Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-1215-AC3, 2021

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Pollution trace gases C_2H_6, C_2H_2, HCOOH, and PAN in the North Atlantic UTLS: observations and simulations" *by* Gerald Wetzel et al.

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Received and published: 15 March 2021

Response to Referee #3:

First of all we thank the referee for the effort to carefully reading the manuscript and for all comments. Citations mentioned below are included in the manuscript.

General comments:

The presentation of these new observations, along with the description of the measurement technique, is worthwhile. However, the goals of the paper should be made

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Discussion paper

clearer. The uniqueness of the observations could be more strongly emphasized. It would be helpful to have some sort of validation of the observations through comparison to aircraft data from established measurement techniques.

We included additional motivation in the introduction and in the conclusions to better emphasize the goals of this paper. Since no co-incident in-situ observations enabling a direct comparison (in the sense of a validation) are available, we have checked in detail published airborne datasets concerning the pollution trace gases derived from GLORIA. We can conclude that retrieved GLORIA amounts of these trace gases are within the spread of values measured by in-situ instruments. Related citations are now provided in Section 3.1 individually for each species.

The introduction seems rather awkward, with the discussion of the measured compounds seeming rather disjointed. Perhaps more discussion of the measurement technique and its uniqueness would be more appealing to readers, and then an explanation of why these species are discussed - driven by the capability to measure them. The explanation of their role in atmospheric chemistry could be saved for the analysis discussion. At l. 41, PAN is a 'secondary pollutant', not 'secondary order'. At l. 51, in what sense is ethane 'most important'?

We include and modified sentences in the introduction to make the text and the goal of these measurements clearer. A comprehensive description of the technical issues of GLORIA is given in the papers by Friedl-Vallon et al. (2014) and Riese et al. (2014) and references therein. This is written now more clearly in the text. We find that the description of the pollutant species is better suited for the introduction than for the data analysis section (of course, this approach may be a matter of taste). In line 41 we omitted "order". Line 51: It is the most important non-methane hydrocarbon constituent of natural gas. We modified this clause accordingly.

The purpose of the model results in the paper should be made clearer. Are they being used to provide validation of the observations? It would be more appropriate to just *use the model to explain the distributions and identify the sources of high mixing ratios.*

Of course, the use of the models is not intended to validate the measurements. It should be understood as a kind of intercomparison. One goal was to see if the models EMAC and CAMS are capable to reproduce the locations of the enhanced amounts of pollutants. Another goal was to quantify the differences between measured and simulated data, especially in the case of EMAC where we performed different emission scenarios. Concerning the origin of the detected enhancements, we used backward trajectories and artificial tracers of air mass origin calculations. We tried to make this issue clearer in the revised text.

Using 60-day back trajectories seems rather a stretch. I would not think they are reliable that far back. The forward CLAMS simulations of various regional tracers seem more reliable, so the back trajectories seem unnecessary.

We agree with the referee, that in general, trajectory calculations have limitations due to trajectory dispersion depending on the trajectory length. Therefore the uncertainty of a single trajectory is increasing with the trajectory length, however the variability of a cluster of trajectories starting in the same region, reflects the impact of mixing processes. Therefore, in this study a plenty of back-trajectories are started in the marked regions with enhanced VMR levels. Frequently employed trajectory lengths to study transport processes in particular in the Asian monsoon region range from a couple of weeks to a few months depending on the transport times from Earth's surface to atmospheric altitudes (e.g., Chen et al., 2012; Bergman et al., 2013; Garny and Randel, 2016; Müller et al., 2016; Li et al., 2018; Vogel et al., 2019; Legras and Bucci, 2020; Hanumanthu et al., 2020). In particular the CLaMS backward calculations to analyse the regions with enhanced PAN between 13 and 14.5 km (about 400 K) demonstrate that the transport times from the planetary boundary layer in Asia to the extratropical UTLS over the Atlantic are between 40 and 60 days. Therefore, trajectories up to a lengths of 60 days are necessary in our study to infer the possible source regions of PAN. Further, the endpoints of the trajectories in the planetary boundary layer show a good overall agreement to the results of forward CLaMS simulations for various regional tracers demonstrating that trajectories of a length up to 60 days are suitable for our study.

Moreover, back trajectory calculations have an added valued to the 3-dimensional forward calculations with CLaMS because they demonstrate the detailed transport pathways from the boundary source to the locations of the measurement and its transport times. The trajectory calculations show that the air parcels with enhanced PAN are uplifted by diabatic heating in the upward spiralling range (e.g., Vogel et al., 2019) of the Asian monsoon anticyclone up to about 400 K within about 40 days and subsequent transport (within about 20 days) occurred along the subtropical jet to the extratropical UTLS over the Atlantic. This detailed transport pathway and its transport time cannot be inferred from CLaMS 3-dimensional forward calculations, therefore the back-trajectory analysis is an added value to our study.

The conclusions seem to discuss more the model evaluation aspects of the observation-model comparisons, which I do not find fully justified by the presentation of the results.

The conclusion consists of two main parts. First the findings of the measurements and second, the findings connected with the comparison to the models EMAC and CAMS. We included some text to make this issue more clearly.

References

Bergman, J. W., Fierli, F., Jensen, E. J., Honomichl, S., and Pan, L. L.: Boundary layer sources for the Asian anticyclone: Regional contributions to a vertical conduit, J. Geophys. Res. Atmos., 118, 2560–2575, https://doi.org/10.1002/jgrd.50142, 2013.

Chen, B., Xu, X. D., Yang, S., and Zhao, T. L.: Climatological perspectives of air transport from atmospheric boundary layer to tropopause layer over Asian monsoon regions during boreal summer inferred from Lagrangian approach, Atmos. Chem. Phys., 12, 5827–5839, https://doi.org/10.5194/acp-12-5827-2012, 2012.

Friedl-Vallon, F., Gulde, T., Hase, F., Kleinert, A., Kulessa, T., Maucher, G., Neubert, T., Olschewski, F., Piesch, C., Preusse, P., Rongen, H., Sartorius, C., Schneider, H., Schönfeld, A., Tan, V., Bayer, N., Blank, J., Dapp, R., Ebersoldt, A., Fischer, H., Graf, F., Guggenmoser, T., Höpfner, M., Kaufmann, M., Kretschmer, E., Latzko, T., Nordmeyer, H., Oelhaf, H., Orphal, J., Riese, M., Schardt, G., Schillings, J., Sha, M. K., Suminska-Ebersoldt, O., and Ungermann, J.: Instrument concept of the imaging Fourier transform spectrometer GLORIA, Atmos. Meas. Tech., 7, 3565–3577, https://doi.org/10.5194/amt-7-3565-2014, 2014.

Garny, H. and Randel, W. J.: Transport pathways from the Asian monsoon anticyclone to the stratosphere, Atmos. Chem. Phys., 16, 2703–2718, https://doi.org/10.5194/acp-16-2703-2016, 2016.

Hanumanthu, S., Vogel, B., Müller, R., Brunamonti, S., Fadnavis, S., Li, D., Ölsner, P., Naja, M., Singh, B. B., Kumar, K. R., Sonbawne, S., Jauhiainen, H., Vömel, H., Luo, B., Jorge, T., Wienhold, F. G., Dirkson, R., and Peter, T.: Strong day-to-day variability of the Asian Tropopause Aerosol Layer (ATAL) in August 2016 at the Himalayan foothills, Atmos. Chem. Phys., 20, 14273–14302, https://doi.org/10.5194/acp-20-14273-2020, 2020.

Legras, B. and Bucci, S.: Confinement of air in the Asian monsoon anticyclone and pathways of convective air to the stratosphere during the summer season, Atmos. Chem. Phys., 20, 11045–11064, https://doi.org/10.5194/acp-20-11045-2020, 2020.

Li, D., Vogel, B., Müller, R., Bian, J., Günther, G., Li, Q., Zhang, J., Bai, Z., Vömel, H., and Riese, M.: High tropospheric ozone in Lhasa within the Asian summer monsoon anticyclone in 2013: influence of convective transport and stratospheric intrusions, Atmos. Chem. Phys., 18, 17979–17994, https://doi.org/10.5194/acp-18-17979-2018, 2018.

Müller, S., Hoor, P., Bozem, H., Gute, E., Vogel, B., Zahn, A., Bönisch, H., Keber, T., Krämer, M., Rolf, C., Riese, M., Schlager, H., and Engel, A.: Impact of the Asian monsoon on the extratropical lower stratosphere: trace gas observations during TACTS over Europe 2012, Atmos. Chem. Phys., 16, 10573–10589, https://doi.org/10.5194/acp-16-10573-2016, 2016.

Riese, M., Oelhaf, H., Preusse, P., Blank, J., Ern, M., Friedl-Vallon, F., Fischer, H., Guggenmoser, T., Höpfner, M., Hoor, P., Kaufmann, M., Orphal, J., Plöger, F., Spang, R., Suminska-Ebersoldt, O., Ungermann, J., Vogel, B., and Woiwode, W.: Gimballed Limb Observer for Radiance Imaging of the Atmosphere (GLORIA) scientific objectives, Atmos. Meas. Tech., 7, 1915–1928, https://doi.org/10.5194/amt-7-1915-2014, 2014.

Vogel, B., Müller, R., Günther, G., Spang, R., Hanumanthu, S., Li, D., Riese, M., and Stiller, G. P.: Lagrangian simulations of the transport of young air masses to the top of the Asian monsoon anticyclone and into the tropical pipe, Atmos. Chem. Phys., 19, 6007–6034, https://doi.org/10.5194/acp-19-6007-2019, 2019.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1215, 2020.