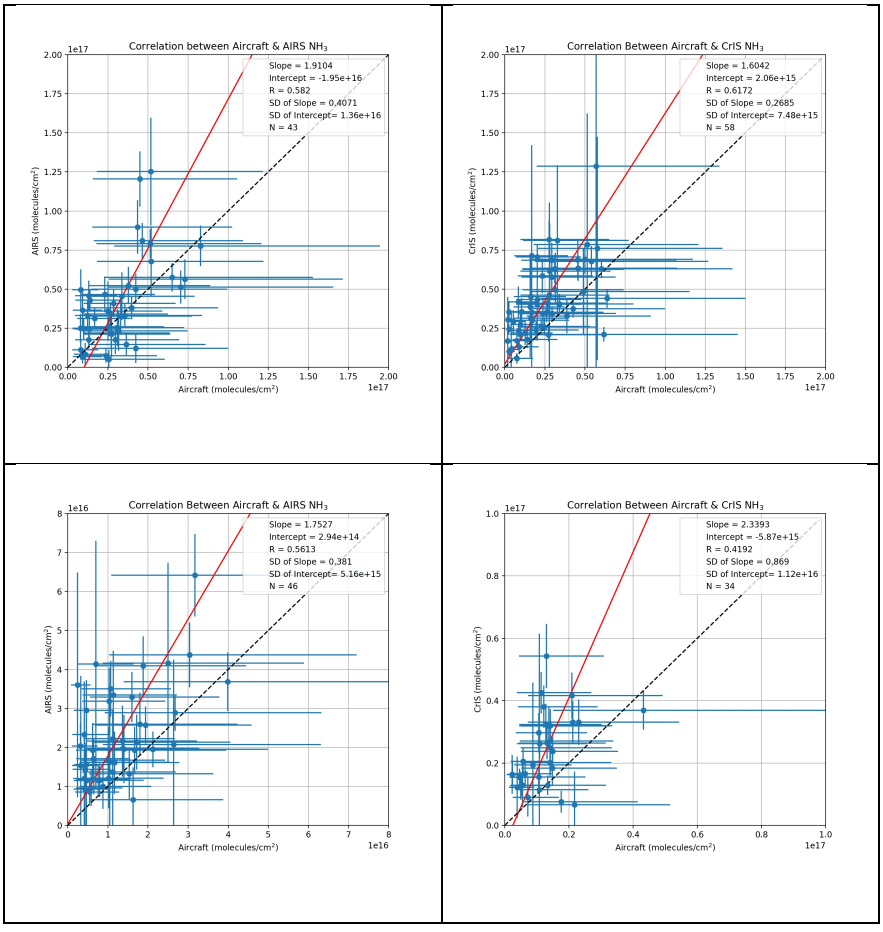

Figure 7: total NH3 columns from AIRS (left) and CrIS (right) versus aircraft columns during DISCOVER-AQ in California (top panels) and Colorado (bottom panels); dashed line shows the 1:1 line, red line the linear fit; vertical and horizontal lines indicate the uncertainties in the aircraft and satellite data, respectively.

In this method the in situ NH_3 total column is estimated by integrating the aircraft NH_3 profile to the top of the mixed layer; above this level, NH_3 amounts are assumed to be zero. This is a reasonable assumption, since NH_3 has a short lifetime (on the order of hours or days), and is rarely transported to the middle or upper troposphere; it has been measured above the mixed layer, but at low levels (less than 1 ppbv; e.g., Höpfner et al., 2019; Nowak et al., 2010). Furthermore, as stated in section 3.1, there are large uncertainties in the PTR-MS measurements above the mixed layer.

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The MUSES total columns are compared against the integrated aircraft columns, using orthogonal linear regression (Figure 7, top panels); the intercept has been allowed to vary, as both AIRS and CrIS have detection limits, as does IASI. Note that the instrument operators have not been applied to the aircraft column data. The correlation coefficients are 0.58 for AIRS and 0.62 for CrIS, within the range of results from previous studies. However, the slopes are very much greater than one (1.9 for AIRS and 1.6 for CrIS), which was not the case in the IASI evaluation of Guo2021. That study showed that assuming NH_3 concentrations are zero above the mixed layer led to slopes close to the 1:1 line. If this assumption of zero amounts NH_3 above the mixed layer is no longer valid at the later (13:30 LST) CrIS and AIRS overpass times, the aircraft columns would be biased low with respect to CrIS and AIRS; on the other hand, the profile differences shown in Figure 3 suggest that AIRS and CrIS are overestimating NH_3 above 910 hPa. Given the uncertainty in the aircraft measurements at these higher altitudes it is not possible to currently resolve this issue.

4.2 Colorado

DISCOVER-AQ Colorado took place during July and August of 2014, in the Colorado Front Range; while this is also a region with strong NH_3 sources, the aircraft data showed lower values than in the California Central Valley (Figure 2, lower left panel); maximum values are on the order of 20 ppbv, with most near surface values ranging from 5 to 10 ppbv. Also, in contrast to the Central Valley there were no thermal inversions during this period (Figure 2, lower right panel). Applying the co-location criteria yielded 46 AIRS profiles but only 34 CrIS profiles, in part due to some poor quality CrIS retrievals. Direct comparisons of the AIRS and CrIS profiles with aircraft data (Figure 3, lower panels) show both AIRS and CrIS biased high by 3.8 ppbv (AIRS) and 3.4 ppbv (CrIS) near the surface (~844 hPa) and by 2.2 ppbv (AIRS) and 1.9 ppbv

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(CrIS) at 825 hPa, but the bias is close to zero at 750 hPa. Note that this is in direct contrast to the California results, which showed both AIRS and CrIS NH_3 biased very low at and near the surface and biased slightly high at greater altitudes.

The sum of the rows of the averaging kernel plots (Figure 4, lower panels) present similar maximum and surface values as in California, and applying the instrument operator again eliminates the bias, but does not reduce the spread in the satellite-aircraft differences dramatically (Figure 5, lower panels), while the large bias seen against the a priori profiles (~-12 ppbv for AIRS and ~-10 ppbv for CrIS) is removed by the retrieval, confirming again that the retrieval process provides information content beyond the a priori.

The uncertainty analysis (Figure 6, lower panels) shows that the estimated uncertainty is quite close to the measured uncertainty below 700 hPa, but is less than the measured value above this level, indicating an unaccounted for error source. Finally, the total column comparisons also indicate that the AIRS and CrIS NH_3 (Figure 7, lower panels) columns are correlated with the aircraft values (with correlation coefficients of 0.56 for AIRS and 0.42 for CrIS), but are biased high, most notably for CrIS, for which the regression presents a slope of 2.3. In this region the high column biases are likely due to the bias in the retrieved surface and near surface values seen in Figure 10, rather than differences between the retrieved and aircraft profile shape. Again, sub-pixel heterogeneity and water vapor retrieval errors could also contribute to the differences.

5.0 Magic Valley Analysis

5.1 USDA network

The USDA-ARS established and maintained an NH_3 monitoring network along two transects (North-South and West-East) across the Magic Valley region of south-central Idaho during the February 2018-December 2020 period (Figure 8). The Magic Valley region is heavily dominated

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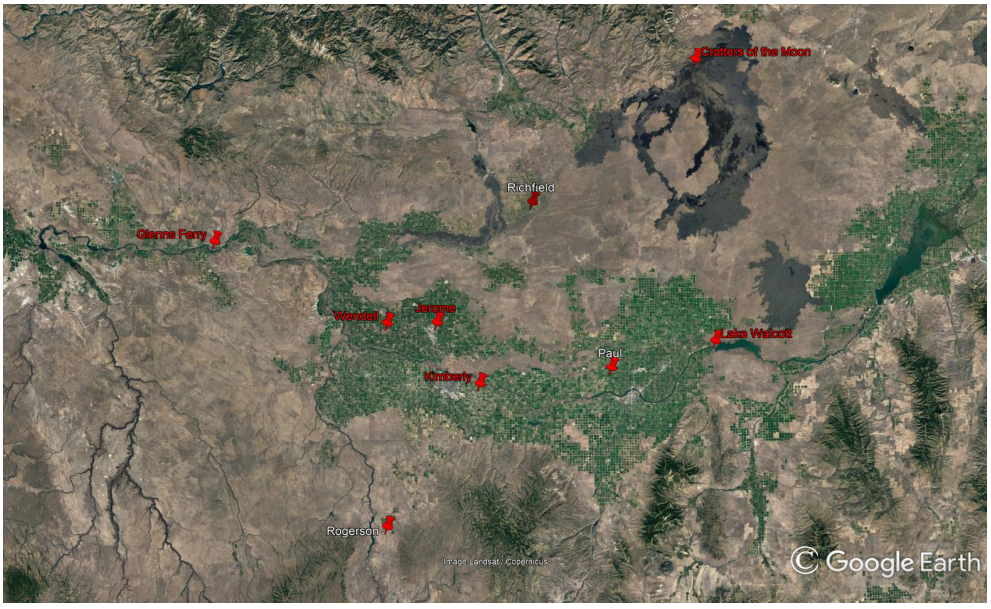

Figure 8: USDA Magic Valley network showing the nine measurement sites

by irrigated agriculture and is one of the most concentrated dairy production regions in the US Research in this region has reported that NH_3 emissions from agricultural and dairy production contribute approximately 44,000 MT N yr^{-1} to the atmosphere (Leytem et al., 2021) and that NH_3 emissions fluctuate by season following trends in temperature (Leytem et al., 2011, 2013). The network was established to gain a better understanding of the spatial variablity of ambient NH_3 concentrations and transport within the region. The network consisted of 8 sampling locations (7 until 2020, when an additional site was added), and also utilized data from the NADP AMoN site located to the north of the region at Craters of the Moon National Monument. Ammonia concentrations were measured with passive diffusive NH_3 samplers (Radiello), which were deployed bi-weekly, and generated two week mean surface NH_3 concentrations. Radiello samplers have been shown to be approximmately 9% biased high (Puchalski et al., 2015)These

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data provided a unique opportunity to evaluate the seasonal signals measured by CrIS NH_3, as well as its capability to capture small scale (on the order of a few kilometers) spatial variability.

5.2 Evaluating CrIS NH_3 against the surface data

CrIS NH_3 data are available for most of the 2018-2020 period, with a gap in the spring of 2019 due to an instrument malfunction. CrIS data within 15 km of each site were compared with the ground data at that site. Only the CrIS observations at CrIS ~13:30 LST were analyzed. The NH3 retrievals at 1:30 LST have weaker signals (due to lower thermal contrast), and would add uncertainty to the results; furthermore, while there are strong diurnal cycles in the NH_3 emitted from the dairy facilities (Letyem et al., 2011, 2013) the average daily emissions are close to the early afternoon value. Figure 9 shows the individual CrIS observations (small red triangles), the two-week averages of the CrIS data (large red triangles connected with a red line) and the ground data (blue triangles) at each site.

The time series in Figure 9 are sorted by the peak values of the ground data. At every site, CrIS clearly captures the seasonal cycle, though winter values are usually underestimated (possibly, and a few strong warm season peaks in the ground data are not observed. At Craters of the Moon, a National Monument, and at Rogerson, in land administered by the Bureau of Land Management, CrIS returns consistently higher values than the ground site during the warmer months (May to October). At present we have no explanation for this high bias. At the next four site (Glenns Ferry, Richfield, Lake Wolcott and Kimberley), which are in areas of mixed agricultural activity, CrIS and the ground data are in good agreement during the warmer months, though CrIS underestimates the June 2018 maximum at the Richfield site. At all four sites there is a peak in the ground data in November 2019 that is either matched in the CrIS data (Glenns Ferry) or at least visible (Richfield, Kimberley, Lake Wolcott). This peak is also evident in the

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data from Paul and possibly Wendell, though the variability at this latter site makes it difficult to

570 confirm. This suggests an area wide change in meteorological conditions, such as an inversion,

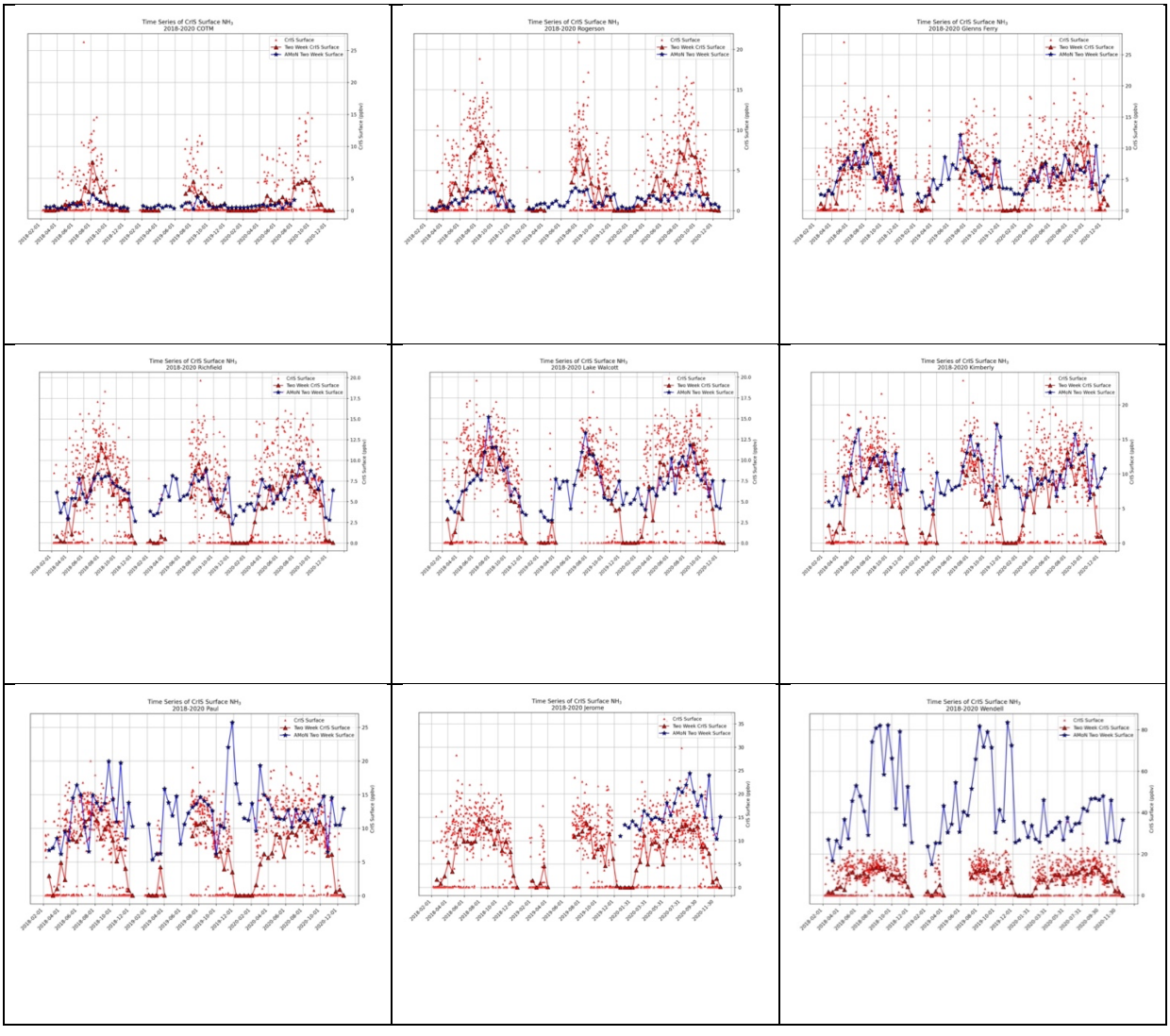

Figure 9: Time series of the in situ data (blue triangles, and the collocated CrIS surface values; red dots indicate daily values, red triangles two week means.

that led to increased NH_3 near the surface: however, there is no evidence in the meteorological

575 data for such an inversion. The last three sites (Paul, Jerome and Wendell) are close to and/or downwind of multiple dairies, which likely leads to greater sub-pixel inhomogeneity. CrIS

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underestimates the warm season maximum, and does not capture many of the peaks in the ground data, most notably at the Wendell site, where CrIS did not observe the over 80 ppbv peaks in 2018 and 2019; these extremely high concentrations were not observed in 2020. The sudden change suggests that a strong source that was dominating the signal in the ground data stopped emitting in 2020.

Analyzing these data in aggregate, first spatially then temporally, provides some useful insights.

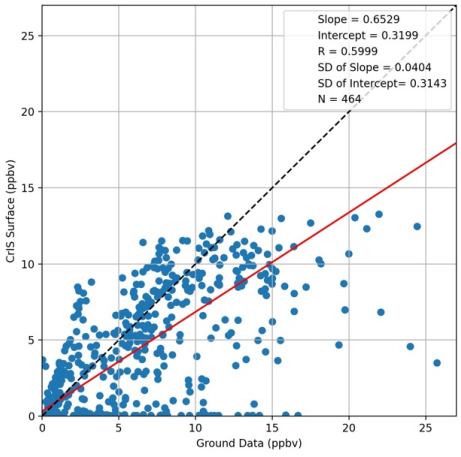

Figure 10: two week means of surface NH3 from CrIS and the in situ instruments; dashed line shows the 1:1 line, red line the linear fit.

Plotting the CrIS two week averages against all the ground data values (Figure 10), excluding those from the Wendell site, which are extreme outliers, shows a correlation of 0.6, at the high end of the values reported in the literature and slope of 0.65, indicating that CrIS NH_3 is biased low overall. This result is in line with the low biases found in the surface values of the AIRS and CrIS California data, though here not necessarily caused by thermal inversions, and with the low biases in the CFPR results at high NH_3 FTIR values seen by Dammers et al. (2017). It provides a quantitative measure of the agreement seen in the seasonal cycles shown in Figure 9.

Three years of fairly dense data over a region with many sources with fixed locations are an excellent candidate for spatial oversampling algorithms, which trade temporal resolution for greater information on spatial variability. Here we applied the physics based oversampling algorithm developed by Sun et al. (2018), which uses the instrument spatial response function to

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600 weight the contributions of each satellite observation to a fine grid (here 0.002 deg), to each of the three years of CrIS data taken over the Magic Valley region (Figure 11). The in-situ data are overlaid on the CrIS maps; note that the Wendell values are shown in the upper right corner of the maps, as otherwise they would distort the color scale; note also that the Jerome data are blank in 2018 and 2019, when this site was not operational.

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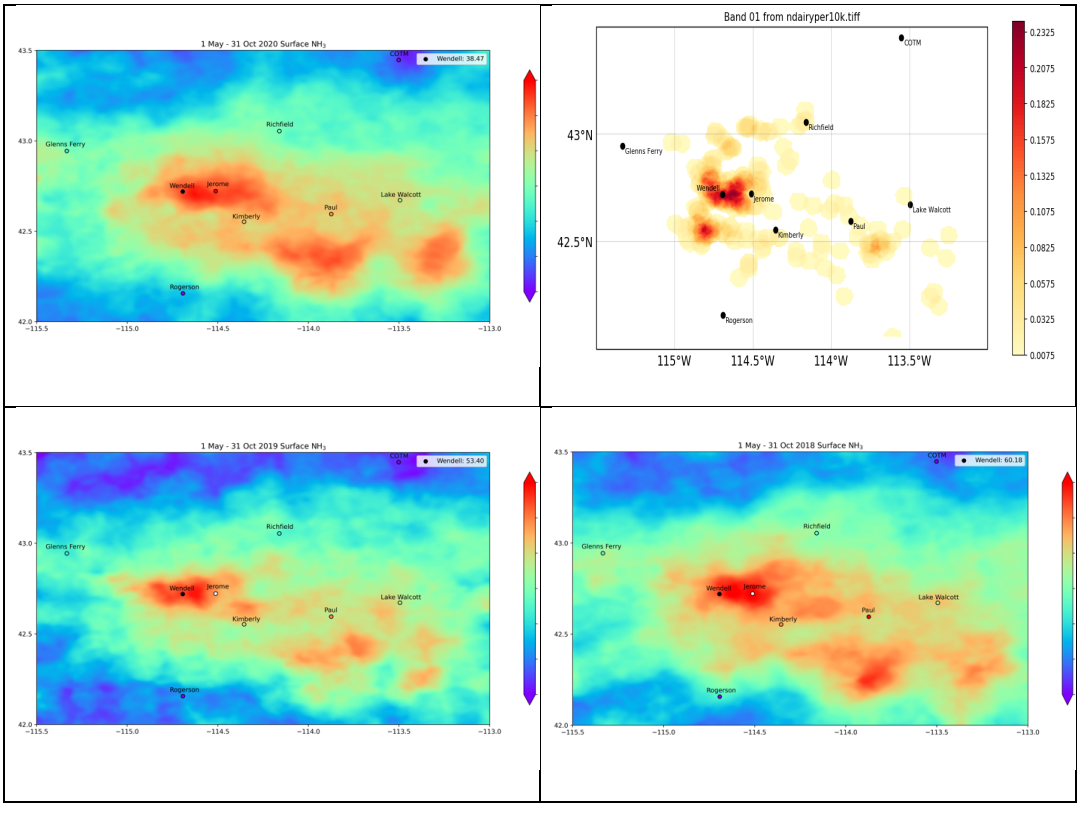

Figure 11: CrIS surface NH3 data from the May to October period oversampled onto a 0.002 deg grid for each of three years: 2020 (upper left), 2019 (lower left), 2018 (lower right); number of dairies per square kilometer (upper right)

The location of the CrIS NH_3 "hotspots" and the gradients in NH_3 are very consistent from year

610 to year, though NH_3 concentrations are lower in 2019, possibly due to the CrIS data gap between March and June. There is good qualitative agreement with the in situ data, with the exception of

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the Wendell site, which was discussed above. Moreover, the hotspots are very well correlated with the areas of high dairy density (Figure 11, upper right). These maps illustrate the power of the CrIS data to provide context to in situ measurements and far more information on the spatial variability in NH_3 than is normally available from emissions databases, which are frequently at county level. Data from these gridded maps could be used to constrain emission inventories over much larger areas and at more frequent intervals than is currently possible. This oversampling analysis demonstrates the utility of providing users with Level 2 products, since they then choose the sampling periods and resolution that are most appropriate for their purposes and are best suited to the times and regions under investigation. For example, a user might want to study the variability of NH_3 over just the urban area of Mexico City. Level 3 products provide no such flexibility.

6.0 Conclusions and future work

While the DISCOVER-AQ datasets are limited in their temporal and spatial coverage, and in challenging conditions in California, they do provide some useful information for end-users who would like to use CrIS and AIRS data over strong source regions. The AIRS and CrIS profiles individually have large uncertainties, which are driven by local conditions, most significantly temperature profiles and sub-pixel heterogeneity. However, average biases, after smoothing errors are accounted for, are below or close to 1 ppbv. The error analysis in Figures 7 and 13 indicates that the a posteriori estimated error underestimates the actual uncertainties: this is likely caused by differences in the air masses sampled by the satellite and the aircraft measurements., and to a lesser extent by unaccounted for error sources. The column data suggest that either there is more NH_3 than expected above the mixed layer, or the retrievals are overestimating NH_3 above

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this layer, maybe because the a priori profile shape is very different from the true profile shape. More measurements of NH_3 in this altitude region are required to resolve this issue.

The Magic Valley analysis clearly demonstrates the importance of having more than a few dozen data point measurements to obtain useful information from space-based retrievals of NH_3. With 464 observations over three years, over a limited region, it was possible to obtain a clear picture of the source distribution in the Magic Valley through the application of a physics based oversampling algorithm. Further work will apply this approach to other regions and times and use the resulting maps to estimate emissions and to improve reactive nitrogen deposition estimates.

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Data availability

MUSES AIRS and CrIS NH_3 products from S-NPP are available via the GES-DISC from the NASA Tropospheric Ozone and Precursors from Earth System Sounding (TROPESS) project at https://disc.gsfc.nasa.gov/datasets/TRPSDL2NH3AIRSFS_1/summary?keywords=tropess%20nh3, (Bowman, 2021a) and https://disc.gsfc.nasa.gov/datasets/TRPSDL2NH3CRSFS_1/summary?keywords=tropess%20nh3, (Bowman 2021b) respectively. The AIRS and CrIS datasets matched with aircraft used here for validation are available from the authors on request.

Author contributions

KCP designed the project, carried out the MUSES retrievals, created the preliminary plots, analyzed the results and wrote the manuscript. XG, RW and MZ provided the co-located aircraft data and made suggestions for the manuscript. AL provided the Magic Valley data and contributed relevant text. KS provided the oversampling code and revised the manuscript. VP and MS provided useful insights and revised the manuscript. CC and EB created all the plots. MM and AW obtained the DISCOVER-AQ data. VK designed and built the MUSES software.

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