EGUSPHERE-2025-2505 | Research article

Ammonia exchange flux over a tropical dry deciduous forest in the dry season in Thailand

Xu et al. (2025)

Response to Referee's Comments

Report #2, Anonymous Referee #1, 16 Oct 2025

I thank the authors for their effort to improve the manuscript. Some of my concerns on the flux calculation, however, remain. The issue is that the measurement set-up is inadequate to calculate reliable fluxes, which is especially concerning during the nighttime. The authors recognize this themselves by stating in Section 3.5 that at least they are sure of the direction of the flux, but should discuss this in a more structured and defined way. Moreover, they should emphasize what this means for the interpretability of their results and also recognize this in the conclusions. As the direction of the flux has more confidence than the size of the flux, the authors could place more emphasis on the gradients rather than the flux. They can e.g. evaluate correlations such as in Figures 5 and 6 for the gradients. Please find below my remaining concerns on the methodology, and some specific comments.

We appreciate the Referee #1's comments and suggestions. We have revised and improved the manuscript as follows:

[Comments on methodology]

Though the used alpha-factor is a good first order estimate for the correction, in reality the alpha-factor in not only height-dependent, but is also affected by canopy structure (Melman et al., 2024). The authors should indicate in Section 3.5 that the value of 0.75 is a first order of estimate, arbitrarily based on the height above the canopy. Secondly, the effect of the RSL on the AGM is different under leafy or leafless conditions (Shapkalijevski et al., 2016) (which may also play a role to explain Figure 8 and the phenomena on lines 323-326). I realize that the measurement set-up is not suited to derive an alpha factor for the two different periods, but the authors should at least discuss this in Section 3.5, and note that this causes a large uncertainty in the comparison of fluxes between the leafy and leafless periods.

We fully agree with this comment and appreciate the suggestion. We have added further explanation about the α factor and corresponding cautions in Section 3.5 (Line 469-477 in the Author's tracked changes version).

The authors indicate in point 1.2 of Referee #1 that they calculate the transfer velocity at 10 minute intervals. However, choosing a short period will lead to a sampling error of large eddies, which may lead to an underestimation of the flux, especially if measured high above the surface where large eddies dominate (e.g. when measuring over forest) (Finkelstein and Sims, 2001). Usually, turbulent statistics are calculated at 30 minute intervals to prevent this error from getting too large, why did the authors choose to calculate D at 10 minute intervals? Moreover, did they apply any additional processing steps, including coordinate rotation, detrending (Gash and Culf, 1996), statistical tests and raw data screening (Vickers and Mahrt, 1997), and flux damping (Moncrieff et al., 2005)? See also Burba (2022).

We agree with the Referee #1 that the failure to capture large eddied typically leads to an underestimation of the flux. In our case, ΔC was measured at 4-h during the daytime, and 10-min transfer velocity was averaged over the same sampling time to calculate the flux. Using 30-minute intervals would result in data loss when even a small number are missing. For example, two missing data within the daytime sampling would reduce data completeness to 75% for 30-minute intervals (n = 6/8), whereas it remains 92% for 10-minute intervals (n = 22/24). Therefore, we adopt 10-min intervals in this study to maintaining sufficient data coverage. Moreover, we also recognize that longer intervals increase random errors of flux due to inadequate sample size and risks of mesoscale variability. Vickers and Mahrt (1997) indicated that the choice of interval involves this trade-off. Because our data is not based on high time-resolution and long-term measurements, and the number of samples is limited, it is important to reduce random errors as well as to maximize data availability. We have added explanations about this in the manuscript (Line 121-122).

In this study, we just conducted low time-resolution fluxes using AGM as a first step. The EC system has not yet been installed at the present study site in Thailand. Therefore, additional processing steps commonly conducted for EC or high time-resolution measurements by software were not applied at this stage. In this study, we used a simple self-made program to record micrometeorological elements at 10 Hz and determined 10-min transfer velocity by calculating friction velocity (u*) and the Obukhov length (L) using the covariances of (i) the east–west and vertical wind components, (ii) the north–south and vertical wind components, and (iii) the vertical wind component with the sonic virtual temperature. Outliers and values with near-zero kinematic heat flux were removed to avoid numerical instability. On the other hand, at our forest research sites in Japan, CO_2 flux measurement using EC is operated, and QA/QC processing is routinely performed using EddyPro software (https://www.licor.com/products/eddy-covariance/eddypro). As a preliminary check, we also confirmed that the u* and L calculated from our simple program has good agreement with those processed by EddyPro, and the difference in transfer velocity on the order of only a few percent. This is likely because averaging over longer periods will minimize the influence of high-frequency noises. Moreover, the QC flag did not show poor quality data particularly during the nighttime, which concerns the Referee #1.

・Vickers and Mahrt (1997) https://doi.org/10.1175/1520-0426(1997)014%3C0512:QCAFSP%3E2.0.CO;2

I'm concerned for the quality of the nighttime fluxes, as in this case the gradients contain very mixed stability classes. The authors should make different groups in their analysis (e.g. in Figure 10) such that their daytime fluxes and nighttime fluxes can be judged individually.

We appreciate this comment. To address this concern, we revised Fig. 10 to separately present daytime and nighttime fluxes so that their behaviors can be evaluated individually (Line 456-462). As we have repeatedly emphasized in the manuscript, the total number of samples is limited in our study. Therefore, focusing solely on nighttime data does not provide sufficiently robust information. As shown in the revised Fig.10 and Table A2, nighttime fluxes exhibit a narrower range than the daytime fluxes, and the variability of the meteorological elements is also smaller. Although some weak tendencies can still be seen, we consider it is difficult to draw conclusions from the nighttime data alone at this stage.

[Specific comments]

1. Line 141: Please also indicate that you will also deal with other sources of uncertainty in Section 3.5.

   We appreciate this suggestion. We have added a brief explanation (Line 139).

2. Figures 5 and 6: I suggest to keep the correlations on the 34m level measurements, and to replace the current column with correlations at the 26m level with correlations between the gradient (dNH3) and the respective meteorological variables. The difference in relations between the 34 and 26 m levels is only limited (as mentioned in the manuscript) and should be due to flux (and thus visible in the gradient), which makes this a good alternative. This could be a good place to place some more emphasis on the found gradients (which have a higher confidence than the flux). Moreover, perhaps relations between dNH3 and temperature are less dependent on wind direction (see also point 6) and could even reveal a relation with RH.

   We appreciate this suggestion. The Referee #1 also recommends us to place more emphasis on ΔC rather than the flux in the general comment. Since only ΔC determines the direction of flux, we agree that qualitatively examining relationship between ΔC and meteorological elements qualitatively helps identify the conditions under which emission and deposition occur. As shown in Fig. R1, the relationship between ΔC and meteorological elements is less clear than that between concentration and temperature and wind speed. What the results suggest is simply that emissions occur under a wide range of meteorological conditions. This lack of a clear correlation is also consistent with the flux. Therefore, we conducted analysis presented in Fig. 10, as already described in the manuscript.

   The Referee #1 recommends us to do this analysis possibly because we state "the direction of the daytime NH_3 flux tended to show upward throughout the observation period. Therefore, the conclusion that the tropical DDF acts as a net source of NH_3 during the dry season remains robust" in Section 3.5. Our intention was to clarify that the measured flux at least represents emission although there are some uncertainties in the magnitude. This state is supported by the results that ΔC was predominantly negative (i.e. upward) and larger than the random errors throughout the observation period. We agree that some uncertainties that could not be evaluated remain in the flux, and it may be large at nighttime. However, as shown in Fig. 9 and discussed in the manuscript, we still believe that the "uncertainty does not alter our overall conclusion that NH_3 emissions predominantly occur during the daytime". As also discussed in the manuscript, the diurnal variation of our measured flux is consistent with previous studies. Generally, during stable conditions (nighttime) the air mixes very slowly and ΔC become relatively large, while during unstable conditions (daytime) the enhanced air mixing leads to lower ΔC (Andersen et al., 1993; Pryor et al., 2001). Even when considering these general characteristics, the fact that ΔC in our observations is substantially larger during the daytime than at nighttime supports the robustness of our interpretation.

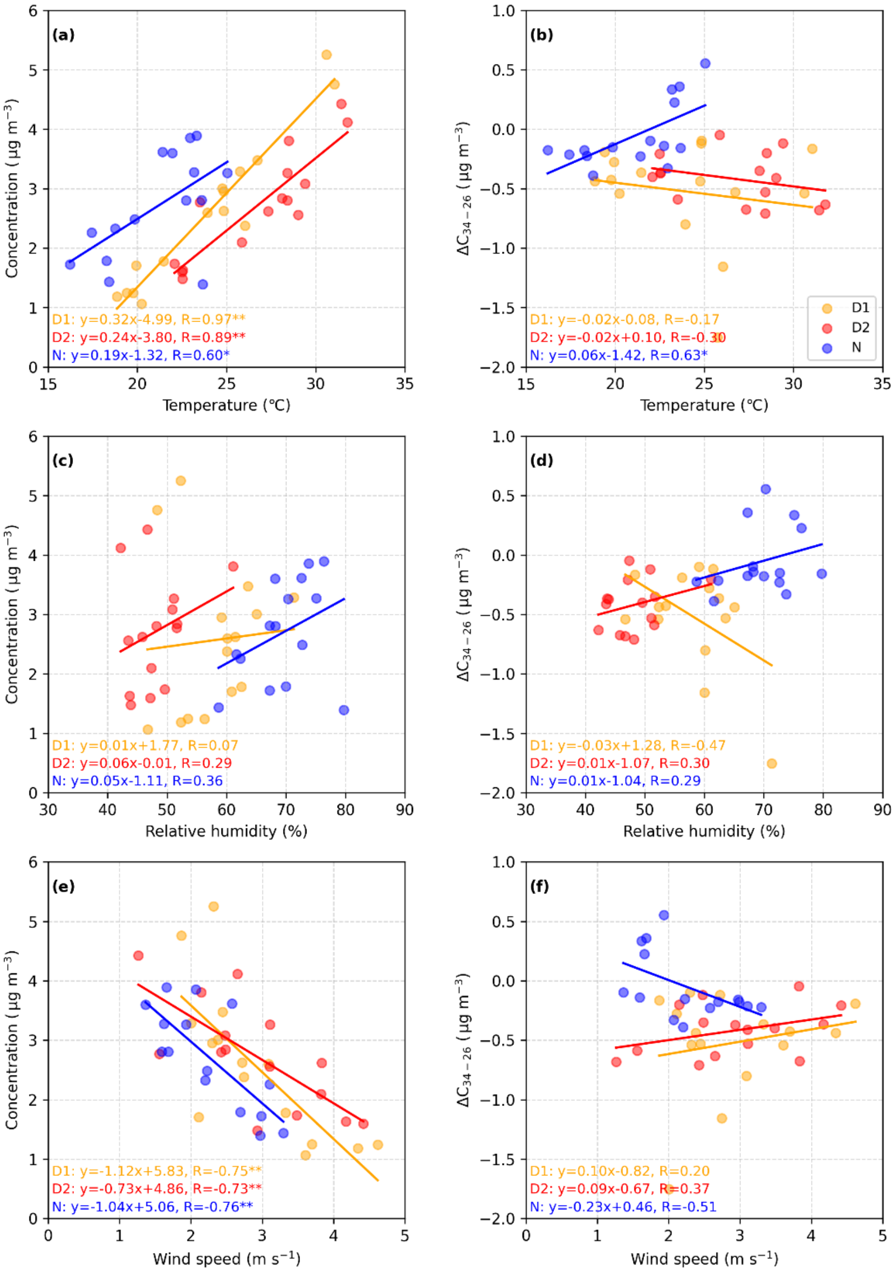

Fig. R1. Relationship between NH_3 concentration at 34 m and ΔC with air temperature, relative humidity, and wind speed during the leafy period. * and ** indicate significance levels of $p < 0.05$ and $p < 0.01$, respectively.

Moreover, <u>our concern is that focusing solely on ΔC would not adequately represent the NH_3 exchange process</u>. Flux has both direction and magnitude and is fundamentally governed by ΔC and the transfer velocity in AGM, both of which can vary. Ignoring the variations in transfer velocity, even though which has a certain uncertainty, surely compromises the essential processes governing the flux (i.e. the bidirectional exchange). As cited in the manuscript, Wolff et al. (2010) demonstrated that most uncertainties of flux originates from ΔC rather than from transfer velocity. We would also like to emphasize that the relationship between the measured fluxes and meteorological elements is generally

consistent with previous studies. In response to this comment, we also thoroughly investigated previous studies using the AGM over forest sites (most of which are summarized in the Table 1 of Xu et al. (2023)) again. These studies examined the relationships between NH_3 fluxes and/or deposition velocity with meteorological elements, even when uncertainties were present or not quantified. We could not find analysis sorely focused on ΔC. Therefore, we have retained our focus on fluxes, while clarifying the uncertainty and the influence on our interpretation in the Conclusion section following the Referee #1's general comment (Line 536-540).

・Andersen et al. (1993) https://doi.org/10.1016/0960-1686(93)90350-8

・Pryor et al. (2001) https://doi.org/10.1016/S1352-2310(01)00259-X

・Xu et al. (2023) https://doi.org/10.1016/j.atmosenv.2023.120144

・Wolff et al. (2010) https://doi.org/10.5194/amt-3-187-2010

3. Lines 267-273: The fact that sign of the correlation changes when incorporating data from certain wind directions aligns with comment 6 of Referee #2; Being that in this case the wind direction, NH3 concentration, and air temperature are correlated. Does that mean that the found relation is only valid under (north)easterly winds? The authors should include a discussion on multicollinearity in the manuscript. Moreover, can the authors speculate on the causality between air temperature and NH3 concentrations?

Although we appreciate this suggestion, based on our current data, it is difficult to conclude that the correlation between concentration and temperature is valid only under specific wind directions. As we have repeatedly explained in the previous revised manuscript and responses, the predominant wind direction in Thailand during the dry season is typically northeasterly. To provide more context, we showed spatial distribution of the 10 m wind speed and direction around Thailand including the leafy and leafless periods in Fig. R2 and Fig R3. As shown in the leafy period, northeasterly winds were dominant (particularly in the northeastern part). On the other hand, the shift in wind direction from northeasterly to southwesterly during the leafless period was an irregular event in the dry season, which made interpretation of the data during this period challenging (Fig. R3(e)). Given the limited number of samples (11–12 per sampling time during the leafless period), even two abnormal data (i.e. 1–2 February) can change the sign of the correlation coefficient. It is also important to note that (i) neither correlation coefficient is statistically significant even though the sign changes, and (ii) data other than northeasterly wind conditions except 1–2 February is also included in the results (as can be seen in Fig. 7).

To thoroughly address the concern of the Referee #1, we showed the relationship between concentration and temperature for northeasterly winds and other wind conditions during the leafy and leafless periods in Fig. R4. Although the number of samples under conditions other than northeasterly winds is limited (a typical feature of the dry season), the correlation holds for the leafy period (Fig. R4(c)). In contrast, there was no clear correlation during the leafless period, regardless of the wind direction. As described in the original manuscript, concentrations during the leafless period are possibly associated with other processes that are possibly driven by structural changes in the forest due to leaf fall. These results are also consistent with those in our studied at a Japanese deciduous forest. Without data obtained from the

wet season when southwesterly winds dominate, it may be an overinterpretation to conclude that the relationship between concentration and temperature is dependent on specific wind direction and to indicate the presence of multicollinearity at this stage.

As already discussed in our previous response to Referee #2, the emission potential of NH_3 is known to be highly sensitive to temperature. Therefore, an increase in temperature during the daytime could enhance emissions from stomata and/or ground and consequently increase concentration. Furthermore, under high temperature conditions, the thermodynamic equilibrium between NH_3, HNO_3, and NH_4NO_3 shifts toward the gas phase, which could also contribute to increase NH_3 concentration. However, it should be noted that our study does not have the materials to provide a firm conclusion regarding this causality.

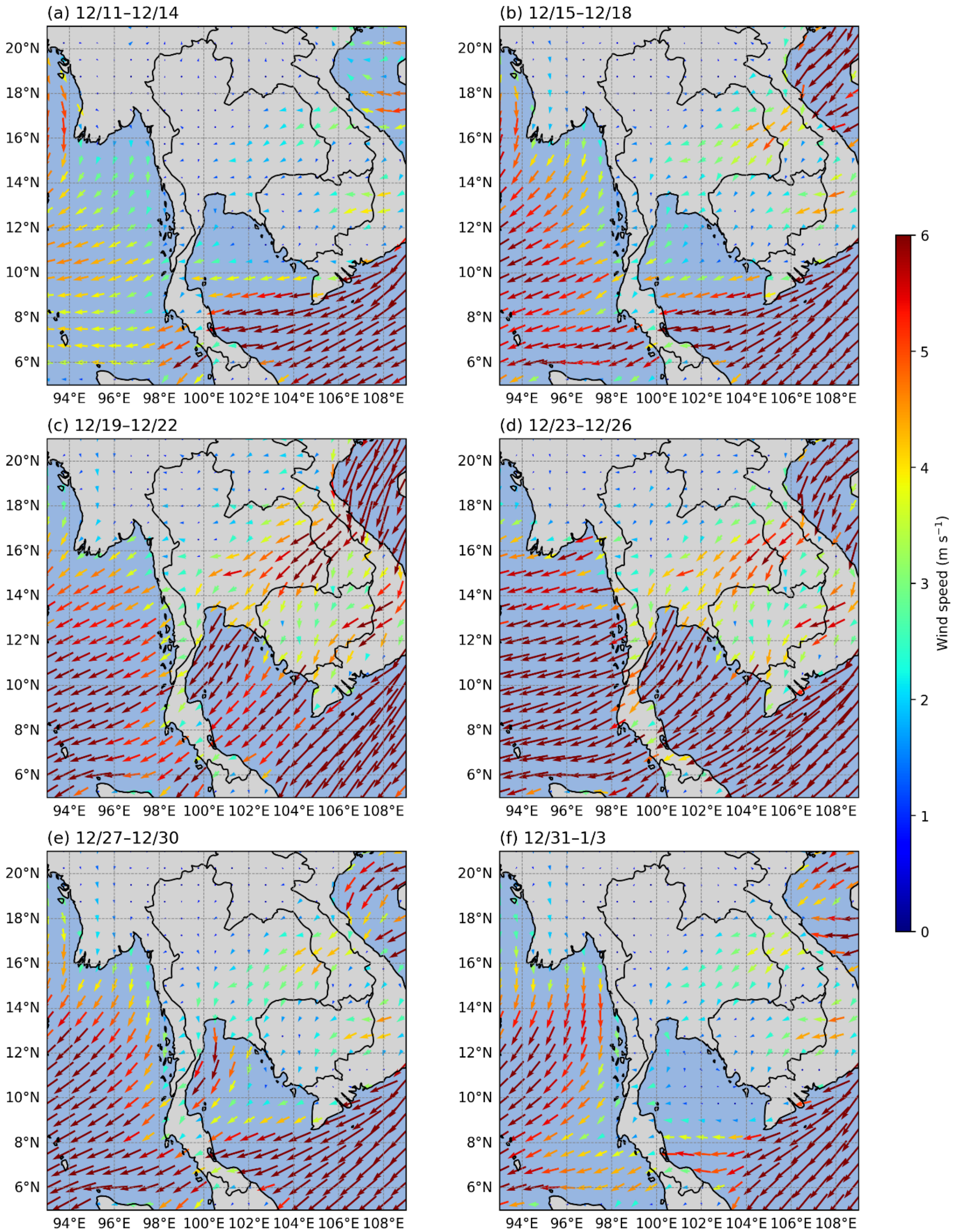

Fig R2. Spatial distribution of the mean wind speed at 10 m around Thailand from 11 December 2023 to 3 January 2024 (leafy period). Eastward and northward components of the 10 m wind over Thailand and neighboring countries were extracted from the ERA5 global reanalysis dataset produced by the Copernicus Climate Change Service at the European Centre for Medium-Range Weather Forecasts. The distributions of mean wind speed and direction are shown for six periods.

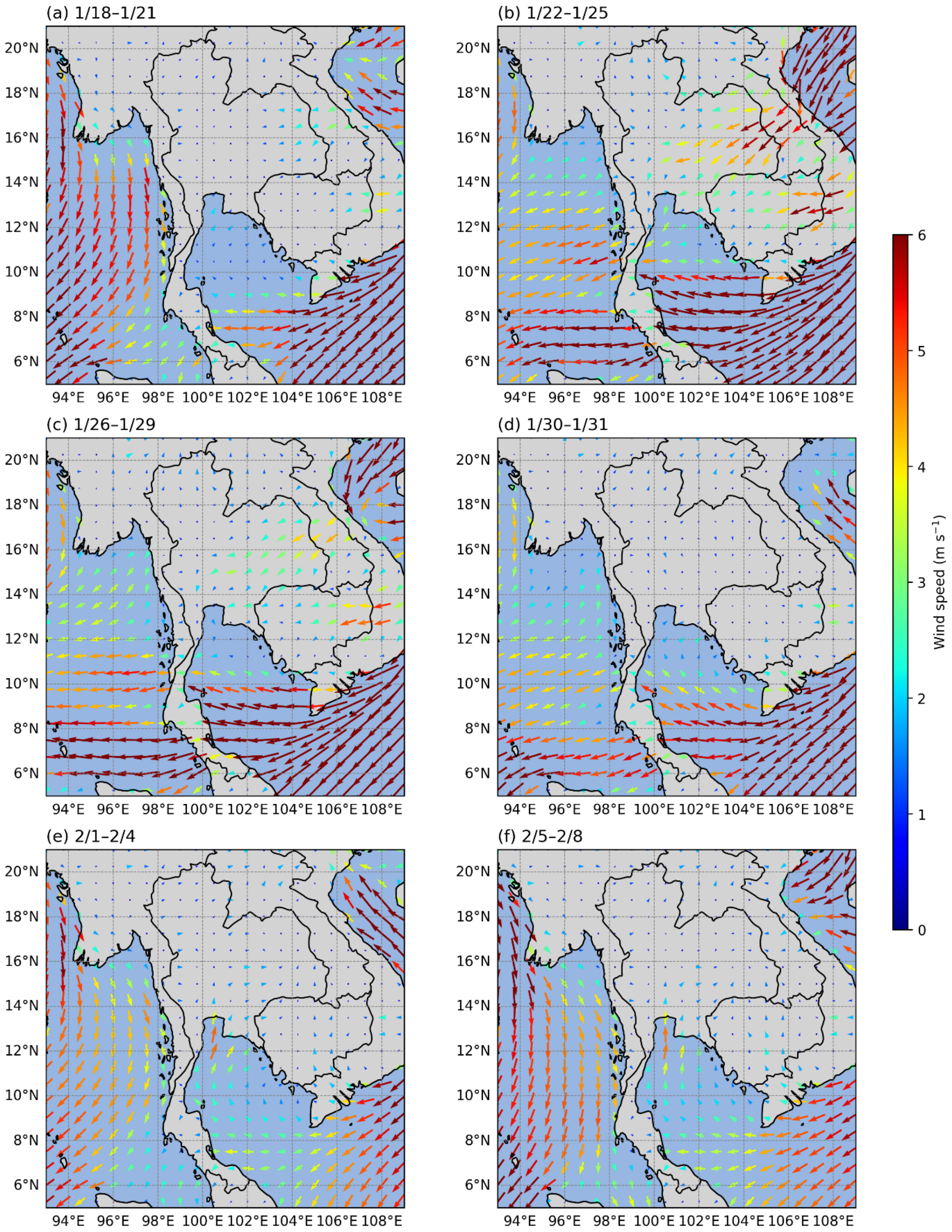

Fig R3. Spatial distribution of the mean wind speed at 10 m around Thailand from 18 January 2024 to 8 February 2024 (leafless period).

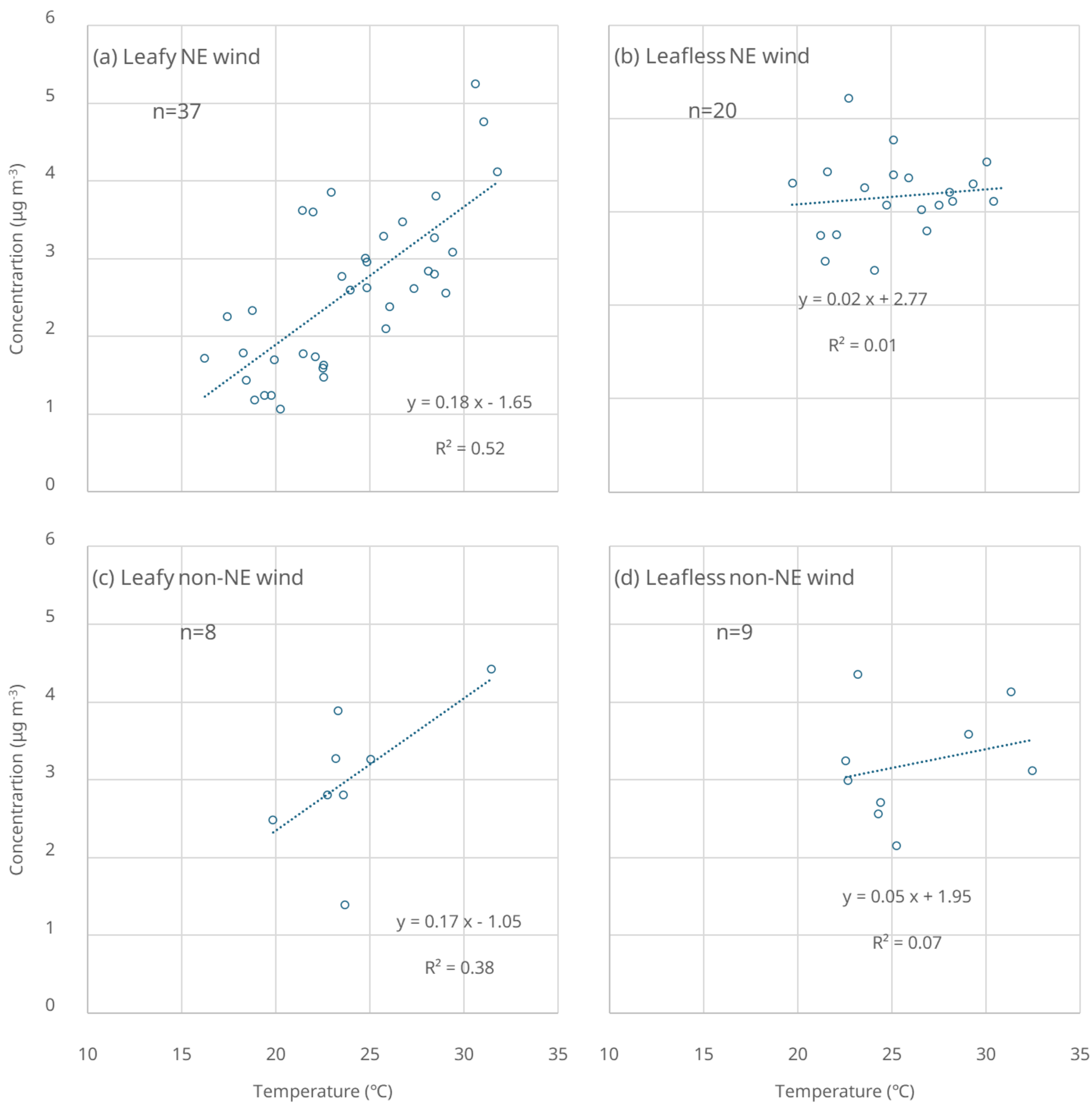

Fig R4. Relationship between NH_3 concentration at 34 m and air temperature for northeasterly winds (NE wind) and other wind conditions (non-NE wind) during the leafy and leafless periods. The data during 1–2 February (n = 6) and the forest fire (n = 8) were excluded.

4. Lines 271-273: In the authors response to comment 10 of Referee #1 they also remove data from February 2nd. Did they also do this in the manuscript?

   In the manuscript, "excluding data from 1 February" mean that the data from 1 February onward were excluded. We have revised the sentence to avoid any misunderstanding (Line 258).

5. Figure 7: Please indicate which measurements where obtained on February 1st and 2nd, such that it is easily visible to support your statements on lines 271-273.

   We think this suggestion is not for Fig. 7 but for Fig. 6 to support our statements regarding the exclusion of data on 1 February and 2 February. We have revised Fig. 6 following the suggestion (Line 275-276).

6. Lines 336-338: Does this mean that the authors found only a significant gradient for in total two or four of the cases?

   We conducted paired t-tests on concentrations measured at 34 m and 26 m for the total observation, leafy, and leafless periods, respectively. For the total observation period, we also performed the tests for all D1, D2, and N samples, respectively. We have revised the sentence to avoid any misunderstanding (Line 317-319).

7. Lines 360-361: In the previous sentence the authors refer to a difference between leafy and leafless, but in this sentence the difference seems to be about daytime or nighttime. Please rewrite for more clarity.

   In the original manuscript, we simply first described the difference between observation periods and then referred to the diurnal variation. We have added a brief explanation (Line 338).

8. Figure 10: The xticks in panels (d) and (h) show values up to 1.5 kW m-2, while in Figure 4(e) and (j) maximum solar radiation is around 0.7 kW m-2. I suspect erroneous xticks. Additionally, I suspect that solar radiation may not be the sole explanatory variable here, but rather the fact that during daytime (high solar radiation) there is a higher transfer coefficient (as in Figure 8). Can the authors reflect on the causality (or multicollinearity) between solar radiation, transfer velocity and NH3 fluxes?

   The x-axis values in Fig. 10 are correct. As clearly stated in the figure caption, Fig. 10 presents integrated 1-hour values during each sampling period, whereas Fig. 4 shows hourly variations. Therefore, the higher values in Figure 10 are natural.

   We showed the relationship between daytime transfer velocity and solar radiation in Fig. R5. The transfer velocity exhibits a strong correlation with solar radiation during both leafy and leafless periods. In addition, as describe in the manuscript, solar radiation is known to facilitate stomatal opening, enhancing emission from plants. Direct sunlight also accelerates decomposition of litter, contributing to emission and volatilization from surface. Therefore, we consider solar radiation to be an explanatory factor controlling NH_3 fluxes.

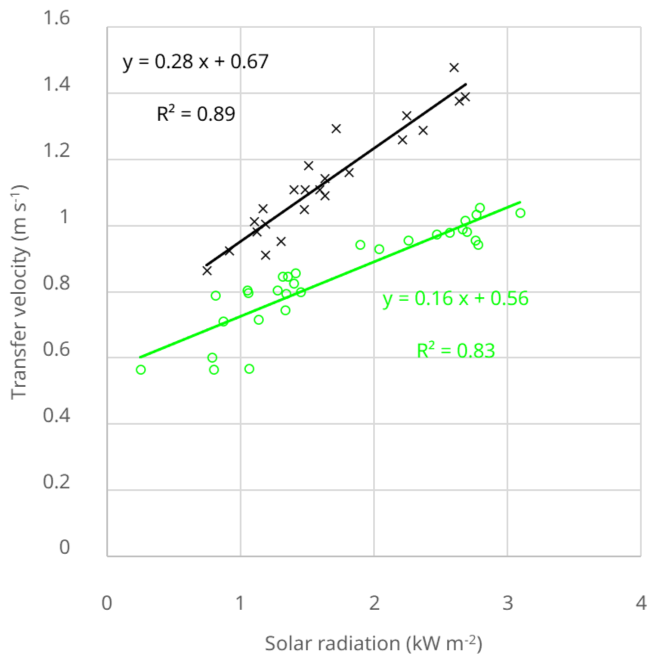

Fig R5. Relationship between daytime (D1 and D2) transfer velocity and integrated solar radiation during the leafy (circles) and leafless (crosses) periods.

9. Section 3.5: This section should start a general statement that the calculated fluxes come with major uncertainties. I propose to start this section with: "Due to several limitations of the measurement set-up, the calculated fluxes contain major uncertainties", followed by a systematic listing of the limitations and the induced errors, being: "(i) Instrumental set-up did not allow us to validate the roughness sublayer effect on the AGM. Therefore, we tentatively used the alpha factor of (…). This introduces an error of (…). (ii) The long measurement of ΔNH_3 interval introduces two uncertainties: (ii-a) ignorance of the cross term and (ii-b) averaging over multiple stability classes. To estimate the error due to ignoring the cross-term (ii-a), we (…). This introduces an error of (…). During daytime, the effect of (ii-b) is limited because (…). However, during nighttime (…). This introduces an error of (…). Finally, (iii) the use of AGM leads to larger errors compared to REA or EC (…). This introduces an error of (…)" The authors should end this part with an estimate of the total error for the random error and for the systematic error (as a quadratic sum to all separate errors), such that the it is easier to evaluate the reported results. The authors could even do that separately for each measurement interval (i.e. D1, D2 and N), as I suspect that during nighttime errors are much larger than during daytime. After that, they can continue with the rest of Section 3.5.

We appreciate this detailed suggestion. Following the comment, we have revised Section 3.5 to provide a better explanation of the limitations and uncertainties associated with our measured flux (Line 466-489). However, we should note that it is difficult to follow the proposed revision exactly, given the nature of our results and the overall manuscript structure. In addition, point (i) is only an evaluation using a tentative value. For point (ii), the magnitude of the uncertainty cannot be quantified from our data. We could only

address point (iii) following the established formulations in previous studies. Therefore, we consider it is not appropriate to combine points (i) and (iii) into a total error given these differences in reliability.
We also evaluated point (iii) separately for each measurement interval (D1, D2, and N). As a result, the random error did not show a large bias between daytime and nighttime samples (Line 502-503).

[References]

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