Air Pollution in The Upper Troposphere: Insights from In-Situ Airplane Measurements (1991-2018) by Wang et al.

Summary

Wang et al. provides a characterization of tropospheric carbon monoxide (CO) based on the analysis of various in situ observations of CO, O_3, and H_2O by commercial aircraft (IAGOS) and research aircraft campaigns. Complemented by CO data from ground-based observations at Mauna Loa, satellite (MOPITT), emission inventory (EDGAR), and chemistry transport model simulation (IMS), they investigate seasonal variations, vertical profiles and long-term trends of lower and upper tropospheric CO for different periods and various regions of the world. They conclude that CO mixing ratios were increasing in the upper troposphere over the North Pacific between 1991 to 2018 (2004–2022) due to increased ground-level CO emissions while other parts of the world show decreasing trends.

General comments

Combining aircraft in situ observations with various datasets to better understand the processes driving changes and variations in tropospheric CO and its vertical distribution is an important approach and fits within the scope of ACP. However, major revisions are needed before it can be considered for publication in ACP.

The manuscript is difficult to follow, and the results are not clear to me. I'm not always convinced by the argumentation. Therefore, the main results should be more clearly summarized in the abstract and the conclusion, with a focus on highlighting the new findings and their implications. A clearer, more concise structure and logical progression of chapters or paragraphs, along with a deeper discussion, is needed to ensure that the reader is not left to interpret the content alone. This includes improving the quality and clarity of figures by using more readable labels, and by maybe reducing the number of figures. The discussion should also compare the findings with more previous studies. Detailed proofreading would eliminate many oversight errors.

The main difficulty I have is that the datasets are not well described (e.g., instruments, sampling frequency, precision) and the methodology part on how the data are processed is missing (e.g., averaging, filtering/quality check, trend analysis, definition of the upper troposphere, how to compare in situ data with satellite data, etc.). Conclusions were made from trend analysis, although sometimes no statistically

significant trend is seen. Together with missing error bars, it is difficult to fully believe the validity of the interpretation. Limitations and uncertainties of the data should be discussed and accounted for in the conclusions.

Specific comments

Title

The title implies a general view on air pollution in the upper troposphere. But the manuscript specifically deals with CO. Can you make the title more specific?

Is this manuscript the extension/follow-up study of Wang et al. (2024), https://www.researchsquare.com/article/rs-3938611/v1?

Abstract

Lines 4–5: Please use consistent time frames or make the periods you are focusing on clearer. In "We utilize extensive in-situ CO measurements spanning 2012-2023, supplemented by prior airplane campaigns from 1991-2019…", the focus of the study seems to be on the period 2012–2023, and data from the period 1991–2019 are maybe used for comparison. This is not consistent with the title (1991-2018). On the other hand, in the introduction, lines 35–37, it is said "In this study, we present routine in-situ measurements of CO from the PGGM/IAGOS air-based measurement project conducted during the period 2012–2018. Combining these measurements with previous in-situ data collected in various years between 1991 and 2019…".

Additionally, various periods are discussed throughout the text, with slight variations in the timeframes, which is confusing for the reader (e.g., Line 183: 2001-2018 and Line 185: 2000-2018; Line 289: 2004-2021 and Line 292: 2004-2020).

Line 8: Could you briefly summarize the main results and clarify what the "…significant impact of chemistry on CO profiles and trends" is?

Introduction

Lines 23–33: To improve the understanding of the manuscript, it would be helpful to briefly describe the role of CO in upper tropospheric chemistry, particularly in connection to O_3, hydrocarbons, and H_2O, is briefly described, including its major sources, lifetime, and key reactions.

Lines 35–39: "In this study….", does this sentence refer to the current study and describes the results in the following paragraphs (lines 37–54)? Or does it refer to a previous study? Please make it clearer. You could start with "Previous studies revealed…". Results of the current study should not be described in the introduction.

Line 42: Wang et al., 2024 is still under review. This should be clarified. E.g. (Wang et al., 2024, under review). In the reference list, please add the preprint link (https://www.researchsquare.com/article/rs-3938611/v1 ?).

Lines 40–45: Is there a reference for it? I couldn't find it in Wang et al. (2024).

Lines 46–54: To get a better understanding of what is already known, more discussion of past studies, including trend analysis, can be added.

Data and Methods

Generally, I recommend reviewing other relevant papers to see how the datasets are typically described (e.g., Cohen et al., 2018, Osman et al., 2016).

After the part "data description", a "methodology" part should follow. For example, with the sub-sections:

-Study region

-Data processing

Data processing should include how the data is prepared for analysis. For example, monthly averaging within the selected regions, data statistics of each region, how the upper and lower troposphere are defined, how the trend analysis is performed, how the uncertainties are evaluated, and how the data are prepared for comparison with MOPITT data (e.g., Osman et al., 2016, ACP). For the analysis, I also recommend reviewing previous papers on trend analysis to see how the trends are calculated, and

how trends and uncertainties are visualized (Gaudel et al., 2020, Sci. Adv.; Worden et al., 2013, ACP).

Lines 62–74: Please add more details about the dataset: a short description of which instrument is used to measure which constituents (measurement principle). What is the measurement precision? How often is data collected in the regions of interest? In which height does the aircraft usually fly when cruising? For further details, you can refer to previous studies.

Line 73: Add reference to Petzold et al. (2015).

Lines 76–103: Please provide a more concise description of the datasets. In addition, check which information can be omitted, or belong to other parts of the manuscript. For example, lines 76–80 belong to chapter 2.1.3; lines 81–85 belong to the introduction; lines 99 – 103, I would put it in a new chapter "Methodology" with sub chapter "Study regions". As mentioned in the previous comment, more information about the measurement instruments and datasets themselves is needed.

Lines 105–109: As in the previous comments, please give more details about the dataset. Also include information about the location of Mauna Loa and the height of the measurement station.

Lines 111–115: Please add information what instrument MOPITT is (gas correlation radiometer), what it measures (columnar amount of CO) in which wavelength range, footprint size, and its altitude dependent sensitivity to CO. This is important to know when comparing satellite observations with situ data at a specific location and altitude level. Please also include the information of which data version is used and if filtering/pre-processing of some kind is performed.

Lines 129–143: Could you shortly explain why you chose the IMS over other models? What are the advantages?

Are there studies by other researchers using the model, not just your own work? It would demonstrate its relevance and strengthen the choice.

Figure 1 (e): The regions are difficult to distinguish. The use of filled squares or partly thicker lines might improve readability. A table with the definitions of each region, e.g. as Region 1, Region 2, etc., would make it easier for the reader to follow the text. At least, the names should be consistent throughout the entire text.

Results

Figures a described in detail but discussed only briefly. A figure legend is often missing. Figure captions can be made more concisely but should contain explanations about the symbols and time intervals. Figure labels can be increased for readability. Generally, I would place sub figure labels like (a), (b) at the left top instead of the bottom.

Figure 2: There are many sub figures which are difficult to read. Please increase the labels and figures. A legend for the different symbols and the trendline is missing. Labeling the sub figures with (a), (b), etc. is easier to follow than "upper panel, second from left". It might be possible to summarize the sub figures for example by showing the monthly median (50% percentile) as thick line with the 25% and 75% percentile as shaded areas. The caption should describe the symbols and period more concisely.

Lines 165–166: Please provide more detail on how you arrived at this conclusion.

Figure 3: Add a legend with the symbols and trendline; add labels (a), (b), etc.. Why is the trendline of the lower tropospheric data (<1 km, red) partly blue, the upper tropospheric data (>9 km) partly black?

Figure 4: For clarity, it is better to place the figure not directly under the title of section 3.2.

- Figure labels like (a), (b) would be helpful.
- Add a legend.
- I guess the data are from IAGOS in Fig. 4 starting in 2001, and the earlier data of Fig. 3 are from other aircraft campaigns? This can be clarified, too.
- It would be interesting to combine the data from Fig. 3 and 4 to increase the number of datapoints and analyze the specific trends from 1991–2012 and 2012–2018 to investigate if there is a change following the reduction of technological CO emissions over East Asia in 2012.
- According to the figure caption and the text, Fig. 3 and 4 should be the same region. However, the label of Fig. 4 describes different latitudes: "10 N 70N". Please correct.
- The label of the right scatter plot is for the blue symbols and trendline is "<2008…". What is the difference between the red and blue trendline? Does it mean trendlines

for different periods: a) before 2008 (blue), b) after 2008 (red)? Please add this information to the legend and caption.

- Error bars (standard deviation) should be included or at least discussed if they make the figure unclear.
- R and p values are shown, but not discussed. Often, the R values indicate a very weak relation. Some of the p values are around 0.5 or higher, which means, there is no statistically significant correlation. For example, I'm not convinced about the positive and negative trends described in Lines 183–188 or the evident negative correlation as described in Lines 191–193 etc.

Lines 191–209: The vertical pumping mechanism is interesting. Besides a more careful interpretation of the figures, more explanation and discussion are needed. Otherwise, the reader cannot follow the argumentation. For example, explain in more detail what is the implication of a negative correlation between CO and O_3, combined with a negative trend of O_3 in the upper troposphere, and a positive correlation between CO and H_2O. Discuss why the results are different for Western Europe.

Figure 5, Lines 226–227: "Figure 5 clearly illustrates…". Based on the profiles alone, it is not clear that CO is transported upward through vertical pumping. Please provide more explanation.

Figure 6: Please also explain the abbreviations of the figures in the caption. E.g. Euro-Asia region (dashed blue, EuAS).

Lines 228–231: It might be helpful to better highlight the higher variability in the graphics as it is difficult for the reader to see. More explanation and discussion are needed regarding the coincident timing of the higher variability and the reduction of surface emissions of CO, which may suggest a causal relation.

Line 239: How are the trends calculated? Please add this to the methodology part. Are the trends significant? What are the uncertainties?

Lines 244–263: A more specific discussion would be helpful. For example, the importance of chemical sources and sinks for controlling CO levels in the upper troposphere is described, but you can be more specific in the discussion what are the sources and possible sinks.

Figure 7, Line 269: For MLO, please add the height information of the measurement station (middle troposphere) e.g. in a methodology part. This helps to interpret Fig. 7 (a) and the following sub figures when comparing results from MLO with other data of the upper? or lower troposphere.

Figure 7 (b): The IMS simulations are for which altitude?

Lines 274, 277: For clarity, add the months in brackets. E.g. summer (JJA), autumn (SON).

Lines 274–279: The different vertical distribution of CO during Asian summer monsoon and Asian winter monsoon are interesting. Please provide more discussion/explanation on possible processes leading to these characteristics.

Lines 294–296: More explanation is needed to follow this argumentation.

Figure 9 to Figure 12: How are MOPITT data prepared for comparison with model and in situ data? The spectrometer and the retrieval of CO have different sensitivities to the various altitude levels, which are captured in the averaging kernel. Additionally, the footprint of MOPITT needs to be considered for the comparison. These points can cause systematic biases.

Lines 313–316: Do you have any explanation? Are there similar results from previous studies (also see comment for Figure 10)? The satellite instrument has different sensitivities depending on altitude level. This needs to be considered in the comparison.

Line 320: This is a strong statement. How about GEOS-Chem, IASI (Infrared Atmospheric Sounding Interferometer), TROPOMI (TROPOspheric Monitoring Instrument)?

Figure 10: For clarity and readability, can you add a legend describing which color is upper and lower troposphere?

Lines 332–335: Can you add more discussion on the reasons? What are the results from previous studies?

Line 345–347: Please add more discussion why IAGOS is underestimated.

Figure 12: Please try to make the figure clearer by using larger labels, clearer symbols and simple labels for the regions.

Lines 373–375: Can you explain and discuss the result in more detail by keeping in mind the significance of the trends?

Figure 13: For the legend, use a red dot for MLO, black dot for MOPITT.

For clarity, can you add the information for which altitude the IMS simulations were performed?

Figure 14: Better change the order of panels so that time increases from left to right: (a) (a) 1948–1978 (b) 1984–2003

Can you explain more clearly the differences between (a) and (b) and why you have 2 panels? For which region is the simulation performed?

Lines 391–411, 412–425: After the paragraphs which are comparing MOPITT data with in situ and model results, the CO budget calculated by IMS is discussed, followed by a summary of the findings from the trends shown in Fig. 11 and 13. For clarity and to improve the flow, it would be helpful to add some sentences that make a clearer connection between these paragraphs.

Conclusions

Lines 441–457: The conclusion section is very general. Please write down the specific conclusion in detail. For example, our results indicate increasing trends of CO in the upper troposphere over the North Pacific between 1991 to 2018 (2004–2022) due to increased ground-level CO emissions, while other parts of the world show decreasing trends which could be explained by [...].

Limitations of the data and analysis should also be mentioned, and maybe pointed out that further studies are needed.

Technical comments

Line 2: "...CFCs...", please spell out the abbreviation as you did with carbon dioxide (CO_2).

Line 7: "...chemistry budget simulations covering the period 1948–2003..."

Line 36: "…PGGM/IAGOS …": Please spell out the abbreviation when it's used the first time. Also check other parts of the manuscript to see if abbreviations are explained.

Line 45: 68 pptv → 68 ppbv

Figure 1, caption: (a) routues → routes

(e) dwonwind → downwind; North Paicific → North Pacific

(f) UAS → USA, OECD should be spelled out when using the first time.

Line 109: Integrated Modelling System (IMS). Spell out the abbreviation for IMS here when it's used the first time, not in line 129.

Figure 2, caption: measreuements → measurements

Figure 3, label: 123 E–142 E → 123 E–142 W

Figure 4, label: 123 E–142 E → 123 E–142 W; 10N 70N → according to the caption it should be 31°N–42°N. (Generally, I would use the format 31–42°N).

Figure 4 caption: western Pacific → Western Pacific; scattered plot → scatter plot

Line 199: Please add the missing reference.

Figure 5, caption: (a) dwonstream → downstream; ares → areas

Figure 7, caption: (c) Scattered plot analysis → Scatter plot analysis. This also occurs in other parts of the manuscript.

Line 265: MLO is not explained in the text.

Line 283: in (c)→ in (d)

Figure 12, caption: liner gression → linear regression

Line 413: zeros → zero

Lines 548–550: In the current manuscript, HYSPLIT is not used. This and many other mistakes can be avoided by careful proofreading.

Please add a section "Data availability"