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Surface Ozone Distribution & Trends Over Ireland : Insights from long-term measurement record and source attribution modelling

Nikhil Korhale [1], Tabish Ansari [2], Tim Butler [2], Jurgita Ovadnevaite[1], Colin D.O'Dowd[1] , Liz Coleman[1]

1 School of Natural Sciences, Physics, Ryan Institute's Centre for Climate & Air Pollution Studies, University of Galway, Galway, Ireland

2 Research Institute for Sustainability - Helmholtz Centre Potsdam, Potsdam, 14467, Germany

Corresponding author :

Name - Dr. Liz Coleman

Email Id – liz.coleman@universityofgalway.ie

Abstract

Surface ozone (O_3) pollution is assessed across Ireland with a focus on long-term trends with a specific focus on the Mace Head atmospheric research station which monitors background O_3 advected into Europe via prevailing South Westerlies. Using innovative trajectory analysis, O_3 concentrations, exceedances and were identified by sectors, revealing distinct seasonal and spatial patterns. Findings show a significant rising trend in surface O_3 at Irish urban sites over the past two decades but without a similar trend at coastal sites. Highest O_3 levels and exceedances were observed at remote coastal sites, less influenced by local emissions, and heavily influenced by meteorological processes, including transboundary pollution and stratospheric intrusion. At Mace Head, springtime O_3 levels show a declining trend, with a

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rising winter-time trend. Looking only at the clean sector, the springtime decline remains significant; but without rising wintertime trends, implying the rising winter trends are a response to declining European emissions. Advanced modelling tools are used to quantify O_3 source contributions, elucidating key drivers behind the observed changes. Characteristic springtime O_3 maxima at Mace Head are attributed to stratospheric transport, influences from westerly transboundary air pollution, and lightning NO_x. Combined trend and sectoral observational analysis reveals that total spring-time concentrations are in decline, with exceedances from the UK & continental sector declining at a greater rate. This research highlights the importance of seasonal factors in air quality management across Ireland, emphasising the need for a multi-faceted approach to control O_3 levels and reduce exceedances through global and regional emission reductions.

Keywords - Meteorology, NO_x, Climate, CH_4, Emissions, VOC.

1. Introduction

Surface Ozone (O_3) has significant implications for health, vegetation, and climate. As O_3 is highly reactive, its chemical production is driven by complex photochemical processes, responding non-linearly to pollution control, creating challenges for its effective regulation. Elevated O_3 levels cause severe health issues, prolonged exposure to high O_3 levels is linked to respiratory issues, cardiovascular problems, and reduced lung function, particularly in sensitive populations such as children, the elderly, and individuals with pre-existing respiratory conditions (Lin et al., 2018; Todorović et al., 2019; Zhang et al., 2019, WHO, 2021). O_3 pollution can adversely impact vegetation by reducing agricultural productivity (Ashmore et al., 2005; Paoletti et al., 2006). O_3 is also the third most significant greenhouse gas after Carbon Dioxide (CO_2) and methane (CH_4), contributing to climate instability (IPCC, 2021). The lifetime of O_3 in the free troposphere is on the order of several weeks, and it is affected by

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large-scale atmospheric circulation patterns (Wespes et al., 2017), and meteorological factors such as temperature, solar radiation, wind speed, and atmospheric stability play a significant role in O_3 formation. (Ding et al., 2023; Khiem et al., 2010).

O_3 is formed in the atmosphere from precursors Nitrogen Oxides (NO_x), carbon monoxide (CO), and volatile organic compounds (VOCs) through photochemical reactions. The reactive, interdependent atmospheric chemistry leads to a non-linear relationship between O_3 and its precursors (Seinfeld and Pandis, 2016), and effective O_3 mitigation requires an understanding of processes influencing O_3 production and removal mechanisms (Fowler et al., 2013). NO_x can suppress or enhance O_3 formation, depending on the atmospheric chemistry regime. In polluted urban environments, high NO_x emissions can lead to O_3 dissociation, retarding formation whereas in relatively clean environments, O_3 formation is correlated with NO_x concentration (Seinfeld and Pandis, 1997;Tavella & da Silva Júnior, 2021). Seasonal and regional variations further complicate the regulation, with higher O_3 levels observed in summer across the northern hemisphere due to increased temperatures, solar radiation, and abundant precursors (Moiseenko et al., 2021; Sicard et al., 2016). In marine boundary layers, O_3 levels are generally lower than in continental regions, though specific oceanic environments can exhibit high O_3 concentrations due to inflows from polluted areas (Boylan et al., 2014; Girach et al., 2020). Another factor which influences O3 level is the North Atlantic Oscillation (NAO which influences O3 levels in Western Europe, with positive phases enhancing the transport of O3 and precursors from North America. This effect is particularly notable in southwest, central, and northern Europe (Bonaccorso et al., 2015; Creilson et al., 2003; Pausata et al., 2012). While Ireland's air quality is mostly governed by the influx of clean maritime air from the Atlantic Ocean (Tripathi et al., 2010), particular synoptic scenarios allow for the intrusion of polluted air masses from continental Europe. These events, though infrequent, can bring

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substantial amounts of ozone and its precursors (NO_x, VOCs), contributing to short-term O_3 pollution episodes.

The World Health Organisation (WHO) publishes Air Quality Guidelines (AQGs) as a non-legally binding global target for governments to achieve within their jurisdictions. These AQGs comprise evidence-based recommendations of limit values to protect public health. The current recommended AQGs for O_3 is expressed as a daily maximum of 8-hourly running average O_3 value of 100 μg/m³. Days when O_3 levels exceed the recommended AQGs are classified as exceedance days. Factors contributing to exceedances include high solar radiation, stagnant air masses, and local emissions and regional and transboundary transport of O_3 and precursors.

Over the past 150 years, there has been a 40% increase in O_3 levels owing to rising precursor emissions. (Archibald et al., 2020; Griffiths et al., 2021; Young et al., 2013). Despite European Union emission reduction policies, O_3 pollution remains a problem, with over 94% of those living in European cities exposed to O_3 levels exceeding the WHO AQGs in 2022 (EEA 2024, WHO 2021). Over 22,000 premature deaths in the EU were attributable to short-term exposure to O_3 in 2021(Soares et al., 2023).

Long-term data from the Mace Head research station in Ireland reveal seasonal peaks in O_3 during spring and lows in summer. (Derwent, 1998; Derwent et al., 1994, 2018a). Historical trends show increasing baseline O_3 levels in the 1980s and 1990s, stability in the 2000s, and a decline in the 2010s.(Derwent et al., 2013; Derwent, Manning, Simmonds, Spain, et al., 2018). Recent observational and modelling data have identified a broad O_3 maximum in spring and early summer, aligning with peak stratospheric transport (Ansari et al., 2024; Lin et al., 2012; Russo et al., 2023). O_3 dynamics are complex, and studies reveal discrepancies between model output and observations (Bessagnet et al., 2016; Vautard et al., 2012), highlighting the need for further understanding of factors governing O_3 levels and trends.

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This study investigates the distribution and trends of O_3 and its precursors across Ireland, providing valuable insights into the regional and hemispheric impact on Irish surface O_3 levels and exceedances. By analysing a long term observational dataset, this research highlights significant seasonal and temporal variations and long-term trends in O_3 concentrations. Advanced modelling results using the Tropospheric Ozone Attribution of Sources with Tagging 1.0 (TOAST 1.0) framework (Butler et al., 2018; Butler et al., 2020) were applied to determine the drivers of O_3 trends in Ireland. Additionally, trajectory analysis is used to trace the origins of air masses, revealing the impact of transboundary pollution and atmospheric transport. This integrated approach not only enhances our understanding of the drivers of O_3 concentrations, trends and exceedances over Ireland but also underscores the importance of global and regional contributions to O_3.

2. Data and methodology

2.1 Observational Network and Analysis Approach

Measurement data is obtained from the Environmental Protection Agency, Ireland (EPA) (https://eparesearch.epa.ie/safer/). The O_3 monitoring network shown in Figure 1 has been operational in Ireland since 1994. O_3 is measured using an API M400 and O_3 analyser based on UV photometry at all monitoring sites. Measurements of O_3 precursors from EPA air quality monitoring sites are also monitored. The details of measurements site are shown in table 1 . Numerous previous studies have analysed this data, with a particular focus on the analysis of Mace Head data to assess background levels of (Carslaw, 2005; Derwent, 1998; Derwent et al., 1994, 1998, 2001, 2004, 2008, 2013; Derwent, Manning, Simmonds, & Doherty, 2018; Derwent, Manning, Simmonds, Spain, et al., 2018; Oltmans et al., 2013; Simmonds et al., 2004; O. P. Tripathi et al., n.d., 2010, 2012, 2013). Additionally, the CH_4 data is obtained from the

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Integrated Carbon Observation System (ICOS) network, accessible at https://www.icos-cp.eu/data-products/ATM_NRT_CO2_CH4.

For this analysis, the observational sites were classified into three categories: Coastal, Rural, and Urban, as shown in Figure 1. The classification of the sites is based on Spohn et al., 2022, with the addition of the coastal category. Hourly data were used to evaluate annual trends based on monthly mean concentrations. Seasonal analysis conducted for the four main meteorological seasons in Ireland, namely Spring (March, April, May), Summer (June, July, August), Autumn (September, October, November), and Winter (December, January, February). O_3 exceedances were calculated based on the WHO AQGs, indicating that the maximum daily average over eight hours (MDA8) should not exceed 100 µg/m³. A significant analysis was performed on data measured at the Mace Head Atmospheric Research Station (53°33′N, 9°54′ W), which is exposed to pristine marine air masses approximately half of the time. (Grigas et al., 2017; O'Dowd et al., 2014).

Table 1. Details of Environmental Protection Agency Ireland (EPA) O_3 measurement sites over Ireland, with location information, and the data period used for the study.

Site	Data availability	Type	Latitude	Longitude
Mace head	1994-2022	Coastal	53.3253	-9.9036
Valentia	2001-2022	Coastal	51.9385	-10.24
Monaghan	1995-2022	Rural	54.0661	-6.883
Laois	2005-2022	Rural	53.1076	-7.1983
Kilkenny	2012-2022	Rural	52.6383	-7.2676
Rathmines	2002-2022	Urban	53.322	-6.2672
Clonskeagh	2008-2022	Urban	53.3118	-6.2353
Mayo Castlebar	2009-2022	Urban	53.851	-9.3003
Swords	2009-2022	Urban	53.4631	-6.2222
Wicklow Bray	2009-2022	Urban	53.1873	-6.122
Cork South link road	2014-2022	Urban	51.8785	-8.4649
Cork Bishops town	2016-2022	Urban	51.8858	-8.53321
Cork UCC	2018-2022	Urban	51.9	-8.4863

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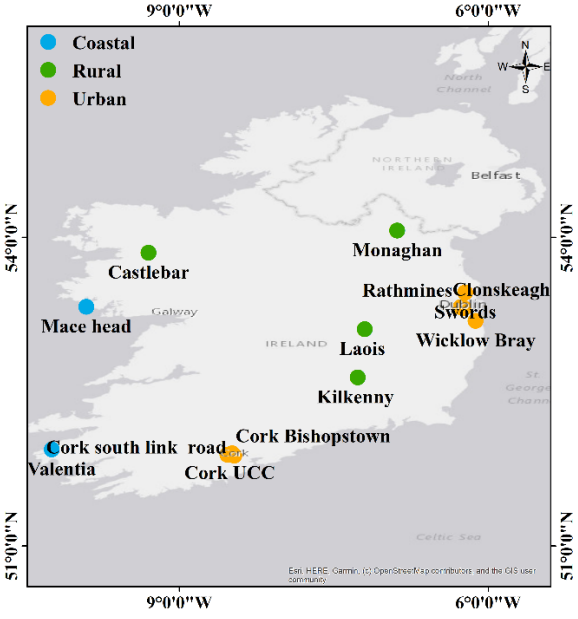

Figure 1. – The map of EPA O_3 measurement sites over Ireland with classification of backgrounds.

Trend analysis was conducted using the Openair package in R. This software tool is designed for the analysis of atmospheric composition data. Trends were determined using the Theil-Sen slope estimator and Mann-Kendall tests to quantify significance, in accordance with the Tropospheric Ozone Assessment Report (TOAR) guidelines (Lefohn et al., 2018). 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. Uncertainty or reliability of the trend is calibrated according to the p-value, as outlined by (Chang et al., 2023), consistent with the best statistical practices for analysis used in the second phase of TOAR.

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2.2 Clean air sector identification from Back trajectories

Baseline O_3 refers to the concentration of O_3 in air masses minimally influenced by local or regional anthropogenic emissions. Back-trajectory methods are widely used to estimate baseline O_3 levels by analysing the origins and transport pathways of air masses reaching observation sites. Typically, Lagrangian dispersion models are used to trace air parcels backwards in time and identify their origin.

For this study, air mass trajectories arriving at Mace Head were calculated using the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) (Draxler et al.,2003; Stein et al., 2015) in conjunction with R software. The air masses were classified into two categories the clean sector and EU-influenced sector. An air mass was considered part of the clean sector consider when air mass trajectories remained over the ocean surface for the previous 72 hours. And the remaining air mass trajectories are classified as the EU-influenced sector. Meteorological data for the analysis were derived from NOAA reanalysis data (Stunder et al., 2004). Calculations were performed for 6:00 UTC each day, with a final trajectory height of 100 meters, covering the years 2000 to 2022. The O_3 concentrations observed during the clean sector were averaged to derive baseline levels, consistent with previous studies on baseline O_3 trends and sources (Derwent et al., 2013; Oltmans et al., 2006).

2.3 CAM4-Chem Model

The CAM-Chem air quality model, part of the Community Earth System Model (CESM), simulates atmospheric chemistry and the interactions among chemical constituents, meteorology, and climate. It incorporates detailed chemical mechanisms, emission inventories, and meteorological data to simulate pollutant dispersion, thereby allowing us to determine air quality trends. CAM-Chem has been applied in numerous studies, significantly contributing to

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the understanding of regional and global atmospheric processes. (Lamarque et al., 2012; Tilmes et al., 2016). The model features a flexible chemical pre-processor to allow for detailed handling of atmospheric chemistry. Studies have demonstrated that CAM-Chem accurately represents conditions in both the troposphere. (Aghedo et al., 2011; Lamarque et al., 2010) and the stratosphere (Lamarque et al., 2008; Lamarque and Solomon, 2010), including temperature structure and dynamics (Butchart et al., 2011). Offline CAM-Chem has also been utilised in the Hemispheric Transport of Air Pollution (HTAP) assessments. (Anenberg et al., 2009; Fiore et al., 2009; Jonson et al., 2010; Shindell et al., 2008;Tan et al., 2018).

For the current study, we analyse simulations of the Community Atmospheric Model version 4 CAM4-Chem (Community Atmosphere Model version 4 with chemistry) ((Lamarque et al., 2012). The model simulations were carried out at a horizontal resolution of 1.9° × 2.5°, featuring 56 vertical levels for the 2000-2018 period, with specified dynamics derived from MERRA2 reanalysis. (Molod et al., 2015). Tagged source attribution of tropospheric ozone (TOAST 1.0) is a novel tagging methodology developed for the CESM to quantify source contributions to O_3 . Unlike traditional methods that rely on sensitivity simulations, TOAST uses an online tagging approach to track O_3 production from specific NO_x and VOC sources (e.g., anthropogenic, biogenic, biomass burning, lightning) directly within the model, allowing for efficient attribution of O_3 to regional and sectoral emissions while maintaining full chemical coupling. The tool has been validated against observations and demonstrates utility in disentangling the impacts of different emission sectors on O_3 pollution. (Butler et al., 2018, 2020; Lupaşcu et al., 2022; Nalam et al., 2024)

Global CAM4-Chem model simulations are performed for the years 2000-2018 with NO_x and VOC tagging (as described in Ansari et al., 2025; Nalam et al., 2025), with the base chemical mechanism (MOZART; Emmons et al., 2012) and source code modified to account for extra tagged species representing regional and sectoral identities. Anthropogenic emissions of NO_x,

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CO, and non-methane volatile organic compounds (NMVOCs) are incorporated from the Hemispheric Transport of Air Pollution version 3 emissions inventory. (HTAPv3; Crippa et al., 2024), which includes land-based emissions, international shipping emissions, and aircraft emissions. Biomass burning emissions are sourced from the GFED-v4 inventory (Van Der Werf et al., 2010), while biogenic NMVOC emissions are derived from CAM4-GLOB-BIO-v3.0. The O_3 source attribution technique used for this study is described in (Butler et al., 2020).

3. Results and discussions

3.1 Yearly variation of O_3

Figure 2 shows box plots, illustrating the average O_3 concentrations for 13 sites over the duration of the available dataset, as discussed in section 2.1 providing a comprehensive overview of the variability and distribution of O_3 concentration. Coastal sites, such as Mace Head, show higher O_3 levels compared to other sites, with an annual average concentration 77 µg/m³. Similarly, Valentia shows higher mean concentrations at 78 µg/m³ (in 2003). In urban areas like Rathmines, Dublin, O_3 concentrations remained consistently lower, with averages ranging from 39 to 56 µg/m³. Similarly, South link road and Bishopstown sites in Cork city, recorded relatively lower concentrations compared to coastal and rural locations, reflecting the impact of high urban NO_x emissions. Rural sites like Laois and Kilkenny showed intermediate O_3 concentrations, less influenced by urban emissions. These sites consistently show O_3 averages ranging between 50 to 57 µg/m³, with little variability, highlighting the predominant role of steady background O_3 contributions in rural sites. O_3 concentrations vary significantly with proximity to emission sources – adjacent to urban areas, O_3 levels can be lower due to titration, where O_3 reacts with NO, causing O_3 depletion, but the transport of precursors can

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cause an increase in O_3 concentration downwind of the sources (Jeon et al., 2014; Monks et al., 2015;Zhu et al., 2012)

The red line over the box shows a clear seasonal pattern in O_3 concentration for each site. With a spring-time (March-April) peak and summer-time (June-July), dip, with the highest peaks in the coastal sites, and lowest dips in urban sites, influenced by local emissions e.g. Cork South Link Road and Swords.

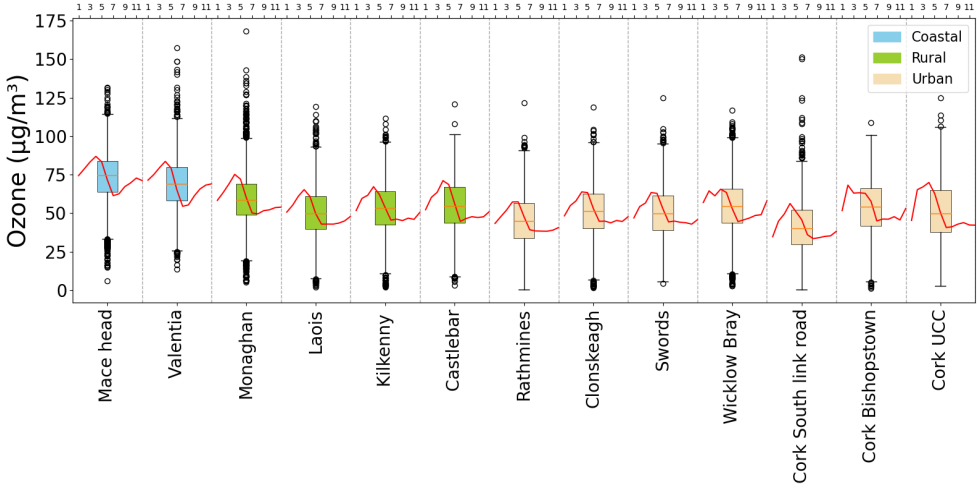

Figure 2. Annual average O_3 concentration at different sites in Ireland. In each box, the lowest whisker level represents the 5th percentile, the box spans from the 25th to the 75th percentile, the horizontal line within the box represents the median 50th percentile, and the upper whisker represents the 95th percentile. The average of monthly O_3 values calculated for the entire period of each station, and the red line shows the average monthly O_3 variation of all sites top axis shows the month (1– 12).

3.2 O_3 Trend analysis

3.2.1 Yearly trend

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Table 2. summarizes the Theil-Sen trends in O_3 concentration (in µg/m³ per year) across 13 monitoring sites in Ireland over different periods: 5 years (2018-2022), 10 years (2013-2022), 15 years (2008-2022), and the available years of data for each site. In the coastal regions, Mace Head shows a consistent decrease in O_3 levels over the 5, 10, and 15-year periods, although the entire dataset exhibits a small rising trend 0.02 µg/m³ per year. These trends are mostly in agreement with previous studies, where there was a positive trend observed in background O_3 up to the mid-2000s, which stabilised and began to decline in the 2010s (Derwent et al., 2018). Valentia shows a long-term decreasing trend of -0.23 µg/m³ per year, consistent with the previous study by Tripathi et al..2010.

In rural areas, Monaghan exhibits a declining trend in O_3 concentrations across all time periods, indicating an overall reduction. Laois shows an upward trend over the 10 and 15-year periods, though there is a slight decline in the most recent 5 years. Kilkenny presents slight negative trends over the 5 and 10-year periods (-0.29 and -0.01µg/m³ per year). Negative trends are observed in Castlebar-(0.71 and -0.05 µg/m³ per year).

The Dublin urban area sites (Rathmines, Clonskeagh, Swords) predominantly show increasing trends in O_3 levels, indicative of changes in urban pollution or local emissions, with decreased suppression of O_3 levels in urban regions due to decreased local emissions. (Derwent et al., 2024). This is consistent with the "weekend effect," as observed by (Atkinson-Palombo et al., 2006)whereby a reduction in NO_x due to reduced weekend traffic decreases O_3 removal by NO_x titration, leading to higher surface O_3 levels, likely to occur in wintertime, and in regions with low photochemical production due to low insolation such as Ireland. Mixed results are observed at the urban stations of Cork. This suggests variable factors affecting Cork O_3 levels. Coastal sites like Mace Head and Valentia generally show decreasing trends, potentially due to less local emission sources but with more significant impacts from regional and long-range

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transport, However, a detailed analysis of the trends requires consideration of seasonal effects.

Table 2. - Trends in surface O3 concentration (µg/m³ per year) calculated for 13 sites in Ireland over different periods over the complete dataset: 5 years (2018-2022), 10 years (2013-2022), 15 years (2008-2022), and the available measurement record for the site. The p-value evaluates the reliability of the trend, whereas a lower p-value indicates trend certainty. Adopting the trend reliability scale defined for TOAR-II studies (Chang et al., 2023), trends with very high certainty will be marked by ***($p \leq 0.001$), trends with high certainty with ** ($p \leq 0.01$), and low to medium certainty with *($p \leq 0.05$).

Site No	Site name (Classification)	Measurement Record	Trend over record µg/m³ per year	5-year trend 2018-2022 µg/m³ per year	10-year trend 2013-2022 µg/m³ per year	15-year trend 2008-2022 µg/m³ per year
1	Mace Head (C)	1994-2022	0.02	-0.25	-0.31*	-0.11
2	Valentia (C)	2001-2022	-0.23***	-1.15***	-0.84***	-0.32**
3	Monaghan (R)	1995-2022	-0.19***	-0.74**	-0.35*	-0.09*
4	Laois (R)	2005-2022	0.39***	-0.15	0.3**	0.46***
5	Kilkenny (R)	2012-2022	0.02	-0.29	-0.01	
6	Castlebar (R)	2009-2022	0.18*	-0.71*	-0.05	
7	Rathmines (U)	2002-2022	0.27***	1.72***	1.15***	0.48***
8	Clonskeagh (U)	2008-2022	0.33***	0.97**	0.12	0.33***
9	Swords (U)	2009-2022	0.6***	0.07	0.33***	
10	Wicklow Bray (U)	2009-2022	0.14*	0.04		
11	Cork South-link Road (U)	2014-2022	0.51*	-0.44		
12	Cork Bishopstown (U)	2016-2022	1.05**	-1.81*		
13	Cork UCC (U)	2018-2022	-0.94	-0.94		

3.2.2 Monthly trend

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Figure 3. shows the monthly trend for 10 years from the period 2012-2022. Mace Head (coastal) and Monaghan (rural) sites predominantly show a rising trend in winter/early spring , with a decreasing trend in late spring to summer. Valentia shows a decreasing trend in every month except February when levels are significantly impacted by long-range transport and stratospheric sources (Auvray and Bey, 2005; Pan et al., 2018). Urban sites show a general increasing trend, as yearly trend but with a seasonal signal in Clonskeagh an increase in winter-spring and a decrease in late spring or summer. Seasonal trends of the 15-year dataset are supplied in supplementary figure S2, where coastal stations exhibit a pronounced increase in early spring and a decrease in late summer, with a consistent near-year-round increase in urban stations of Rathmines and Laois.

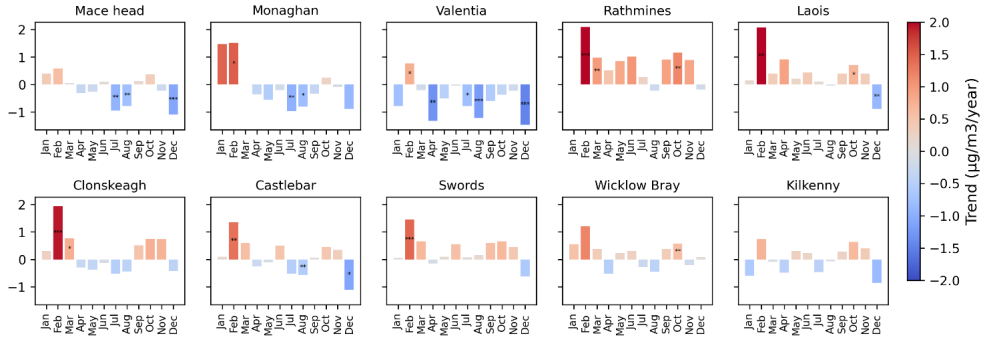

Figure 3. Monthly trend analysis of O_3 at different sites for 10 year period. (2012-2022) Adopting the trend reliability scale defined for TOAR-II studies (Chang et al., 2023), trends with very high certainty are marked by ***($p \leq 0.001$), , trends with high certainty with **($p \leq 0.01$), and low to medium certainty with **($p \leq 0.05$). Positive trends are in red shade and negative trends are in blue shade.

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3.3. O_3 Exceedance

The O_3 exceedance are identified according WHO criteria and the results shown in figure 4. It shows the monthly O_3 exceedances at 13 sites in Ireland over the available measurement dataset. The highest and lowest numbers of O_3 exceedances were observed at Mace Head and Rathmines, representing coastal and urban sites, respectively. Most exceedances occurred in spring when O_3 concentrations were at a maximum.

Most sites recorded elevated spring-time occurrence in exceedances. E.g. Rathmines had its highest number of exceedances in April 2019, while Laois reached a peak of 13 exceedances in May 2017. Castlebar and Swords show increased exceedance occurrences in spring and early summer, particularly notable spikes occurring in 2010, 2013, 2016, and 2019. Conversely, Wicklow Bray exhibited a different pattern, showing significant spikes in February and March 2022, alongside occasional exceedances during March, April, and May, for example, in 2012 and 2018. Cork South link Road also recorded exceedances, particularly in March and June, with significant spikes in 2018 and 2019. Cork Bishoptown shows exceedances, especially in February and March 2019, while Cork UCC experienced spikes, particularly in April and May 2019. Kilkenny consistently exhibited exceedances during spring and summer, with April and May often recording the highest number, particularly in 2019. This highlights the impact of seasonal atmospheric conditions on O_3 levels. It is noted that summertime exceedances, although less frequent in occurrence, indicate significant photochemical production that would be required to elevate O_3 levels from the annual dip in the seasonal cycle to exceed the WHO AQG threshold. These episodic spikes are characteristic of unique climatic or pollution events and warrant further study.

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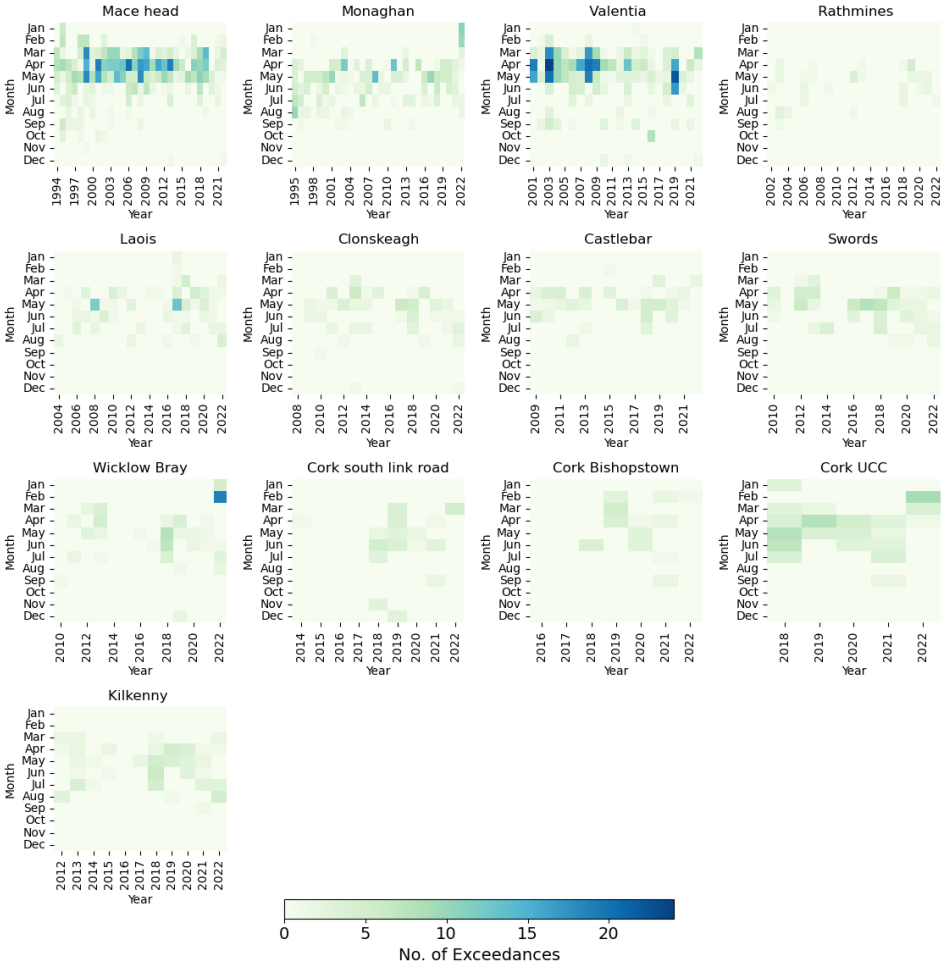

Figure 4. - Monthly O_3 exceedance at different sites in Ireland.

Figure 5 depicts trends in NO_2 and CH_4 concentrations across various Irish measurement sites. Most monitored sites exhibit a decreasing trend in NO_2 concentrations because of pollution control on transportation, industrial activities, and energy production in the EU and North America, in line with previous studies (Coleman et al., 2013; Donlon et al., 2024).In contrast, CH_4 levels at three sites Mace Head, Malin Head, and Carnsore Point indicate a significant and persistent rise in CH_4 concentrations. Mace Head, known for its clean Atlantic air. Malin Head, situated at Ireland's northern tip near the UK border, offers a unique position to observe both

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clean marine air and transboundary pollution whereas Carnsore Point in the southeast, is capture air masses from both the UK and mainland Europe, Carnsore Point receives the majority of air masses from the land (Spohn et al., 2022). These NO_2 and CH_4 trends reveal a dual dynamic: while NO_2 levels are decreasing due to effective emission controls, CH_4 levels are rising unabated, highlighting the need for enhanced mitigation strategies targeting CH_4.

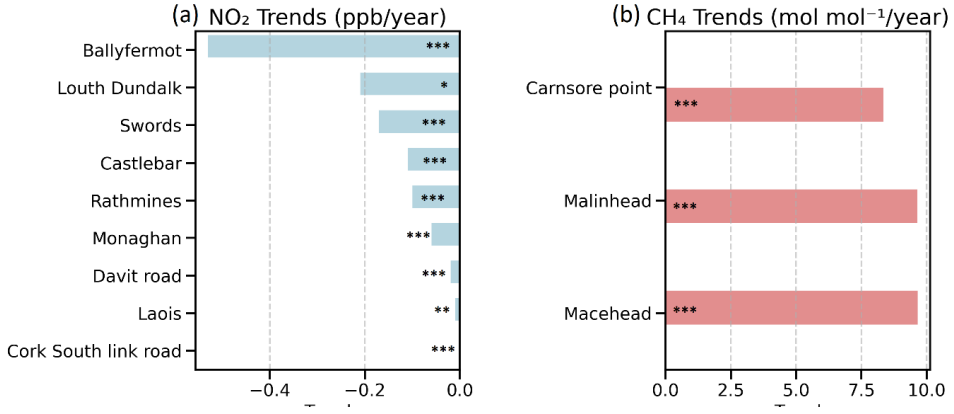

Figure 5.- Trend in O_3 precursors NO_2 (a), and CH_4 (b) at different sites. Trends with very high certainty are marked by ***($p \leq 0.001$), , trends with high certainty with **($p \leq 0.01$),, and low to medium certainty with *($p \leq 0.05$).

To evaluate the relationship between NO_x and O_3 concentrations in an Irish context and the potential benefit of abrupt enforcement of NO_x control measures, we assess the impact of the COVID-19 2020 lockdown, Spring 2020, whereby the lockdown period saw a prominent relative decrease in NO_2, yet an increase in surface O_3 compared to average measurements for the same months 2017-2019 in most national monitoring stations (Figure 6). The negative correlation between O_3 and NO_2 is indicative of a NO_x-saturated regime, normally associated with polluted urban environments and NO_x titration events. Similar results are discussed by (Spohn et al., 2022) with meteorology. This effect was widely observed during the COVID

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lockdown (Ordóñez et al., 2020; Tavella & da Silva Júnior, 2021; C. Zhang & Stevenson, 2022). Significant enhancement of O_3 occurs at the inland measurement sites, despite a 2020 spring-time 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)

The atmospheric conditions in Ireland do not align with the interpretation of the atmosphere as being either a NO_x-controlled regime for clean environments or a NO_x-saturated regime in polluted environments. The negative correlation between NO_x and O_3, due to NO_x titration, observed in Ireland occurs under relatively clean atmospheric conditions, but it is consistent with low-insolation conditions, which are characteristic of Irish meteorology and frequent cloud cover ((Pall E E and Butler, n.d.)

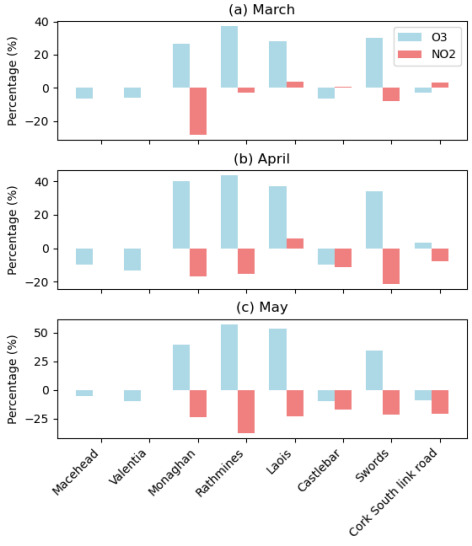

Figure 6.- Percentage change in NO_2 and O_3 during the lockdown period of 2020 as compared to the 2017-2019 average at different sites in Ireland for (a) March (b) April (c) May Month.

3.4 Model and Observations Comparison

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3.4.1 Comparison between CAM4 – Chem Model and Observations

Global simulations were performed with the CAM4-Chem model enabled with source tagging (Butler et al., 2018) for 2000-2018 and the modelled O_3 over Ireland was compared with surface O_3 measurements at five sites. Figure 7 shows the comparison of monthly O_3 CAM4-Chem and ground station O_3 data. From this figure, it is observed that CAM4-Chem exhibits negative (positive) bias in rural and coastal (urban) sites. The underestimation at Mace Head is probably caused by the coarse grid resolution, covering a large area not representative of Mace Head conditions. The influence of coastal meteorology also leads to an underestimation of O_3 (Yerramilli et al 2012). Coastal meteorology, including cool sea surface temperatures and persistent clouds, suppresses O_3 formation. McVeigh et al., 2010 explained this through eddy correlation measurements showing downward ozone fluxes over coastal waters west of Ireland. The dry deposition rate over land would exceed that over the ocean leading to a lower simulated O_3 concentration for the entire grid cell. Dry deposition is enhanced by solar radiation (Coleman et al.,2012; Coleman et al., 2013; Pio et al., 2000) hence model measurement discrepancy is at a maximum in late summer months. Overestimation of O_3 in Clonskeagh and Cork South link Road is likely due to coarse grid handling of localised emissions and subsequent atmospheric chemistry.

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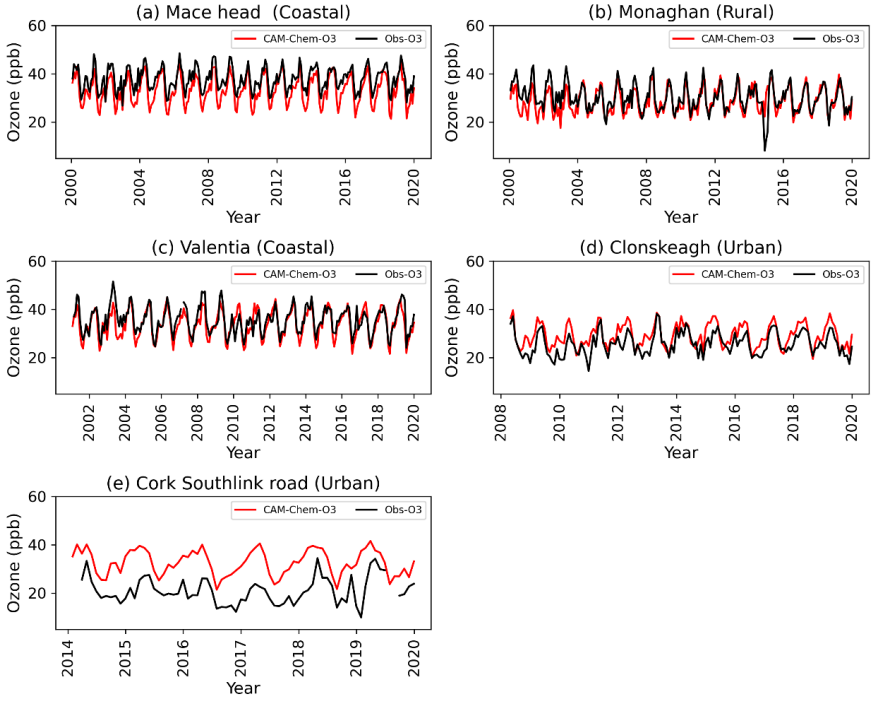

Figure 7. - The comparison of Monthly CAM4 – Chem O_3 and Monthly O_3 observations at five sites in Ireland**.**

At Mace Head, the model shows a negative mean bias of - 4.42 (-11.68% normalized mean bias) but strong correlation (r =0.83). In Monaghan and Valentia, the model shows smaller biases of −1.43 and −1.54, normalized mean biases of −4.74% and −1.54%, and correlation coefficients of 0.73 and 0.72, respectively. These high correlations are in line with (Tilmes et al., 2015). However, at Clonskeagh and Cork South Link Road, the model overestimates, with positive biases (3.38 and 11.98) and weaker correlations (0.68 and 0.49). Statistics are discussed in more detail in the supplementary material table S1. These results suggest better model performance at coastal/rural sites and greater discrepancies in urban areas as expected at this model resolution.

3.4.2 Source attribution using CAM4-Chem

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To quantify the contribution of various precursor emission sources to modelled O_3 concentrations, the TOAST1.0 dual NO_x and VOC tagging technique is utilised as described in (Butler et al., 2018). This allows us to attribute the modelled O_3 to the emissions of NO_x and VOC precursors across different source sectors and regions as listed in Table 3. The NO_x and VOC precursor emissions across different source sectors and regions, as shown in the Table, are responsible for the attribution of the modelled O_3.

Table 3. - List of tags used in NO_x and VOC tagging.

Regional Land-Based Tags		**Regional Oceanic Tags**		**Global Sector/Process-Based Tags**	
ARC	Arctic	**NAL**	North Atlantic	**AIR**	Aircraft
CAS	Central Asia	**ENA**	Eastern North Atlantic	**BIO**	Biogenic
EAS	East Asia	**NAE**	North America East Coast	**BMB**	Biomass Burning
EUR	Europe	**NAW**	North American West Coast	**LGT**	Lightning
MCA	Mexico & Central America	**NPA**	North Pacific	**STR**	Stratospheric Intrusion
MDE	Middle East	**BNS**	Baltic and North Seas	**XTR**	Extra untagged O_3
NAF	North Africa	**HBY**	Hudson Bay	**CH4**	Methane
NAM	North America	**IDO**	Indian Ocean	**OCN**	Oceanic Sources (DMS)
RBU	Russia-Belarus-Ukraine	**MBC**	Mediterranean, Black, and Caspian Seas	**SHP**	Shipping
SAS	South Asia	**SHO**	Southern hemispheric oceans	**AIR**	Aircraft
SEA	Southeast Asia			**INI**	Initial Condition O_3
VRW	Rest of the World				

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The monthly tagged major precursor contributions to surface O_3 at Mace Head, averaged over the 2000-2018 simulation period, are shown in Figure 8. The stratospheric source of O_3 dominates in Winter-Spring, contributing to the spring-time maxima due to vigorous stratospheric transport. European NO_x emissions contribution peaks in May, while lightning NO_x has the greatest impact in winter. North American emissions contribute 3.5 to 5.25 ppb, peaking in April, and aviation emissions contribute 1 to 3 ppb, with the highest contributions in winter and spring. Biogenic NO_x, significant between June and October, contributes an average of 3.6 ppb, with higher contributions during August and September. Biogenic VOC sources contribute slightly more, averaging over 4 ppb during late autumn and maintaining a more sustained contribution throughout the year. East Asian NO_x emissions, contributing up to 3.6 ppb, show a minimum contribution in July and August. North Atlantic shipping NO_x (NAL) accounts for up to 2.4 ppb of O_3 during July month. The total shipping NOx (SHIP) also contributes significantly and shows the highest contribution in June month.

Methane (CH_4) is the dominant reactive carbon molecule contributing to O_3 formation. VOC emissions from biomass burning also play a measurable role, contributing 1 to 2 ppb, with their largest contributions in August and September. Finally, European VOC emissions contribute 1 to 3 ppb, with the largest impact from March to May, coinciding with the spring-time peak in surface O_3. These findings allow quantification of specific sources amidst the complex interplay of regional and global sources in driving seasonal variations in surface O_3 levels over the Irish domain, highlighting the roles of stratospheric processes, anthropogenic emissions, biogenic sources, and lower-latitude contributions in shaping the observed patterns at background monitoring sites such as Mace Head.

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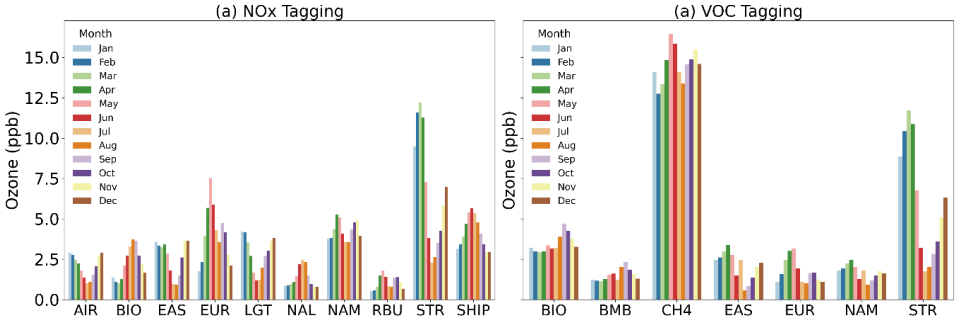

Figure 8.- Absolute contribution of major NO_x sources (a) (NO_x Tagging) and VOC source (b) (NO_x Tagging) to the CAM4-Chem simulated surface O_3 for the Mace Head grid cell between 2000-2018.

Figure 9 shows the monthly changes in contributions to surface O_3 at Mace Head over the simulation period (2000-2018). A negative (blue) trend indicates that the contribution of the source to simulated surface O_3 in this grid cell has declined over the simulation period, whereas a positive trend (red) indicates the contribution to surface O_3 has risen. Figure 9 (a) indicates that the amount of simulated O_3 at Mace Head originating from European or North American NO_x decreases during the simulation period, consistent with EU & North American emission reductions over the period (, (Guerreiro et al., 2014;US EPA 2027), with more significant reduction occurring in late Spring through late summer, when EU NO_x contributions are most significant to Mace Head O_3 concentrations (as seen in Figure 8).

There is a rising trend in simulated surface O_3 originating from NOx emissions from global aviation, from East Asia, and to a lesser extent from South Asia, which is more pronounced in the wintertime. This seasonality in source contributions explains the observed reduction in spring-time maxima and increase in winter-time levels from the measurement record. East-Asian and South-Asian VOCs also contribute to a rising trend in simulated O_3, with a more pronounced increase in winter and spring. This highlights a different pattern in hemispheric O_3 contributions, where emission reductions in Europe and North America are accompanied by

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increased influence from lower latitudes. This increasing contribution could become a more important source of background O_3 in the future. The contribution of CH_4 also has a positive trend over the simulation period, but the CH_4 trend has a reliable correlation only in the December and spring periods, with no observed trend during the summer months when atmospheric CH_4 trends have very low certainty (correlation coefficient, $p>0.33$) consistent with decline in local NOx emissions in Europe. The Anthropogenic VOC contributions from Europe (EUR) and North America (NAM) show a negative trend for all months.

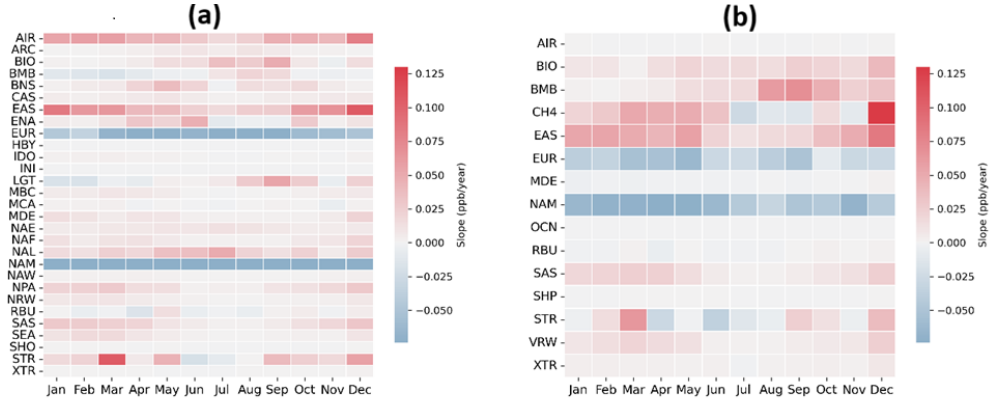

Figure 9. Trends in contributions to monthly average modelled Mace Head grid cell surface O_3 at for the 2000-2018 period derived from (a) NO_x tagging and (b)VOC tagging.

Table 4. shows the overall trend in the main contributors to NO_x and VOC tagging. It is observed that there is an increase in simulated surface O_3 originating from NO_x contributions, from aviation and East Asia, while there is a decrease in European (EUR) and North American (NAM) NO_x contributions. In VOC tagging, Methane (CH_4) and East Asian anthropogenic VOC (EAS) contribute to a rising trend over the simulation period, whereas anthropogenic VOC contributions from Europe (EUR) and North America (NAM) show a negative trend.

Table 4 - Overall Trend in contributions to Mace Head grid cell O_3 simulated by CAM4-Chem for NO_x tagging and VOC tagging over the simulation period in units of ppb per year. The

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trend with very high certainty is marked by ***($p \leq 0.001$), , trends with high certainty with **($p \leq 0.01$), and low to medium certainty with *($p \leq 0.05$).

NO_x Tagging		VOC Tagging	
	Slope (ppb/year)		Slope (ppb/year)
AIR	0.0467^{***}	CH_4	0.0590^{***}
EAS	0.0491^{***}	EAS	0.0333^{***}
EUR	-0.0900^{***}	EUR	-0.0553^{***}
NAM	-0.1243^{***}	NAM	-0.0670^{***}

3.5 O_3 Trends in Background and EU influenced sector Airmasses at Mace Head

Although Mace Head is classified as a global background site, quantification of the baseline pollution levels requires filtering the data to limit the data to that arriving from the clean sector. Based on the trajectories filtering of Mace head O_3 measurement data discussed in section 2.5. From figure 10 O_3 observations shows clean sector consistently has higher O_3 concentrations than the EU influenced sector especially during the annual spring-time high, indicating a longer lifetime for O_3 over the North Atlantic, and the land mass and pollution sources are acting as a sink for O_3 in the Irish context (Fowler et., al 2008). A decreasing trend in spring-time levels is observed for both clean and EU influenced sectors, consistent with the decrease in precursor emissions in Europe and North America. There is a significant difference between clean, and EU influenced sector measurements, the influence of EU influenced sector air to scavenge O_3 via NO_x titration, leading to higher O_3 in clean-sector air masses, as also found in previous studies (Coleman et al., 2013).An increasing trend is observed in the winter-time EU influenced sector, which is not observed in the clean sector. This would infer a decrease in winter-time O_3 depletion events due to decreasing European emissions (from the EU influenced sector), consistent with the conclusions from previous studies of Mace Head surface O_3 (Derwent et al., 2024). Summer-time values do not exhibit a notable trend or a discrepancy between clean

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and EU influenced sector measurements, indicating that there is little O_3 advected into Europe from the west in the summer months. Autumn values show higher O_3 in the clean sector, but without a significant slope.

The model results indicate that the clean sector consistently exhibits higher O_3 concentrations than the EU-influenced sector, except during the summer season. In the winter season, a significant increasing trend is observed for both sectors, which aligns well with the O_3 observations. A decreasing trend is observed during the summer season, consistent with the observations for both sectors. In spring and autumn, a positive trend is observed.

Trends in contributors of model O_3 during the different seasons for the clean and EU-influenced sector are shown in Tables S3 to S4 in the supplementary material. Aviation and East Asian NO_x shows consistently positive and significant trends in both sectors, while North America NO_x shows strong negative trends throughout the year in the clean sector. For the EU-influenced sector in NO_x tagged (Table S2), similar positive trends observed for aviation and East Asian, with North America NO_x remaining negative and European NO_x showing more significant declines in spring and winter. In case of VOC tagged O_3, the East Asian VOCs shows an increasing trend and North America VOCs negative across all seasons in both sectors (Table S3 and S4). European VOCs also show a consistent negative trend, particularly strong in the EU-influenced sector. Methane trends are seasonally positive, especially in spring and winter.

It is observed that the model consistently simulates O_3 at lower concentrations than that observed at Mace Head. This is not surprising, considering the coarse resolution of the model, which limits its ability to represent fine-scale processes and dry deposition accurately. Dry deposition is typically higher over land, and the grid cell covering Mace Head includes land area, as shown in Figure S5 of the supplementary material. Further, as explained by Fiore et al.

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(2009), models average the landscape characteristics within a grid cell, which can enhance O_3 deposition and result in lower simulated O_3 concentrations; hence, the discrepancy is more pronounced in the clean sector data.

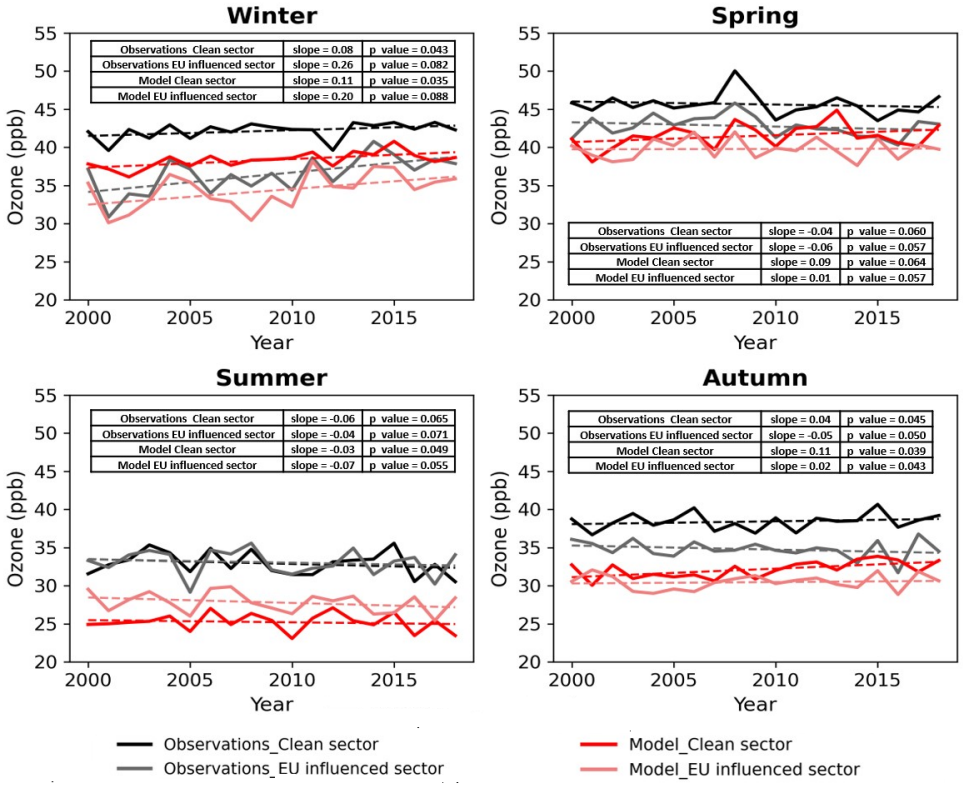

Figure 10. - Trend in seasonal Average of observed O_3 (black) and Model O_3 (red) at Mace head, separated into clean sector and EU-influenced sector.

3.6 Exceedances from the clean and EU-influenced sector at Mace Head

Exceedances observed at Mace Head between 2000 and 2022 are separated into clean and EU-influenced sectors based on trajectory air masses and shown in Figure 11. 33% of all exceedances for this period occurred in clean air masses, the remainder occurring when O_3-EU

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influenced sector air is advected over Ireland and local land masses to enhance surface O_3, which is already elevated at Mace Head compared to inland and urban sites.

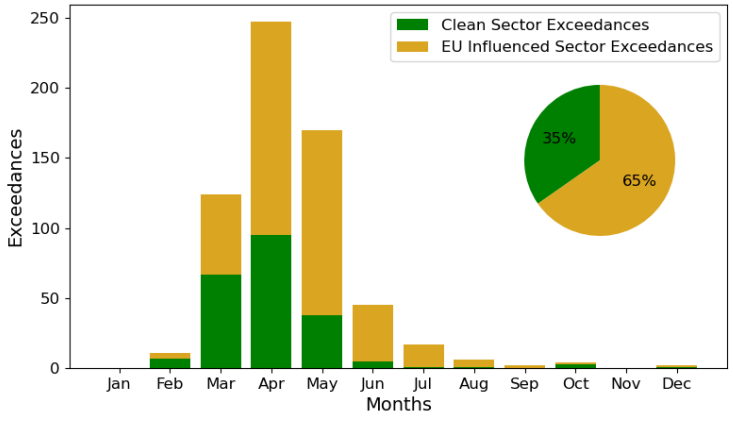

Figure 11. - Exceedances measured at Mace Head per month from 2000 until 2022, during the clean air sector(green) and EU influenced sector (yellow). The percentage of both to total exceedances is shown in the inlay.

Figure 12 shows the trend in spring-time exceedances and the 95th percentile Spring-time O_3 measured at Mace Head between 2000 and 2022. A decreasing trend in exceedances and clean sector spring-time exceedances is observed, with a greater decreasing trend in the total number of exceedances. This indicates that the changes that are driving the reduction in the exceedances in Europe are coming into effect at a quicker rate than the changes that are driving O_3 event reduction over the North Atlantic. The trends in the exceedance counts are not significant, according to the criteria in Chang et al., 2023, but there is a statistically significant decreasing trend in the 95th percentile springtime surface O_3 over the measurement record. Figure 12 (b) shows the trend in spring-time surface O_3 measured at Mace Head segregated into Clean and EU-influenced sector. The trend is more significant both in magnitude and statistical certainty for the EU-influenced sector, indicating EU emission changes having a more pronounced effect

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on spring-time O_3 measured at Mace Head O_3 as compared to changes affecting O_3 transported or formed over the North Atlantic.

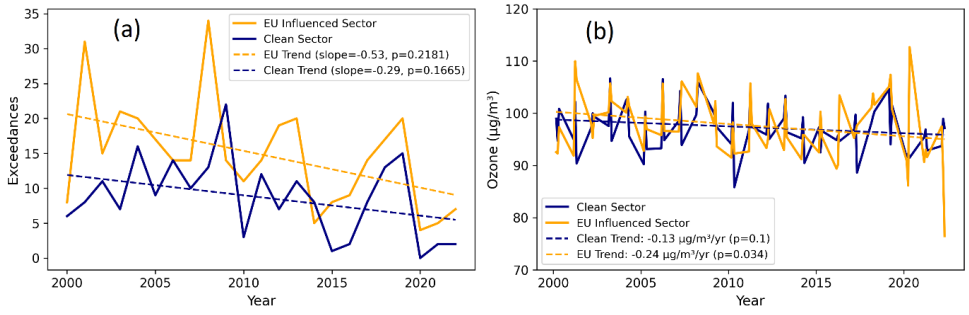

Figure 12. (a) The trend in Spring-time exceedances measured at Mace Head between 2000 and 2022 (blue) with the clean-air exceedances (gold), and (b) The trend in 95th percentile of spring (Mar- May) O_3 measured in µg /m^3 for the clean sector (blue) and the EU-influenced sector (gold).

Figure 13 shows monthly cumulative contributions to simulated O_3 concentrations within the Mace Head grid cell for NO_x and VOC tagging during O_3 exceedance, which is observed from O_3 observations calculated as discussed in section 3.3. In addition, these exceedances are categorised into clean and EU-influenced sectors. The maximum exceedances are observed in March to May month. From Figure 13 (a), it is clear that stratospheric intrusion, North American NO_x, European NO_x, and East Asian NO_x are the major contributors driving exceedances at Mace Head during the spring months (March to May). Among these, European emissions dominate the supply of NO_x precursors in April, reaching their peak in May. Figure 13 (b) shows that CH_4 is the most dominant source, followed by stratospheric intrusion and Biomass burning. North American and European VOC emissions also contribute significantly to O_3 formation during this period. Collectively, these findings highlight the complex interplay of regional and global sources in driving surface O_3 exceedances over the Irish domain.

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The cumulative O_3 contributions to EU-influenced sector and clean sector exceedances for NO_x tagging and VOC tagging, it is clear that the North American NO_x also contributes significantly to exceedance in both clean and EU-influenced sectors at Mace Head during March to May months. It may be due to transport and mixing, regional stagnation or synoptic-scale recirculation. In the case of VOC tagging, stratospheric intrusion, and CH_4 show notable contributions. Biomass burning, East Asian emissions and North American VOC emissions also play a role in O_3 exceedances.

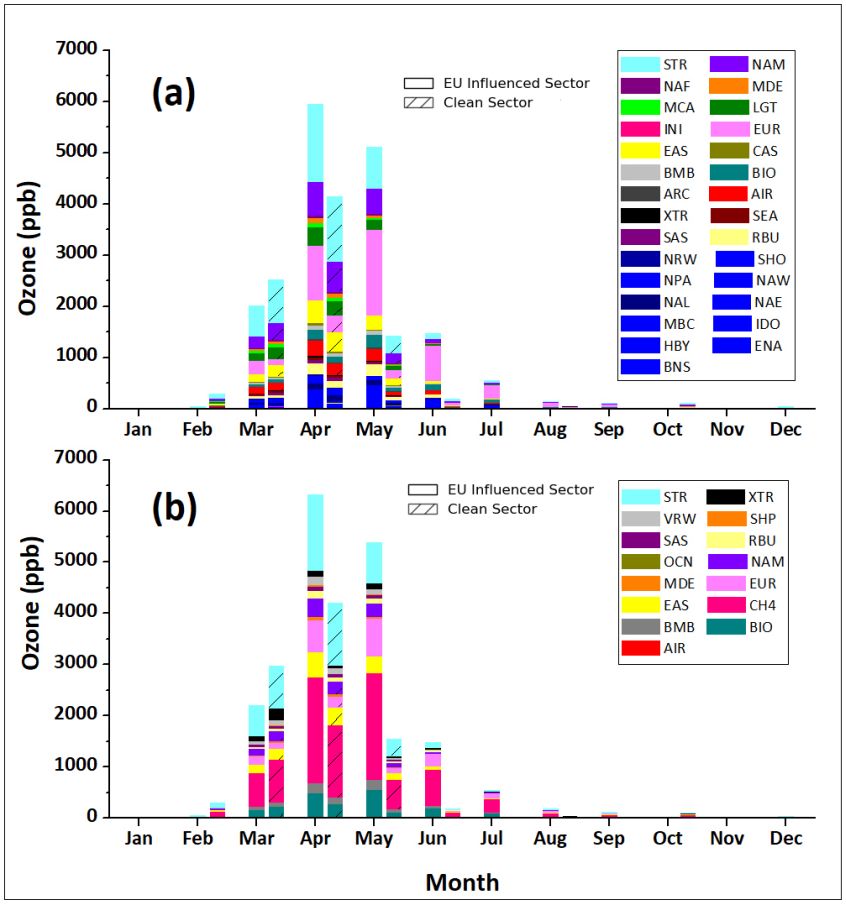

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Figure 13 - Monthly cumulative Mace Head grid cell O_3 contributions to EU influenced sector and clean sector exceedances (a) NO_x tagging and (b) VOC tagging Mace Head grid cell.

4. Conclusion

This study highlights the complexities of O_3 pollution in Ireland, revealing that coastal areas experience higher O_3 concentrations than rural and urban environments, attributed to the effect of transboundary pollution and stratospheric intrusion. Over the last two decades, urban sites have shown a significant increasing trend in O_3 levels, likely influenced by decreasing nitrogen oxides (NO_x) in Europe, including Ireland, and North America. The analysis also points out that exceedances at coastal monitoring sites correlate with years of higher spring maxima, driven mainly by hemispheric transport and stratospheric influences(16%). Utilising the advanced capabilities of the CAM4-Chem model with dual NO_x and VOC tagging, we identified key factors affecting seasonal O_3 variations, such as the spring-time peak and summer dip, driven by a mix of stratospheric intrusion, hemispheric transport, and regional emissions. Trend analysis from simulation results identified East Asian and aviation emissions as significant contributors to the rising winter trends in O_3, while reductions in North American and European emissions accounted for the decrease in spring peaks. This study provides a comprehensive understanding of the various factors affecting O_3 levels in Ireland, offering important insights for the development of O_3 pollution control policies.

Data availability

All data are available upon request.

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Author contributions

LC designed the study. NK analyzed the data and wrote the manuscript. TA and TB provided CAM-Chem model results and reviewed the manuscript.. JO and CD reviewed the manuscript and edited it.LC edited it with contributions from all coauthors

Competing interests

The authors declare that at least one of the authors sits on the editorial board of ACP.

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Figure Captions

Figure 1. The map of EPA O_3 measurement sites over Ireland with classification of backgrounds.

Figure 2. Annual average O_3 concentration at different sites in Ireland. In each box, the lowest whisker level represents the 5th percentile, the box spans from the 25th to the 75th percentile, the horizontal line within the box represents the median 50th percentile, and the upper whisker represents the 95th percentile. The average of monthly O_3 values calculated for the entire period of each station, and the red line shows the average monthly O_3 variation of all sites top axis shows the month (1– 12).

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 with very high certainty are marked by ***($p \leq 0.001$), , trends with high certainty with **($p \leq 0.01$), and low to medium certainty with **($p \leq 0.05$). Positive trends are in red shade and negative trends are in blue shade.

Figure 5.- Trend in O_3 precursors NO_2 (a), and CH4 (b) at different sites. Trends with very high certainty are marked by ***($p \leq 0.001$), , trends with high certainty with **($p \leq 0.01$),, and low to medium certainty with *($p \leq 0.05$).

Figure 6. Percentage change in NO_2 and O_3 during the lockdown period of 2020 as compared to the 2017-2019 average at different sites in Ireland for (a) March (b) April (c) May Month.

Figure 7. The comparison of Monthly CAM4 – Chem O_3 and Monthly O_3 observations at five sites in Ireland.

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Figure 8. Absolute contribution of major NOx sources (a) (NOx Tagging) and VOC source (b) (NOx Tagging) to the CAM4-Chem simulated surface O_3 for the Mace Head grid cell between 2000-2018.

Figure 9. Trends in contributions to monthly average modelled Mace Head grid cell surface O3 at for the 2000-2018 period derived from (a) NO_x tagging and (b)VOC tagging.

Figure 10. Trend in seasonal Average of observed O3 (black) and Model O_3 (red) at Mace head, separated into clean sector and EU-influenced sector.

Figure 11. Exceedances measured at Mace Head per month from 2000 until 2022, during the clean air sector(green) and EU influenced sector (yellow). The percentage of both to total exceedances is shown in the inlay.

Figure 12. (a) The trend in Spring-time exceedances measured at Mace Head between 2000 and 2022 (blue) with the clean-air exceedances (gold), and (b) The trend in 95th percentile of spring (Mar- May) O_3 measured in µg /m^3 for the clean sector (blue) and the EU-influenced sector (gold).

Figure 13 Monthly cumulative Mace Head grid cell O_3 contributions to EU influenced sector and clean sector exceedances (a) NOx tagging and (b) VOC tagging Mace Head grid cell.