

## Ammonia variability and trends from globally distributed FTIR measurements and model simulations

### Response to Anonymous Referee #1

#### Summary

This study uses ground-based FTIR measurements at 22 worldwide sites to track atmospheric ammonia (seasonality, diurnal patterns, trends). The authors also compare the measurements against two models. As a detailed measurement report, the paper is a valuable addition to the existing literature on atmospheric ammonia. The release of the dataset is a milestone. The trend and seasonal analysis complement results established from satellite observations. The diurnal characterisation is a useful contribution that is harder to obtain from satellite data. The GCHP/TCR-2 NH<sub>3</sub> evaluations would arguably deserve a dedicated study, in which a more in-depth analysis could be made and/or attempts made to reconcile the simulations with the observations.

**I can recommend publication of this paper subject to a major revision taking into account the following comments:**

Response: We thank the reviewer for the careful and constructive assessment of our manuscript and for recommending publication subject to revision. The comments helped us strengthen the statistical treatment, clarify the interpretation of the Izaña enhancements, improve the model evaluation, and improve the presentation of the figures and tables. We have addressed each comment below and revised the manuscript accordingly.

#### Main comments

**- As the authors acknowledge, NH<sub>3</sub> enhancements originating from volcanoes are rarely reported. In view of the exceptional nature, the authors should present robust evidence of this, for instance in the form of a physical fit over the spectrum showing the residuals with and without NH<sub>3</sub>/SO<sub>2</sub>. This would exclude retrieval artefacts.**

Response: We thank the reviewer for raising this important point. We agree that direct volcanic NH<sub>3</sub> emissions are exceptionally rare and that our original wording could be interpreted as implying a direct magmatic source. We have therefore revised the manuscript to avoid this interpretation and now refer to the observed NH<sub>3</sub> enhancements as associated with the Tajogaite eruption rather than as direct volcanic NH<sub>3</sub> emissions.

Regarding the possibility of NH<sub>3</sub>/SO<sub>2</sub> spectral interference or retrieval artefacts, the SO<sub>2</sub> and NH<sub>3</sub> products at Izaña are retrieved independently from different, non-overlapping spectral regions. The SO<sub>2</sub> retrieval uses the 1120–1180 cm<sup>-1</sup> region, whereas the three NH<sub>3</sub> microwindows are those described in Section 2.1.2 of the manuscript. SO<sub>2</sub> is therefore not fitted as an interfering species within the NH<sub>3</sub> microwindows. The principal SO<sub>2</sub> vibrational modes ( $\nu_1 \approx 1152$  cm<sup>-1</sup>,  $\nu_2 \approx 518$  cm<sup>-1</sup>, and  $\nu_3 \approx 1362$  cm<sup>-1</sup>) also do not coincide with the principal NH<sub>3</sub> vibrational modes used for the retrieval. Consequently, a combined fit comparing NH<sub>3</sub> residuals “with and without SO<sub>2</sub>” is not directly applicable to our retrieval setup because SO<sub>2</sub> and NH<sub>3</sub> are not retrieved from the same spectral fit.

To address the broader concern about source attribution, we substantially revised the discussion and added an independent tracer analysis. SO<sub>2</sub> is used as a tracer of volcanic-plume influence and HCN as a tracer of biomass-burning influence. We compare standardized daily-maximum anomalies using both eruption-period statistics and longer reference periods. The revised analysis identifies NH<sub>3</sub>–HCN coincident enhancements on 22 September and 2 October 2021 and an NH<sub>3</sub>–SO<sub>2</sub> coincident enhancement on 28 September 2021. The lack of a systematic one-to-one correspondence between NH<sub>3</sub> and SO<sub>2</sub>, together with the NH<sub>3</sub>–HCN coincidences and the independent evidence of enhanced NH<sub>3</sub> emissions from agricultural areas affected by the lava flows (López-Darias et al., 2025), supports an eruption-associated secondary source rather than direct magmatic NH<sub>3</sub> emission. Two more references were added López-Darias et al., (2025) and Mather et al., (2004).

Changes to the manuscript

## Abstract

### Previous text:

“Significant enhancements were attributed to biomass burning, and  $\text{NH}_3$  emissions from volcanic eruptions were detected at the Izaña site.”

### Revised text:

“Significant enhancements were attributed to biomass-burning events, including enhancements associated with the 2021 Tajogaite eruption detected at the Izaña site.”

## Section 3.1

### Previous text:

“For the Izaña site, the enhancements observed in 2012 are from the Tenerife Island fire that reached the site in July 2012 (Santamarta Cerezal, 2013). In addition, in September 2021, a volcanic eruption on the island of La Palma (Canary Islands) affected the Izaña site (García et al., 2023; Campeny et al., 2023). Studies on  $\text{NH}_3$  emissions from volcanic eruptions are limited, but simultaneous enhancements of  $\text{NH}_3$  and  $\text{SO}_2$  from volcanoes have been reported (Uematsu et al., 2004; Adams et al., 2017). Figure A5 shows  $\text{SO}_2$  and  $\text{NH}_3$  enhancements during the Tajogaite eruption from 19 September 2021 to 13 December 2021, during which volcanic plume reached the Izaña site.”

### Revised text:

“For the Izaña site, the enhancements observed in 2012 are from the Tenerife Island fire that reached the site in July 2012. In addition, in September 2021, a volcanic eruption on the island of La Palma (Canary Islands) affected the Izaña site. Magmatic  $\text{NH}_3$  emissions are unlikely, as volcanic nitrogen is degassed almost exclusively as  $\text{N}_2$  (Mather et al., 2004), and, to our knowledge, only one study has reported simultaneous  $\text{NH}_3$  and  $\text{SO}_2$  enhancements from a volcano (Uematsu et al., 2004). Figure A2a shows  $\text{NH}_3$  and  $\text{SO}_2$  enhancements from 19 September to 13 December 2021, when the volcanic plume reached Izaña; given the near-zero  $\text{SO}_2$  background at Izaña,  $\text{SO}_2$  unambiguously traces the volcanic plume. However, enhanced  $\text{NH}_3$  emissions from agricultural areas of La Palma were reported during the eruption, when hot lava flows reached cultivated fields and a fertilizer-storage facility (López-Darias et al., 2025), events associated with biomass burning. A complementary analysis of HCN over this period also supports a biomass-burning contribution in the Izaña measurements (Fig. A2b), and thus a non-volcanic origin of the observed  $\text{NH}_3$  enhancements. ”

### Updated appendix A

Updated Figure A2 (previously figure A5) showing  $\text{NH}_3/\text{SO}_2$  time series and standardized anomalies.

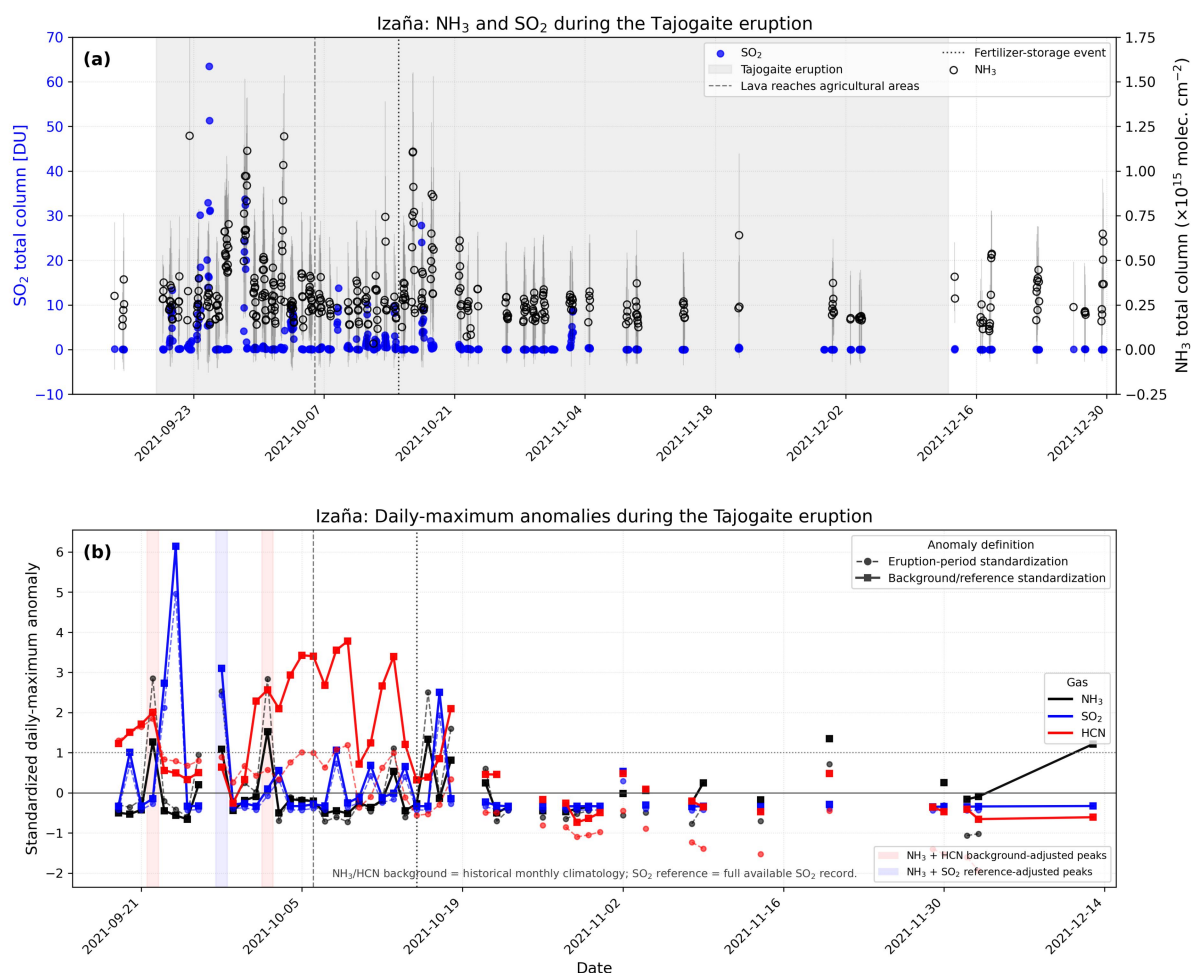


Figure A2. (a) NH<sub>3</sub> and SO<sub>2</sub> total columns measured at Izaña from September to December 2021 during the Tajogaite eruption. Episodic enhancements of both gases are observed during periods when the volcanic plume reached the Izaña site. The vertical lines indicate the onset of the reported increase in NH<sub>3</sub> associated with lava flows reaching agricultural areas of La Palma (6 October 2021) and the explosion of a fertilizer-storage facility affected by the lava flows (15 October 2021) (López-Darias et al., 2025). (b) Standardized anomalies of the daily maximum NH<sub>3</sub> (black), SO<sub>2</sub> (blue), and HCN (red, NDACC product) total columns during the Tajogaite eruption. SO<sub>2</sub> is used here as a tracer of volcanic-plume influence, whereas HCN is used as a tracer of biomass-burning influence. Dashed lines represent anomalies standardized using only observations obtained during the eruption period. Solid lines represent anomalies calculated using longer reference periods. For NH<sub>3</sub> and HCN, daily maxima are standardized relative to the corresponding monthly climatology derived from the multi-year records excluding 2021. Because a multi-year SO<sub>2</sub> record was not available, the SO<sub>2</sub> reference anomaly was calculated from the daily maxima over the complete available SO<sub>2</sub> record (September 2021–February 2022). Shaded intervals indicate dates on which NH<sub>3</sub> and HCN or NH<sub>3</sub> and SO<sub>2</sub> simultaneously exceeded the selected standardized-anomaly threshold ( $z > 1$ ). The NH<sub>3</sub>–HCN coincident enhancements on 22 September and 2 October, and the NH<sub>3</sub>–SO<sub>2</sub> enhancement on 28 September, are identified consistently using both the eruption-period and longer-reference standardizations.

Added anomaly methodology to appendix A

The standardized anomalies of the daily maxima shown in Figure A2b were calculated using two reference definitions. First, daily maxima were standardized relative to the mean and standard deviation calculated using only observations during the Tajogaite eruption period (dashed lines). Second, longer reference periods were used to account for the normal variability of each species (solid lines). For NH<sub>3</sub> and HCN, this was achieved using month-specific climatological means and standard deviations derived from the corresponding multi-year records, excluding 2021. Because a multi-year SO<sub>2</sub> record was unavailable, the SO<sub>2</sub> reference statistics were instead calculated from the complete available SO<sub>2</sub> record (September 2021–February 2022). Thus, the latter is an available-record reference rather than a climatological background.

For each species  $X$ , the daily maximum is defined as

$$X_{\max}(d) = \max_{t \in d} X(t), \quad (\text{A1})$$

where  $X(t)$  represents the individual FTIR total-column retrievals obtained during day  $d$ . For the eruption-period standardization,

550 the mean daily maximum for species  $X$  is

$$\mu_{X,\text{eruption}} = \frac{1}{N_{\text{eruption}}} \sum_{d \in \text{eruption}} X_{\max}(d), \quad (\text{A2})$$

and the corresponding standard deviation is

$$\sigma_{X,\text{eruption}} = \sqrt{\frac{1}{N_{\text{eruption}} - 1} \sum_{d \in \text{eruption}} [X_{\max}(d) - \mu_{X,\text{eruption}}]^2}, \quad (\text{A3})$$

where  $N_{\text{eruption}}$  is the number of days with available observations during the eruption. The standardized eruption-period

555 anomaly is then

$$Z_{X,\text{eruption}}(d) = \frac{X_{\max}(d) - \mu_{X,\text{eruption}}}{\sigma_{X,\text{eruption}}}. \quad (\text{A4})$$

This definition was applied independently to  $\text{NH}_3$ ,  $\text{SO}_2$ , and  $\text{HCN}$ .

For  $\text{NH}_3$  and  $\text{HCN}$ , a second set of anomalies was calculated relative to their historical monthly climatologies. For each calendar month  $m$ , the climatological mean of the daily maxima is

$$560 \quad \mu_{X,m} = \frac{1}{N_m} \sum_{d \in m, y \neq 2021} X_{\max}(d), \quad (\text{A5})$$

and the corresponding standard deviation is

$$\sigma_{X,m} = \sqrt{\frac{1}{N_m - 1} \sum_{d \in m, y \neq 2021} [X_{\max}(d) - \mu_{X,m}]^2}, \quad (\text{A6})$$

where  $N_m$  is the number of daily maxima available for calendar month  $m$  in the historical record. All observations from 2021 were excluded from the climatological reference. The climatology-adjusted anomaly during the eruption is therefore

$$565 \quad Z_{X,\text{clim}}(d) = \frac{X_{\max}(d) - \mu_{X,m(d)}}{\sigma_{X,m(d)}}, \quad (\text{A7})$$

where  $m(d)$  denotes the calendar month corresponding to day  $d$ .

For SO<sub>2</sub>, a multi-year climatological reference was not available. Instead, the mean and standard deviation were calculated from the daily maxima over the complete available SO<sub>2</sub> record,

$$\mu_{\text{SO}_2,\text{ref}} = \frac{1}{N_{\text{ref}}} \sum_{d=1}^{N_{\text{ref}}} \text{SO}_{2,\text{max}}(d), \quad (\text{A8})$$

$$\sigma_{\text{SO}_2,\text{ref}} = \sqrt{\frac{1}{N_{\text{ref}} - 1} \sum_{d=1}^{N_{\text{ref}}} [\text{SO}_{2,\text{max}}(d) - \mu_{\text{SO}_2,\text{ref}}]^2}, \quad (\text{A9})$$

where  $N_{\text{ref}}$  is the number of days with available SO<sub>2</sub> observations between September 2021 and February 2022. The resulting available-record standardized anomaly is

$$Z_{\text{SO}_2,\text{ref}}(d) = \frac{\text{SO}_{2,\text{max}}(d) - \mu_{\text{SO}_2,\text{ref}}}{\sigma_{\text{SO}_2,\text{ref}}}. \quad (\text{A10})$$

- The influence of SO<sub>2</sub>/NO<sub>x</sub> on the trends is stated a few times but never demonstrated. For example a figure of collocated trends of NH<sub>3</sub>/SO<sub>2</sub>/NO<sub>x</sub> could be shown and discussed. Table 9 contradicts partially the lifetime argument (showing very small correlations between NH<sub>3</sub> and SO<sub>2</sub>/NO<sub>x</sub>).

Response: We thank the reviewer for this suggestion. We added a new Figure 8 showing collocated GCHP trends in NH<sub>3</sub>, SO<sub>2</sub>, and NO<sub>x</sub> over 2003–2021. Thirteen sites show the combination of a significant positive NH<sub>3</sub> trend and significant negative SO<sub>2</sub> and NO<sub>x</sub> trends. This long-term trend comparison addresses a different timescale from the correlations reported in Table 9, which quantify covariability over the matched time series and can remain small even when long-term trends have opposite signs. We revised the discussion to make this distinction explicit.

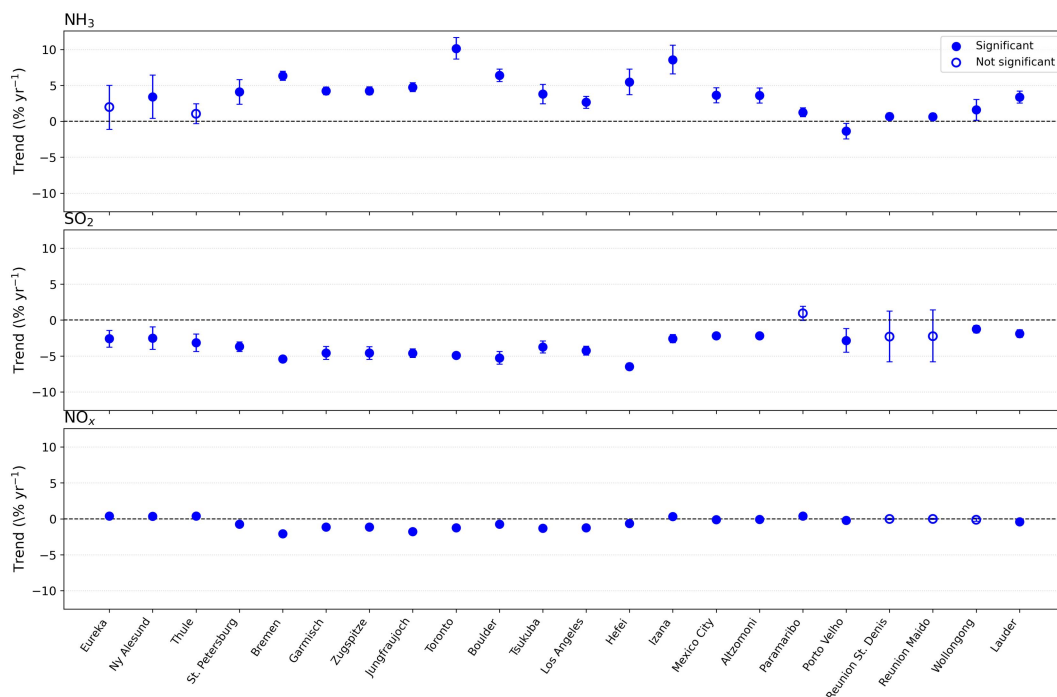


Figure 8. Collocated trends of daily means of total columns of NH<sub>3</sub>, SO<sub>2</sub>, and NO<sub>x</sub> derived from the GCHP 2003–2021 time series at each site. Vertical lines represent the 95% CI.

Added new text on section 4.4

“The collocated GCHP trends (Figure 8) show that at many sites, particularly in the Northern Hemisphere, increasing NH<sub>3</sub> coincides with significant decreases in SO<sub>2</sub> and NO<sub>x</sub>, which is consistent with reduced acidic precursor availability contributing to increased gaseous NH<sub>3</sub> lifetime. However, this relationship is not observed at all sites, indicating that changes in NH<sub>3</sub> emissions, meteorology, transport, and other processes also contribute to the observed trends.”

**- Can you report the error (budget) per site (currently only a range and average is given)?**

Response: We thank the reviewer for this suggestion. The mean total retrieval error is now reported for each site in Table 2 and the manuscript text Section 2.1.2 was revised.

. We also clarified in Section 2.1.2 that the total retrieval error is calculated as the square root of the sum of the squares of the systematic and statistical error components. Across all retrievals, the total error ranges from 0.1 to  $2.0 \times 10^{15}$  molec. cm<sup>-2</sup>, with an average of  $0.93 \pm 1.30 \times 10^{15}$  molec. cm<sup>-2</sup>.

Previous text

“The total error on the NH<sub>3</sub> total columns was calculated as the square root sum of the squares of all systematic and statistical errors (García et al., 2021), ranging between 0.1 and 2.0 with an average value for all sites of  $0.93 \pm 1.30 \times 10^{15}$  molec. cm<sup>-2</sup>.”

Revised text

“The total error on the NH<sub>3</sub> total columns was calculated as the squareroot sum of the squares of all systematic and statistical errors (García et al., 2021), ranging between 0.01 and  $4.32 \times 10^{15}$  molec. cm<sup>-2</sup> with an average value for all sites of  $0.93 \pm 1.30 \times 10^{15}$  molec. cm<sup>-2</sup>, the mean total retrieval error per site is reported in Table 2.”

**Table 2.** NH<sub>3</sub> mean total columns  $\pm 1\sigma$  and absolute trends with 95% confidence intervals for the FTIR sites. Bold indicates a significant trend.

| Station                | Measurement | NH <sub>3</sub> Mean Total Column<br>(molec. cm <sup>-2</sup> ) $\times 10^{15}$ | NH <sub>3</sub> Mean Total Error<br>(molec. cm <sup>-2</sup> ) $\times 10^{15}$ | NH <sub>3</sub> Trend and 95% CI                             |                              |
|------------------------|-------------|----------------------------------------------------------------------------------|---------------------------------------------------------------------------------|--------------------------------------------------------------|------------------------------|
|                        | Period      |                                                                                  |                                                                                 | (molec. cm <sup>-2</sup> yr <sup>-1</sup> ) $\times 10^{13}$ | % yr <sup>-1</sup>           |
| Eureka                 | 2006 – 2020 | 0.17 $\pm$ 0.24                                                                  | 0.01                                                                            | <b>0.58</b> (0.13 – 1.02)                                    | <b>3.0</b> (0.73 – 5.21)     |
| Ny Alesund             | 2017 – 2019 | 0.12 $\pm$ 0.12                                                                  | 0.06                                                                            | –                                                            | –                            |
| Thule                  | 1999 – 2022 | 0.46 $\pm$ 0.34                                                                  | 0.03                                                                            | <b>-2.10</b> (-2.50 – -1.70)                                 | <b>-7.7</b> (-9.1 – -6.25)   |
| St. Petersburg         | 2010 – 2022 | 3.07 $\pm$ 2.85                                                                  | 0.87                                                                            | <b>11.70</b> (3.54 – 19.60)                                  | <b>4.67</b> (1.44 – 7.81)    |
| Bremen                 | 2004 – 2018 | 16.80 $\pm$ 14.70                                                                | 0.93                                                                            | 28.20 (-8.20 – 63.80)                                        | 3.55 (-1.03 – 8.02)          |
| Garmisch               | 2004 – 2007 | 3.56 $\pm$ 3.97                                                                  | 0.54                                                                            | 31.10 (-13.80 – 76.20)                                       | 10.64 (-6.39 – 28.04)        |
| Zugspitze              | 1995 – 2023 | 0.32 $\pm$ 0.55                                                                  | 0.11                                                                            | <b>0.84</b> (0.58 – 1.10)                                    | <b>2.13</b> (1.43 – 2.84)    |
| Jungfraujoch           | 2008 – 2023 | 0.20 $\pm$ 0.16                                                                  | 0.03                                                                            | <b>0.38</b> (0.13 – 0.63)                                    | <b>1.89</b> (0.66 – 3.07)    |
| Toronto                | 2002 – 2023 | 7.98 $\pm$ 8.53                                                                  | 0.97                                                                            | <b>28.40</b> (21.20 – 35.40)                                 | <b>4.68</b> (3.53 – 5.82)    |
| Boulder                | 2010 – 2022 | 9.00 $\pm$ 7.42                                                                  | 1.25                                                                            | <b>37.30</b> (26.30 – 48.30)                                 | <b>5.93</b> (4.11 – 7.75)    |
| Tsukuba                | 2014 – 2022 | 4.64 $\pm$ 3.45                                                                  | 0.63                                                                            | <b>19.10</b> (7.02 – 31.40)                                  | <b>5.16</b> (1.82 – 8.46)    |
| Los Angeles            | 1985 – 2024 | 11.31 $\pm$ 7.07                                                                 | 0.21                                                                            | <b>23.4</b> (19.60 – 27.40)                                  | <b>4.86</b> (4.07 – 5.69)    |
| Hefei                  | 2017 – 2022 | 19.20 $\pm$ 13.50                                                                | 4.32                                                                            | <b>73.80</b> (16.60 – 132)                                   | <b>4.36</b> (0.78 – 7.90)    |
| Izana                  | 2000 – 2022 | 0.42 $\pm$ 0.47                                                                  | 0.34                                                                            | <b>0.38</b> (0.03 – 0.74)                                    | <b>0.88</b> (0.07 – 1.69)    |
| Mexico City            | 2012 – 2022 | 14.50 $\pm$ 6.89                                                                 | 3.14                                                                            | <b>93.0</b> (78.8 – 107)                                     | <b>12.8</b> (10.86 – 14.79)  |
| Altzomoni              | 2012 – 2022 | 1.63 $\pm$ 2.36                                                                  | 0.46                                                                            | <b>7.08</b> (2.56 – 11.50)                                   | <b>10.73</b> (3.94 – 17.46)  |
| Paramaribo             | 2018 – 2019 | 3.02 $\pm$ 4.85                                                                  | 0.38                                                                            | –                                                            | –                            |
| Porto Velho            | 2019        | 11.80 $\pm$ 9.82                                                                 | 1.83                                                                            | –                                                            | –                            |
| Reunion St. Denis      | 2004 – 2011 | 1.21 $\pm$ 1.04                                                                  | 0.27                                                                            | –                                                            | –                            |
| Reunion Maïdo          | 2013 – 2023 | 0.34 $\pm$ 0.29                                                                  | 0.07                                                                            | 0.16 (-0.53 – 0.84)                                          | 0.49 (-1.73 – 2.59)          |
| Wollongong             | 2007 – 2022 | 2.62 $\pm$ 2.12                                                                  | 3.03                                                                            | <b>-3.78</b> (-7.10 – -0.35)                                 | <b>-0.93</b> (-1.79 – -0.08) |
| Lauder                 | 1996 – 2022 | 3.15 $\pm$ 2.95                                                                  | 0.95                                                                            | <b>2.37</b> (0.86 – 3.85)                                    | <b>1.63</b> (0.60 – 2.61)    |
| <b>Median (Q1-Q3)*</b> | –           | –                                                                                | –                                                                               | 4.725 (0.38 – 24.6)                                          | 3.27 (1.44 – 4.94)           |

\* The cross-site median was calculated for FTIR records spanning at least 8 years.

**- There are several problems with the reported statistics:**

**(1) a mean of absolute trends does not make sense, as these trends are calculated from data from stations whose average columns span three orders of magnitude. The mean trend is mainly representative of the high-column stations.**

Response: We agree with this observation. The all-site mean absolute trend has been removed. Cross-site FTIR trends are now summarized using the median and interquartile range (Q1–Q3). In addition, to reduce the influence of short records, we introduced a minimum record-span criterion of 8 years for inclusion in the FTIR cross-site trend summary, supported by Liu et al. (2024). Garmisch and Hefei are therefore excluded from the cross-site summary, while their individual trends remain reported. The median and interquartile range (Q1–Q3) was added to Table 2, and the mean all row was removed from Table 7, as this table is intended as a site by site comparison.

New text added to Section 2.3

“To obtain a robust cross-site summary of the relative trends reported in Tables 2 and 7, only long FTIR records (Liu et al., 2024) spanning at least 8 years were considered. The cross-site relative trends are reported as the median and interquartile range (Q1–Q3).”

Tables 2 and 7 were updated accordingly.

**Table 2.** NH<sub>3</sub> mean total columns  $\pm 1\sigma$  and absolute trends with 95% confidence intervals for the FTIR sites. Bold indicates a significant trend.

| Station                | Measurement | NH <sub>3</sub> Mean Total Column<br>(molec. cm <sup>-2</sup> ) $\times 10^{15}$ | NH <sub>3</sub> Mean Total Error<br>(molec. cm <sup>-2</sup> ) $\times 10^{15}$ | NH <sub>3</sub> Trend and 95% CI                             |                              |
|------------------------|-------------|----------------------------------------------------------------------------------|---------------------------------------------------------------------------------|--------------------------------------------------------------|------------------------------|
|                        | Period      |                                                                                  |                                                                                 | (molec. cm <sup>-2</sup> yr <sup>-1</sup> ) $\times 10^{13}$ | % yr <sup>-1</sup>           |
| Eureka                 | 2006 – 2020 | 0.17 $\pm$ 0.24                                                                  | 0.01                                                                            | <b>0.58</b> (0.13 – 1.02)                                    | <b>3.0</b> (0.73 – 5.21)     |
| Ny Alesund             | 2017 – 2019 | 0.12 $\pm$ 0.12                                                                  | 0.06                                                                            | –                                                            | –                            |
| Thule                  | 1999 – 2022 | 0.46 $\pm$ 0.34                                                                  | 0.03                                                                            | <b>-2.10</b> (-2.50 – -1.70)                                 | <b>-7.7</b> (-9.1 – -6.25)   |
| St. Petersburg         | 2010 – 2022 | 3.07 $\pm$ 2.85                                                                  | 0.87                                                                            | <b>11.70</b> (3.54 – 19.60)                                  | <b>4.67</b> (1.44 – 7.81)    |
| Bremen                 | 2004 – 2018 | 16.80 $\pm$ 14.70                                                                | 0.93                                                                            | 28.20 (-8.20 – 63.80)                                        | 3.55 (-1.03 – 8.02)          |
| Garmisch               | 2004 – 2007 | 3.56 $\pm$ 3.97                                                                  | 0.54                                                                            | 31.10 (-13.80 – 76.20)                                       | 10.64 (-6.39 – 28.04)        |
| Zugspitze              | 1995 – 2023 | 0.32 $\pm$ 0.55                                                                  | 0.11                                                                            | <b>0.84</b> (0.58 – 1.10)                                    | <b>2.13</b> (1.43 – 2.84)    |
| Jungfraujoch           | 2008 – 2023 | 0.20 $\pm$ 0.16                                                                  | 0.03                                                                            | <b>0.38</b> (0.13 – 0.63)                                    | <b>1.89</b> (0.66 – 3.07)    |
| Toronto                | 2002 – 2023 | 7.98 $\pm$ 8.53                                                                  | 0.97                                                                            | <b>28.40</b> (21.20 – 35.40)                                 | <b>4.68</b> (3.53 – 5.82)    |
| Boulder                | 2010 – 2022 | 9.00 $\pm$ 7.42                                                                  | 1.25                                                                            | <b>37.30</b> (26.30 – 48.30)                                 | <b>5.93</b> (4.11 – 7.75)    |
| Tsukuba                | 2014 – 2022 | 4.64 $\pm$ 3.45                                                                  | 0.63                                                                            | <b>19.10</b> (7.02 – 31.40)                                  | <b>5.16</b> (1.82 – 8.46)    |
| Los Angeles            | 1985 – 2024 | 11.31 $\pm$ 7.07                                                                 | 0.21                                                                            | <b>23.4</b> (19.60 – 27.40)                                  | <b>4.86</b> (4.07 – 5.69)    |
| Hefei                  | 2017 – 2022 | 19.20 $\pm$ 13.50                                                                | 4.32                                                                            | <b>73.80</b> (16.60 – 132)                                   | <b>4.36</b> (0.78 – 7.90)    |
| Izana                  | 2000 – 2022 | 0.42 $\pm$ 0.47                                                                  | 0.34                                                                            | <b>0.38</b> (0.03 – 0.74)                                    | <b>0.88</b> (0.07 – 1.69)    |
| Mexico City            | 2012 – 2022 | 14.50 $\pm$ 6.89                                                                 | 3.14                                                                            | <b>93.0</b> (78.8 – 107)                                     | <b>12.8</b> (10.86 – 14.79)  |
| Altzomoni              | 2012 – 2022 | 1.63 $\pm$ 2.36                                                                  | 0.46                                                                            | <b>7.08</b> (2.56 – 11.50)                                   | <b>10.73</b> (3.94 – 17.46)  |
| Paramaribo             | 2018 – 2019 | 3.02 $\pm$ 4.85                                                                  | 0.38                                                                            | –                                                            | –                            |
| Porto Velho            | 2019        | 11.80 $\pm$ 9.82                                                                 | 1.83                                                                            | –                                                            | –                            |
| Reunion St. Denis      | 2004 – 2011 | 1.21 $\pm$ 1.04                                                                  | 0.27                                                                            | –                                                            | –                            |
| Reunion Maïdo          | 2013 – 2023 | 0.34 $\pm$ 0.29                                                                  | 0.07                                                                            | 0.16 (-0.53 – 0.84)                                          | 0.49 (-1.73 – 2.59)          |
| Wollongong             | 2007 – 2022 | 2.62 $\pm$ 2.12                                                                  | 3.03                                                                            | <b>-3.78</b> (-7.10 – -0.35)                                 | <b>-0.93</b> (-1.79 – -0.08) |
| Lauder                 | 1996 – 2022 | 3.15 $\pm$ 2.95                                                                  | 0.95                                                                            | <b>2.37</b> (0.86 – 3.85)                                    | <b>1.63</b> (0.60 – 2.61)    |
| <b>Median (Q1–Q3)*</b> | –           | –                                                                                | –                                                                               | 4.725 (0.38 – 24.6)                                          | 3.27 (1.44 – 4.94)           |

\* The cross-site median was calculated for FTIR records spanning at least 8 years.

**(2) There is similar objection for the mean of relative trends. In contrast to the absolute trends, these can be driven by outliers typically seen in stations with a low column average (e.g. Eureka and Ny Alesund in table 8 in the TCR-2 % column). I would propose dropping the all-site mean of absolute trends and providing the**

**median + Q1-Q3 quartiles for the relative trends. In addition, stations with a short record (e.g. Hefei) should be excluded from this by applying some threshold over the number of years covered.**

Response: We agree. The all-site mean relative trend and the corresponding cross-site 95% confidence interval were replaced by the median and interquartile range (Q1–Q3). The same  $\geq 8$ -year record-span criterion is applied to the FTIR cross-site summary. Individual site trends are still reported so that short records are not discarded from the site-by-site analysis.

Table 8 was updated accordingly.

**Table 8.** GCHP and TCR-2 trends in  $\text{NH}_3$  total columns derived from full time series of each model along with 95% confidence intervals. Bold indicates a significant trend. Model values correspond to the nearest model grid cell to each site (no interpolation applied).

| Station                             | GCHP (2003–2021) $\text{NH}_3$ trends and 95% CI            |                              | TCR-2 (2005–2018) $\text{NH}_3$ trends and 95% CI           |                              |
|-------------------------------------|-------------------------------------------------------------|------------------------------|-------------------------------------------------------------|------------------------------|
|                                     | (molec. $\text{cm}^{-2} \text{ yr}^{-1}$ ) $\times 10^{13}$ | %                            | (molec. $\text{cm}^{-2} \text{ yr}^{-1}$ ) $\times 10^{13}$ | % $\text{yr}^{-1}$           |
| Eureka                              | 0.87 (-0.42 – 2.19)                                         | 1.99 (-0.97 – 5.01)          | 0.03 (-0.01 – 0.07)                                         | 72.90 (-11.00 – 156.00)      |
| Ny Alesund                          | <b>0.90</b> (0.12 – 1.67)                                   | <b>3.42</b> (0.44 – 6.38)    | 0.05 (-0.01 – 0.11)                                         | 29.64 (-3.90 – 62.00)        |
| Thule                               | 0.81 (-0.24 – 1.91)                                         | 1.05 (-0.31 – 2.47)          | <b>0.03</b> (0.00 – 0.07)                                   | <b>31.11</b> (2.06 – 58.90)  |
| St. Petersburg                      | <b>3.02</b> (1.69 – 4.27)                                   | <b>4.09</b> (2.29 – 5.79)    | <b>-1.12</b> (-2.13 – -0.14)                                | <b>-1.77</b> (-3.36 – -0.21) |
| Bremen                              | <b>43.80</b> (39.60 – 48.30)                                | <b>6.34</b> (5.73 – 6.98)    | <b>5.46</b> (0.98 – 9.99)                                   | <b>0.59</b> (0.11 – 1.08)    |
| Garmisch                            | <b>19.10</b> (16.80 – 21.70)                                | <b>4.23</b> (3.72 – 4.79)    | <b>4.10</b> (0.97 – 7.16)                                   | <b>0.92</b> (0.22 – 1.60)    |
| Zugspitze                           | <b>19.10</b> (16.80 – 21.50)                                | <b>4.23</b> (3.70 – 4.76)    | <b>3.58</b> (0.68 – 6.38)                                   | <b>0.90</b> (0.17 – 1.61)    |
| Jungfraujoch                        | <b>25.30</b> (21.80 – 28.60)                                | <b>4.74</b> (4.10 – 5.36)    | 1.38 (-1.36 – 4.22)                                         | 0.37 (-0.36 – 1.12)          |
| Toronto                             | <b>12.20</b> (10.50 – 14.00)                                | <b>10.14</b> (8.73 – 11.60)  | 2.58 (-0.01 – 5.12)                                         | 0.68 (0.00 – 1.35)           |
| Boulder                             | <b>12.50</b> (10.90 – 14.30)                                | <b>6.40</b> (5.54 – 7.29)    | 0.30 (-0.30 – 0.91)                                         | 0.54 (-0.54 – 1.62)          |
| Tsukuba                             | <b>8.18</b> (5.50 – 11.00)                                  | <b>3.80</b> (2.55 – 5.10)    | <b>12.80</b> (9.49 – 16.10)                                 | <b>5.04</b> (3.74 – 6.34)    |
| Los Angeles                         | <b>7.75</b> (5.24 – 10.20)                                  | <b>2.66</b> (1.80 – 3.50)    | -1.55 (-4.85 – 1.81)                                        | -0.26 (-0.80 – 0.30)         |
| Hefei                               | <b>41.20</b> (27.70 – 54.40)                                | <b>5.48</b> (3.69 – 7.24)    | <b>23.70</b> (9.56 – 37.20)                                 | <b>1.50</b> (0.61 – 2.36)    |
| Izana                               | <b>3.91</b> (2.99 – 4.81)                                   | <b>8.58</b> (6.57 – 10.60)   | -0.19 (-0.51 – 0.12)                                        | -1.59 (-4.38 – 1.05)         |
| Mexico City                         | <b>6.20</b> (4.41 – 7.98)                                   | <b>3.62</b> (2.57 – 4.66)    | <b>-1.77</b> (-2.37 – -1.18)                                | <b>-2.58</b> (-3.46 – -1.72) |
| Altzomoni                           | <b>6.16</b> (4.32 – 7.95)                                   | <b>3.60</b> (2.52 – 4.64)    | <b>-1.77</b> (-2.37 – -1.15)                                | <b>-2.58</b> (-3.46 – -1.68) |
| Paramaribo                          | <b>0.64</b> (0.32 – 0.95)                                   | <b>1.24</b> (0.62 – 1.84)    | <b>-0.13</b> (-0.21 – -0.05)                                | -2.10 (-3.42 – -0.81)        |
| Porto Velho                         | <b>-5.84</b> (-10.60 – -1.12)                               | <b>-1.36</b> (-2.47 – -0.26) | -5.55 (-11.00 – 0.03)                                       | -1.63 (-3.23 – 0.01)         |
| Reunion St. Denis                   | <b>0.11</b> (0.04 – 0.19)                                   | <b>0.67</b> (0.21 – 1.12)    | <b>1.09</b> (0.94 – 1.24)                                   | <b>6.51</b> (5.61 – 7.43)    |
| Reunion Maïdo                       | <b>0.11</b> (0.04 – 0.18)                                   | <b>0.65</b> (0.21 – 1.09)    | <b>1.09</b> (0.94 – 1.24)                                   | <b>6.51</b> (5.61 – 7.44)    |
| Wollongong                          | <b>2.69</b> (0.29 – 5.17)                                   | <b>1.60</b> (0.17 – 3.08)    | <b>-1.65</b> (-2.46 – -0.85)                                | <b>-1.95</b> (-2.90 – -1.01) |
| Lauder                              | <b>4.42</b> (3.31 – 5.54)                                   | <b>3.36</b> (2.52 – 4.22)    | -0.02 (-0.74 – 0.68)                                        | -0.02 (-0.59 – 0.54)         |
| <b>Median (Q1-Q3) for all sites</b> | 5.29 (0.87 – 12.43)                                         | 3.61 (1.70 – 4.61)           | 0.04 (-0.89 – 2.28)                                         | 0.56 (-1.62 – 4.15)          |

**(3) mean NRMSE/MRD are likewise determined by outliers. For instance in Table 3, the mean of MRD is close to 0, but only because of a fortunate cancellation of several outliers above 100% with otherwise mostly negative values. I would recommend replacing these means with median and/or Q1-Q3 quartiles.**

Response: We agree. In Table 3, cross-site means of the site-level bias and error metrics were replaced by the median and interquartile range (Q1–Q3). We also replaced the pointwise mean relative difference (MRD) with a site-level normalized mean bias (NMB), which avoids the disproportionate influence of individual very small FTIR columns on pointwise percentage differences. NRMSE is likewise summarized across sites using the median and Q1–Q3.

The manuscript was updated accordingly.

Previous text

“To compare the simulated and FTIR total columns, correlations for the simulation-measurement pairs were performed, in addition to the calculation of simulation minus measurement relative differences (RD) as defined by Equation 3 (Dammers et al., 2016), where  $\text{TCS}_i$  and  $\text{TCF}_i$  are the total columns for the simulations and FTIR, respectively. The mean is calculated to obtain the mean relative difference (MRD) for site  $i$  between the simulation and the FTIR measurements:”

Revised text:

“The comparisons between the FTIR and simulated NH<sub>3</sub> total columns were first performed by calculating the normalized mean bias (NMB) of the temporally matched time series as defined by Equation 3. This formulation provides an overall measure of systematic bias while avoiding the disproportionate influence of individual very small FTIR columns on point-wise relative differences. Here,  $TCS_{i,j}$  and  $TCF_{i,j}$  are the mean simulated and FTIR total columns, respectively, calculated from the temporally matched observations at site  $i$ . An  $NMB=0$  indicates no net bias,  $NMB > 0$  indicates model overestimation, and  $NMB < 0$  model underestimation.”

**(4) The "Mean for all sites" row in Table 3 uses two different methods: the mean slope and R (not sure as the text doesn't specify it) were calculated from all the individual datapoints while the mean MRD and NRMSE are per-site averages.**

Response: We clarified this distinction explicitly. The R value and regression slope in the “All pooled coincidences” row are calculated from all temporally matched model–measurement pairs pooled across sites, whereas NMB and NRMSE are site-level metrics summarized across sites using the median and Q1–Q3. The original mixed “Mean for all sites” row has been removed.

Updated Table 3:

**Table 3.** Results of the NH<sub>3</sub> total column comparisons of GCHP and TCR-2 with the FTIR measurements.

| Station                 | GCHP     |       |                 |             | TCR-2    |       |                  |             |
|-------------------------|----------|-------|-----------------|-------------|----------|-------|------------------|-------------|
|                         | <i>R</i> | Slope | NMB<br>%        | NRMSE       | <i>R</i> | Slope | NMB<br>%         | NRMSE       |
| Eureka                  | 0.02     | 0.03  | -30.22          | 1.96        | 0.81     | 0.05  | -97.22           | 1.14        |
| Ny Alesund              | 0.08     | 0.13  | 106.90          | 2.13        | 0.85     | 0.06  | -73.60           | 1.19        |
| Thule                   | 0.09     | 0.48  | 318.13          | 6.58        | 0.12     | 0.02  | -97.62           | 1.59        |
| St. Petersburg          | 0.51     | 0.44  | -18.06          | 0.95        | 0.39     | 0.16  | -67.67           | 1.19        |
| Bremen                  | 0.12     | 0.07  | -25.31          | 1.14        | 0.23     | 0.11  | -14.79           | 1.02        |
| Garmisch                | 0.29     | 0.50  | 222.00          | 2.66        | 0.18     | 0.20  | 112.14           | 1.66        |
| Zugspitze               | 0.35     | 0.54  | 139.31          | 1.71        | 0.36     | 0.32  | -0.32            | 1.07        |
| Jungfraujoch            | 0.33     | 0.35  | -18.08          | 1.22        | 0.26     | 0.08  | -91.97           | 1.46        |
| Toronto                 | 0.66     | 0.39  | -44.23          | 0.87        | 0.36     | 0.22  | -16.72           | 0.98        |
| Boulder                 | 0.49     | 0.34  | -12.41          | 0.91        | 0.46     | 0.08  | -88.02           | 1.40        |
| Tsukuba                 | 0.49     | 1.15  | -6.60           | 2.06        | 0.45     | 0.47  | -23.66           | 1.12        |
| Los Angeles             | 0.56     | 0.24  | -64.32          | 1.37        | 0.45     | 0.26  | -42.75           | 1.21        |
| Hefei                   | 0.40     | 0.87  | -17.82          | 1.99        | 0.07     | 0.15  | 97.44            | 2.56        |
| Izana                   | 0.19     | 0.19  | -15.73          | 1.29        | -0.04    | -0.01 | -96.18           | 1.32        |
| Mexico City             | 0.38     | 0.13  | -79.48          | 1.90        | 0.16     | 0.03  | -88.12           | 2.13        |
| Altzomoni               | 0.47     | 0.10  | -73.64          | 1.04        | 0.22     | 0.00  | -96.01           | 1.21        |
| Paramaribo              | 0.07     | 0.01  | -84.97          | 1.13        | 0.09     | 0.00  | -97.07           | 1.72        |
| Porto Velho             | 0.87     | 0.69  | -30.24          | 0.62        | NA       | NA    | NA               | NA          |
| Reunion St. Denis       | 0.23     | 0.05  | -80.48          | 1.34        | -0.02    | 0.00  | -74.56           | 1.33        |
| Reunion Maïdo           | 0.59     | 0.11  | -83.89          | 1.33        | 0.13     | 0.00  | -98.86           | 1.60        |
| Wollongong              | 0.51     | 0.38  | -8.15           | 0.89        | 0.23     | 0.09  | -62.58           | 1.26        |
| Lauder                  | 0.27     | 0.12  | -56.90          | 1.15        | 0.25     | 0.09  | -55.46           | 1.13        |
| All pooled coincidences | 0.36     | 0.27  | –               | –           | 0.28     | 0.22  | –                | –           |
| Median across sites     | –        | –     | -21.69          | 1.31        | –        | –     | -73.60           | 1.26        |
| (Q1–Q3)                 | –        | –     | (-62.46– -9.22) | (1.06–1.95) | –        | –     | (-96.01– -23.66) | (1.14–1.59) |

**- P3 reviews the literature on NH<sub>3</sub> trends, could cite/discuss the following (if found relevant)**

- (1) <https://doi.org/10.1021/acs.est.4c14020>
- (2) <https://doi.org/10.1016/j.scitotenv.2024.176846>
- (3) <https://doi.org/10.3390/atmos14071056>
- (4) <https://doi.org/10.1016/j.envint.2024.108519>

Response: We thank the reviewer for suggesting these studies. We reviewed all four references and considered their relevance to the scope of the present work. Of these, Liu et al. (2024; <https://doi.org/10.1016/j.envint.2024.108519>) was incorporated into the revised manuscript because of its direct relevance to the analysis of long-term NH<sub>3</sub> trends from multi-year observational records. In particular, this study helped support the introduction of a minimum record-length criterion for the cross-site trend summary. The other suggested studies were also reviewed but were not incorporated, as their specific focus was less directly relevant to the methodological changes and discussion retained in the revised manuscript.

**- Figures:**

**(1) All the "full page" figures are in-between single column and double column figures. Some of them are very squashed, so I would strongly recommend increasing their width, height or both to cover the entire page.**

Response: We reviewed all full-page figures and adjusted their dimensions to use the available page width more effectively and improve readability. In particular, Figures 1–5 were reformatted and enlarged where appropriate.

**(2) Figures A1-A5 could be combined in a multipanel figure**

Response: We combined the former Figures A1–A4 into a single four-panel Figure A1 showing the NH<sub>3</sub>–CO enhancements at Porto Velho, Reunion Mado, Los Angeles, and Mexico City. The former Figure A5 addresses a distinct volcanic-event analysis and was therefore revised separately as Figure A2, as described in our response to the volcanic NH<sub>3</sub> comment above.

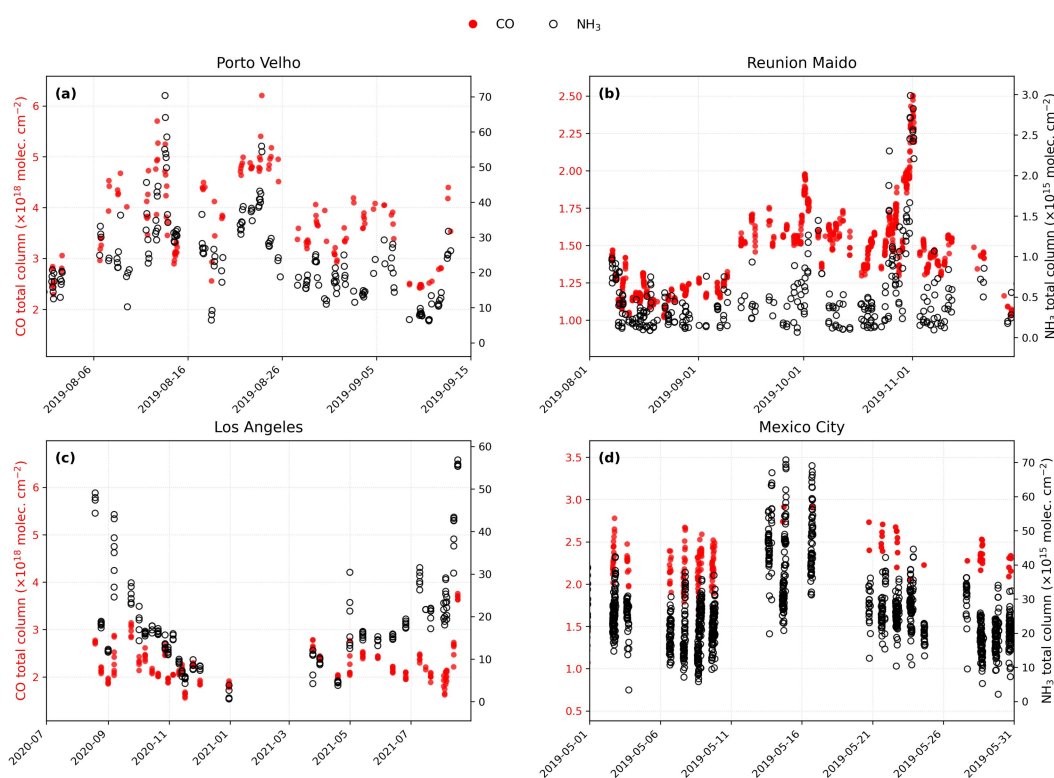


Figure A1. Comparison of NH<sub>3</sub> and CO total columns measured at (a) Porto Velho, (b) Reunion Mado, (c) Los Angeles and (d) Mexico City. Simultaneous enhancements can be observed, attributed to biomass burning events.

### Typos and inconsistencies

**P5, Table 1: Lauder should be 169.68°E not W; For Eureka, the ° misplaced**

Response: Thank you. The longitude values were corrected to 169.68°E for Lauder and 86.42°W for Eureka.

**P6, line 145: remove "very"**

Response: Thank you. The word "very" was removed.

**P6, Sect. 2.2.1–2.2.2: HTAP is cited as Janssens-Maenhout et al. (2015) for both the HTAPv3 GCHP sensitivity run and HTAPv2 TCR-2, but that reference is HTAPv2.2. The 0.1°, 2000–2018 coverage corresponds to HTAPv3 (Crippa et al., 2023), which is not cited. Please clarify which inventory/version and years were used for each simulation and correct the citation.**

Response: Thank you for pointing out the inconsistency in the inventory versions and citations. We have clarified the inventories used in the two model configurations and corrected the corresponding references. The Crippa et al., (2023) reference was added.

For the GCHP sensitivity experiment, we used HTAPv3 emissions (Crippa et al., 2023), which provide monthly emissions at  $0.1^\circ \times 0.1^\circ$  resolution for 2000–2018. A one-month sensitivity simulation was performed for February 2013, replacing CEDS emissions with the corresponding HTAPv3 emissions while retaining the same model configuration and meteorology. We have clarified that this simulation is a single-month case study intended to examine the sensitivity of the simulated NH<sub>3</sub> columns to the choice of emissions inventory under the February 2013 conditions, rather than a climatological assessment of the differences between HTAPv3 and CEDS.

For TCR-2, the anthropogenic emissions are based on HTAPv2.2 (Janssens-Maenhout et al., 2015), which provides  $0.1^\circ \times 0.1^\circ$  monthly emission fields for 2008 and 2010. In the TCR-2 configuration used here, the 2008 emissions were applied for 2005–2008 and the 2010 emissions for 2009–2018. The manuscript has been revised to distinguish clearly between HTAPv3 used in the GCHP sensitivity experiment and HTAPv2.2 used in TCR-2

The manuscript was updated as follows for

Section 2.2.1:

Previous text

"In addition to the 2003–2021 GCHP run, a one-month simulation was performed to explore the effect of changing the global emissions, by using the Hemispheric Transport of Air Pollution (HTAP) v3 emissions (Janssens-Maenhout et al., 2015), instead of CEDSv2, maintaining the same model configuration."

Revised text

"In addition to the 2003–2021 GCHP simulation, a one-month sensitivity experiment was performed for February 2013 using the Hemispheric Transport of Air Pollution (HTAP) v3 anthropogenic emissions (Crippa et al., 2023) instead of CEDS, while retaining the same model configuration and meteorological conditions. The HTAPv3 emission inventory has a  $0.1^\circ \times 0.1^\circ$  resolution between 2000 and 2018."

Section 2.2.2

Previous text

"A priori anthropogenic emissions used in the reanalysis are from the HTAPv2 emissions (Janssens-Maenhout et al., 2015), biomass burning emissions from GFED v4 (Randerson et al., 2017), and soil emissions from GEIA

(Graedel et al., 1993). The HTAP emission inventory has a  $0.1^\circ \times 0.1^\circ$  resolution between 2000 and 2018 and is used to study trends and transport of air pollutants.”

Revised text

“A priori anthropogenic emissions used in the reanalysis are from the HTAPv2.2 emissions (Janssens-Maenhout et al., 2015), biomass burning emissions from GFED v4 (Randerson et al., 2017), and soil emissions from GEIA (Graedel et al., 1993). The HTAP emission inventory provides emissions fields for 2008 and 2010 at a  $0.1^\circ \times 0.1^\circ$  resolution, data for 2008 was used for 2005 2008 and 2010 emissions for 2009-2018.”

Section 4.3

The terminology in Section 4.3 was revised throughout to distinguish explicitly between HTAPv3 used in the GCHP sensitivity experiment and HTAPv2.2 used in TCR-2.

**P13, line 301: duplicate "over Europe"**

Response: Thank you. The duplicate phrase “over Europe” was removed.

**P16: The NRMSE is missing a 1/N**

Response: Thank you. The missing 1/N term was added to the NRMSE equation.

**P18: the stated TCR-2 minimum is incorrect, it is -0.04 for Izaña and not -0.02 at Reunion**

Response: Thank you. The manuscript was corrected to: “For TCR-2, R varies from a minimum of -0.04 at Izaña to a maximum of 0.85 at Ny Ålesund.”

**P25, line 415: "despite the HTAP emissions being smaller than the CEDS emissions" contradicts what is written on line 412.**

Response: We agree that the original wording was inconsistent. The sentence was removed. Section 4.3 now treats the February 2013 HTAPv3–CEDS experiment strictly as a one-month sensitivity test and emphasizes that the response is strongly site dependent. Because TCR-2 uses HTAPv2.2 rather than HTAPv3, the experiment is not used to directly attribute the GCHP–TCR-2 differences to the emissions inventory. The revised text no longer states that HTAP emissions are generally smaller than CEDS emissions.

Previous text

“One difference between TCR-2 and GCHP is the a priori input emissions inventory used: CEDS for GCHP and HTAP for TCR-2 as described in Section 2.2. To explore the impact of the emissions inventory, we performed sensitivity tests for GCHP by running the model using HTAP emissions for one month (February 2013). In addition, the stand-alone input emissions from HTAP and CEDS were analyzed independently. The MRD difference for the GCHP-HTAP vs. GCHP-CEDS NH<sub>3</sub> total columns and between HTAP and CEDS NH<sub>3</sub> total emissions are presented in Table 6 and were calculated using Equation 5 (MRD). The total HTAP NH<sub>3</sub> emissions are 10.85% larger than the total CEDS emissions for all sites, with the biggest differences for Los Angeles, Eureka, Hefei, Boulder and St. Petersburg. Among the emissions, the contribution from anthropogenic and seabird emissions the most different sectors.

This sensitivity test suggests that the larger underestimation in the NH<sub>3</sub> total columns from TCR-2 compared with GCHP is not solely due to the a priori input emissions, since the total columns of GCHP-HTAP were, on average, larger than GCHP-CEDS despite the HTAP emissions being smaller than the CEDS emissions.”

Revised text

“One difference between TCR-2 and GCHP is the a priori input emissions inventory used: CEDS for GCHP and HTAP for TCR-2 as described in Section 2.2. To explore the sensitivity of simulated NH<sub>3</sub> columns to the choice of anthropogenic emissions inventory, we performed a one-month GCHP sensitivity experiment in which CEDS was replaced by HTAPv3 emissions, while retaining the same model configuration and meteorological conditions. Because HTAPv3 differs from the HTAPv2.2 inventory used in TCR-2, this experiment is not intended to reproduce the TCR-2 emissions configuration, but rather to provide a case-study assessment of the sensitivity of GCHP NH<sub>3</sub> to the use of an alternative global emissions inventory. In addition, the stand-alone

input emissions from HTAPv3 and CEDS were analyzed independently. The relative difference for the GCHP-HTAPv3 vs. GCHP-CEDS NH<sub>3</sub> total columns and between HTAPv3 and CEDS NH<sub>3</sub> total emissions are presented in Table 6 and were calculated using: Equation 6 (RD)....where X represents the February 2013 mean of either the NH<sub>3</sub> total column or the NH<sub>3</sub> emission at site j.

Across the sites, the median relative difference between the February 2013 mean GCHP-HTAPv3 and GCHP-CEDS NH<sub>3</sub> total columns was 9.43% (Q1–Q3: -12.50% to 161.57%), while the median relative difference between HTAPv3 and CEDS NH<sub>3</sub> total emissions was -4.11% (Q1–Q3: -14.24% to 24.77%). The sign and magnitude of the relative differences varied substantially among sites, indicating strong spatial variability in the effect of changing the emissions inventory. The sites with negative HTAPv3-CEDS RD in the total columns were Eureka, Tsukuba, and all sites with a latitude south of Izana, while Bremen, Garmisch, Zugspitze, Jungfraujoch, Toronto, Boulder and Los Angeles presented large positive relative differences for the total columns.

This sensitivity test showed that changing the anthropogenic emissions inventory can substantially affect simulated NH<sub>3</sub> total columns and that the response is site dependent. However, because this experiment uses HTAPv3 rather than the HTAPv2.2 inventory used in TCR-2 and is limited to February 2013, it cannot directly quantify the contribution of emissions-inventory differences to the discrepancies between GCHP and TCR-2. "

**P26/29: "NH<sub>3</sub> prevails in the particulate phase", more accurate is to say that the particulate form dominates.**

Response: We agree and revised the wording accordingly. The manuscript now states that the particulate form dominates, rather than that NH<sub>3</sub> "prevails in the particulate phase." The revised text reads: "The NH<sub>3</sub>/NH<sub>4</sub><sup>+</sup> ratio is less than one for all sites except Porto Velho and Lauder, meaning that the particulate form dominates." A corresponding statement in the discussion was also revised to refer explicitly to particulate NH<sub>4</sub><sup>+</sup>.

**P27, Table 7: The 20.9 for Los Angeles is not inside the CI**

Response: Thank you. The upper confidence-interval endpoint for Los Angeles was corrected from 3.85 to 38.5.

**P28, Table 8: Can you double check the identical TCR-2 values of Mexico city and Altzomoni? The 2.58 should probably be -2.58, otherwise the CI are wrong**

Response: Thank you for pointing this out. We rechecked the TCR-2 trends for Mexico City and Altzomoni. The relative trend should indeed be negative at both sites; the positive sign in the original table was a typo. The corrected relative trend is -2.58% yr<sup>-1</sup> for both sites. The identical trend values are expected because Mexico City and Altzomoni are represented by the same nearest TCR-2 grid-cell time series at the model resolution. The corresponding 95% confidence intervals are -3.46 to -1.72% yr<sup>-1</sup> for Mexico City and -3.46 to -1.68% yr<sup>-1</sup> for Altzomoni.

**P29: The seasonal 2.66-6.92 and diurnal 0.20-6.74 numbers in the conclusion do not correspond to what is given in the paper.**

Response: We rechecked the values used in the Conclusions and corrected the quoted ranges so that they match the final seasonal and diurnal figures. The seasonal range is now 0.07–42.4, and the diurnal range is 0.02–39.4.

**P33: Tarjogaite should be Tajogaite**

Response: Thank you. The typo "Tarjogaite" was corrected to "Tajogaite" in the caption of Figure A2.