Responses to referee 1:

We would like to thank the referee for the useful comments and constructive suggestions. In the following, we address the referee's comments and describe corresponding changes we have made to the manuscript. The referee's comments are listed in *italics*, followed by our response in blue. New/modified text in the manuscript is in **bold**.

The authors have addressed most of my previous comments, by adding a few appendixes to clarify the features, assumptions and caveats of the method. One critical thing I would like to point out is that, the wind divergence term, which the authors argued to be unphysical, is indeed physical and necessary in order to ensure mass conservation. The physical meaning is that when there is wind divergence, the same volume of air parcel expands, and thus the density of NO2 decreases. This term, together with the density gradient term, makes sure the mass conservation of an air parcel when there are no temporal change of density (as assumed in the study), loss and emissions. The negative emissions are NOT a result of including the wind divergence term, but from other simplifications of the flux divergence method, including but not limited to the use of temporally (a few hours) averaged wind data. Therefore, although there maybe value of excluding the wind divergence term, it is certainly not because it is unphysical. Because exclusion of the wind divergence term is a foundational assumption of this study, the justifications and caveats must be clarified in the main text.

We again appreciate the referee's efforts to scrutinize the directional derivative methodology established in Sun (2022), which contains detailed derivations, justifications, and evaluations in its main text and supporting information and has already undergone a rigorous peer review process of the journal *Geophysical Research Letters*. Although the goal of this current ACP manuscript is to apply the established directional derivative approach to unveil post-COVID emissions, instead of reinventing the methodology, we recognize that the readers who are interested in the details do not necessarily have to go back reading Sun (2022). As such, we add more clarification as the second paragraph of Section 3.2 of the manuscript, which is included at the end of this response. Before that, we would like to address the referee's comments specifically.

The referee asserted that "*the wind divergence term, which the authors argued to be unphysical*". It is unclear how the referee got this impression. We copy the relevant part in our previous response:

"The consequence of including the wind divergence term (highlighted by red in the equation above) is clearly demonstrated by Fig. 1a in Sun (2022), where large negative emissions are found over coastal land, positive emissions are found over coastal ocean, and the Gulf Stream appears as a strong sink due to its wind convergence. All of those are highly unrealistic and apparently not real sources/sinks."

To paraphrase that, it is the inclusion of the wind divergence term that leads to unrealistic emissions (how can the Gulf Stream be a huge sink of trace gases larger than all known flux values?). We did not say that the wind divergence itself is unphysical. In fact, we did not even use the word "unphysical".

We generally agree with the referee's following physical interpretation of the wind divergence term. To clarify, although the wind divergence term does not appear in the directional derivative equation (Eq. 1) in the manuscript, it does not simply vanish or get ignored. Instead, it is cancelled out by the vertical flux at z_1. Both Sun (2022) and the added Appendix A in the manuscript emphasized that the air flow is incompressible (it is compressible when flow becomes as fast as 0.3 Mach number)(Smits, 2000). As a result, if we take a column-shaped control volume from surface to z_1, the density of air in the control volume does not change, and the volume of the control volume, by construction, does not change. The horizontal wind divergence draws air down at z_1, and the horizontal wind convergence pushes air up at z_1. The following was stated in Appendix A in the previous revision, and we move these sentences to the main text:

"Conceptually, the upward flux of the observed species at z_1 would not be due to emissions, as z_1 is chosen not to "feel" the emission impact; the only cause of this flux is the convergence of air in the column below that squeezes air upwards or the divergence of air below that draws air downwards."

The equations that incorporate this and lead to the directional derivative equation are Eqs. 9–13 in Sun (2022). We summarize these in the newly added text to this manuscript at the end of this response.

The referee further alleged that *"The negative emissions are NOT a result of including the wind divergence term, but from other simplifications of the flux divergence method, including but not limited to the use of temporally (a few hours) averaged wind data"*, which we respectfully disagree. See the added Eq. A1:

$$\begin{aligned}\langle E\rangle &= \big\langle \nabla\cdot(\Omega\vec{u})\big\rangle + \frac{\langle\Omega\rangle}{\tau}\\ &= \big\langle \vec{u}\cdot(\nabla\Omega)\big\rangle + \big\langle \Omega(\nabla\cdot\vec{u})\big\rangle + \frac{\langle\Omega\rangle}{\tau}.\end{aligned}$$

Ignoring the chemical term $\langle\Omega\rangle/\tau$, the emission calculated that way is just the sum of the wind divergence and directional derivative term. If the wind divergence is a large negative number, the calculated emission will be a similar large negative number. Additionally, we are unclear how the referee had the impression of the *"use of temporally (a few hours) averaged wind data"*, which we did not. Both Sun (2022) (Section 2) and this manuscript (Section 2.1) clearly state that the horizontal winds are spatiotemporally interpolated/sampled at TROPOMI level 2 observations from ERA5, where ERA5 winds are instantaneous at hourly resolution. We have thoroughly discussed the simplifications of both the flux divergence and the directional derivative approach in Appendix A, and none of these simplifications could explain the large, oscillatory positive/negative "emissions" calculated from the flux divergence, except the wind divergence term.

Finally, we summarize the clarifications and justifications above as the second paragraph of Section 3.2 of the revised manuscript:

"The most important difference between the flux divergence and directional derivative approach is, at flat surface and without chemical loss, whether the emissions equal the divergence of horizontal flux ($\langle \nabla \cdot (\Omega \vec{u}) \rangle$) or a directional derivative of the column amount ($\langle \vec{u} \cdot (\nabla \Omega) \rangle$). The mathematical and physical justifications of using the directional derivative instead of the flux divergence to estimate emission are provided in detail by Sun (2022), and we further list the key assumptions made by the flux divergence and directional derivative approaches in Appendix A. In brief, we assume an altitude z_1 that divides the lower troposphere where emissions are mixed within and the upper troposphere where emissions are not "felt" at the satellite pixel scale, and horizontal variability is much smaller than the lower part. Together with the incompressible flow assumption (Smits, 2000), these enable us to cancel out the wind divergence term from surface to z_1 ($\Omega_b(\nabla \cdot \vec{u})$, where Ω_b is the subcolumn from surface to z_1) with the vertical flux at z_1. Conceptually, the upward flux of the observed species at z_1 would not be due to emissions, as z_1 is chosen not to "feel" the emission impact; the only cause of this flux is the convergence of air in the column below that squeezes air upwards or the divergence of air below that draws air downwards. Ultimately, this leads to the only appearance of the directional derivative term in Eq. 1, instead of the flux divergence term that can be decomposed to the sum of the directional derivative term and a wind divergence term (see Eq. A1). Moreover, this study includes in general more advanced considerations of atmospheric physical and chemical processes in comparison with previous studies, which we summarize in Appendix B."

References

Smits, A. J.: A physical introduction to fluid mechanics, John Wiley & Sons Incorporated, 2000.

Sun, K.: Derivation of Emissions from Satellite-Observed Column Amounts and Its Application to TROPOMI NO2 and CO Observations, Geophysical Research Letters, 49, e2022GL101 102, https://doi.org/https://doi.org/10.1029/2022GL101102, 2022.