Supplementary material

1. Monthly variation in O3

The monthly variation in O3 levels was calculated using a five-year average to understand whether any significant changes were observed during this period. It was observed that O3 levels at all sites were highest during April and May, which correspond to the spring season. This seasonal peak is attributed to increased sunlight and heightened photochemical activity during this time. Following the spring months, O3 levels began to decrease as summer progressed.

Coastal sites, such as Mace Head and Valentia, showed higher monthly variations compared to other sites. Additionally, a noticeable reduction in O3 levels was observed over the five years. This decline was particularly pronounced during July, with a similar trend evident at the Valentia site. Across all sites, the pattern of monthly O3 variation was consistent, highlighting higher concentrations in spring and lower levels during July and August.

The characteristic spring-time high and summer-time low is apparent in all stations, owing to the favourable conditions in springtime for hemispheric transport, stratospheric in-flux and photochemical production, whereas the summer, with elevated temperature and insolation favouring photochemical destruction and prevailing westerlies and lower windspeeds leading to less transported O3 arriving at Ireland, The seasonal cycle is more pronounced in clean and rural stations due to lack of influence of local emissions. Looking at the 5-yearly averages, there appears to be a decrease in the spring and summer values with time in some of the clean sites (Mace Head, Monaghan, Valentia), and a general increase with time in Rathmines. Trends will be discussed in the subsequent section

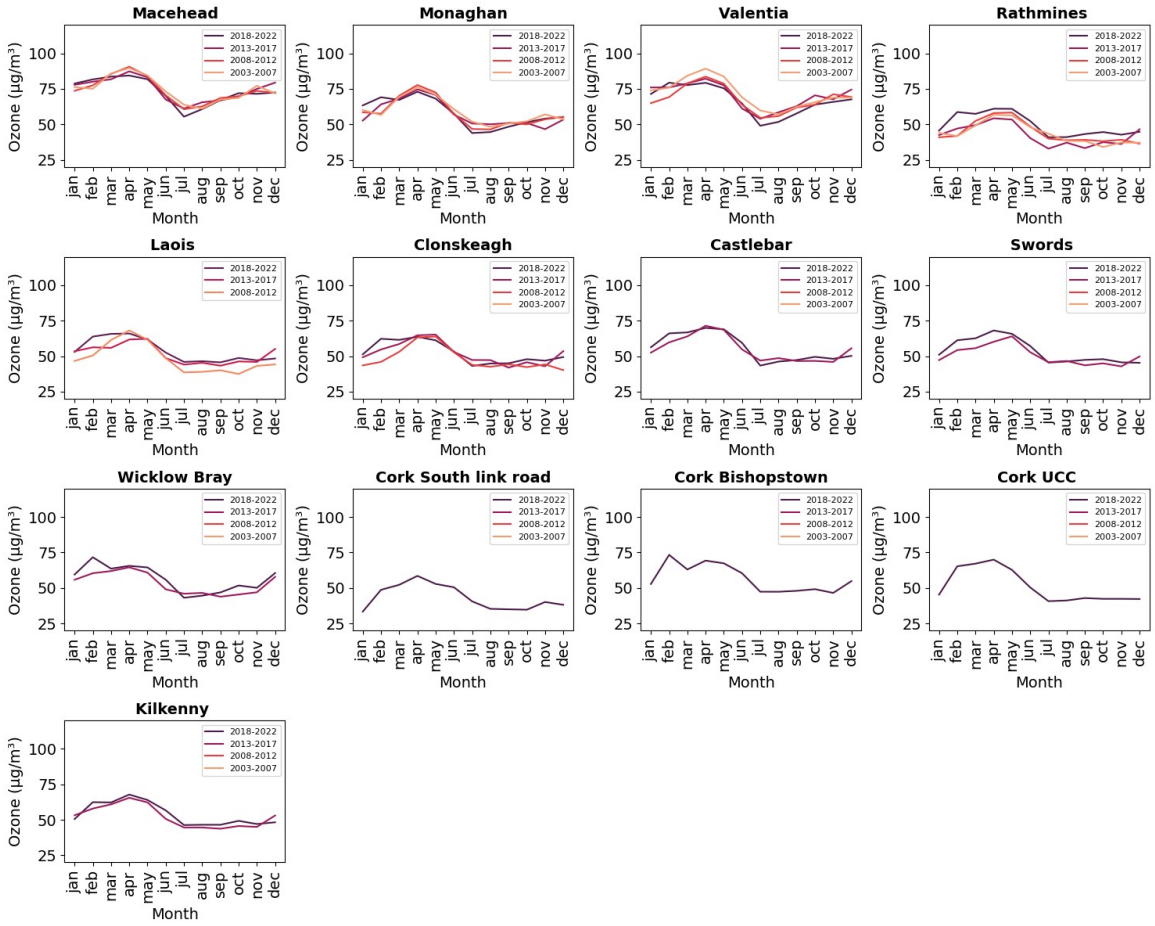

Figure S1 Five-year monthly variation in O3 concentration at different sites in Ireland.

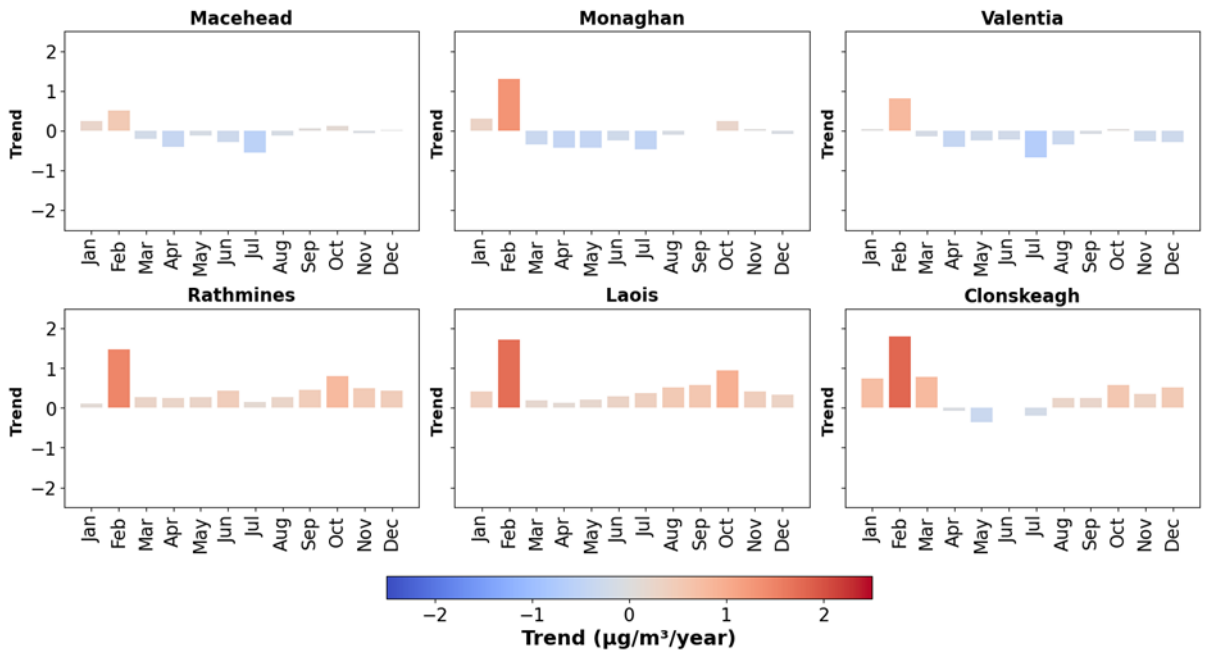

Figure S2 Monthly trend analysis of O3 at different for 15 years

2. Trend in O_3 Percentiles

Percentile statistics are valuable for understanding the distribution of O levels. The 5th percentile represents the value below which 5% of the ozone measurements fall, indicating low or background O_3 levels. This measure helps identify baseline O_3. The 50th percentile, or median, marks the midpoint of the data, providing a robust measure of typical O_3 levels without being skewed by extreme values. It is often used to represent average exposure conditions in a region. The 95th percentile captures the value below which 95% of O_3 measurements fall and highlights the upper extremes of ozone concentrations. This measure is crucial for understanding high O_3 pollution events. Percentiles to the analysis of O_3 in different scientific studies (e.g., Lefohn et al., 1998; Monks et al., 2015).

Figure 5 shows the percentile trend observed at different stations. The percentiles considered are the 5th, 50th, and 95th percentiles, which provide insights into the lower, median, and upper trends respectively. Mace Head exhibits a slight positive trend at the lower percentile and no change at the median percentile. However, the upper percentile shows a minor negative trend, indicating some variability in higher values. Monaghan shows a consistent negative trend across all percentiles, with the trend becoming more pronounced at higher percentiles. This suggests a general decline in the observed values. Similar to Monaghan negative trend across

all percentiles was observed at Valentia. The negative trend is stable and does not vary significantly between the percentiles. Rathmines shows a positive trend at all percentiles, though the upper percentile trend is slightly less pronounced. Laois shows a uniform positive trend across all percentiles. Clonskeagh exhibits a positive trend, with the median percentile showing slightly higher than the lower and upper percentiles. Mayo Castlebar shows no change at the lower percentile but a positive trend at the median and upper percentiles, indicating an overall positive shift. Swords demonstrates a significant positive trend, especially at the median and upper percentiles, and Wicklow Bray exhibits an increasing positive trend from lower to upper percentiles. Cork South link road shows a consistent positive trend, with a more substantial increase at the upper percentile. Cork Bishopstown shows a strong positive trend, particularly at the median and upper percentiles, Cork UCC shows a substantial negative trend across all percentiles, with the trend becoming more pronounced at higher percentiles, indicating a notable decline. Kilkenny shows a negative trend at the lower and median percentiles but a slightly positive trend at the upper percentile, indicating some variability in the observed values. The trend analysis reveals diverse patterns across different stations. While some sites show consistent positive trends, others exhibit negative trends or mixed results.

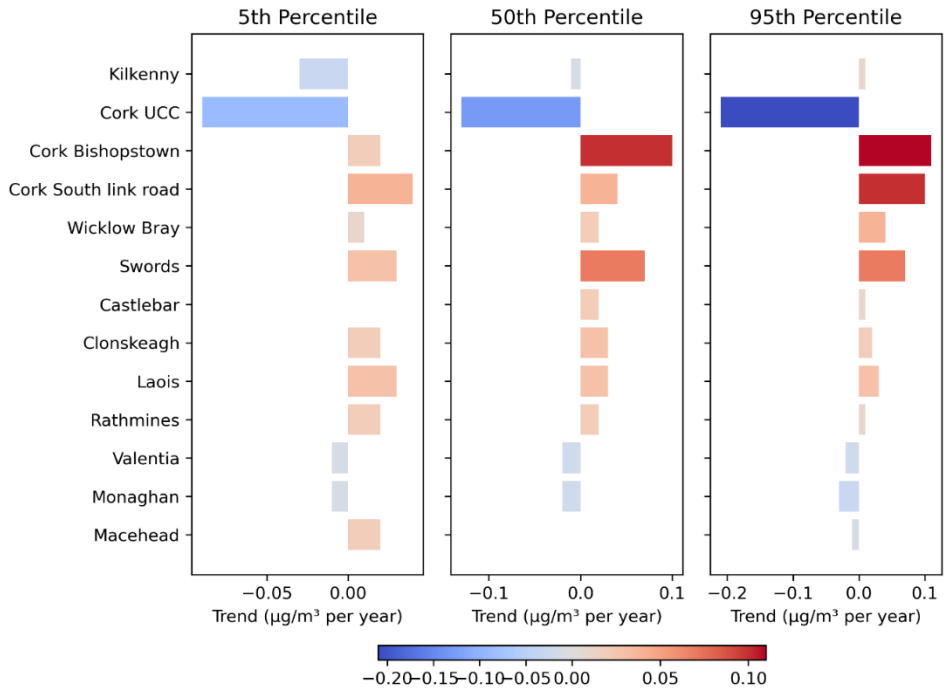

Figure S3

4.Exceedances in AOT40 –

AOT40 (Accumulated Ozone exposure over a Threshold of 40 ppb) is the key indicator for the protection of the vegetation (Ashmore et al., 2004). The AOT 40 is defined as the sum of the differences between hourly mean ozone concentrations (in ppb) and 40 ppb (80 μg/m3) for each hour when the concentration exceeds 40 ppb, accumulated during daylight hours**,** The EU ozone directive defines an accumulated ozone exposure of 3000 ppb*h for 3 months period as a critical level for the protection of vegetation. Therefore, daily AOT40 values in general should not exceed ∼33 ppb.

Figure shows a number of AOT40 exceedances at different locations in Ireland from 1994 to 2022. Mace head consistently shows a high number of exceedance days, with the peak occurring in 1999 (162 days).The number of exceedances generally declines over the years, with fluctuations, reaching a low of 84 days in 2022. Monaghan shows considerable variability with peaks . The highest number of exceedance days was 64 in 1995.The values tend to be lower in the 2000s and 2010s, around 20-48 days in recent years. At Valentia the number of exceedance days peaked in 2003 (162 days) and then showed variability, declining to 52 days in 2022. O_3 exceedances in Rathmines are generally low. The highest recorded value is 24 days in 2021, but most years have less than 20 exceedance days. At Laois a notable peak of 63 days occurred in 2017, with recent values ranging from 12 to 39 days. Similar to Rathmines, Clonskeagh generally reports low exceedances. A noticeable increase to 47 days was recorded in 2013.

Mayo Castlebar, showing moderate levels. There was an increase around 2019 and 2020 with values reaching 44 days.at Dublin Swords .The highest levels were observed in 2019 (41 days).Bray shows low initial values with increasing trends in recent years. The highest recorded value was 67 days in 2022. Exceedances in Cork south link road are generally low, with a peak of 44 days in 2013.Values remain consistent around 9-30 days in recent years. Similar to South Link Rd, Bishopston and CORK UCC shows low exceedances. Kilkenny, showing moderate levels. The highest recorded level was 51 days in 2021.

Mace head, due to its coastal location and exposure to transboundary air pollution, consistently reports the highest number of exceedance days. Urban areas like Dublin and Cork generally report fewer exceedance days, likely due to local atmospheric conditions and O_3 precursors .

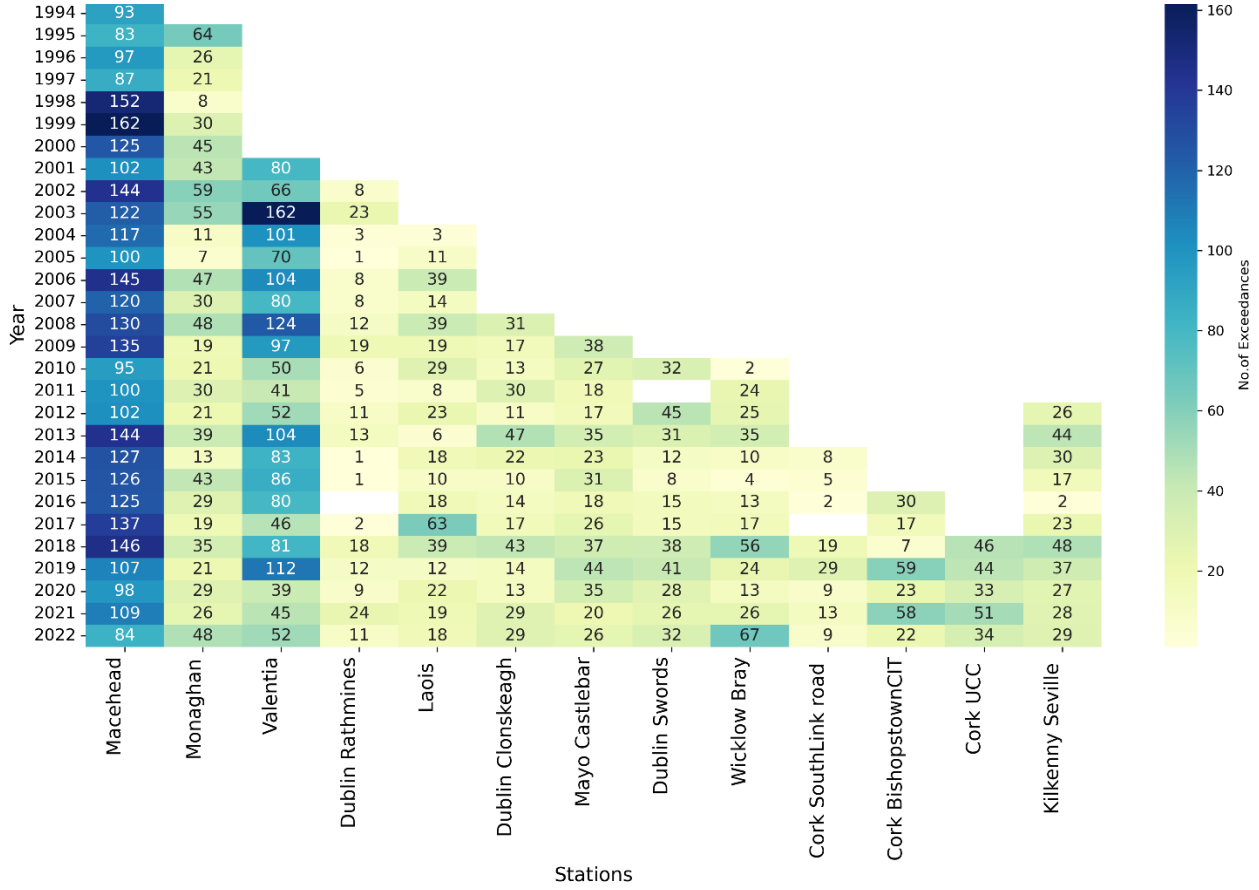

Figure – S4 - AOT40 exceedances at different locations in Ireland from 1994 to 2022.

5. CAM4- Chem Model grids

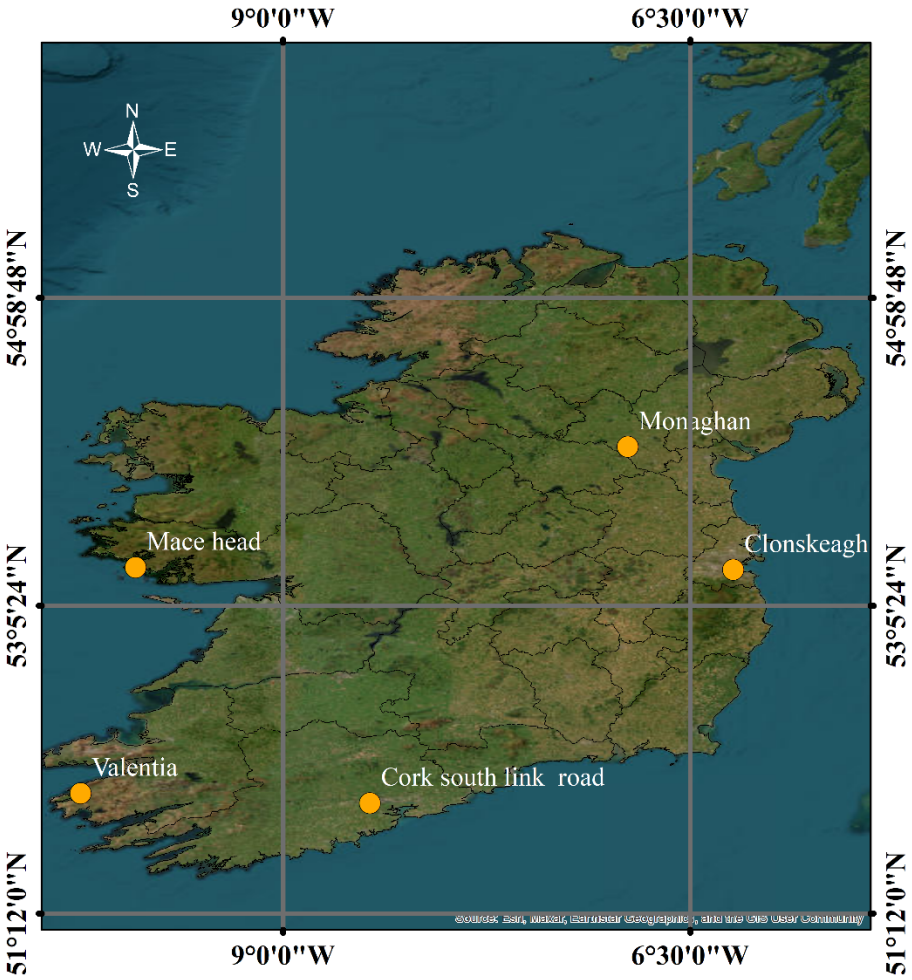

Figure – S5 - CAM4- Chem Model grids over Ireland and the O_3 observation sites.

Table S1 . The statistical analysis between the CAM4-Chem model and observations mean bias (MB), normalized mean bias (NMB), Mean Gross Error (MGE), and Mean Normalized Gross Error (MNGE).

	Mace Head	Monaghan	Valentia	Clonskeagh	Cork South link road
Bias	4.42	1.43	1.54	-3.38	-11.98
Normalized Bias (%)	11.68	4.74	4.33	-13.15	-57.58
Gross Error	5.28	4.36	4.71	5.54	12.45
Normalized Gross Error (%)	13.95	14.44	13.28	21.52	59.85
Correlation	0.83	0.73	0.72	0.68	0.49

6. The average contribution in model surface o3

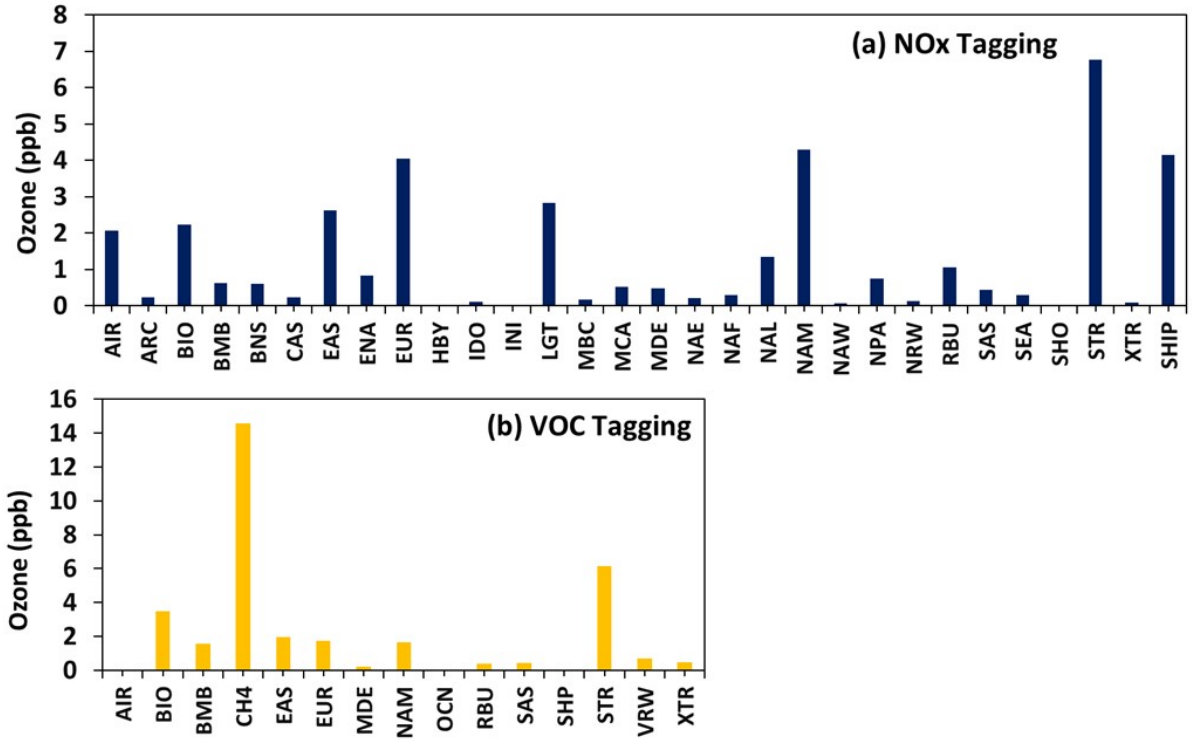

Figure S6 – The average contribution in model surface o3 for Mace head grid cell (a) NOx Tagging (b) VOC tagging .

7. Trends in contributors of Model O3 during the different season for clean and EU influenced sector

Table S2 – Trends in NOx Tagged contributors of model O_3 during the different season for clean sector .

	Autumn		Spring		Summer		Winter	
	slope	p value	slope	p value	slope	p value	slope	p value
AIR	0.058	0	0.049	0	0.023	0	0.058	0
BIO	0.015	0.076	-0.002	0.77	0.025	0.015	-0.001	0.775
EAS	0.062	0	0.058	0.004	0.023	0.006	0.083	0
EUR	-0.062	0.002	-0.067	0.002	-0.06	0.061	-0.044	0
LGT	0.034	0.003	-0.005	0.408	0.013	0.008	-0.021	0.036
NAL	0.015	0.204	0.027	0.022	0.035	0.013	0.023	0
NAM	-0.15	0	-0.152	0	-0.138	0	-0.138	0
RBU	0	0.998	-0.005	0.496	0	0.979	-0.003	0.186
STR	0.045	0.018	0.066	0.158	-0.006	0.692	0.036	0.13

Table S3 – Trends in NOx Tagged contributors of model O3 during the different season for EU influenced sector.

	Autumn		Spring		Summer		Winter	
	slope	p value	slope	p value	slope	p value	slope	p value
AIR	0.046	0	0.048	0	0.03	0	0.066	0
BIO	0.012	0.152	0.011	0	0.025	0.013	0.007	0.135
EAS	0.042	0.004	0.036	0.031	0.019	0.049	0.086	0
EUR	-0.092	0.062	-0.118	0.028	-0.147	0.04	-0.041	0.02
LGT	0.014	0.204	-0.006	0.477	0.01	0.148	-0.005	0.733
NAL	0.016	0.039	0.018	0.003	0.034	0.012	0.016	0
NAM	-0.131	0	-0.137	0	-0.108	0	-0.1	0
RBU	0.001	0.933	-0.001	0.83	-0.008	0.488	-0.001	0.566
STR	0.013	0.4	0.015	0.782	-0.007	0.678	0.043	0.269

Table S4 – Trends in VOC Tagged contributors of model O_3 during the different season for clean sector

	Autumn		Spring		Summer		Winter	
	slope	p value	slope	p value	slope	p value	slope	p value
BIO	0.016	0.059	0.008	0.146	0.011	0.207	0.013	0.001
BMB	0.054	0.064	-0.003	0.758	0.018	0.323	0.012	0.158
CH4	0.048	0.083	0.063	0	0.017	0.555	0.063	0
EAS	0.033	0	0.06	0	0.011	0.032	0.061	0
EUR	-0.03	0	-0.049	0	-0.027	0.011	-0.033	0
NAM	-0.066	0	-0.087	0	-0.055	0	-0.075	0
STR	0.04	0.019	0.069	0.13	-0.006	0.673	0.036	0.099

Table S5 – Trends in VOC Tagged contributors of model O_3 during the different season for EU influenced sector.

	Autumn		Spring		Summer		Winter	
	slope	p value	slope	p value	slope	p value	slope	p value
BIO	0.009	0.284	0.019	0.045	0.003	0.873	0.025	0.004
BMB	0.052	0.052	0.001	0.891	0.014	0.424	0.017	0.033
CH4	0.011	0.714	0.08	0	0.024	0.422	0.112	0.006
EAS	0.024	0.003	0.046	0.004	0.011	0.066	0.06	0
EUR	-0.045	0.04	-0.097	0	-0.07	0.028	-0.034	0.002
NAM	-0.06	0	-0.084	0	-0.049	0	-0.058	0
STR	0.012	0.374	0.016	0.761	-0.006	0.689	0.039	0.274