Referee 1

Clara M. Nussbaumer et al. presented the NOx fluxes from airborne measurements in Los Angeles during the aircraft campaign RECAP-CA. They showed both NOx concentrations and fluxes were higher in the weekdays and lower in the weekend. They also showed the difference between their calculated NOx fluxed and NOx emissions from the CARB inventory. The observations are valuable and very useful to the emission community. The paper is generally well rewritten. However, I still have some minor concerns before it can be published.

We would like to thank Referee 1 for taking the time to review our manuscript and the valuable feedback. We have corrected our manuscript according to the referee's comments and think it is now improved.

line 94: please add what is NOy, what species are included in the NOy?

NO_y describes the sum of all reactive nitrogen species including NO_x and higher nitrogen oxides: HNO_3, HONO, peroxy nitrates (RO_2NO_2), alkly nitrates ($RONO_2$), etc. We have clarified this in the manuscript.

Lines 98 ff.: Reactive nitrogen species ($NO_y \equiv NO_x$, HNO_3, HONO, $RONO_2$, RO_2NO_2, ...) were detected through thermal dissociation at ~500°C to NO_2 in the third channel (Day et al., 2002).

line 160-162: it was mentioned that the values of the NOx flux are dominated by the atmospheric variability. Can you explain a little more about it?

The uncertainty of the nitrogen oxides measurements used for calculating the NOx fluxes over Los Angeles is not dominated by the measurement uncertainty (typically around 7% for the used instrument), but rather by the atmospheric variability (around 30% for this study) induced by factors like varying meteorological conditions and the time-of-day. Additional uncertainties arise from the method applied (wavelet transformation) for determining the NOx emissions, for which a detailed error analysis is provided in Zhu et al. (2023). We agree that this was worded somewhat confusingly in the text and have rephrased it for clarification.

Lines 173 ff.: The overall uncertainty of the calculated NO_x flux is composed of the uncertainty of the measurement of the NO_x concentration and the vertical wind speed. We find that the NO_x median and average values are dominated by the atmospheric variability and not the measurement uncertainties. The observed atmospheric variability of NO_x is in the order of 30% (1σ) which is around 4 times higher than the instrumental precision of <7% (1σ). Additional uncertainty is associated with the presented method of performing the wavelet transformation, including random and systematic errors (Lenschow et al., 1994; Mann and Lenschow, 1994; Wolfe et al., 2015; Vaughan et al., 2021). A detailed error analysis for these observations is provided in Zhu et al. (2023).

line 174 you are using the boundary layer height, where did you get the boundary layer height? Is it measured or modeled boundary layer height? What is the uncertainty of the boundary layer height?

The boundary layer height was determined from changes in water vapor, the dew point and toluene concentrations, which are usually high in the boundary layer and decrease promptly in the free troposphere. We have clarified this in the text.

Line 85 ff.: The planetary boundary layer (PBL) height was determined from changes in water vapor and toluene concentrations, the dew point and temperature, which decrease rapidly at the boundary between the BL and the free troposphere (Pfannerstill et al., 2023). The aircraft crossed the top of the PBL at several times during each flight providing these direct observations.

Line 177 'the fit' is the the linear fit of Fz and z/zi. Please mention it here.

We have added this information in the text.

Line 193 ff.: In order to investigate the influence of vertical divergence, we compare an analysis with a correction of the fluxes using the linear fit of the NO_x flux (F_z) and the dimensionless altitude (z/z_i) as shown in Figure S4 to our analysis assuming the divergence is zero, which we will refer to as 'sensitivity study' in the following.

Line 187. Please make it more clear what is the sensitivity study. Can you also provide a figure of the vertical divergence versus the dimensionless which excluding data points within the upper 20 % of the boundary layer? I get very confused by looking at Figure s4 and Figure s5. It would be nice if you also use different colors to indicate the density of the points.

With the sensitivity study, we attempt to investigate the impact of a range of choices for vertical flux divergence (including zero) on the interpretation of our measurements. With the available data, unfortunately we cannot perform an unambiguous correction of the flux divergence, whose existence is indicated by the Figure S4 (plotting the flux versus the dimensionless altitude). However, we apply a correction factor derived from the linear fit of F_z versus z/z_i to show the potential effect of the vertical divergence compared to analyses which assume the divergence to be zero. We have clarified what we mean with 'sensitivity study' in the text.

Lines 193 ff.: In order to investigate the influence of vertical divergence, we compare an analysis with a correction of the fluxes using the linear fit of the NO_x flux (F_z) and the dimensionless altitude (z/z_i) as shown in Figure S4 to our analysis assuming the divergence is zero, which we will refer to as 'sensitivity study' in the following.

We have added Figure of F_z vs z/z_i, excluding data point in the upper 20% of the boundary layer to Figure S7 of the Supplement. The resulting linear fit shows a more vertical course (to be expected after vertical divergence correction) compared to Figure S6 which included data points throughout the entire boundary layer.

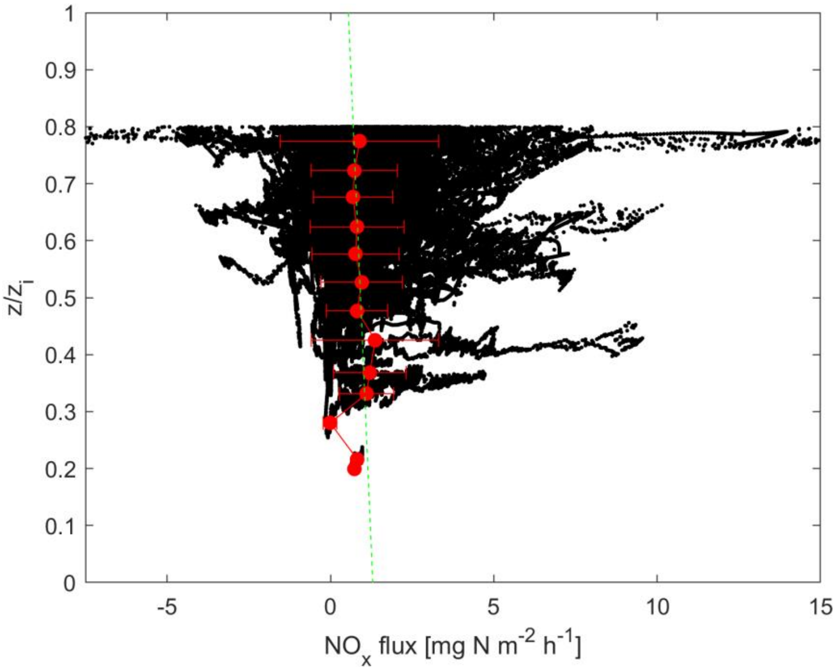


Figure S7. Dimensionless altitude z/z_i versus the corrected NO_x flux according to Figure S4, omitting data in the upper 20% of the boundary layer, which are most strongly affected by uncertainties in the vertical divergence correction. Black dots represent all data points. The green dashed line shows the linear fit of all data points. The red points and error bars represent the binned means with the 1σ variability.

Figure S4 presents all available data points of the calculated NO_x flux versus the dimensionless altitude. We take the resulting linear fit to correct the NOx fluxes for vertical divergence and the result is presented in Figure S6 (S5 before). Due to large uncertainties of the correction in the upper 20% of the BL we omit these data in the following sensitivity study. We now show the corrected fluxes for the lower 80% of the BL in Figure S7 of the Supplement. We have also created a density plot to indicate the data distribution which we show in Figure S5.

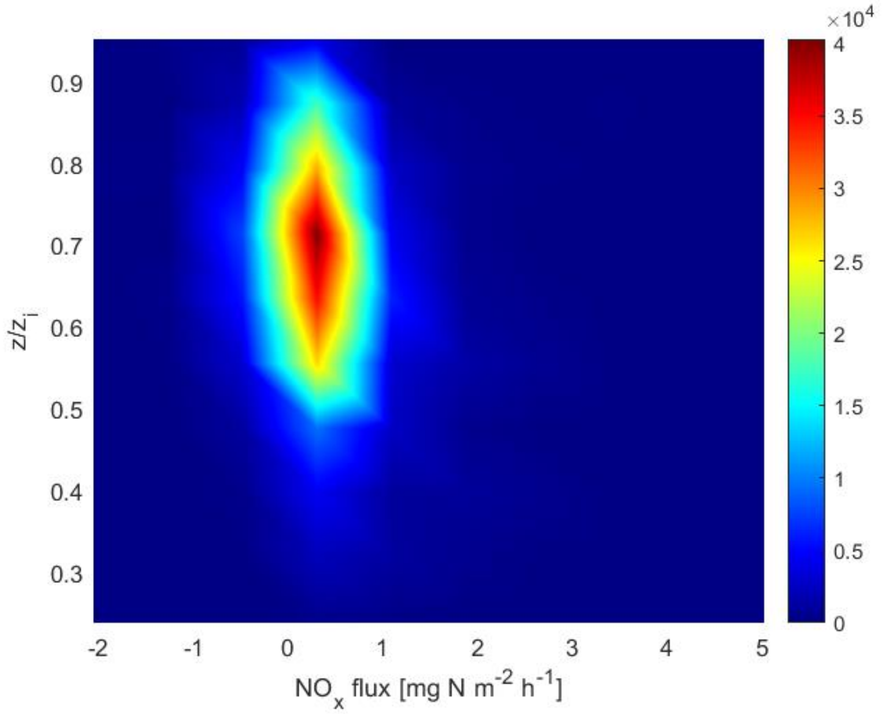


Figure S5. Density plot of the dimensionless altitude z/z_i versus the NO_x flux to show the distribution of the data presented in Figure S4.

Line 195-199: the footprint calculation is dependent on many variables and using the KL04 model. What are the meteo input for the model? Do you use measured data or data from meteo models?

The meteorological inputs for the footprint calculations including the wind direction, the crosswind fluctuations and the vertical wind fluctuations were obtained via a radome flow angle probe which provided 3D wind data. The aircraft's altitude was measured via a C-MIGITS. These measurements are described in detail in Karl et al. 2013). The boundary layer height was determined as described above via changes of water vapor, toluene, temperature and dew point. We have added a reference to Section 2.2, where we describe the acquisition of the meteorological data.

Lines 222 f.: Please find details on the acquisition of the meteorological inputs in Section 2.2.

Figure 4. What do the black lines indicate in figure 4? Are they the flight paths? Please mention it in the caption.

The black lines indicate the contour of the 90% footprints. The flight paths are colored by the calculated NO_x flux. We have clarified this in the Figure caption and in the text.

Figure 4. Flight segments colored by the NO_x flux in geographic proximity on two weekend days, (a) 6 June and (b) 12 June, with different footprint size. The black lines represent the contour of the 90 % footprints. © Google Maps 2023.

Lines 245 f.: The two panels present selected flight segments colored by the NOx flux in geographic proximity (…)

Lines 249 f.: At the same time, the footprint size for these segments, represented by the black lines, was more than 4 times larger (…)

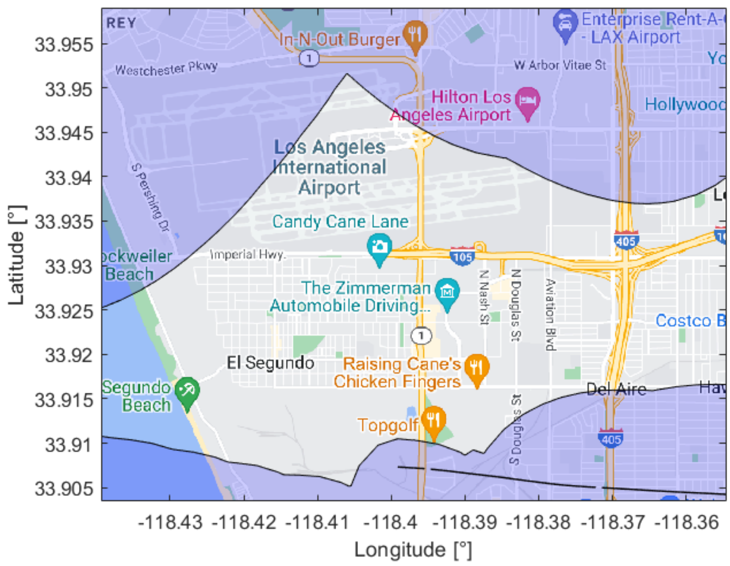
Line 300, what are ‘area emissions’?

Area emissions are fluxes that originate from a larger area, instead of a point source e.g. an industrial facility. They usually represent small individual emissions which accumulate to have a significant contribution. For NO_x, area sources mostly include residential fuel combustion processes such as heating or cooking. We have indicated this in the manuscript and added a reference by the California Air Resources Board.

Lines 331 ff.: We show the NO_x fluxes as predicted by CARB separated into (a) on-road emissions, (b) aircraft emissions, (c) area sources (e.g. residential heating or cooking emissions which accumulate over a larger area (CARB, 2023)) and (d) emissions from ocean going vessels in Figure S11 of the Supplement.

Line 310-313, I don’t understand this part. The airport emissions were not captured by the measurements. How about comparing your NOx fluxes with the CARB emissions excluding aircraft emissions?

The airport emissions were likely only captured to a small extent, which can be seen when looking at the footprints, e.g. around LAX:



The CARB aircraft emission inventory also includes aircraft emissions in further distance to the airport, as well as ground handling equipment and vehicle traffic around the airport. We agree that this was not clear in the text and we have clarified this.

Lines 328 ff.: The large NOx flux in proximity to the coast (~34.0°N, 118.4°W) with a value close to 3.5 mg m^{-2} h^{-1} was associated with aircraft emissions, as well as ground handling equipment and vehicle traffic, from and around Los Angeles International Airport (LAX). Additionally, emissions from aircraft not only at the surface but also at elevated altitudes could contribute to the observed value.

and

Lines 344 ff.: Due to lively air traffic, the research aircraft could not approach the airport closely and the footprints only covered a minor area of LAX airport. As a result, the differences in the vicinity of the airport should not be interpreted as meaningful.

Line 325-330: In Figure 6 and Figure s10, NOx fluxes are quite different. Which one shows better results? Is it necessary to include the correction of vertical divergence in the flux calculation? Why are the emissions enhanced over Downtown Los Angeles and the inland highways in San Bernardino, but lower in the coastal region and Santa Ana? Please Add more discussion about the influence of vertical divergence.

Figure S12 (previously Figure S10) represents the results of a sensitivity study in an attempt to investigate the influence of vertical flux divergence and underlines that this effect could be quite large. While Figure S4 (F_z vs z/z_i) indicate that vertical flux divergence can play a role for example through entrainment from above or horizontal advection, we do not perform a correction of the calculated NO_x fluxes because the correlation between the flux and the dimensionless altitude does not provide significant results. The linear fit of F_z vs z/z_i exhibits an R^2 of only 6% which likely arises from the surface heterogeneity experienced over Los Angeles. This includes heterogeneity in time (e.g. rush-hour traffic) and space (a high variety of sources). Therefore, the results shown in Figure S12 should not be interpreted as unambiguously supporting a specific value for the flux divergence, rather only an idea of the impact of vertical divergence. Unfortunately, with the available data set, we cannot convincingly determine the flux divergence over Los Angeles, and we have to acknowledge this drawback in our analysis. We therefore strongly suggest the characterization of vertical flux divergence over heterogeneous sources to be subject to future studies. We have added some discussion in the manuscript regarding this topic.

Lines 363 ff.: We do not correct the fluxes for vertical divergence as our data set does not provide significant or unambiguous indication for its occurrence and extent. This is likely an outcome of the source heterogeneity experienced across Los Angeles as most emissions are highly variable in time and space. In previous studies, the vertical divergence has been successfully characterized via the correlation of the flux and the dimensionless altitude over homogeneous surfaces, which is not applicable to Los Angeles. Instead, carefully planned stacked race track flights could provide insights into vertical flux divergence. This sensitivity analysis emphasizes how important the characterization of the vertical flux divergence is and should be subject to future studies.

Line 332 change ‘in-situ’ to ‘airborne’

We have changed this.

Section 4: The conclusion section is only a short summary of the results. Please also indicate the implication of the study. What can we learn from the difference between the estimated NOx fluxes and the CARB inventory? What is your conclusion after investigating the influence of vertical divergence. Also discuss the limitation of the study and recommendations for future study.

We have added some discussions in the conclusion section.

Lines 379 ff.: Spatially, the emission inventory overestimated the fluxes in coastal proximity and over Downtown Los Angeles, which could be due to COVID-19 related reductions, such as a shift to more remote work and less commuter traffic, general emission reductions not yet captured by the emission inventory, or misallocation of emission sources in the inventory. In contrast, the emission inventory underestimated the NO_x fluxes over the Eastern part of the San Bernardino valley where an increased activity of trucks going to and from warehouses due to the exponential growth of online retailers, such as Amazon, lead to higher NO_x emissions in recent years. A single uniform correction for vertical divergence could locally lead to improved agreement in this part of the domain, but would at the same time increase the difference in other parts of the studied area. Being an important tool in air quality regulation, we encourage further investigation of the accuracy of local emission inventories with observations from aircraft, towers or dense networks. For flux measurements from aircraft or towers, a particular focus on improving vertical divergence characterization, in order to provide accurate emission predictions would be especially beneficial.