



Urban Ozone Trends in Europe and the USA (2000-2021)

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Abstract. Trends in urban O₃ and NO₂ across Europe and the United States of America were explored between 2000-2021. Using surface monitoring site data from the TOAR-II and European Environment Agency databases, piecewise quantile regression (PQR) analysis was performed on 228 O₃ time series (144 European, 84 USA) and 322 NO₂ times series (245 European, 77 USA). The PQR analysis permitted 2 break points over the 23 year period to balance the intent to describe changes over a large time period, while still capturing the abrupt changes that can occur in urban atmospheres. Regressions were performed over quantiles ranging from 0.05 to 0.95 and indications of a slowing in the increase of high European O₃ levels was observed. In Europe, more trends were found to having an increasing O₃ trend between 2015-2021 compared with 2000-2004. The reverse was true in the USA, with a reduction in the number of sites with increasing O₃ trends when the same periods were compared. An analysis of the change points revealed a large proportion of sites in Europe, were the second change point in NO₂ switched from a positive to negative trend, occurred in 2020 (41/43 second change points in this year). This was attributed a reduction in NO₂ due to the COVID-19 pandemic, however, in some cases these increasing trends have sustained beyond the recovery from restrictions.

1 Introduction

Tropospheric ozone (O₃) is a greenhouse gas and air pollutant harmful to human health, and plant growth (Fleming et al., 2018; Mills et al., 2018; Szopa et al., 2021). It is a secondary air pollutant, formed from the photochemical reactions of primary pollutants NO_x (NO + NO₂) and volatile organic compounds (VOCs). The chemistry of O₃ formation is non-linear and the effect of changing precursor concentrations can cause O₃ concentrations to increase or decrease depending on the photochemical regime found at a given location (Sillman et al., 1990; Sillman, 2002). Despite global successes in reducing primary pollutant emissions over the past few decades, global exposure to O₃ has been increasing throughout the 21st century. This is particularly observed in urban areas, where the vast majority of the global population live, projected to increase to 68% in 2050 from 55% in 2018 (UN, 2019). In a study of 12946 cities located worldwide, the average mean weighted O₃ concentration increased by 11% between 2000 and 2019, and the number of cities exceeding the WHO peak season O₃ standard increased from 89% to 96% (Malashock et al., 2022).



Due to the complexity of O_3 production, its trend direction, magnitude, and significance varies by location. A previous study
 25 calculated trends in two O_3 metrics globally over the period of 2000 - 2014: 4th highest daily maximum 8-hour O_3 (4MDA8),
 and the number of days with MDA8 > 70 ppb O_3 (NDGT70) (Fleming et al., 2018). The study used data from 4801 global
 monitoring sites over this time period. For both of these metrics, downward trends were observed for most of the USA, and
 some sites in Europe. However, over the period of 2010-2014 (2,600 sites utilised), sites located in regions with the highest O_3
 precursor emissions across North America, Europe and East Asia had the highest values in 4MDA8 and NDGT70. In North
 30 America and Europe, this was particularly true for California and parts of southern Europe (Fleming et al., 2018).

Since the 1990s, a general downward trend in urban O_3 pollution has been observed in the United States (He et al., 2020).
 This reducing trend has been linked to stricter limiting regulations on the emissions of primary pollutants such as NO_x and
 VOCs. Although NO_x and VOC emissions in Europe have also been declining since the late 1980s, the trend in O_3 is less clear
 due to large inner-annual variation, driven by climate variability and the dispersion and transport of pollutants from other re-
 35 gions (Jonson et al., 2006; Yan et al., 2018). Between 1995 and 2014, negative trends in the highest O_3 levels across urban sites
 in Europe were identified due to pollutant emission restrictions across Europe. However, increasing background levels, particu-
 larly in northern and eastern Europe, make it difficult to identify strong trends in urban O_3 when transboundary effects are
 considered (Yan et al., 2018). Despite this, a study of 93 suburban and urban sites across Europe identified notable enhance-
 ments in O_3 seasonal and annual means between 1995 - 2012, even with the continuous downward trend in anthropogenic
 40 emissions across the continent (Yan et al., 2018).

Since these earlier studies, much of the literature focuses on the impact of the COVID-19 pandemic on air pollutant concen-
 trations and trends (Lee et al., 2020; Shi et al., 2021; Grange et al., 2021; Sokhi et al., 2021). A study of eleven cities across
 the world, all of which implemented stringent lockdown measures in early 2020, captured sudden decreases in deweathered
 NO_2 concentrations, concurrent with sudden increases in O_3 , in most locations (Shi et al., 2021). Another study employing
 45 machine learning models to predict business-as-usual levels of pollutants, estimated that NO_2 concentrations were 32% lower
 than expected across 102 European urban background locations, whereas O_3 was 21% higher (Grange et al., 2021). It is highly
 likely that this global event has resulted in perturbations of both long-term NO_2 and O_3 trends, particularly in locations where
 lockdowns were stringent or lengthy.

This study examines the trends in urban O_3 and NO_2 using monitoring site data from 2020 - 2023. It employs quantile
 50 regression and change point detection to construct trends that capture the broad structure of a complex time series, while
 remaining explainable with concise statistics. Section 2 details the method used to create the trends, section 3.1 provides a
 high-level overview of the trends with section 3.1.1 focusing on the significance of the trends and highlighting features that
 are discussed in detail later on. Section 3.2 explores the information that can be gained from trends calculated over a range of
 quantiles and finally section 3.3 provides details on some of the features revealed earlier in the analysis.



55 2 Methodology

Hourly NO_2 and O_3 data were obtained for urban sites in the USA and Europe using the TOAR-II (Schröder et al., 2021) and European Environment Agency (EEA) (European Environment Agency, a, b) databases, using the r-packages toarR (Drysdale, 2024) and saqgetr (Grange, 2019) respectively. Those obtained from the EEA are categorised as urban traffic, urban background and urban industrial, while those from the TOAR database are categorised as urban and suburban. A full list of sites can be found in the supplementary information (table S1). TOAR-II data was retrieved between 2000-01-01 and 2021-12-31 (the latest available) and EEA data between 2000-01-01 and 2023-12-31, the latter being extended longer so the effects of the COVID-19 pandemic could be observed more clearly. Time series were required to have 90 % data coverage between 2000-01-01 and 2021-12-31 and retained time series were averaged to daily medians. Additionally, a small number of time series were removed following visual inspection - these are listed in table S2. This resulted in 228 O_3 time series (144 Europe, 84 USA), 322 NO_2 time series (245 Europe, 77 USA) and 126 O_x ($\text{NO}_2 + \text{O}_3$) time series (101 Europe, 25 USA).

Firstly, the time series were de-seasoned by calculating a monthly median climatology and subtracting this from the time series, producing an ‘anomaly’ time series. The NO_2 and O_3 anomaly time series were summed to produce O_x anomaly time series for sites that have both NO_2 and O_3 data available. Following this, several methods were defined to help describe trends. As the time series are non-linear Locally Estimated Scatterplot Smoothing (LOESS) was applied first. This method works by calculating many least squared fits over a moving weighted subset of the data – with this subset tuned based on the desired ‘smoothness’ of the result. In this case with a smoothing parameter (α) chosen as 0.5, which was determined by inspection to capture a balance of broad trends and medium-term non-linearity. LOESS works well for drawing the eye to features of the time series but does not allow for broad trends to be described numerically as the resulting slopes are changing continuously.

As such, quantile regression (QR) was used to define trends following the methodology in and using code provided by Chang et al. (2023). QR calculates a linear model that seeks to minimise the residuals with a defined proportion (τ) of the points above and below the fit line. For example, the scenario $\tau = 0.5$ splits the data 50:50 above and below the line. QR has the advantage of being insensitive to outliers and can be considered analogous to a “median” trend line. This is desirable for the longer-term trends being investigated here. The 1-sigma uncertainty for the QRs were calculated via a moving block bootstrapping method, where the data are subdivided into overlapping blocks that are $n^{1/4}$ points wide (or ~20 points for 20 years of hourly data). These blocks are shuffled to generate a new time series, and replicated 1000 times, and the standard deviation of these replicates calculated. This uncertainty was subsequently used in the determination of the p-value, providing a metric for the significance of the trends.

As described the QR will be a poor representation of a time series with non-linearities (hence first investigating the LOESS) as it will be using a single slope to define the whole trend across a complex time series. As urban concentration trends can change sharply (e.g. with policy intervention), the model requires freedom to represent these, therefore they were extended to use piecewise quantile regressions (PQR) with up to two break points (resulting in three trend lines). This allows the model to have some changes in the trends calculated, capturing sufficiently large-scale changes while still being able to describe a ~20 year time series with a small number of coefficients.



To determine what break points to use on each time series, 109 scenarios per time series defined by 1, 2 or 3 sequential QRs were created, separated by break points occurring on the 1st of January in up to two years excluding the first and last 2 years of a time series. In the cases where there were 2 break points, these were not allowed to occur within 5 years of each other. The cases with 2 or 3 regressions were combined into a single PQR, (PQR1 and PQR2 respectively). All regressions were calculated at $\tau = 0.05, 0.10, 0.25, 0.50, 0.75, 0.95$ and 0.95 .

To select 'change points' from the array of break points available, the model performance was evaluated via Akaike information criterion (AIC). The piecewise scenario with the minimum AIC at $\tau = 0.5$ were selected as the change points for a given time series. This is a decision that trades off between there being potential change points that only exist in cases where $\tau \neq 0.5$, and retaining the ability to relate trends to one another across all quantiles. As this study aimed to explore the trends across a large number of sites, defining the change points only using $\tau = 0.5$ is more appropriate, but future studies that reduce the number of time series could consider investigating how change points differ with τ in more detail.

Figure 1 demonstrates this process on a single time series. Change points could be assessed for their efficacy (i.e. is the effect of introducing a change point important, or has it only provided a trivial reduction in the AIC?) by comparing the QR and the best PQR1 and PQR2 cases, but as this does not substantively change the results presented here, this was not done and the PQR2 fits were determined to be the most appropriate in all cases.

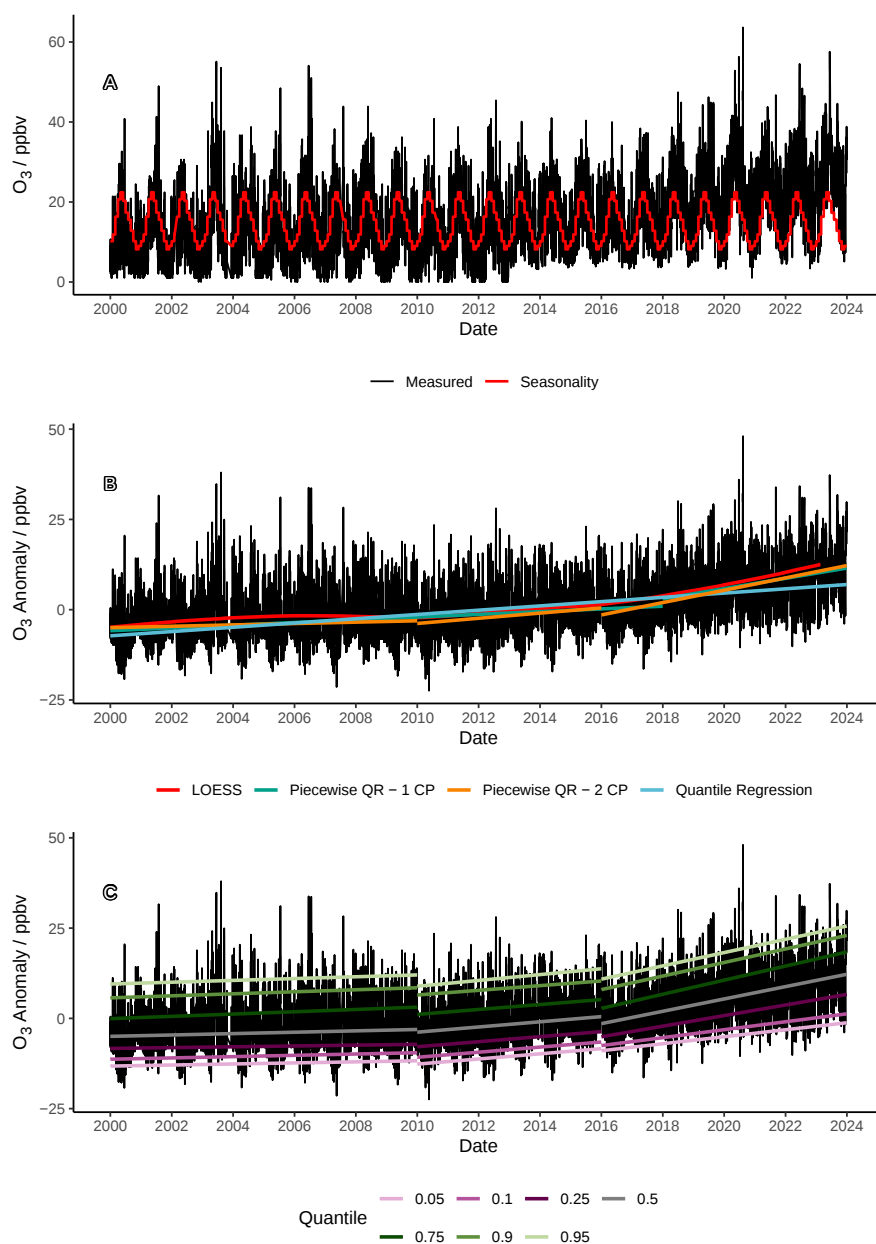


Figure 1. Example trend determination for an O₃ time series (London Bloomsbury - GB0566A). A - O₃ concentration time series (black) and monthly median climatology (red). B - Anomaly time series (black, concentration minus climatology) and 4 trend options, LOESS (red), QR (blue), PQR1 (green) and PQR2 (yellow). C - Anomaly time series (black) and PQR2 across $\tau = 0.05, 0.10, 0.25, 0.50, 0.75, 0.90, 0.95$. (purple to green)

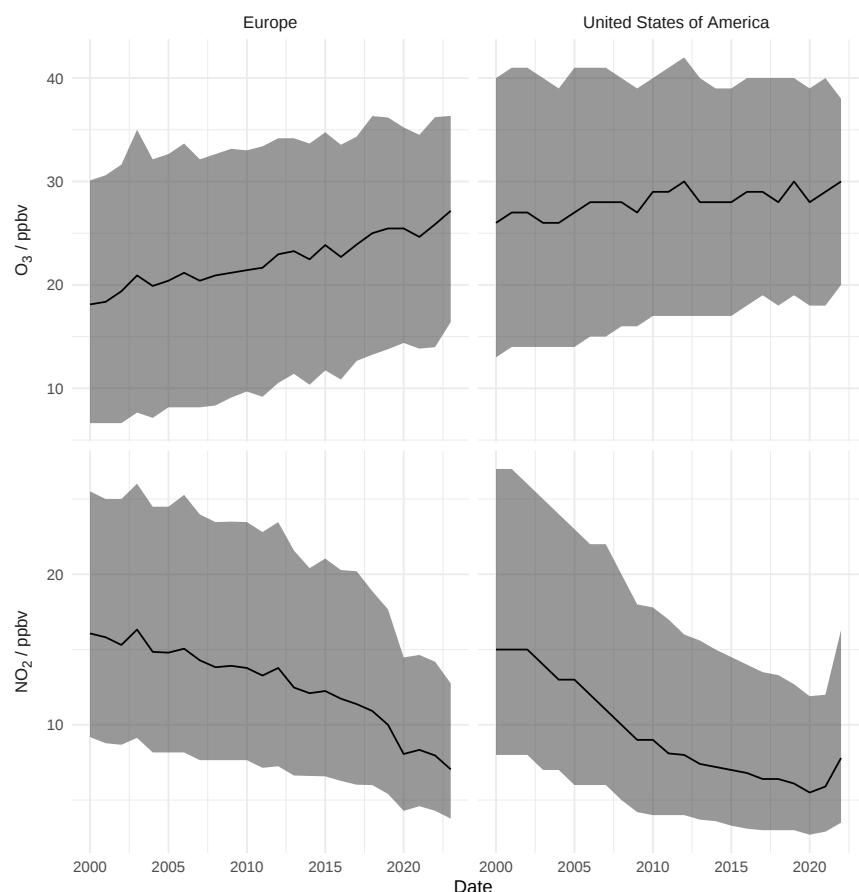


Figure 2. Median concentrations of O_3 (top) and NO_2 (bottom) across all sites in this study, separated into sites in Europe (left) and sites in the USA (right). Shaded regions represent the 25th and 75th percentiles.

3 Results and Discussion

105 To provide context for the subsequently calculated trends, figure 2 shows the annual median, 25th and 75th percentile concentrations of O_3 and NO_2 across all the urban sites used in this study. Europe shows median O_3 concentrations increasing from 18 ppb to 26 ppb between 2000 and 2019, and USA concentrations increasing, from 26 ppb in 2000 to 30 ppb in 2019. Both Europe and the USA show decreasing NO_2 over the same period (16 ppb to 10 ppb in Europe, 15 ppb to 6 ppb in the USA). The USA does see a return to increasing NO_2 concentrations after 2020, reaching 7.8 ppb in 2022. In section 3.3 evidence for
 110 this increase is discussed.

The dataset that results from the application of the method described in section 2 has several degrees of freedom to bear in mind during discussion. For a given time series (one species at one site) there are two break points that are common for all the trend lines, and as there are 7 quantiles, this results in 21 slopes, each with an associated p-value. When comparing between



sites (or even species within a site) the change points can differ. This is necessary to capture the changing trends observed at
 115 urban sites but does complicate describing them. For this discussion we have grouped the sites into those from Europe and
 those from the United States, though details on individual European countries are available in table S1. To aid in summarising,
 in some cases the results have been grouped into time periods of a few years – in these cases if a trend changes due to a change
 point both trends have been counted. For example, if between 2000 and 2004 a site were to go from increasing O₃ to decreasing
 O₃, this would be described as ‘between 2000 and 2004 there was one increasing O₃ trend and one decreasing O₃ trend’. In the
 120 case where a site had a change point in 2003, but the trend remained increasing, this will be counted as one increasing trend.
 This has the benefit that no one category can count more than the number of available time series. In visualisation, both trends
 would be shown.

Trends have been collated into significance categories: $p \leq 0.05$ (high certainty), $0.05 < p \leq 0.10$ (medium certainty), 0.10
 $< p \leq 0.33$ (low certainty) and $p > 0.33$ (very low certainty or no evidence) based off of the guidance of Chang et al. (2023).
 125 For the most part slopes where the p-value is > 0.33 are treated as ‘no trend’ regardless of their magnitude (generally we
 observe that as the magnitude of the trend decreases so does its significance), though sometimes are given with a direction
 when required by a visualisation.

3.1 Overview of Trends

The $\tau = 0.5$ trends of O₃ and NO₂ were examined and are presented on maps in figures 3 and 4.

130 In Europe, between 2000 and 2004 there were 108 trends where O₃ was increasing, and 41 where it was decreasing, 32
 showed no trend. By 2015-2021, 123 had an increasing trend, 32 decreasing and 41 with no trend, indicating generally increas-
 ing O₃ concentrations across Europe. This is somewhat mirrored by the trends in NO₂, where in 2000-2004 69 trends where
 increasing, 181 decreasing and 59 with no trend, changing to 29 increasing, 241 decreasing and 39 with no trend by 2015-2021.
 This would suggest a general picture that urban locations in Europe are still on the VOC limited portion of the O₃ production
 135 isopleth, and decreasing NO_x increases O₃ production, however, it is not quite as ubiquitous as this, as shown in 3.3.1 which
 discusses how O₃ responded to changing NO₂ following restrictions in 2020.

This contrasts with the USA, where in 2000-2004, there were 10 sites with increasing NO₂, and 0 sites increasing between
 2005 and 2014, but in 2015-2021 17 sites had begun increasing in NO₂ again – though notably as seen from figure 4 these
 are different sites to those that were increasing at the beginning of the century. This increase in NO₂ is not clearly reflected
 140 by the O₃ trends with 62 increasing, 23 decreasing and 22 showing no trend in 2000 – 2004 and 44 increasing, 34 decreasing,
 and 25 showing no trend in 2015-2021. A decreasing number of sites with a positive trend in O₃, suggests that the majority of
 decreasing NO₂ trends are slowing the increase in O₃. The counts of sites at all values of τ can be found in tables 2 and 3.

Across the entire study period, for O₃ in Europe, at sites with increasing trends the median slope was 0.36 ppb yr⁻¹, slightly
 lower than in the US, where they were increasing by 0.40 ppb yr⁻¹. For NO₂, the median rate of increase was the same in
 145 Europe (0.36 ppb yr⁻¹), but higher at 0.71 ppb yr⁻¹ in the US. For decreasing trends, O₃ and NO₂ in Europe and the US were all
 similar, O₃ in Europe decreasing at -0.40 ppb yr⁻¹, (-0.36 ppb yr⁻¹) in the US and for NO₂ -0.39 ppb yr⁻¹ in Europe and -0.47
 in the US. This information along with the 5 th and 95 th percentile (still at $\tau = 0.5$) are shown in table 1.

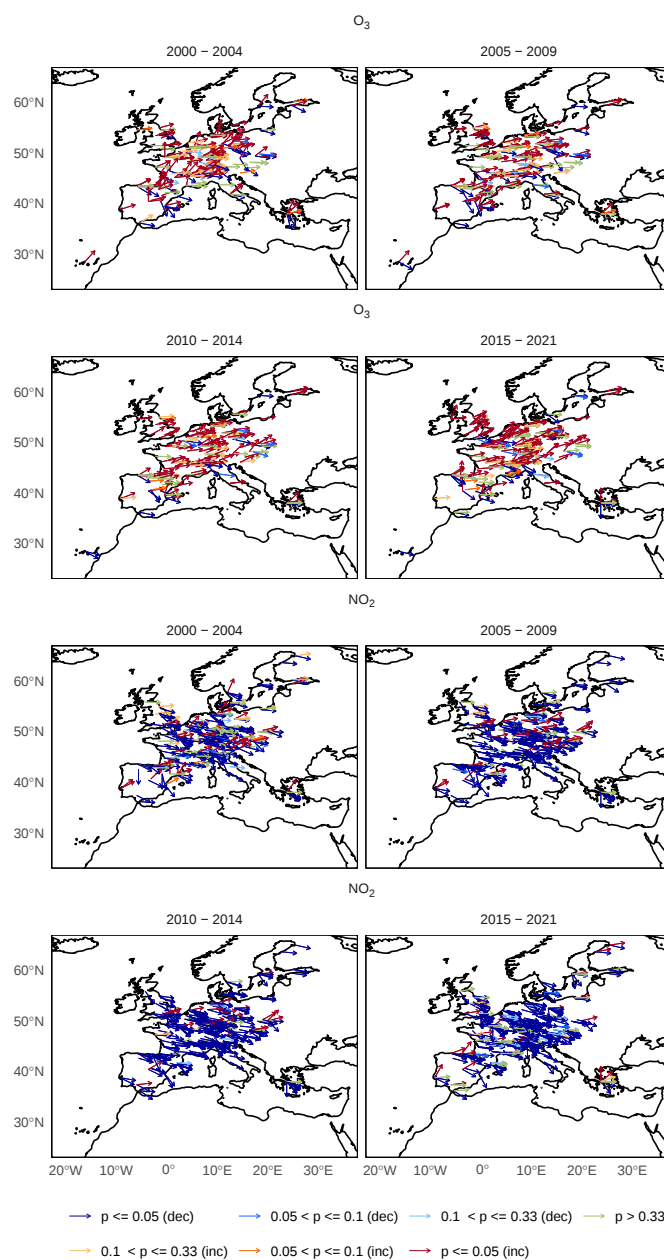


Figure 3. Summary of trends across Europe. Angle of arrow indicates magnitude and direction, with the following orientations: vertical up - $+2.5 \text{ ppb yr}^{-1}$, horizontal - 0 ppb yr^{-1} and vertically down - -2.5 ppb yr^{-1} . The few sites with absolute trends $> 2.5 \text{ ppb yr}^{-1}$ have been clamped to $\pm 2.5 \text{ ppb yr}^{-1}$. Colour indicates significance and direction, darkest colours being most significant ($p < 0.05$) and green the least ($p > 0.33$). Blues are decreasing trends and reds are increasing trends. The upper 4 plots show O_3 trends, and the lower 4 show NO_2 trends. Each group of 4 groups the trends into segments of the study period. If a change point occurs in a group an arrow for both trends is shown.

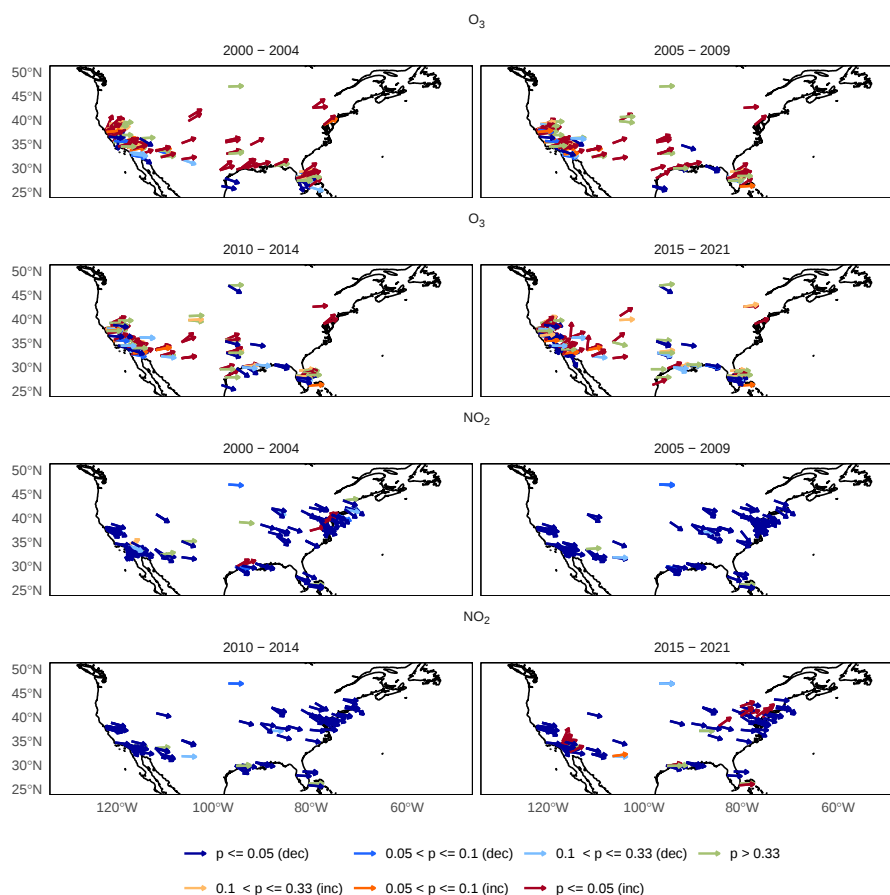


Figure 4. Summary of trends across the United States of America. Angle of arrow indicates magnitude and direction, with the following orientations: vertical up - $+2.5 \text{ ppb yr}^{-1}$, horizontal - 0 ppb yr^{-1} and vertically down - -2.5 ppb yr^{-1} . The few sites with absolute trends $> 2.5 \text{ ppb yr}^{-1}$ have been clamped to $\pm 2.5 \text{ ppb yr}^{-1}$. Colour indicates significance and direction, darkest colours being most significant ($p < 0.05$) and green the least ($p > 0.33$). Blues are decreasing trends and reds are increasing trends. The upper 4 plots show O_3 trends, and the lower 4 show NO_2 trends. Each group of 4 groups the trends into segments of the study period. If a change point occurs in a group an arrow for both trends is shown.



Table 1. 5 th, 50 th and 95 th percentiles of the $\tau = 0.5$ slopes in Europe and the USA.

		Slope / ppbv year ⁻¹					
		Europe			United States of America		
Species	Percentile	Increasing	Decreasing	No Trend	Increasing	Decreasing	No Trend
O ₃							
	5 th	0.09	-1.55	-0.11	0.11	-1.21	-0.14
	50 th	0.36	-0.40	0.00	0.40	-0.36	0.00
	95 th	1.46	-0.10	0.14	1.11	-0.09	0.13
NO ₂							
	5 th	0.07	-1.32	-0.12	0.15	-1.36	-0.16
	50 th	0.35	-0.39	0.00	0.71	-0.47	0.00
	95 th	1.39	-0.10	0.11	1.35	-0.10	0.19
O _x							
	5 th	0.10	-1.16	-0.07	0.19	-1.41	-0.13
	50 th	0.41	-0.29	0.01	0.36	-0.31	0.00
	95 th	1.66	-0.06	0.19	1.22	-0.11	0.11

Table 2. Trends in O₃, NO₂ and O_x at sites in Europe in annual groups between 2000 and 2021 inclusive. If a site as a change point within a group, both slopes are added to the tally. Those classed as 'No Trend' are the slopes where $p > 0.33$

Species	τ	Increasing					Decreasing					No Trend				
		00 - 04	05 - 09	10 - 14	15 - 21		00 - 04	05 - 09	10 - 14	15 - 21		00 - 04	05 - 09	10 - 14	15 - 21	
O ₃	0.05	112	114	119	123	23	23	21	15	24	45	46	33	47		
	0.10	114	119	124	127	26	26	23	16	23	36	36	26	44		
	0.25	107	110	115	127	32	32	30	19	30	40	42	31	41		
	0.50	108	108	115	123	41	41	40	23	32	32	36	30	41		
	0.75	104	99	107	120	40	40	38	24	36	38	44	39	45		
	0.90	99	93	104	111	43	43	43	23	32	44	50	41	53		
	0.95	93	84	96	101	44	44	43	25	33	50	60	53	61		
NO ₂	0.05	59	48	31	23	169	169	197	223	242	80	54	33	67		
	0.10	68	45	31	27	169	169	205	230	243	73	51	26	50		
	0.25	66	43	29	30	183	183	217	236	243	59	33	18	49		
	0.50	69	42	27	29	181	181	224	238	241	59	32	14	39		
	0.75	73	44	22	22	175	175	220	237	242	60	34	18	35		
	0.90	73	43	20	17	166	166	218	238	241	72	40	21	39		
	0.95	78	44	18	15	157	157	207	236	241	83	50	29	42		
O _x	0.05	52	27	28	38	52	52	58	61	62	41	39	34	44		
	0.10	55	29	33	37	61	61	68	67	64	33	26	25	43		
	0.25	58	29	36	44	59	59	67	67	62	29	28	25	35		
	0.50	59	31	40	51	59	59	66	66	65	26	25	19	28		
	0.75	59	30	41	49	56	56	62	61	59	31	27	21	32		
	0.90	62	29	37	50	58	58	62	61	57	27	27	26	33		
	0.95	62	26	30	39	63	63	65	58	52	25	29	37	48		

Table 3. Trends in O₃, NO₂ and O_x at sites in the United States of America in annual groups between 2000 and 2021 inclusive. If a site as a change point within a group, both slopes are added to the tally. Those classed as 'No Trend' are the slopes where $p > 0.33$

Species	τ	Increasing				Decreasing				No Trend			
		00 - 04	05 - 09	10 - 14	15 - 21	00 - 04	05 - 09	10 - 14	15 - 21	00 - 04	05 - 09	10 - 14	15 - 21
O ₃	0.05	68	64	54	49	10	11	18	20	28	23	30	38
	0.10	71	67	55	50	9	9	19	24	25	18	27	32
	0.25	69	63	54	47	13	10	25	32	24	21	28	27
	0.50	62	57	46	44	23	19	33	34	22	24	29	25
	0.75	55	50	41	39	25	24	41	40	30	28	23	24
	0.90	42	38	35	36	30	30	43	41	36	35	31	28
NO ₂	0.95	36	33	29	31	37	36	44	39	34	33	31	34
	0.05	11	3	3	10	70	74	68	60	16	10	14	24
	0.10	11	3	5	12	74	77	69	62	13	6	10	20
	0.25	13	3	4	14	71	75	74	67	10	4	5	17
	0.50	10	0	0	17	74	77	76	72	6	2	5	7
	0.75	9	0	0	16	75	77	77	74	5	1	3	6
O _x	0.90	6	0	0	9	75	77	77	74	15	0	2	14
	0.95	7	0	0	7	74	77	76	74	15	3	3	17
	0.05	8	7	6	8	12	14	15	14	15	11	8	13
	0.10	9	6	5	8	11	13	17	15	16	13	10	14
	0.25	9	7	5	5	15	14	16	19	11	9	9	10
	0.50	8	5	4	3	17	17	19	20	10	8	8	10
	0.75	9	4	3	3	19	19	20	20	6	7	6	9
	0.90	8	4	4	4	20	19	21	22	7	6	5	7
	0.95	6	4	3	3	20	20	21	20	8	6	7	10

3.1.1 Significance of Trends

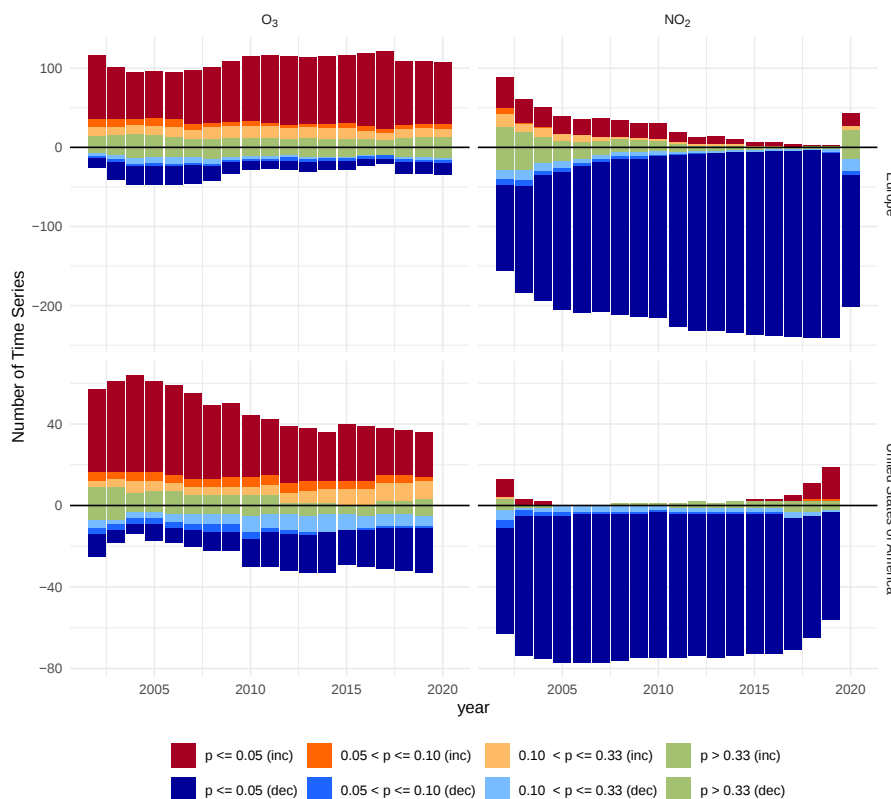


Figure 5. Time series of trends in O_3 (left) and NO_2 (right) for Europe (top) and the USA (bottom). Positive trends are contained in the bar above 0 and negative trends are contained in the bar below 0. Colour indicates significance and direction, darkest colours being most significant ($p < 0.05$) and green the least ($p > 0.33$). Blues are decreasing trends and reds are increasing trends. In this case trends with $p > 0.33$ have been sorted by their direction.

Figure 5 counts the trends by significance and direction per year. 2000, 2001 and 2021 onward in Europe and 2000, 2001, 2020 and 2021 onwards for the USA were excluded from this year on year analysis, as it is not possible to capture changing trends in these regions due to the limitations placed on where change points can occur as detailed in section 2.

This reveals the majority of trends are of high certainty. It also reinforces the observations from section 3.1, where both Europe and the US see O_3 increasing at number of sites, with there being proportionally more sites with a decreasing trend in the US. This additionally shows that the number of high certainty decreasing trends in O_3 has been present since ~2010, joined by a slow decline in increasing trends.

For NO_2 the feature of some sites beginning to show increasing trends in the US is clear, with this pattern beginning in 2015-2016. Between 2000 and 2019 nearly all European sites moved to a trend of decreasing NO_2 , however, this reversed for



several sites in 2020. This trend reversal can be attributed to the reduction in NO_2 concentrations in Europe which were a side effect of public health measures (Lee et al., 2020; Grange et al., 2021), and is discussed in more detail in 3.3.1.

160 3.2 Trends Across Quantiles

To investigate how the trends in urban O_3 are generally changing across quantiles in Europe and the USA, the distribution of slopes in each region was determined for each τ . This section necessarily needs to discuss two sets of quantiles referring to different calculations, as such τ is used to refer to the quantile associated with the quantile regression, and other references to quantiles are associated with the analysis of the distribution of slopes within a given τ .

165 The distributions at the start (year = 2000) and end (year = 2019) of the 20-year period were compared (Figure 6). Generally, across all values of τ , the interquartile range (IQR = 75th quantile minus 25th quantile) of the slope distributions in O_3 was greater in 2000 compared with 2019 ($\tau = 0.5$, IQR = 0.87 and 0.58 ppb yr^{-1} for 2000 and 2019 respectively). This is particularly true for Europe, where a clear reduction in the higher τ O_3 slopes is observed between 2000 and 2019. For $\tau = 0.95$, a much larger proportion of the slopes were $> 1.25 \text{ ppb yr}^{-1}$ in 2000 than 2019. For $\tau = 0.95$ in the USA, there is a reduction in the
 170 number of slopes with very negative ($< 1.00 \text{ ppb yr}^{-1}$) slopes between 2000 and 2019.

In Europe, the bulk of NO_2 slopes become more negative across all τ (Figure 6). However, in the USA, despite the majority of slopes being negative in both 2000 and 2019, there is still a proportion of slopes with positive slopes in 2019, not observed in the European distribution. This is particularly true for $0.5 \geq \tau \leq 0.9$. The IQR of the slope distributions was comparable between 2000 ($\tau = 0.5$, IQR = 0.50 ppb yr^{-1}) and 2019 ($\tau = 0.5$, IQR = 0.40 ppb yr^{-1}) for Europe, comparable to the reduction
 175 in spread observed in the USA in 2019 ($\tau = 0.5$, IQR = 0.44 ppb yr^{-1}) compared to 2000 ($\tau = 0.5$, IQR = 0.57 ppb yr^{-1}).

Annual changes in τ slopes were used to evaluate how absolute urban O_3 mixing ratios and trends changed across Europe and the USA between 2000 and 2021. For both Europe and the USA, the cumulative sum of the annual median of the trends across all sites was calculated for each value of τ . This was followed by the subtraction of the 2000 value to get relative change since 2000. Using this analysis, Figure 7 shows the change in median O_3 and NO_2 mixing ratios since 2000. The median of
 180 these results for Europe and the United States of America was then derived for each year and for each τ . From this, Figure 8 describes how the magnitude and direction of the trend line changes annually.

In both Europe and the USA, median trend-derived O_3 mixing ratios have generally increased between 2000 and 2021 (figure 7). This is most clearly observed in Europe, where all percentiles demonstrated a consistent increase in median O_3 across the two decades. Larger increases in O_3 were observed in the lower τ cases (+6.33 ppb , $\tau = 0.05$) compared to the higher τ cases
 185 (+5.08 ppb , $\tau = 0.95$), indicating that the lowest ambient O_3 levels are continuing to increase while higher levels show some signs of reducing their rate of increase. The picture is more mixed in the USA. Similarly to Europe, the largest increases in median O_3 since 2000 were observed in the lower τ cases (+4.95 ppb , $\tau = 0.05$). In the $\tau = 0.50$ case, median O_3 increased to ca. +2.5 ppb by 2011 before plateauing between 2011 and 2021. In contrast, in the highest τ value cases, median O_3 was lower in 2021 than in 2000 (-0.76 and -0.60 ppb , $\tau = 0.90$ and 0.95 respectively). This suggests that higher ambient levels of O_3 have
 190 reduced in magnitude since 2000, compared to lower ambient levels which are continuing to increase. In both Europe and the USA, median ambient NO_2 mixing ratios have reduced, with the largest reductions observed in the $\tau = 0.95$ case (-11.49 ppb

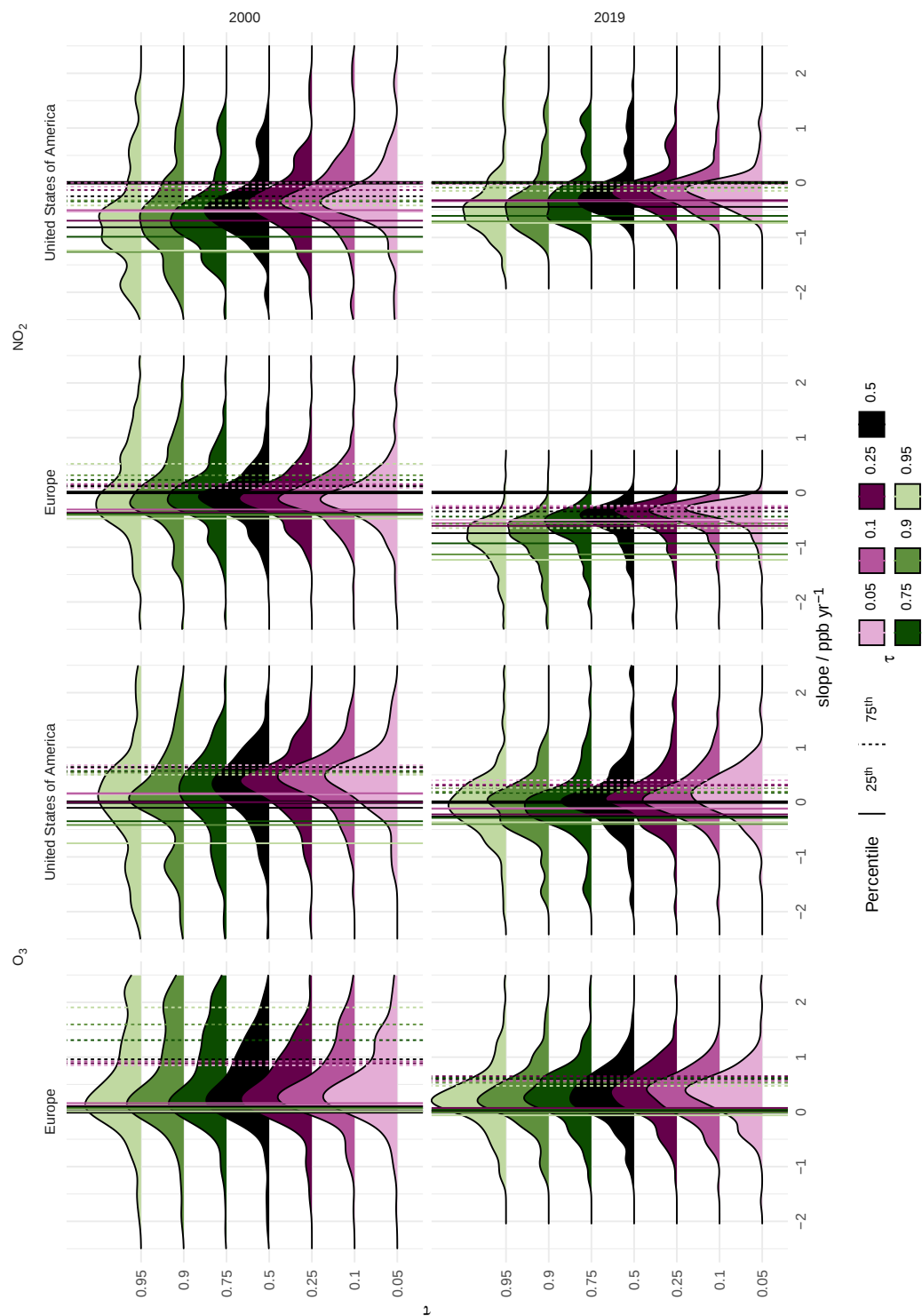


Figure 6. Slope density for NO₂ (left) and O₃ (right) for Europe (cols 1 and 3) and the USA (cols 2 and 4) in 2000 (top) and 2019 (bottom) for $\tau = 0.05, 0.1, 0.25, 0.5, 0.75, 0.9$ and 0.95



and -15.94 ppb for Europe and the USA respectively). Smaller reductions were observed in the $\tau = 0.05$ case (-4.48 and -5.78 ppb for Europe and the USA respectively).

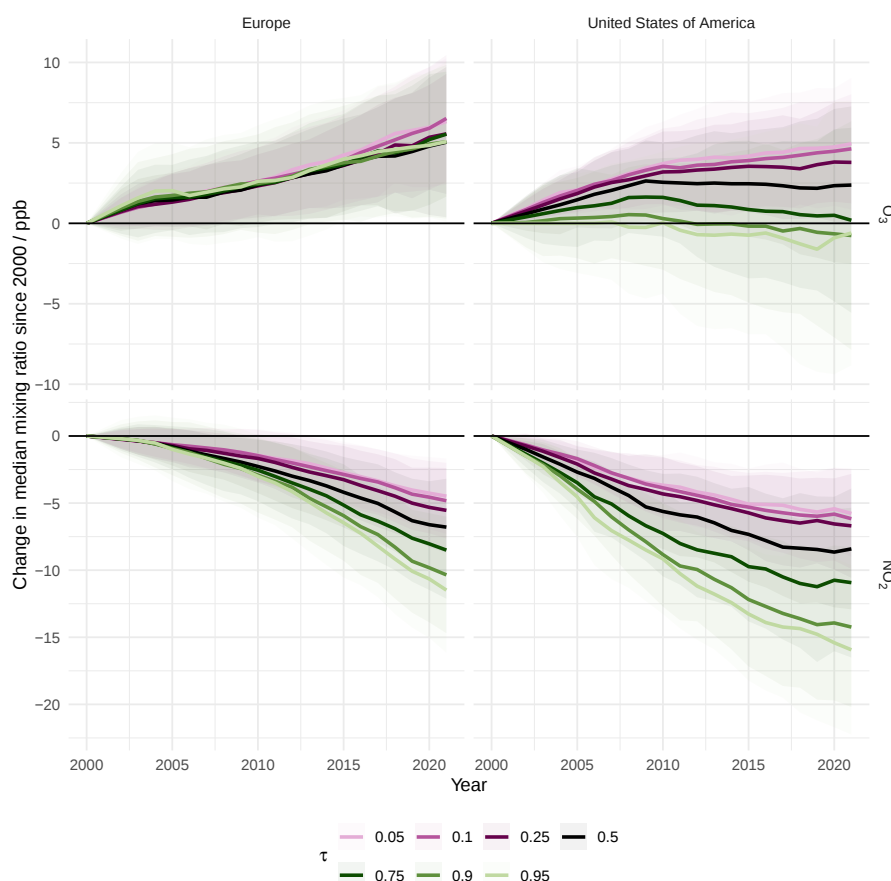


Figure 7. Absolute change in trend line-derived median NO₂ and O₃ mixing ratios for Europe and the USA, coloured by trend line percentile. The shaded region represents the median absolute deviation.

Taking a closer look at the annual magnitude and direction of the trends allows for a closer inspection of how O₃ and NO₂ trends have changed between 2000 and 2021 for each τ value (Figure 8). For Europe, all median O₃ trends were positive for all τ values across all years. For higher τ values ($\tau = 0.75, 0.90$ and 0.95), the slope dropped sharply from ca. 0.54 ppb yr^{-1} to ca. 0.07 ppb yr^{-1} ($\tau = 0.95$) between 2000 and 2004. A smaller decrease in slope was observed for the lower τ cases (from ca. 0.35 ppb yr^{-1} to 0.18 ppb yr^{-1} for the $\tau = 0.05$ case). From 2005 onward, the slope in the O₃ trend generally continued to increase up to 2021. In the lower τ cases, the median slope returned to 2000-2002 levels in 2021. For the higher τ cases, the magnitude of the median slope was between ca. $0.29 - 0.46 \text{ ppb yr}^{-1}$ lower in 2021 than 2000. In the USA, a larger spread in the changing median slopes was observed between different τ values. Generally, and in contrast to Europe, the median slope in O₃ steadily decreased between 2000 and 2014, before plateauing between 2014 to 2021. For the lower τ values, the slope in

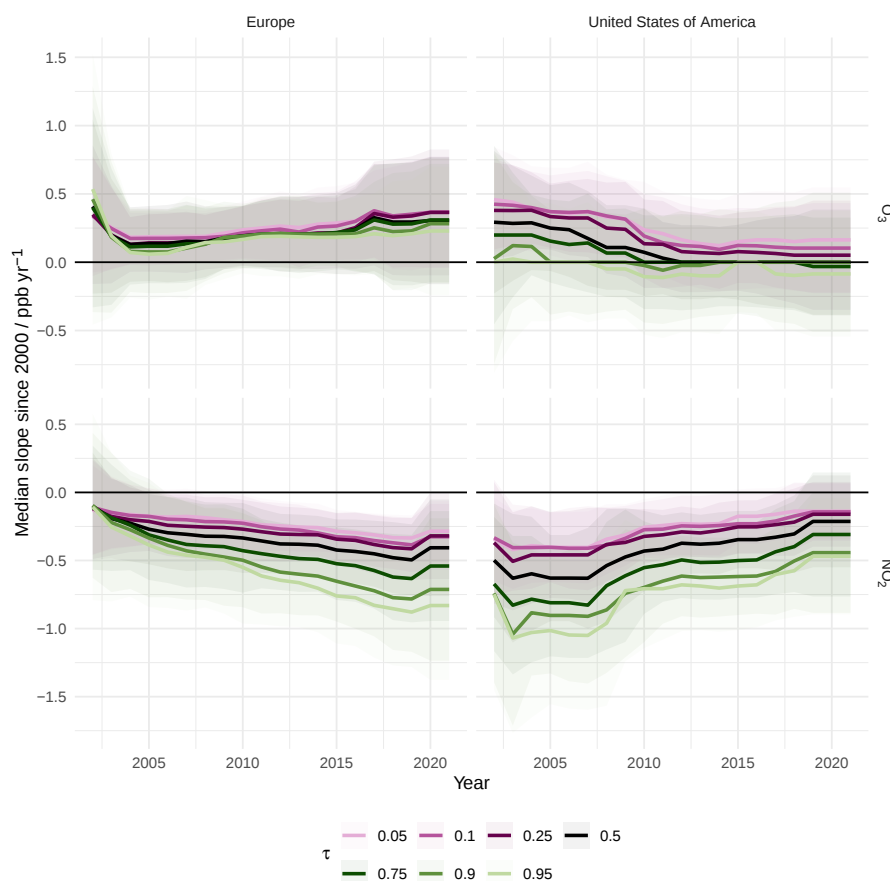


Figure 8. Median trend line slopes (ppb yr^{-1}) in NO_2 and O_3 for Europe and the USA, coloured by trend line τ value. The shaded region represents the median absolute deviation.

O_3 remained positive between 2000 and 2021, but reduced by ca. 0.34 ppb yr^{-1} between 2000 and 2014 ($\tau = 0.05$). Trends in the higher τ cases were positive at the beginning of the century, with a transition to a negative trend observed for $\tau = 0.75$ (in 2013), and $\tau = 0.90$ and 0.95 (both in 2007). In the $\tau = 0.50$ case, median trends in O_3 plateau around 0 ppb yr^{-1} from 2012, indicating no overall direction of trend.

Trends in NO_2 in Europe and the USA between 2000 and 2021 are also contrasting. In both Europe and the USA, median trends remain negative between 2000 and 2021, indicating NO_2 is decreasing in both cases, as observed in Figure 7. However, the direction of the trend is different in each region. In Europe, the slope of the trend is becoming increasingly more negative with time, particularly for the higher τ cases (reduction of ca. 0.72 ppb yr^{-1} for the $\tau = 0.95$ case). However, in the USA the negative trends are becoming increasingly more positive (or less negative) between 2000 and 2021 (0.29 ppb yr^{-1} higher, $\tau = 0.95$).



3.3 Spatio-temporal Distribution of Change points

The direction and magnitude of the change points in each location were investigated to identify locations where a change in direction of the trend (i.e. a negative to positive trend) occurs. In Europe, 21 sites observed a switch from a negative O₃ trend line to a positive one in the first change point. The biggest switches occurred in southern Europe, in Spain and Greece ($\Delta 3.28$ ppb in *gr0022a*, 2004). For a larger number of sites (41), mainly in central Europe, the first change point in O₃ switched from positive to negative, up to a maximum swing of 4.55 ppb (*fr31013*, 2003). In contrast, a larger number of sites observed a negative to positive switch (39) in the second change point, compared to a positive to negative switch (28). Eight of the top ten biggest switches from negative to positive in the second change point occurred in Spain and Italy ($\Delta 3.99$ ppb in *es0124a*), whereas the biggest switch from positive to negative showed no distinct spatial pattern. This suggests that generally across Europe, more trends in O₃ were switching from positive to negative in the first 5-10 years of the 21st-century, whereas more trends were switching from negative to positive between 2010-2021. In both decades, southern European sites showed the biggest changes in O₃ trend from negative to positive. Similarly to O₃, more sites showed a positive to negative trend (84) in NO₂ compared to negative to positive (28) for the first change point, with the largest change points again observed in Spain ($\Delta 3.25$ ppb, *es1453a*). A comparable number of sites showed changes in trend direction in the second change point, (43 negative to positive, and 33 positive to negative). The positive to negative trends were generally distributed around 2010-2014, whereas the vast majority of negative to positive switches occurred in 2020 (41 out of 43 sites). The cause of this switch in 2020 across Europe is discussion in further detail in section 3.3.1.

In the USA, a comparable number of directional switches in O₃ trend in the first change point occurred in the positive to negative direction (21), compared to negative to positive (17). The three largest magnitude trend changes from negative to positive occurred in Florida ($\Delta 2.43$ ppb, *15292*), with all of the ten highest magnitude cases in either Florida or California. Trends switching from positive to negative in the first change point scenario were smaller in magnitude, the biggest switch being in California ($\Delta 1.53$ ppb, *8197*), but with some noticeable contributions from the central states. The majority of second change points occurred between 2010-2019, though it is important to note that due to a lack of data availability, it is not possible for this analysis to pick out change points from 2020 onward in the USA dataset. In total, 14 sites switched from positive to negative, with a maximum change of $\Delta 2.25$ ppb (*8169*, 2019). A comparable number of sites (12) showed trends switching from negative to positive, with a maximum change of $\Delta 2.46$ ppb (*1959*, 2019). In the NO₂ dataset, there is only one site showing a switch from negative to positive in the first change point ($\Delta 1.30$ ppb, *1369*, 2008), compared to 13 sites showing a positive to negative switch. Of these 13 sites, the largest magnitude is in Virginia ($\Delta 2.99$ ppb, *15024*, 2003). In contrast, 19 sites show a switch in NO₂ trend from negative to positive after the second change point, with the biggest change in California ($\Delta 2.16$ ppb, *1206*, 2019), with only one site showing a switch from positive to negative ($\Delta 0.48$ ppb, *1369*, 2013).

Generally, the largest changes from negative to positive O₃ trends in the USA were in California and Florida in the first half of the 20 year period, with the reversal of this trend seen across California and Florida in the second change point. 8/14 and 4/14 of the sites showing a positive to negative transition in the second change point were located in California and Florida

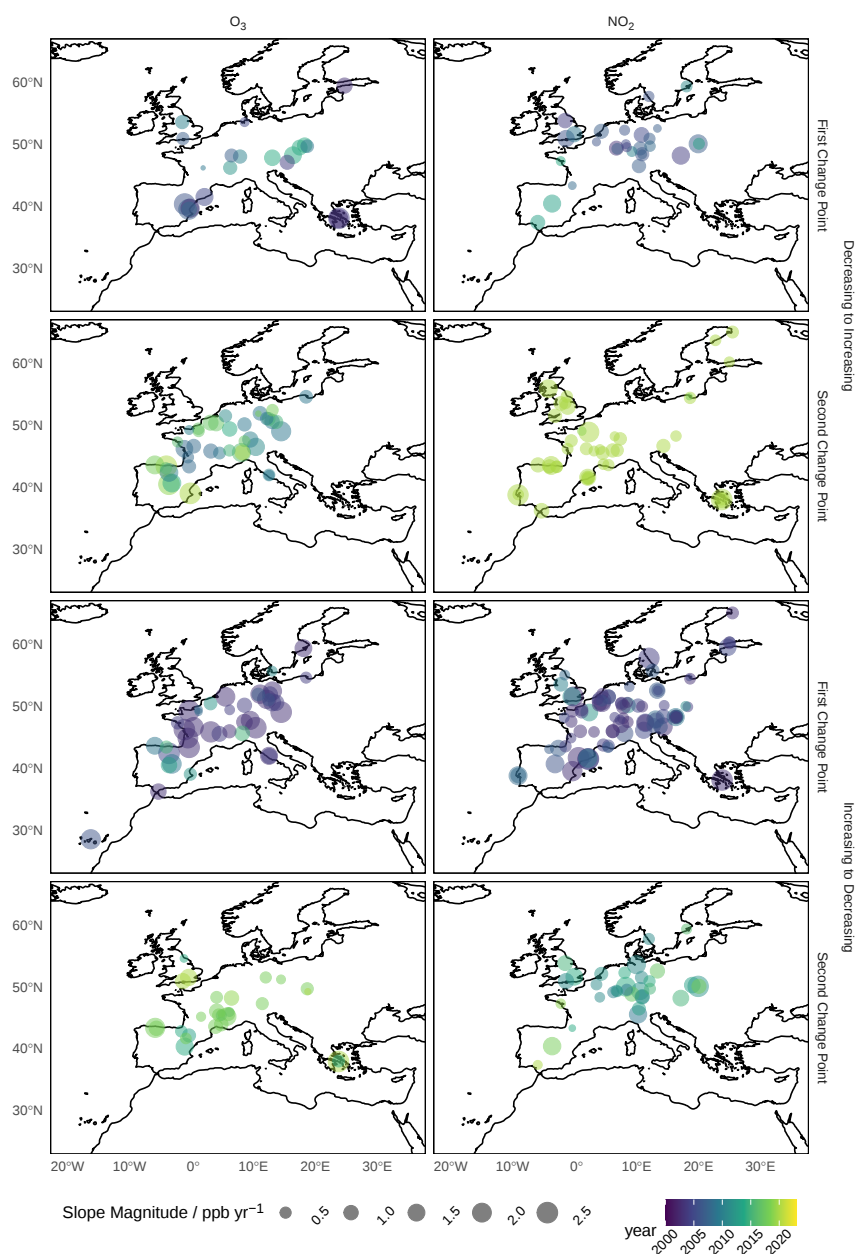


Figure 9. European sites with a change in direction of slope (O₃ - left, NO₂ - right) of the first (rows 1, 3) second (rows 2, 4) change points. The size of the slope is relative to the magnitude of the change in slope (slopes > 2.5 ppb yr⁻¹ have been clamped), coloured by the year of the change point. Negative to positive change points (rows 1, 2) are presented separately to positive to negative change points (rows 3, 4).

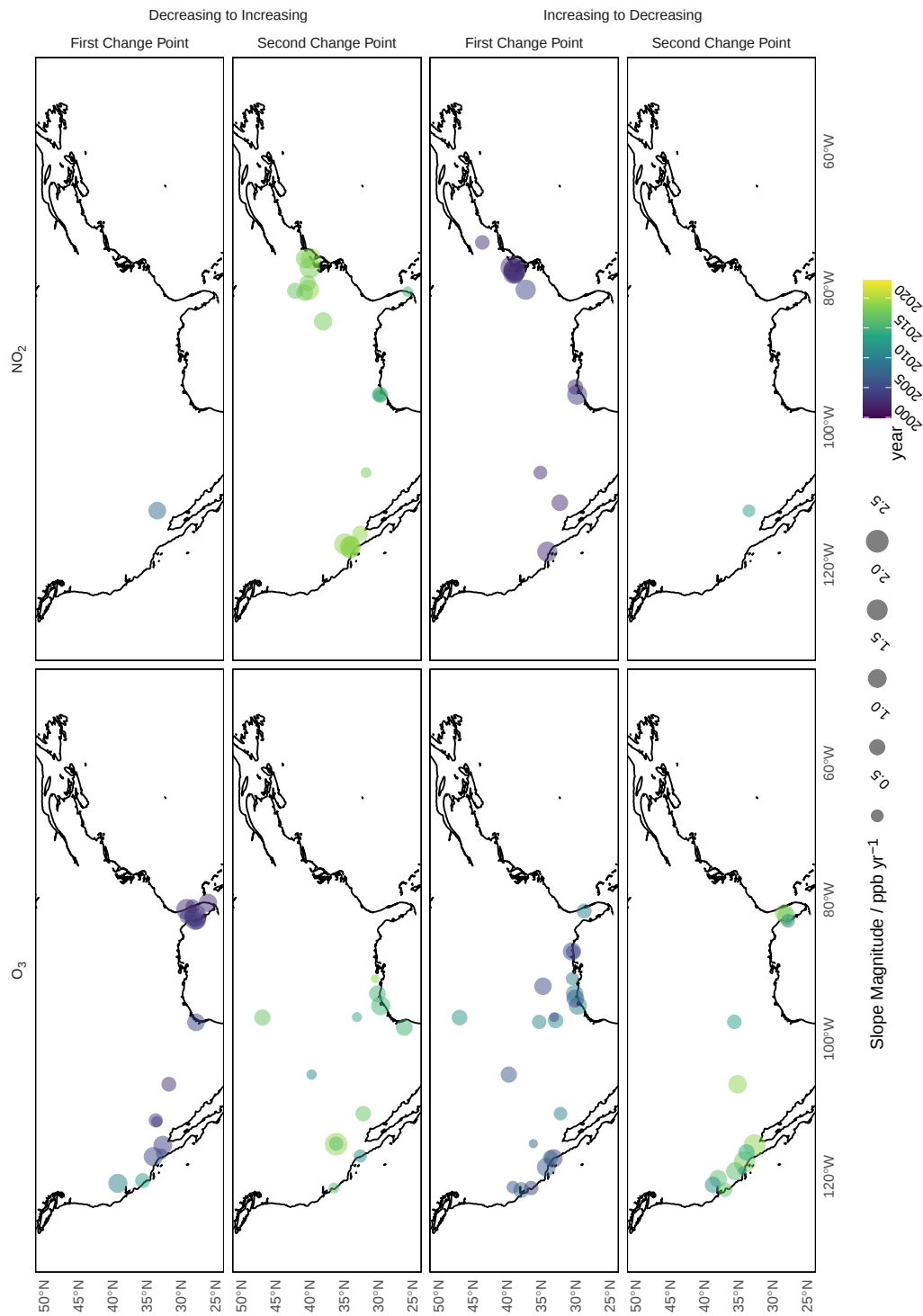


Figure 10. US sites with a change in direction of slope (O_3 - left, NO_2 - right) of the first (rows 1, 3) second (rows 2, 4) change points. The size of the slope is relative to the magnitude of the change in slope (slopes $> 2.5 \text{ ppb yr}^{-1}$ have been clamped), coloured by the year of the change point. Negative to positive change points (rows 1, 2) are presented separately to positive to negative change points (rows 3, 4).



respectively. This is important, since a previous study highlighted that 4th highest daily maximum 8-hour O₃ (4MDA8) values between 2010-2014 were particularly high in California (Fleming et al., 2018).

To supplement this analysis, the 4MDA8 was calculated using the mixing ratio data for each site and slicing the fourth highest value on an annual basis. 4MDA8 is an important metric used by the US for determining compliance with the National Ambient
 250 Air Quality Standards for Ozone (Environment Protection Agency, 2015; Berrocal et al., 2014). To allow for comparison with previous studies, only the 6-month warm season (April - September) was used to calculate 4MDA8 values, which is a reasonable approach in the USA and Europe since the highest O₃ values are likely to occur in the warmest months.

Figure 11 shows how the 4MDA8 value varies between 2000, 2007, 2014 and 2021 in the USA. Across the US, 33 sites had a 4MDA8 $\geq$ 85 in 2000, compared to 19 in 2007 and 6 in 2014. In 2016 there was an up-tick to 10 sites with 4MDA8 values at
 255 this level. Of these 10 sites, all are located in California, compared to 20/33 Californian sites in 2000 and 18/19 in 2007. All 11 sites in 2021 with 4MDA8 $\geq$ 85 were located in California. This highlights that although fewer Californian sites are exceeding 4MDA8 $\geq$ 85 ppb with time over the 2-decade period, the Californian 4MDA8 "hotspot" identified in Fleming et al. (2018) is still present. The more general picture across the USA is that 4MDA8 values are coming down with time, particularly strongly in the central and Eastern portion of the USA but less successfully in the west.

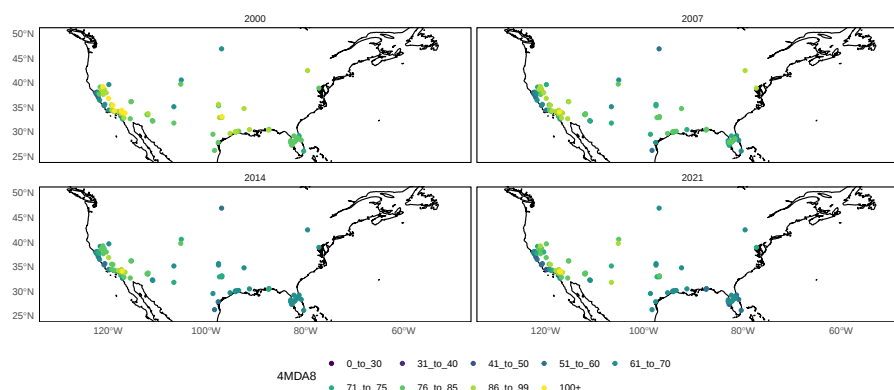


Figure 11. 4MDA8 values in ppb for sites in the United States in 2000, 2007, 2014 and 2021

260 Southern Europe, particularly Greece and Northern Italy, were also highlighted in Fleming et al. (2018) as being 4MDA8 hotspots. Figure 12 shows how the 4MDA8 values are changing between 2000, 2007, 2014 and 2021 across Europe. In 2000, there were 6 sites in which 4MDA8 $\geq$ 85 ppb. This slightly increased to 7 sites in 2007 before decreasing to 4 sites in 2014.



By 2021, no sites had 4MDA8 values exceeding 85 ppb. Since the measurement sites in this dataset are not evenly distributed across the USA and Europe, it is difficult to attribute high 4MDA8 values to particular states or countries. However, it was noticeable that a large proportion of the sites were 4MDA8 ≥ 85 ppb were in Italy and Greece. In 2000 3/6 of high 4MDA8 sites were located in Italy, and 1/6 in Greece. In 2007, 3/7 were located in Italy and 1/7 in Greece; and in 2014 3/4 sites were located in Italy. Consistent with the Fleming et al. (2018) study, the southern European 4MDA8 hotspots are apparent throughout the 20-year period.

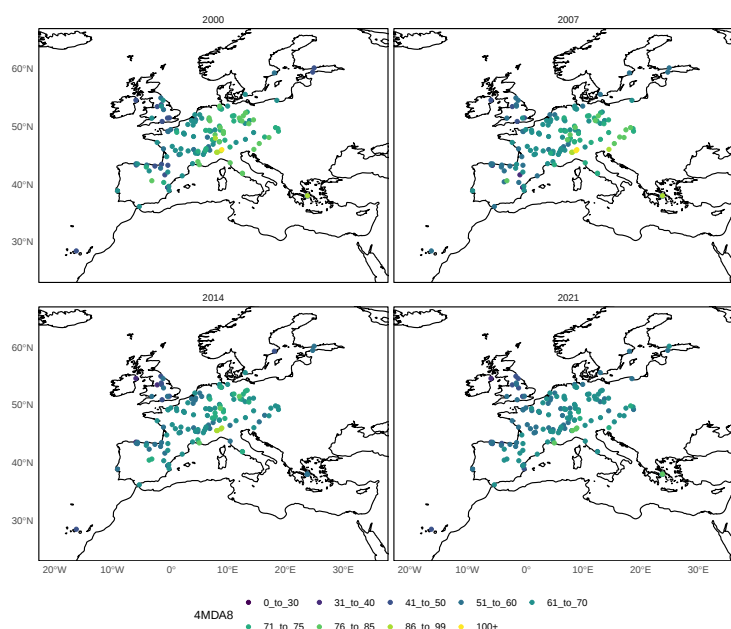


Figure 12. 4MDA8 values in ppb for sites in Europe in 2000, 2007, 2014 and 2021

3.3.1 2020 In Europe

As noted in section 3.1.1, in 2020 in Europe several sites switched from a long term decrease in NO_2 to increasing NO_2 again. The public health measures enacted due to the COVID-19 pandemic had the side effect of decreasing NO_2 concentrations due to reduced traffic emissions. The removal of these measures led to an increase in NO_2 mixing ratios as normal activities resumed, resulting in a positive trend in NO_2 concentrations up from their lowest during the restrictions. There are 42 sites that had a negative trend in 2019 and a non-negative trend in 2020. The 2019 trends ranged from 1.47 to $-0.04 \text{ ppb yr}^{-1}$ and the 2020 trends ranged from 0.00 to 3.74 ppb yr^{-1} . There were two sites that already had non-negative trends (*fr25039* and *pl0048a*) by 2020, but neither were strong or significant ($p > 0.33$). The effect of this on O_3 concentrations at the sites is varied. 21 of the 42 sites also measured O_3 , though only 3 (*es1038a*, *fr33120* and *gr0031a*) also had 2020 as one of the change points for the PQR. To compare the 2020 onwards trends, new PQRs for each site were selected by requiring the second change point



to occur in 2020, and then selecting where the first occurred on a time series by time series basis by minimising the AIC as
 280 before. Furthermore, only sites where the resulting slope for the 2020 onwards O_3 trend had p values < 0.33 were selected.
 One final site (*gr0031a*) was removed as although it meets the data coverage criteria for the main analysis, its missing data
 is focused around 2019 and 2020 which has a strong influence on the trends derived. This led to 15 sites remaining for this
 case study. The resulting alternative trends for these sites are shown in figure 13. Additionally, as the focus is on sites with
 both NO_2 and O_3 , O_x is displayed. If a site is under a VOC limited regime, the trend in O_x provides an indication whether any
 285 changes in O_3 are only due to changes in NO_2 , or other factors. Whether a change in NO_2 concentrations results in an increase
 or decrease in O_3 concentrations depends on the if a site is sensitive to locally produced O_3 (which an urban site, in general,
 would be expected to be), and what photochemical regime (NO_x or VOC limited) the site is under. For example, in a VOC
 limited regime, if NO_2 increases, O_3 decreases but O_x remains the same, the change in O_3 can be attributed to the change in
 NO_2 . If NO_2 and O_3 increase, O_x will also increase, indicating a NO_x limited regime. 14 of the sites saw increasing O_3 with
 290 increasing NO_2 between 2020 and 2023 - this is expected in NO_x limited regimes. Due to the need for both O_3 and NO_2 to
 be measured at both sites 13 of the sites were urban background and 2 were urban traffic, as O_3 is less frequently measured at
 traffic sites. Urban background sites are more likely to be under NO_x limited regimes as they are, by design, situated further
 from major roads, which are still a significant source of NO_x . This does not necessarily mean that urban areas in Europe are
 completely NO_x limited and will depend on the specific location as to whether an urban traffic or urban background site is
 295 more representative of a location. At one urban traffic site (*es1529a*) has seen decreasing O_3 with increasing NO_2 over the
 same period. This is more indicative of a VOC limited regime, and is more similar to the overall trends seen across Europe in
 the 21st century, with more increasing O_3 trends in 2015-2021 than in 2000-2004.

The sites identified by this method do not appear to have resumed their pre-2020 trends, though this should be verified
 when sufficient data has been collected post-2020 where change points will have more freedom to occur. The pattern of sites
 300 switching to an increasing trend in NO_2 will have varied effects on urban O_3 in Europe if they continue, though as background
 sites are designed to be more representative of residential areas, which make up a large portion of urban areas then it could be
 expected to see urban O_3 continue to increase.

3.3.2 Recent NO_2 in the USA

In the final years of the USA NO_2 dataset, an up-tick the number of positive NO_2 trends can be seen. From 2015 onwards there
 305 is an increasing number of sites with this trend reaching 17 sites trends (16 high, 1 medium certainty) in 2021, up from 0 in
 2014. Anomaly time series and trends of can be viewed in figure 14. Trends have reversed from $-0.98 - -0.03$ ppb yr^{-1} to 0.1
 $- 1.78$ ppb yr^{-1} . Unfortunately, only 4 of the 17 sites have concurrent measurements of O_3 so it is difficult to investigate this
 changing trend in NO_2 from the time series analysed in this study. Only one site (8169) O_3 trend has its original second change
 point occurring alongside its NO_2 increasing change point, so the same method has been applied as in 3.3.1 where the second
 310 change point has been forced to occur in the same year as the NO_2 change point. Doing this reveals that sites 8169, 14386 and
 15197 have reasonably significant ($p = 0.01, 0.15, 0.002$ respectively) O_3 trends after change point 2, when this change point is
 the same as that in NO_2 . 8169 and 14386 have decreasing O_3 trends (-1.70 and -0.55 ppb yr^{-1}) with increasing NO_2 and 15197

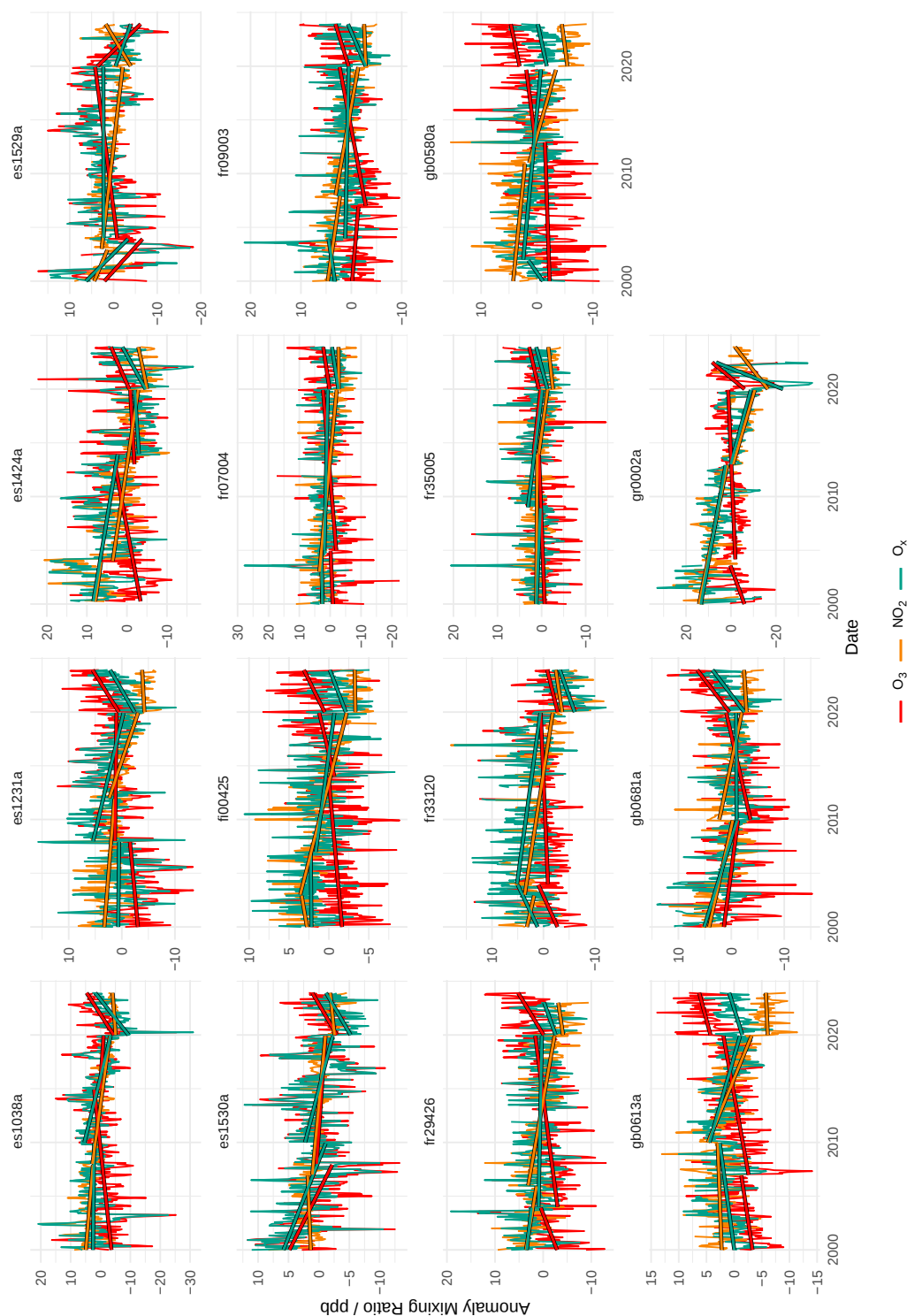


Figure 13. Sites in Europe that had decreasing NO₂ in 2019 but increasing NO₂ from 2020 onwards. Monthly averaged O₃ (red), NO₂ (orange) and O_x (green) anomaly are shown. O₃ and O_x trends differ from those presented elsewhere in the manuscript as the piecewise quantile regression where the second change point has been restricted to be in 2020.



saw increasing O_3 at 1.15 ppb yr^{-1} . These data suggest that the O_3 concentrations will be sensitive to this increase in NO_2 , but without the co-located O_3 measurement it does not provide definitive information on the direction on any changes. Indeed
 315 Jhun et al. (2015) note that reductions in NO_x suggested that the 99.9th quantile O_3 concentrations were also reduced between 1994 and 2010. While this study limits itself to the 95th quantile, there is some indication in figure 7 that there has been a small increase in high O_3 levels during the time increased NO_2 is observed.

4 Conclusions

Trends in urban O_3 and NO_2 were calculated from de-seasoned monitoring site data across both Europe and the USA. A
 320 quantile regression analysis was utilised to assess long-term trends, with the method extended to a piecewise approach. This allowed for some freedom to capture changes in a complex dataset, whilst being able to describe the trends with a small number of coefficients. Break points were limited to two per time series to keep the focus on long-term trends and the largest changes. One limitation to this approach is that time series with 3 or more large changes will not be well described by this method - however, after inspection of the regressions and comparison with a LOESS, two points were found to capture the
 325 structure well. The trends constructed from the piecewise regressions were evaluated by their statistical significance, and those poorly described using this methodology ranked lower in significance and were subsequently investigated less in the analysis. Piecewise segments were required to have ≥ 2 years of data, which limited the ability to define the most recent changes in a given time series, and such is why only Europe was analysed for the effects of the COVID-19 restrictions.

In Europe, more sites were found to have an increasing O_3 trend between 2015-2021, compared to 2000-2004, with NO_2
 330 trends showing the reverse effect (fewer sites increasing, and more decreasing in 2015-2021 compared to 2000- 2004). This broadly suggests that a VOC-limited relationship for O_3 formation could be common across urban sites in Europe. In the USA, the reverse is true, with a reduction in the number of sites with increasing O_3 trends in 2015-2021 compared with the 2000-2004 period, but with more sites with increasing NO_2 in 2015-2021 compared with 2000-2004. The majority of O_3 trends are positive and of high certainty ($p \leq 0.05$) in Europe across the 20-year period. This is also broadly true for the USA, but
 335 the proportion of sites showing this trend reduces with time, alongside an increase in the number of sites with high-certainty negative trends in O_3 between 2010-2021. Previous studies have attributed positive urban O_3 trends in the USA to increasing winter O_3 concentrations (Simon et al., 2015). However, differences in seasonality would not be observed in this analysis as the trends are derived from de-seasoned data. From the trends described here, the median ($\pm$ median absolute deviation) change in O_3 over the last two decades were calculated as $5.0 \pm 4.6 \text{ ppb}$ in Europe and $2.4 \pm 4.9 \text{ ppb}$ in the USA, with corresponding
 340 changes in NO_2 as -6.8 ± 3.6 and -8.4 ppb .

Given the nature of its long lifetime and the complexity of its chemistry, when describing trends in O_3 it is important to look beyond the 50th percentile. A study of European ozone trends between 1995-2014 found that whilst the 50th percentile data did not show significant trends, significant trends were observed in the 5th and 95th percentiles (τ values) (Yan et al., 2018). An exploration of the τ values in this study revealed smaller reductions in trend-derived change in O_3 mixing ratios in 2021 since
 345 2000 for the lower τ values compared to higher values in Europe. This, coupled with the fact that generally O_3 trends across

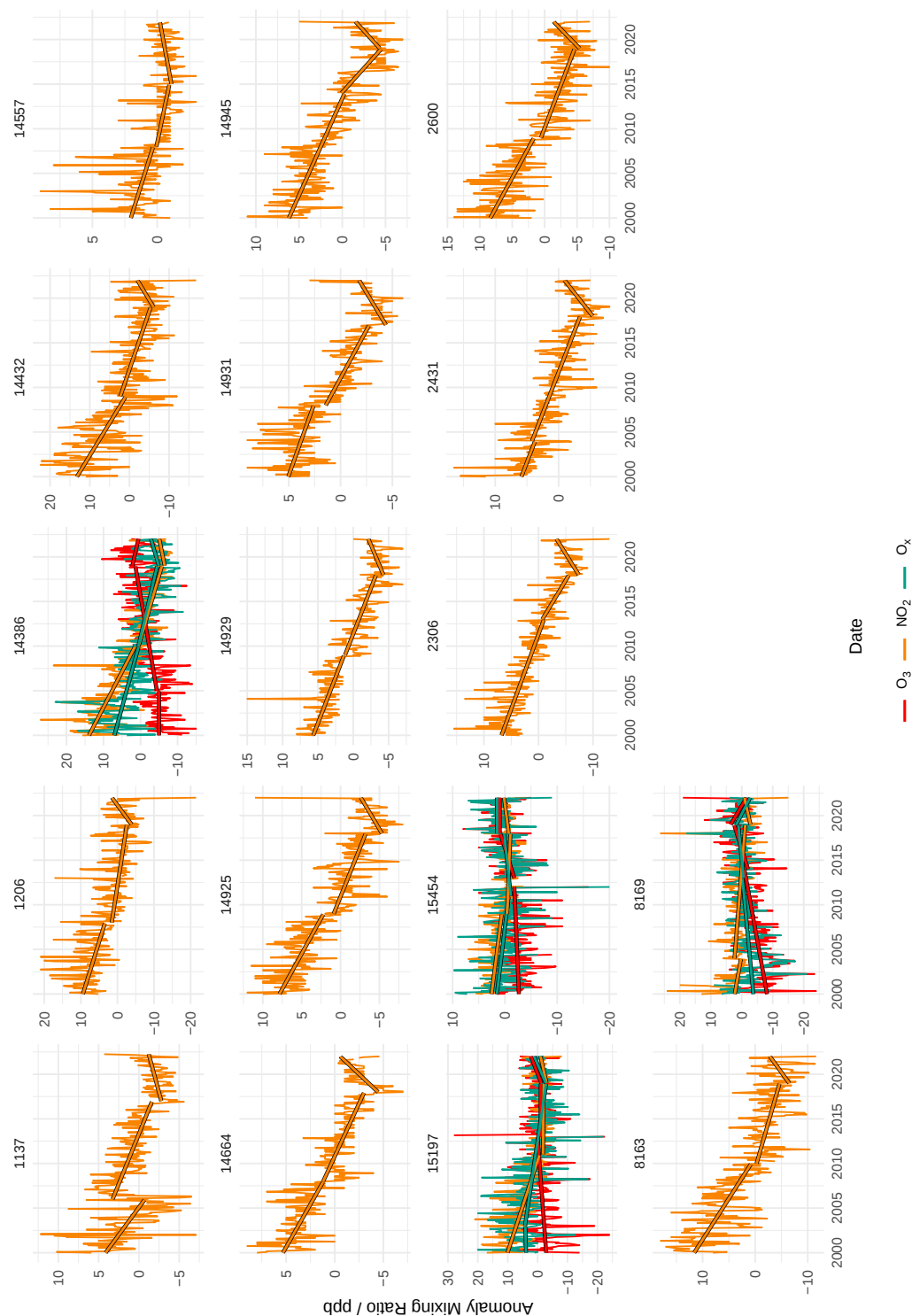


Figure 14. Sites in the USA that had increasing NO₂ since 2015. Monthly averaged O₃ (red), NO₂ (orange) and O_x (green) anomaly are shown. O₃ and O_x trends differ from those presented elsewhere in the manuscript as the piecewise quantile regression where the second change point has been restricted to match that of the NO₂ at each site.



Europe are positive, suggests that the lowest ambient O_3 mixing ratios are increasing more rapidly than the highest values. This is consistent with the findings of Yan et al. (2018), who found that although peak O_3 concentrations were coming down across urban and suburban sites between 1995 - 2014 in Europe, an increase in the lower O_3 concentrations was observed in more than 75 % of sites.

350 An assessment of the spatio-temporal distribution of the first and second change points revealed more sites with a directional switch in O_3 trend across Europe were in the positive to negative direction in the first change point, but with more sites with a negative to positive second change point. In the USA dataset, the biggest switches from negative to positive in the first change point occurred in California and Florida, but a reversal of this trend from positive to negative was also observed across these states in the second change point. To supplement this, a calculation of the 4th highest daily maximum 8-hour running mean
355 for O_3 (4MDA8) revealed high 4MDA8 hotspots in parts of southern Europe, but particularly in California, consistent with previous findings of Fleming et al. (2018). Furthermore, the USA has seen significant increasing trends in NO_2 since 2015 at several sites. However, there are limited sites that have been used in this study with co-located NO_2 and O_3 , and the O_3 response is not consistent. This is not unexpected due to the wide range of conditions that an 'urban' site can be found in (e.g. variations in local VOC concentrations), but it should be expected that in some locations O_3 will be sensitive to this increase
360 in NO_2 , especially if it persists.

The impact of COVID-19 on ambient mixing ratios of NO_2 is well studied, with one previous study estimating that NO_2 concentrations across urban background sites in Europe were 32 % lower than expected in 2020 (Grange et al., 2021). The impact of this COVID-19 effect was observed in the trend analysis of Europe described in this study, with the vast majority of directionally switching second change points in NO_2 occurring in 2020 in the negative to positive direction. This can be
365 attributed to lower ambient NO_2 mixing ratios in 2020, leading to an increasing trend in the subsequent years as business-as-usual conditions resumed. These increasing NO_2 concentrations have in some cases been accompanied by increasing O_3/O_x and should continue to be studied, since in many cases the prior downward trend in NO_2 has not resumed.



Code and data availability. All data for this study can be downloaded from the relevant databases. Code for performing the download as well as the analysis can be found at [10.5281/zenodo.14538198](https://zenodo.org/doi/10.5281/zenodo.14538198) (Drysdale and Nelson, 2024)

370 *Author contributions.* BSN and WSD equally contributed to all aspects of this manuscript's production

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