RC1: 'Comment on egusphere-2026-2838', Anonymous Referee #2, 19 Aug 2026

Summary:

This manuscript presents simultaneous measurements of NH3, CH4, CO, N2O, and other trace species using a mobile laboratory to distinguish between agricultural and non-agricultural emission sources in the Netherlands. Although the dataset is constrained by limited sampling (4–5 hours on a single day), this study provides a valuable contribution to the literature, where mobile-platform, multi-species strategies for source separation are currently underrepresented. While this work establishes a useful foundation for future studies, the manuscript requires additional detail in specific sections and refinement to improve clarity. I recommend publication in AMT following the suggested revisions below.

Response to Reviewer 2

We sincerely thank the reviewer for the careful evaluation of our manuscript and for the constructive and encouraging comments. We particularly appreciate the reviewer for pointing us to several recent studies from North America, which provide valuable context for comparison with our measurements in the Netherlands. We also appreciate the reviewer's recognition of the value of our multi-species mobile measurement approach for characterizing agricultural and non-agricultural emission sources.

We acknowledge the limitations associated with the relatively short measurement period and have further clarified the scope and limitations of the study. In response to the reviewer's suggestions, we have addressed a number of comments through additional methodological details, revised figures, updated literature comparisons, and clarification of the interpretation, while several additional checks and revisions are still in progress. Our detailed responses to each comment, including completed and planned revisions, are provided below.

General Comments:

Comment 2.1

While the specific deployment of this instrument suite aboard this mobile laboratory in this region may be unique, characterizing the entire approach as "novel" in the title seems misleading. Given a history of similar mobile sampling techniques reported in the literature, I suggest adjusting the title to more accurately reflect the study's objectives. See additional comments below.

Response 2.1:

We thank the reviewer for this comment. We agree that the term "novel" may overstate the uniqueness of the overall mobile measurement approach, given that similar mobile sampling strategies have previously been reported in the literature. The novelty lies mainly in the application of a mobile ammonia device in the European hotspot using a new commercial type ammonia open-path analyzer. We will revise the title to more accurately reflect the specific objectives and scope of this study, while avoiding a broader claim of novelty.

Comment 2.2

The manuscript lacks a dedicated section on measurement uncertainty. Reported results should incorporate propagated uncertainties associated with measurement systems, analysis methods, and statistical variability.

Response 2.2:

We thank the reviewer for this important comment and agree that the sources and implications of uncertainty should be more explicitly addressed.

There are several studies that check the emission of one or a few sources. Here we try to assess a large group of sources in a single day which means there is less time for repetition of plume drives at each source. So only one or two downwind transects were available for most individual agricultural sources. Previous mobile plume-quantification studies generally rely on multiple repeated transects to characterize the variability of individual emission estimates.

Consequently, the present dataset does not provide sufficient repeated observations to empirically quantify the statistical variability of individual source estimates. Although individual components of uncertainty, such as instrumental precision, can be characterized, a complete propagation of uncertainty through the Gaussian plume inversion would also require a robust estimate of the sampling and transect-to-transect variability. Assigning formal propagated uncertainty bounds to individual source estimates based on only one or two transects could therefore imply a level of statistical confidence that is not supported by the present measurements.

Nevertheless, we agree with the reviewer that the individual sources of uncertainty should be identified and discussed more systematically. We have therefore substantially revised and expanded Section 4.3, now entitled "**Uncertainties and limitations of mobile emission estimates**." The revised section is organized into three subsections: (i) measurement and calibration uncertainty (Section 4.3.1), including instrumental precision, calibration, plume identification, and background determination; (ii) model and sampling uncertainty (Section 4.3.2), including atmospheric transport, Gaussian plume model assumptions, meteorological inputs, and the limited number of repeated transects; and (iii) interpretation of measurement–inventory differences (Section 4.3.3), in which we discuss how these uncertainties, together with temporal source variability and inventory uncertainty, affect the comparison between mobile measurement-based and inventory-based emission estimates.

We have also clarified that the individual agricultural emission estimates should be interpreted as proof-of-concept estimates rather than precise source-specific emission determinations. Nevertheless, comparison with inventory-based estimates remains useful for assessing whether the mobile-derived estimates are of a comparable magnitude, while acknowledging their substantial uncertainty. A more complete quantitative uncertainty assessment will require future measurements with repeated transects of individual sources under comparable conditions.

The situation is different for the traffic analysis, for which approximately 630 individual plume events were available. Statistical variability is therefore characterized directly from the observed distributions within the speed classes; Table 3 reports the geometric mean, geometric standard deviation, median, interquartile range, minimum, and maximum. We have clarified this distinction between the traffic analysis and the more sparsely repeated agricultural-source measurements in the revised manuscript.

Comment 2.3

The manuscript neglects to address the impact of NH_3 gas-particle transformation between emission and sampling. This is an important consideration for atmospheric NH3 sampling given that atmospheric lifetime and transformation timescales can be quite short under certain conditions (like those during the sampling period). The implications of this process should to be discussed accordingly in the context of this work.

Response 2.3:

We agree that atmospheric NH_3 can be converted to particulate NH_4^+ through reactions with acidic species during atmospheric transport, potentially affecting the measured NH_3 concentrations.

In the present study, however, measurements were made close to the identified sources, with source-to-road distances of approximately 50–800 m, corresponding to transport times ranging from seconds to a few minutes depending on wind speed. Over these short distances, we expect gas-to-particle conversion to be limited, particularly in this NH_3-rich agricultural environment where conversion is constrained by the availability of acidic species such as HNO_3 (Schaap et al., 2004).

We therefore consider gas-to-particle transformation to be a secondary uncertainty for the near-source plume measurements, although it cannot be quantified directly because HNO_3 and particulate NH_4^+ were not measured. We will add a brief statement acknowledging this process in the revised uncertainty discussion. Other near-source loss processes, particularly dry deposition, may be more relevant over these distances and are discussed separately in Section 4.3.

Reference:

Schaap, M., van Loon, M., ten Brink, H. M., Dentener, F. J., and Builtjes, P. J. H.: Secondary inorganic aerosol simulations for Europe with special attention to nitrate, Atmospheric Chemistry and Physics, 4, 857–874, 2004. ACP. DOI: https://doi.org/10.5194/acp-4-857-2004.

Comment 2.4

The main paper lacks sufficient cross-referencing to the SI.

Response 2.4:

We thank the reviewer for this comment. We will review the manuscript and add cross-references to the Supplementary Information at the relevant locations throughout the main text, including references to the corresponding supplementary texts, figures, and tables. This should make the supporting information easier to locate and improve the connection between the main manuscript and the Supplement.

Specific Comments:

Comment 2.5

Abstract, L23 ish: It should be acknowledged that the sample size for traffic plumes is significantly smaller than for other sources.

Response 2.5:

We thank the reviewer for pointing this out. We have revised the Abstract to report the number of plumes included for each source category in the ΣNH_3:ΣCH_4 analysis. This makes the differences in sample size among the source categories, including the smaller number of traffic plumes used in this comparison, explicit to the reader. The current text reads:

"Plume-integrated enhancement ratios (ΣNH_3:ΣCH_4) showed systematic differences among source types, based on 31 livestock farms, 4 industrial, 7 manured-field, and 3 traffic plumes. The ratios werewith low ratios for livestock farms (~0.04 for cattle and goats), intermediate values for industrial sources, and substantially higher ratios for manured fields (>2) and traffic plumes (>4)."

Comment 2.6

*P2, L36: Expand the **introduction** to include NH3's role in fine particle formation and its broader implications for regional air quality, rather than focusing solely on its contribution to reactive nitrogen deposition.*

Response 2.6:

We agree that the Introduction focused primarily on the role of NH_3 in reactive nitrogen deposition and did not sufficiently describe its broader relevance to regional air quality through secondary particulate matter formation.

We have therefore expanded the first paragraph of the Introduction to describe the role of atmospheric NH_3 as an important precursor of secondary inorganic aerosol. In the atmosphere, NH_3 reacts with sulfuric and nitric acids to form ammonium sulfate and ammonium nitrate, which contribute to fine particulate matter ($PM_{2.5}$). This pathway links NH_3 emissions not only to nitrogen deposition and ecosystem impacts, but also to regional $PM_{2.5}$ concentrations and associated air-quality and human-health impacts. The revised text therefore provides a broader motivation for improved characterization and quantification of NH_3 emission sources.

Comment 2.7

P2, L70: Clarify that Sun et al. (2014) focused on US environments, while Sun et al. (2017) provided a US-China comparison. Consider adding Miller et al., 2015, https://agupubs.onlinelibrary.wiley.com/doi/10.1002/2015JD023241.

Response 2.7:

We have revised this part of the Introduction more broadly to provide a clearer and more concise overview of previous mobile NH_3 studies. Rather than describing the geographical scope of individual studies, the revised text now groups previous applications according to their scientific focus, including agricultural NH_3 emissions and relationships with co-emitted species (Miller et

al., 2015; Eilerman et al., 2016; Golston et al., 2020), and traffic-related emissions and urban source contributions (Thornhill et al., 2010; Sun et al., 2014, 2017; Pu et al., 2023).

Following the reviewer's suggestion, Miller et al. (2015) has been added as an important earlier application of mobile multi-species measurements to agricultural NH_3 sources. This revised literature context also supports the more precise positioning of the present study described in our response to Comment 2.8.

Comment 2.8

P3, L78: The manuscript overstates the novelty of the mobile sampling framework. While the specific deployment of this instrument suite in the Netherlands may be unique, the broader approach of using open- and closed-path in-situ spectrometers on a mobile platform to distinguish between agricultural and non-agricultural sources is not new and there are publications in the literature reflecting this. Some examples are already identified and referenced in this work (e.g., Golston and Sun). Since there are a finite number of these publications, I recommend acknowledging and expanding the literature review to include:

Miller et al., 2015, https://agupubs.onlinelibrary.wiley.com/doi/10.1002/2015JD023241.

Eilerman et al., 2016, https://doi.org/10.1021/acs.est.6b02851.

Juncosa Calahorrano et al., 2024, https://doi.org/10.1021/acs.est.3c10902.

Tao et al., 2015, https://link.springer.com/article/10.1007/s00340-015-6069-1.

Thornhill et al., 2010, https://acp.copernicus.org/articles/10/3629/2010/.

Reply 2.8

We thank the reviewer for highlighting these previous studies. We agree that the general concept of using mobile open-path and multi-species measurements for source characterization is not new, and we have revised the Introduction to more clearly acknowledge this previous work, including applications to agricultural emissions (Miller et al., 2015; Eilerman et al., 2016; Golston et al., 2020), traffic and urban sources (Thornhill et al., 2010; Sun et al., 2014, 2017; Pu et al., 2023), and the development of multi-species open-path mobile sensing platforms (Tao et al., 2015).

We have also revised the novelty statement to distinguish the established general measurement concept from the specific contribution of the present study. In particular, the open-path NH_3 systems used in several of the previous mobile studies are research-grade instruments and are distinct from the commercial HT-8700E used here. The present work integrates the commercial HT-8700E with a high-resolution TILDAS multi-gas analyzer and evaluates the resulting system through field calibration, data synchronization, plume identification and integrated multi-species analysis. The platform is subsequently applied within a single campaign to characterize both agricultural and traffic-related NH_3 sources, derive NH_{34} relationships and agricultural emission estimates, and estimate traffic NH_3 emission factors.

Accordingly, we no longer describe the general mobile sampling approach itself as novel. Instead, the revised manuscript more specifically identifies the contribution of this study as the integration, field evaluation, and application of the commercial HT-8700E within a multi-species mobile

measurement framework for agricultural and traffic source characterization under real-world conditions in the Netherlands.

Comment 2.9

Section 2.1: Some additional information about the study area would be helpful for readers unfamiliar with the types of sources in this region. For example, it would help to clarify the nature of local livestock operations (e.g., dairy vs. beef; confined/concentrated operations vs. open pasture). Furthermore, identify other potential regional emission sources, such as wetlands, which could significantly influence NH3, N2O, and CH4 signals.

Reply 2.9

We will expand Section 2.1 to provide additional context on the agricultural landscape and relevant emission sources in the study area. The Veluwe is predominantly a dry sandy landscape characterized by forests, heathlands and agricultural areas along and within its margins. Agricultural NH_3 emissions in the region are strongly associated with livestock housing and manure management, with dairy-cattle housing and manure application identified as important agricultural sources in the Veluwe region (RIVM, 2026).

Along our measurement route, the cattle sources were predominantly dairy farms, with only a small number of identified beef-cattle farms; some farms contained mixed cattle types or could not be further classified. Other livestock sources encountered included swine and poultry farms. Because the campaign was conducted in February, the cattle associated with the sampled farms were housed indoors rather than grazing on open pasture. We will clarify these characteristics in the revised description of the study area.

The broader Veluwe also contains localized wet habitats, including fens, wet heath and stream valleys. However, these were not located along or in the relevant upwind footprint of the agricultural plume measurements considered here and were therefore not identified as plausible sources of the observed NH_3–CH_4 plume enhancements. We will clarify this point rather than implying that wetlands are entirely absent from the Veluwe. In addition to agriculture, traffic and the identified industrial sources represent the main non-agricultural sources considered in this study.

Reference:

Ministerie van Landbouw, Natuur en Voedselkwaliteit: Natura 2000-gebied Veluwe (gebied 57), Natura 2000, available at: https://www.natura2000.nl/node/9903 (last access: 29 September 2026).

Comment 2.10

Section 2.2.1: This section could benefit from additional details around the following concepts:

- *Why not use a closed-path NH3 TILDAS instead of the open-path instrument? The closed path NH3 TILDAS has been seen to achieve 1 Hz time resolution or faster in prior works like these: Pollack et al., 2019, https://doi.org/10.5194/amt-12-3717-2019;*

Roscioli et al., 2015, https://doi.org/10.1021/acs.jpca.5b04395. Especially since all measurements are averaged to 1-Hz in the end for analysis.

- *There are distinct trade-offs between open and closed path sampling systems. Some factors include sensitivity to rain, dust, particles, clouds/fog, bugs, temperature, pressure, etc.; ease of calibration and maintenance; power, space, and weight considerations; and of course time resolution. Those differences should be explicitly stated here and again in the discussion in the context of the findings. For example, P8, L232 says that atmospheric conditions were unstable. How does this impact the measurements?*
- *Do the advantages outweigh the disadvantages of an open path measurement if the data are ultimately averaged to 1-Hz for analysis?*
- *Even though the greenhouse gases are not "sticky", did the 6 m long sampling inlet on the TILDAS compromise the time resolution of these measurements? What was the flow rate? Did the sample air go directly from the inlet tip to the analyzer or did the TILDAS sample air from a larger inlet with a larger flow rate? Is the 5 second correction for inlet time delay measured or calculated?*
- *More details are needed on how the open-path NH3 system and the closed-path TILDAS were calibrated. For example for the TILDAS, how and where were cal gases introduced along the sampling line? How many calibration gas mixtures were used? What were their target mixing ratios? Was the inlet tip forward or rear facing? Did you use any filters for dust and particles (Yes)? These details can go in the SI, but it should be available somewhere and called out where to find them in the main paper.*

Reply 2.10

We thank the reviewer for these detailed comments. We agree that additional information on the instrument configuration, sampling system, time response, and calibration procedures is useful for evaluating the mobile measurements. We have therefore expanded **Section 2.2.1** and added further technical details to the Supplement 2.

- **Choice of open-path NH_3 measurement**

We agree that closed-path NH_3 TILDAS instruments can achieve rapid response times of 1 Hz or faster when appropriately configured, as demonstrated by Pollack et al. (2019) and Roscioli et al. (2015). We agree that appropriately configured closed-path NH_3 TILDAS instruments can achieve response times of 1 Hz or faster, as demonstrated by Pollack et al. (2019) and Roscioli et al. (2015). The choice of the open-path HT-8700E for this campaign was primarily based on practical considerations related to the mobile platform, particularly constraints on available power and payload. The open-path instrument could be operated without the additional pumping and cooling capacity required by a closed-path configuration. We have revised the manuscript to clarify this practical motivation and removed statements that could imply that the open-path configuration is generally superior to closed-path NH_3 measurements. A detailed comparison of the advantages and disadvantages of the two configurations is beyond the scope of this study.

- **Advantage & disadvantage of averaged 1-Hz data for open-path analyzer.**

We agree that averaging the measurements to 1 Hz reduces the importance of the higher native time resolution of the open-path instrument, and that suitably configured closed-path NH_3

instruments can also provide measurements at this time resolution. Therefore, we do not consider the 1 Hz time resolution itself to be an advantage of the open-path configuration in this study. As clarified above, the choice of the open-path instrument was primarily based on practical constraints of the mobile platform, particularly limitations in available electrical power and the weight capacity of the mobile platform. We will revise the manuscript to avoid suggesting otherwise.

- **TILDAS inlet configuration and time response**

We have added additional details on the closed-path TILDAS sampling system. Ambient air was drawn through an approximately 6 m sampling line at a flow rate of approximately 5 L min^{-1}. A particle filter was used to protect the sampling system. The TILDAS measurements were recorded at 2 Hz and subsequently averaged to 1 Hz for synchronization with the other measurements. The approximately 5 s transport delay through the inlet was determined experimentally using a tracer response rather than calculated solely from the tubing dimensions and flow rate. This measured delay was applied when synchronizing the TILDAS measurements with the open-path NH_3 observations and GPS data. We additionally verified the relative timing of the open-path NH_3 and closed-path TILDAS measurements using plume encounters in which both systems passed through the same source plume. This provided a field-based check of the synchronization between the two measurement systems.

- **Calibration and inlet settings**

We agree that the calibration and inlet configurations were insufficiently described in the original manuscript. We have therefore added a summary to Section 2.2.1 and provide further technical details in Section S2 of the Supplement. For the TILDAS, two ICOS working standards, calibrated against the ICOS station standards at the Cabauw tall-tower facility, were used. The standards contained approximately 2000 and 2360 ppb CH_4, respectively, together with calibrated concentrations of the other measured species CO, CO_2 and N_2O concentrations. Calibration gas was supplied at ambient pressure through a three-way valve located immediately upstream of the analyzer, allowing switching between ambient sample air and calibration gas. We have also added details of the inlet configuration and particle filtration. The forward-facing inlet was mounted at the upper front of the vehicle, approximately 1.5 m ahead of the trailer. Sample air passed through an approximately 6 m stainless-steel inlet and 1.2 m of 1/4-inch PTFE tubing to a Balston particle filter (~30 mL volume) before entering the analyzer. These details are now provided in Supporting material S2 and referenced from the main Methods section.

Comment 2.11

P5, L122 says that Met data was acquired from a nearby airport. Is there a reason the mobile lab did not have a met sensor onboard during drives?

Reply 2.11

The mobile laboratory was equipped with an onboard meteorological sensor. However, wind measurements obtained while driving were strongly influenced by vehicle motion and vehicle-induced airflow and tended to overestimate wind speed. We therefore did not consider these measurements sufficiently reliable for quantitative plume interpretation and Gaussian plume modelling.

Instead, meteorological data from the nearby weather station were used to provide continuous regional wind speed and direction during the campaign. In addition, a portable 3-D sonic anemometer was deployed at selected locations near agricultural source areas while the vehicle was stationary, providing independent ground-level measurements of local wind speed and direction.

We will clarify this measurement strategy in the Methods section, including the reason for not using the onboard wind measurements and the complementary use of the portable 3-D sonic anemometer.

Comment 2.12

Section 2.3.1: It would help to have more details in the SI about how the open-path NH3 system was calibrated. This is also a good place to explicitly discuss accuracy and precision (or total uncertainty) of the measurements used in this analysis compared to the other instruments mentioned on L158. Along the same lines, what are the uncertainties of the other supporting measurements from the TILDAS?

Reply 2.12

The calibration and validation of the HT-8700E are already described in detail in Supporting material S2, including the co-located comparison with miniDOAS, data-quality filtering, calibration range, derived correction function, and post-correction agreement (Figs. S4–S5). We will refer more explicitly to this section from the main manuscript.

We agree that the instrumental performance of the supporting measurements should be more clearly documented. We have therefore added a concise summary of the precision and calibration information for the HT and TILDAS measurements, with additional TILDAS calibration details provided in the S2 as well.

As discussed in our response to Comment 2.2, we will distinguish these instrument-level uncertainties from the overall uncertainty of the derived agricultural emission estimates. The latter additionally includes uncertainties from plume sampling, meteorological conditions, Gaussian plume modelling, and source variability. Because most agricultural sources were sampled by only one or two transects, a robust propagation of all these components into source-specific uncertainty intervals is not supported by the present dataset. These limitations will be explicitly discussed in the revised Section 4.3.

Comment 2.13

P6, L161: For measurements with OSS<10%, what does this translate to in terms of a detection limit in ppb or ug/m3?

Reply 2.13

OSS is not directly related to the NH_3 detection limit and therefore cannot be converted to a concentration threshold in ppb or $\mu g\ m^{-3}$. OSS is an operational indicator of optical signal quality and decreases when the laser beam is partially obstructed or attenuated, for example by precipitation, dust, or contamination of the mirror surface. Measurements with OSS < 10% were

therefore excluded as a data-quality criterion rather than because NH_3 concentrations were below the instrument detection limit. We have clarified this distinction in Section 2.3.1.

Comment 2.14

P6, L171: What are the other potential sources you found? Can you list them here or later in the results and discussion?

Reply 2.14

Here, "potential sources" refers to nearby upwind sources identified from aerial imagery that could plausibly contribute to the observed plume. For approximately three-quarters of the selected plumes, a single source could be identified, with several source assignments additionally confirmed by farm operators. For the remaining plumes, two or more nearby sources could not be individually resolved from the mobile measurements and were therefore represented as multiple sub-sources in the plume model. We have revised Section 2.3.2 to make this distinction clearer rather than implying that additional unidentified source categories were observed.

Comment 2.15

P6, L178: The findings of Lassman et al., 2020, https://doi.org/10.1016/j.agrformet.2020.107989, are an important consideration for this work and would be worth discussing in the framework of this analysis for context.

Reply 2.15

Thanks for the reviewer pointing out this reference. Lassman et al. (2020) is directly relevant to our analysis as it demonstrates the distance-dependent effect of NH_3 deposition on NH_3 : CH_4 ratios downwind of agricultural sources. We've already discussed near-source NH_3 removal and its potential influence on the observed NH_3:CH_4 ratios in Section 4.2. We had added Lassman et al. (2020) to this discussion and also cite it in Section 2.3.2, where dry deposition and gas–particle partitioning are introduced as processes that may reduce NH_3 plume enhancements with distance.

Our analysis is restricted to well-attributed near-source plumes, typically measured within 50–800 m of the source. We will therefore use the findings of Lassman et al. (2020) to provide additional context for the magnitude of potential NH_3 loss over the source–receptor distances relevant to this study.

Comment 2.16

P7, L208: Can you justify using this approach over the other methods, particularly regression analysis? Can you perform a sensitivity test to show how the results and uncertainties are impacted if one method is used over another? Could the differences in methodology of analysis be used to provide some estimate of the overall uncertainties on the results?

Reply 2.16

The integrated plume ratio was selected because the mobile measurements frequently contain asymmetric and multi-peaked plume structures, for which integration across the complete plume is less sensitive to the exact location and magnitude of individual concentration maxima. In addition, the relatively short duration and limited number of observations within individual mobile plume encounters can make regression slopes sensitive to a small number of points. The integrated approach therefore provides a consistent metric that can be applied across the heterogeneous plume structures encountered in this campaign and is consistent with previous mobile NH_3–CH_4 plume analysis (Miller et al., 2015).

We agree that regression analysis represents an alternative approach. However, we have retained a single, consistent plume-integration method rather than combining estimates derived using different analysis approaches. Differences between integrated ratios and regression slopes would reflect not only measurement uncertainty, but also differences in the statistical treatment and weighting of plume structure; we therefore do not consider their difference to provide a robust quantitative estimate of the overall uncertainty. We have clarified the rationale for selecting the integrated plume approach in Section 2.4.1 and acknowledge the methodological dependence of the derived ratios as a limitation.

Comment 2.17

P8, L245: Can the database be further disaggregated to distinguish between dairy and beef cattle, which can have very different NH3:CH4 ratios?

Reply 2.17

No. The sampled cattle farms in this region were predominantly dairy farms. Three sources were clearly identified as meat-cow farms, while several others were classified more generally as cattle farms because detailed cattle-type information was unavailable or the farms contained mixed cattle types.

The three identified meat-cow farms had plume-integrated NH_3:CH_4 ratios of 2%, 5%, and 9%, which overlap with the range observed for the dairy farms. Given the small number of identified meat-cow farms, we do not consider a separate dairy–beef classification in the present analysis to be statistically supported. We will clarify the composition of the cattle category and this limitation in the revised manuscript.

Comment 2.18

P9, L263: How do processes like respiration and uptake influence the use of CO2 in this context?

Reply 2.18

Respiration and photosynthetic uptake can influence the ambient CO_2 background. However, our traffic analysis is based on background-corrected CO_2 enhancements (ΔCO_2) associated with individual on-road plumes. The local background was determined and subtracted before calculating the NH_3:CO_2 (or NH_3:COx) enhancement ratios. Thus, the more slowly varying influence of respiration and uptake is largely represented in the background, whereas the short-duration CO_2 enhancement above this background is attributed to the passing vehicle plume.

Comment 2.19

P9, L266: Is this a reasonable assumption given that enhanced NH3 emissions have been associated with diesel vehicles outfitted with selective catalytic reduction (SCR) systems? What did the on-road and off-road fleet look like in this region (e.g., are vehicles predominantly gasoline, diesel, EV, hybrid) during the time period of this study? Have SCR systems been implemented in this area?

If yes, when did this happen in relationship to the timeframe of the Burgard 2006 study? See also, Farren et al., 2020, https://doi.org/10.1021/acs.est.0c05839.

Reply 2.19

We agree that the assumption based on Burgard et al. (2006) is too restrictive for the contemporary Dutch vehicle fleet. Diesel vehicles equipped with SCR systems can also emit NH_3 through ammonia slip, and such vehicles were already present well before our 2023 campaign (Farren et al., 2020).

The Dutch passenger-car fleet was still dominated by gasoline vehicles around the measurement period. According to Statistics Netherlands (CBS), passenger cars powered by gasoline or gasoline-hybrid vehicles accounted for approximately 84.0 billion vehicle-km, compared with 19.7 billion vehicle-km for diesel and diesel-hybrid vehicles, with additional contributions from electric and plug-in hybrid vehicles (CBS, 2023). However, vehicle type, fuel, and after-treatment technology were not recorded for the individual ~630 plume encounters, and the measurements may also include commercial and heavy-duty vehicles.

We will therefore remove the assumption that diesel vehicles contribute negligibly to NH_3 emissions and interpret the derived emission factors as representative of the sampled mixed on-road fleet. We will also evaluate the sensitivity of the carbon-balance calculation to the gasoline (WC = 0.85) and diesel (WC = 0.87) carbon fractions; because these values differ by only ~2.4%, this choice is expected to have only a small effect on the derived emission factors compared with the observed traffic variability.

Reference:

CBS (Statistics Netherlands): Verkeersprestaties personenauto's; brandstof uitgebreid, leeftijd, StatLine, available at: https://www.cbs.nl/nl-nl/cijfers/detail/85405NED (last access: 29 September 2026).

Comment 2.20

P10, L292: Are wetlands a potential source in this area?

Reply 2.20

No wetlands were present along the measurement route or in the relevant upwind source areas. Although wetlands can emit CH_4 and under some conditions exchange NH_3, they therefore cannot explain the NH_3–CH_4 plumes attributed to agricultural/manure sources in this campaign. These

source assignments were based on plume location and wind direction relative to identifiable agricultural sources, aerial imagery, and field observations by experienced measurement operators.

Comment 2.21

P10, L299: Simultaneous enhancements in N_2O corroborate this statement. See Eilerman et al., 2016, https://doi.org/10.1021/acs.est.6b02851.

Reply 2.21

The reviewer is correct that the simultaneous N_2O enhancement provides additional evidence for the agricultural source attribution. We will add this observation to the Results and cite Eilerman et al. (2016), who similarly reported co-emitted NH_3, CH_4, and N_2O from livestock operations and demonstrated the usefulness of their enhancement ratios for source characterization.

Comment 2.22

Figure 5: Can you further refine dairy vs beef cattle as the ratios can be quite different for these types of facilities? For example see Juncosa Calahorrano et al., 2023, https://doi.org/10.1029/2023JD039043.

Reply 2.22

We agree that cattle type can influence NH_3:CH_4 ratios, as discussed by Juncosa Calahorrano et al. (2023). However, the cattle sources sampled in our study area were predominantly dairy farms. Only three sources were clearly identified as meat-cow farms; their plume-integrated NH_3:CH_4 ratios were 2%, 5%, and 9%, which fall within the variability observed for the dairy farms. The limited number of meat-cow farms therefore does not support a separate statistical category in Figure 5.

Several additional cattle sources, particularly near the provincial border, could not be classified reliably as dairy or meat cattle from the available information. In addition, one source was identified as a mixed dairy- and meat-cow farm and showed a higher NH_3:CH_4 ratio of 26%. We will therefore retain the broader cattle category in Figure 5, but clarify its composition and the associated classification uncertainty.

We also note that the beef-cattle facilities studied by Juncosa Calahorrano *et al.* (2023) were large Colorado feedlots and therefore differ substantially from the cattle sources sampled here. During our February campaign, the sampled Dutch cattle were housed indoors, whereas the US study represents facility-scale emissions from large feedlot systems under summer conditions. Differences in housing and manure management, as well as meteorological and management conditions, limit a direct comparison of the beef-cattle ratios between the two studies. We will add this distinction when discussing the cattle-source variability.

Comment 2.23

P14, L351: Could a simple Multivariate Linear Regression model (or a Positive Matrix Factorization model if there are enough observations) be applied to these measurements for a source apportionment analysis (maybe something like CO = traffic, N2O = manured fields, NH3 = cattle livestock, CH4 = enteric fermentation, ethane = petrochemical)? MLR examples, include Kille et al., 2019, https://doi.org/10.1029/2019GL082132; McCabe et al., 2023, https://acp.copernicus.org/articles/23/7479/2023/; Pollack et al., 2022, https://doi.org/10.1021/acs.est.1c07382; and Lill et al., 2025, https://doi.org/10.1029/2025JD044019.

Reply 2.23

We thank the reviewer for this interesting suggestion. We have reviewed the suggested applications of multivariate approaches (Kille et al., 2019; Pollack et al., 2022; McCabe et al., 2023; Lill et al., 2025) and agree that MLR and PMF can provide useful source-apportionment information when the available tracers and sampling design provide sufficient constraints. However, we decided not to apply quantitative MLR or PMF source apportionment because the present dataset does not provide a sufficiently constrained basis for robust interpretation.

The measurements were obtained during a single mobile campaign of approximately 4-5 h, during which the platform sequentially intercepted short-lived plumes from individual agricultural, traffic, and industrial sources. Consequently, although the high-frequency measurements provide a large number of data points, the temporal coverage and diversity of source conditions represented in the dataset remain limited.

For MLR, the main limitation is that the measured species cannot be uniquely assigned to individual source categories. The proposed associations (e.g. CO with traffic, N_2O with manured fields, NH_3 with cattle, CH_4 with enteric fermentation, and C_2H_6 with petrochemical sources) are useful indicators for source identification, but several of these compounds are emitted by multiple source types in our dataset. In particular, NH_3, CH_4, and N_2O enhancements were associated with different agricultural source categories. Quantitatively interpreting regression coefficients as source contributions would therefore require source-specific assumptions that cannot be independently constrained by the present measurements.

The ability of PMF to resolve and physically interpret source factors depends on the chemical information contained in the input dataset, including the number and selection of measured species and their associated uncertainties, as well as the temporal coverage of the measurements (e.g. Thera et al., 2019; Frischmon and Hannigan, 2024). In the present study, only a limited set of gaseous species was measured, and the single mobile campaign provided limited temporal coverage of the different source conditions. Therefore, although a large number of high-frequency data points is available, the combination of limited chemical dimensionality and temporal coverage does not provide a sufficiently constrained basis for a robust and physically interpretable PMF analysis. We therefore retain the plume-based source classification used in this study.

References:

Thera, B. T. P., Dominutti, P., Öztürk, F., Salameh, T., Sauvage, S., Afif, C., Çetin, B., Gaimoz, C., Keleş, M., Evan, S., and Borbon, A.: Composition and variability of gaseous organic pollution in the port megacity of Istanbul: source attribution, emission ratios, and inventory evaluation, Atmos. Chem. Phys., 19, 15131–15156, https://doi.org/10.5194/acp-19-15131-2019, 2019.

Frischmon, C. and Hannigan, M.: VOC source apportionment: How monitoring characteristics influence positive matrix factorization (PMF) solutions, Atmos. Environ. X, 21, 100230, https://doi.org/10.1016/j.aeaoa.2023.100230, 2024.

Comment 2.24

Section 3.4 (and Figure 7): It would help to report and compare absolute values of NH3 and CH4. One way to exemplify this could be to add a box plot (or violin plot) of the observed mixing ratios for all species. This could be a multipaneled figure broken down by source like Figure 7.

Reply 2.24

We agree that showing the magnitude of the measured concentration enhancements would provide useful context for the ratio-based analysis in Section 3.4. However, because the analysis is based on background-subtracted plume signals, we consider the background-corrected enhancements (ΔNH_3, ΔCH_4, etc.) more informative than the absolute ambient mixing ratios.

We will therefore add a supplementary figure showing the distributions of the observed plume enhancements for the measured species, separated by source category where sufficient observations are available. The corresponding figure will be referenced in Section 3.4 to provide context for the inter-species relationships shown in Figure 7.

Comment 2.25

Table 1: Are traffic NH3 to CH4 ratios biased by low CH4 emissions from vehicles? How would a similar analysis of NH3 to CO ratios change your conclusions about agricultural vs. non-agricultural sources in this region?

Reply 2.25

The high NH_3:CH_4 ratios observed for the three traffic plumes are indeed partly related to the relatively weak CH_4 emissions from vehicles. We will clarify that the high traffic NH_3:CH_4 ratio should therefore be interpreted primarily as a source signature rather than as indicating unusually large absolute NH_3 emissions.

We also examined the complementary use of CO. Traffic plumes showed strong and coincident NH_3 and CO enhancements, whereas CO enhancements were generally small for the agricultural sources. Consequently, NH_3 is useful for identifying and characterizing traffic emissions but is not a robust ratio for distinguishing among agricultural source types. Conversely, CH_4 provides a much stronger tracer for livestock sources but is weak in traffic plumes.

We will clarify this complementary role of CH_4 and CO in the revised manuscript. Rather than relying on a single ratio across all sources, the simultaneous NH_3, CH_4, and CO measurements provide complementary information for distinguishing agricultural and traffic-related emissions.

Comment 2.26

Section 3.5: See comment above about SCR. Also, did you ever just sit and sample in an exclusively urban area to characterize predominantly traffic-related ratios?

Reply 2.26

Regarding SCR-equipped diesel vehicles, please see our response to Comment 2.19. We will revise Section 3.5 accordingly to avoid attributing the observed traffic NH_3 exclusively to gasoline vehicles.

We did not conduct a dedicated stationary sampling period in an exclusively urban area as part of this campaign. The campaign was designed as a regional mobile survey, with the objective of characterizing as many different emission sources as possible within a single measurement day, with particular emphasis on agricultural sources.

However, the traffic analysis in Section 3.5 is based on approximately 630 individual on-road vehicle plume events identified from coincident short-duration NH_3, CO, and CO_2 enhancements. These events therefore provide a substantially larger traffic-specific dataset than the few longer-duration traffic plumes summarized in Table 1. We will clarify this distinction in Section 3.5.

Comment 2.27

P21, L459: I disagree with this phrase: "an advantage not achievable with closed-path instruments". Prior literature shows that closed-path NH3 TILDAS instruments are capable of achieving adequate response times for this type of sampling and analysis (See Pollack et al., 2019, https://doi.org/10.5194/amt-12-3717-2019; Roscioli et al., 2015, https://doi.org/10.1021/acs.jpca.5b04395; and others coming soon using a mobile lab). A more objective discussion of the advantages and disadvantages of open and closed-path instruments is recommended. The differences in data coverage reported in this analysis on P10, L304 highlights some of those differences.

Reply 2.27

We agree that the statement "an advantage not achievable with closed-path instruments" was too absolute, and we have removed it. As noted in our response to Comment 2.10, appropriately configured closed-path NH_3 TILDAS systems can achieve response times suitable for mobile plume measurements (Roscioli et al., 2015; Pollack et al., 2019). We have therefore revised the manuscript to avoid suggesting that the open-path configuration is generally superior to closed-path NH_3 measurements.

The choice of the open-path HT analyzer in this campaign was primarily based on practical constraints of the mobile platform, particularly available electrical power and weight load for the van, rather than on an inherent advantage in measurement response. We also acknowledge the reduced data coverage of the open-path measurements under unfavorable environmental conditions, as described in Section 3.1. A detailed comparison of open- and closed-path instrument configurations is beyond the scope of the present study, and we have revised the discussion accordingly.

Comment 2.28

P21, L477: Should be extent not extend.

Reply 2.28

Corrected.

Comment 2.29

Section 4.4: You could similarly compare NH3 to carbon ratios with NH3:CO ratios reported for North American cities by Lill et al., 2025, https://doi.org/10.1029/2025JD044019.

Reply 2.29

We thank the reviewer for pointing us to this relevant study. As a check, we calculated an approximate ΔNH_3:ΔCO ratio of ~0.025 for our traffic dataset. This is comparable in magnitude to several of the traffic-related ratios compiled in Fig. 9 of Lill et al. (2025), which includes airborne, on-road, and roadside measurements. However, our traffic analysis is based on carbon-balance emission factors derived from the combined NH_3, CO, and CO_2 enhancements, rather than on ΔNH_3:ΔCO ratios. We therefore prefer not to introduce this additional metric as a new quantitative analysis in the manuscript. Instead, we will cite Lill et al. (2025) in Section 4.4 as complementary context for the variability of traffic-related NH_3 emissions across different vehicle fleets and measurement approaches. Direct quantitative comparison should be treated cautiously because of differences in fleet composition, emission-control technologies, driving conditions, and measurement methodology.

Comment 2.30

Figure S1: The color scales are not particularly useful for distinguishing enhancements. One way to highlight datapoints, especially if one or two high points are skewing the scale, is to reduce the full-scale range and report an off-scale maximum.

Reply 2.30

We thank the reviewer for this suggestion. We have revised Figure S1 to improve the visibility of moderate concentration enhancements. The revised figure uses linear color scales with the upper limits set to the 95th percentile of the displayed enhancements (33 ppb for ΔNH_3, 823 ppb for ΔCO, and 108 ppm for ΔCO_2), preventing a small number of high values from dominating the color range. Measurements exceeding these limits are retained and displayed using the uppermost color, with the color bars indicating the off-scale range ($\geq$). We have also added the CO_2 enhancement panel (as suggested by reviewer 1) and updated the figure caption accordingly.

Comment 2.31

Figure S3: Add a 1:1 line to guide the eye.

Reply 2.31

Agreed. We have added a 1:1 line to both panels of Fig. S3 to facilitate visual comparison between the measurement- and inventory-based emission estimates.

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