Response to Reviewers' comments

We sincerely appreciate the thoughtful and constructive feedback from editors and all reviewers, which has significantly improved the manuscript. We have carefully revised the manuscript in response to their suggestions. Below, we provide a point-by-point response, with our replies highlighted in blue.

RC1

This manuscript conducts continuous eight-year (2016–2023) observations of submicron particulate matter (PM1) chemical composition in an urban background environment in Dublin; this dataset is solid and possesses significant scientific value for evaluating the long-term effectiveness of solid fuel emission reduction policies in Europe. The paper introduces a supervised machine learning model to accurately distinguish between the local formation and transboundary transport sources of oxygenated organic aerosol (OOA), which holds significant innovative merit in terms of methodology. However, several profound scientific loopholes remain in the manuscript regarding the representativeness of certain typical case studies, the analytical methods for long-term trends, and the socio-economic attribution arguments for key conclusions. These issues must be carefully revised or supplemented with additional analysis before the manuscript can be considered for publication

1. Page 5, Line 150-155: The text mentions that the equivalent black carbon (eBC) data from 2018 to 2020 were missing due to instrument failure, and the missing data were estimated and reconstructed by multiplying the existing organic aerosol (OA) data by a strongly correlated ratio. However, the data presented in Table S1 show only partial annual eBC/OA correlations and ratios. Given that the paper itself has demonstrated significant seasonal differences in aerosol source composition, the rationality of using a single annual average ratio to reconstruct seasonal eBC concentrations needs to be clearly justified. It is recommended that the authors additionally provide seasonal eBC/OA ratios and their coefficients of variation (CV), and re-evaluate the uncertainty that this reconstruction method introduces to the overall trend analysis.

We thank the reviewer for this important suggestion. We agree that the possible seasonal variability in the eBC/OA ratio needs to be evaluated before using this ratio to reconstruct the missing eBC data. We have therefore re-examined the eBC/OA relationship using all periods with valid eBC measurements, including 2016-2017, 2021, 2022, and 2023.

First, we calculated the monthly mean eBC/OA mass ratios from the available measured data. As shown in Fig. R1a, the eBC/OA ratio indeed exhibits clear seasonal variability, with lower values in summer and higher values during colder months. The coefficient of variation of the monthly average eBC/OA ratios was 22 % (CV = standard deviation / mean × 100 %), indicating moderate seasonal variability in the eBC/OA relationship.

To evaluate whether this variability affects the eBC reconstruction, we compared two reconstruction approaches: (1) the original method using a single annual average eBC/OA ratio of 0.24, and (2) a revised method using monthly average eBC/OA ratios. The performance of the two methods was assessed using the periods with measured eBC data, by comparing reconstructed eBC with observed eBC. As shown in Figs. R1b-d, the eBC values reconstructed using the annual-ratio and monthly-ratio methods are generally very similar, while the monthly-ratio method shows slightly better agreement with the measured eBC. This indicates that monthly ratios better capture the seasonal variability in the eBC/OA relationship, but the overall reconstruction is not strongly sensitive to the choice of annual or monthly ratios. The

reconstruction performance was relatively weaker for 2016-2017, likely due to the lower eBC-OA correlation and higher eBC/OA ratio during this period compared with 2021-2023, suggesting potential interannual uncertainty in the eBC reconstruction.

We then assessed if this difference affects the long-term eBC trend analysis. The resulting long-term eBC trends were nearly identical between the two reconstruction methods. The Mann-Kendall trend significance remained unchanged, and the fitted slope for annual mean eBC was −0.07 μg m^{-3} yr^{-1} for both methods. For the yearly maximum hourly eBC concentration, the fitted slope changed only from -12.6 to -11.7 μg m^{-3} yr^{-1} when monthly ratios were used, corresponding to a relative difference of less than 10 %.

These results indicate that although the eBC/OA ratio shows measurable monthly variability, the use of a single annual eBC/OA ratio has only a limited influence on the overall long-term eBC trend conclusions. We have now added a separate section in the Supplement (S1.1) summarizing the details of the eBC data reconstruction and the associated sensitivity tests. We have also added a clearer justification for using the annual eBC/OA ratio to reconstruct the missing eBC data for 2018-2020 in Sections 2.1 and 3.2.1 of the revised manuscript.

Table R1. Long-term trend results for annual eBC from 2016 to 2023 based on two different reconstruction methods for the missing eBC data in 2018-2020: using a single annual eBC/OA ratio and using monthly average eBC/OA ratios, respectively. The Mann-Kendall test results are shown together with linear regression slopes and 95% confidence intervals (95% CI) calculated from annual average eBC concentrations.

Methods	P-value	Slope (95% CI) (μg m^{-3} $year^{-1}$)	Slope of annual hourly maxima* (95% CI) (μg m^{-3} $year^{-1}$)
Annual eBC/OA	<0.05	-0.07 (-0.10 – -0.04)	-12.6 (-19.1 – -6.0)
Monthly eBC/OA	<0.05	-0.07 (-0.09 – -0.04)	-11.7 (-20.8 – -2.7)

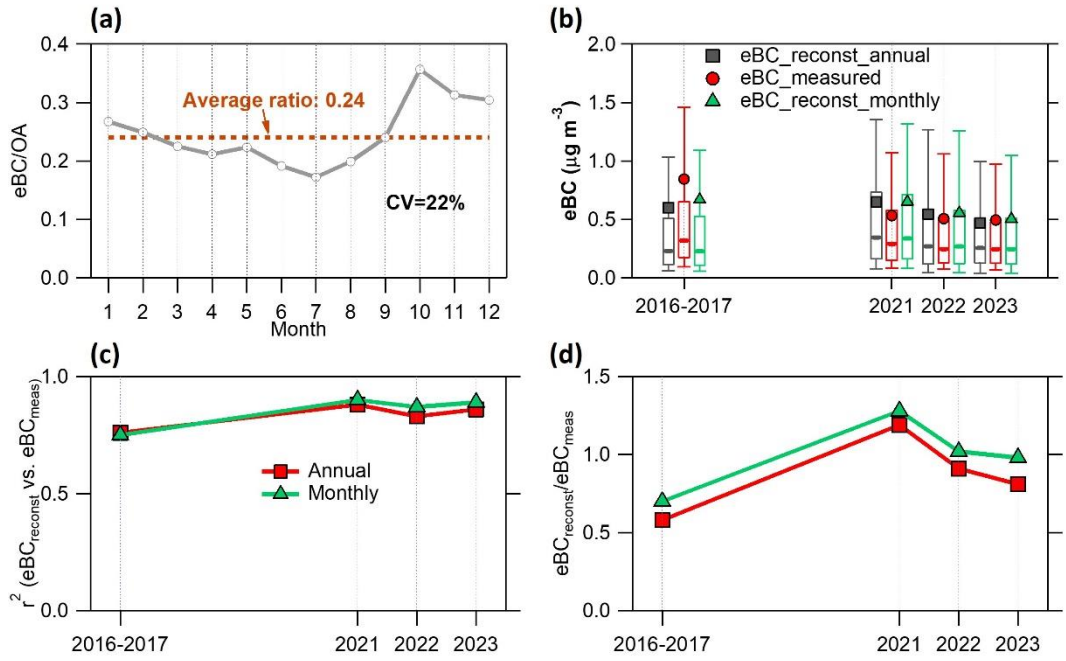

Figure R1. Monthly mean eBC/OA mass ratios calculated from all available eBC measurements in 2016-2017, 2021, 2022, and 2023. The annual mean eBC/OA ratio and the corresponding coefficient of variation (CV = standard deviation / mean × 100 %) of the monthly ratios are also shown. Panels b-d compare the reconstructed eBC with the measured eBC using either a single annual eBC/OA ratio or monthly eBC/OA ratios, showing box plots of the overall distributions, fitted slopes and correlation coefficients (r^2), respectively.

The revised text is provided below:

Section 2.1

"*To allow for more continuous and comparable long-term trend analysis, eBC mass concentrations during this period were estimated from concurrent OA mass concentrations using a single annual average eBC/OA ratio (0.24) derived from periods with valid eBC measurements. This approach is supported by the typically strong correlation and relatively stable mass ratios between eBC and OA (see Table S1), due to their common sources. The uncertainty associated with the eBC reconstruction, including the possible influence of seasonal variability in the eBC/OA ratio and the use of reconstructed data in the long-term trend analysis, was evaluated through sensitivity tests described in the Supplement (S1.1). Overall, the potential uncertainty introduced by reconstructing eBC from OA is expected to be largely reduced when focusing on monthly or annual scales, making this approximation suitable for long-term analysis. However, compared with components based entirely on direct measurements, the long-term eBC trend, particularly its fitted magnitude, remains subject to greater uncertainty.*"

Section 3.2.1

"*… The robustness of the long-term eBC trend, despite the reconstruction of missing eBC data for 2018-2020 using a single annual eBC/OA ratio, was validated by a sensitivity test using monthly eBC/OA ratios, which produced the same annual mean eBC trend slope and unchanged Mann-Kendall significance (see Fig. S10 and Table S3 for details). An additional sensitivity test excluding the reconstructed years also produced a comparable decreasing trend, indicating that the eBC trend was not significantly biased by the reconstructed data.*"

2. Page 6-7, Line 199-216: To elucidate the characteristics of two typical pollution events, the authors selected only one case study each from March 2021 for analysis in Section 3.1, indicating a local combustion source and a transport source, respectively. However, it is questionable whether using a brief peak triggered by an occasional cold wave and lasting only a few hours to represent a typical, multi-day winter persistent heating pollution event with high background levels is scientifically rigorous. At the same time, this study spans an eight-year period during which solid fuel regulatory policies in Ireland were intensively tightened. Existing studies have shown significant differences in the carbonaceous components emitted by different solid fuels; therefore, using the chemical composition compounding ratios from a single 2021 case study cannot reflect the statistical characteristics of similar events prior to (2016–2018) and following (2023) the evolution of these policies. The authors should explain the generalizability of this case study, or provide a multi-sample composite analysis or statistical average characteristics across different policy stages.

We would first like to clarify the intended purpose of the two selected cases shown in Fig. 1. These cases were not used to derive statistically representative chemical composition ratios for all pollution events, but to illustrate the contrasting temporal and chemical characteristics

of two frequently observed pollution types in Dublin. Based on the long-term observations, local domestic heating pollution in Dublin is typically characterized by rapid and intense PM_1 increases during heating hours (generally from evening to midnight) under cold and stagnant weather conditions, followed by a relatively fast decrease when emissions weaken or dispersion improves. Therefore, the short duration of the local case in Fig. 1 is not an exceptional feature of the selected episode, but reflects a common behaviour of local heating emission-driven pollution events in Dublin. In contrast, transboundary transport events are usually associated with persistent easterly air masses and tend to last for several days, resulting in less intense but sustained PM_1 enhancement.

However, we agree that the two examples alone are insufficient to demonstrate the statistical characteristics of similar events across the full eight-year period, and the representativeness of the two case studies should be more clearly demonstrated, particularly with changes in solid-fuel regulations. We have therefore added two complementary analysis to the revised manuscript and Supplement.

First, we classified all 157 polluted days during 2016-2023 (as shown in previous Fig. S8, now Fig. S11), defined as days with daily average $PM_1 > 15$ μg m^{-3}, according to their dominant pollution type. To provide a relatively objective, although still simplified, classification of polluted days, we used the OA composition as the primary criterion, together with NO_3 as an additional tracer for transboundary transport. Specifically, a polluted day was classified as local home heating dominated when local OA, defined as the sum of solid fuel OA, HOA and OOA_{local}, contributed more than 80 % of total OA mass. A polluted day was classified as transboundary dominated when OOA_{TBT} contributed more than 40 % of total OA and NO_3 contributed more than 20 % of total PM_1. Days that did not meet either criterion were classified as mixed events with occasional contributions from other sources. This classification was designed to identify the dominant pollution sources in a consistent way across the full dataset, rather than to provide an exact source attribution for each polluted day. The OA factors were used as the main basis because their local and transboundary contributions have been clearly resolved, while the inorganic species have more mixed origins and are therefore less suitable as primary classification indicators. For transboundary events, an additional NO_3 contribution threshold was applied as elevated NO_3 is a recurring feature of regional transport pollution in Dublin, using it together with OOA_{TBT} reduces the chance of classifying weak or ambiguous regional influence as a transboundary dominated event. Overall, this provides a statistical overview of the occurrence of different pollution regimes over the full observation period, rather than relying only on the two examples shown in Fig. 1. The results are summarized in Table R2, showing that 96 polluted days were local home heating dominated (61 %), 31 days were transboundary dominated (20 %), and 30 days were mixed/less typical events, including only 3 days showing less clear characteristics of either of the two main pollution processes. These 3 less-typical days were characterized by relatively high SO_4 and OOA contributions and longer-lasting but moderate PM_1 enhancements, possibly related to enhanced local secondary formation during summer.

Second, we added additional pollution episode examples from different years. For local-dominated pollution, we selected the major local-heating-driven episodes associated with the three highest daily PM_1 concentrations in each year, and if additional local emission dominated pollution events occurred continuously before or after the selected polluted day, these periods were also included to better show the common pollution characteristics. For transboundary transport dominated pollution, representative episodes with more pronounced PM_1 enhancement were selected in each year when available, based on daily

PM_1 > 20 μg m^{-3} and chemical signatures consistent with transboundary influence. In addition, one example each is also shown for the mixed events and the less typical events. These added examples, although with event-to-event variability, show that local emission dominated pollution repeatedly exhibits intense but relatively short-lived PM_1 enhancements dominated by eBC and local OA factors, while transboundary events are generally more persistent and characterized by enhanced NO_3, NH_4, and OOA_{TBT}. In addition, although the peak concentrations of the most extreme local pollution events decreased over time, consistent with the overall long-term trend, comparisons among events from different years show that the main characteristics of each pollution regime remained largely unchanged, for both types of pollution events.

To avoid possible misunderstanding and to further support the generality of the interpretation, we have added a clear clarification in Section 3.1 that the two cases are intended as illustrative examples, rather than as statistical representations of all pollution events during the study period as below:

"*…Figure 1 shows illustrative examples selected to demonstrate the characteristic temporal evolution and chemical signatures of the two typical pollution episodes frequently observed in Dublin.*"

In addition, a short summary of the classification results and added examples has been added after the two illustrative examples in Section 3.1, with the corresponding table and figure also included in the Supplement as Table S2 and Figs. S6-7.

Table R2. Summary of the number of polluted days dominated by local home-heating emissions, transboundary transport (TBT), or mixed influences in each year.

Year	Local-dominated	TBT-dominated	Mixed
2016-2017	19	7	5
2018	19	6	5
2019	18	0	6
2020	13	6	2
2021	10	7	3
2022	9	4	8 (3*)
2023	8	1	1

*Three of the eight mixed events showed less typical pollution characteristics, with a high SO_4 fraction.

3. Page 8, Figure 2 merges and averages the same-month data for all years from 2016 to 2023, but the paper simultaneously demonstrates a significant long-term downward trend for each component. This means there are systematic differences in the orders of magnitude between the same-month data of earlier years (2016–2018) and later years (2021–2023), and simple averaging produces an artificial result that does not represent any actual year. It is suggested to provide at least a comparison of seasonal curves between two sub-periods (the early and late stages) to evaluate whether the seasonal pattern itself evolves over time.

We would like to clarify that Fig. 2 was intended to summarize the general seasonal characteristics of PM_1 composition in Dublin based on a multi-year average, rather than the seasonal cycle of any specific year. We agree that interannual variability, particularly in orders of magnitude, is expected. However, the seasonal analysis in this study focuses mainly on the recurring seasonal behaviour of different PM_1 components and source-related processes.

At the same time, we acknowledge that, because many of the major PM_1 components show long-term decreases, the absolute concentrations, and thus the relative contributions, in the early and later periods may differ. It is therefore useful to test whether the seasonal patterns themselves changed over time. Following the reviewer's suggestion, we compared the seasonal curves between the early period (2016-2018) and the later period (2021-2023) after the concentrations decreased substantially. As shown in Fig. R2, the absolute concentrations were generally lower in the later period, consistent with the long-term decreasing trends, and some differences were observed in terms of absolute concentrations. For example, the monthly mean PM_1 concentration during the colder months decreased from 7.2-12.4 μg m^{-3} in 2016-2018 to 4.6-6.4 μg m^{-3} in 2021-2023. Some differences in the monthly patterns were also observed, for example, October showed a particularly high PM_1 concentration (8.0 μg m^{-3}) during 2016-2018, whereas the highest PM_1 levels during 2021-2023 occurred in March-April (6.6-7.5 μg m^{-3}). Despite these differences, the dominant seasonal features remained broadly consistent over the years. Colder months, especially January, November, and December, were generally characterized by enhanced PM_1 concentrations and stronger local heating contributions, with October acting as a transition month in some years. Spring, particularly March-April, remained the period most strongly affected by transboundary transport, as indicated by the highest relative contributions of NO_3 (> 20%), with this influence sometimes extending into May. In contrast, June-September consistently represented the cleanest period, with relatively low PM_1 concentration.

We have added this comparison to the Supplement as Section S1.2. We have also added a clearer clarification in Section 3.1 and included a short discussion after the long-term trend analysis in Section 3.2.1 in the revised manuscript, which are also cited below:

Section 3.1

"*Driven by the two aforementioned pollution types, or in some instances, their combination, PM_1 concentrations and chemical composition in Dublin show clear seasonal patterns. Here, monthly averages were calculated using available data from all years to provide a representative overview of the dominant seasonal variations in PM_1 concentration and composition in urban Dublin.*"

Section 3.2.1

"*…Besides, potential changes in PM_1 seasonal patterns associated with the long-term concentration trends were also evaluated and discussed in detail in Section S1.2. Overall, the comparison between 2016-2018 and 2021-2023 shows that the long-term decreases mainly affected the absolute concentration levels, while the dominant seasonal structure was preserved.*"

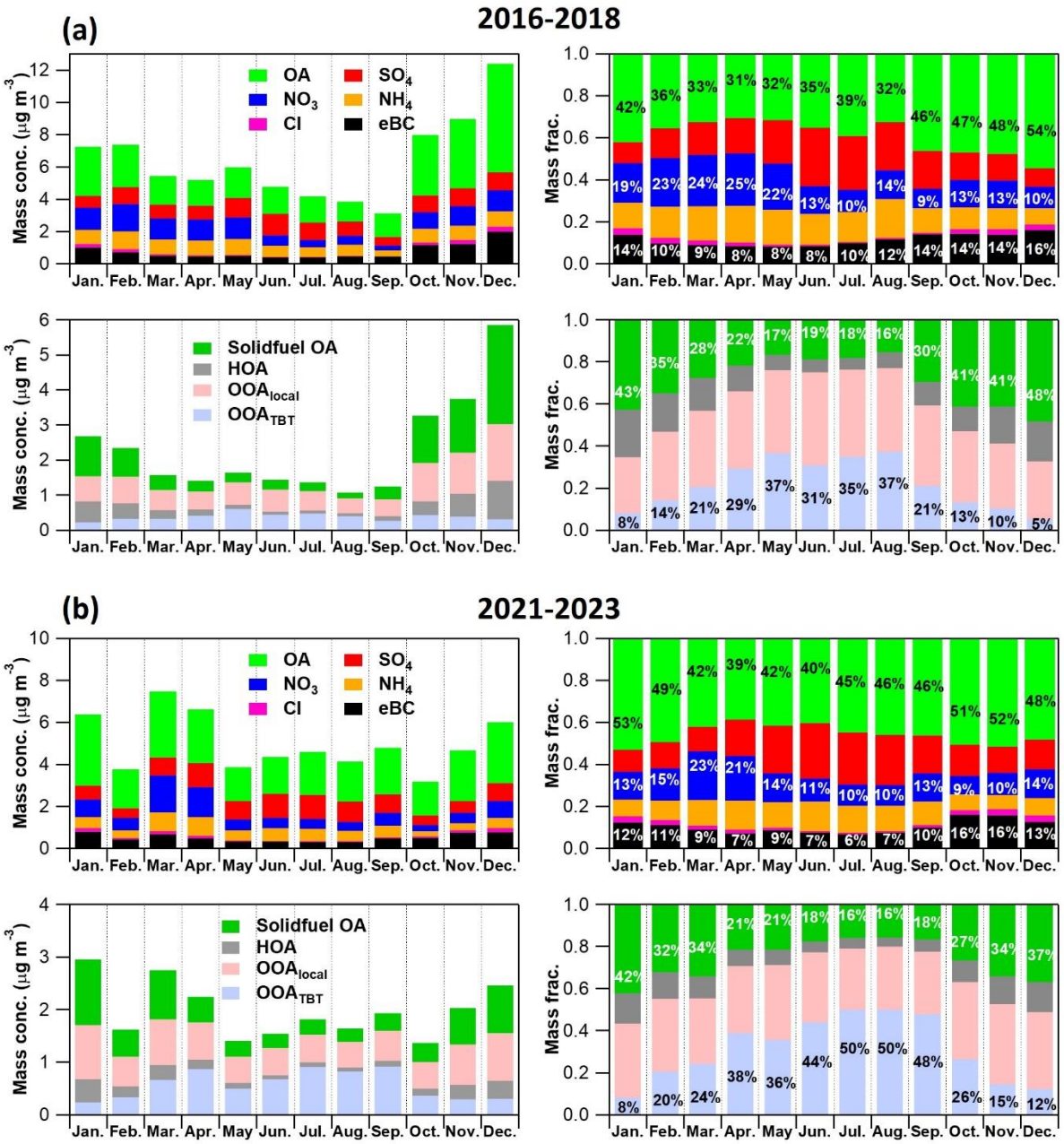

Figure R2. Comparison of monthly mean PM_1 and OA mass concentrations and chemical composition in Dublin between 2016-2018 and 2021-2023. The percentage contributions of major PM_1 and OA components are shown.

4. Page 11, Line 376-380; Page 15, Line 488; Page 16, Line 510–515: The authors attribute the significant decline in extreme peaks of solid fuel OA and eBC in 2021 to "increased public awareness before the official implementation of the solid fuel regulations in October 2022". The authors attribute the rebound of local pollutants (especially wood-burning OA) in 2023 to increased solid fuel use driven by the European energy crisis and the delayed manifestation of policy effectiveness. Similarly, the rebound of SO42– in 2022–2023 is attributed to the "use of low-cost but high-sulfur fuels". These inferences lack direct evidence, and some arguments present potential self-contradictions.

We thank the reviewer for pointing this out. We agree that the links between the observed changes in local pollutants and potential drivers such as public awareness, fuel-use changes during the European energy crisis, and the use of lower-cost fuels are interpretative hypotheses rather than directly demonstrated causal relationships. Since direct household fuel-use, fuel-sales, or behavioural data are not available in this study, these factors cannot be conclusively identified as the causes of the observed changes. We therefore agree that the original wording was too strong in places and could be interpreted as over-attribution.
In the revised manuscript, we have retained these discussions because they provide plausible contextual explanations for the timing of the observed changes, but we have revised the wording throughout to make the statements more cautious and to clearly acknowledge the need for further evidence. The revised text now reads:
Section 3.2.1:
"*The timing of the sharp decline in extreme local primary pollutants coincides with the period of intensified public awareness campaigns and preparation for the new solid fuel regulations before the official implementation in October 2022 (Department of Climate Energy and the Environment, 2021). This suggests that the public outreach efforts may have contributed to raising public awareness and reducing extreme pollution events. However, this interpretation needs further confirmation with more direct evidence, such as household fuel-use or behavioural data.*"
Section 3.2.2:
"*…possibly linked to shifts in fuel usage under the influences of the European energy crisis, such as potential increased use of lower-price fuels with higher sulfur contents (Fazelianov, 2023; Urbano et al., 2023). Such a shift may have occurred in some households despite the new solid fuel regulations and public awareness efforts, as economic pressures could still have influenced fuel choices. However, this interpretation needs to be further validated with direct fuel-use data in future studies.*"

5. Page 4, Line 113-129; Page 18, Line 563: Both the abstract and conclusion claim that this study is the "first long-term source apportionment study of PM1 pollution in Dublin". However, the text simultaneously cites multiple PM1 source apportionment studies already conducted in Dublin or Ireland (Lin et al., 2018, 2019a, 2019b, 2022, 2023, etc.), some of which also cover a substantial time span. What exactly is the core innovation of this study in terms of study period, methodology, or scientific questions compared to the aforementioned existing works? It is recommended to more explicitly articulate the specific knowledge gaps filled by this study in the introduction, rather than generalizing it loosely as the "first".

We fully acknowledge that previous studies have already provided important source-apportionment and source-characterization results for PM_1 in Dublin and Ireland, including those cited by the reviewer, as we have summarized in the Introduction. However, we would like to clarify that the manuscript did not intend to claim that this is the first PM_1 source apportionment study in Dublin or Ireland. The original statement was intended to emphasize the long-term, source-resolved characterization of PM_1 pollution in Dublin, particularly the multi-year trends in chemically resolved PM_1 components and source-related factors. Therefore, the main contribution of this study is not source identification itself, but the long-term assessment of how PM_1 chemical composition and source-related contributions have changed in Dublin from 2016 to 2023.

To avoid possible misunderstanding, we have revised the relevant statements in both the Introduction and Conclusions and the word “first” has been removed to better keep the focus on the long-term assessment of source-resolved PM_1 chemical composition and pollution-regime evolution in Dublin.

6. Page 4, Line 145; Page 14,Line 443 "nitrogen dioxides" is misspelled as "nitrogen deoxidizes"; "reduced" is misspelled as "recued". A thorough language check throughout the manuscript is recommended.

The misspellings of “nitrogen dioxide” and “reduced” have now been corrected. We have also carried out a thorough spelling and language check throughout both the manuscript and the Supplement to identify and correct other potential typographical errors.

RC2

General comments

This manuscript presents a long time series of ACSM data (~7 years) in Dublin, and uses this to assess trends in the influence of different aerosol species and the impact of specific sources during this time period. The authors demonstrate that local emissions from domestic heating have been reducing in response to changes in policy, while the influence of transboundary pollution is increasing. These two sources, together, are identified as the most important drivers of high pollution events in the city.

The long time series presented here is a strength of this study; datasets of real-time aerosol composition with this large a span are still relatively uncommon, making this a valuable addition to the literature. The results are important for understanding the drivers of aerosol pollution in European cities. Results are presented clearly and the conclusions are sound. I would recommend that this paper is suitable for publication in ACP, after the comments below have been addressed.

Specific comments

1. In lines 150–158, the authors describe using a ratio between eBC and OA to reconstruct the eBC time series where data were unavailable during 2018–2020. This is justified by citing the relatively stable ratio between the two species, with the reconstructed data then being used to analyse eBC trends. However, I would argue that the variation in ratio over the time series (between 0.18 and 0.31) is not negligible, and that this presupposes stability in the aerosol sources over the study period. Given the large amount of data missing, the trend that is identified is likely more a reflection of changes in OA than it is a true observation of a trend in eBC. I would suggest that there is not enough information available here to carry out a genuine trend analysis for eBC.

We thank the reviewer for raising this important concern. We agree that reconstructing eBC from OA introduces additional uncertainty, and that the variability in the eBC/OA ratio should not be neglected. However, we retained eBC in the long-term analysis because eBC is an important PM_1 component and a key tracer of combustion-related emissions. We also note that the reconstructed eBC data are not used here to interpret short-term or real-time eBC variability. The eBC-related analysis focuses on long-term annual trends and monthly mean patterns averaged over multiple years. Therefore, although the OA-based reconstruction

introduces uncertainty at shorter time scales, its influence is expected to be reduced for the monthly-to-annual scale analysis in this study.

To evaluate whether the reconstructed eBC data could bias the long-term trend, we performed two sensitivity tests. First, we compared the original reconstruction based on a single annual average eBC/OA ratio with a reconstruction based on more detailed monthly eBC/OA ratios derived from all periods with valid eBC measurements. This test accounts for the seasonal variability in the eBC/OA relationship. As shown in Fig. R1 and Table R1, using monthly ratios produced the same annual average eBC trend slope as the annual ratio method and did not change the Mann-Kendall trend significance, indicating that the use of a single annual ratio has only a limited influence on the long-term eBC trend.

Second, we repeated the eBC trend analysis after excluding the reconstructed eBC data for 2018 and 2020 and using only directly measured eBC data. As shown in Table R3, the resulting trend remained significant ($p < 0.05$), and the fitted slope was very similar to that obtained when the reconstructed data were included (-0.06 vs. -0.07 μg m^{-3} yr^{-1}). This indicates that the decreasing eBC trend is not simply an artefact of the OA-based reconstruction.

Table R3. Long-term trend results for annual eBC from 2016 to 2023 with the reconstructed eBC data in 2018 and 2020 included and excluded. The Mann-Kendall test results are shown together with linear regression slopes and 95% confidence intervals calculated from annual average eBC concentrations.

2018 and 2020 (Reconstructed eBC data)	**P-value**	**Slope (95% CI) (μg m^{-3} $year^{-1}$)**
Included	<0.05	-0.07 (-0.10 – -0.04)
Excluded	<0.05	-0.06 (-0.10 – -0.01)

Overall, while the reconstructed eBC data inevitably carry larger uncertainty than directly measured data, the sensitivity tests suggest that the long-term decreasing eBC trend is robust and is not primarily driven by the OA-based reconstruction. We therefore retain eBC in the long-term trend analysis, but have added the details of the sensitivity tests to the Supplement (Section S1.1). We have also revised Section 2.1 and 3.2.1 of the manuscript to clearly acknowledge the additional uncertainty and to interpret the eBC trend more cautiously.

2. In section 3.1, the authors present a case study period to identify typical characteristics of pollution events dominated by transported aerosol, in contrast to those dominated by local heating emissions. How representative were these chosen examples? How many pollution events occurred throughout the full period, and how many of these were associated with each of these sources? Were there any other sources that led to similar events? It would be valuable to look at these case studies in the context of the full dataset. This could also be used to support the authors' decision to focus primarily on these two sources.

We agree that the two typical types of pollution events should be interpreted in the context of the full long-term dataset, rather than as isolated examples. To evaluate how representative the selected examples are within the full dataset, we have now classified all 157 polluted days during 2016-2023, defined as days with daily average PM_1 > 15 μg m^{-3} based on their dominant pollution sources. These days were classified as local home heating dominated, transboundary

dominated, or mixed/other events based on OA composition, with NO_3 fraction as an additional criterion for transboundary transport. A more detailed response to a similar concern has been provided in our response to Reviewer 1 under comments No. 2. The results are summarized in Table R2, showing that 96 polluted days were local home heating dominated (61 %), 31 days were transboundary dominated (20 %), and 30 days were mixed/other events, with only 3 polluted days showing less clear characteristics of either of the two main pollution processes.

In summary, this full-dataset classification shows that local domestic heating and transboundary transport, or events with mixed contributions, were the two main sources associated with polluted periods in urban Dublin, which supports our focus on these two pollution types in Section 3.1. To further demonstrate that the two examples are representative of recurring pollution processes in the full dataset, we have also added multi-year examples of local- and transboundary-dominated pollution episodes, together with one mixed event and one less typical event. A short discussion on the classification results and newly added examples has been added in Section 3.1 after the two case studies in the revised manuscript.

3. Lines 256–7: How do you know that 15–17% of HOA originates from oil heating?

We apologize for the ambiguity in the original wording. The value of 15-17% refers to the contribution of HOA to total OA mass during these cold months, not to the fraction of HOA originating from oil burning. We have revised this sentence, as shown below

"*Indeed, OA during these months comes almost entirely from local sources (around 90%), primarily from solid fuel OA (39-43%), followed by* OOA_{local} *(around 32%) and* ***HOA (15-17%, mainly originated from oil heating Lin et al., 2019b)***",

The relevant citation has also been added to support the attribution of HOA mainly to oil-heating emissions in Dublin during winter.

4. Lines 478–80: Please clarify whether this is referring to an increase in oxidative capacity in the region that the aerosol is originally emitted from, or an increase at the measurement site. This increase in local/regional O3 concentrations and oxidative capacity is mentioned several times – is there an understood or hypothesised reason for this increase? Given that this increasing trend in transboundary pollution is an important outcome of this paper, it would be valuable to explore the reasons for and implications of this in more depth.

We acknowledge that there was some ambiguity regarding the increased O_3 concentrations and enhanced atmospheric oxidative capacity in the original manuscript. We have now revised the relevant content (originally Lines 478-480) to clearly state that the slower reduction in OOA_{local}, compared to primary species, could be linked to enhanced oxidative capacity in urban Dublin, while the upward trend of OOA_{TBT} may also be related to enhanced oxidative processing in the upwind source regions and/or during transport from the UK and continental Europe, associated with rising regional O_3 levels. We have also added a brief discussion in Section 3.2.1, following the discussion of the long-term O_3 trend, about the potential reasons for the increased O_3 concentrations and the resulting enhancement in atmospheric oxidative capacity, as follows:

"*…In particular, as shown in Fig. S9b and Table S4, the* O_3 *concentration in Dublin increased from 38.7 µg* m^{-3} *in 2016-2017 to 49.4 µg* m^{-3} *in 2023 (2.21 µg* m^{-3} yr^{-1}*, 95% CI: -0.11 – 4.53). As one of the most important atmospheric oxidants and a key species closely linked to photochemical chemistry, the significant increase in* O_3 *may indicate enhanced atmospheric*

oxidative capacity in urban Dublin, which may potentially offset the benefits of reduced primary emissions through more effective secondary production (Huang et al., 2021; Sun et al., 2020; Nassau and Jaeglé, 2025). The increase in O_3 concentrations may be related to the non-linear response of O_3 chemistry to declining nitrogen oxides (NOx) emissions, leading to a lower O_3 titration by NO (Korhale et al., 2026). Indeed, NO_2 concentrations in Dublin showed a continuous decreasing trend, from around 20 μg m^{-3} in 2016-2018 to around 14 μg m^{-3} in recent years (Fig. S9b). Similar NOx-O_3 responses have been reported in the UK (Lee et al., 2020; Finch and Palmer, 2020) and many other urban areas (Yan et al., 2018; Sicard et al., 2020), where volatile organic compound (VOC)-limited photochemistry can lead to increased O_3 or slower O_3 decreases despite reductions in NOx emissions. In addition, the continuous decline in PM_1 concentrations may also indirectly enhance O_3 formation by reducing the heterogeneous scavenging of hydroperoxy radicals and reactive nitrogen species that are involved in photochemical O_3 production (Li et al., 2019). This highlights the need for coordinated control of PM, O_3, and their precursors as reductions in primary PM emissions alone may not be sufficient to achieve better air quality."

5. Lines 488–9: The SO2 trend is associated with "shifts in fuel usage under influences of the European energy crisis". A citation is needed here.

Thanks for the helpful suggestion. Relevant citations have now been added, and the statement has been slightly revised as follows:

"*…possibly linked to shifts in fuel usage under influences of the European energy crisis, such as potential increased use of lower-price fuels with higher sulfur contents (Fazelianov, 2023; Urbano et al., 2023). Such a shift may have occurred in some households despite the new solid fuel regulations and public awareness efforts, as economic pressures could still have influenced fuel choices. However, this interpretation needs to be further validated with direct fuel-use data in future studies.*"

6. Given the time period that is presented here, I was surprised that there was no mention of the impact of the COVID pandemic on emission patterns. Was any influence observed, and is there a chance that data from 2020 and 2021 may be biased as a result of behaviour change during this period?

We thank the reviewer for raising this important point. We agree that COVID-19 related restrictions may have affected human activity patterns and air pollutant emissions during 2020 and 2021 and may therefore have biased the derived long-term trends.

To evaluate whether the COVID-19 period affected our long-term trend conclusions, we performed a sensitivity test by repeating the trend analysis with the 2020 and 2021 data excluded. Specifically, the Mann-Kendall tests were repeated using daily averages after excluding these two years, and the annual linear slopes were recalculated using annual mean concentrations without 2020 and 2021. This comparison was used as a sensitivity check rather than as an independent trend estimate, since excluding these two years reduces the number of annual points available for the linear fitting.

Table R4. Comparison of Mann-Kendall test results and fitted slopes for major PM_1 species using the full dataset and a sensitivity dataset excluding the COVID-19 affected years (2020 and 2021).

Species	P-value (20-21 excluded)	Slope (95% CI) (μg m^{-3} $year^{-1}$) (20-21 excluded)	P-value (all years)	Slope (95% CI) (μg m^{-3} $year^{-1}$) (all years)
PM_1	<0.05	-0.42 (-0.65 – -0.19)	<0.05	-0.46 (-0.70 – -0.23)
Org	0.14	-0.13 (-0.42 – 0.18)	0.20	-0.14 (-0.34 – 0.06)
SO_4	<0.05	-0.03 (-0.08 – 0.02)	0.06	-0.04 (-0.09 – 0.02)
NO_3	<0.05	-0.09 (-0.14 – -0.03)	<0.05	-0.11 (-0.18 – -0.03)
NH_4	<0.05	-0.08 (-0.13 – -0.02)	<0.05	-0.09 (-0.16 – -0.03)
eBC	<0.05	-0.06 (-0.09 – -0.03)	<0.05	-0.07 (-0.10 – -0.04)
Solid fuel OA	<0.05	-0.07 (-0.20 – 0.06)	<0.05	-0.08 (-0.16 – -0.02)
HOA	<0.05	-0.03 (-0.05 – -0.01)	<0.05	-0.04 (-0.06 – -0.02)
OOA_{local}	0.89	-0.02 (-0.14 – 0.08)	0.64	-0.03 (-0.10 – 0.03)
OOA_{TBT}	<0.05	0.02 (-0.01 – 0.05)	<0.05	0.02 (-0.08 – 0.13)

The results, as summarized in Table R4, show that excluding 2020 and 2021 does not substantially change the main trend conclusions. The decreasing trends in PM_1, NO_3, NH_4, eBC, solid fuel OA, and HOA remain significant, while total OA and OOA_{local} remain non-significant, with the fitted slope variations within 10%. The only minor change is observed for SO_4, for which the trend becomes significant after excluding 2020 and 2021, however, the original p value was already close to the significance threshold (p = 0.06), and the fitted slope remains very similar. Overall, the trend directions are unchanged and the fitted slopes are generally comparable with and without the 2020-2021 data.

Therefore, while COVID-19 related behavioural changes may have had source-specific effects, particularly for traffic-related emissions (detailed discussion is out of the scope of this study), our sensitivity analysis suggests that the main long-term PM_1 and chemical component trend conclusions are not primarily driven by the inclusion of the COVID affected years. We have included Table R4 in the Supplement and have also added a brief discussion of this point in the revised manuscript in Section 3.2.1 as below:

"*Since the study period includes 2020 and 2021, COVID-19 related restrictions may have altered human activity patterns and emission intensities (Sun et al., 2020; Shi et al., 2021), potentially influencing the derived long-term trends. To assess this effect, we performed a sensitivity test by repeating the trend analysis after excluding the 2020-2021 data. However, as shown in Table S5, the overall direction and statistical significance of the trends remained largely unchanged, and the fitted slopes were generally comparable to those obtained using the full dataset. This suggests that, although COVID-19 related behavioural changes may have affected short-term concentrations and chemical composition, the main long-term trend conclusions are not significantly biased by the inclusion of the COVID affected years.*"

In addition, we also acknowledge that COVID-19 may have led to longer-lasting behavioural changes after the main restriction period, such as changes in working-from-home patterns and residential heating demand. However, such effects cannot be robustly evaluated within the scope of the present dataset, particularly given concurrent influences from fuel-use changes, regulatory developments and broader long-term emission changes.

Technical comments

1. Line 319: ‘annual’ is missing an ‘l’.

Corrected.

2. 6: The purpose of the grey markers should be explained in a legend or in the figure caption.

We agree that the original Figure 6 caption did not explain the meaning of the grey markers explicitly enough. We have now revised the caption to clarify that the grey open-circle markers indicate the number of hourly data points, as shown below:

“Figure 6. Long-term trends in average mass concentrations and relative contributions of PM1 and OA components under (a) low, (b) moderate and (c) high pollution conditions from 2016 to 2023 in Dublin. Data from 2019 are excluded from the trend analysis due to biased coverage and are shown in grey. ***The grey open-circle markers in the top panels indicate the number of hourly data points in each year under each pollution level, read from the right-hand axis.***”

3. All figures with trendlines: the significance of the trendline should be visible somewhere on the figure.

The significance (p-value) of the long-term trends has now been added to all figures with trendline.