Review of , The Turbulent Enhancement Ratio as a novel Approach for Characterizing Local Emission Sources in Complex Environments' by Lamprecht et al.,

Summary statement:

The paper presents a novel method of examining sources of pollutants (in this case NOx) in an urban environment using a 'Turbulent Enhancement Ratio (TER)'. The quantity is calculated from fast response analysers, thus resolving the turbulent motion of the pollutant. The TER is compared to a flux ratio (FR) of the pollutants (NOx / CO2), calculated using the widely used eddy covariance technique for each species. Once it has been shown that the agreement between the TER and FR is good, the authors use TER to examine sources of NOx in the Innsbruck area, in particular using the difference between COVID19 lockdown and 'normal' times.

This is an interesting paper describing a novel technique that could lead to important insights into air pollutant emissions in urban environments. I recommend publication subject to the following revisions and additions.

REPLY:
We thank the reviewer for his valuable assessment. Below we address all comments grouped by their relevance. We fixed some errors that were pointed out.

General comments

COMMENT 1 + 2:
I think the manuscript needs more discussion as to why the TER is preferable to standard Eddy covariance fluxes? It seems that they essentially give the same thing (especially when looking at ratios), and both require high time resolution measurements. So what are the main benefits of TER.

On page 8 line 10 it is stated that TER is the preferred methodology (compared to the more widely used normalised emission ratio (NER)). It is not clear to me why this is the case, and this section would benefit from an expansion to better explain this.

REPLY:
We revise this statement to clarify that TER should be seen as an extension to NER by taking into account the turbulent part of the correlation between two tracers. As noted by reviewer one, mathematically the approach should be more seen as a spatial filtering approach to NER. In this context we interpret TER as a spectral similarity ratio, similar to approaches used to filter data in turbulent flows (Antonia et al., 1987, Nappo, 2012) This is particularly relevant in urban areas and other complex environments, where the background can not be easily separated from plume enhancements. This leads to biases and has been discussed in the literature (e.g. Yokelson, 2013). To overcome these limitations, we leverage the advantages of turbulence to distinguish between local and non-local sources, allowing for a more accurate representation of enhancement ratios in dynamic and complex environments. We have also expanded the theory and discussion sections to provide greater clarity and detail.

CHANGES:
As suggested by Reviewer 1, we have expanded the discussion to provide a more detailed context on spectral analysis. Accordingly, we have revised Fig. 2 and introduced Fig. 3, which presents the co-spectra and ogives of CO_2 and NO_x. In this context, we also emphasize

that the importance of experimentally resolving TER becomes particularly pronounced when the measurement site is close to an emission source, as turbulent mixing plays a dominant role in determining enhancement ratios. This effect is especially critical during morning and evening hours, when the planetary boundary layer (PBL) undergoes strong transitions, leading to shifts in vertical mixing conditions that directly influence the observed ratios.

COMMENT 3:
Page 13 line 8. This paragraph would benefit from expansion. It says that higher CO_2 fluxes are seen in the colder seasons the result of increased domestic heating in these seasons. This is almost certainly true but why do the NOx fluxes not also increase in the colder season? Presumably there is also a NOx emission from heating – is this not observed at all? Please comment on this.

REPLY:
It is true that NO_x emissions also originate from heating systems. However, within our footprint, most heating systems are gas-powered, emitting approximately 1,000 times more CO_2 than NO_x per TJ of energy (based on the emission factor inventory of Austria). Since more than 90% of the NO_x emissions at the field site area are traffic-related - remaining relatively stable between summer and winter - the impact of temperature or the "heating period" on NO_x fluxes is minimal. Instead, other factors, such as slight shifts in the footprint, play a more significant role in influencing NO_x flux variations.

CHANGES:
In contrast, the NO_x flux demonstrates a different pattern. It exhibits only a very moderate positive temperature dependence ($dNO_x/dT = +0.14$ nmol/K), which remains relatively constant across the entire temperature range examined. This suggests that NO_x emissions, primarily influenced by traffic (Lamprecht et al. 2021), do not fluctuate significantly with temperature changes. A key factor in this weak temperature dependence is the composition of heating systems within the turbulent footprint. Most heating systems in this area are gas-powered, which produce negligible NO_x emissions compared to the traffic sector, thus the impact on NO_x fluxes are largely decoupled from temperature-driven heating demand.

The interplay of these two trends is reflected in the Turbulent Enhancement Ratio, which decreases at lower temperatures. This decline can be attributed to the increasing dominance of CO_2 sources, particularly from heating systems, as temperatures drop. Consequently, the relative contribution of NO_x to the total emissions diminishes, altering the NO_x/CO_2 ratio.

COMMENT 4:
In section 4.3.2, the authors could comment on whether their data shows a change of dominant source during the lockdown period (i.e. does the NOx / CO2 ratio suggest a change from traffic dominated to domestic heating dominated emissions), as has been seen in other studies (e.g. Cliff et al., ACP, 2023).

REPLY:
we agree that the NO_x/CO_2 ratio shifts more towards other sources (e.g. residential) exhibiting a lower ratio during the lockdown, has been observed for other locations (e.g. London). As pointed out by the reviewer these findings are similar to the study by Cliff et al. from central London, which we now consider in our discussion.
Significant shifts from road- to non-road sources have also been observed in London during pandemic restrictions (Cliff et al. 2023); (p13)

COMMENT 5:
The authors state that they have a data series from mid 2018 to early 2022 but do not comment on any longer term trend. It would be nice to see if the NOx / CO_2 ratio has changed from the start to the end of their dataset (when COVID restrictions had been lifted) and what any change could be attributed to

REPLY:
In Innsbruck the vehicle fleet still includes a significant proportion of diesel-powered cars, and unlike cities such as London, there are no low-emission zones where high-emitting vehicles exceeding predefined thresholds are restricted. While the transition to lower-emission vehicles with improved Euro standards is currently underway, longer and more extensive measurement campaigns at our field site will be necessary to accurately quantify the impact of this transformation.

Specific comments

COMMENT 6:
2.2 Instrumentation section: It is not clear which of the two NO_2 measurements is used for the calculations (of TER, NER and fluxes). If it is the Moly converter NO_2, have the authors investigated any potential issues with the residence time of air in the converter and the validity of 5Hz measurements?

REPLY + CHANGES:
We added a hint that the CLD899Y was used to perform the calculations for Flux and TER.

We confirm that the converter is a molybdenum converter, and its performance was assessed during an instrumentation characterization after the instrument was delivered. Our tests determined that the residence time of air in the converter is less than 1s. We have previously tested the response time for measuring NOx EC fluxes using the CLD899 and found a damping timescale of 0.8 s (e.g. Karl et al., 2017. doi: s41598-017-02699-9). This represents a high frequency loss of about 13%. From our analysis, here we find that in an urban area a 1s response time is still sufficient to capture the relevant turbulent co-variant parts.

COMMENT 7:
Table 1: what do 'dd' and 'ff' represent in the measured parameters from the sonic anemometer? It is probably obvious but should be made clearer.

REPLY:
Thank you. More specified by adding a footnote below the table. The abbreviation is commonly used in the field of meteorology.

COMMENT 8:
P 13 line 24: It says ‘the TER remains higher’ but higher than what. Please better explain this sentence.

REPLY:
Replaced the sentence for greater clarity. "Remain higher" referred to the 55% traffic reduction, whereas the TER did not decrease by the same proportion:
Compared to the approximately 55% reduction in weekday traffic during the lockdown, the decrease in TER is less pronounced (p13)

COMMENT 9:
Section 2.3: I think it would benefit from a slightly more detailed description of how the eddy covariance fluxes are calculated, including the key parameters and filtering methodology used. I realise it is in the literature but just a few sentences would greatly benefit this paper.

REPLY:
good point, the key parameters for filtering are given now in the section "2.3 Dataset":
We used the following quality assurance/quality control (QA/QC) criteria: a signal-to-noise ratio > 3, a steady-state criterion ≤ 0.5, a noise RMSE ≤ 20 for NOx and CO2 fluxes and a correlation coefficient ≥ 0.5 for TER while TER and FR were calculated for 30 min averages.