

Answer to reviewer 1

This document is the list of our responses to the reviewer's comments and a revised version of the text is also attached to this response to show the changes in red and the deleted sentences using strikethrough text

The manuscript by Ancellet et al. (2024) analyzes results from the summer 2022 ACROSS (Atmospheric ChemistRy Of the Suburban foreSt) measurement campaign. This field campaign was conducted in, and around, the city of Paris and focused on observations of the diurnal and day-to-day variability of ozone (O₃) in the lower troposphere. Vertical profiles of atmospheric constituents were obtained from O₃ and aerosol lidars and commercial aircraft and were combined with radiosondes, IASI satellite retrievals, and CAMS model simulations to understand the processes driving spatiotemporal variability, vertical distributions, and magnitudes of O₃ in the planetary boundary layer (PBL). The manuscript presents in detail the physicochemical characteristics of numerous high O₃ events which occurred between June 13 and July 13, 2022. The text goes to great lengths to describe the agreement in different features (e.g., PBL and RL heights, O₃ concentrations, etc.) observed or simulated by the numerous measurement and modeling tool applied in this study. Four main O₃ events were intercompared for the physicochemical associated with the observed pollution values. The work highlights the importance of ground based O₃ lidars for better understanding air quality. I appreciated the effort the authors have gone through to provide all the details of results from the observations and modeling tools used during the campaign; however, the text does become dense at times. It would be nice if the authors could focus more on the main results of the study without discussing and intercomparing each observation/modeling data source for all 4 pollution events. Also, the novelty of this study is not immediately apparent. The manuscript is generally well-written; however, numerous typos were identified. Please see the minor and major comments below which I think would improve the overall manuscript.

We warmly thank the reviewer for his/her suggestions and comments.

In the introduction the objectives of the paper have been presented more explicitly with the following paragraph:

“The presentation of the O₃ vertical observations available during this period as well as a preliminary analysis of the respective contribution of the urban boundary layer structure and of the O₃ plume regional transport are the main objectives of this paper. The latter has been extensively discussed for North American campaigns listed hereabove, but it is not clear if similar conclusions can be drawn for the Paris area about the role of elevated ozone concentrations transported from outside the megacity area. The Paris area is also different from the places with complicated pollution plume recirculation due to orography or land-sea breeze meteorological forcing where many previous campaigns took place in Europe or North America. Therefore it is relevant to present a study specific to the development of ozone pollution episode in the Paris area.

The overall description of the O₃ variability during the ACROSS campaign and the selection of the pollution events analyzed in this work are presented in section 3.1. This section focusses on lidar observations and their comparison with aircraft and model data. The comparison of the ACROSS O₃ vertical profiles and satellite observations, as well as a comparison of the pollution events in term of regional O₃ transport and PBL dynamical development are discussed in section 4. Section 4.1 first shows to what extent the O₃ measurements discussed in this work are relevant for studying the summer day-to-day variability of ozone in the lower troposphere in Paris, including the potential input from satellite observations. Section 4.2 presents the analysis of the regional O₃ transport during ACROSS since this process has been recognized during the past campaigns as a significant source of variability. Sections 4.3 and 4.4 summarize the main characteristics of the summer pollution episodes encountered during ACROSS and put the results into a broader perspective by comparing them with those of past measurement campaigns”

The structure of the paper has been modified to make the contribution of the work more readable with firstly a section 3 presenting the measurements discussed in the paper with fewer figures and more synthetic and with secondly a section 4 discussing the analysis of the results. We have modified figures 5 to 12 (now figures 5 to 7) and have moved the microlidar data presentation in the supplementary document to focus on the ozone data analysis as requested by the reviewer. A summary table (Table 3) has been added to present the main characteristics of the summer pollution episodes encountered in Paris during ACROSS in section 4.3 and this section has been expanded to present the 3 main findings derived from this work. A new subsection 4.4 is added discussing similarities and differences with results obtained during past campaigns. A careful copy editing of English writing has been made.

Minor Comments

1. Line 1. “profile” not “profiles”
2. Line 10. “shows” not “show”
3. Line 48. “relative contribution of”...
4. Line 57. Is the last comma in this line supposed to be a period? The sentence between Line 55-60 needs some work. It is very hard to follow.
5. Line 62. The impact of long-range transport of O₃ has been shown in studies using ground-based lidar and satellites as well (e.g., Langford et al., 2019, 2022; Johnson et al., 2021).
6. Line 121. “remnants” instead of “remain”.
7. Line 151. There is an extra “)”.
8. Line 154. “The” Copernicus...; and “concentration” should be “concentrations”. 9. Line 151. 10 km × 10 km
10. Line 156-157. Does the author mean “In this work, CAMS model analysis was conducted at 3 daily time steps...? This sentence needs some editing.
11. Line 157. October needs to be capitalized.
12. Line 202. “and” not “or”.
13. Line 229. The authors start to use “O₃” for ozone about halfway through the paper. For consistency, it would be good to just use the chemical formula throughout the manuscript.
14. Line 158. I don’t think you need “downloaded in october 2023” in this sentence. This information is better for the Data Availability section at the end of the manuscript.
15. Line 352. Missing a “)”.

We thank the reviewer for his careful editing of the paper and all these minor corrections are included in the new version.

Major Comments

1. How were the IAGOS and lidar O₃ partial columns calculated? Was the IASI observational operator (averaging kernel and a priori profile) used to calculate these values from IAGOS and lidar data? Same question about the CAMS data shown in Fig. 13. This is an important step in order to have directly comparable information between satellite products and other observed/modeled data.

In the revised manuscript, we have applied the IASI observational operator to the IAGOS, LIDAR and CAMS data. We have changed Figure 13 (new figure 8), to show both the raw and smoothed IAGOS, LIDAR and CAMS data. Finally, we have also modified Table 2 to directly compare raw and smoothed values of O₃ partial columns between IASI and the other observed/modeled data.

Table 2. Mean and standard deviation of O₃ 0-3km partial columns in Dobson Unit (DU) derived from raw and smoothed IAGOS, DIAL, and CAMS data, as well as IASI observations during the ACROSS campaign between June 13 to July 13 2022.

O₃ column (0 - 3 km DU)

	raw			N	smoothed			N
IAGOS	11.56	± 1.93		49	8.53	± 0.40		28
DIAL	12.88	± 2.38		52	8.55	± 0.49		42
CAMS	12.00	± 1.77		32	7.83	± 0.12		19
IASI AM	7.75	± 1.37		19				
IASI PM	6.25	± 0.98		19				
IASI	7.00	± 1.40		38				

We thank the reviewer for his careful editing of the paper and all these minor corrections are included in the new version.

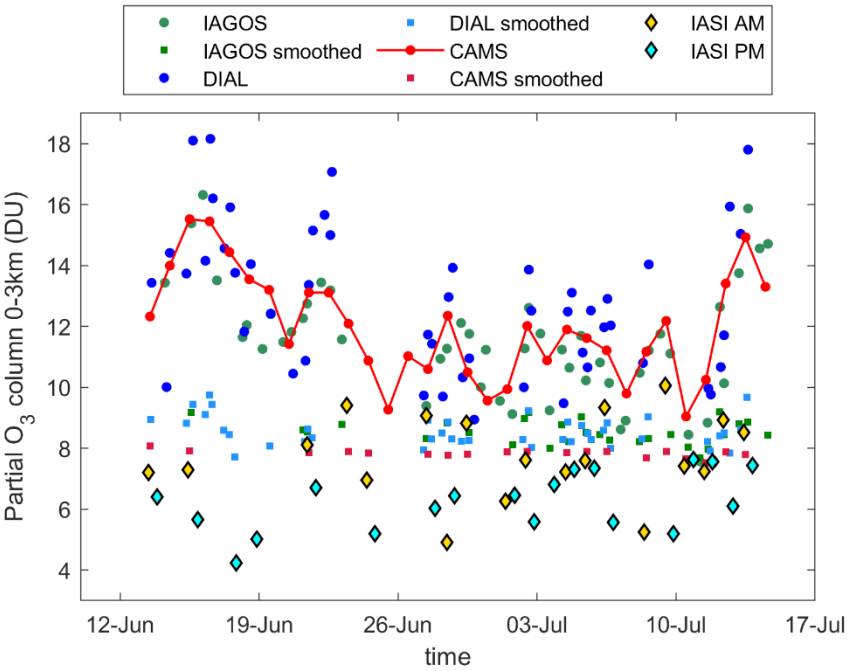


Figure 8. Comparison of tropospheric lowermost O₃ column derived from the ACROSS observations (DIAL in blue and IAGOS in green), CAMS data (in red), and IASI satellite observations (morning – yellow diamonds, and evening – cyan diamonds) calculated in the [48.84°N- 49°N, 2°E-2.5°E] box between June 13 to July 13 2022. Circles and squares correspond to the 0-3km O₃ partial columns and smoothed partial columns, respectively. The orange boxes show the pollution days discussed in section 4.

2. Figure 13. The authors compare IASI 0-3 km partial O3 columns to IAGOS, lidar, and CAMS 0-3 km and 1.2-3 km partial O3 columns. This figure shows that IASI 0-3 km partial O3 columns are much lower compared to the IAGOS, lidar, and CAMS 0-3 km products; however, are more comparable to the 1.2-3 km partial O3 columns from these three products. I am confused why the authors state this is such a good agreement. The IASI 0-3 km partial O3 columns compared to IAGOS, lidar, and CAMS 0-3 km data suggests nearly a 100% underestimation by satellite data. The authors state that satellites have limited sensitivity to lower tropospheric O3, which is true; however, the a priori information in the retrievals still exists. The limited sensitivity only limits the retrieval from deviating from the a priori state. The text reads as if the authors are saying the lowermost tropospheric O3 values in the satellite retrievals will be near zero due to the limited sensitivity. Is this why the authors focus on the comparison of IASI 0-3 km partial O3 columns to IAGOS, lidar, and CAMS 1.2-3 km partial O3 columns? This is not correct.

We agree with the referee and we have removed the comparison with the 1.2-3 km O₃ partial columns in the revised manuscript. Instead, we have analyzed the sensitivity of the O₃ partial columns derived from IASI in terms of deviation from the a priori states, and Degrees Of Freedom for Signal (DOFS). Figure R1 shows that the O₃ 0-3 km partial columns and variabilities derived from

IASOS, DIAL and CAMS smoothed data are systematically lower than those calculated without taking into account the IASI averaging kernels. Figure R1 is only included in the answer to the reviewer. The following text has been added in section 4.1:

Smoothing with the IASI AKs reduces ozone columns and variability because part of the signal information comes from the *a priori* profile which is constant over time. However, IASI observations exhibit a variability of ~ 5 DU (mean of 7.00 ± 1.40 DU) over Paris during the ACROSS campaign, demonstrating that atmospheric signal is present in the retrieval information content with an averaged DOFS of 0.22 and 0.08 for morning and evening measurements, respectively.

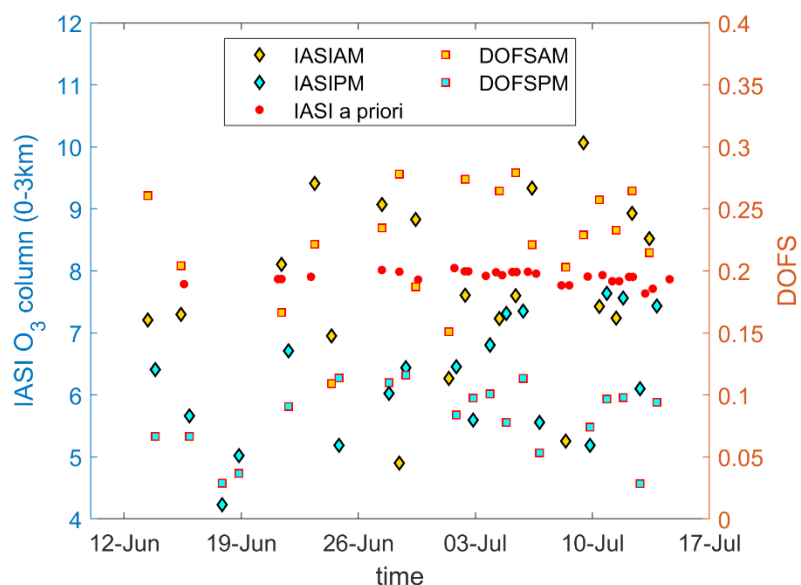


Figure R1: Timeseries of O₃ 0-3km partial columns of the retrievals (diamonds) and the *a priori* states (red dots), as well as Degrees Of Freedom for Signal (DOFS, squares) derived from IASI morning (yellow) and evening (cyan) observations.

3. Line 305-310. The authors are starting to touch on the true limitations of satellite sensitivity to lowermost tropospheric O₃ here; however, don't quite complete the statement. A main reason the satellite data agrees with observations on low O₃ days is that the *a priori* information for IASI is likely based on climatological information. Given the limited sensitivity of satellites to PBL pollution, the retrieval will result in values very similar to the *a priori*. The authors should expand upon this and reference the numerous studies that have been published on this.

Figure R1 above clearly show that IASI retrievals vary with time while the *a priori* column is constant over time. We show, in the new figure 8, that the day-to-day variability of IASI columns is of the same order of magnitude as that of O₃ IAGOS, DIAL and CAMS (5 DU). The following text has been also included in section 4.1:

IASI O₃ columns are overall lower than IAGOS, DIAL and CAMS raw and smoothed columns, with biases of the order of 1-3 DU, in particular when ozone partial columns above 2 km are low, such as between June 14th and 19th, and between June 29th and July 5th. Inversely, IASI and the smoothed IAGOS/DIAL O₃ columns are similar in the case of a high PBL (> 2.5 km) or in the case of high ozone above 2km (> 100 $\mu\text{g}/\text{m}^3$), which are the cases on June 22th, June 28th, and July 12th.

4. For back-trajectory calculations, are there higher spatial resolution meteorological data that could be used to drive these simulations? $1^\circ \times 1^\circ$ ECMWF meteorological data cannot capture the

city-scale features being observed during ACROSS. The entire domain shown in the supplemental figures only encompasses $\sim 2 \times 4$ ECMWF grids.

We agree with the reviewer that the resolution of the ECMWF used for the FLEXPART simulations may limit the analysis of city-scale features but these simulations are only used in section 4.2 focussing on the regional scale transport of the ozone plume. The final horizontal resolution of the FLEXPART simulation output product (here map of PES) is also smaller than the ECMWF grid as Lagrangian model are in principle independent of the initial wind horizontal resolution data (Stolh and Seibert, 1998; Stohl et al. 2002). We do not aim at using such simulations for a detailed description of the city-scale micrometeorological features. The city-scale ozone vertical features are only discussed on the basis of the microlidar data and the Paris radiosoundings. Fig. 9 shows now an example of the output of a FLEXPART simulation in the main paper as requested by Reviewer 2 and the corresponding domain encompasses 10×25 ECMWF grids. This is good enough for our objective.

The following sentence has been included in section 4.2:

“The $1^\circ \times 1^\circ$ horizontal resolution of the ECMWF wind analysis is obviously limited for fine tracking of the city plume, but the PES FLEXPART distributions remain very accurate to check to what extent long range transport must be taken into account in the analysis of the city plume.”

5. This work highlights the importance of O₃ lidar data to better understand air quality and PBL dynamics throughout the day. It would be good to reference the many studies in the literature that have demonstrated this in the past especially those from observations made by the Tropospheric Ozone Lidar Network (TOLNet, <https://tolnet.larc.nasa.gov/>) (e.g., Langford et al., 2017, 2019, 2022; Sullivan et al., 2016, 2017; Johnson et al., 2021). Similar to the work here, these past studies, many conducted during large field campaigns, have shown the impact of local emissions, long-range transport of pollution, PBL heights, RL heights, meteorological conditions, and other physicochemical elements on local O₃ concentrations. These referenced works have focused on UV O₃ lidar observations, combined with ancillary observations and model simulations, to study nearly identical topics focused on in this work. It would be good for the authors to review these past studies and determine the similarities and differences between them and the work presented here by the authors.

We fully agree that the first version of the paper did not sufficiently detail the contribution of the numerous past campaigns, e.g. the results obtained in North America since the setup of the TOLNET network. We apologize for not having been explicit enough on this point, even if the previous introduction already recalled the numerous existing contributions on the role of processes controlling the intensity of pollution episodes. The introduction has been updated with the following text:

“Several campaigns took place in North America to characterize high O₃ summer concentrations: Texas Air Quality Study (TexAQS) 2000 and 2006 and TRacking Aerosol Convection Experiment - Air Quality (TRACER-AQ) 2021 in Southwestern US (Daum 2004, Senff 2010, Liu 2023), California Research at the Nexus of Air Quality and Climate Change (CalNex), California Baseline Ozone Transport Study (CABOTS) 2016, Las Vegas Ozone Study (LVOS) 2016 and 2017 in California (Ryerson2013, Langford2022, Faloona2020), Long Island Sound Tropospheric Ozone Study (LISTOS) 2018 and 2019 in New York City (Couillard 2021). During these campaigns extensive use of aircraft and lidar were conducted to better understand the sources and formation mechanism of O₃ plumes (Langford 2019). Results of LISTOS, CABOTS and TRACER-AQ show that meteorology and boundary layer heights are significant parameters influencing the vertical distribution of O₃ in these areas. Sullivan (2017) demonstrated that residual O₃ layer reincorporation with mixed layer development contributes to a significant part of surface O₃ concentration increase in the afternoon. Contribution of long range transport of O₃ has been also analyzed using airborne differential absorption LIDAR

(DIAL) and satellite. For example it was shown that regional transport of O₃ from Asian emissions over the North Pacific Ocean to California is responsible for a significant part of lower tropospheric O₃ increase in Summer (Lin2012, Langford2017) and that stratospheric-tropospheric exchanges (STE), forest fires and Asian pollution significantly control baseline ozone and therefore O₃ pollution in urban area in North America (Langford 2022, Wang 2021, Faloona 2020).”

A new section 4.4 is now devoted to comparing ACROSS results with those of previous campaigns, in particular those with the TOLNET network:

“LISTOS 2018-2019 and Southwestern USA campaigns took place in places and time periods which can be best compared with ACROSS, i.e. with limited fire and intercontinental pollution and STE. The main difference with LISTOS is the lack of land-sea breeze recirculation for Paris. Ozone concentrations exceeded 200 µg.m⁻³ during LISTOS with stagnation and land-sea breeze recirculation not seen during ACROSS (Couillard et al., 2021). The regional advection of European continental O₃ plume and of Saharan dust outbreak frequently associated to heat wave and pollution episode are also specific of the Paris area. Regarding the comparison with the TEXAQS and TRACER-AQ Southeastern USA campaigns, large O₃ concentrations > 200 µg.m⁻³ are observed near Huston due to the contribution of numerous petrochemical plants in addition to the city emissions (Parrish et al., 2009; Senff et al., 2010), while such O₃ concentrations have never been reached during ACROSS. The same conclusion can be drawn from the comparison with the ESCOMPTE campaign O₃ observations when petrochemical plant and ship emission contributions to O₃ plume formation are comparable to the Houston area (Drobinski et al., 2007). The O₃ long range transport observed during the Southwestern USA campaigns (CABOTS, LVOS) is different from the conditions encountered during ACROSS since STE, fire emission and Asian pollution plume transport significantly contributed to the O₃ inflow upstream of the local emission sources especially at altitudes above 2 km (Langford et al., 2022, 2017; Faloona et al., 2020). The latter makes difficult a direct comparison with the level of O₃ pollution encountered during ACROSS. The main similarity with the ACROSS results is the good agreement between the wide extension of the O₃ streamers shown by both the chemical transport models and the lidar and aircraft observations (Langford et al., 2022; Zhang et al., 2020). Indeed the CAMS model analysis during ACROSS are consistent with the O₃ observations presented in this paper and also show that the role of easterly flow from continental Europe replaces that played by the long range transport of fires and Asian pollution plumes during the Southwestern USA campaigns.”

6. The authors go through great lengths to discuss the physicochemical conditions observed and simulated during the ACROSS. However, the manuscript lacks discussion about what new has been found compared to past field campaigns and publications. The authors state at the end of the paper that “...interaction between the urban layer dynamical development and the O3 plume formation during the day, this work is a first study”. However, there are many studies which have discussed the impact of PBL/RL dynamics, local emissions, and long-range transport on observed O3 formation. Just a small sample of these studies are referenced above. I think the authors could reduce the very lengthy text describing and intercomparing each observation/modeling tool for all four O3 events in order to expand more on the novelty of this study. What new results were found during ACROSS? How does this advance the understanding of air quality? This needs to be discussed in detail because it is not clear to this reviewer that any novel findings were found. The authors should do a much more thorough literature review of this topic in order to identify the novelty of this work.

Again we apologize for not having been explicit enough on the high value of the results available from past campaigns. The use of the word “first study” and “first analysis” in the introduction and conclusion is a grammatical error made by a non-native English writer, we only maint that the paper is a preliminary analysis of the city-scale dynamical feature. This has been corrected.

As said earlier, section 3 has been significantly shortened to keep mainly the presentation of the ozone observations and the CAMS simulations. Section 4 has been expanded to summarize the main findings and add a new summary table (Table 3). The new version of section 4.3 now includes the following text:

“Table 3 summarizes the main characteristics of the summer pollution episodes encountered in Paris. The diversity of long range transport and its role in O₃ variability means that this table can be considered sufficiently representative of the conditions that lead to a summer O₃ increase in a city like Paris. Three main conclusions can be drawn from our analysis:

– Westward advection of the pollution plume from continental Europe enhance the O₃ increase over the city of Paris. The contribution of an increase in O₃ background has already been widely demonstrated for other megacities in North America, such as deep stratospheric intrusions or forest fire plumes (see next section). Deep stratospheric intrusions are rare from May to September in North Western Europe in comparison with North America (Akritidis et al., 2021). Long range transport of forest fire plumes are also detected in Europe, but at higher altitude (>5km) than in North America (Baars et al., 2021) with less contribution to the low troposphere O₃ background. Therefore westward advection of the pollution plume from continental Europe is a significant contribution for the Paris area.

– High temperatures in Paris are often accompanied by a southerly flow carrying Saharan dust in the 2-5 km altitude range over northern France (Israelevich et al., 2012). This study show that the downward entrainment of the low O₃ plume at the top of the polluted PBL must be accounted for to understand a possible mitigation of the PBL ozone increase during a summer heat wave.

– The maximum altitudes of the O₃ plume change from 1.5 km up to 3 km. The capability of IR satellite observations can be assessed using the ACROSS O₃ profile observations. Our study shows that IASI 0-3 km tropospheric O₃ column is sensitive to the day-to-day O₃ variability in the lower troposphere, especially when using the AM IASI observations. The significant underestimate of the 0-3 km partial column when the O₃ plume remains below 1.5 km, is reduced as soon as the plume maximum altitude exceeds 2 km.”

Table 3. Characteristics of the Paris ozone episodes in summer 2022.

Date	14-18 June	21-22 June	28 June (or 2 July)	11-13 July
O ₃ plume altitude, km	<1.5	<2.5	<2.5	<3
O ₃ plume maximum, µg.m ⁻³	170	150	110	150
O ₃ 0-3 km column, DU	14-16	12-13	12	13-15
High temperature, No clouds	Yes	No	No	Yes
PBL height maximum, km	1.5	1.5	2.5	3.0
PBL O ₃ and NO ₂ regional increase	Yes	Yes	No	13 June only
Regional plume above PBL	Dust plume	European pollution	No	No
Bias IASI vs O ₃ profiles, DU	-1.5 to -5	0 to 1.5	-2 to -3.5	0 to -2

7. At times it feels there are too many figures in the paper. All 14 figures in the main text have multiple panels and become overwhelming. It would be easier for the reader if the authors focused their discussion on new findings and condense the figures in order to show the main results. The text is very dense when intercomparing every measurement and modeling tool for each case study. Perhaps the authors could improve the readability of the manuscript by only focusing on main findings instead of discussing every piece of information for every day throughout the campaign. At times it starts to read more like a field campaign report and less like a journal manuscript.

As said earlier we strongly modified Fig. 5 to 14. There are now only 3 figures in section 3 (Fig. 5, 6, 7) to present the DIAL ozone data (including the height of the RL and PBL height as in the first version). The comparison between IAGOS, CAMS and DIAL vertical profiles are now shown in Fig. 7. We keep only the days where the comparison of IAGOS and DIAL is meaningful and we take into account only the lidar data that can be best compared with IAGOS (measurement times are now included in Fig. 7).

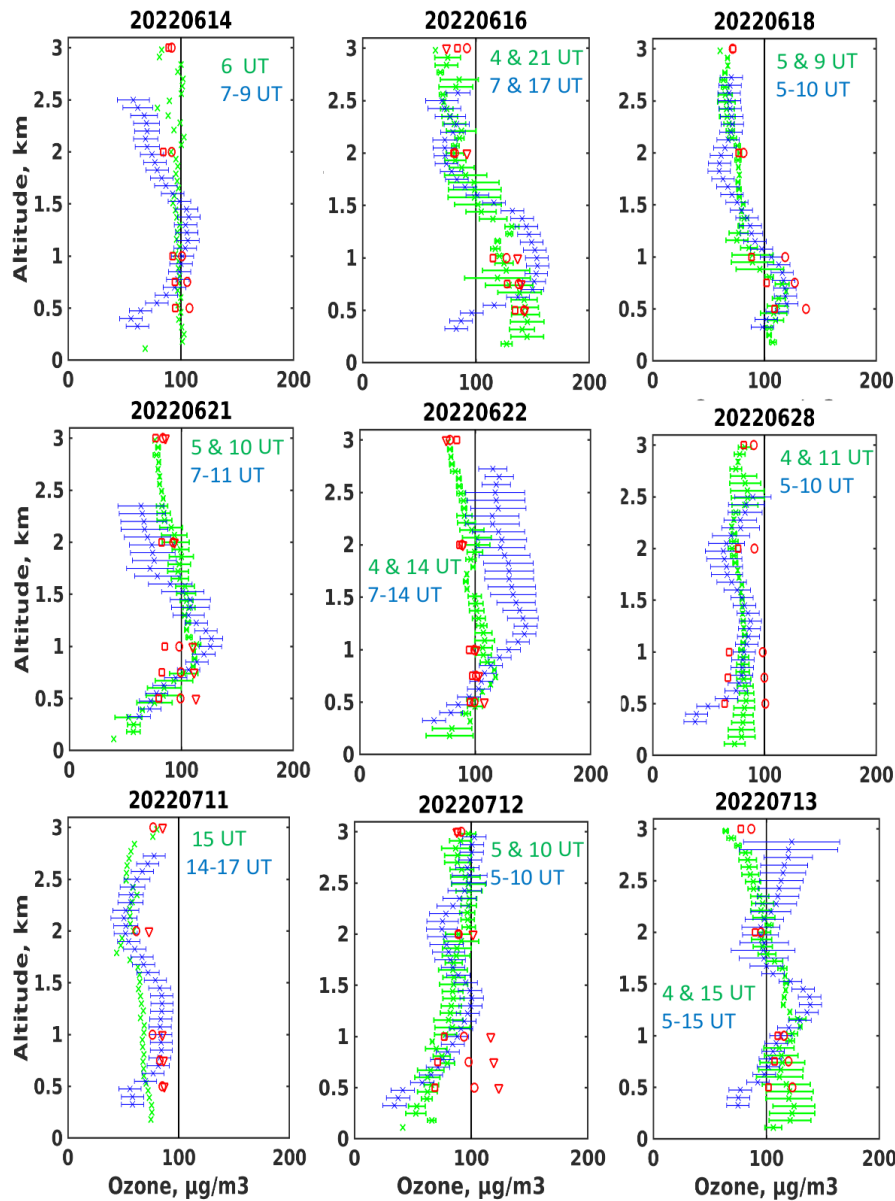


Figure 7. Daily mean O_3 vertical profiles in $\mu g \cdot m^{-3}$ for the IAGOS aircraft (green) and the corresponding DIAL observations (blue) shown in Fig. 5 to 6. Green times in UTC labeled within the figures are the IAGOS measurement times above Paris (two profiles per day except on June

14 and July 11). Blue times below the IAGOS flight times show the selection of the DIAL observations. CAMS model vertical profiles are also shown using horizontal averages of the model concentrations included in the Fig.1 area. CAMS profiles are shown at 6 UT (red □), 12 UT (red ◐) and 18 UT (red ▽).

We agree that the level of detail in the presentation of the different measurement days makes it more difficult to read the summary section 4. However, as in the numerous papers describing measurement campaigns, including those listed by the reviewer, it remains important to provide the reader with the information needed to contextualize the observations. We did our best to balance section 3 and 4 to show that the paper goes beyond a campaign report.

8. The final version of the paper should improve the quality of the figures. Some of the figures appear to have low resolution and some of the symbols used in them are not easy to see.

Fig. 5 to 7 have been changed to make them more readable.

References

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