A point-by-point response to Referee #2

We sincerely appreciate Referee #2 for the valuable and constructive comments, which are very helpful for the improvement of the manuscript. We have revised the manuscript carefully according to the referee's comments. The following is a point-by-point response to address the referee's comments. The original comments are presented in *black* and our responses are in *blue*, respectively. The new or modified contents in the revised manuscript are marked in *red*.

Comments from Referee #2:

Meng et al. conducted flux measurements of HONO, NO and NO₂ over paddy fields. The measured period covered several agricultural activities. They found relatively higher HONO and NO emissions during the rotary tillage period but lower emissions during irrigation/fertilization periods. NO₂ flux was generally negative as soil represents a NO₂ sink. Through correlation analysis, they found the co-existence of soil biogenic emissions and NO₂ conversion which dominate the daytime HONO budget. HONO contribution to OH was also estimated.

This study was focused on an interesting topic and the dataset benefits our understanding of soil HONO emissions. Some descriptions are still not clear, which needs to be improved before publication. **Response:** Many thanks for your careful review and constructive suggestions, which are quite valuable to us and greatly improve the quality of the manuscript.

Major comments:

The conclusion that fertilizer doesn't significantly affect HONO emissions is not convincing, mainly due to the short period of measurements. As reported in several studies listed below, peak HONO emissions after fertilization were observed in a wide range of 3-15 days after fertilization and could still sustain at a high level within 3 weeks after fertilization. Therefore, I would suggest the authors to be careful when drawing this conclusion.

https://acp.copernicus.org/articles/23/15733/2023/ https://pubs.acs.org/doi/10.1021/acs.est.4c01070 https://linkinghub.elsevier.com/retrieve/pii/S0048969720343965 https://www.sciencedirect.com/science/article/pii/S1352231018306599?via%3Dihub

Response: Thanks for your great comments. We agree with the referee that fertilizers affect HONO emissions and modified the conclusions presented in the manuscript accordingly. Compared to the continuous peaks observed during rotary tillage, no significant HONO emissions were measured after fertilization. Unlike the studies by Tang et al. (2019) and Xue et al. (2019), which focused on maize and wheat fields, the paddy fields were consistently inundated before and after fertilization. The prolonged waterlogging may affect HONO emissions. Additionally, our study employed the aerodynamic gradient method, focusing on a large area of farmland with no disturbance to the underlying surface, while the dynamic chamber method used by Tang et al. (2019) and Xue et al. (2019) has limitations when applied to large areas due to its focus on specific points. Different measurement methods may lead to discrepancies in results. Furthermore, the continuous peaks of HONO flux observed following topdressing (Fig. 3 in the manuscript) suggest that fertilization can promote emissions, which aligns with the aforementioned studies. However, the exploration of the fertilizer effect in this observation is brief, and more detailed research is needed to understand the impact of fertilization in paddy fields.

Abstract: We have deleted the following sentence as suggested.

"During paddy cultivation, the flooded environment with a higher water-filled pore space (~80 %) significantly suppressed the HONO emission, and the fertilization did not have a significant promoting impact on HONO fluxes."

Conclusion: We made revisions and it now reads as follows.

"Under this inhibitory effect, no significant peaks in HONO flux were observed after fertilization compared with that during the rotary tillage."

The description of the AG method is not clear enough. There is no information on how the 3D winds were measured and validated; the impact of sampling pressure change due to NO_x switching was not discussed.

Response: Thanks for your great comments. The introduction of the AG method has been revised to enhance clarity as suggested by the referee.

Lines 178-192 in the tracked changes manuscript:

"The HONO, NO and NO₂ fluxes were calculated by the AG method at time intervals of 30 min, which has been elaborated upon in previous studies (Laufs et al., 2017; Meng et al., 2022; Stella et al., 2012) and will be briefly introduced here. The flux (F_{χ}) of trace gas is calculated from the friction velocity (u_*) and the mixing ratio scaling parameter (χ_*) as follows:

$$F_{\chi} = -u_* \chi_* \tag{1}$$

where u_* is calculated from eddy covariance measurements and χ_* is defined from stability-corrected gradient of the scalar mixing ratio (χ) with height (z) as:

$$\chi_* = \kappa \cdot \frac{\partial \chi}{\partial \left[\ln(z-d) - \Psi_{\rm H} \left(\frac{z-d}{L} \right) \right]}$$
(2)

The fluxes ($F_{\text{HONO,NO and NO}_2}$) of trace gases at geometric mean height can be expressed as:

$$F_{\text{HONO,NO and NO}_2} = -\kappa \cdot u_* \cdot \frac{\partial c(\text{HONO,NO and NO}_2)}{\partial \left[\ln(z-d) - \Psi_{\text{H}}\left(\frac{z-d}{L}\right)\right]}$$
(3)

where κ is von Kármán constant ($\kappa = 0.4$), *z* is the height above the ground, *d* is zero plane displacement and was taken as $2/3 \cdot h_c$ (h_c is the canopy height), *L* is the Obukhov length and Ψ_H is integrated stability correction function for scalars (Sutton et al., 1993)."

Eddy covariance measurements, including 3-D wind velocity (u, v, w), were performed with an integrated 3-D ultrasonic anemometer and open-path CO₂/H₂O analyzer (IRGASON, Campbell Sci. Inc., USA), which was detailed in Text S1 in the supplementary material. The IRGASON is routinely maintained and calibrated to guarantee data quality. Quality control was implemented following the CarboEurope methodology (Aubinet et al., 2000), which encompasses the removal of spikes, application of planar fit to correct for the sonic anemometer tilt and frequency response correction, etc.

The pressure changes caused by the NO_x analyzer switch were insignificant, with a change of merely 5×10^{-4} kPa (<0.001%) before and after the switch, having no impact on the BBCEAS measurement (see response question L164-165 for more details).

Correlations between HONO flux and NO_2*JNO_2 were used to support light-induced NO_2 conversion. As NO_2 flux was also measured here, it would be great to see the correlation between HONO flux and NO_2 flux* JNO_2 .

Response: Thanks for your great comments. As the referee suggested, we have added the correlations between HONO flux and NO₂ flux, as well as the product of $J(NO_2) \times NO_2$ flux (Fig. S4), in the supplementary material. The HONO flux is weakly correlated with NO₂ flux only (R = 0.25), however, it exhibited a moderate correlation with the product of $J(NO_2) \times NO_2$ flux (R = 0.41). The correlation between HONO flux and NO₂ flux, as well as the product of $J(NO_2) \times NO_2$ flux, is not only influenced

by the chemical processes but also by the physical. Nevertheless, the increasing correlation between HONO flux and the product of $J(NO_2) \times NO_2$ flux indicates the source of light-induced NO₂ conversion during daytime.

Lines 346-349 in the tracked changes manuscript:

"Although the correlations of HONO flux with NO₂, $J(NO_2)$ and NO₂ flux were found to be low (R = 0.28, 0.12 and 0.25), we observed a significant correlation (R = 0.60) between HONO flux and the product of $J(NO_2) \times NO_2$ (Fig. 6), as well as a moderate correlation (R = 0.41) with the product of $J(NO_2) \times NO_2$ flux (Fig. S4)."

Supplementary material:



Figure S4. Correlations of the daytime HONO flux with (a) NO₂ flux and (b) the product of $J(NO_2) \times NO_2$ flux during rotary tillage.

Specific comments:

L30-31, emission processes refer to mechanism?

Response: Yes, the emission processes here refer to the mechanisms, and we have modified the expression.

L33, be careful with using "the first time" as there are already many flux measurements. I don't think it is interesting to indicate the first measurement at a specific location. Otherwise, there will be too many first-time...

Response: Thank you for your suggestion. The phrase "for the first time" has been deleted in the revised manuscript. As the referee suggested, we have also revised other parts of the manuscript and removed pertinent expressions.

Lines 32-33 in the tracked changes manuscript:

"Herein, we performed measurements of HONO and NO_x fluxes over paddy fields in the Huaihe River Basin."

L39, "HONO and NO fluxes, and they exhibited" ...

Response: Revision has been made as the referee suggested.

Lines 40-41 in the tracked changes manuscript:

"Continuous peaks in HONO and NO fluxes were observed during the rotary tillage, and they exhibited a significant correlation (R = 0.77)."

L41-42, maybe it's better to name "soil release mechanisms from biological processes" as "soil biological emissions"?

Response: Thank you for your suggestion. The sentence has been modified as suggested.

Lines 42-45 in the tracked changes manuscript:

"The results indicate that both soil biological emissions and light-driven NO₂ conversion are likely active, collectively influencing the diurnal pattern of HONO flux."

L46, what's "flux rates". The unit of nmol m⁻² s⁻¹ was used in previous sentences.

Response: The "flux rates" refer to the atmospheric HONO production rate with a unit of ppb h⁻¹, which is derived from the release of HONO from soil to the atmosphere (Meng et al., 2022; Su et al., 2011; Tang et al., 2020; Xue et al., 2019). The calculation of the HONO production rate based on HONO flux

 (P_{soil}) has been detailed in the supplementary material, and we have revised the expression here to enhance comprehension for the reader.

Lines 46-50 in the tracked changes manuscript:

"Sensitivity tests demonstrated that photo-enhanced NO₂ conversion on the ground could adequately explain P_{unknown} , while the nocturnal HONO production derived from soil emission fluxes (ranging from 0.32 ppbv h⁻¹ to 0.79 ppbv h⁻¹) was sufficient to elucidate the nighttime P_{unknown} ."

Abstract: in general, I think the abstract needs to be improved to be more concise.

Response: Thank you for your suggestion. We have revised the abstract to make it more concise as suggested.

Lines 29-52 in the tracked changes manuscript:

"Abstract: Significant amounts of nitrous acid (HONO) released from soil affect the chemistry of the troposphere, as a major precursor of hydroxyl radical. However, the scarcity of in-situ data on soilatmosphere HONO exchange flux has constrained the comprehension of emission mechanisms and reactive nitrogen budget. Herein, we performed measurements of HONO and NO_x fluxes over paddy fields in the Huaihe River Basin. The entire experiment experienced various agricultural management activities, including rotary tillage, flood irrigation, fertilization, paddy cultivation and growth, and topdressing. HONO and NO exhibited upward fluxes, whereas NO₂ deposited to the ground, with average hourly fluxes of 0.07 ± 0.22 , 0.19 ± 0.53 and -0.42 ± 0.44 nmol m⁻² s⁻¹, respectively. Continuous peaks in HONO and NO fluxes were observed during the rotary tillage, and they exhibited a significant correlation (R = 0.77). Moreover, a significant correlation (R = 0.60) between HONO flux and the product of $J(NO_2) \times NO_2$ was also observed during the daytime. The results indicate that both soil biological emissions and light-driven NO₂ conversion are likely active, collectively influencing the diurnal pattern of HONO flux. Source analysis revealed that the unknown HONO source $(P_{unknown})$ exhibited a diurnal pattern with higher daytime and lower nighttime values. Sensitivity tests demonstrated that photoenhanced NO₂ conversion on the ground could adequately explain P_{unknown}, while the nocturnal HONO production derived from soil emission fluxes (ranging from 0.32 ppbv h⁻¹ to 0.79 ppbv h⁻¹) was sufficient to elucidate the nighttime $P_{unknown}$. Our study emphasized the variability of HONO fluxes across various agricultural management activities, as well as the importance of heterogeneous NO₂ conversion on the ground surface and soil emissions for HONO production."

L60-74, emissions from the combustion process should also be included here.

Response: Thank you for your suggestion. We have supplemented the combustion process and relevant literature in the revised manuscript.

Lines 65-76 in the tracked changes manuscript:

"Several potential mechanisms have been proposed to explain atmospheric HONO levels, including direct emissions from combustion processes (Nakashima and Kajii, 2017; Nie et al., 2015); the chemical equilibrium between soil nitrite (NO₂⁻) and hydrogen ions (Su et al., 2011); photosensitized reactions of NO₂ on organic substances (George et al., 2005), humic acids (Han et al., 2016; Stemmler et al., 2006), soot (Monge et al., 2010), minerals (Ndour et al., 2008), urban grime (Liu et al., 2019a), plant leaves (Marion et al., 2021), etc.; photolysis of adsorbed nitrates or nitric acid (Ye et al., 2017; Zhou et al., 2003; Zhou et al., 2011) and ortho-nitrophenols (Bejan et al., 2006; Guo and Li, 2022); direct emission from ammonia oxidizing bacteria and other microorganisms (Oswald et al., 2013; Scharko et al., 2015); desorption of adsorbed HONO from the surface by acid displacement processes (Vandenboer et al., 2013; Vandenboer et al., 2014; Vandenboer et al., 2015), and chemical reactions of hydroxylamine on the surface of soil particles (Ermel et al., 2018)."

L164-165, When NO_x analyzer is switching between the upper channel and the lower channel, gas flows as well as the gas pressure in both channels change. What's the corresponding impact on BBCEAS measurements?

Response: Thanks for your great comments. The sample flow rate of BBCEAS instruments is controlled by a mass flow controller, and the switching solenoid valve does not affect the flow rate within the cavity. The changes in cavity pressure before and after switching the solenoid valve, which only changes by 5×10^{-4} kPa (<0.001%), have no impact on BBCEAS measurement.



Figure 1. The changes in cavity pressure with just the BBCEAS instrument versus with both the BBCEAS instrument and NO_x analyzer.

L170: Normally the sampling tube needs to be heated to above ambient temperature. 30 °C seems not to satisfy this demand through the measurement period, see Figure 2.

Response: Thanks for your great comments. The ambient temperature ranged from 17.0 to 36.6 °C throughout the campaign, with average values of 26.8 ± 3.5 °C. The heating tape with a heating temperature of 30 °C was encased in thermal insulation cotton. This configuration will make the temperature within the sampling tube beyond 30 °C, thereby preventing the occurrence of condensation within the tube. Throughout the measurement periods, no condensation was observed when the temperature of the heating tape was maintained at 30°C, thereby fulfilling the requirements for measurement.

L237, pls also indicate the period for each activity in Table 1.

Response: Thank you for your suggestion. We have supplemented the period for each activity in the revised manuscript.

Lines 222-224 in the tracked changes manuscript:

"The field campaign was performed across various agricultural management activities, including rotary tillage (June 2–13), flooding irrigation (June 13–19), fertilization (June 19–21), paddy cultivation (June 26–27), and top-dressing (July 10)."

L242, the minor ticks for the x-axis in Figure 3 look weird.





Figure 3. Time series of O₃, NO, NO₂, HONO and the fluxes of HONO, NO, and NO₂ were determined by the aerodynamic gradient method. The mixing ratios of HONO, NO, NO₂ (lower level: 0.2/0.3 m, upper level: 1.6 m), and O₃ were measured above a crop rotation field and averaged for 30 min intervals. Periods of agricultural management activities (rotary tillage, flood irrigation, fertilization, after

fertilization, paddy cultivation and growth, top-dressing) are denoted at the top of the graph.

L250, what is the level of 80% WFPS in water holding capacity? Was there still liquid water over the paddy fields after irrigation and fertilization? See the lab work below where high HONO emissions were observed at high WHC.

https://pubs.acs.org/doi/10.1021/acs.est.4c01070

https://pubs.acs.org/doi/10.1021/acs.est.2c07793

https://onlinelibrary.wiley.com/doi/10.1029/2021JD036379

Response: Thanks for your great comments and the reference literature. In some laboratory studies, high HONO emissions were found under high moisture conditions. During the paddy season, there was liquid water on the paddy fields after irrigation and fertilization. For 80% WFPS, the corresponding WHC is about 72%.

In our laboratory study, we referred to the research by Oswald et al. (2013) and Huang et al. (2024) to calculate the relationship between paddy soil HONO emission flux and WHC and WFPS. Figure 2 shows the trend of WHC and WFPS over time and their corresponding relationship (WHC = $0.90 \times$ WFPS). Approximately 80% WFPS corresponds to a WHC of about 72%. This relationship corresponds closely to the results demonstrated by Behrendt et al. (2019). (WHC = $0.80 \times$ WFPS).



Figure 2. Time series and correlation of water holding capacity (WHC) and water-filled pore space (WFPS).

L278, pls indicate that 0.19 nmol m⁻² s⁻¹ is the measured value in this study.

Response: As the referee mentioned, the value is our actual measurement. In order to make it clearer for readers, we have modified the sentence.

Lines 290-291 in the tracked changes manuscript:

"The agricultural field acted as a well-known source of atmospheric NO, with an average flux of 0.19 \pm

 $0.53 \; nmol \; m^{\text{-}2} \; s^{\text{-}1}$ in this study."

L283, Another point to be discussed. When soil is waterlogged, HONO can't be evaporated from the soil due to its high water solubility.

Response: Thank you for your suggestion. We have supplemented the discussion about HONO in the revised manuscript.

Lines 298-299 in the tracked changes manuscript:

"Similarly, we also did not observe significant emissions of HONO under sustained high moisture conditions."

L295, Table 2, there are quite a lot of flux measurements in below study.

https://pubs.acs.org/doi/10.1021/acs.est.4c01070

Response: Thank you for your suggestion. We have supplemented the recent research of Xue et al. (2024)

in Table 2 in the revised manuscript.

 Table 2 Summary of the maximum and minimum of HONO flux in field measurements over different soil types at remote/rural/suburban sites.

Soil type	Method	HONO flux ^a		HONO flux ^b		Reference
		(nmol m ⁻² s ⁻¹)		(nmol m ⁻² s ⁻¹)		
		Min	Max	Min	Max	-
Grassland	AG	-0.09	0.53	_	_	Harrison and Kitto (1994)
		-0.21	0.70			
Forest	AG	0.02	0.07	—	—	Sörgel et al. (2015)
Maize	AG	—	—	0.01	0.16	Laufs et al. (2017)
Wheat	AG	-0.39	1.10	-0.003	0.20	Meng et al. (2022)
Agricultural	REA	-0.30	0.50	-0.007	0.10	Ren et al. (2011)
Forest	REA	-0.50	1 31	0.03	0.19	Zhou et al. (2011)
Forest	REA	0.03	0.19	-	_	Zhang et al. (2012)

Grassland	REA	-0.06	0.16	0.02	0.07	Von Der Heyden et al.
						(2022)
		0.04	0.23	—	_	
Maize	OTDC	0.41	2.89	—	_	Xue et al. (2019)
		_	108.21	—	_	
Maize	OTDC	_	2.84	-0.06	1.45	Tang et al. (2019)
Wheat	OTDC	-0.09	0.55	_	_	Tang et al. (2020)
Maize	OTDC	-0.61	22.79	0.01	10.86	Song et al. (2023)
		_	0.33	_	_	
Maize	OTDC	_	11.50	—	_	Xue et al. (2024)
		_	24.86	—	_	
Paddy	AG	-0.70	1.86	0.01	0.15	This study

L311: I don't think the negative NO₂ flux was due to NO₂ photolysis.

Response: Thanks for your great comment. As noted by the referee, the downward NO_2 flux could be attributed to its dry deposition. A greater downward NO_2 flux during daytime was observed in comparison to nocturnal NO_2 fluxes, which could be ascribed to an increased deposition velocity. The sentence has been modified in the revised manuscript.

Lines 323-325 in the tracked changes manuscript:

"A greater downward NO₂ flux of -0.85 ± 0.27 nmol m⁻² s⁻¹ (-0.57 ± 0.23 nmol m⁻² s⁻¹ at night) was observed during the daytime, potentially due to an increase in the dry deposition velocity of NO₂ during the day."

L332, maybe add the correlation between HONO flux and NO₂ flux*JNO₂.

Response: Thank you for your suggestion. We have added the correlations between HONO flux and NO₂ flux, as well as the product of $J(NO_2) \times NO_2$ flux (Fig. S4), in the supplementary material. Additionally, we also modified the corresponding sentences in the revised manuscript.

Lines 346-349 in the tracked changes manuscript:

"Although the correlations of HONO flux with NO₂, $J(NO_2)$ and NO₂ flux were found to be low (R = 0.28, 0.12 and 0.25), we observed a significant correlation (R = 0.60) between HONO flux and the product of $J(NO_2) \times NO_2$ (Fig. 6), as well as a moderate correlation (R = 0.41) with the product of $J(NO_2) \times NO_2$ (Fig. 6), as well as a moderate correlation (R = 0.41) with the product of $J(NO_2) \times NO_2$ (Fig. 6), as well as a moderate correlation (R = 0.41) with the product of $J(NO_2) \times NO_2$ (Fig. 6), as well as a moderate correlation (R = 0.41) with the product of $J(NO_2) \times NO_2$ flux (Fig. S4)."

L395-398, those results largely rely on MLH estimations. Better to show how MLH was estimated.

Response: Thank you for your suggestion. The detailed calculation for MLH can be found in Text S3, and its diurnal variation is shown in Fig. S6 in the supplementary material. To help readers better understand the calculation results, we have added guidance before the HONO budget.

Lines 372-374 in the tracked changes manuscript:

"The calculation of HONO sources and sinks, as well as the estimates of the mixing layer height (MLH) is described in detail in Text S3 in the Supplemental Material."

L438, first, it was not a long-term measurement; second, there were several more studies measuring flux at high water content;

Response: Thank you for your suggestion. We have modified the expressions of "first" and "long-term observation" as suggested by the referee.

Lines 460-462 in the tracked changes manuscript:

"Considering limited field observation of HONO flux under high soil water content, future studies should pay more attention to paddy fields to validate the mechanisms observed in the laboratory."

Supplementary

Aerosol surface area, Sa, was used to calculate P_{aerosol} . Sa was stated to be calculated based on aerosol size distribution. However, there is no mention of how the aerosol size was measured.

Response: We apologize for the unclear expression in the manuscript and have revised the sentence in Text S1 in the supplementary material.

"The particle matter concentration and aerosol size distribution of 0.25–32 μm were measured using a particulate analyzer (EDM180, Grimm, Germany)."

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