Point-to-point responses to anonymous Referee #2

Thank you for your comments concerning our manuscript. Below are point-to-point responses to the comments. Changes are made in the resubmitted manuscript accordingly and are marked in blue.

Comment on egusphere-2026-2234, Anonymous Referee #2

The manuscript by Hua et al. addresses an important flux research gap through the self-designed Relaxed Eddy Accumulation (REA) system: the air-surface exchange of multiple inorganic acidic species over cropland chronically influenced by industrial emissions. The authors comprehensively evaluated the precision and detection limit of the REA system. They analyzed the bidirectional flux characteristics of target species, and further estimated the gross upward emission fluxes of HNO_3 and HONO. The topic is original and practically valuable, and key findings are meaningful for regional air-soil-water pollution mitigation. The manuscript aligns with the scope of Atmospheric Chemistry and Physics. I recommend publication after the authors address the following questions.

(1) This manuscript employs the REA method to calculate fluxes and presents a relatively comprehensive uncertainty analysis. The β coefficient serves as a key parameter in flux estimation, but the current error estimation does not fully account for the β variability. I encourage the authors to include a complete the calculation process and uncertainty analysis for the β coefficient.

Re:

Thank you for your suggestion. Now we provide an uncertainty analysis of β and incorporate it into the overall uncertainty estimate of flux. In line 191-195, we add:

"*The β coefficients for 33 4-hour sampling periods fell within the typical range of 0.47 to 0.62. While a single β coefficient was used for each 4-hour sampling period, β exhibited intra-period variability. We therefore calculated β at 30-minute intervals, and used σ_β/β within each 4-hour period as the relative uncertainty of the corresponding period-averaged β. The relative uncertainties of β for the 33 sampling periods ranged from 2.80% to 26.14% (Table S1)*"

Table S1 and Table 2 are updated accordingly.

Table 2. Concentration and flux detection limits, mass analysis precision and flux measurement precision of the acidic species.

	Concentration detection limit (μg m^{-3})	Mass analysis precision (RSD, %)	Flux measurement precision (RSD, %)	Flux detection limit (μg m^{-2} s^{-1})
HNO_3	0.027	4.4	8.5–37.7	1.5×10^{-2}–2.1×10^{-1}
HONO	0.034	4.6	8.8–28.0	1.5×10^{-2}–1.2×10^{-1}
SO_2	0.121	5.8	9.1–30.3	9.5×10^{-3}–1.3×10^{-1}
HCl	0.047	4.7	11.6–32.3	8.2×10^{-3}–1.1×10^{-1}
NO_3^-	0.027	2.7	7.3–31.6	2.2×10^{-2}–2.4×10^{-1}
NO_2^-	0.034	3.0	5.4–26.3	6.1×10^{-4}–2.8×10^{-2}
SO_4^{2-}	0.121	4.6	6.7–28.6	1.2×10^{-2}–1.3×10^{-1}
Cl^-	0.047	3.2	7.1–21.6	6.6×10^{-3}–1.0×10^{-1}

Table S1. Relative uncertainties of β coefficient and sampled M induced by lag time inaccuracy in different samples.

Sample run No.	Relative deviation of β (%.±)	Relative deviation of M (%.±)					
		0.1s		0.3s		1s	
		Up	Down	Up	Down	Up	Down
1	3.13	/	/	/	/	/	/
2	4.91	1.69	-1.68	4.19	-4.26	7.14	-8.22
3	5.91	-3.94	3.95	-10.01	10.07	-18.26	19.41
4	6.81	-1.64	1.65	-4.20	4.23	-7.55	8.15
5	6.63	3.29	-3.28	8.15	-8.32	13.02	-16.04
6	6.55	2.82	-2.81	7.01	-7.14	11.48	-13.77
7	26.14	-0.75	0.76	-1.95	1.96	-3.47	3.77
8	5.26	0.79	-0.78	1.93	-1.96	3.34	-3.78
9	5.10	-1.09	1.10	-2.80	2.81	-5.00	5.42
10	8.16	-1.09	1.10	-2.80	2.81	-5.00	5.42
11	3.91	-0.65	0.66	-1.69	1.70	-3.01	3.27
12	6.83	0.07	-0.06	0.12	-0.13	0.20	-0.25
13	8.55	0.85	-0.84	2.07	-2.10	3.59	-4.06
14	4.90	0.85	-0.84	2.07	-2.10	3.59	-4.06
15	2.80	1.59	-1.58	3.95	-4.01	6.75	-7.74
16	3.26	-0.76	0.77	-1.98	1.99	-3.52	3.83
17	8.65	1.13	-1.12	2.79	-2.83	4.82	-5.47
18	6.43	2.44	-2.43	6.07	-6.17	10.10	-11.91
19	4.51	2.50	-2.49	6.20	-6.30	10.29	-12.16
20	9.47	1.07	-1.06	2.63	-2.67	4.54	-5.15
21	13.40	0.52	-0.51	1.26	-1.28	2.19	-2.47
22	9.32	-1.61	1.62	-4.10	4.13	-7.37	7.96
23	5.90	0.96	-0.95	2.37	-2.41	4.10	-4.65
24	7.70	-2.01	2.02	-5.13	5.16	-9.23	9.94
25	3.11	-0.73	0.74	-1.90	1.91	-3.38	3.67
26	2.92	0.48	-0.47	1.16	-1.18	2.02	-2.28
27	9.33	-0.20	0.21	-0.55	0.55	-0.97	1.04
28	8.55	-9.80	9.81	-24.88	24.98	-46.30	48.15
29	6.21	-0.85	0.86	-2.19	2.20	-3.90	4.24
30	12.97	1.27	-1.26	3.14	-3.18	5.39	-6.14
31	7.44	-0.01	0.02	-0.09	0.08	-0.17	0.16
32	5.67	1.09	-1.08	2.69	-2.73	4.64	-5.27
33	10.52	-6.89	6.90	-17.50	17.58	-32.31	33.89

(2) Why was the threshold for distinguishing updrafts and downdrafts set to $\pm 0.6\sigma_w$ instead of $\pm 0.5\sigma_w$ or $\pm 0.7\sigma_w$?

Re:

According to the conditional sampling theory of REA, a higher triggering threshold of wind speed enhances the concentration difference between the updraft and downdraft reservoirs ($C_{up}-C_{down}$), thereby improving the precision of its measurement. However, if the threshold is set too high, an excessive fraction of low-vertical-velocity samples will be excluded, reducing the representativeness of the conditional samples relative to actual turbulent exchange. This may increase the uncertainty in fluxes derived from the REA method.

We have adapted the recommendations of Businger and Oncley (1990) and used a threshold of $\overline{w}\pm0.6\sigma_w$. The threshold of $\overline{w}\pm0.6\sigma_w$ has been widely adopted in relaxed eddy accumulation (REA) studies (Bai et al. 2015, Sarkar et al. 2020) and has been demonstrated in previous experiments to provide reliable flux estimates (Bowling et al. 1998, Desjardins 1977, Ge et al. 2022, Velentini et al. 1997).

In lines 116−120 of the revised manuscript, we revised:

"*Three-minute running mean ($\overline{w}$) and standard deviation (σ_w) of w was calculated in real time to trigger solenoid valve (Numatics LS02M6F00B, Emerson Electric Co., St. Louis, Missouri, USA) switching: updraft sampling at $w>\overline{w}+0.6\sigma_w$, downdraft sampling at $w<\overline{w}-0.6\sigma_w$, and dead-band airflow within the $w\pm0.6\sigma_w$ range. Wind speed triggering threshold selection involves a trade-off between the precision of $C_{up}-C_{down}$ measurements and sample representativeness. As an empirical threshold commonly used in REA studies, $0.6\sigma_w$ has been demonstrated to provide reliable flux estimates (Bowling et al. 1998, Businger and Oncley 1990, Bai et al. 2015, Sarkar et al. 2020).*"

References:

Bai, J., Guenther, A., Turnipseed, A. and Duhl, T. (2015) Seasonal and interannual variations in whole-ecosystem isoprene and monoterpene emissions from a temperate mixed forest in Northern China. Atmospheric Pollution Research 6(4), 696-707.

Bowling, D.R., Turnipseed, A.A., Delany, A.C., Baldocchi, D.D., Greenberg, J.P. and Monson, R.K. (1998) The use of relaxed eddy accumulation to measure biosphere-atmosphere exchange of isoprene and other biological trace gases. Oecologia 116(3), 306-315.

Businger, J.A. and Oncley, S.P. (1990) Flux Measurement with Conditional Sampling. Journal of Atmospheric and Oceanic Technology 7(2), 349-352.

Desjardins, R.L. (1977) Description and evaluation of a sensible heat flux detector. Boundary-Layer Meteorology 11(2), 147-154.

Ge, H., Wei, Z., Zhang, H. and Kang, L. (2022) Experimental Research of Methane Flux Measurement by the Relaxed Eddy Accumulation Method. Acta Scientiarum Naturalium Universitatis Pekinensis 58(3), 434-442.

Sarkar, C., Turnipseed, A., Shertz, S., Karl, T., Potosnak, M., Bai, J., Serça, D., Bonal, D., Burban, B., Lopes, P.R.C., Vega, O. and Guenther, A.B. (2020) A portable, low-cost relaxed eddy accumulation (REA) system for quantifying ecosystem-level fluxes of volatile organics. Atmospheric Environment 242, 117764.

Velentini, R., Greco, S., Seufert, G., Bertin, N., Ciccioli, P., Cecinato, A., Brancaleoni, E. and Frattoni, M. (1997) Fluxes of biogenic VOC from Mediterranean vegetation by trap enrichment relaxed eddy accumulation. Atmospheric Environment 31, 229-238.

(3) This manuscript should incorporate more detailed background information on the sampling site and farmland conditions. For example, the manuscript should provide the exact latitude and longitude of the flux tower site, as well as the duration over which the cropland has been continuously affected by chemical industrial emissions. In addition, field conditions during the sampling campaign should be described in greater detail, including the crop growth stage and records of fertilization and irrigation practices. The absence of these critical details limits the comparability with previous agricultural flux studies.

Re:

We revise the paragraph in 142-155 to include more descriptions:

"*The Wuhan Chemical Industrial Park (WCIP) is located northeast to the urban area of Wuhan. The WCIP covers 71.64 km^2 and hosts petrochemical, fine chemical, and building material industries. Since its full operation in 2013, the WCIP has been identified as a major industrial emission source influencing atmospheric composition in the surrounding region. The flux tower (114.53° E, 30.65° N) was installed at the center of a vegetable cropland within the 7.3 km^2 Beihu Farm, which is immediately adjacent to the WCIP. The nearest petrochemical emission source lies approximately 1.0 km north of the tower (Figure S1). The cropland was cultivated with Raphanus sativus L. var. longipinnatus (white radish), a typical cold-season vegetable in this region. The tower is situated on flat terrain with no obstructing buildings or trees within a 500 m radius of the tower, ensuring undisturbed airflow measurements.*

Field sampling was performed from 29 October to 30 December 2025, with 11 valid sampling days in cloudy or sunny rain-free weather. Each sampling day included three 4-hour sampling periods (morning: 8:00–12:00; afternoon: 12:30–16:30; early night: 17:00–21:00). The sampling was conducted during the vegetative growth and harvest periods of R. sativus, and no fertilization activities were conducted throughout the sampling period."

(4) Figure captions should be checked for consistency. For example, the main text states that horizontal lines in Figure 2 mark means, whereas the supplementary figures use medians.

Re:

We corrected the supplementary figures by using mean values. Figure captions were double-checked throughout the manuscript to remove any inconsistency.