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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimates of free-tropospheric NO_2 and HCHO mixing ratios derived from high-altitude mountain MAX-DOAS observations in the mid-latitudes and tropics" *by* S. F. Schreier et al.

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We would like to thank the reviewer for his / her useful comments.

This article reports on MAX-DOAS observations of free troposheric nitrogen dioxide and formaldehyde volume mixing ratios measured from two high-altitude locations: Zugspitze (Germany) and Pico Espejo (Venezuela). Accurate observations of this part of the atmosphere are sparse because with ground-based (i.e. from within the boundary layer) or satellite remote sensing it is almost impossible to do accurate retrievals

of this part of the atmosphere. Therefore the study is quite relevant as a first-order quantification of trace gas abundances in the free troposphere, despite the fact that the retrieved values are at best indicative for volume mixing ratios at other locations around the world.

The paper describes the applied methodology in a clear and concise way and the quality of English writing is high.

It would have been nice if the authors would also have reported free tropospheric trace gas partial columns above the location of the instrument. This complementary information would have been equally unique and relevant and is contained in the MAX-DOAS data set used in this work.

A drawback of the work discussed in this paper is the fact that results are almost not compared to other data sets (except for some references to papers reporting similar values for other locations). However, as measurements of tropospheric trace gas abundances with other sensors are so rare, the lack of comparison with other data sets should not be a reason to reject this study.

In my view, the paper can be published after minor changes described below.

- p.31782, l.7: Please replace “close to the instrument” by “close to the instrument altitude”. Same for the sentence in the conclusions (p.31799, l.22)

We have now replaced “close to the instrument” to “close to the instrument altitude” in the Abstract and Conclusions Sections.

- p.31785, l.1: Please remove “The authors of”.

We have now removed “The authors of”.

- p.31791: About Fig. 2: The residual spectrum of the HCHO fitting window appears to have a considerable amount of structure in it (it does not look like random noise). Are the residual spectra similar for other cases? What could be the cause of these

ACPD
15, C11331–C11334, 2016

structures?

We have tried many different settings during the fitting procedure. We found strong correlation between the residual spectra and HCHO differential optical density when using other fitting windows and/or absorption cross sections. The settings used for HCHO retrieval as presented in the manuscript are by far the best. We agree that there is still some structure in the residual spectrum. We do observe similar patterns for other cases. However, the magnitude of the residual spectrum is lower than the magnitude of the differential optical density of HCHO, which makes us confident to use those settings.

- p.31797: First paragraph: I find the reasoning to exclude the month of June not so convincing. I would say that the NO2 life time in July and May is not so very different from June. Did the authors explore back-trajectory calculations to support the hypothesis that air masses measured in June can be linked to different (pollution) source regions?

We agree that June should not be too different from May and July and have followed the suggestion of the referee and checked backward trajectories for the days of measurements that remain after applying the filter criteria and did not find any month-specific patterns or origin of air masses. We therefore can only speculate that in June, air masses with less NO2 crossed the light path of the instrument. We have now rephrased this paragraph to:

“This slight deviation could be related to the fact that air masses with less NO2 and/or more aerosols crossed the light path of the instrument in June. However, no significant differences in the origin of air masses between the different months could be found by exploring back-ward trajectories for the days of measurements that remain after applying the filter criteria to the data (not shown).”

- p.31797, l.25: Please replace “Due” by “Due to”

ACPD

15, C11331–C11334, 2016

We have now replaced “Due” by “Due to”.

- p.31798, l.1-8: In my view this paragraph should be part of the previous section (as it is about NO2). Please consider to copy it to that section, and then remove the first part of the first sentence: “Before . . . HCHO,”

We have now deleted the first part of the first sentence and copied the paragraph to the end of the previous section.

- p.31801, l.12-15: “This implies . . . free troposphere.” This sentence is difficult to understand and appears gramatically incorrect. Please rephrase.

We have now rephrased this sentence to:

“This implies that there is sufficient NOx for oxidizing hydrocarbons through photooxidative catalytic cycles. As a consequence, production of some tropospheric ozone (O3) is occurring at mid-latitudes in the free troposphere.”

In addition to the reviewers’ comments, we have performed some minor changes: - In the ACPD version, References from Table 2 are not included in the reference list. We have now added them to the reference list.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 31781, 2015.

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15, C11331–C11334, 2016