Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-671-RC2, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evolution of NO_x in the Denver Urban Plume during the Front Range Air Pollution and Photochemistry Experiment" *by* Carlena J. Ebben et al.

Anonymous Referee #2

Received and published: 4 October 2017

The manuscript by Ebben et al. describes the photochemical evolution of NOx in the Denver urban plume, taking into account photochemistry, dilution and dry deposition. It thus addresses an important topic, investigation NOx transformation and loss in the continental boundary layer. The topic itself is highly relevant and thus suitable for ACP. Unfortunately, I feel that the paper is unnecessary short, in particular with respect to the presentation of the measurement data and a thorough discussion of the limitations of the analysis. This makes it difficult to judge on the significance of the results from this study. Thus I recommend that the paper should only be published after some major modifications.

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Discussion paper

Major points:

The whole analysis relies on the data presented in Fig. 1. Unfortunately, no information is provided about the original data (number of flights, flight patterns, time series) and no reference is given for the individual flights. Figure 1 shows only median values, with no information on atmospheric variability. I consider it essential that more information is given on the input data, at least please provide flight patterns and information on the variability.

The analysis itself relies on a quasi-Lagrangian approach assuming a spatial and temporal connection between the emission source (Denver), transport and photochemical processing of the plume. Using a statistical analysis (median over several flights?) of measurements at various distances from the Denver source, might establish the spatial connection but not necessarily a temporal relation. I think it is fair to assume, that NOx emissions (and e.g. CO emissions) will exhibit a significant diurnal variation, so that any analysis of the plume evolution has to make sure that this temporal variability of the source has been taken into account. Unfortunately, this question is not addressed in the manuscript. Can we assume that a measurement at a distance of e.g. 50 km away from Denver is indeed related to the NOx emissions at the source 4 hours previously? If not, the limitations of a statistical approach with respect of NOx diurnal variations should be discussed.

Equation 1 relies on the assumption that secondary production of CO can be neglected and I am not sure that this assumption is justified. It can be expected that e.g. HCHO emissions due to mobile sources will lead to enhanced mixing ratios of several ppbv, that will produce additional CO within a time period of several hours. This source might be of similar magnitude as the photochemical sink of CO (1 – 1.5 ppbv according to the estimate made in line 23 on page 4). There might be other secondary sources of CO that could contribute. In any case the limitations and consequences of neglecting secondary CO production on the dilution studies should be discussed.

The discussion on page 6, line 18 – 20 dealing with the derivation of the NOx lifetime from a correlation between NOx and its oxidation products is too short. Please show a least a correlation plot of the data used for this analysis.

More details about the production of peroxy nitrates (LaFranchi et al., 2009) should be provided: e.g. which oxygenated VOC were taken into account?

Minor points:

Equation 2: Is it justified to neglect photochemical sinks for HNO3, e.g. reaction with OH?

Equation 3: Is it justified to neglect physical removal processes (dry deposition, particle production) in the summation of the PNs sinks?

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