

Response to Reviewer Comments – responses in red text.

We are extremely grateful to the reviewers for their valuable and constructive comments. We have carefully revised the manuscript and addressed all comments. We believe these suggestions have improved the manuscript.

Reviewer 1- Comments

General Comments

RC1 1

This is a nice study of the factors contributing to ozone levels in Ireland. It illustrates the different trends at urban, rural and coastal sites. The addition of the ozone tagging is helpful to understand the contributing factors to these trends. This will be suitable for publication after addressing some points below. Some of the descriptions around NO_x-saturated vs NO_x-limited could do with clarifying, particularly with regard to season. It seems reasonable that urban sites will always be NO_x-saturated, but it would be surprising if rural sites in Ireland were NO_x saturated outside winter. It would be useful to compare with other studies in similar latitudes (maybe UK?) to see if NO_x saturation has been seen in other rural sites. The patterns in figure 3 show rural sites decreasing throughout the spring and summer (except for Laois) which would be consistent with NO_x-limited chemistry, but with very strong increases in February which presumably ends up causing a positive annual trend in table 2. Since February ozone rarely causes exceedances, describing these sites as having an increasing trend might imply pollution is getting worse at these sites when it probably isn't getting worse in the peak seasons. Some discussion of why Laois is behaving more similarly to the urban sites would be useful. Similarly, discussion of the contributions of EU and N. American NO_x seem to emphasise the importance of NO_x titration which is mostly wintertime rather than the increase springtime production. The discussions in the supplement should be brought into the main paper if they are important or removed if not.

AR -We are very grateful to the reviewer on providing these insightful comments that have guided us towards improvement of the manuscript.

Referring to the general comment above, we acknowledge that Irish conditions rarely lend themselves to photochemical ozone production, hence we do not infer that there is a NO_x saturated photochemical production regime in Ireland. We instead suggest that reduction of NO_x leads to decreased O₃ removal, as seen in Finch and Palmer 2020, a comparable study in the UK. The manuscript has been updated to reflect this point. Similarly, we have revised the wording to ensure not to imply that pollution is worse in periods when exceedances are not an issue. We have also commented on the reason Laois is influenced by local emissions.

Changes in Manuscript:

L312-315

It is duly noted that NO_x driven O₃ removal dominates over photochemical production in these sites. A comparable study in the UK carried out by Finch and Palmer (2020) attributed similarly rising trends in surface O₃ between 1999 and 2019 to decreasing NO_x, characterising UK observation sites as VOC-limited.

L344 -L349

Laois is characterised as a rural site yet exhibits rising trends like the urban sites for all months except December, indicating that the measurement station is affected by nearby emissions. Seasonal trends of the 15-year dataset are supplied in supplementary figure S2, where coastal stations exhibit a pronounced increase in late winter, and a decrease throughout the spring and summer, with a consistent near-year-round increase in Rathmines and Laois.

Specific comments

RC1 2

Line 66-68: This should comment on whether the NAO increases or decreases ozone levels.

AR - During a positive NAO phase, surface ozone levels increase. In contrast, during a negative NAO phase (NAO-low), ozone levels decrease. This has been added to the revised manuscript

Changes in Manuscript:

L81 -85

Another factor which influences O₃ levels is the North Atlantic Oscillation (NAO), which influences O₃ levels in Western Europe. During a positive NAO phase, O₃ levels increase. In contrast, during a negative NAO phase (NAO-low), O₃ levels decrease. This effect is particularly notable in southwest, central, and northern Europe (Bonaccorso et al., 2015; Creilson et al., 2003; Pausata et al., 2012).

RC1 3

Line 82: Is this 40% increase global, EU or Ireland?

AR - It is a global increase and now mentioned it revised manuscript.

Changes in Manuscript:

L98 -100

Globally, over the past 150 years, there has been a 40% increase in O₃ levels owing to rising precursor emissions. (Archibald et al., 2020; Griffiths et al., 2021; Young et al., 2013).

RC1 4

Line 183: It might be useful to give the grid resolution in km over Ireland

AR- Yes, the details of CAM -Chem Model grid in km is added in revised manuscript.

Changes in Manuscript:

L225-227

The model simulations were carried out at a horizontal resolution of 210 km × 280 km, with 56 vertical levels for the 2000-2018 period, with specified dynamics derived from MERRA2 reanalysis. (Molod et al., 2015).

RC1 5

Section 3 figures: It would be useful to keep the same order of stations throughout the tables and figures, and to group coastal, rural and urban so that a reader doesn't have to repeatedly refer to table 1.

AR - Yes, the figures have been revised according to the order in Table 1 and have been added to the revised manuscript

RC1 6

Figure 2: This figure averages over different time periods for different stations. Given the trend in concentrations over time this can distort the comparison. The same period should be used as far as possible.

Response - Yes, the time periods differ for these sites due to the start dates of measurements.

The data period is mentioned in in table 1.

RC1 7

Line 212: It is not obvious why a single year (2003) has been highlighted for Valentia when all other values are time period means.

Response - Yes, the values represent the mean over the specified time period. This is now included in the revised manuscript.

Changes in Manuscript:

L266-268

Coastal sites, Mace Head and Valentia, show higher O₃ levels compared to other sites, with annual average concentrations of 77 µg/m³ and 69 µg/m³, respectively.

RC1 8

Line 277-278: Figure S2 shows coastal increases in February which is conventionally "Late winter" rather than "early spring". And then decreases throughout the spring and

Summer.

AR – The sentence in the manuscript has been revised accordingly

Changes in Manuscript:

L 346 – 349

Seasonal trends of the 15-year dataset are supplied in supplementary figure S2, where coastal stations exhibit a pronounced increase in late winter, and a decrease throughout the spring and summer, with a consistent near-year-round increase in Rathmines and Laois.

RC1 9

Figure 3 needs error bars. I couldn't see the *** markings.

AR - This figure shows monthly trend values, so plotting error bars is not feasible. Instead of using asterisks to show significance, the significant values are highlighted using colour for better visualisation.

Changes in Manuscript:

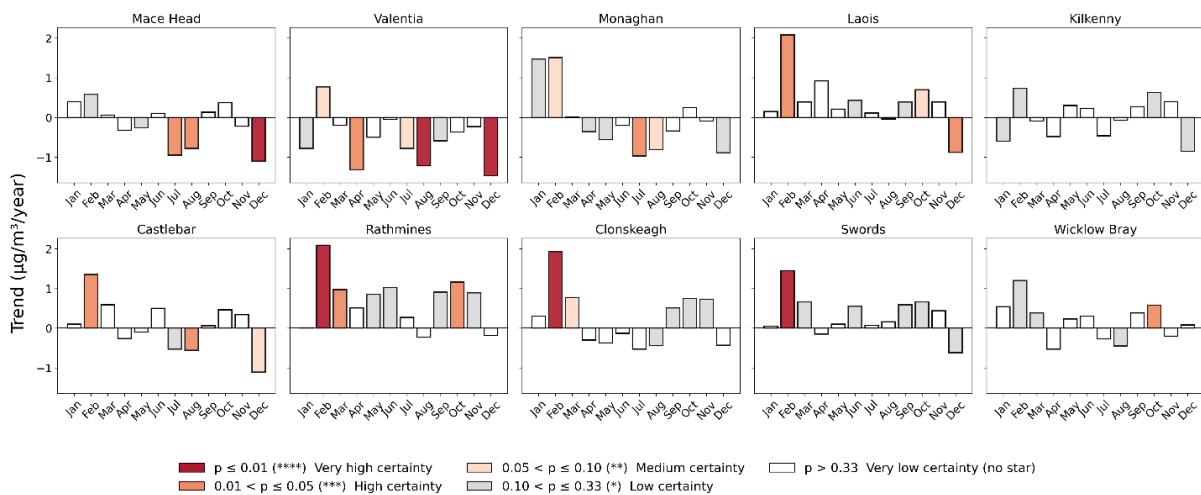


Figure 3. Monthly trend analysis of O_3 at different sites for 10 year period. (2012-2022) Adopting the trend reliability scale defined for TOAR-II studies (Chang et al., 2023), trends significance highlighted by colour.

RC1 10

Figure 5: What years are these trends calculated over? Are they the same or different. As with figure 2 the use of different time periods might distort the comparison.

Response - . NO₂ trend calculations are based on site-specific data periods , Cork South Link Road (2014–2022), Ballyforemont (2003–2022), Davitt Road (2018–2023), Rathmines (1995–2024), Swords (2011–2025), Laois (2014–2026), Castlebar (2003–2027), Louth Dundalk (2019–2028), and Monaghan (2001–2029). For CH₄, the data cover the period 2010–2022.

Changes in Manuscript:

L378-381

NO₂ trend calculations are based on site-specific data periods , Cork South link Road (2014–2022), Ballyfermot (2003–2022), Davit Road (2018–2023), Rathmines (1995–2024), Swords (2011–2025), Laois (2014–2026), Castlebar (2003–2027), Louth Dundalk (2019–2028), and Monaghan (2001–2029). For CH₄, the data cover the period 2010–2022.

RC1 11

Line 329 to 347: The percentages changes in figure 6 need some uncertainty analysis – are they at all significant? Could much of the change be due to meteorology in 2020 vs that in 2017-2019? Could there be a circulation change that reduced ozone at the coast but increased it inland? Or could changes in insolation enhance ozone production inland but enhance destruction over the ocean? The stronger change at rural sites rather than urban doesn't support NO₂ titration being the major contribution to the 2020 increases. The % changes in rural ozone seem very large compared to the NO₂ decrease – it would be good to see this in ppb where presumably the concentration change would be even larger. It is not obvious that there is sufficient decrease in NO₂ to explain the ozone increase through titration.

AR - The strongest ozone changes observed at Laois, Swords, Rathmines, and Monaghan are all inland and affected by European and surrounding local emissions, whereas the coastal stations are influenced by long-range transport. It is noted that the NO_x titration is not the dominant removal mechanism. However, to confirm this we would require concurrent O₃, NO and NO₂ measurements at each site, which are unavailable.

The discussion has been expanded to consider the unique meteorological conditions associated with 2020, with relevant citations and the competing factors governing changes in O₃ during the lockdown period, notably the effect of transport – hence, it is highlighted that it is not the local NO₂ changes, but EU wide changes that influenced the increase in O₃ during lockdown.

Changes to Manuscript

L409 - 415

It is noted that April and May 2020 had unique meteorological conditions compared to previous years, with lower wind speed, less rain and significantly higher solar radiation, see Figure 12 in Spohn et al. (2022). These meteorological conditions would potentially facilitate photochemical O₃ production, contributing to positive O₃ anomalies during the lockdown period in addition to NO_x reduction, also potentially enhancing dry deposition to the ocean. Further investigation into this topic would warrant model sensitivity studies, beyond the scope of this current work.

RC1 12

Line 342: This sentence needs to be clearer. Is it referring to urban or rural sites? Average NO_x concentrations in each need to be stated and compared with literature studies of NO_x-limited vs NO_x-saturated conditions.

AR - We have now revised this sentence and included a relevant reference

Changes to Manuscript:

L 416 -420

The negative correlation between NO_x and O₃ under relatively clean atmospheric conditions indicates that O₃ levels are influenced predominantly by transport and chemical removal, and local photochemical production does not represent a significant surface O₃ source owing to periods of low-insolation periods and low temperature, which are characteristic of Irish meteorology and frequent cloud cover (Pallé and Butler, 2002).

RC1 13

Line 346: Did Ireland have low-insolation and frequent cloud cover in March, April or May 2020?

AR – The explanation refers to the general ozone chemistry over Ireland, which is influenced by both insolation and frequent cloud cover. Low insolation and frequent cloudiness are a general feature of Irish meteorology for any year, not specifically 2020. This means that Ireland is not a region of vigorous ozone photochemistry and the ozone abundance is controlled more by transported ozone formed elsewhere and by loss processes, as explained in revised manuscript, L 421 – 425, as above.

RC1 14

Line 356: Figure S5 should be referred to here to illustrate the collocation of the grid and the sites.

AR - The reference of Figure S5 has been incorporated into the revised manuscript

Changes to Manuscript:

L429

The grid details are shown in Figure S5

RC1 15

Line 428-433: This should discuss that the lifetime of ozone is much longer in winter and hence Ireland can receive transport from further distances. Even in winter insolation and temperatures will still be high enough to produce ozone in South Asia. Discussions of transport should cite HTAP 1 and 2 studies.

AR –The discussion has been included and cited HTAP1 and HTAP2 studies in the revised manuscript

Changes to Manuscript:

L508-511

Wintertime temperatures in South Asia are still high enough to sufficiently produce local ozone especially when NO_x emissions are rising (Crippa et al., 2023). The relatively longer atmospheric lifetime of O₃, in the free troposphere, during winter enables longer-range intercontinental transport. (Huang et al., 2017; Yu et al., 2013).

RC1 16

Figure 10: It would be useful to see the annual average trends shown too. It looks as if this would be positive for both clean and EU sectors. It looks as if most of the slopes are not significant at the $p < 0.05$ level. This should be commented on.

AR - The annual average trend is now shown in the supplementary material, Figure S7 which show a small positive slope in EU-influenced sector, without any statistical reliability of trend., and text has been added to the manuscript to comment on the significance of the slopes according to TOAR slope reliability criteria

Changes to Manuscript & Supplementary Material:

L 561 – 563

It is noted that the seasonal trends exhibit slopes with p-values ranging between $0.1 > p > 0.01$, which denote trends of medium to high certainty, as defined by TOAR assessment criteria of Chang et al., 2023

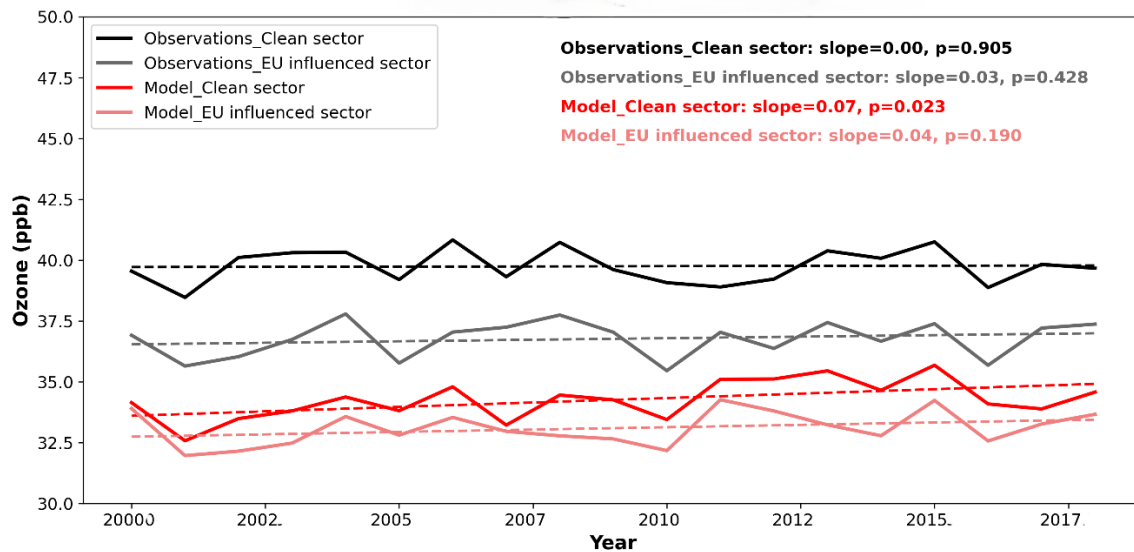


Figure S7

RC1 17

Line 462-463: It is not evident that there is a net chemical sink for ozone over Ireland in spring since ozone decreases with decreasing emissions. A dry deposition sink seems more plausible.

AR – The dry depositional sink is made explicit in the revised manuscript .

Changes to Manuscript:

L549 - 551

O₃ originating from EU airmasses is susceptible to higher rates of dry deposition and removal via local pollution while traversing the land-mass westwards towards Mace Head, leading to higher O₃ in clean-sector air masses consistent with previous studies (Coleman et al., 2013).

RC1 18

Line 464: Decreasing spring ozone from the EU influenced sector is not consistent with the increases in rural O₃ during COVID. This suggests that the COVID ozone change was not due to emissions.

Response - The trends in clean sector and EU influenced sector are calculated for sustained period of decades and the increase in lockdown O_3 are observed in the short-term at sites around Ireland, confounded by unique meteorology of 2020. Further, in this section, we focus only on observations at Mace Head, where local sources and hence NO_x titration has little influence. This is reflected in the fact that the increase in O_3 observed over lockdown was not observed at Mace Head or Valentia, where there was a decrease in mean O_3 compared with average of previous years, as discussed in manuscript.

L405-409

Significant enhancement of O_3 occurs at the inland measurement sites, despite a 2020 springtime decrease in O_3 observed at background coastal sites, Mace Head and Valentia. These coastal stations are less sensitive to changes in European NO_x emissions than inland sites and more sensitive to stratospheric and hemispheric transport (Tan et al., 2018).

RC1 19

Lines 466-467: The scavenging of ozone via NO_x titration is only true in the winter months. In spring and summer, the decrease in ozone with decreasing emissions indicates net chemical production in the EU sector.

AR – The manuscript has been changed to reflect this point

Changes to Manuscript:

L545– 548

The figure shows that the clean sector has consistently higher O_3 concentrations than the EU influenced sector for Winter, Spring and Autumn, with the most significant disparity between clean and EU sectors in winter/spring when stratospheric intrusion and lightning NO_x contribute most significantly to O_3 , as discussed in Section 3.4.2.

RC1 20

Lines 472-474: It is not clear what is meant by “there is little O_3 advected into Europe

from the west in the summer months”. Is this just saying that there is no east-west gradient in ozone therefore eastward or westward advection will have no effect.

AR – The intention was to state that the air flow from the west is not a dominant source in the summer months, but the sentence has been removed for clarity.

RC1 21

Line 481: How are the clean and EU sectors separated in the model data?

AR- Both observational and the model data are separated into clean/EU-influenced datasets according to the clean/EU-influenced days identified by the trajectory analysis. The trajectory analysis is described in Section 2.2 and the application of the analysis to split both measured and modelled data has been clarified in the manuscript.

Changes to Manuscript:

L540-543

Although Mace Head is classified as a global background site, quantification of the baseline pollution levels requires trajectory analysis, whereby both measured and modelled data is filtered to limit the data to that arriving from the clean sector, unaffected by land-based emission sources, as discussed in Section 2.2.

RC1 22

Line 505: It should be explained why the exceedances are higher for the EU sector even though the mean concentrations are lower – presumably the variance is higher, which would be interesting to discuss.

AR This discussion has been added to the manuscript

Changes to Manuscript:

L597-602

It is notable that there is a higher proportion of exceedances that occur from EU- influenced sector, despite higher mean observations from the clean sector for all seasons, bar summer. This occurs because of an enhancement of surface O₃ occur during an influx of polluted air from EU, UK or local sources. The EU influence on exceedance becomes more proportionally prominent in late Spring and summer, with more frequent easterly airflow when there is a higher occurrence of stagnation events.

RC1 23

Line 515: Should clarify that the greater decreasing trend is for the EU sector.

AR – this is addressed accordingly

Changes to Manuscript:

L609-610

A decreasing trend in exceedances is observed, with a greater decreasing trend in from the EU and locally influenced sector.

RC1 24

Line 539: Add “VOC” after “dominant”.

AR - Added in revised manuscript.

Changes to Manuscript:

L638-639

Figure 13 (b) shows that CH₄ is the most dominant VOC source, followed by stratospheric intrusion and Biomass burning

RC1 25

Line 543: The first clause in this sentence seems incomplete.

AR- It is revised in the revised manuscript.

Changes to Manuscript:

L644 – 646

North American NO_x also contributes significantly to exceedance in both clean and EU-influenced sectors at Mace Head during March to May month, likely due to long-range transport and mixing, regional stagnation or synoptic-scale recirculation.

RC1 26

Figure 13: Explain how the quantify plotted is derived. Is it the mean ozone above 100 ug/m³ summed for each day exceeded. The units on the y-axis must include a time dimension, presumably ppb days or maybe ug/m³ days if the exceedance criterion is in ug/m³? I suggest thinking about the colours used, it is difficult to see which shade refers to which region.

AR - First, the exceedances were identified, and then these exceedances were divided into two sectors the EU-influenced sector and the clean sector. Figure 13 presents the hourly ozone exceedance cumulative concentrations in ppb, along with the contributions from different parameters. It indicates which parameters contribute more to the exceedances, in both EU-influenced sector or the clean sector. In this the ug/m³ to ppb conversion is considered for the exceedances.

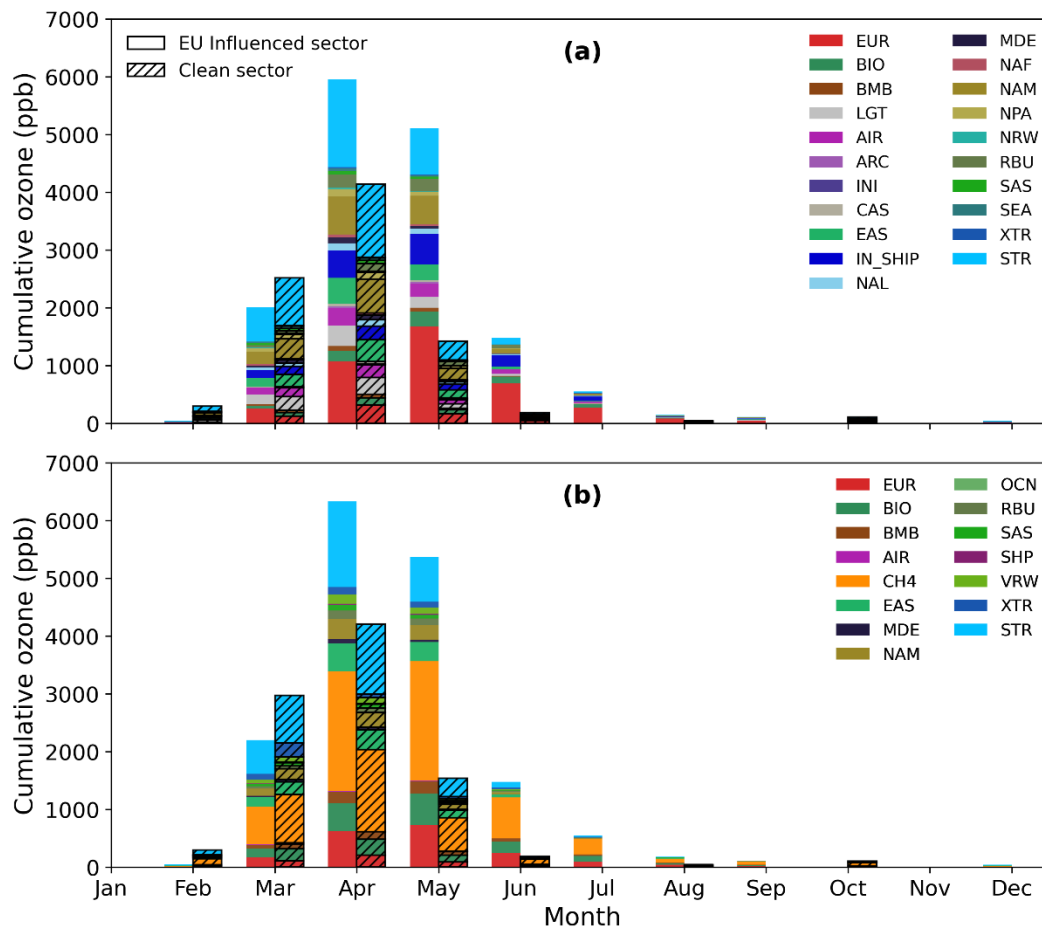
Changes to manuscript:

L626-633

Figure 13 shows monthly cumulative contributions to simulated O₃ concentrations within the Mace Head grid cell for NO_x and VOC tagging during O₃ exceedance. First, the exceedances were identified from O₃ observations as discussed in Section 3.3 and then these exceedances were divided into two sectors the EU-influenced sector and the clean sector. In Figure 13, the

hourly O_3 exceedance cumulative concentrations in ppb, along with the contributions from different parameters, are shown. It indicates which parameters contribute more to the exceedances, in both the EU-influenced sector or the clean sector. In this the ug/m^3 to ppb conversion is considered for the exceedances.

Revised colour scheme:



RC1 27

Lines 558-560 The increased urban ozone is likely to be dominated by the local NO_x changes rather than from Europe and North America as suggested here. The exception might be winter, when ozone is low anyway. This study hasn't demonstrated that European and North American NO_x are important in urban areas.

AR – the manuscript has been adapted to explicitly acknowledge the role of local NO_x, and we have amended the text to allude to the reduction in winter time ozone depletion events due to regional transport of European outflow of CO & CFCS (Derwent et al 2024, Simmonds and Derwent 1991)

Changes to manuscript:

L657 - 661

Over the last two decades, urban sites have shown a significant increasing trend in O₃ levels, particularly in winter, influenced by decreasing anthropogenic pollution in Europe, the UK, North America and local to the observation sites, representing a decline in chemical removal mechanism (Derwent et al., 2024; Simmonds and Derwent, 1991).

RC28

Lines 560-562: This claim of correlation of exceedances at coastal sites with years of higher spring maxima is not supported by any of the text in section 3.3. Note that most of the exceedances come from the EU sector which would suggest an EU rather than hemispheric source

AR – Thanks, this was badly articulated – the majority of exceedances occur in the spring months, coinciding with spring maximum which peaks in April. As discussed above, the spring maximum influenced by stratospheric transport, hemispheric and LRT, and EU pollutions pushes over the exceedance threshold, but this was determined from the simulation results, so this sentence has been revised.

Changes to manuscript:

L661-662

The analysis also points out that the majority of exceedances at coastal monitoring sites coincide with the annual spring maxima.

Reviewer 2 - Comments

This paper addresses an important topic: surface ozone (O₃) trends across Ireland, using longterm observational data and model (CAM4-Chem + tagging via TOAST 1.0) source-attribution analyses. The combination of measurements, trajectory / sector classification, and modelling is commendable. The regional focus on Ireland particularly the background coastal site at Mace Head adds value, since many O₃ studies focus on larger continental areas. The methods are reasonably described, and the results are of interest for air-quality / atmospheric chemistry / policy communities. That said, the manuscript requires substantial improvement in several areas before it is ready for publication: grammar & sentence structure need polishing, some methodological choices need clarification, some results could be better contextualized, and a number of references appear missing or inconsistent.

AR - We sincerely thank the reviewer for their valuable and constructive comments. We have carefully revised the manuscript to address the concerns raised regarding grammar and sentence structure and all references have been checked and updated for accuracy and consistency.

Major concerns

RC2 1

The abstract and introduction present the study aims, but the phrasing is sometimes confusing. For example: “Using innovative trajectory analysis, O₃ concentrations, exceedances and were identified by sectors...” (L17–19) – the sentence is awkward, and a verb seems missing (“and ... were identified”).

AR - This sentence is rephrased in revised manuscript and the abstract has been rewritten to improve flow

Changes to manuscript:

L16-36

We present an analysis of long-term trends in surface ozone (O₃) across Ireland, with specific focus on the Mace Head atmospheric research station, representative of Northern hemispheric background atmospheric conditions. Surface O₃ dataset was characterised using advanced trajectory analysis and seasonal decomposition, revealing distinct seasonal and spatial

patterns. Findings show a significant rising trend in surface O₃ at Irish urban sites over the past two decades but without a similar trend at coastal sites. Highest O₃ levels and exceedances were observed at remote coastal sites, which are less susceptible to influence from local and easterly emissions but heavily influenced by transboundary pollution and stratospheric intrusion.

At Mace Head, springtime O₃ levels exhibit a declining trend, whereas wintertime levels show a rising trend. Focussing on the clean sector, the springtime decline remains significant, but without corresponding clean sector rising wintertime trends, implying the rising winter trends occur in response to declining local, United Kingdom (UK) and European emissions. Advanced modelling tools are used to quantify O₃ source contributions, elucidating key drivers behind the observed changes. Characteristic springtime O₃ maxima at Mace Head are predominantly attributed to stratospheric transport, hemispheric and long-range transport and lightning NO_x. The complementary trend and sectoral observational analysis reveal a decline in total springtime concentrations, with a more rapid decline in exceedances from the UK & continental sector.

This research highlights the importance of seasonal factors in air quality management across Ireland, emphasising the need for a multi-faceted approach to control O₃ levels and reduce exceedances through global and regional emission reductions.

RC2 2

It would help to explicitly state the hypotheses (e.g., “We hypothesise that urban sites will show increasing O₃ due to reduced NO_x titration while background coastal sites will show declining O₃ thanks to precursor reductions”). A clear statement of hypotheses will strengthen the framing.

AR - The study aims to identify trends, patterns and drivers of changes, rather than testing a hypothesis. In this context, our article is more investigative in nature. To clarify the methods, we have included a description of the analysis in terms of surface ozone sources at the end of the introduction

Changes to manuscript:

L114-133

This study investigates the distribution and trends of O₃ and its precursors across Ireland, providing valuable insights into the regional and hemispheric impact on Irish surface O₃ levels and exceedances. We analyse the long term O₃ observational dataset for Ireland, identifying the mean and range of O₃ levels and seasonal variation at each site. We identify the long-term annual and monthly trends at each site, quantifying the significance of each trend according to TOAR guidelines (Chang et al., 2023). We also identify the frequency and seasonality of exceedance of the WHO AQGs for the protection of human health from O₃ pollution for each site. The trends and seasonality are contextualised by looking at trends in the Irish instrumental record of dominant precursors of NO_x and CH₄ and we discuss the relationship between NO_x and O₃ by comparing anomalies between monthly average NO_x and O₃ during lockdown compared to the average values for the three years prior to lockdown. Advanced modelling results using the global Tropospheric Ozone Attribution of Sources with Tagging 1.0 (TOAST 1.0) framework (Butler et al., 2018; Butler et al., 2020) are validated against measurements at various sites for simulation period 2000-2018, and the simulation results analysed to determine the geographical and sectoral source of precursors that contribute to simulated O₃ at Mace Head, identifying seasonality and long-term trends in the sources. Finally, the observational data are classified using advanced trajectory clustering methods to separate air masses from the clean sector from those influenced by local, UK or EU emission sources, with seasonal

trends identified for both clean and polluted sectors, and the exceedances classified as coming from either the clean or polluted sector.

RC2 3

The authors mention “innovative trajectory analysis” but more precisely explain what is new compared to previous work. Many previous studies have done back-trajectory classification. Clarify what is novel.

AR– In this work, we used 22-year, 72-hour back trajectories, which were filtered into “Clean sector ” and “EU Influenced sector ” air masses. This approach is particularly important due to the unique geographic location of Mace Head, situated on the west coast of Europe and in close proximity to the Atlantic Ocean. Understanding the origin and transport mechanisms of clean air masses is crucial in this context. These clean air pathways establish an essential baseline. By characterising them, our method goes beyond tracking pollution, it quantifies the exposure to natural background conditions and reveals how large-scale circulation patterns influence ozone levels. This allows for a more precise attribution of ozone exceedance events, distinguishing between changes driven by transport from the North Atlantic Ocean and from Europe. It builds upon previous non-hierarchical clustering mechanisms by differentiating between trajectories that have passed over land, and those uninfluenced by land-based emissions - allowing exclusion of continental and local emission sources on ozone concentrations instead of clustering via air-mass origin. Therefore, we mentioned it as this innovative methodology, but it is perhaps more accurate to describe it as advanced trajectory analysis. We adopt this more accurate terminology in the revised manuscript.

Changes to manuscript:

L129-133

Finally, the observational data are classified using advanced trajectory clustering methods to separate air masses from the clean sector from those influenced by local, UK or EU emission sources, with seasonal trends identified for both clean and polluted sectors, and the exceedances classified as coming from either the clean or polluted sector.

RC2 4

I suggest a thorough pass with a native English speaker or professional editing service to improve readability, grammar, and logical flow.

AR- We would like to thank the reviewer for the constructive suggestions to improve to the paper's readability and structure. The entire paper has been thoroughly revised by authors for readability.

RC2 5

The trajectory classification: The criteria (72h over ocean) for “clean sector” should be justified more clearly. Why 72 h? Why 100 m height at 6:00 UTC? Are results sensitive to these thresholds?

AR - The 72h duration captures regional/long-range transport without trajectory error from meteorological uncertainties. The 100 m height was used to represent the well-mixed flow of the boundary layer above the surface. The 06:00 UTC aligns with synoptic times and can match daily ozone cycles or measurement periods.

Changes to manuscript:

L200-204

The 72h duration captures regional/long-range transport without trajectory error from meteorological uncertainties. The 100 m height was used to represent the well-mixed flow of the boundary layer above the surface. The 06:00 UTC aligns with synoptic times and can match daily O₃ cycles or measurement periods

RC2 6

The modelling: The CAM4-Chem grid resolution is coarse ($1.9^{\circ} \times 2.5^{\circ}$) (L229). This is acknowledged by the authors in discussing biases (L486–490). However, the implications for interpretation (especially for urban sites) should be more emphasised. Is the coarse resolution adequate for urban site comparisons?

AR – We are in agreement that CAM-Chem has a coarse spatial resolution, it is still appropriate for this study because it provides a reliable representation of the regional background atmosphere influencing the urban site. The model captures large-scale features of atmospheric transport, seasonal variability, and background ozone levels, all of which are essential for interpreting urban observations. By comparing urban measurements with regional-scale CAM-Chem outputs, we can distinguish local pollution effects from broader atmospheric processes. The purpose of using CAM-Chem here is not to reproduce fine-scale urban hotspots, but to understand the larger chemical environment surrounding the city.

Changes to manuscript:

L227-232

The coarse spatial resolution, it is still appropriate because it provides a reliable representation of the regional background atmosphere influencing the urban sites. The model captures large-scale features of atmospheric transport, seasonal variability, and background O₃ levels, all of which are essential for interpreting urban observations. Comparing urban measurements with regional-scale CAM-Chem outputs allows local pollution effects to be distinguished from regional atmospheric influences.

RC2 7

The source tagging via TOAST is described, but I would recommend including a validation of the tagging method (or refer to validation studies) in a little more detail. For instance, what is the error/uncertainty associated with the tagging?

AR - The references for the validation studies of the TOAST tagging method, including information on errors and uncertainties, have been added. The sum of all tagged contributions very closely matches the total simulated ozone (which is simulated independently and is not just an algebraic sum of the tagged contributions) is a good validation of the tagging mechanism. (Butler et al., 2018 and 2020.)

Changes to manuscript:

L246-248

The sum of all tagged contributions very closely matches the total simulated ozone (which is simulated independently and is not just an algebraic sum of the tagged contributions) is a good validation of the tagging mechanism (Butler et al., 2018 and 2020).

RC2 8

Statistical trend methods: The authors use Theil-Sen slopes (L180–185). It may help to compare with alternative methods or at least to discuss the limitations (e.g., nonstationarity, autocorrelation).

AR - In this trans analysis, Theil-Sen method was chosen to quantify the trends magnitude. Mann-Kendall primarily provides a p-value, while Theil-Sen directly provides robust slope estimate, providing a reliable measure of change over time for direct interpretation and comparison. This provides the trend's actual impact, such as the rate of increase per year. The limitations of Mann-Kendall tests are added in revised manuscript (line 181 -185).

Changes to manuscript:

L181-185

Theil-Sen directly provides a robust slope estimate. It is a reliable measure of change over time for direct interpretation and comparison. It is a robust method for estimating trend slopes in time series data, preferable to traditional least-squares regression, which can be sensitive to extreme values and outliers.

RC2 9

In modelling vs observations comparison (Section 3.4.1), only five sites are used. Why these? Are they representative? Could more sites be used or reasons given for the selection?

AR - The CAM-Chem model grid covers Ireland in 4–5 grid cells. The five selected sites are located in different grid cells., represent different Irish regions and have the advantage of continuous long-term measurements.

RC2 10

The result that urban sites show increasing O₃ trends (e.g., Rathmines +0.27 μg m⁻³ yr⁻¹ for full period) (L363–365) is interesting. But the mechanism is briefly mentioned (“weekend effect”, NO_x titration) (L311–313); this could be developed further, perhaps linking to local emission inventories or changes in VOC/NO_x ratios over time in Ireland.

TAR-This is expanded on in the manuscript, and it is emphasised that the effect is related to the role of NO_x in the removal of ozone, noting that Irish meteorological conditions allow limited

photochemical production of ozone due to frequent cloud cover and low insolation. This effect is supported by referring to a UK study (Finch and Palmer, 2020) which observed similar surface ozone increase coinciding with NO_x reduction.

Changes to manuscript:

L312-315

It is duly noted that NO_x driven O₃ removal dominates over photochemical production in these sites. A comparable study in the UK carried out by Finch and Palmer (2020) attributed similarly rising trends in surface O₃ between 1999 and 2019 to decreasing NO_x, characterising UK observation sites as VOC-limited.

RC2 11

The discussion of the 2020 COVID-19 lockdown (Section 3.3) is interesting (L451–458). But the data are limited (March-May 2020) and a more nuanced discussion of meteorology confounding effects would strengthen the claim.

AR - The manuscript has been updated to discuss the confounding effect of meteorology during lockdown.

Changes to manuscript:

L409-415

“It is noted that April and May 2020 had unique meteorological conditions compared to previous years, with lower windspeed, less rain and significantly higher solar radiation, see Figure 12 in Spohn et al. (2022). These meteorological conditions would potentially facilitate photochemical O₃ production, contributing to positive O₃ anomalies during the lockdown period in addition to NO_x reduction, also potentially enhancing dry deposition to the ocean. Since our model simulations presented here end in 2018, further investigation into this topic would warrant model sensitivity studies, beyond the scope of this current work.”

RC2 12

The authors report a decline in spring-time O₃ at Mace Head but an increase in winter trends (L465–470). This is a key finding. However, the discussion linking these to emission

reductions, stratospheric intrusion, hemispheric transport is somewhat speculative and could be better supported by citations or sensitivity tests.

AR - We have amended the manuscript to relate the springtime O₃ decline at Mace Head to the tagged contributions from the CAM-Chem model simulations which differentiates between the different sources – stratospheric intrusion and emissions from different geographical sources. Tagged simulations are considered an advancement to sensitivity simulations, as described in L234-238 in manuscript, hence the link to the Mace Head spring time decline to decreasing EU and North American emissions are supported by the modelling results displayed in Figure 9.

Changes to manuscript:

L552-554

A decreasing trend in mean spring-time levels is observed for both clean and EU influenced sectors, consistent with the sustained decrease in precursor emissions in Europe and North America as displayed in Figure 9.

RC2 13

It would help to compare the Irish trends with trends elsewhere (Western Europe, North Atlantic) more thoroughly — e.g., are the trends consistent with broader European background ozone literature?

AR - The comparison of this trend with other are discussed in revised manuscript to show consistency with the most relevant previous studies in the European context, citing Yan et al 2018 and Nelson and Drysdale 2025. The North Atlantic context is supported by references to studies of Derwent.

Changes to manuscript

L322-327

Previous studies indicate that in northeast Europe, peak surface ozone concentrations have generally declined, reflecting the effectiveness of emission control measures (Yan et al., 2018).

In contrast, background and lower-level O₃ concentrations have continued to increase, particularly at rural and suburban sites. Consistent with this, urban observations from 2000–

2021 show increasing trends in median and lower-percentile O_3 levels, while the highest extremes have mostly decreased (Nelson et al., 2025).

RC2 14 The uncertainties in modelling and measurement (especially for attribution of sources) should be more explicitly discussed.

AR – A more detailed discussion is added in the methodology section for both measurements and modelling is added in revised manuscript

Modelling

L241-248

The model results have inherent standard uncertainties common in any modelling exercise, i.e., uncertainties in emission inventories in terms of magnitude and spatial accuracy; uncertainties in model parameters, e.g., surface resistance for deposition for various surfaces, boundary layer mixing, photolysis, chemical kinetic parameters (Wild et al., 2025) and structural deficiencies such as a coarse resolution and missing processes (e.g., halogen chemistry; Saiz-Lopez et al., 2025). The sum of all tagged contributions very closely matches the total simulated ozone (which is simulated independently and is not just an algebraic sum of the tagged contributions) is a good validation of the tagging mechanism (Butler et al., 2018 and 2020).

Measurements

L146-147

Measurement operating accuracy is within 1.0 ppb, based on precision, calibration and drift characteristics.

Minor Comments:

RC2 15

Line 229 - It would be useful to maintain the same order of stations throughout all tables and figures.

AR - The order of stations in all figures and tables is maintained in revised manuscript.

RC2 16

Line 326 - Figure 5 Why these NO₂ and CH₄ sites are selected.

AR - These sites were selected as the only available with long-term NO₂ and CH₄ datasets

RC2 17

Line 342 - This sentence needs to be rewritten for clarity; the current wording is difficult to follow.

AR - Sentence has been removed from the manuscript, but is clarified by stating that local photochemical production does not represent a major source.

Changes to manuscript:

L416-420

The negative correlation between NO_x and O₃ under relatively clean atmospheric conditions indicates that O₃ levels are influenced predominantly by transport and chemical removal, and local photochemical production does not represent a significant surface O₃ source owing to periods of low-insolation periods and low temperature, which are characteristic of Irish meteorology and frequent cloud cover. (Pallé and Butler, 2002).

RC2 18

Line 353 Include details of the model grid used over Ireland to support the interpretation of spatial results.

AR - Details of the model grid used over Ireland now it is added to supplementary material

Changes to the manuscript

L433

The grid details are shown in Figure S5

.

RC2 19

Line 415 Figure 8 Clarify what the SHIP parameter represents. It is not defined in Table 3.

AR -The SHIP parameter is used as the addition of all oceanic emissions now it is added to the revised manuscript

Changes to the manuscript

L481-482

The total shipping NO_x (SHIP) also contributes significantly. It is the addition of all oceanic emissions and shows the highest contribution in June month.

RC2 20

Line 456 - Please provide details of the method used to define background and EU- influenced airmasses. This information is essential.

AR - The detailed methodology for define background and EU- influenced airmasses is in section 2.2 of the manuscript, and the separation of measurement and modelling data according to the trajectory sectoral classification is explicitly referenced in the manuscript.

Changes to Manuscript:

L540-543

Although Mace Head is classified as a global background site, quantification of the baseline pollution levels requires trajectory analysis, whereby both measured and modelled data is filtered to limit the data to that arriving from the clean sector; unaffected by land-based emission sources, as discussed in Section 2.2.

RC2 21

Line 543 Check this sentence; it appears incomplete and needs revision.

AR- The sentence is revised in the manuscript.

Changes to manuscript:

L644-646

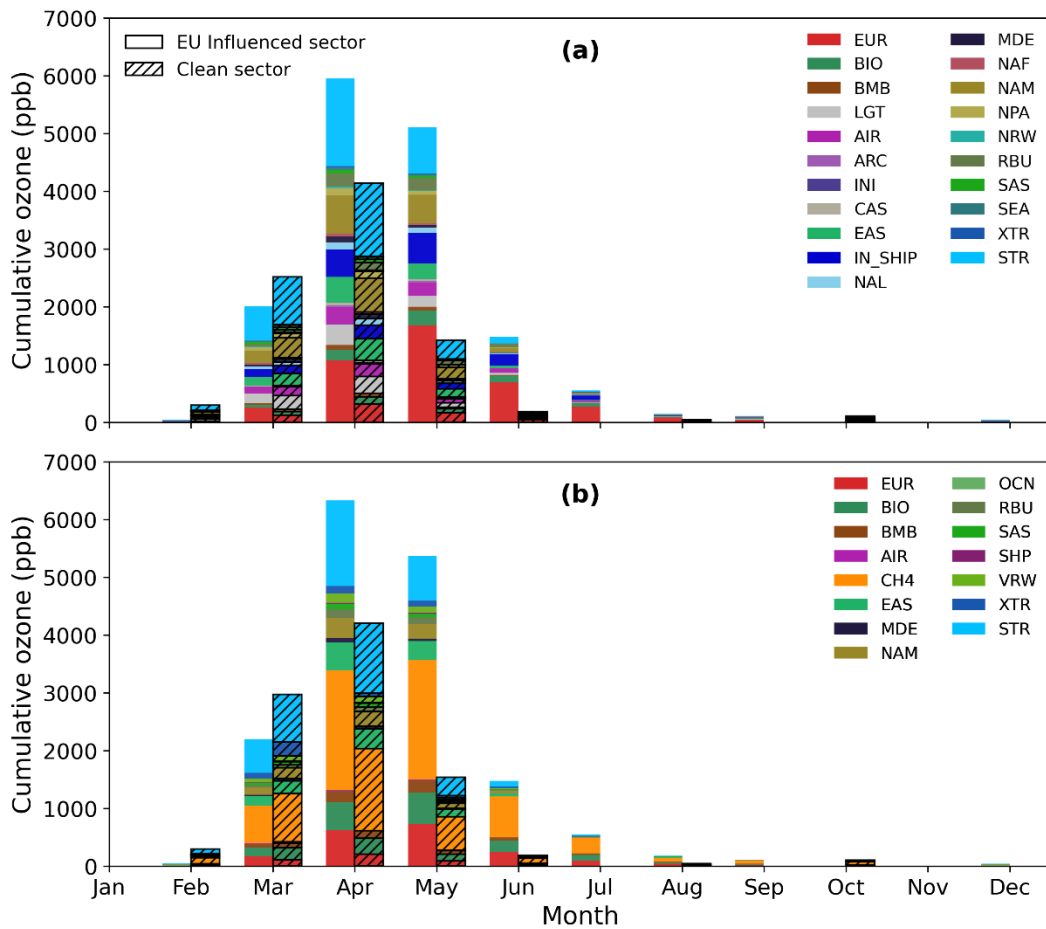
North American NO_x also contributes significantly to exceedance in both clean and EU-influenced sectors at Mace Head during March to May month, likely due to long-range transport and mixing, regional stagnation or synoptic-scale recirculation.

RC2 22

Line 551 Figure 13: Change the colour scheme to improve readability.

AR - The colour scheme is changed in the revised manuscript

Changes to manuscript:



RC2 24

Supplementary material - Section 3 is missing. Please correct the numbering and ensure figures.

AR -The supplementary material placed in corrected order in revised manuscript.