RC2: 'Comment on egusphere-2026-2838', Anonymous **Referee #1**, 28 Aug 2026

This manuscript by Zhang et al. reports mobile measurements of ammonia, methane, CO, and CO2 in a 160-km trip in the central Netherlands. Ammonia was measured by a commercially available open-path instrument (HT hereafter) aboard a customized mobile platform, together with multiple other tracers measured by an Aerodyne TILDAS. The results are presented as the enhancement ratios of ammonia and methane, point-source emission rates of ammonia and methane inverted using Gaussian plume model, and emission factor of ammonia from on-road sources. The main novelty seems to be the application of the commercial HT sensor in a mobile setting. The overall rigor and quality of this manuscript should be improved before publication.

*One critical issue is the **QA/QC** of the ammonia measurements, as it is tied to most of the emission-related results of this work.*

Response to reviewer 1:

We thank the reviewer for the careful evaluation of our manuscript and for the constructive comments and suggestions. We appreciate the reviewer's recognition of the novelty of applying the commercial open-path HT analyzer in a mobile measurement setting. We also acknowledge the concerns regarding the overall rigor of the manuscript, particularly the QA/QC of the NH_3 measurements. In response, we have addressed these comments through additional analyses and revisions where possible, while several further checks and revisions are in progress. These changes include a more comprehensive description of the instrument calibration and validation, clarification of the measurement and data-analysis procedures, and strengthened interpretation of the emission-related results. Our detailed responses to each comment, including completed and planned revisions, are provided below.

Comment 1.1

Section 2.3.1 (HT calibration) of the main text is very vague, only referring to Swart et al. (2023) without providing any details. Text S2, HT calibration and validation, is much more informative and should be incorporated into the main text.

Response 1.1

We agree with Reviewer 1's comment. We will incorporate some detailed description of the HT calibration and validation, previously provided in Text S2 of the Supplement, into **Section 2.3.1** of the main text. This revision can provide the relevant calibration procedure and validation results directly in the main text rather than referring only to Swart et al. (2023).

Comment 1.2

The Swart et al. (2023) paper intercompared the miniDOAS and HT and showed reasonable agreement (its Figs. 5 and 6), in contrast to the nonlinear relationship in Figs. S4 and S5 where HT is ~3 times lower. In addition, the HT results are corrected to match mini-DOAS with a quadratic polynomial function.

This is over a range of ~0-60 microgram/m3, much lower than the on-road peaks, up to over 1000 ppb. Extrapolating a quadratic function will likely cause very large errors. According to Fig. S4a, the data points in the upper range (30-60 microgram/m3) are very sparse, making the calibration curve quite questionable. ***What if a linear curve is fitted using miniDOAS data from 20 to 60?***

The reported ammonia concentration may get significantly lower. How will that change the conclusions?

Response 1.2

We agree that extrapolation of the polynomial correction beyond the concentration range constrained by the HT–miniDOAS comparison requires further evaluation, particularly because relatively few calibration observations were available at the upper end of the comparison range. We will therefore perform the suggested sensitivity analysis by fitting a linear relationship to the higher-concentration miniDOAS observations (20–60 µg m^{-3}) and comparing the resulting corrected HT concentrations with those obtained using the polynomial correction.

Importantly, the HT–miniDOAS field comparison extended to approximately 60 µg m^{-3} NH_3 (~80 ppb), which encompasses the large majority of the NH_3 enhancements observed during the mobile campaign. Among all 7142 enhanced NH_3 observations (>3 ppb), the median, P90, P95, and P98 enhancements were 4.7, 20.3, 33.0, and 88.9 ppb, respectively. Based on this distribution, only approximately 2–3% of the enhanced observations exceeded ~80 ppb. The very high concentrations (P99 = 235.9 ppb; maximum = 1,298.4 ppb) therefore represent a small fraction of the dataset. The concentration distribution is also illustrated in the revised Fig. S1a, where the color scale is capped at P95 (33 ppb) to prevent these relatively few high values from dominating the visualization.

The same pattern is evident for the source-specific analyses. Of the 55 plumes used for source characterization, 43 (78%) had NH_3 concentrations below 60 µg m^{-3}, including all 31 single-species livestock plumes, all four mixed-species livestock plumes, all three traffic plumes, two of seven manured-field plumes, and three of four industrial plumes. Extrapolation beyond the field-comparison range therefore primarily concerns a subset of the higher-concentration manured-field and industrial plumes rather than the livestock sources that constitute the majority of the source-characterization dataset.

For the separate traffic emission-factor analysis, only 4 of 627 traffic plume events exceeded ~80 ppb. The calibration extrapolation therefore has very limited relevance for this analysis.

We will assess the sensitivity of the higher-concentration observations and the resulting NH_3:CH_4 ratios and emission estimates to the alternative linear calibration suggested by the reviewer. Given that approximately 97–98% of the enhanced observations, and almost all traffic events, fall within or close to the concentration range covered by the field comparison, we expect the influence of the extrapolation method to be concentrated in the relatively small number of high-NH_3 events. It will be explicitly included in the revised uncertainty discussion.

Comment 1.3

*The **TNO** report cited in line 158 provides more details. It should either support the measurements in this manuscript or be removed as it is distractive and irrelevant if applied on a different instrument.*

Response 1.3

We agree with the reviewer that the calibration results reported in Zhang et al. (2025) should not be used to support or validate the measurements presented in this study, as those experiments were conducted using a different HT unit. We have therefore removed the discussion of those calibration results from the manuscript. However, we retain Zhang et al. (2025) solely as a reference for the VSL traceable NH_3 calibration setup, which became available after the 2023 campaign. We have revised the text to clearly distinguish this later calibration capability from the miniDOAS-based field calibration applied to the HT instrument used in the present study. No calibration results obtained with the different HT unit were used to correct or validate the 2023 campaign measurements. The revised text reads:

"At the time of the 2023 campaign, the later traceable VSL NH_3 calibration approach (Zhang et al.,2025) was not available to us; therefore, the co-located miniDOAS measurements served as the available independent reference for evaluating and correcting the HT measurements. The specific HT unit deployed during the campaign was subsequently replaced and was no longer available for retrospective calibration."

Comment 1.4

The Gaussian plume model roughly introduced in Section 2.4.2 could be extremely uncertain. So many assumptions would break with single drive by measurements. A few references for more rigorous consideration of Gaussian plume-based inversion for mobile measurements:

https://doi.org/10.1021/acs.est.5b05059,

https://doi.org/10.5194/acp-18-15145-2018,

https://doi.org/10.1021/acs.est.2c05373.

Response 1.4

We thank the reviewer for this important comment and for suggesting these relevant references. We agree that Gaussian plume inversion based on only one or two mobile transects can have substantial uncertainty, particularly due to atmospheric turbulence and plume meandering, as well as assumptions regarding steady emissions and meteorological conditions. Previous studies have shown that repeated transects improve the robustness of individual source estimates (Albertson et al., 2016; Caulton et al., 2018; Moore et al., 2023).

Our measurement strategy involved a trade-off between repeated measurements at individual sources and coverage of a large number of agricultural sources within a single measurement day. Consequently, only one or two suitable transects were available for most sources. The Gaussian-derived emissions are therefore intended as indicative snapshot estimates for comparison with inventory-derived emissions rather than precise or temporally representative source emission rates.

Following the reviewer's suggestion, we have added the recommended references and clarified the assumptions and limitations of the Gaussian plume inversion in Section 2.4.2. The uncertainty associated with the limited number of transects is also discussed in Section 4.3.

Comment 1.5

Throughout the text, CO_2 seems to be downplayed, but it is more important than CO in quantifying the emission factor of ammonia from on-road traffic. CO2 should be mentioned and data should be shown in the context of fossil fuel sources.

Response 1.5:

We agree that CO_2 plays a more direct quantitative role than CO in the calculation of the on-road NH_3 emission factor. In the carbon-balance approach used here, the emission factor is derived from the background-corrected NH_3 enhancement relative to the total carbon enhancement represented by CO_2 and CO. We will therefore revise the Abstract, Methods, and traffic analysis to make the role of CO_2 more explicit.

In our mobile measurements, however, short-duration NH_3 peaks generally showed a clearer correspondence with CO than with CO_2. CO therefore remains useful for identifying combustion-related plume events because it has a relatively low and stable background, whereas ambient CO_2 has a larger and more variable regional background. For the emission-factor calculation, we use local background-subtracted CO_2 enhancements (ΔCO_2) rather than absolute CO_2 concentrations. We have revised Figure S1 to show the spatial distributions of ΔNH_3, ΔCO, and ΔCO_2 together, providing a consistent visualization of the three species used in the traffic emission analysis.

Minor comments:

Comment 1.6

Abstract: CO2 should be brought up. Otherwise it is unclear how the emission factor is possible.

Response 1.6:

We have added CO_2 to the Abstract (Lines 15 and 28) to clarify its role in the calculation of the NH_3 emission factor.

Comment 1.7

Lines 69-70: the open-path ammonia instruments presented in the Miller, Sun, and Golston papers are different designs than HT and predate HT, so it is ***improper to refer to them as "HT-based"****. Double check Vecchi et al. 2023 whether it should be included here.*

Response 1.7:

We thank the reviewer for pointing this out. We agree that our previous wording was inaccurate. The open-path NH_3 instruments used in Miller et al. (2015), Sun et al. (2014, 2017), and Golston et al. (2020) are different instrument designs that predate the commercial HT-8700E used in the present study and should therefore not have been described as "HT-based."

We also re-checked Vechi et al. (2023). This study did not use the HT-8700E, but employed mobile optical remote-sensing techniques, specifically Solar Occultation Flux (SOF) for NH_3 emission quantification and mobile extractive FTIR (MeFTIR) for ground-level NH_3 and CH_4 measurements. We have therefore removed Vechi et al. (2023) from this statement.

The Introduction has been revised accordingly. Previous studies are now cited as examples of established mobile NH_3 and multi-species measurement approaches, while the commercial HT-8700E used in the present study is described separately. This distinction also allows us to more precisely position the contribution of the present work without implying that the broader concept of mobile open-path NH_3 measurements is new.

Comment 1.8

Lines 84 and 90: use consistent flux unit.

Response 1.8:

Done. We have converted the NH_3 emission densities to mol N ha^{-1} yr^{-1} to ensure consistency with the nitrogen deposition flux units used in line 90. The revised text in line 84 reads:

"In Gelderland province, where the Veluwe is located, NH_3 emission densities reach approximately 5,300–7,000 mol N ha^{-1} yr^{-1}, roughly three to four times the national average (~1,700 mol N ha^{-1} yr^{-1}) (Dutch National Institute for Public Health and the Environment [RIVM] & CBS, 2024)."

Comment 1.9

Line 118: here is a good place to review and compare different versions of compact, open-path ammonia instruments. Miller et al. (2014, https://doi.org/10.5194/amt-7-81-2014) should be cited here as the first paper describing the instrument. The HT instrument (Wang 2021) is a similar commercially available alternative, but these two should not be confused.

Response 1.9:

We thank the reviewer for this clarification. We have revised the text to distinguish the HT instrument from the earlier open-path QCL NH_3 analyzer described by Miller et al. (2014) and have added the corresponding reference. The revised text reads:

"The HT is a compact open-path quantum cascade laser (QCL) absorption spectrometer designed for fast-response NH_3 measurements at frequencies of up to 10 Hz (Wang et al., 2021). The instrument is conceptually similar to, but independently developed from, the open-path QCL NH_3 analyzer first described by Miller et al. (2014)."

Comment 1.10

Line 122: how is the 5s determined? Does it stay the same in the field?

Response 1.10:

The approximately 5 s delay represents the sample transport time through the closed-path TILDAS inlet and was determined experimentally from the response to a tracer introduced at the inlet, rather than calculated solely from the tubing dimensions and flow rate. During the field measurements, the inlet configuration and sampling flow rate (~5 L min^{-1}) were kept constant; therefore, the transport delay was expected to remain relatively stable. As an additional field check, the relative timing between the TILDAS and open-path HT measurements was verified using coincident plume encounters detected by both instruments. The ~5 s correction was subsequently applied when synchronizing the TILDAS measurements with the open-path NH_3 measurements and GPS data. We have clarified the determination and application of this delay in Section 2.2.1, with further details provided in Supporting material S2.

Comment 1.11

Line 158: Zhang et al. 2025 is a report. Similar calibration applied to the instrument in this study will strengthen this manuscript.

Response 1.11:

We thank the reviewer for this suggestion. At the time of the 2023 measurement campaign, the VSL TRACTOR calibration system was not yet available to us. Therefore, the miniDOAS measurements provided the available independent reference for evaluating and correcting the HT NH_3 measurements, following an approach similar to Swart *et al.* (2023). The specific HT instrument used during the 2023 campaign has since been replaced by a new instrument and is no longer available for additional calibration. Although the VSL TRACTOR system is now available, a calibration performed on the replacement HT instrument cannot be directly transferred to the instrument used during the campaign because the calibration response may differ between individual instruments. We have clarified this limitation in the revised manuscript. (*c.f.* **response 1.3**)

Comment 1.12

Line 161: define OSS. → optical signal strength / value calibration

Response 1.12:

We have defined optical signal strength (OSS) in Section 2.3.1 and clarified that it represents the relative intensity of the laser signal received by the detector and is used as an indicator of measurement quality. The new text reads:

"OSS represents the strength of the laser signal received by the detector and serves as an indicator of the optical performance of the analyzer; low OSS values can occur when contamination of the exposed mirrors reduces the transmitted laser signal (Wang et al., 2021)."

Comment 1.13

Line 190: include CO2 here and similar situations.

Response 1.13:

We have included CO_2 in the relevant parts of the manuscript and revised Section 3.1 to explicitly discuss the observed CO_2 enhancements together with the other measured trace gases.

Comment 1.14

Section 2.5: please clarify what quantity was used in the statistical test.

Response 1.14:

We thank the reviewer for pointing out this ambiguity. We have revised Section 2.5 to clarify that the ΣNH_3:ΣCH_4 ratio was the quantity used in the Mann–Whitney U-test. Specifically, the test was used to assess differences in this ratio between agricultural and non-agricultural source categories, with the ratio derived for each identified plume treated as an individual observation**.**

Comment 1.15

Line 467: should use correlation coefficient, "r" instead of "R2".

Response 1.15:

We thank the reviewer for pointing this out. We have replaced R^2 with the correlation coefficient (r) in the revised manuscript.

Citation**: *https://doi.org/10.5194/egusphere-2026-2838-RC2***