Response to reviewer 2

The authors thanks the reviewers for their thoughtful evaluation of our manuscript and for their constructive comments and suggestions. We appreciate the time and effort devoted to reviewing our work, which have helped us improve its quality and clarity. Below, we provide point-by-point response addressing the concerns raised.

Main comments

1. Photochemical age and conversion to absolute time.

The conversion of the hydrocarbon ratio into an absolute photochemical age relies on several assumptions that require greater justification and uncertainty analysis.

Most importantly, the use of a constant [OH] = 5×10^6 molecules cm^{-3} directly determines the calculated age in hours and therefore all subsequent rates expressed in h^{-1}. Please provide the basis for this value and quantify how potential variation in OH affects the calculated ages and associated temporal slopes.

Reporting the underlying OH exposure in addition to the converted age would help distinguish what is directly constrained by the VOC measurements from what depends on the assumed OH concentration.

Please also define precisely what is included in the PTR-ToF-MS "C8 aromatics" signal and justify the use of a single reaction rate coefficient with OH ($1.4x10^{-11}$ cm^3 $molec^{-1}$ s^{-1}), since OH reaction rate coefficients for C8 aromatic isomers vary by roughly a factor of 2-3.

Figure S1 clarifies that a value of approximately 0.80 is derived from an upper-envelope fit of C8 aromatics against benzene. However, the physical basis for treating this upper-envelope slope as the zero-age source ratio should be explained. The non-zero intercept also raises the possibility that background concentrations and dilution influence the ratio.

Finally, the chemically derived quantity should be described primarily as an OH-equivalent photochemical age rather than directly as elapsed transport time unless independently validated. Since HYSPLIT trajectories have already been calculated, comparison between trajectory transport times and hydrocarbon-derived ages would provide a useful independent check.

Response:

We thank the reviewer for this insightful comment.

The value of [OH] = 5×10^6 molecules cm^{-3} was selected as a representative average daytime OH concentration for summer urban plumes in Paris during field campaigns, following established literature (Michoud et al., 2012; Borbon et al., 2013; Freney et al., 2014). To make the results independent of the assumed [OH], we now report both the OH exposure ($t \cdot [OH]$ in units of molecules cm^{-3} s) and the converted OH-equivalent photochemical age in hours (Figures 4 and 5a). Additionally, as requested by the reviewer, we performed a sensitivity analysis varying [OH] across a range (2×10^6 to 8×10^6 molecules cm^{-3}). We have included the test in the supporting information (Text S2 and Figure S5a) and we clarified the used of the selected [OH] value in section 2.5 (lines 298-300).

In our PTR-TOF-MS measurements, the C8 aromatics signal at m/z 107.086 ($C_8H_{11}^+$) represents the

sum of aromatic isomers, primarily ethylbenzene and xylene isomers. The effective rate constant used (k_{C8} = 1.4x10^{-11} cm^3 molec^{-1} s^{-1}) corresponds to the weighted-average OH reactivity based on typical urban C8 isomer emission ratios, reported in prior hydrocarbon-age studies in urban plumes (Warneke et al., 2007; Borbon et al., 2013). We have added a sensitivity test using values ranging from ethylbenzene (7 x10^{-12}) to *m*-xylene (2.3 x10^{-11}) in the supporting information (Text S2 and Figure S5b). Clarifying text has been added to section 2.1 (lines 157) and section 2.5 (lines 300-303).

Results of the sensitivity analysis varying [OH] and the reactivity of the chosen C8 isomer (Figure S5) show that, although the absolute OH-equivalent photo-chemical age expressed in hours varies depending on the assumed [OH] concentration (0.13 h to 0.52) and the reactivity of the chosen C8 isomer (0.12h to 0.46 h), the relative chronological order of aging of the plume air samples remains identical (R = 1.0). Furthermore, the integrated exposure to OH (OH_{exp} = [OH] · Δt) remains constant regardless of [OH]. To explicitly quantify the sensitivity, varying [OH] from 2×10^6 to 8×10^6 molecules cm^{-3} modifies the absolute OH-equivalent photochemical ages by -37.5% to +150% around the nominal median (0.21 h, spanning from 0.13 h to 0.52 h). Similarly, the associated temporal slopes scale linearly with [OH], resulting in a proportional uncertainty range of 60% relative to the baseline rates. However, because this scaling factor is uniform across all data points, the relative chronological aging sequence and correlation coefficients among samples remain completely unaffected.

The physical basis for treating the upper-envelope slope as the zero-age source ratio is now explained in section 2.5 (lines 289-296). To answer whether background and dilution bias our zero-age ratio (t_0), we evaluated alternative formulations using background-corrected VOC enhancements (ΔC8 aromatics / Δbenzene), where regional background concentrations measured outside the plume were subtracted prior to fitting (Figure S4), yielding in an upper-envelope slope of 0.93 pptv pptv^{-1}. Furthermore, as characterized in the instrumental overview of the HR-PTR-ToF-MS by Michoud and Bouzidi (2024), technical challenges between flights (such as power losses and vacuum fluctuations) introduced variable instrumental background levels and baseline offsets for several volatile organic compounds. Applying t_0 = 0.93 or forced background-subtracted ratios to our specific airborne dataset introduced numerical artifacts at low mixing ratios. Specifically, it compressed young air mass observations and yielded non-physical near-zero or negative ages for fresh plume transects (affecting 77% of the data due to underlying instrumental noise and limits of detection). Consequently, maintaining t_0 = 0.80 pptv pptv^{-1} provided the most robust and stable value to calculate the OH-equivalent photochemical age. We have added a discussion in the supporting information (Text S1 and Figure S4) detailing this sensitivity test justifying the non-zero intercept.

Following the reviewer's suggestion, we compared the hydrocarbon-derived OH-equivalent photochemical ages with physical transport times calculated from HYSPLIT forward-trajectories. The trajectory-derived physical transport time shows a statistically significant positive correlation (Pearson coefficient R of 0.59, $p < 0.001$, N = 16) with the chemically derived photochemical age. The positive slope (0.63) support that VOC-derived aging tracks physical transport. The slope offset indicates that the campaign-average effective OH concentration encountered along the trajectory pathways was slightly lower ($[OH]_{effective}$ ~ 3.15 ×10^6 molecules cm^{-3}) than the nominal 5.0 × 10^6 molecules cm^{-3} baseline. Adjusting the photochemical age to $[OH]_{effective}$ yields a near 1:1 agreement with HYSPLIT physical transport times. A scatter plot comparing trajectory transport time vs. OH-equivalent photochemical age has been added to the supporting information (Figure S8), and we discuss these results in section 3.3 (lines 410–417). Additionally, temporal ages recalculated under this effective [OH] range are provided bellow Table S4.

2. Derivation and interpretation of the reported 0.25 h^{-1} AAE growth rate.

Table S4 raises an important issue regarding the reported AAE growth rate.

The three reported binned values are approximately:
(t,AAE)=(2.0,1.1);(6.7,1.6);(8.0,2.6).

The stated parameterisation, AAE=0.25t+0.60 appears to be obtained by connecting the first and last binned medians:
(2.6-1.1)/(8-2)=0.25 h^{-1}.

However, this equation predicts an AAE of approximately 2.28 at 6.7 h, whereas the reported median at that age is only 1.6. The three median points therefore do not appear to support a particularly linear growth.

Please clarify exactly how the 0.25 h^{-1} slope was derived. I recommend calculating the relationship directly from the paired underlying observations, with an uncertainty/confidence interval, rather than from the difference between the first and last binned medians.

Table S4 also shows that the number of observations contributing to different variables within the same O_X/BC bin is not consistent across variables. Please clarify whether the photochemical-age and AAE values used to derive the reported slope are based on temporally matched observations. Ideally, the AAE-photochemical-age relationship should be calculated only from matched measurements.

Addressing these points, together with the uncertainty in the assumed OH concentration, I recommend describing 0.25 h^{-1} as an empirical AAE growth slope only if it remains supported after reanalysis. It should not be referred to as a BrC production rate.

Response:

We thank the reviewer for rising this point regarding the reported AAE_{aer} growth rate.

First, in the revised manuscript (section 2.1, lines 176-181, section 2.4 lines 261-264), we clarified how we temporally matched observations to evaluate relationships within all the variables described in the manuscript. Specifically, in Table S4 we now described that bins were populated based strictly on the availability of synchronised data for each specific chemical or optical property analysed. In addition, we clarified why the number of observations contributing to different variables within each bin is not consistent across variables.

We calculated the empirical slope directly from the AAE_{aer} and OH-equivalent photo-chemical age all individual paired observations and by aggregated at the plume-intercept level using Ordinary Least Squares (OLS) and Pearson linear regressions. However, the linear fit yields a low coefficient of determination across the individual paired observations (Pearson coefficient R = 0.18, p = 0.199, N = 51). Thus, as the reviewer suggested, we removed the empirical parameterization (AAE = 0.25t + 0.60) and all references to a "BrC production rate of 0.25 h^{-1}" from the abstract, main text, and conclusions. Instead, we now present our results as observational evidence for coordinated chemical and optical changes during plume processing.

3. Mathematical coupling between central variables.

Several of the principal relationships involve variables that are not mathematically independent.

Most importantly, the correlation between ΔOx/ΔBC and ΔOA/ΔBC uses the same ΔBC denominator in both variables. Variation in BC can therefore induce or strengthen correlations between these ratios. The same issue remains when the data are binned according to ΔOx/ΔBC and changes in ΔOA/ΔBC are subsequently examined. Please assess whether the relationship remains when the influence of the common BC denominator is accounted for.

Likewise, photochemical age is calculated from C8 aromatics/benzene while C8 aromatics/toluene is subsequently used as another ageing indicator and as a separate PCA input variable. These two metrics share the same C8 aromatic measurement and therefore do not constitute independent evidence.

I recommend demonstrating that the main relationships remain robust using analyses that avoid these shared variables, for example by examining the relationship between the underlying ΔOx and ΔOA enhancements while treating ΔBC as a covariate, or using an equivalent partial-correlation.

This issue is especially important because the observed (R=0.71) relationship between O_X/BC and OA/BC is subsequently used as evidence linking oxidation and OA production.

Response:

We thank the reviewer for raising this point regarding potential mathematical coupling when both normalized ratios share a common denominator (rBC) and for recommending the necessary types of analysis.

To address this point and evaluate whether the relationship between OA/rBC accumulation and O_x/rBC is influenced by mathematical coupling from shared rBC normalization, we tested the unnormalized variables (OA, O_x, and rBC) across individual points (N = 93) using partial correlation and multiple linear regression with rBC treated as a covariate.

First, controlling for the influence of rBC yields a statistically significant positive partial correlation between O_x and OA (partial r = 0.58, $p < 0.001$, N=93). Furthermore, a multiple-variable model ($OA = \beta_0 + \beta_1 \cdot O_x + \beta_2 \cdot BC$) accounted for half of the overall OA variance ($R^2 = 0.51$), highlighting the link between atmospheric oxidation processes and organic aerosol enhancement within the plume. The positive partial regression coefficient for O_x ($\beta_1 = 0.049$, $p < 0.001$) when controlling for rBC is significant, suggesting that each incremental unit of photochemical processing enhances OA relative to rBC within the plume. The results of additional statistical Pearson test – such as the comparison of the relative increase in OA to rBC as a function of OH-equivalent photochemical age ($p < 0.05$) and the use of different normalization variables such as CO ($p < 0.05$) – also support this (Table S3).

We have included these tests in the supporting information (Figure S9) and discussed the results in section 3.4 (lines 446-456).

We acknowledge the reviewer's observation that OH-equivalent photchemical age (derived from C8 aromatics/benzene) and C8 aromatics/toluene share the same C8 aromatic measurement. The inclusion of both metrics in the initial PCA was intended to reflect varying photochemical timescales due to differences in OH reactivity (k_{OH}) across aromatic species, rather than to introduce artificial variable redundancy.

To verify that this variable sharing does not distort the multivariate PCA structure or artificially inflate PC1, we performed a PCA sensitivity test comparing the base set (4 variables, N = 28) with a reduced model retaining only O_x/BC, OA/BC and OH-equivalent photochemical age. In this 3-

variable configuration (N = 33 vs 28), which avoids unnecessary data, PC1 continues to capture the dominant variance (56.24% with PC1+PC2 accounting for 86.94%, N = 28). Additionally, the resulting PC1 vector is virtually identical to the base set PC1 (Pearson correlation R = 0.96, $p < 0.001$, N = 28). This alignment suggested that the multivariate axis reflects OH-equivalent photochemical aging in the Paris outflow rather than an artifact of shared C8 aromatic measurements. Therefore, in the revised manuscript (section 3.4, lines 504-512) and supporting information (Figure S10) we adopted the 3-variable PCA as the primary framework, retaining all valid observations without compromising the multivariate structure.

4. Statistical independence and effective sample size

Table S1 is useful in showing the measurement time resolutions and raw data counts per flight. However, it also reinforces the need to address statistical independence.

The underlying instruments generate measurements at 1-90 s resolution and Table S1 reports thousands of observations per flight. Consecutive measurements within a single plume crossing are unlikely to represent independent atmospheric samples because of temporal and spatial autocorrelation.

Please state clearly what temporal matching/averaging was applied before combining measurements from instruments operating at different time resolutions, and report the actual number of observations entering each correlation, PCA, and significance test.

More importantly, please explain how the effective number of independent observations was determined. Treating every high-frequency point as statistically independent may overestimate the nominal sample size and produce very small p-values.

This applies particularly to the repeated ($p<0.001$) results in Figs. 3-4 and the Kruskal- Wallis/Dunn analyses. Analysis at the plume-intercept level, flight level, or using an appropriate autocorrelation-adjusted effective sample size would provide stronger evidence.

Table S2 should also report the Kruskal-Wallis result as, for example, ($p<0.001$), rather than "p-value = 0.00000".

Response:

We appreciate the suggestion of the reviewer of including some additional test to provide stronger evidence to the results in Figs 3-4 and the Krustal-Wallins/Dunn analysis.

To combine measurements from instruments operating at different high-frequency time resolutions (ranging from 1 s to 90 s), all fast response data were aligned to the sampling time grid of the PTR-TOF-MS. Specifically, for each PTR-TOF-MS timestamp t_{VOC}, a symmetric moving time window with a tolerance of ±90 s was applied. High-frequency measurements falling within this window—including 1-s SP2 (rBC), 10-s CAPS (extinction and scattering at 450 and 630 nm), 15-20 s AMS (non-refractory PM_1 species), 1-s/5-s gas monitors (O_3, NO_x, CO), and 1-s meteorological parameters—were averaged to generate a synchronized merged dataset. If no high-frequency observations were present within the tolerance window, NaN values were assigned to preserve temporal integrity without interpolation. We have stated this in the revised manuscript (section 2.1, lines 176-181, section 2.4 lines 261-264) and we have thoroughly updated our data matching, sample size reporting, and statistical testing framework across the manuscript and supporting information (Table S3).

Following the reviewer's suggestion, we performed an aggregation of our dataset to the discrete plume-intercept level for all the variables reported in the text (N = independent plume transects across the campaign flight, Table S3). At this plume-intercept level, the relationships of OH-equivalent photochemical age with O_x/BC (Pearson coefficient R of 0.88, $p < 0.001$, N = 15) and toluene mixing ratio (Pearson coefficient R of -0.82, $p < 0.05$, N = 16) remained statistically significant, indicating that the oxidation evolution presented in Figure 4 (before Figure 3) is not an artifact of high-frequency autocorrelation.

For the Kruskal-Wallis/Dunn analysis, to account for temporal autocorrelation within individual points without discarding the fine-scale spatial gradients captured by our binning framework, we calculated the effective sample size (N_{eff}) using the lag-1 autocorrelation coefficient (ρ_1) for the distributions presented in Table S5 as follows:

$$N_{eff} = N \cdot [(1- \rho_1) / (1 + \rho_1)]$$

Results show low intra-bin lag-1 autocorrelation (ρ_1 values ranging from 0.02 to 0.39), yielding target effective sample sizes across the oxidation bins of 11, 8, and 18 for bins 1, 2, and 3, respectively. After applying a systematic sub-sampling strategy to match these effective sample sizes and ensure observation independence, the analysis no longer finds significant differences between the groups after correcting for the issue of data dependency. Although subtle variations may exist, the method used does not allow them to be detected with confidence due to the reduction in independent data points. We have incorporated this test in the revised manuscript (section 3.4, lines 495-502) and the supplementary information (Text S4, Table S6). Additionally, the new test and we have corrected all instances of unscientific p-value notation (such as "p = 0.00000") across the supplementary information, and the main text, replacing them with standard inequalities (e.g., $p < 0.001$ or exact scientific notation).

5. Relative BrC contribution versus actual production of new absorbing material

The manuscript should distinguish more clearly between an increase in the relative BrC contribution and actual production of additional absorbing material.

For example, the manuscript notes that flights with lower BC concentrations exhibit larger fractional BrC contributions. However, because BrC is expressed as a fraction of total absorption, a reduction in BC can mathematically increase the BrC fraction even if absolute BrC absorption remains unchanged.

Similarly, increasing AAE demonstrates increasingly wavelength-dependent absorption but does not by itself demonstrate formation of additional BrC.

If possible, I encourage the authors to examine how the inferred BrC absorption coefficient changes with photochemical age both in absolute terms and after normalisation to an independent dilution tracer (e.g. CO). This would help distinguish the formation of additional absorbing material from changes in the relative BC/BrC contribution caused by plume dilution or varying BC abundance.

The same distinction applies to OA. An increase in (ΔOA/ΔBC) is not equivalent to an increase in OA mass; it represents an increase in OA relative to BC and requires assumptions regarding the behaviour of BC before it can be interpreted quantitatively as SOA production.

Response:

We thank the reviewer for urging a clearer distinction between BrC and actual production of

absorbing material.

Following the reviewer's suggestion, we examined the evolution of BrC absorption coefficient—both in absolute terms and normalized by an independent dilution tracer (CO)—as a function of OH-equivalent photochemical age. Similarly, the relationship between the BrC absorption coefficient and OH-equivalent photochemical age exhibited considerable data scatter without a simple dominant linear trend likely associated to the combined influence of competing simultaneous atmospheric mechanisms within the sampled Paris outflow (e.g., plume dilution, varying BC abundance, secondary production versus photobleaching). Therefore, in the revised manuscript we acknowledge the complex interplay of competing processes within the sampled plume and have specifically addressed the relative BrC contribution to light absorption in section 3.2. Furthermore, we have refined our interpretation in the manuscript and removed strong causal conclusions based on relative changes, such as "SOA production" and "production of new absorbing material within the urban plume".

6. Association versus causal attribution to secondary BrC/SOA formation

The combined changes in AAE, aromatic VOC depletion, OA/BC, and oxidation state provides compelling evidence for coordinated chemical and optical changes during plume processing.

However, the manuscript frequently goes beyond this observational result and states that SOA formation "drives" the browning or that secondary BrC production has been demonstrated.

The present aircraft measurements do not directly identify the molecular species responsible for enhanced absorption. The discussion of nitro-aromatics provides a potential mechanism based on previous laboratory and field work, but CHON compounds or aromatic VOC depletion do not directly establish that nitro-aromatic chromophores account for the observed AAE increase.
Similarly, newly formed OA is not necessarily light absorbing.

I therefore recommend maintaining a clear distinction between:

a. the directly observed increase in wavelength-dependent absorption;
b. the BrC contribution inferred using the optical attribution assumptions; and
c. the proposed mechanism of secondary light-absorbing OA formation.

Wording such as "drives", "demonstrates", "production rate", and "producing new absorbing material" should be moderated unless directly supported by the measurements. A formulation such as "the observations are consistent with an increasing contribution from secondary light-absorbing OA during photo-chemical processing" would remain scientifically strong while better reflecting the evidence.

Response:

We thank the reviewer for this thoughtful recommendation, we agree that a clear distinction must be maintained between directly observed optical properties, inferred BrC contributions, and proposed chemical mechanisms, avoiding overstatements regarding causal attribution.

In response to this comment, we have thoroughly revised the manuscript to moderate deterministic terminology (such as "drives", "demonstrates", "production rate", and "producing new absorbing material") throughout the text, abstract, and conclusions.

Specifically, we have updated the text and structure of the manuscript to ensure that:

1. The directly observed increase in wavelength-dependent absorption is presented as an observation in section 3.1.
2. The BrC contribution and BrC absorption derived through optical attribution assumptions are clearly framed as inferred estimates in section 3.2.
3. The discussion concerning nitro-aromatics, CHON species, and SOA formation is explicitly contextualized in section 3.4 as a proposed mechanism utilizing previous literature, rather than a direct molecular identification.

We believe these revisions strengthen the clarity of the manuscript, aligning with the reviewer's recommendations.

7. Plume attribution and interpretation of transport/ageing

The manuscript would benefit from a clearer distinction between identifying air masses influenced by Paris and demonstrating temporal transformation of the same plume.

The percentile-based plume definition (>75th percentile BC) is operationally reasonable but guarantees a high-BC subset for every flight regardless of absolute Paris influence. In addition, (ΔBC/ΔCO) is not a fully independent validation because BC is used both in the initial selection and the validation metric.

I suggest making greater quantitative use of the WRF-CHIMERE Paris-source attribution across all flights and showing that the selected plume and background populations differ systematically in simulated Paris influence.

The HYSPLIT analysis also requires further clarification. Please specify the forward-trajectory starting height(s), explain what is meant by "arrival heights corresponding to the aircraft altitude", and define the horizontal, vertical, and temporal criteria used to determine an aircraft-trajectory intersection. HYSPLIT is described as a complementary check that the sampled air masses were transported from Paris, but the corresponding trajectory results should also be shown, potentially in the Supplement. Please also explain whether the aircraft data were screened for possible contamination from the ATR42 exhaust or previously emitted wake, particularly during turns and repeated low-altitude transects. Figure 1 contains repeated BC, CO, and optical enhancements. The spatial agreement with WRF-CHIMERE provides evidence for an ambient Paris plume, but the manuscript should nevertheless describe the self-contamination screening procedure and explain the yellow excluded periods. Finally, wording such as "tracking plume evolution" and "transport time" should reflect that the analysis represents an ensemble of Paris-influenced plume intercepts from different flights and meteorological conditions rather than continuous tracking of a single parcel.

Response:

To ensure that the subsets selected using the percentile-based approach (>75th percentile of rBC) reflect Paris urban influence rather than arbitrary local variations, we quantified the WRF-CHIMERE Paris-source attribution across all flights. The selected in-plume population exhibits a systematically high simulated Paris influence with a campaign-average ratio of 21.23%, whereas the out-of-plume (background) population consistently shows a negligible Paris contribution averaging 2.92%, confirming that both populations differ.

As requested by the reviewer, we now provide a quantitative comparison using the WRF-CHIMERE Paris-to-regional BC ratio across all flights (Table S2). Additionally, we have expanded the text in section 2.4 (lines 254-259) to describe that the selected in-plume and out-of-plume

populations exhibit systematic and statistically significant differences in simulated Paris source contributions.

Regarding the BC\CO ratio we removed the text to clarify that the emission ratio itself serves to characterize the combustion source mix (fossil fuel vs. other sources) rather than acting as a fully independent meteorological validation metric.

We have updated section 2.2 (lines 196-203) to clarify the HYSPLIT configuration. Additionally, as suggested we now included representative HYSPLIT forward-trajectory results and maps in the supporting information (Figure S1) taken from our previous work.

To prevent artifacts from aircraft exhaust or previously emitted wakes (particularly during sharp turns, spirals, and low-altitude repeated transects), all in situ time series were screened. Data points recorded during aircraft maneuvers exceeding quantitative attitude thresholds—specifically absolute roll angles (roll > 6 °) or turn rates (> 1.5 $°s^{-1}$)—were flagged and removed from the analysis. In Figure 1b, the yellow-shaded areas initially marked take-off, landing, and vertical profiling maneuvers. To fully address the reviewer's point, we have updated the text and added a supplementary figure (Figure S2) that specifically highlights the additional flight intervals filtered out due to sharp turns and potential self-exhaust recirculation during horizontal transects. We now described the screening procedure in section 2.4 (lines 244-247) and we clarified the excluded periods in the caption of Figure 1 of the revised manuscript.

Finally, as suggested by the reviewer, we have state in revised the manuscript text (introduction, lines 105-106) that the plume analysis in this study represents an ensemble of Paris-influenced plume intercepts from different flights and meteorological conditions.

Minor comments

1. Introduction, lines 43-45: Please separate direct atmospheric absorption, snow/ice albedo effects following deposition, and cloud/boundary-layer adjustments rather than grouping these mechanisms together.
The changes have been made (lines 44-46).

2. Introduction, lines 63-66: Please broaden the discussion of secondary BrC formation beyond gas-phase VOC oxidation and clarify what is meant by “BrC lifetime”. In many cases, this is more accurately a wavelength-dependent absorption-decay or photobleaching timescale.
In the revised version of the manuscript, we discussed the formation of brown carbon beyond the gas phase. Specifically, reviewer #3 suggested discussing the formation of BrC in the aqueous phase (introduction, lines 64-65, lines 82-84, section 3.4 lines 475-485, section 4 lines 541-548). Furthermore, we specified in line 67 that the term “BrC lifetime” refers to photo-bleaching timescale.

3. Introduction, lines ~90-102: The novelty relative to Yu et al. (2026b) should be made more explicit, since that study already reported SOA formation, increasing AAE, and enhanced BrC contribution using substantially overlapping ACROSS airborne measurements. Please state clearly which analyses and conclusions areunique to the present manuscript.
In the revised version of the manuscript, we state clearly (lines 100 -113) which analysis are unique

to the manuscript of Yu et al. (2026b).

4. End of Introduction: The objective to provide "recommendations for incorporating BrC dynamics into climate models" appears somewhat stronger than what is directly evaluated. The study does not perform a model sensitivity experiment. "Discuss implications for the representation of BrC ageing in atmospheric and climate models" may more accurately describe the scope.
In the revised we have removed objective 3. Instead, we have reworded the objectives to emphasize what is evaluated in the manuscript.

5. Section 2.1: Table S1 provides the individual instrument time resolutions. Please explain how these datasets were temporally matched before ratios and multivariate statistics were calculated.
We have stated this in the revised manuscript section 2.1 (lines 176-181).

6. Section 2.1, lines ~127-134: I suggest using the term refractory black carbon (rBC) for the SP2-derived mass concentration. This would distinguish the SP2 measurement from modelled BC and optically inferred BC absorption used elsewhere in the manuscript.
This comment was also suggested by reviewer #1, thus we have used the acronym rBC to refer to the refractory black carbon measured by the SP2 thorough the revised manuscript.

7. Section 2.2: Please briefly explain how Paris-derived BC was tagged or separated from regional/background BC in WRF-CHIMERE.
We have reworded the text in the revised manuscript to clarify how BC_{Paris} and $BC_{Regional}$ were identified (line 193-194).

8. Lines 190-199: The BrC attribution assumes, assigns absorption at 630 nm entirely to BC, extrapolates this contribution to 450 nm, and attributes the residual absorption to BrC. A close to unity AAEBC is a reasonable nominal assumption, but BC mixing state and coating can modify its wavelength dependence, particularly during atmospheric ageing. I therefore suggest a sensitivity analysis using a reasonable range of values to indicate how strongly the inferred BrC fraction depends on this assumption. The assumption of negligible BrC absorption at 630 nm should also be acknowledged.
In the revised version of the manuscript, as suggested by the Reviewer #2 we acknowledge the influence of BC mixing state and coating in the AAE_{aer}. Additionally, we included a sensitivity analysis in which we varied the AAE_{BC}. The values were obtained from ambient collected datasets in Europe (Rovira et al., 2026). The sensitivity analysis showed that the estimated BrC contribution varies ~4% across the entire range, suggesting that the choice in the AAE_{BC} value have a minor impact in our apportionment results. Finally, we acknowledged the hypothesis that BrC absorption at 630 nm is negligible. These revisions have been made to the lines 224-232 of the revised manuscript.

9. Lines 120-126 and 185-190; Table S1: Please provide or propagate the uncertainty associated with the A2S2-derived absorption through the AAE and BrC calculations, since absorption is obtained from extinction minus scattering before the subsequent spectral attribution. Table S1 also indicates missing/problematic A2S2 450 nm measurements for some flights. Please clarify which flights are included in the AAE/BrC analysis and whether the missing optical data systematically alter the range of conditions represented in this part of the study.
As we indicated in our responses to reviewer #1, in the revised manuscript we discussed (section 3.1, lines 315-219) the propagation errors, acknowledged instrument's sensitivity limit. Furthermore, we clarified that specific multi-instrument comparison or chemical ratios involving collocated variables. Consequently, each specific statistical subset strictly relies on mutually valid, quality-assured data points, ensuring that the integrity and representativeness of the findings remain unaffected (section 2.4, lines 261-264).

10. Sections 2.3-2.4: Please define explicitly how the background concentrations used for each Δ quantity were determined, particularly for observations already classified as "background" using the lower BC quartile.
We thank the reviewer for pointing out this source of confusion. As we indicated in our responses to reviewer #1, we have removed the sentence that previously appeared on line 206, which defined Δ and caused ambiguity. We also revised relevant figures and their captions to ensure consistent terminology.

11. Section 2.6: PCA describes covariance rather than causal "drivers"; please revise the corresponding wording.
The words have been reviewed in section 2.6 (line 311).

12. LOWESS analysis / Fig. S6: The apparent plateau at advanced ageing should remain explicitly tentative. The data are sparse in this regime and LOWESS behaviour at the edge of the dataset can depend strongly on the smoothing parameter.
As we indicated in our responses to reviewer #3, in the revised version of the manuscript, we have reworded the discussion (section 3.4, lines 514-518), acknowledging the dispersion of the data in Figure S10a.

13. Section 3.1: Higher SSA supports a more scattering-dominated aerosol but does not by itself demonstrate that the aerosol is more OA-rich.
We agree that a high SSA value does no, by itself demonstrate that the aerosol is richer in organic aerosol particles. We have reworded the correspondent sentences in sections 3.1 (lines 335-337) and 3.2 (lines 365-366).

14. Section 3.1: A weak relationship between (ΔBC/ΔCO) and BrC absorption does not by itself establish a secondary origin for BrC.
We have reworded the related sentences in sections 3.1 (lines 323-329).

15. Section 3.2, lines ~305-310: The quantity described as an "oxidant production rate" is more accurately the slope of BC-normalised OX enhancement against inferred photochemical age. Please revise the terminology and retain the BC normalisation in the reported units, including in the Conclusions.
We have revised the related sentences and the reported units in sections 3.3 (lines 393-395) and section 4 (522-524).

16. Section 3.2, line 308: The statement that oxidant production remains "highly efficient" is stronger than supported by the metric. The data indicate continued Ox enhancement relative to BC but do not quantify efficiency.
We have revised the related statement in the revised manuscript (section 3.3, lines 393-395).

17. Section 3.2, line 305 and Section 3.3, lines ~340-342: Figure 3 appears to span photochemical ages up to approximately 12.5 h, whereas the AAE change and reported 0.25 h slope are based on the 2-8 h interval. Please clarify why the optical analysis is restricted to this shorter age range (e.g. due to availability of temporally matched optical and VOC measurements) and state this distinction explicitly when discussing the 6 h processing interval in the Abstract and Conclusions. In addition, "photochemical lifetime" at line 305 would be better replaced by "photochemical age" or "age range".
In the new version of the manuscript, Figure 4 (before Fig 3) has been updated. Instead of showing the distribution of individual data points within the plume, the figure shows the data grouped by transects, with their respective statistics covering the same time intervals as Figure 5 (before Fig 4)

and Table S4. Section 2.4 (lines 261–264) provides details on data availability based on time-synchronized optical and chemical measurements.

18. Section 3.2: Temperature and RH correlations should be interpreted more cautiously. These parameters likely covary with time of day, photochemical age, solar radiation, boundary-layer evolution, and flight-to-flight variability.
We have revised the related paragraph in section 3.3 (lines 419-430). We recommend caution when interpreting these relationships, as a more in-depth analysis is required to distinguish the independent contributions.

19. Figure 4 caption: A decrease in C8 aromatics/toluene should not be described as "depletion of toluene". It represents preferential depletion of the more reactive C8 aromatics relative to toluene.
We have revised the related statements in the revised manuscript (abstract, section 3.3 lines 404-408, section 4 lines 524-525).

20. Section 3.3 / Table S2: The statistical analysis demonstrates a significant difference between the lowest and highest OX /BC groups, whereas adjacent groups are not statistically different. I therefore suggest replacing "statistical progression" with wording describing an endpoint difference across the oxidation range.
After conducting the rigorous tests recommended in the Major Comments section (point 4), we have reworded the paragraph. In this new paragraph (section 3.4, lines 493-495), we described the endpoint difference between the oxidation states.

21. PCA / Table S3: OA/BC loads strongly on PC2 (0.893), whereas the primary photochemical-processing metrics dominate PC1. Please discuss what this indicates about OA variability rather than presenting OA production as completely coincident with the principal ageing axis.
In the revised version of the manuscript, we have reworded the discussion (section 3.4, lines 505-512) on this point, emphasizing the variability of OA, rather than presenting OA production as something that fully aligns with the main axis of aging.

22. Table S4: The final sentence describing the different AAE "sensitivities" appears to contain copy/paste errors in the derivative labels. Please check the labels for OA/BC, Ox /BC, and C8 aromatics/toluene carefully.
We have revised the related copy/paste errors in the derivative labels.

23. Table S4 / main text: Please clarify the units/scaling of C8 aromatics/toluene. The main text refers to ratios of approximately 0.2-0.3, whereas Table S4 reports values of 351.5, 115.7, and 70.9. If these values are scaled, this should be state explicitly.
We have revised the report values of toluene normalised by rBC (Figure 4) and in absolute terms (Figure 5) within the main text and reported in Table S4.

24. Conclusions, line 411: The statement that photochemical ageing "leads to higher SSA values" does not appear to be directly demonstrated by the ageing analysis. Please either show this relationship or soften/remove the statement.
In the revised version of the document, we removed that statement.

25. Conclusions/model implications: OA/BC and SSA are useful observational covariates but are not yet demonstrated to be transferable model parameterisations of BrC ageing. I suggest describing them as observational relationships that may inform future model development.
In the revised version of the document, we reworded the conclusions and we describe our results as observational relationships that may inform future model development.

Overall recommendation

The manuscript presents a valuable airborne dataset and identifies combined changes in aromatic VOC depletion, OA relative to BC, oxidation state, and wavelength-dependent aerosol absorption in air masses influenced by emissions from the Paris metropolitan area. I believe the study has clear merit and could make a valuable contribution to ACP after the points above are addressed.

However, the quantitative BrC attribution and photochemical-age estimation require additional sensitivity analysis, the derivation of the reported rate of 0.25 h^{-1} in AAE should be revisited using temporally matched observations and appropriate uncertainty, some key correlations should be reassessed because the variables share common measurements and the data are autocorrelated, and the causal interpretation should be brought into closer alignment with what is directly constrained by the measurements.

Addressing these issues would, in my view, substantially strengthen both the scientific interpretation and the broader relevance of the manuscript.

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