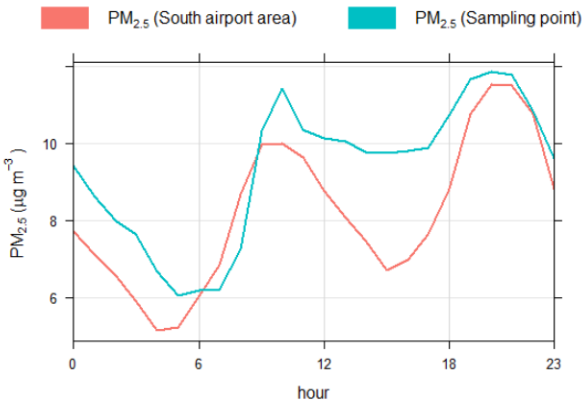
Thanks to the reviewer for their thorough examination of the manuscript and their valuable feedback. We have carefully considered all the comments provided and have incorporated the suggested improvements into the updated version of the manuscript.

RC1

1- With mention of source apportionment, more attention should be paid to understanding the urban contributions. An attempt to quantify the relative contributions of urban vs aviation would be welcome but, if not possible, an explanation of what would be needed to do so would be needed.

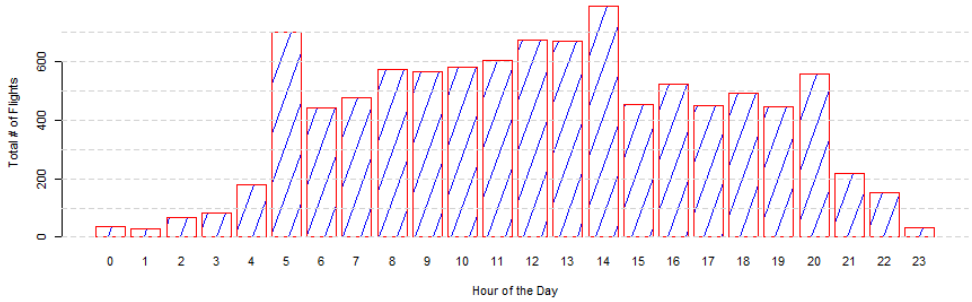
Response: Thank you for pointing this. We provided background particle matter concentrations PM2.5 in comparison with PM2.5 at sampling point. We have added these lines to discuss the potential background impact as suggested by the referee comment.

This is supported by the variance in background particulate matter concentrations located south of the airport compared to those at the sampling point, approximately 6 km apart, as illustrated in Fig. S3. (Fig. S3) reveals that PM2.5 levels at both locations experience significant increases during morning and evening rush hours, with the sampling point consistently showing higher concentrations than the background location. The diurnal pattern of the background location demonstrates a rapid decrease in PM2.5 levels in the afternoon, unlike the measurements at the sampling point. Additionally, there is a noticeable lag of about an hour between the peak concentrations at the sampling point and those in the background, suggesting the influence of additional combustion sources of PM2.5, notably aviation-related activities, particularly during periods of increased airport traffic.



2- an illustrative figure of a time series of 1) an aircraft take-off and 2) an aircraft landing would provide insight into the detailed contribution of aviation compared to the more diffuse background contributions.

Response: Diurnal flight activity been provided to be compared with PM2.5 concentrations located south of the airport and at the sampling point as well as the AlkOA factor.



3- Terminology: Use of SV-OOA—this should be rephrased to "less oxidized oxidized organic aerosol" (LO-OOA) as there is no specific volatility data presented (such a thermal denuder) to determine if it is semi volatile or more volatile than MO-OOA.

Response: Thank you for pointing this out. SV-OOA been rephrased to LO-OOA and we have added these lines for clarity.

Another study by Smith et al. (2022), investigated the chemical composition of organic aerosols emitted by gas turbines and identified a Semi-Volatile Oxygenated Organic Aerosol (SV-OOA) factor, which forms through oxidative processes near the engine exit. A strong correlation (R = 0.91) and similarity in mass spectra between the LO-OOA in this study and the SV-OOA described by Smith et al. (2022) were observed. Owing to the absence of volatility measurements during this period and the limited time for aging (no more than a few minutes), we consider the LO-OOA factor in our analysis to be the most accurate estimate available, rather than the SV-OOA as suggested by Smith et al. (2022).

4- Line 59-60: Jet A1 (in Europe and Jet A in US) is a mix of BOTH aliphatic HCs and **aromatic species** (not just aliphatics). A minimum aromatic content is prescribed, in fact, which is relevant as sustainable fuels are considered. So the aromatic content cannot be ignored.

Response: Thank you for the comment. We rephrased the sentence based on referee comment. We have added these lines about the aromatic fraction.

Aviation fuel Jet A1 is the most common type of fuel that is used in civil aviation. It's a complex mixture of aliphatic hydrocarbons and aromatic compounds, characterized by a mean C/H ratio of ~ 0.52 (with an average empirical molecular formula of C12H23) (Lee et al., 2010). The paraffins fractions in jet fuel typically make up over 75% of the fuel by weight, while the aromatic content is less than or equal to 25% (Liu et al., 2013).

5- Line 91: "is the main international airport" , perhaps add "in Spain"? Otherwise it could be interpreted more broadly.

Response: Thank you for the suggestion. We rephrased it as Adolfo Suárez Madrid-Barajas Airport is the main international airport in Spain.

6- Section 2.3 **Data Analysis**: This section is poorly written and needs to be edited. The verb tense changes throughout ("was operating" vs "were analyzed", etc.) There is significant redundancy and 149-151 is an incomplete sentence or missing verb, etc. Line 148 should read SQUIRREL "operated within Igor Pro", not "supplied by Igor Pro".

Response: Thank you for your comment. We improved the writing of this section based on the referee comments.

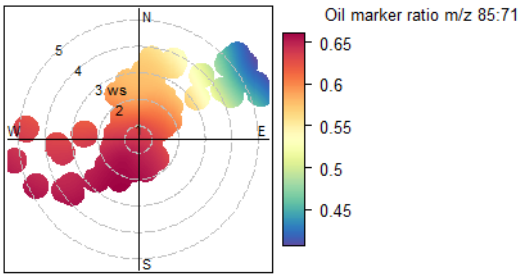
7- Regarding the apparent lack of observation of oil emissions (lines187-202), it might be valuable to cite Ungeheuer et al., 2021 and Fushimi 2019 in line 187

Response: Thank you for suggesting these references to include. They have been added to the manuscript.

8- Some insight on this might be gained by coloring the data stream in figure 3 with the wind direction. If the "little oil" region can be attributed to runway or terminal influenced air and the "unlikely" region is more attributable to urban background, it may well be that the oil (and aviation signal more generally) is swamped by urban background.

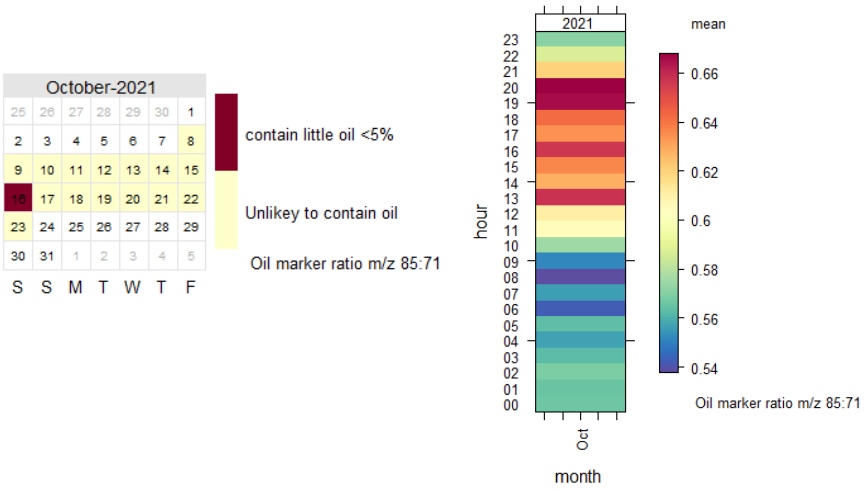
Response: Thank you for your feedback. Bivariate polar plots of lubrication oil ratio measured by AMS is provided to describe the oil contribution. These lines been added to the manuscript for more clarity.

Additional information on how the lubrication oil ratio, as measured by AMS, varies with wind speed and direction, is provided in the supplementary material (Fig.S4). During the AVIATOR autumn campaign, measuring oil was challenging due to the prevalent urban background. A "little oil" region was identified at low to moderate wind speeds (2~5 m/s) originating from the southwest, encompassing terminal buildings (T1, T2, T3, T4, and TS4), two runways (14R/32L and 18R/36L), and a hangar zone. In contrast, a region "unlikely to contain oil" was noted when winds came from the northeast of the airport, near runways 18L/36R, with relatively higher wind speeds (above 5 m/s).



Moreover, Calendar plots for lubrication oil ratio with annotations highlighting the days where the ratio of lubrication oil > 0.66 are also provided. These lines been added to the manuscript for more clarity.

Fig.S5 displays the daily lubrication oil ratio throughout the sampling period, pinpointing Sunday, October 16th, as the only day when the lubrication oil ratio surpassed 0.66. On other days, the ratio suggested a minimal likelihood of oil presence. An hourly analysis within Fig.S5 reveals that the lubrication oil ratio exceeded 0.66 only at 20:00, aligning with the evening peak in PM2.5 concentrations Fig.S3. This suggests a significant influence of urban background aerosols on the lubrication oil measurements.



9- the last sentence (line 202) might better say that the "organic mass may be under-represented in this study".

Response: Thank you for the suggestion. sentence been rephrased.

10- line 227: "an earlier studies" singular/plural mismatch.

Response: Typo been corrected.

11- line 248 "Aliphatic#1" is unclear. If this is referencing PMF analysis from a separate paper, this needs to be rephrased and referenced. (vs AlkOA already described in this manuscript).

Response: Thank you for your comment. This been adjusted to primary aliphatic factor and referenced in the manuscript.

12- line 287-288 discussion of SV-OOA and MO-OOA formation: might be worth emphasizing here that this processing occurs primarily for urban contributions in this study since there is too little time for significant photochemical oxidation of aviation emissions so close to the source.

Response: Thank you for your feedback. This been mentioned in the manuscript and these lines been added for more clarity.

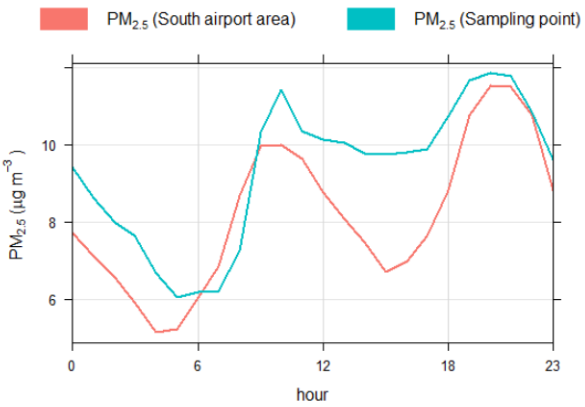
In this study, urban contributions are predominantly subject to this processing, as there is insufficient time for significant photochemical oxidation of aviation emissions in such close proximity to the source.

RC2

1- The discussion overall lacks consideration of the effect of background urban pollution on the measurements performed in this work. Particularly, given the diurnal trend of the pollutants presented in Figures 5 and 6, it is evident that many pollutants show a strong influence from urban emissions. This strong influence from urban emissions could potentially alter some of the main conclusions in the current manuscript.

Response: Thank you for bringing this to our attention. We have included information on background particle matter concentrations (PM2.5) and compared them with the PM2.5 levels at the sampling point. These additions address the potential background impact, as suggested by the referee's comments.

This is supported by the variance in background particulate matter concentrations located south of the airport compared to those at the sampling point, approximately 6 km apart, as illustrated in Fig. S3. (Fig. S3) reveals that PM2.5 levels at both locations experience significant increases during morning and evening rush hours, with the sampling point consistently showing higher concentrations than the background location. The diurnal pattern of the background location demonstrates a rapid decrease in PM2.5 levels in the afternoon, unlike the measurements at the sampling point. Additionally, there is a noticeable lag of about an hour between the peak concentrations at the sampling point and those in the background, suggesting the influence of additional combustion sources of PM2.5, notably aviation-related activities, particularly during periods of increased airport traffic.



2- Section 2.1: Please note the distance from sampling location to the runways, terminals, and nearest highway in Fig.1 or text.

Response: Thank you for your comment. This been addressed in the following lines.

The distance from sampling location to the runways 18L/36R, 18R/36L, 14L/32R and 14R/32L are 680 m, 620 m, 3.2 km, and 4.1 km respectively. Furthermore, the distance between sampling location and adjacent terminals T1, T2, T3 is approximately 5 km whereas 3 km and 1.5 km to the terminals T4 and TS4 respectively. Nearest highway is located around 2.6 km away from the sampling location.

3- Section 2.2: Please give a more detailed description of AMS, including the AMS inlet used here (PM1 or PM2.5?), and the time resolution of AMS.

Response: Thank you for the suggestion. More detailed description of AMS been included to the manuscript.

An aerodynamic lens is used to draw aerosols into a vacuum chamber. Particles are focused into a narrow beam and accelerated to a velocity inversely related to their vacuum aerodynamic diameter. The particles impact on a tungsten surface, heated to 600 °C, which causes them to flash vaporise. A 70-eV electron is used to ionize the vapours before they are analysed by mass spectrometry. During the measurement period, AMS was sampling with 1μm cut-off inlet and at 30 s time resolution.

4- Section 2.3: Please add more details of how the authors selected the PMF factors and explain the technical terms used (e.g. FPEAK, Q/Qexp).

Response: These lines been added to the manuscript for more clarity

Seed runs and fpeak are rotational techniques in ME-2 tool and they are one of the unconstrained PMF run approaches used for the exploration of the solution space.

Furthermore, the obtained solution exhibited the most favorable results, characterized by distinct diurnal trends and dissimilarities in time series and mass-to-charge ratios among the factors.

5- Rewrite sentence line 149-151.

Response: sentence been rewritten and added to the manuscript

The Source Finder (SoFi) is a software package designed to analyse multivariate data using state-of-the-art source apportionment techniques to understand the sources of various pollutants (Canonaco et al., 2013)

6- Section 3.1: are the signal at m/z 85 and m/z 71 used here all from AMS UMR analysis?

Response: Yes, signal at m/z 85 and m/z 71 from AMS UMR. As suggested by (Yu et al., 2012) the m/z 85 signal is an oil signal in the AMS mass spectrum. The ratio of m/z 85:71 is used as a marker for oil. The value of 0.66 is used as a benchmark for oil lubrication identification.

7- When comparing the results from this study to those presented by Yu et al. in 2012, it's important to consider other factors that may be contributing to the very low presence of oil-related aerosols in your findings. These factors could include the overall mass loading of aerosols, the influence of urban aerosol emissions, or the proximity of the sampling location to the nearest runways.

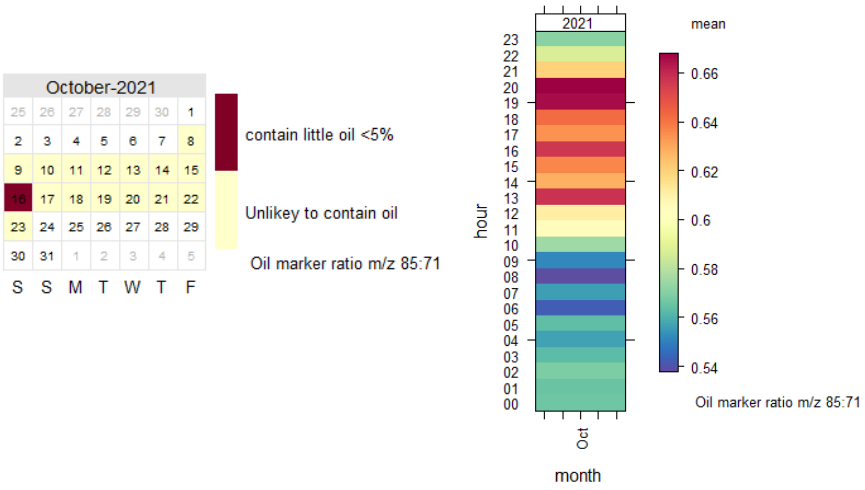
Response: Thank you for your suggestions. This been addressed as follow:

Additional factors that could potentially impact the minimal presence of oil lubrication in this analysis might involve the overall mass loading of aerosols, the influence of urban aerosol emissions, or the proximity of the sampling point to the nearest runways.

Calendar plots for lubrication oil ratio with annotations highlighting the days where the ratio of lubrication oil > 0.66 are also provided. These lines have been included in the manuscript to enhance clarity.

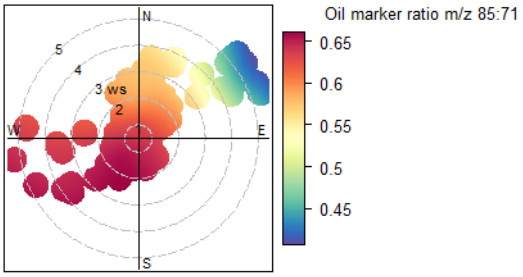
Calendar plots displays the daily lubrication oil ratio throughout the sampling period, pinpointing Sunday, October 16th, as the only day when the lubrication oil ratio surpassed 0.66. On other days, the ratio suggested a minimal likelihood of oil presence. An hourly analysis within Calendar plots

reveal that the lubrication oil ratio exceeded 0.66 only at 20:00, aligning with the evening peak in PM2.5 concentrations Fig.S3. This suggests a significant influence of urban background aerosols on the lubrication oil measurements.



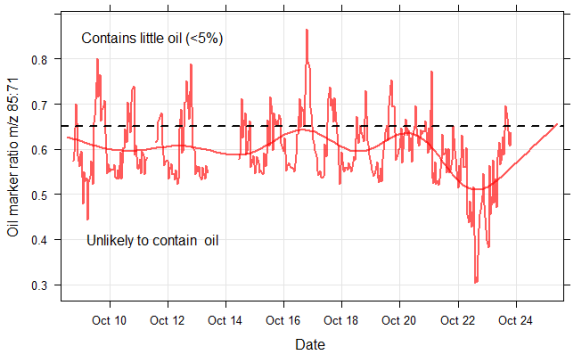
Moreover, Bivariate polar plots of lubrication oil ratio measured by AMS is provided to describe the oil contribution. These lines been added to the manuscript for more clarity.

Additional information on how the lubrication oil ratio, as measured by AMS, varies with wind speed and direction, is provided in the supplementary material (Fig.S4). During the AVIATOR autumn campaign, measuring oil was challenging due to the prevalent urban background. A "little oil" region was identified at low to moderate wind speeds (2~5 m/s) originating from the southwest, encompassing terminal buildings (T1, T2, T3, T4, and TS4), two runways (14R/32L and 18R/36L), and a hangar zone. In contrast, a region "unlikely to contain oil" was noted when winds came from the northeast of the airport, near runways 18L/36R, with relatively higher wind speeds (above 5 m/s).



8- The values displayed in Fig 3 are noisy and show a broad range. It may be beneficial to further average or smooth these values for a clearer presentation of the data.

Response: Thank you for your comment. Oil marker plot has been adjusted accordingly.



9- Section 3.2: the main conclusions in this part are same as previous studies. Are there any new findings from this work?

Response: Thank you for your feedback. Yes, there are. We measured and identified indene as a marker at Madrid Airport in the near runway ambient samples. The following lines been added to discuss the findings.

The first factor in Fig.4, LO-OOA, stands for Less Oxidised Oxygenated Organic Aerosol. It is a type of secondary organic aerosol (SOA) characterized by its low degree of oxidation. LO-OOA are formed in the atmosphere through the oxidation of volatile organic compounds (VOCs), which can originate from a variety of anthropogenic sources. In this analysis LO-OOA shows the presence of an aromatic marker at m/z 115, a marker used for identifying indene (C9H8) ion in previous studies focusing on aviation emissions (Timko et al., 2014; Smith et al., 2022). LO-OOA is associated with aromatic fragments at m/z 77, 91, 105, 115 and presents a high relative intensity (0.13) at m/z 43

(characteristic of LO-OOA) and a lower relative intensity (<0.04) at m/z 91 which is related to toluene ion (C7H7) (Smith et al., 2022). Ambient temperature plays a crucial role in influencing the LO-OOA factor, displaying significant diurnal fluctuations. The lowest concentrations of LO-OOA are recorded at midday, coinciding with the peak in ambient temperatures (Fig. 5).

10- The sources of MOOOA need to be discussed.

Response: Thank you for your comment. A more detailed discussion on MOOOA been added to the manuscript.

The third factor, More Oxidised Oxygenated Organic Aerosol (MO-OOA), is a type of secondary organic aerosol (SOA) that can form from various origins and processes, such as photochemical processing of aged SOA and the regional-scale transport of chemical reactions. MO-OOA has a spectral fingerprint that consists of more oxidised ions (compared to LO-OOA and AlkOA), indicating a secondary aerosol fraction in the sample. MO-OOA is characterized by its notably high relative intensities (>0.18) at m/z 29 and 44, which serve as markers for its identification. Given that MO-OOA has the highest f44/43 ratio among the three factors, it is expected to be the most oxygenated (in terms of chemical content) factor. Being more oxidised potentially makes MO-OOA less volatile than LO-OOA (Jimenez et al., 2009; Smith et al., 2022). MO-OOA in this analysis indicates the formation of aged secondary organic aerosols with no significant diurnal variation (Fig. 5), often associated with air masses transported from polluted regions.

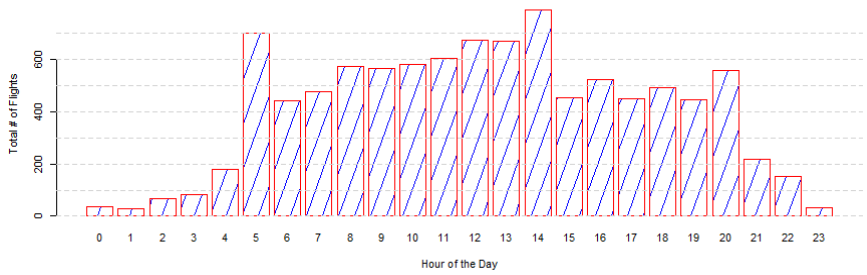
11- Line 248: the term “aliphatic #1” is from the reference, please avoid using it directly without explanation.

Response: Thank you for your comment. This been adjusted to primary aliphatic factor and referenced in the manuscript.

12- Section 3.3: concentration of AlkOA is suggested to be influenced by flight activity, please include such information in main text or SI.

Response: Thank you for your comment. Diurnal flight activity data has been provided for comparison with the AlkOA factor. These lines been added to the manuscript

The concentration of AlkOA factor is relatively higher in the afternoon compared to the morning and midday. The pattern of diurnal AlkOA closely resembles that of diurnal flight activities, suggesting that the surge in AlkOA levels beginning at noon is linked to primary particles released by aircraft. Further details on daily aircraft activities can be found in the supplementary material (Fig. S2)



13- Line 291: please further explain the method of normalization.

Response: Thank you for your comment. This been explained as the following.

Normalising is accomplished by dividing the concentrations of the pollutants by their average value.

14- Line 298-300: the authors suggested the diurnal trend of the pollutants listed in Fig 6 were all similar, but it is very clear that BC, NOx, and CO have two peaks, total number concentration starts to increase mid-morning and maintains high during the daytime, while AlkOA only has one big peak around evening. Please provide further evidence for the conclusion. In Fig 6, at hour 23, the normalized level of AlkOA is about 1.9, but at hour 0, it's about 0.8, can you please explain the big change here? I'm assuming the change from 1.9 to 0.8 happened within one hour since this plot describes diurnal cycle.

Response: Thank you for your comment. This has been addressed as below:

The daily trend of eBC, NOx and CO are mostly similar, with very pronounced increases in concentrations during the morning and evening rush hours. The average concentrations were 1.07 μg/m3, 22.7 μg/m3 and 0.23 mg/m3 for eBC, NOx and CO respectively (Table S1). AlkOA gradually increases during the morning, with multiple minor peaks observed in the morning hours. The average concentration of AlkOA is higher at night than during the day. This increase is potentially related to daily aircraft activities. AlkOA began to increase, reaching a maximum during the afternoon rush hour from 12:00-18:00. a second rapid increase occurred around 20:00, potentially caused by an increase in the number of flights at this time (Fig. S2). Early morning AlkOA concentrations are significantly lower compared to those of eBC, NOx and CO. This difference could be attributed to reduced emissions resulting from decreased aircraft activities in early mornings (Fig. S2). The rise in trace gases and eBC observed in the early morning hours could originate from various airport operations. Such operations might encompass emissions from auxiliary power units, vehicle traffic, and the use of ground service equipment at the airport (Masiol and Harrison, 2014). The total number concentration exhibited a temporal pattern similar to that of AlkOA from 15:00–21:00. Likewise, the temporal profiles of AlkOA and trace gases were similar during the afternoon period (17:00-21:00). This similarity in temporal profiles suggests common source origins, which may be temporally associated with aircraft activity or the influence of background urban pollution.

15- Section 3.4 Fig.8 and Fig.9: Please explain why the authors selected AlkOA vs. eBC and SVOOA vs. THC for correlation analysis.

Response: Thank you for your comment. This been addressed as follow:

- Based on the strength of the relationship outlined in Table 1 between derived factors and external tracers, the linear correlations (Pearson correlation) between (i) AlkOA with eBC and (ii) LO-OOA with THC

- Both AlkOA and eBC are related to jet fuel emissions, as they are directly emitted by aircraft engines as a result of fuel combustion. eBC emissions are a function of engine power settings, reaching their maximum at full thrust during take-off (Kinsey et al. 2011; Hu et al., 2009).

- THC emissions at airports primarily dependent on the jet engine thrust setting (Anderson et al., 2006; Onasch et al., 2009). When engines operate at low thrust settings (e.g., during landing, taxiing, idling), combustion is less efficient, leading to the emission of higher amounts of hydrocarbons. The association between LO-OOA and THC in certain areas of the airport can be interpreted as indicative of fresh emissions from aircraft in service.

16- why selected SO2/NOx and CO/eBC ratios for analysis of aircraft activity? What do these ratios imply in terms of aircraft emission?

Response: Thank you for your comment. This has been addressed and included in the manuscript as follows:

Given that Barajas airport is situated near Madrid and significantly influenced by external sources, particularly traffic on the southwest side of the airport, it experiences considerable environmental impact. Therefore, the ratios of SO2/NOx and CO/eBC were used in this analysis as indicators of the relative emission strengths associated with aircraft movements.

The SO2/NOx ratio would increase in the case of aviation emissions compared to traffic emissions, since NOx emissions from aircraft are difficult to distinguish due to the major influence of other sources (Yu et al., 2004; Carslaw et al., 2006). Consequently, in situations where there are substantial levels of NOx emissions, the SO2/NOx ratio will be low due to the impact of on-road vehicles emissions. This enables the identification of aircraft's relative contribution at the airport, as shown in Fig.9.

SO2 emissions are primarily associated with the sulphur content of the fuel and emissions from aircraft activities at the airport, such as approach, taxi-idle and climb. As a result, SO2 plays a significant role in tracing aircraft emissions at a local scale (Yang et al., 2018).

Black carbon (eBC) and carbon monoxide (CO) are primarily produced by incomplete or inefficient combustion. Around the airport perimeter, aircraft are a significant contributor to CO emissions. Therefore, it's possible for aircraft engines to emit more CO compared to emissions from road traffic, due to the duration spent at the airport in taxiing /idling mode (Yu et al., 2004; Zhu et al., 2011).

The CO/eBC ratio significantly varies with the source (Bond et al., 2004), indicating the presence of different emission sources in the vicinity of the airport, as previously reported.

17- The authors suggested CO was mainly related to road traffic emissions based on previous paper (line 379-380), then the authors related CO measured by the monitoring site with aircraft activities (line 390-392). Could you further explain your discussion and conclusion here?

Response: Thank you for your comment. Being predominantly emitted by on-road vehicles, at low engine settings (taxi/idle) aircraft also emit significant amount of CO. This has been clarified in the manuscript as follows:

Aviation activities have previously been reported as a significant source of gaseous and vapour-phase pollutants, such as SO2, CO and NOx (Masiol and Harrison, 2014). In the same vein, mobile sources, such as vehicle exhaust, generally contribute to the increase in CO and NOx levels, as motor vehicle emissions are the dominant sources of CO and NOx emissions in urban areas (Yu et al., 2004). Around the airport perimeter, aircraft are a significant contributor to CO emissions. Therefore, it's possible for aircraft engines to emit more CO compared to emissions from road traffic, due to the duration spent at the airport in taxiing /idling mode (Yu et al., 2004; Zhu et al., 2011).