



Recent ozone trends and changepoints in the Greater Region: The role of meteorology and NO_x emission reductions

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Abstract. We analysed long-term records of NO₂ and O₃ in Luxembourg and the Greater Region with the Bayesian Estimator of Abrupt change, Seasonality, and Trend (BEAST) to identify probabilistic changepoints in time series. Seasonal components of NO₂ and long-term components of O₃ revealed a significant changepoint in 2017/2018 that is more prominent than the effect of the short-term emission reductions in 2020/2021 and the subsequent post-Covid rebound in urban areas. NO₂ concentrations decrease by at least 50 % for all station categories from 2009 to 2024. Linear trend slopes for O₃ during this period based on quantile regression and Theil-Sen trend estimator were positive, with highest values at urban stations (~0.75 µg m⁻³ yr⁻¹), while increases at peri-urban, rural, and forest sites were smaller (~0.4–0.6 µg m⁻³ yr⁻¹). Spring and autumn are the dominant seasons contributing to the O₃ increase. The decline of the positive weekend effect (higher O₃ on Saturdays and Sundays than on weekdays), together with the reduced MDA8 O₃ sensitivity to maximal air temperatures above ~20 °C after 2018, indicates an ongoing transition from predominantly VOC-limited towards NO_x-limited chemistry, particularly at peri-urban, rural, and forest stations during summer. However, our results suggest that the impact of reduced O₃ titration by NO, particularly at low to moderate temperatures has sustained increasing O₃ in the Greater Region. At the same time, the reduction in peak O₃ due the influence of NO_x-limited chemistry was partly offset by the increase in daytime maximum boundary layer height observed after the 2017/2018 changepoint.



25 1 Introduction

Tropospheric ozone (O_3) is a secondary air pollutant that plays a central role in tropospheric chemistry, acts as a potent greenhouse gas, adversely affects terrestrial ecosystems, and poses significant risks to human health (Fuhrer et al., 1997; Grulke and Heath, 2020; Malashock et al., 2022; Brauer et al., 2016; Forster et al., 2007). Particularly, under future climate conditions the impacts of O_3 on health during heat waves and droughts would likely confound the health burdens from the drought itself (Lee et al., 2023). It is formed by photochemical oxidation of carbon monoxide (CO), methane (CH_4), and non-methane volatile organic compounds (NMVOCs) in the presence of nitrogen oxides (NO_x , the sum of nitrogen monoxide, NO, and nitrogen dioxide, NO_2). NO_x acts as a key catalyst in the formation of tropospheric O_3 (Crutzen and Lelieveld, 2001; Monks et al., 2015). Depending on the relative availability of NO_x and VOC concentrations, the O_3 formation regime is typically divided into three categories: NO_x -limited, transitional, and NO_x -saturated (VOC-limited), each of which governs the rate of O_3 production (Jung et al., 2022; Sillman, 1999). The diurnal, seasonal and long-term O_3 variations observed at a surface station represents the net effect of several physical, biological and photochemical processes, with the latter being characterized by a non-linear response to precursor emissions (Trebs et al., 2012; Yan et al., 2018; Monks et al., 2015). The overall processes include:

- regional photochemical O_3 formation (Sillman, 1999; Crutzen and Lelieveld, 2001)
- direct emission sources (titration by nearby NO and VOC emissions) (Sillman, 1999; Crutzen and Lelieveld, 2001)
- synoptic and local meteorological conditions, with dependence on solar radiation, air temperature and boundary layer height (Real et al., 2024; Otero et al., 2021; Adame et al., 2022; Haman et al., 2014)
- removal from the atmosphere by dry deposition to surfaces and vegetation (Clifton et al., 2020; Anav et al., 2019)
- stratosphere-troposphere exchange or long-range (hemispheric) transport, which contribute to background O_3 (Monks et al., 2015; Ansari et al., 2025; Fiore et al., 2014)

Climatic variability largely regulates the inter-annual variations of surface O_3 in Europe (Yan et al., 2018) and there is broad scientific consensus that rising air temperatures associated with climate change can partly offset emission-driven O_3 reductions through the so-called climate penalty (Colette et al., 2015). Despite the exceptionally hot summers and severe heatwaves of the last decade, the frequency of large O_3 episodes has declined since 2015, even though the total number of O_3 episodes increased, indicating a shift towards more frequent but less intense O_3 pollution episodes (Crespo-Miguel et al., 2024; Zohdirad et al., 2022). European emission reductions targeting the main O_3 formation precursors, such as NMVOCs and NO_x , implemented over the last three decades (Macdonald et al., 2021; Ojeda-Castillo et al., 2025; Ordóñez et al., 2020; Thürkow et al., 2023; Eea, 2025a, b; Fortems-Cheiney et al., 2024) resulted in declining summer peak O_3 levels, especially in rural areas (Derwent et al., 2010; Monks et al., 2015; Zohdirad et al., 2022; Yan et al., 2019), particularly after the period 1995-2000 (Yan et al., 2019; Boleti et al., 2020). Concurrent with the ongoing long-term NO_2 decline in Western and Central Europe, additional reductions were reported across Europe during the COVID-19 lockdown period (Varga et al., 2024; Grange et al., 2021; Barré et al., 2021; Ordóñez et al., 2020), particularly in traffic-influenced urban environments. However,



at urban and suburban locations measures to reduce NO₂ are counterproductive as indicated by increases in O₃ resulting in relatively constant or even increasing total oxidant (OX = NO_x + O₃) concentrations (Grange et al., 2021; Real et al., 2024).

60 Long-term increases in urban and suburban mean O₃ despite declining NO_x emissions are frequently attributed to reduced NO titration, whereby lower ambient NO decreases local O₃ destruction, especially at traffic-influenced sites (Geddes et al., 2009; Ansari et al., 2025; Monks et al., 2015; Colette, 2024; Real et al., 2024; Boleti et al., 2020; Nelson and Drysdale, 2025; Wang et al., 2025). This substantial O₃ increase at traffic sites reaches almost 20 % in Europe during the period 2005-2019, while the trend was near zero for rural sites (Solberg et al., 2022). However, mean O₃ levels did not decrease as much

65 as expected and much less than precursor emissions because of the nonlinear response of O₃ to precursor changes and the increase in background O₃ over Europe due to hemispheric transport (Colette, 2024; Real et al., 2024; Crespo-Miguel et al., 2024; Monks et al., 2015; Solberg et al., 2022; Ansari et al., 2025; Wang et al., 2025). Concentrations still often exceed the EU limit values or WHO guidelines during hot summer periods (Boleti et al., 2018; Yan et al., 2019). Additionally, long-term trends are characterized by increasing seasonal O₃ average concentrations in the 5th percentile, especially attributed to

70 spring and winter periods (Yan et al., 2019; Boleti et al., 2020; Yan et al., 2018; Nelson and Drysdale, 2025). As NO_x reductions continue the chemical regime in some regions and urban locations is expected to transition from VOC-limited to NO_x-limited (Fadnavis et al., 2025; Wang et al., 2025). On the other hand, positive O₃ anomalies under NO₂ reduction scenarios were associated with elevated air temperatures in the Benelux, Germany and France (Ordóñez et al., 2020). A very recent study covering the period 2000-2024 reports decreasing NO_x levels at all stations in Switzerland as well O₃ decreases

75 at rural and background stations, while O₃ increased at traffic sites (Nussbaumer et al., 2026). The study showed that O₃ formation at rural background sites was recently dominated by NO_x-sensitive photochemistry, while NO titration remained the dominant process controlling O₃ concentrations at traffic sites. Only a limited number of studies have investigated changepoints in long-term O₃ trends across Europe (e.g., Boleti et al., 2020; Nelson and Drysdale, 2025) and the underlying drivers of these abrupt changes remain poorly understood. Previous studies also often focussed on urban trends because of

80 their direct relevance to human exposure and public health (e.g., Nelson and Drysdale, 2025), whereas substantially less attention has been given to rural, and especially forest environments.

The new Ambient Air Quality Directive (DIRECTIVE (EU) 2024/2881), revised in October 2024, tightened the O₃ target value for the protection of human health from 2030 onward by retaining the maximum daily 8-hour average concentration (MDA8) of 120 µg m⁻³, while reducing the permitted exceedance frequency from 25 to 18 days per year (averaged over three

85 years), applying in 2030. It also establishes a long-term objective for 2050 of 100 µg m⁻³, with no more than three exceedance days per year. The Greater Region is a cross-border area in Western Europe, covering Luxembourg and neighbouring regions of three countries (Belgium, France and Germany). It is an underexplored region in terms of long-term air quality studies and exhibits characteristics that make it unique for such an analysis. Amongst all EU member states, Luxembourg has not only the largest NO_x reductions in the last two decades (82% from 2005 until 2023) (Eea, 2025a), but

90 also the highest number of cross-border commuters in the EU (over 230,000 people by late 2024, <https://center-border-studies.uni-gr.eu/index.php/en/taxonomy/term/110>). They commute daily from the neighbouring countries, and the overall



cross-border transport is additionally affected by fuel tourism. Luxembourg has the highest car density and the highest per capita CO₂ emissions from transport in the EU (Eibinger and Fernando, 2026). On the other hand, it was the first country that made public transport free of charge (since March 2020). This study analyses 25 years (2000–2024) of in-situ observations of NO₂ and O₃ for 15 surface stations in the Greater Region, with particular focus on the recent period 2009–2024. The main scientific objectives are to (a) characterize the long-term evolution of NO₂ and O₃ and their coupling in the Greater Region, including seasonal and long-term trend changepoints, (b) quantify the seasonal and diurnal variability in NO₂ and O₃ trends for urban, peri urban, rural, and forest environments, including the assessment of NO_x limited versus VOC limited regimes, and (c) evaluate the influence of boundary layer height and air temperature on NO₂ and O₃ trends, changepoints, and implications for human and ecosystem O₃ exposure.

2 Methods

2.1 Measurement locations, time periods and data preparation

Concentrations of O₃ and NO₂ were obtained from permanent air quality monitoring stations operated by the national and regional environmental agencies of the four countries, which are acknowledged at the end of this paper. In all countries O₃ is measured according to EN 14625 based on the ultraviolet absorption method. NO and NO₂ are measured according to the reference method EN 14211 with photolytic conversion and chemiluminescence. The dataset with hourly time resolution used in this study comprises 15 atmospheric monitoring stations (Figure 1, Table 1) located close to the border of Luxembourg (max. distance ~ 90 km) in the Greater Region. It covers an area of 65.401 km² with about 12 million inhabitants from the territories Wallonia in Belgium, Lorraine in the French region Grand Est, Saarland and Rhineland-Palatinate in Germany as well as the Grand Duchy of Luxembourg (<https://www.granderegion.net/en/>). The selected measurement stations (Figure 1, Table 1) cover a range of environments including four urban, three peri-urban, five rural and three forested sites (clearings). Most stations have been continuously operational from 2000 until 2024. O₃ concentrations are reported in µg m⁻³ (STP: 20°C and 1013 hPa) to ensure consistency with European air quality legislation and monitoring networks, as the ambient air quality standards and target values applied in this study are defined in mass concentration units. After data retrieval, routine quality control procedures were conducted such as range-based plausibility checks and internal consistency checks. For example, negative concentration values or extremely high values that exceed known atmospheric limits are flagged (beyond plausible ambient concentrations, values for NO > 1500 µg m⁻³, NO₂ > 1000 µg m⁻³, O₃ > 500 µg m⁻³). Also, the plausibility of the seasonal cycles was analysed. Early NO₂ data, often before 2008, show seasonal irregularities and were considered as suspicious for several stations and removed from the final data set. Finally, daily (min, max, percentiles), monthly (min, max, percentiles) time aggregation was made and, subsequently, O₃ ecosystem and health metrics were computed.



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Table 1. Overview of air quality monitoring stations in the Greater Region used for this study, including coordinates, country, station category and measurement periods.

Station	Longitude	Latitude	Country	Category	Period
Beckerich	5.84716 E	49.73186 N	Luxembourg	peri-urban	11/2002 – 12/2024
Beidweiler	6.30532 E	49.72225 N	Luxembourg	rural background	10/2007 – 12/2024
Luxembourg-Bonnevoie	6.13760 E	49.59771 N	Luxembourg	urban	01/2000 – 12/2024
Esch-sur-Alzette	5.97689 E	49.50504 N	Luxembourg	urban	01/2000 – 12/2024
Jonville en Woevre	5.78556 E	49.06583 N	France	rural background	08/2001 – 12/2024
Habay La Neuve	5.62911 E	49.71987 N	Belgium	rural background	01/2000 – 12/2024
Hunsrück-Leisel	7.19372 E	49.74103 N	Germany	forest (clearing)	01/2000 – 09/2024
Metz-Centre	6.18083 E	49.11944 N	France	urban	07/2005 – 12/2024
Nancy-Charles III	6.19043 E	48.68817 N	France	urban	01/2004 – 12/2024
Sainte Ode	5.59369 E	50.02736 N	Belgium	rural background	01/2000 – 12/2024
Scy-Chazelles	6.12278 E	49.10750 N	France	peri-urban	10/2000 – 12/2024
Thionville-Garche	6.20139 E	49.39444 N	France	peri-urban	02/2001 – 12/2024
Vianden	6.16884 E	49.94743 N	Luxembourg	rural (water reservoir)	01/2000 – 12/2024
Vielsalm	6.00043 E	50.30378 N	Belgium	rural (forest)	01/2000 – 12/2024
Westeifel-Wascheid	6.37807 E	50.26656 N	Germany	forest (clearing)	01/2000 – 09/2024

Meteorological data (air temperature at 2 m above ground (°C) and boundary layer height (m)) were extracted from the latest ERA5 climate reanalysis produced by the European Centre for Medium Range Weather Forecast (ECMWF) (Soci et al., 2024) that provides hourly data for each latitude and longitude and time period listed in Table 1.

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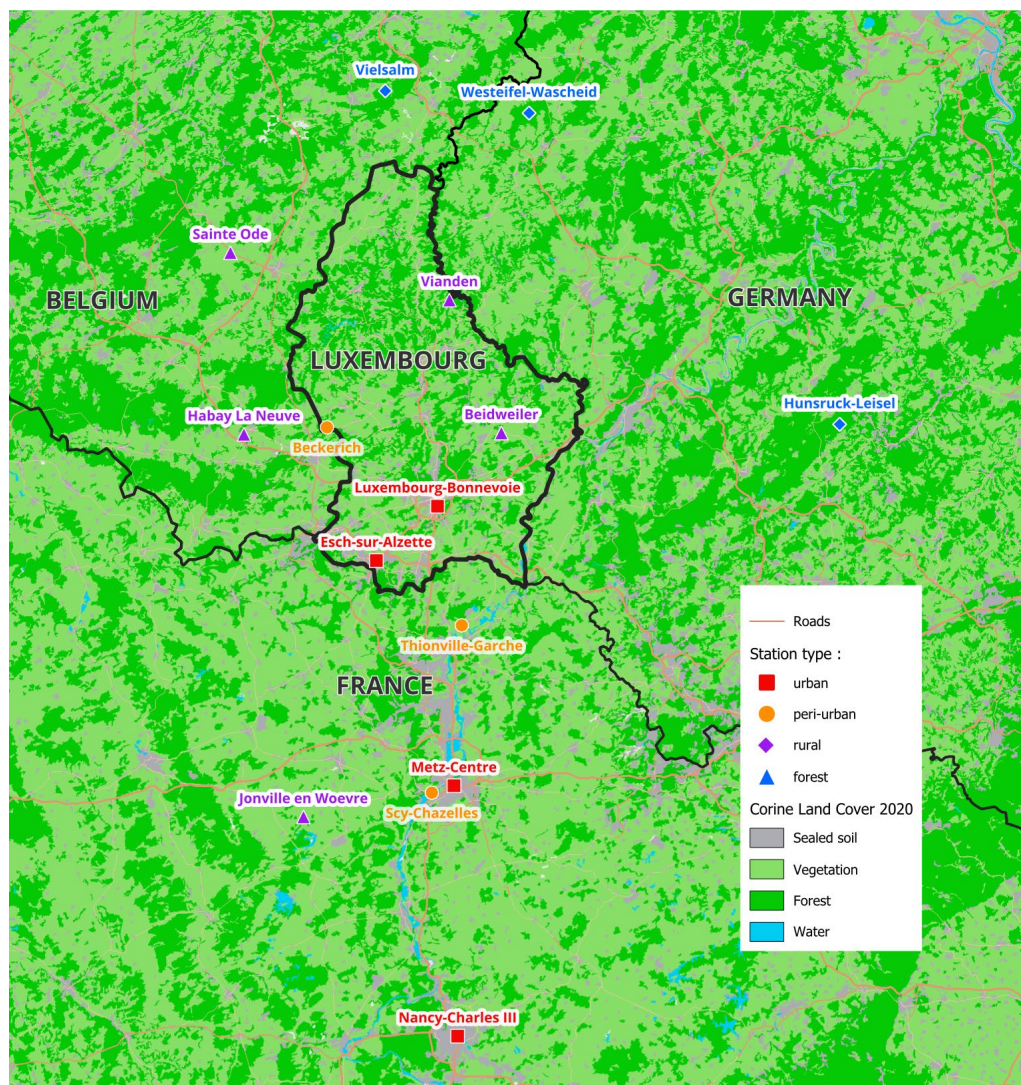


Figure 1: Map with the locations of the measurement sites in the Greater Region of Western Europe. Data sources: CORINE Land Cover 2018 (CLC2018, V2020_20u1), Copernicus Land Monitoring Service; administrative boundaries derived from GADM 4.1; roads © OpenStreetMap contributors, Microsoft, Esri Community Maps contributors, Map layer by Esri. Map design and additional content by the authors. GADM data are used in accordance with the GADM licence for academic research.

2.2 Timeseries decomposition and trend analysis

To infer robust long-term trend slopes and analyse the seasonal variability of the NO_2 and O_3 monthly averages we used three different approaches: (a) Bayesian model averaging (BMA) (Zhao et al., 2019), (b) quantile regression (QR) (Chang et al., 2023) and (c) standard non-parametric methods such as Mann-Kendall analysis (Kendall, 1975; Mann, 1945) and Theil-Sen's slope (Sen, 1968; Theil, 1950) to estimate linear trends. These analyses were performed for the monthly averages and



monthly averages of diurnal minima and maxima of NO₂, O₃, air temperature and boundary layer height (BLH) timeseries of each location and subsequently trend results were clustered according to the four station categories (urban, peri-urban, rural and forest).

2.2.1 BEAST algorithm

145 We applied the Bayesian Estimator of Abrupt change, Seasonal change, and Trend (BEAST) as an ensemble algorithm that fits a Bayesian regression model to match time-series data with time as the independent variable (Zhao et al., 2019). It is a generic Bayesian time-series decomposition algorithm for three contrasting components: abrupt change (changepoints), periodic/seasonal change, and trend implemented via a stochastic sampling algorithm known as Markov Chain Monte Carlo. BEAST enables the representation of nonlinear dynamics inherent to environmental and atmospheric pollutant time series, driven by chemistry, emissions, and meteorology (Wu et al., 2023). Time-series are statistically decomposed into a long-term trend signal and a seasonal signal that is approximated as a piecewise harmonic model (Zhao et al., 2019). BEAST detects the trend changepoints and seasonal changepoints separately and quantifies their posterior probabilities from the Bayesian model (values range from 0 → 1, 0 = almost no evidence of a change, 1 = very strong evidence of a seasonal or trend change at that time). Changepoint selection was performed using an adaptive scheme directly applied to the BEAST outputs. For each time series and component (seasonal or trend), the posterior distribution of the number of changepoints was first used to determine the maximum number of changepoints retained. For the seasonal component, the posterior median number of changepoints and for the trend component, the 10th percentile of the posterior distribution was used, suppressing insignificant slope perturbations. Individual changepoint candidates were then filtered using a relative posterior probability criterion. This approach avoids the use of generic probability thresholds and ensures that changepoint selection adapts automatically to the posterior probabilities of each individual time series. BEAST analyses were performed for the entire data record available for each station (Table 1).

2.2.2 Quantile regression

As recommended in the Tropospheric Ozone Assessment Report, Phase II (TOAR-II) we applied quantile regression (QR) to estimate long-term trends (Van Malderen et al., 2025; Chang et al., 2023), primarily because it enables detecting linear changes within distinct portions of the O₃ distribution, (e.g., 5th percentile, characterizing low O₃ conditions, 50th percentile, or 95th percentile). Here, the method is applied to the median (50th percentile) change in the trends, which is equivalent to the least absolute deviation estimator that is particularly robust when extreme values or outliers are present (Van Malderen et al., 2025; Nelson and Drysdale, 2025). Four harmonic terms (sine-cosine combination) are used to capture the seasonality and subsequently the timeseries were deseasonalised. QR is combined with a moving block bootstrap algorithm taking autocorrelation into account in the trend uncertainty (Chang et al., 2023).



2.2.3 Mann-Kendall analysis and Theil-Sen trend estimator

A combination of non-parametric Mann-Kendall analysis (Kendall, 1975; Mann, 1945) and Theil-Sen trend estimator (Sen, 1968; Theil, 1950), which do not assume any statistical distribution of the data, were used for testing the presence and significance of linear trends in the timeseries. To estimate the long-term trend, the seasonal signal was removed by (a) singular spectrum analysis (SSA) (Golyandina and Zhigljavsky, 2013), which assumes an additive decomposition of the data and (b) trend decomposition using Loess (STL), which is an additive decomposition based on a locally weighted regression (Cleveland and Cleveland, 1990). After decomposition, the non-parametric Mann-Kendall test and the Theil-Sen method were applied to the sum of the long-term trend and the residual signal of the NO₂ and O₃ timeseries to detect monotonic trends over several years. While the Kendall's tau statistic is an indicator of whether a trend exists, the Sen's slope delivers the strength of the trend.

3 Results and Discussion

3.1 Time series overview and extreme events

Table A1 (Appendix A) presents the descriptive statistics (including extreme events) for the timeseries of the individual stations. The annual distributions of NO₂ concentrations in Figure 2a shows an increase between 2000 and 2003, most pronounced at urban, peri-urban and forest stations, while rural sites exhibit only minor changes during the same period. Following this initial increase, NO₂ concentrations remained relatively stable for approximately a decade, after which a progressive decline is evident until 2024 for all station categories. As expected, concentrations were highest in urban areas, which also exhibited the greatest variability in the boxplots, whereas forest sites showed the lowest concentrations with the smallest variability. At urban sites median values decline from approximately 30 to 35 µg m⁻³ in the early 2000s to near 10 to 15 µg m⁻³ after 2020. Mean rural and forest long-term averages were three times lower than at urban locations. Figure 2b indicates highest O₃ medians at forest stations in almost every year, followed by rural, peri-urban, and urban sites. During approximately 2000 to 2016, O₃ levels remain relatively stable for each category with a short upward trend in the early phase 2000-2003 at forest and rural stations. Lowest values were observed at urban locations around 2007–2009, while highest values were measured at forest sites in 2003, 2006, 2018 and 2024. The first three peak years coincide with previously reported O₃ episodes over Europe co-occurring with heatwaves (Crespo-Miguel et al., 2024; Zohdirad et al., 2022). The episode in 2003 occurred in the first half of August driven by the mega-heatwave that affected western Europe. In that month the maximal hourly O₃ concentrations substantially exceeded 200 µg m⁻³ at several stations (Table A1). From approximately 2014–2016 onward, median values cluster around or above 60 µg m⁻³ at rural and forest stations. Between 2016 and 2018 an increase was observed at all station types. After 2018, O₃ medians remain consistently elevated at rural and forest stations. Urban sites also show a gradual increase in the last years but mean urban and peri-urban long-term averages were about 30 % lower than those of forest and rural locations.

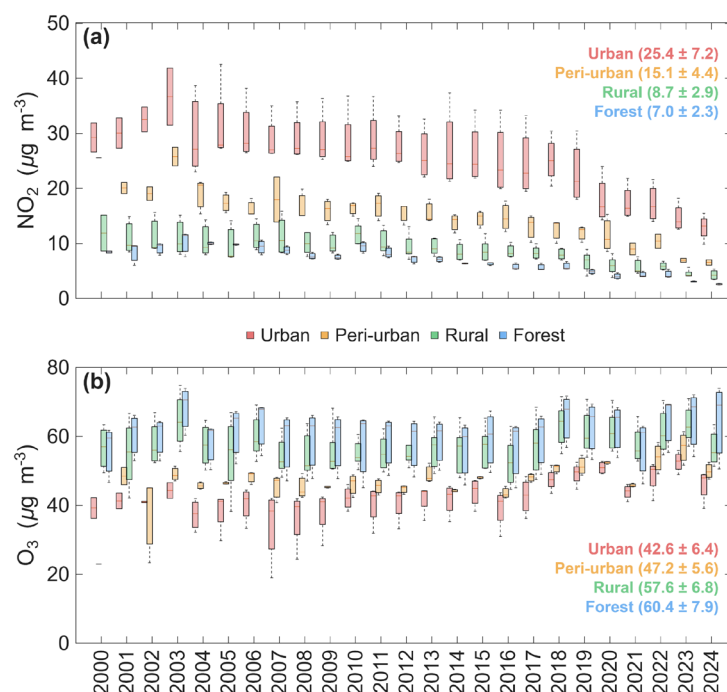


Figure 2: Box-and-whisker plots presenting the annual statistical distribution of (a) NO₂ and (b) O₃ for 15 stations (combined per station category, see Table 1) in the Greater Region from 2000 to 2024. Means and standard deviations for each station category are printed inside the figures. Number of data points (active stations) was nearly equal only from 2006 onwards.

3.2 Analysis of long-term NO₂ and O₃ trends

3.2.1 Results from the BEAST changepoint detection for NO₂ and O₃

Figure 3a-d summarizes BEAST posterior changepoint probabilities for NO₂ and O₃, separated into seasonal and trend components, classified by station category. Seasonal shifts in NO₂ (Figure 3a) occurred predominantly during 2014 and 2017, with the highest number of posterior probabilities detected in autumn 2017. These changepoints occur across all station categories for monthly means and are more dominant for the monthly means of diurnal minima than for the maxima indicating an overall change of the seasonal cycle. In contrast, O₃ exhibits no evidence of changes in the seasonal cycle as only three isolated changepoints with low probability were found by the BEAST algorithm (Figure 3b). This indicates that the seasonal amplitude and phase of O₃ remained comparatively stable over the period 2000-2024.

Trend changepoints for NO₂ (Figure 3c) occur during different periods, an earlier phase around 2006–2012 and a later, more coherent phase around 2019–2020. The later period shows higher posterior probabilities but is only evident for rural and urban stations. O₃ trend changepoints (Figure 3d) show a stronger clustering around 2017/2018 for all station categories and value types (mean, max, min), with moderate to high posterior probabilities.

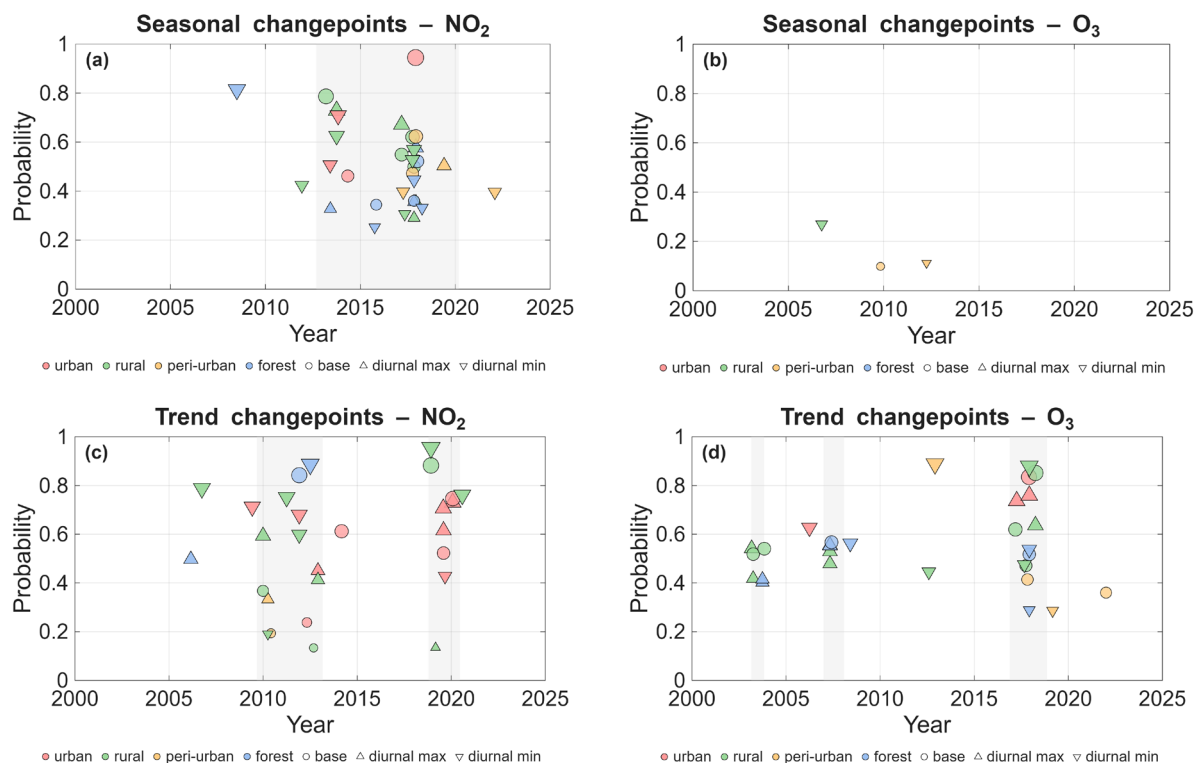


Figure 3. Probability of seasonal (a and b) and trend (c and d) changepoints derived with BEAST from monthly NO_2 and O_3 timeseries between 2000 and 2024 for all 15 stations, clustered by station category. Marker size is scaled by the posterior probability of the changepoint. It quantifies the confidence of the BEAST algorithm in the detected changepoint, with higher values indicating stronger evidence. Circles denote monthly means (base) and triangles monthly means of diurnal maxima and minima (diurnal max, diurnal min). Grey bands indicate periods when changepoints concentrate. Changepoints from 2000-2003 and 2022-2024 were excluded as they are not reliable due to missing data before and after these periods.

Additional earlier signals before 2010 are present but less dominant. The exact trend changepoint occurred in autumn 2017 and coincides temporally with the principal seasonal NO_2 changepoints (Figure 3a), roughly one year before the heatwave and associated O_3 episode days in summer 2018 (Nelson and Drysdale, 2025; Pope et al., 2023). The onset of the change in the NO_2 intra-annual variability and O_3 trend increase in autumn 2017 for most stations coincides with changes in the vehicle fleet composition, particularly the rapid increase in electric vehicle registrations and the reduction in diesel passenger cars, e.g. in Luxembourg (see Figure A1). From the timeseries of monthly diesel vehicle registrations, BEAST detects seasonal changepoints in 2020 and 2021 with posterior probabilities > 0.8 , and trend changepoints in 2017 and 2021 with posterior probabilities > 0.45 . However, NO_x reductions from Euro 6 diesel vehicles had already begun after 2015 with the introduction of AdBlue (selective catalytic reduction) systems. The principal changepoint in 2017 may also partially reflect



other altered emission patterns, including reduced winter combustion contributions and changes in gas consumption associated with warmer winters (Figure A5). Notably, the identified changepoints precede the COVID-19 pandemic and, hence, cannot be attributed to emission reductions related to lockdowns. However, the decline of national NO_x emissions between 2005 and 2023 by 82 % was only partially reflected in the NO_x decline at Luxembourgish monitoring stations, which was ~60 % on average over the same period (Figure A2).

3.2.2 Results from the BEAST trend decomposition for NO₂ and O₃

Data quality and completeness of NO₂ measurements improved after 2009 and most valid BEAST changepoints for NO₂ occurred after 2010 (see Figure 3). Hence, we focussed our trend decomposition analysis on the period 2009-2024. BEAST results show that all station categories exhibit a stable seasonal cycle, with winter maxima and summer minima, consistent with enhanced winter NO_x emissions and reduced photochemistry (Figure 4a). Urban and peri-urban stations show the largest seasonal amplitudes, while rural and especially forest sites reveal smaller amplitudes, indicating the lower influence of local emissions. The amplitude of the seasonal NO₂ component is decreasing over time with a marked break in 2017/2018 (Figure 4a), confirming major seasonal changepoints before the COVID-19 pandemic. During and after this period, the interannual amplitude starts to decline. Peri-urban stations show the most pronounced amplitude reduction after 2017. In contrast, the O₃ seasonal component (Figure 4b) shows a highly regular annual cycle for all station categories, without major changepoints (Figure 3b). Peak O₃ concentrations occur later at peri-urban and urban locations (June/July) compared to rural and forest sites (May).

The extracted long-term component of NO₂ (Figure 4c) shows a persistent downward trend for all station categories. The decline is strongest in absolute terms at urban sites, moderate at peri-urban locations, and smallest at rural and forest sites. Concentrations decrease by at least 50 % for all station categories from 2009 to 2024, with slightly larger relative reductions at rural and forest sites. The previously presented trend changepoints (Figure 3c) are visible in 2014 and 2019/2020, especially for urban sites. The drop in 2020 is likely related to the COVID-19 lockdown. NO₂ concentrations decline on average by ~ 15.4 µg m⁻³ (−0.96 µg m⁻³ yr⁻¹) at urban stations, by ~ 9.3 µg m⁻³ (−0.58 µg m⁻³ yr⁻¹) at peri-urban, by ~ 5.6 µg m⁻³ (−0.35 mg m⁻³ yr⁻¹) at rural and by ~ 5.2 µg m⁻³ (−0.33 mg m⁻³ yr⁻¹) at forest stations during the 16 years (2009-2024).

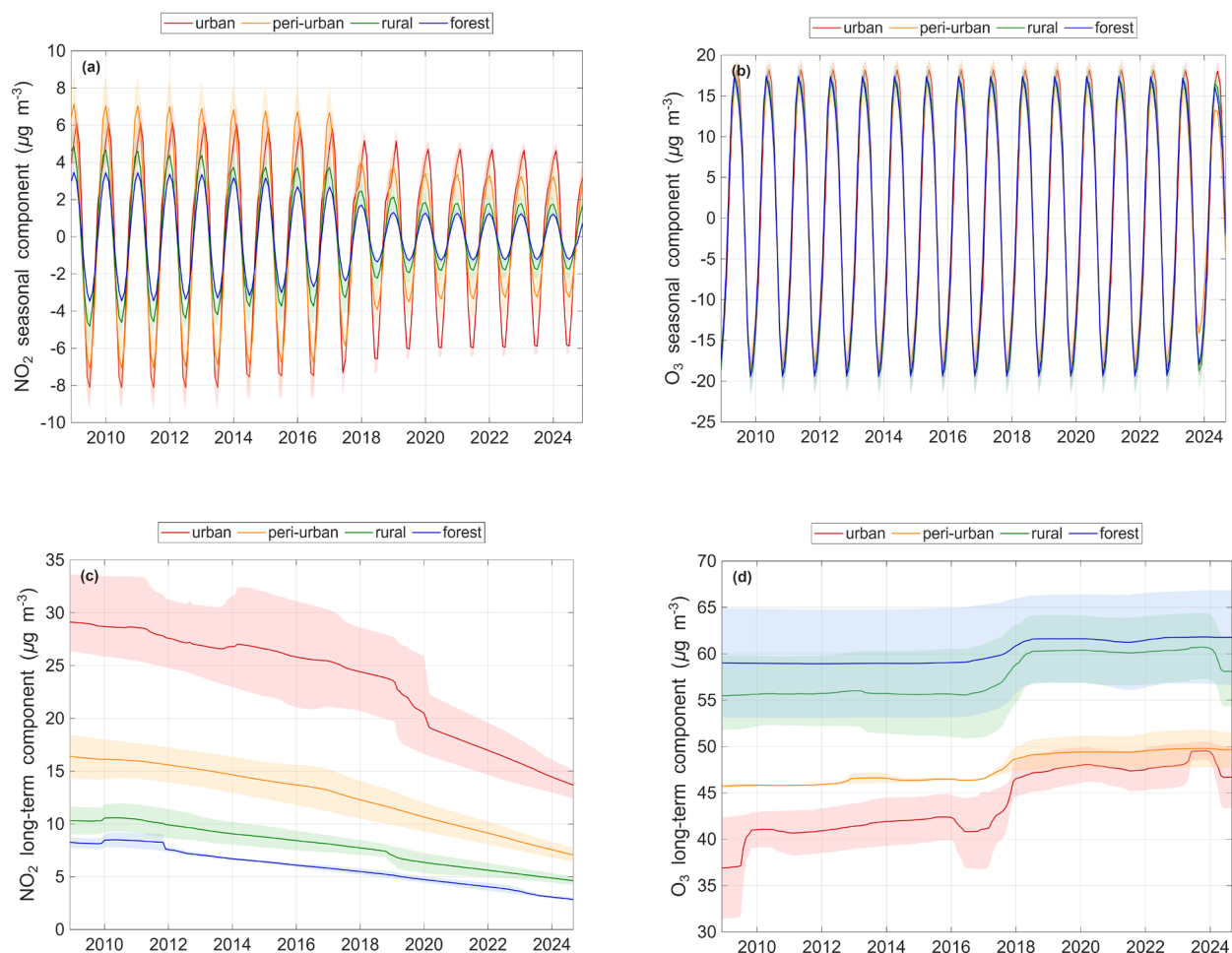


Figure 4: Extracted trend components of monthly mean NO₂ and O₃ concentrations from 2009 to 2024 with the BEAST algorithm averaged for the different station categories. The plots in (a) and (b) show the seasonal component and (c) and (d) show the long-term components. Shaded areas indicate uncertainty ranges after bootstrapping the data (n = 1000, lower bound = 5th percentile, upper bound = 95th percentile).

From 2009 to 2016, O₃ long-term trend components extracted with BEAST remained stable, especially at rural and forest locations, while small increases were evident at peri-urban and urban stations (Figure 4d). The changepoint that indicates the abrupt O₃ increase (Figure 3d) starting at the end of 2017 is clearly visible in Figure 4d. Between 2009 and 2024 O₃ increased on average by $\sim 9.8 \mu\text{g m}^{-3}$ ($0.61 \mu\text{g m}^{-3} \text{ yr}^{-1}$) at urban stations, by $\sim 4.2 \mu\text{g m}^{-3}$ ($0.26 \mu\text{g m}^{-3} \text{ yr}^{-1}$) at peri-urban, by $\sim 5.0 \mu\text{g m}^{-3}$ ($0.31 \mu\text{g m}^{-3} \text{ yr}^{-1}$) at rural and by $\sim 2.8 \mu\text{g m}^{-3}$ ($0.18 \mu\text{g m}^{-3} \text{ yr}^{-1}$) at forest stations. In relative terms, O₃ concentrations increased by $\sim 27 \%$ at urban and by $\sim 10 \%$ at peri-urban, rural and forest stations from 2009 to 2024. The results presented in Figure 4 indicate that the changepoints in 2017 are more prominent than the effect of the short-term emission reductions in 2020/2021 and the post-Covid rebound in urban areas (Varga et al., 2024; Grange et al., 2021).

3.2.3 Changepoints in air temperature and boundary layer height

The long-term component of daily maximum air temperature shows a gradual increase for all station categories from 2009 to 2024, reflecting a coherent regional warming signal (Figure 5a). The increase is largest at urban stations (approximately 0.9°C), followed by peri-urban stations (0.8°C), while rural and forest stations exhibit slightly smaller increases of about 0.7°C. Potential changepoints are visible just before 2014 and in 2018. A similar long-term evolution is evident for the diurnal maximum BLH, although with a different temporal pattern. The maximum BLH remains relatively constant until approximately 2016, followed by a marked increase between 2017 and 2018 that persists for several years before declining slightly after 2022 (Figure 5b). Urban stations consistently exhibit the deepest daytime boundary layers, whereas forest stations show the lowest BLH values. In contrast to the increase in air temperature, the more abrupt, step-like increase in BLH occurs simultaneously to the seasonal and long-term changepoints detected for NO₂ and O₃, respectively (Figures 3 and 4). The BLH development is relevant because it integrates several meteorological controls including surface sensible heat flux, atmospheric stability, cloudiness, soil moisture, and synoptic circulation. Consequently, it is an indirect proxy for the efficiency of photochemistry, vertical mixing and O₃ entrainment from the residual layer (Song et al., 2024). Higher daytime maximum BLHs indicate stronger convective boundary layer development, which favours the entrainment of O₃-rich air from the residual layer into the mixed layer during the morning, contributing to elevated surface O₃ (Kaser et al., 2017).

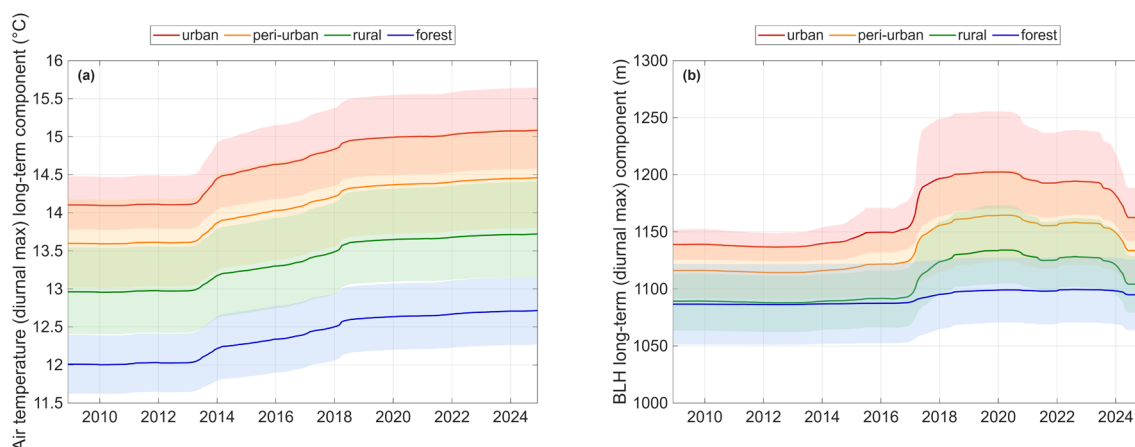


Figure 5: Long-term components of monthly mean diurnal maxima in (a) air temperature (2 m) and (b) boundary layer height (BLH) extracted from ERA5 climate reanalysis data derived with the BEAST algorithm and averaged for different station categories. Shaded areas indicate uncertainty ranges after bootstrapping the data (n = 1000, lower bound = 5th percentile, upper bound = 95th percentile).

Most stations exhibit their highest-probability maximal air temperature changepoints during 2014 and 2018 (Figure 6a), with the latter reflecting most probable changes in the monthly 95th percentile of air temperature (extreme values). This changepoint in extremes occurs just after the BLH long-term changepoint in 2017, which have higher maximum



probabilities than air temperature (reaching values close to 0.7 at some urban stations) and highest probabilities occurred in 2017 for nearly all stations (Figure 6b).

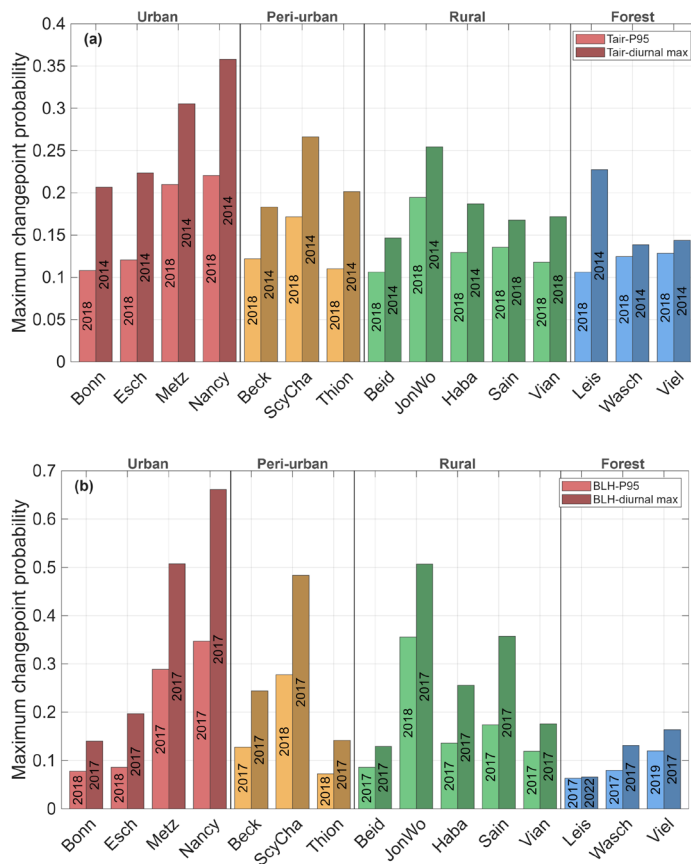


Figure 6. Maximum posterior changepoint probabilities derived from the BEAST method from (a) monthly mean diurnal maxima of air temperature (Tair-diurnal max; dark bars) and the monthly 95th percentile air temperature (Tair-P95; light bars) and (b) monthly mean diurnal maxima boundary layer height (BLH-diurnal max; dark bars) and the monthly 95th percentile BLH (BLH-P95; light bars) at the 15 monitoring sites in the Greater Region, grouped by station category. The labels within the bars indicate the year associated with the most probable changepoint. The posterior probability quantifies the confidence of the BEAST algorithm in the detected changepoint, with higher values indicating stronger evidence.

BLH changepoint probabilities are mostly higher for diurnal maxima compared to P95, suggesting that the long-term change is primarily affected by typical daytime meteorological conditions rather than only the most extreme events. Urban stations generally exhibit the highest changepoint probabilities, with particularly strong evidence at Metz and Nancy, whereas forest stations show lowest probabilities. Changepoints in monthly daytime maximum BLH and the monthly 95th percentile of air



temperature (2017/2018) largely coincide with the detected changepoints in the long-term O₃ trend and the seasonal NO₂ trends (Figures 3 and 4). No significant changepoints in long-term and seasonal trends were detected in minimal air temperatures and BLH values. Changepoint probabilities for means were lower than for diurnal maxima and 95th percentiles.

3.2.4 Trend slopes of NO₂ and O₃ using quantile regression and Theil-Sen method

Tables A2-A4 summarize the trend slopes and associated uncertainty metrics for monthly NO₂ means of all individual stations using the three different linear trend estimation methods described in sections 2.2.2 and 2.2.3 (QR-MBB, Theil-Sen after SSA and STL decomposition). The slopes determined with the QR-MBB method had a very high certainty as indicated by signal-to-noise-ratios above three and $p < 0.001$ (Table A2, Cheng et al., 2023). Also, the Theil-Sen trend slopes were statistically highly significant ($p < 0.001$). Mann-Kendall (MK) rank correlation coefficient (τ) for the individual stations was between -0.65 and -0.4 with highest values for the STL decomposition method (Table A4). We find consistent negative trends for all station categories confirming the substantial decline in NO₂ concentrations from 2009 to 2024 (Figure 7a). Urban stations show decreases in monthly mean concentrations of approximately $-1.1 \mu\text{g m}^{-3} \text{ yr}^{-1}$, consistent with emission reductions in traffic-dominated areas, which is comparable to declines observed in Switzerland (Nussbaumer et al., 2026). In contrast, the absolute decline at forest stations is substantially lower ($-0.35 \mu\text{g m}^{-3} \text{ yr}^{-1}$). All slopes compare relatively well with average BEAST estimates (see Figure 4c). The slopes estimated with the QR-MBB method are slightly lower than those derived with the Theil-Sen method and show the best agreement with the average BEAST slopes. The strongest NO₂ decline occurred in diurnal maxima (Figure 7a middle panel), especially at urban ($-1.8 \mu\text{g m}^{-3} \text{ yr}^{-1}$) and peri-urban ($-1.25 \mu\text{g m}^{-3} \text{ yr}^{-1}$) stations. Hence, peak NO₂ concentrations mainly occurring in the early morning or evening during rush hour periods have declined more strongly than mean or minimum values. Monthly means of diurnal minima show weakest long-term trends, particularly at background (forest and rural) stations.

The trend slopes of O₃ concentrations in Figure 7b are consistently positive for all station categories, confirming the increase in O₃ over the study period. Tables A5-A7 summarize the trend slopes and associated uncertainty metrics for monthly O₃ means of all individual stations. The slopes determined with the QR-MBB method had a high to very high certainty as indicated by signal-to-noise-ratios above two and $p \leq 0.05$ (Cheng et al., 2023) while no evidence for a significant positive slope was found only for the rural station Beidweiler in Luxembourg (Table A5). All Theil-Sen trend slopes were statistically significant ($p < 0.05$). MK- τ for the individual stations was between 0.11 and 0.37 with highest values for the STL decomposition method (Table A7). Slope magnitudes were highest for urban stations ($\sim 0.75 \mu\text{g m}^{-3} \text{ yr}^{-1}$), while increases at peri-urban, rural, and forest sites were smaller but nearly within the same range for the three categories ($\sim 0.4\text{--}0.6 \mu\text{g m}^{-3} \text{ yr}^{-1}$) (Figure 7b). The increases were similar to values found in Saxony (Germany) for urban and rural sites covering a comparable period (Wang et al., 2025), while O₃ trend slopes determined for Switzerland were lower or even decreased at rural background sites (Nussbaumer et al., 2026). The trend in the surface to 300 hPa tropospheric O₃ column over Uccle, a peri-urban site near Brussels, was +1.5 ppb per decade (equivalent to $0.3 \mu\text{g m}^{-3} \text{ yr}^{-1}$) during the period 2000 to 2022 (Van Malderen et al., 2025).

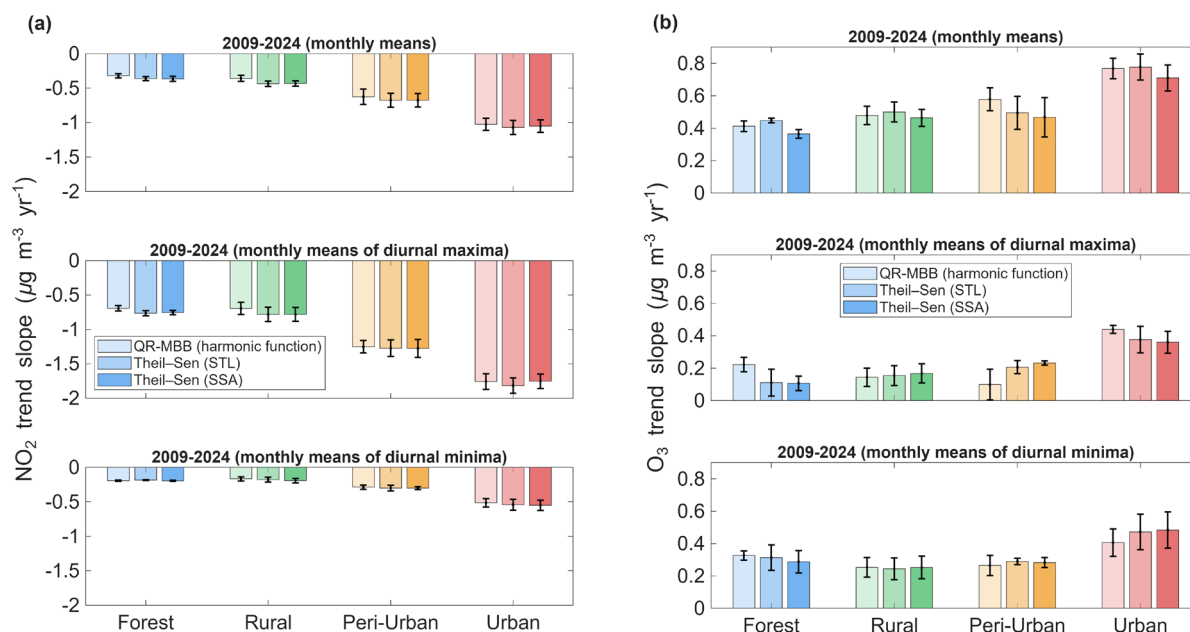


Figure 7: Long-term trend slopes for (a) NO₂ and (b) O₃ computed from monthly means, monthly means of diurnal maxima and minima using quantile regression and a moving-block-bootstrap algorithm (QR-MBB, $\tau = 0.5$) after removing seasonality calculated from harmonic functions and Theil-Sen estimator method after removing seasonality based on the STL and SSA methods, see section 2.2) averaged for each station category. Error bars denote standard errors of category means. Individual monthly mean slopes and associated uncertainty ranges for NO₂ measured at all stations are listed in Table A2 (QR-MBB), Table A3 (Theil-Sen-SSA) and Table A4 (Theil-Sen-STL) and for O₃ in Table A5 (QR-MBB), Table A6 (Theil-Sen-SSA) and Table A7 (Theil-Sen-STL).

The three methods yield comparable slope magnitudes, with higher variations for peri-urban stations. Except for urban stations, the derived average BEAST slopes for O₃ are lower than those of the QR-MBB and Theil-Sen methods, especially for peri-urban and forest sites (see Figure 4d). This can be partly explained by the drop in O₃ during 2024 that is accounted for by BEAST but only partly by the linear trend estimates. For diurnal maxima (middle panel), positive trends are also observed for all station categories, although magnitudes are generally lower than those for monthly means. For diurnal minima (bottom panel), estimated trends are also positive and exceed those of the diurnal maxima for peri-urban, rural and forest stations.

3.3 Seasonal, weekly and diurnal changes in NO₂ and O₃

3.3.1 Seasonal and diurnal trends

To investigate the temporal evolution of the change intensity we analysed five-year moving window differences (ΔNO_2 and ΔO_3) for each season (Figures 8 & 9). The negative five-year moving window differences (ΔNO_2) in Figure 8 indicate sustained NO₂ reductions relative to the previous 5-year periods during all seasons at urban stations. Before 2018, only minor differences between urban, peri-urban, rural and forest stations were detected and the concentrations were relatively constant or even increased. During winter, a slight decrease already started in 2013/2014 (Figure 8d), which is confirmed by the changepoints detected with BEAST (Figure 3c). The pre-COVID changepoint in 2017 is consistently observed in all seasons, indicating a structural shift in the long-term trend and the onset of a sustained decline. In accordance with the effect of NO_x emission reductions, the magnitude of this decline decreases from urban to peri-urban, rural, and forest stations. This strong decrease at urban sites is weakest during summer and autumn (Figure 8b,c), most likely due to lower domestic heating activities. During spring and summer (Figure 8a,b), ΔNO_2 values were close to zero and constant over time for all non-urban sites, revealing relatively stable background NO₂ concentrations.

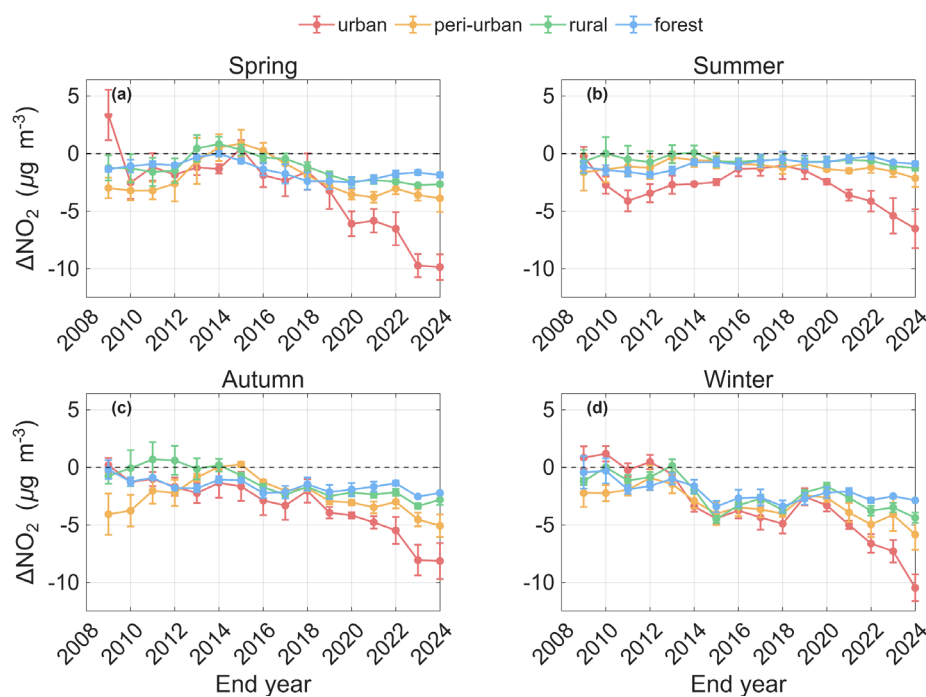


Figure 8: Changes in NO₂ concentrations derived from a moving window of five-year differences, shown separately for different meteorological seasons (a) spring, (b) summer) (c) autumn and (d) winter and each station category. For each end year Y, ΔNO_2 is defined as the difference between the mean NO₂ concentration over years Y–4 to Y and the preceding 5-year period Y–9 to Y–5. Error bars denote the standard error of the mean for all stations within each category.



The five-year differences (ΔO_3) in Figure 9 reveal that spring and autumn are the dominant seasons contributing to the O_3 increase after 2017/2018, with highest contributions for urban sites during spring. In 2016, a minimum appears consistently in spring (Figure 9a), most pronounced at rural and forest sites, indicating a decline until that year, while after 2017/2018, ΔO_3 becomes persistently positive. However, in spring the positive ΔO_3 values are levelling off since 2022 and start to decrease again. During summer, ΔO_3 started to increase already around 2012 at urban sites, remained constant between 2014 and 2022 and afterwards started to decrease for all station categories (Figure 9b). A continuous increase in ΔO_3 is observed during autumn for all station categories which is steepest since 2018 (Figure 9c). The pattern is comparable during winter, when ΔO_3 is positive since 2011 for all, except forest stations. However, the increase since 2018 is less pronounced than during autumn and values increase already between 2011 to 2014, especially at urban and peri-urban sites (Figure 9d). Comparable trends were found at other locations in Europe and showed upward values for winter, spring and autumn with stable or downward values for summer (Adame et al., 2022).

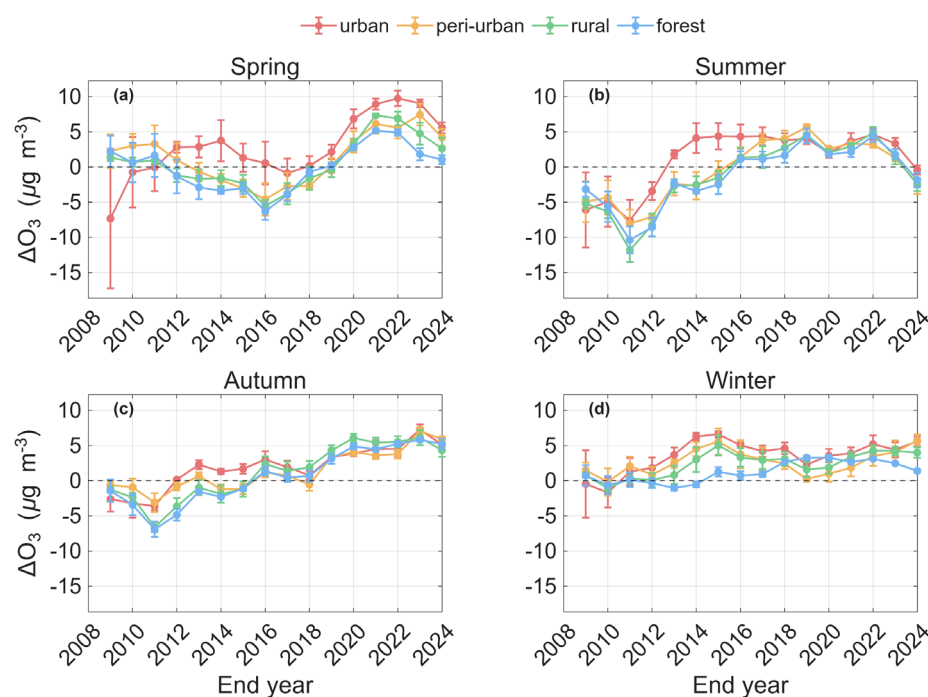
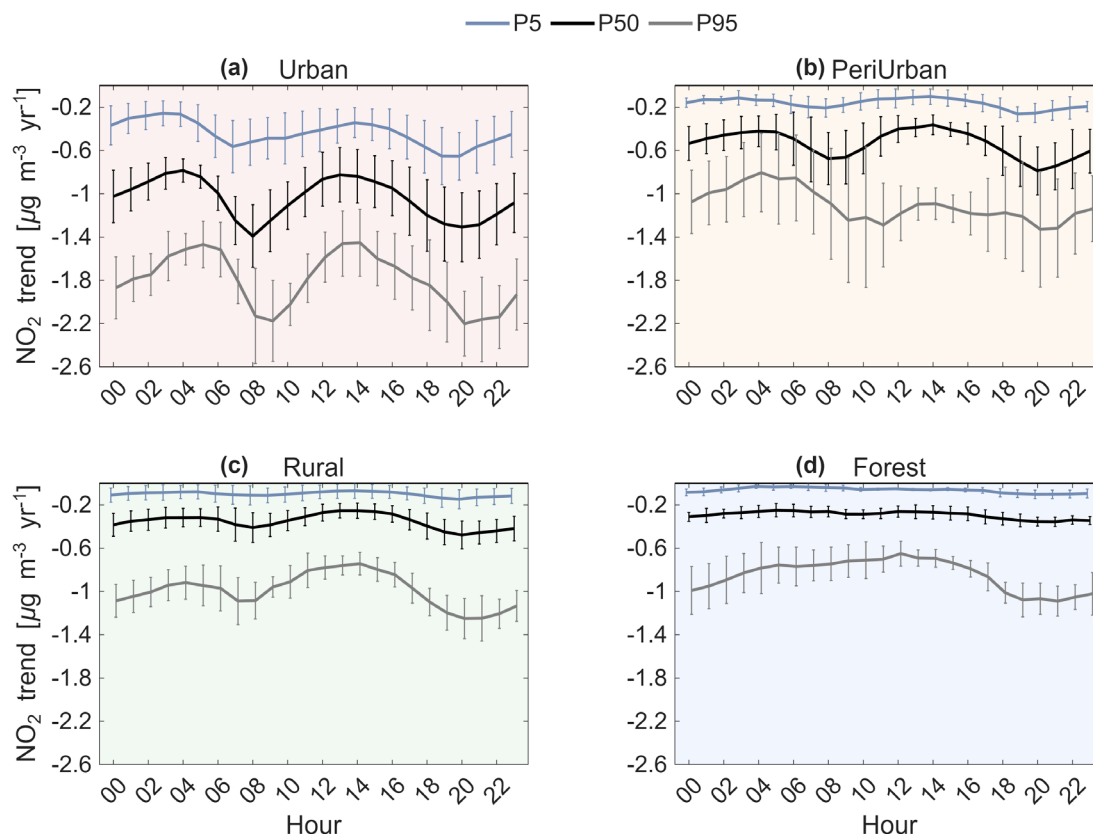


Figure 9: Same as Figure 8, but for O_3 .

NO_2 reveals declining diurnal trends throughout the whole value distribution for the different station categories between 2009 and 2024 (Figure 10, Figure A3). In line with the direct effect of emission reduction measures, highest NO_2 concentrations have decreased more than background concentrations (see also Figure 7a). The negative P95 trend at peri-urban, rural, and forest stations is comparable and approximately half the magnitude of that observed at urban locations (Figure 10a-d). The latter show the strongest diurnal variability, with largest reductions occurring around typical commuting hours 07:00–09:00 and 19:00–21:00 that is additionally affected by the decrease of photolysis rates and BLH in the evening.



395 The comparable trend pattern (P50 and P95) at peri-urban stations indicates that they remain influenced by nearby anthropogenic emissions while also reflecting regional background conditions. Rural and forest stations trends display much weaker diurnal variability, lacking distinct morning peaks related to reduced traffic emissions. The strongest negative trends in these stations occur during the evening and nighttime (approximately 18:00–00:00), whereas the smallest decreases are observed around midday (Figure 10c,d). This pattern is consistent with declining regional NO_x together with stable nocturnal
 400 boundary layer conditions, which suppress vertical mixing and increase the sensitivity of near-surface NO_2 concentrations to changes in regional emissions (Kuhn et al., 2024). P5 varies only modestly throughout the day for all station categories, with slightly stronger declines during the early morning and evening at urban locations. Comparing kernel density estimates of the NO_2 concentration distributions (Figure A3) before and after the detected changepoints in 2017/2018 indicates that the NO_2 distribution has become substantially narrower, with overlap coefficients between 0.71 and 0.86.



405 **Figure 10.** Mean diurnal NO_2 trends derived from 15 monitoring sites in the Greater Region grouped by station category (a-d) for the period 2009–2024. Trends were estimated using the Sen's slope method applied to deseasonalised (STL decomposition) monthly time series of hourly NO_2 percentiles. Curves show the average hourly trends for the 5th (P5), 50th (P50), and 95th (P95) percentiles. Error bars indicate the standard deviation of the trend
 410 estimates (cf. Yan et al., 2018).



The P5 exhibits the largest positive O₃ trends, P50 show intermediate positive trends, while the P95 displays the smallest trends and approaches zero or becomes slightly negative after midday, particularly at rural and forest stations (Figure 11). A pronounced daytime trend divergence between the O₃ percentiles is evident at urban and peri-urban stations, with only P5 increasing substantially from the morning into the afternoon (Figure 11a,b). Hence, the largest daytime increases occur under conditions that historically exhibited low O₃ concentrations because of the reduced titration with NO (e.g., during winter, see Figure A4) (Real et al., 2024; Yan et al., 2019).

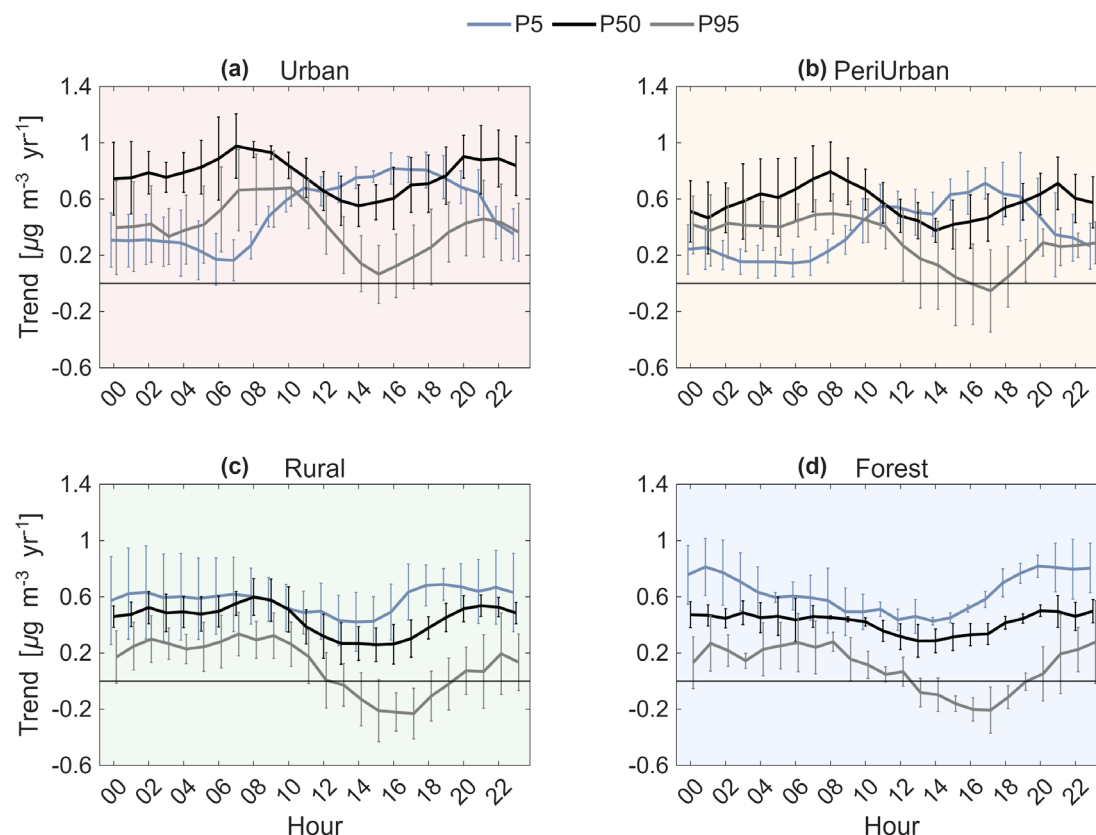


Figure 11. Same as Figure 8, but for O₃.

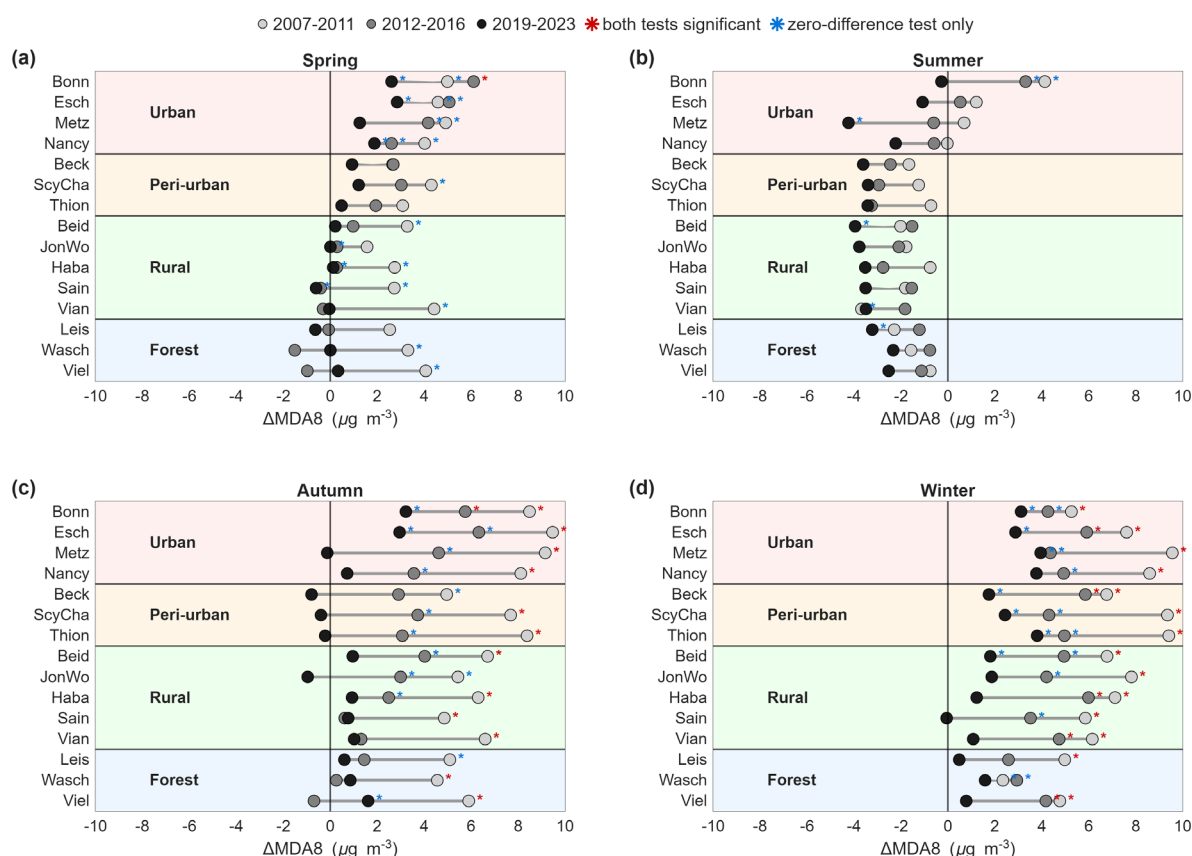
In contrast, rural and forest stations exhibit similar pattern of the diurnal cycle for all trend percentiles (Figure 11c,d). Peak O₃ concentrations remained stable or have slightly decreased only during summer (see Figure A4). Because at our forest clearing sites, biogenic NO emissions from soil are expected to be small compared to traffic emissions, and an increase in mean (or P50) levels can be largely explained by hemispheric transport and reduced titration by NO as a result of overall reduced NO_x levels in the atmosphere (Solberg et al., 2022). Except in the P5 for urban and peri-urban stations, higher increasing trends were found during nighttime, which agrees with previous studies covering different time periods that also attributed the positive surface O₃ trend mostly to the colder months (see Figure A4) and to the nocturnal period (An et al., 2024; Adame et al., 2022). In Western Europe, the most notable change between 2000–2004 and 2014–2018 was a distinct



flattening of the summer O_3 peak alongside an increase in wintertime O_3 levels (Ansari et al., 2025). Other previous studies also identified for both urban and rural background sites a narrowing of the O_3 distribution, hence, a decrease in the 95th percentile (or maximal values during the warm season) and an increase in the 5th percentile of O_3 for the periods 2000–2015, 2000–2021, 1995–2014 and 2000–2024, respectively in Europe (Boleti et al., 2020; Nelson and Drysdale, 2025; Yan et al., 2018; Nussbaumer et al., 2026; Yan et al., 2019). This contrasts with our study, where we show that in the Greater Region, trends for all station categories and throughout the whole O_3 distribution are upwards or near zero from 2009 until 2024, which is similar to results recently obtained for Saxony (Germany) (Wang et al., 2025). Although reductions in regional NO_x levels are expected to reduce O_3 at the upper end of the O_3 distribution in VOC-limited (NO_x -saturated) regimes (Real et al., 2024), decreases in Europe during 2005–2019 were generally small and not statistically significant compared with the earlier period (2000–2017) (Solberg et al., 2022). This is consistent with the findings of the present study, although the effect on peak O_3 was very small in the Greater Region. Kernel density estimates of the mean 5-year O_3 concentration distributions before and after the 2017/2018 changepoints (Figure A4) indicate an overall shift of the O_3 distribution towards higher concentrations, particularly at rural and forest stations, but changes during summer remain comparatively small (overlap coefficients > 0.9). The reduction in peak O_3 concentrations due to chemistry was partly offset by the increase in daytime maximum BLH observed after the 2017/2018 changepoint (Figures 4 and 5), which enhanced vertical mixing and entrainment of O_3 -rich air from the residual layer. The pronounced differences in diurnal percentile trends between urban and rural environments reveal the relative importance of declining precursor emissions, evolving boundary layer dynamics and changes in regional photochemical O_3 production (Boleti et al., 2020; Boleti et al., 2018).

3.3.2 Weekend-weekday effect on MDA8

The concept of the weekend-weekday effect was originally developed by Levitt and Chock (1976) and implies that lower NO_x emissions on weekends, primarily due to reduced vehicle traffic in densely populated and polluted regions, often result in higher O_3 concentrations under VOC-limited conditions. We have computed the seasonal weekend-weekday MDA8 differences ($\Delta MDA8$) to elucidate the dominant chemistry regime (see Otero et al., 2021; Nussbaumer et al., 2026; Abeleira and Farmer, 2017). A positive weekend-weekday effect is consistent with a VOC-limited system, while a negative weekend-weekday effect is indicative of a NO_x -limited system. Mean MDA8 differences were derived for five-year periods (2007–2011, 2012–2016, and 2019–2023) and for different seasons (changepoint years 2017 and 2018 were excluded) (Figure 12). Statistical significance was assessed using the Mann-Whitney U test (Sicard et al., 2020; Otero et al., 2021).



455 **Figure 12. Seasonal weekend-weekday MDA8 differences (ΔMDA8 , weekend minus weekday) for all station categories for the periods 2007-2011, 2012-2016, and 2019-2023 (changepoint years 2017 and 2018 were excluded). Weekly weekend-weekday differences were first computed from daily MDA8 values and subsequently averaged seasonally for spring (a), summer (b), autumn (c), and winter (d). Circles indicate seasonal mean ΔMDA8 values for each period, while connecting lines illustrate the differences between five-year periods at individual stations.**
 460 **Statistical significance was assessed using the Mann-Whitney U test (Sicard et al., 2020) applied to weekly distributions of weekend-weekday differences for each season and for the difference of the ΔMDA8 distribution to zero. Red asterisks indicate significance at $p < 0.05$ for both tests, while blue asterisks indicate significance for the zero-difference test. MDA8 values were computed using the Openair R package (Carslaw and Ropkins, 2012).**

With few exceptions, the positive weekend effect during 2007–2011 progressively weakens and is near zero or becomes
 465 negative by 2019–2023 (Figure 12). During summer at higher temperatures ΔMDA8 values are lowest and predominantly negative (albeit not statistically significant at $p < 0.05$), indicating a transitional chemical regime with a weakening VOC-limited response and increasing influence of NO_x -limited conditions (Figure 12b). Under such conditions, O_3 concentrations



tend to decrease due to NO_x reductions (Otero et al., 2021). The magnitude of the statistically significant weekend effects during 2007-2011 and the decrease over the following periods is largest in autumn and winter for all station categories (Figure 12c,d), indicating that the change intensity towards a NO_x -limited regime is most pronounced during these two seasons. The strongest changes occur at urban and peri-urban stations, where the weekend effect is most sensitive to declining traffic-related NO_x emissions. Changes are smaller at rural and forest stations, with negligible differences between 2012–2016 and 2019–2023 in spring and autumn, indicating a weak $\Delta\text{MDA8 O}_3$ response. This was due to minor regional NO_x reductions at background sites, as indicated by relatively constant NO_2 (Figure 8 a,c) in these seasons. These stations are also more characterized by the increasing impact of hemispheric background O_3 (Solberg et al., 2022) and the temporal changes in regional background photochemical O_3 production (Boleti et al., 2018). Our results agree with Otero et al. (2021), who found that southwestern stations in Germany are transitioning to the NO_x -limited regime from 1999-2008 to 2009–2018, with a significantly decreasing weekend-weekday effect at higher temperatures. Although continued NO_x emission reductions are expected to shift O_3 production towards NO_x -limited conditions and thereby suppress peak photochemical O_3 formation, the often statistically insignificant seasonal ΔMDA8 values for 2019-2023 (Figure 12) suggest that reduced NO titration still plays a major role, especially at low to moderate temperatures. Combined with favourable meteorological conditions, particularly deeper daytime boundary layers (Figure 5b, Figure A5), this explains the recent increases in O_3 diurnal means, minima and maxima (Figures 7b, 8, 11, A3), even at rural and forest sites.

3.4 Trends in O_3 health metrics and ecosystem exposure

3.4.1 Trends in MDA8 ozone values and dependency on BLH and air temperature

The number of MDA8 exceedance days has decreased since 2018 (Figure 13), but considering the whole period since 2001 no clear trend is visible, while the years with extreme heatwaves stand out (e.g., 2003, 2006, 2018). Pronounced peak exceedances occurred in 2003 and 2018 corresponding to years with exceptionally favourable meteorological conditions for O_3 formation. Rural and forest stations consistently experienced the highest numbers of exceedance days, while the fewest were recorded at urban stations. The timeseries show the lowest ever recorded values in 2021 and 2024, with MDA8 exceedance days remaining below the current and future EU threshold for all station categories. The heatwave in 2018 was comparable to the one in 2003 in western Europe (Boboc et al., 2025; Liu et al., 2020), but the number of O_3 exceedance days was lower (Figure 13).

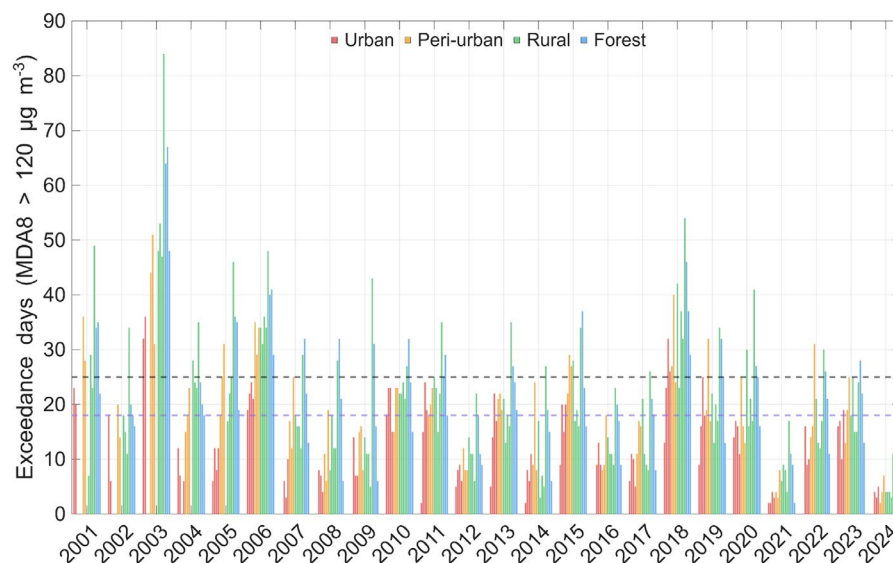


Figure 13. Average annual number of days with maximum daily 8-hour average (MDA8) O₃ concentrations exceeding 120 µg m⁻³ at urban, peri-urban, rural, and forest stations between 2001 and 2024. Horizontal dashed lines indicate the O₃ target values specified in the previous EU Ambient Air Quality Directive (25 exceedance days per year, averaged over 3 years; black) and the revised Directive (EU) 2024/2881 (18 days per year, averaged over 3 years; blue) applying from 2030.

This is consistent with the finding that the daytime trend of O₃ P95 values contained the only negative trend pattern since 2009 (Figure 11), especially during the summer month at rural and forest sites. To investigate the meteorological controls on high-O₃ episodes for five-year periods before and after the main changepoint in 2017/2018, daily maximum air temperature observations from all stations within each station category were pooled and grouped into fixed 2.5°C bins, and the corresponding mean MDA8 O₃ concentration was calculated for each bin (Figure 14a-d). At all four station categories, MDA8 O₃ increases as maximum daily temperature rises. A transition to a steeper temperature dependence and reordering of relative slopes is visible above ~20°C for the periods 2007–2011, 2012–2016 and 2019–2023. The MDA8 O₃ values for 2019–2023 are higher than for 2012–2016 and 2007–2011 at maximal air temperatures below ~20°C especially at urban and peri-urban locations (Figure 14a,b). Hence, the dependence at lower temperatures increased (Welch's t-test significant, $p < 0.05$), likely reflecting the effect of reduced titration with NO during nighttime, autumn, and winter, as supported by the decreasing NO₂ concentrations with increasing maximal winter air temperature (Figures 8 and A4) (Nussbaumer et al., 2026). In contrast, the relationship for the period 2019–2023 is comparable to, or slightly below, the period 2012–2016 at air temperatures above 20°C (Welch's t-test significant for all stations, except urban ($p < 0.05$), see Figure 14). The relative slope of the interpolated MDA8 O₃ response between maximum diurnal air temperature values of 20°C and 30°C varies between ~44 and ~62 %, with highest values at forest stations, and the overall weakest relative temperature response in 2019–2023



(Figure 14a-d). This indicates that above $\sim 20^{\circ}\text{C}$ the air temperature sensitivity of MDA8 O_3 decreased at peri-urban, rural and forest stations after 2018. The weaker response at higher temperatures is consistent with a transition toward more NO_x -limited conditions in summer, under which declining NO_x availability increasingly constrains temperature-enhanced photochemical O_3 production. This is comparable to findings of previous studies and earlier time periods indicating a decline of the climate penalty in southwestern Germany (Otero et al., 2021). Our study shows that this trend is progressing and can explain the lower number of MDA8 exceedance days in recent years.

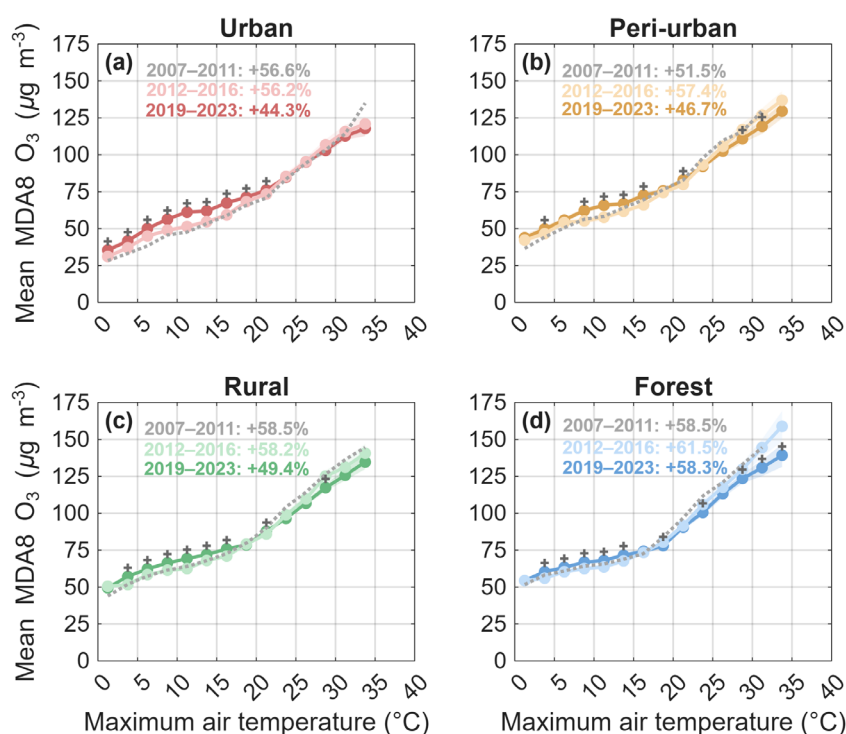


Figure 14. Relationship between maximum daily air temperature and daily maximum 8-hour average (MDA8) O_3 concentration for (a) urban, (b) peri-urban, (c) rural, and (d) forest station categories during 2007–2011, 2012–2016 and 2019–2023. Daily observations from all stations within each station category were pooled and grouped into fixed 2.5°C maximum air temperature bins. Symbols represent the mean MDA8 O_3 concentration for each temperature bin, and shaded areas indicate the 95% bootstrap confidence intervals of the bin means. Crosses denote temperature bins where differences between the 2012–2016 and 2019–2023 periods are statistically significant (Welch's t-test, $p < 0.05$). The annotated values indicate the relative increase in mean MDA8 O_3 between maximum daily air temperatures of 20°C and 30°C (cf. Nussbaumer et al., 2026).

The relationship between MDA8 O_3 and maximum daytime BLH generally shows lowest relative slopes at rural and forest stations (Figure 15). The response in 2019–2023 is characterized by a smaller relative slope between a maximum BLH range



of 1000 and 2000 m, resulting in a lower relative increase in MDA8 O₃ (40–53%) compared with earlier periods (53–65%). As for air temperature, the increase in MDA8 O₃ at higher maximum BLH levels was weaker after the changepoint occurrence (Figure 5b) in 2019–2023 than during 2012–2016 (albeit not always statistically significant), suggesting that the enhanced vertical mixing did not translate into proportionally larger peak O₃ increases. However, maximal BLH values below 1500 m were associated with a significant increase in MDA8 O₃. Hence, the correspondence between the 2017/2018 changepoints in the long-term O₃ component (Figure 4d) and daily maximum BLH (Figure 5b) reflects both increasing maximum BLH, which enhances MDA8 O₃ through deeper daytime mixing and entrainment, and reduced NO titration under shallow to medium maximum BLH conditions (Figure A5), contributing to higher MDA8 O₃ outside the summer season. This effect is most significant at urban and peri-urban stations (Figure 15a,b).

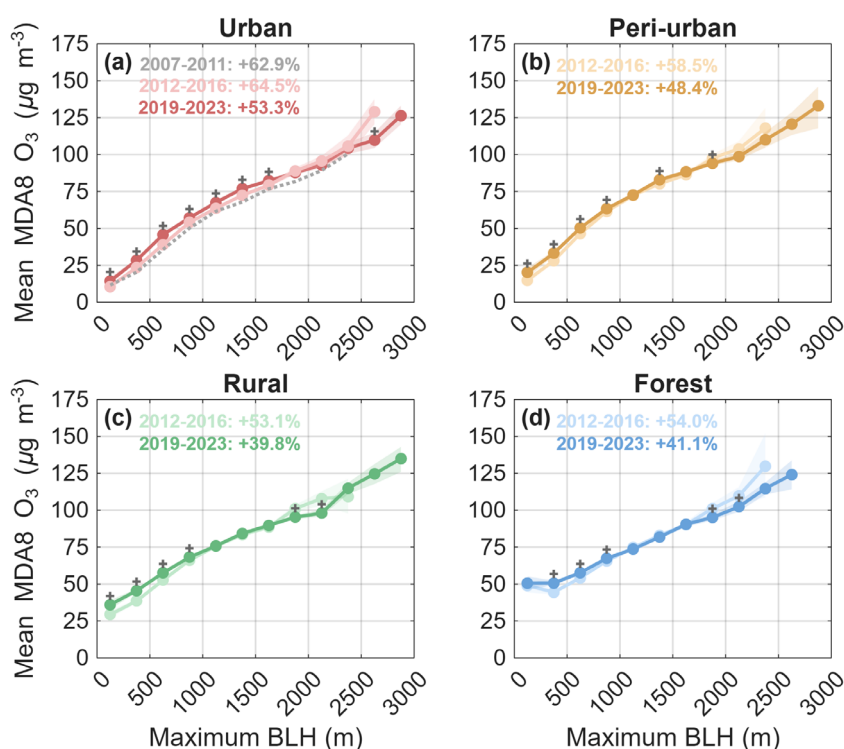


Figure 15. Same as Figure 14, but for the maximum diurnal boundary layer height (BLH). Results for 2007–2011 are only shown for urban stations because for the other station categories no difference to the later periods were observed. Daily observations from all stations within each station category were pooled and grouped into fixed 250 m maximum BLH bins. The annotated values denote the relative slope of the interpolated O₃ response between maximum BLH values of 1000 and 2000 m.



3.4.2 Trends in AOT40

The mean cumulative AOT40 during the growing season (Figure 16) for rural and forest stations in the Greater Region exhibit a very similar temporal evolution. During 2007–2011 (grey), cumulative AOT40 increases rapidly from April onward, indicating that O₃ exposure accumulated earlier in the growing season (Figure 16). In contrast, the 2012–2016 (dashed green) and especially the 2019–2023 (solid green) curves remain lower during spring and only begin to increase more rapidly from approximately June onwards.

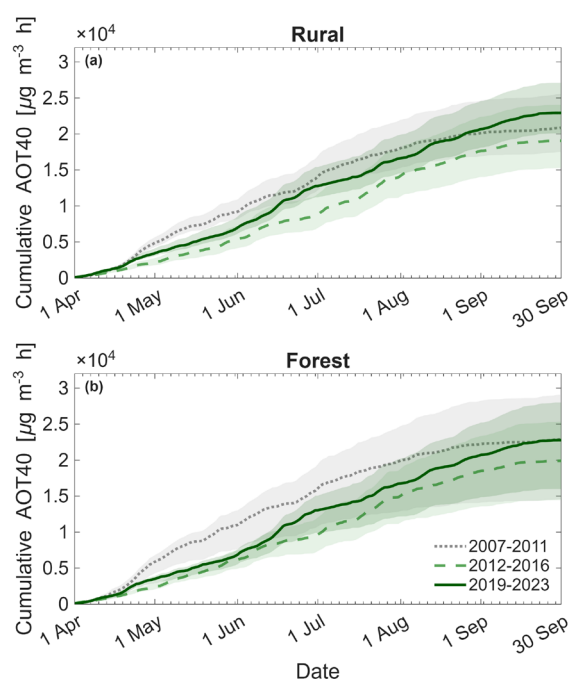


Figure 16. Mean cumulative AOT40 during the growing season (April to September) for (a) rural (a) and (b) forest monitoring sites in the Greater Region. Curves show the cumulative daily AOT40 averaged over the 5-year periods 2007–2011, 2012–2016, and 2019–2023. Shaded envelopes represent 95% bootstrap confidence intervals of the mean ($n = 1000$ resamples of station means). AOT40 was calculated as the cumulative hourly O₃ exposure exceeding 80 $\mu\text{g m}^{-3}$ (40 ppb) during daylight hours using the Openair R package (Carslaw and Ropkins, 2012).

There was a substantial reduction of AOT40 from the period 2007–2011 to 2012–2016, while the cumulative AOT40 again increased after the 2017/2018 changepoint during the 2019–2023 period, especially at rural stations and from mid-June to mid-July. By the end of September, the mean cumulative AOT40 for 2019–2023 again exceeds the one for both previous periods at rural stations and is comparable to the one for 2007–2011 at forest stations. This suggests that NO_x emission reductions alone have not been sufficient to sustain reductions in ecosystem O₃ exposure in the Greater Region. However, the influence of increasing air temperatures on stomatal conductance and O₃ uptake (Anav et al., 2019; Clifton et al., 2020), is not explicitly accounted for in the AOT40 metric.



4 Conclusions

For the first time, a comprehensive long-term analysis of surface O₃ and NO₂ was conducted for urban, peri-urban, rural, and forest locations in the Greater Region, covering the period 2000 to 2024. While Luxembourg has achieved the largest reduction in inventory-based NO_x emissions among EU Member States over the past two decades, this decline is only partially reflected in measured concentration levels, as regional air quality remains strongly influenced by extensive cross-border commuting and associated transport emissions. Using the Bayesian Estimator of Abrupt change, Seasonal change, and Trend (BEAST) we found a pronounced changepoint in the O₃ long-term trend component around 2017/2018, that was accompanied by a change in the seasonal component of NO₂, diurnal maximum boundary layer height and air temperature. Particular attention was given to the period from 2009 to 2024, when the most pronounced changes occurred and when the time series extends substantially beyond those considered in most previous European studies. During this period, NO₂ decreased on average by 50 %, while O₃ increased between 10 % and 27 % in urban, peri-urban, rural, and forest environments. In contrast to the declining O₃ trends reported for other rural parts of Western Europe, surface O₃ concentrations in the Greater Region have generally continued to increase. In agreement with previous studies, the strongest increases occurred at the lower end of the O₃ distribution, particularly at urban and peri-urban stations, whereas trends in the upper tail were substantially weaker. Peak O₃ concentrations remained stable or decreased slightly under some conditions, particularly during summer. Overall, the previously identified narrowing of the O₃ distribution associated with increasing low O₃ concentrations and decreasing peak concentrations in Western Europe was absent in the Greater Region in recent years, where the seasonal distribution was shifted towards higher concentrations. Although summer peak daytime concentrations and exceedances of regulatory target values have not increased, average O₃ concentrations exposure are higher after the 2017/2018 changepoint. We also detected increasing nighttime O₃ trends that may alter nocturnal oxidation chemistry and atmospheric oxidizing capacity. The decline of the weekend effect indicates an ongoing transition from VOC-limited towards NO_x-limited O₃ chemistry, particularly at higher temperatures during summer in rural, and forest locations. However, the small and insignificant ΔMDA8 during 2019–2023 suggests that the transition towards NO_x-limited conditions is not yet complete. Reduced titration of O₃ with NO at low to moderate temperatures, particularly in spring and autumn, together with increasingly favourable meteorological conditions such as deeper daytime boundary layers, continues to offset the benefits of declining NO_x emissions and sustain positive O₃ trends, especially at urban and peri-urban locations. Overall, the substantial reduction in NO_x emissions has been effective in limiting summer peak O₃ concentrations and corresponding MDA8 exceedance days but has not yet produced a corresponding decline in regional background O₃. Cumulative AOT40 for the period 2019–2023 returned to levels comparable to or even exceed those observed in 2007–2011 by the end of the growing season. Sustained and coordinated reductions in NO_x, together with appropriate control of VOC emissions, are therefore required to move the chemical regime towards conditions in which further precursor reductions consistently decrease O₃. At the same time, increasing temperatures and changing boundary layer dynamics demonstrate that future O₃



mitigation must account for a changing climate, which can partly offset the benefits achieved through anthropogenic
 600 emission reductions.

Appendix A

Table A1: Descriptive statistics of O₃ and NO₂ concentrations at all monitoring stations over the study period. For each site, the mean, standard deviation, hourly maximum concentration, and the date of the hourly maximum are reported for O₃ and NO₂ (µg m⁻³).

Station	Mean O ₃ (µg m ⁻³)	Std O ₃	Max O ₃ (µg m ⁻³)	Date O ₃ max	Mean NO ₂ (µg m ⁻³)	Std NO ₂	Max NO ₂ (µg m ⁻³)	Date NO ₂ max
Beckerich	46.9	30.9	234	19/07/2006	11.8	9.1	108	13/03/2005
Beidweiler	56.0	27.9	233	20/06/2017	10.0	7.9	77	11/01/2009
Luxembourg- Bonnevoie	36.9	28.1	199.5	05/08/2003	32.2	17.5	236.9	18/01/2016
Esch-sur- Alzette	45.5	29.8	209.5	09/08/2003	24.8	17.5	265.5	11/03/2004
Habay La Neuve	53.1	28.5	223	15/07/2003	10.6	9.3	251	02/02/2008
Jonville en Woevre	52.3	27.8	221	19/07/2006	7.6	7.5	104.3	01/12/2017
Hunsrück- Leisel	66.1	26.9	232	09/08/2003	7.2	7.1	96	05/03/2004
Metz-Centre	44.2	30.1	197.2	09/08/2003	21.1	15.5	152	22/11/2011
Nancy-Charles III	45.3	29.4	192	04/08/2018	21.3	16.1	528	29/09/2010
Sainte Ode	60.1	25.0	235	11/08/2003	6.4	6.4	69	11/01/2009
Scy-Chazelles	47.6	31.6	244	12/08/2003	16.3	14.5	172	12/02/2008
Thionville- Garche	48.7	30.9	222	19/07/2006	16.1	11.5	124.8	27/02/2003
Vianden	66.6	28.3	281.5	02/07/2015	7.8	7.5	226	04/01/2002
Vielsalm	50.6	27.0	283	08/08/2003	6.7	6.7	163	03/02/2006



Station	Mean O ₃ (µg m ⁻³)	Std O ₃	Max O ₃ (µg m ⁻³)	Date O ₃ max	Mean NO ₂ (µg m ⁻³)	Std NO ₂	Max NO ₂ (µg m ⁻³)	Date NO ₂ max
Westeifel- Wascheid	64.5	25.7	262	08/08/2003	7.0	6.7	116	21/03/2003

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Table A2. Long-term trend slopes in NO₂ concentrations ($\mu\text{g m}^{-3} \text{ yr}^{-1}$) derived from QR-MBB ($\tau = 0.5$). Reported metrics include the slope of the long-term and residual components, the standard error (SE) of the slope, the associated t value (ratio between the estimate and uncertainty or signal-to-noise ratio, SNR) used to determine the p values. For $p \leq 0.01$ and $\text{SNR} \geq 3$ very high certainty and for $0.05 \geq p > 0.01$ and $2 \leq p < 3$ high certainty and $\text{SNR} < 1$ no evidence (Chang et al., 2023). Positive slopes indicate long-term increases in NO₂.

Station	Slope	Slope SE	t value	p value
Beckerich	-0.52	0.064	-8.1	<0.001
Luxembourg- Bonnevoie	-1.27	0.196	-6.5	<0.001
Beidweiler	-0.49	0.050	-9.8	<0.001
Esch-sur-Alzette	-1.00	0.112	-8.9	<0.001
Habay La Neuve	-0.37	0.050	-7.5	<0.001
Jonville en Woevre	-0.22	0.069	-3.2	<0.001
Hunsrück-Leisel	-0.36	0.064	-5.6	<0.001
Metz-Centre	-0.96	0.083	-11.6	<0.001
Nancy-Charles III	-0.87	0.093	-9.3	<0.001
Sainte Ode	-0.34	0.077	-4.4	<0.001
Scy-Chazelles	-0.74	0.095	-7.8	<0.001
Vianden	-0.36	0.070	-5.2	<0.001
Vielsalm	-0.26	0.055	-4.7	<0.001
Westeifel-Wascheid	-0.35	0.047	-7.5	<0.001



Table A3. Long-term trend slopes in NO₂ concentrations (µg m⁻³ yr⁻¹) derived from SSA decompositions. Reported metrics include the Theil-Sen slope of the long-term and residual components, the Mann-Kendall (MK) rank correlation coefficient (τ), associated p value, and the MK hypothesis test result (1 indicates rejection of the null hypothesis of no monotonic trend). Negative slopes indicate significant long-term declines in NO₂.

Station	Theil-Sen slope	MK τ	MK p-value	MK hypothesis
Beckerich	-0.58	-0.577	<0.001	1
Luxembourg-Bonnevoie	-1.30	-0.592	<0.001	1
Beidweiler	-0.56	-0.623	<0.001	1
Esch-sur-Alzette	-0.96	-0.463	<0.001	1
Habay La Neuve	-0.49	-0.569	<0.001	1
Jonville en Woevre	-0.35	-0.420	<0.001	1
Hunsrück-Leisel	-0.41	-0.595	<0.001	1
Metz-Centre	-1.04	-0.627	<0.001	1
Nancy-Charles III	-0.90	-0.528	<0.001	1
Sainte Ode	-0.36	-0.460	<0.001	1
Scy-Chazelles	-0.77	-0.513	<0.001	1
Vianden	-0.41	-0.548	<0.001	1
Vielsalm	-0.29	-0.461	<0.001	1
Westeifel-Wascheid	-0.39	-0.627	<0.001	1



620 **Table A4.** Long-term trend slopes in NO₂ concentrations (µg m⁻³ yr⁻¹) derived from STL decompositions. Reported metrics include the Theil-Sen slope of the long-term and residual components, the Mann-Kendall (MK) rank correlation coefficient (τ), associated p value, and the MK hypothesis test result (1 indicates rejection of the null hypothesis of no monotonic trend). Negative slopes indicate significant long-term declines in NO₂.

Station	Theil-Sen slope	MK τ	MK p-value	MK hypothesis
Beckerich	-0.58	-0.601	<0.001	1
Luxembourg- Bonnevoie	-1.36	-0.637	<0.001	1
Beidweiler	-0.55	-0.660	<0.001	1
Esch-sur-Alzette	-0.98	-0.513	<0.001	1
Habay La Neuve	-0.50	-0.604	<0.001	1
Jonville en Woevre	-0.36	-0.508	<0.001	1
Hunsrück-Leisel	-0.40	-0.639	<0.001	1
Metz-Centre	-1.05	-0.677	<0.001	1
Nancy-Charles III	-0.89	-0.573	<0.001	1
Sainte Ode	-0.35	-0.518	<0.001	1
Scy-Chazelles	-0.78	-0.541	<0.001	1
Thionville-Garche	-0.42	-0.582	<0.001	1
Vianden	-0.30	-0.475	<0.001	1
Westeifel-Wascheid	-0.38	-0.649	<0.001	1

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Table A5. Long-term trend slopes in O_3 concentrations ($\mu g\ m^{-3}\ yr^{-1}$) derived from QR-MBB ($\tau = 0.5$). Reported metrics include the slope of the long-term and residual components, the standard error (SE) of the slope, the associated t value (ratio between the estimate and uncertainty or signal-to-noise ratio, SNR) used to determine the p values. For $p \leq 0.01$ and $SNR \geq 3$ very high certainty and for $0.05 \geq p > 0.01$ and $2 \leq p < 3$ high certainty and $SNR < 1$ no evidence (Chang et al., 2023).

630 Positive slopes indicate long-term increases in O_3 .

Station	Slope	Slope SE	t value	p value
Beckerich	0.51	0.214	2.4	<0.05
Luxembourg- Bonnevoie	0.91	0.210	4.3	<0.001
Beidweiler	0.34	0.238	1.4	0.153
Esch-sur-Alzette	0.63	0.141	4.5	<0.001
Habay La Neuve	0.41	0.150	2.7	<0.05
Jonville en Woevre	0.62	0.168	3.7	<0.001
Hunsrück-Leisel	0.40	0.202	2.0	<0.05
Metz-Centre	0.70	0.186	3.8	<0.001
Nancy-Charles III	0.83	0.188	4.4	<0.001
Sainte Ode	0.61	0.129	4.7	<0.001
Scy-Chazelles	0.65	0.176	3.7	<0.001
Thionville-Garche	0.42	0.181	2.3	<0.05
Vianden	0.47	0.160	3.0	<0.005
Westeifel-Wascheid	0.36	0.137	2.6	<0.05



Table A6. Long-term trend slopes in O₃ concentrations (µg m⁻³ yr⁻¹) derived from SSA decompositions. Reported metrics include the Theil-Sen slope of the long-term and residual components, the Mann-Kendall (MK) rank correlation coefficient (τ), associated p value, and the MK hypothesis test result (1 indicates rejection of the null hypothesis of no monotonic trend). Positive slopes indicate significant long-term increases in O₃.

Station	Theil-Sen slope	MK τ	MK p-value	MK hypothesis
Beckerich	0.35	0.141	<0.005	1
Luxembourg- Bonnevoie	0.92	0.303	<0.001	1
Beidweiler	0.37	0.120	<0.05	1
Esch-sur-Alzette	0.54	0.188	<0.001	1
Habay La Neuve	0.38	0.130	<0.05	1
Jonville en Woevre	0.56	0.198	<0.001	1
Hunsrück-Leisel	0.36	0.110	<0.05	1
Metz-Centre	0.63	0.204	<0.001	1
Nancy-Charles III	0.75	0.235	<0.001	1
Sainte Ode	0.62	0.219	<0.001	1
Scy-Chazelles	0.59	0.180	<0.001	1
Thionville-Garche	0.38	0.118	<0.05	1
Vianden	0.41	0.161	<0.05	1
Westeifel-Wascheid	0.32	0.114	<0.05	1



Table A7. Long-term trend slopes in O₃ concentrations (µg m⁻³ yr⁻¹) derived from STL decompositions. Reported metrics include the Theil-Sen slope of the long-term and residual components, the Mann-Kendall (MK) rank correlation coefficient (τ), associated p value, and the MK hypothesis test result (1 indicates rejection of the null hypothesis of no monotonic trend). Positive slopes indicate significant long-term increases in O₃.

Station	Theil-Sen slope	MK τ	MK p-value	MK hypothesis
Beckerich	0.39	0.182	<0.001	1
Luxembourg- Bonnevoie	0.97	0.371	<0.001	1
Beidweiler	0.45	0.174	<0.001	1
Esch-sur-Alzette	0.61	0.256	<0.001	1
Habay La Neuve	0.37	0.164	<0.001	1
Jonville en Woevre	0.61	0.247	<0.001	1
Hunsrück-Leisel	0.46	0.170	<0.001	1
Metz-Centre	0.69	0.308	<0.001	1
Nancy-Charles III	0.85	0.342	<0.001	1
Sainte Ode	0.68	0.309	<0.001	1
Scy-Chazelles	0.60	0.235	<0.001	1
Thionville-Garche	0.39	0.163	<0.001	1
Vianden	0.46	0.210	<0.001	1
Westifel-Wascheid	0.42	0.185	<0.001	1



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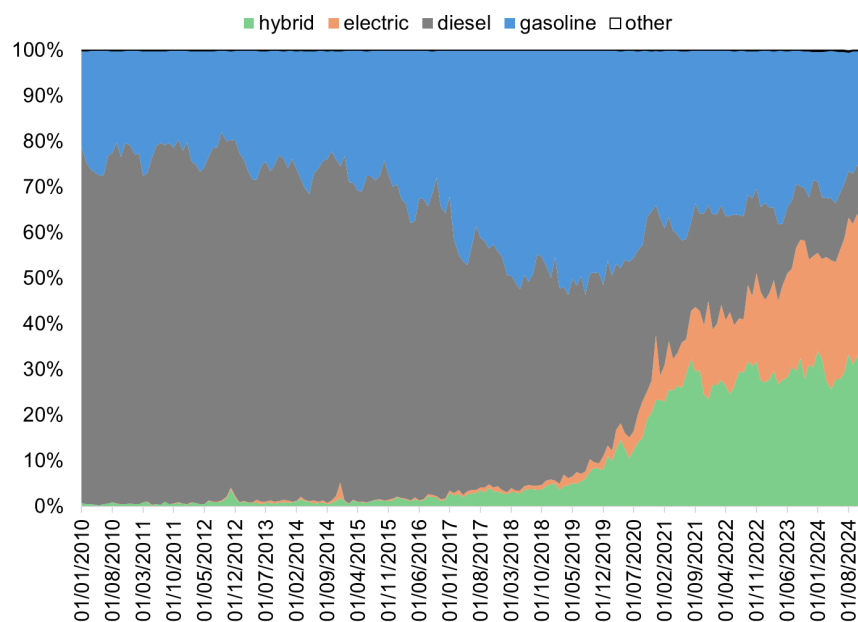


Figure A1. Monthly relative shares (%) of passenger car registrations by propulsion type (hybrid, electric, diesel, gasoline, other) from 2010 to 2024 in Luxembourg (data source: STATEC). Diesel vehicles dominated the fleet composition until approximately 2017, after which their share declined markedly. From 2018 onward, a rapid increase in hybrid and battery-electric vehicles is observed, accompanied by a stabilization of gasoline vehicles.

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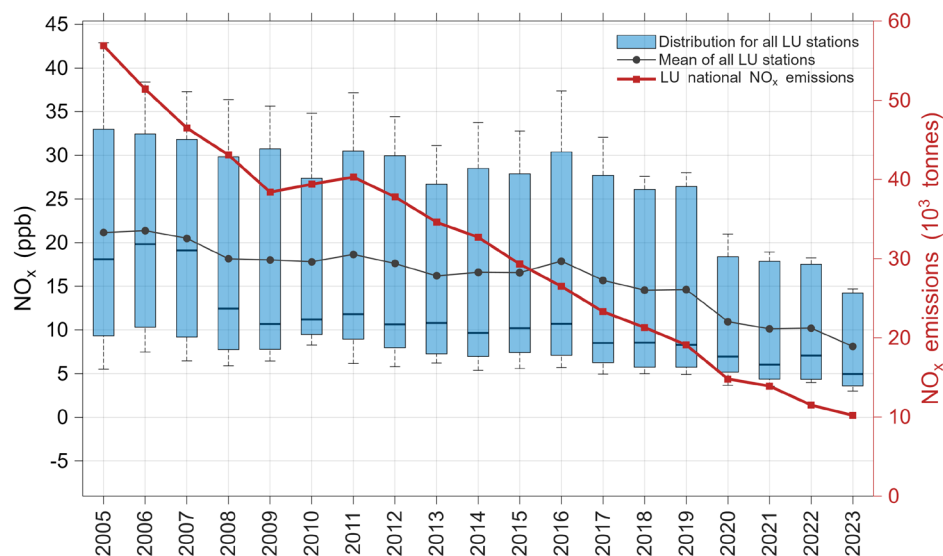


Figure A2. Temporal evolution of ambient NO_x and national NO_x emissions in Luxembourg from 2005 to 2023. Boxplots show the distribution of annual mean NO_x mixing ratios for the available monitoring stations in Luxembourg (Table 1), with the grey line indicating the annual mean for the stations. The red line shows annual national emissions of nitrogen oxides (NO and NO₂, expressed as NO_x) from the emission inventory, reported in 10³ tonnes (source STATEC, <https://statistiques.public.lu>).

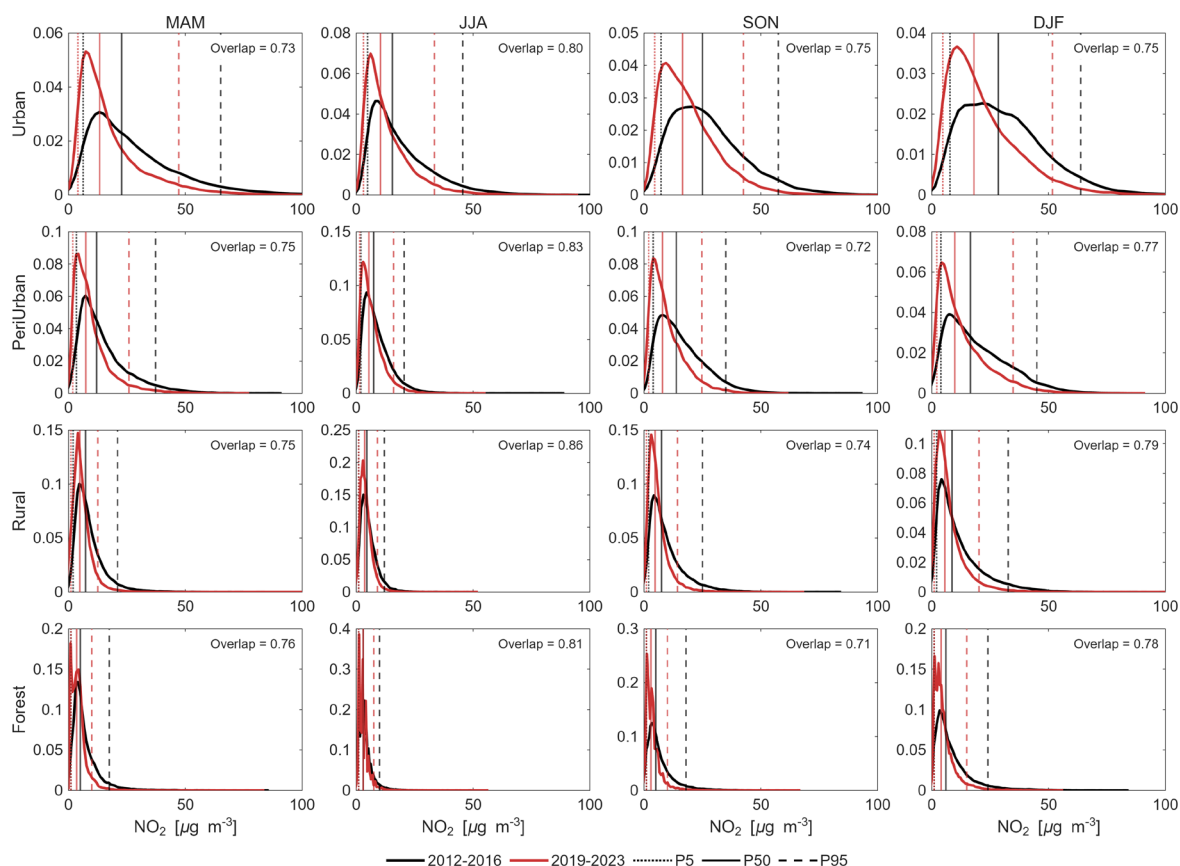


Figure A3. Seasonal probability density functions of hourly NO₂ concentrations for urban, peri-urban, rural, and forest monitoring sites in the Greater Region during 2012–2016 (black) and 2019–2023 (red). Columns represent spring (MAM), summer (JJA), autumn (SON), and winter (DJF). Curves show kernel density estimates of the NO₂ concentration distributions. Vertical lines indicate the 5th (P5, dotted), 50th (P50, solid), and 95th (P95, dashed) percentiles for each period. The overlap coefficient, calculated as the integral of the minimum of the two kernel density estimates, quantifies the similarity between the distributions, with values approaching unity indicating highly similar distributions and lower values indicating changes of the distribution.

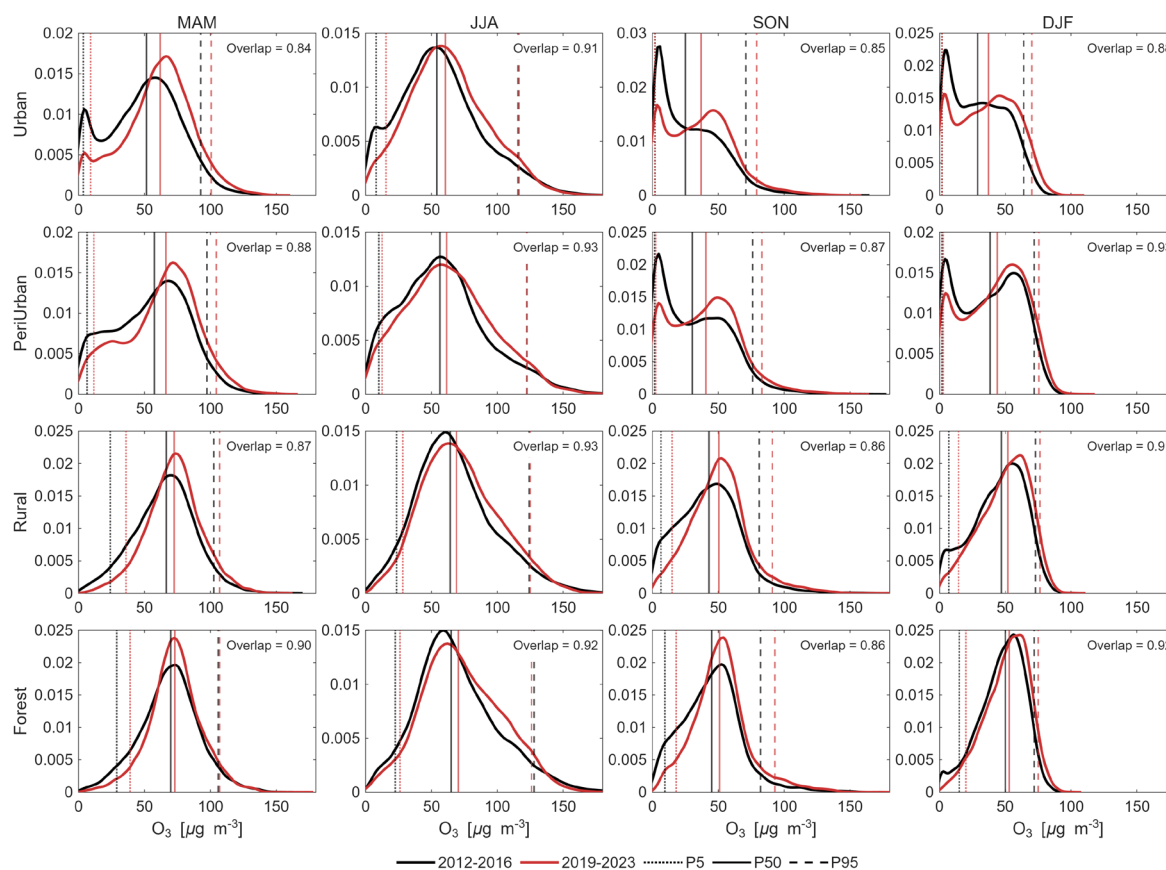


Figure A4. Same as Figure A3, but for O_3 .

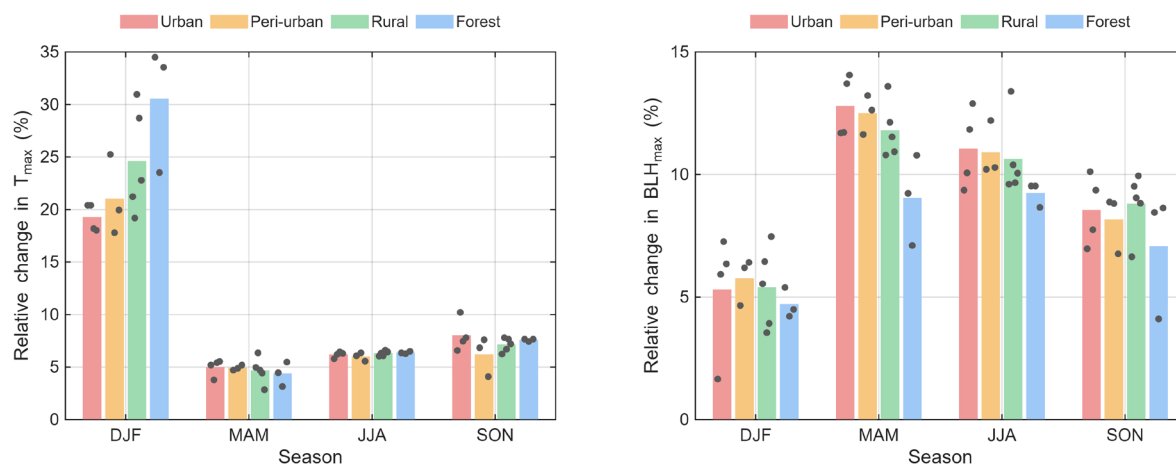


Figure A5. Relative seasonal change in (a) daily maximum air temperature (T_{max}) and (b) daily maximum boundary layer height (BLH_{max}) between 2012–2016 and 2019–2023 for urban, peri-urban, rural, and forest station categories. For each station and season, annual seasonal mean values were first calculated and averaged over each five-year period. Relative changes were calculated with respect to 2012–2016. Bars represent the mean relative change for stations within each category, and grey symbols indicate the individual station values.

Code and data availability

All datasets used in this study, as well as the data products generated during the analysis, are available from the corresponding author upon request.

Author contributions

IT conceptualised the study and conducted the main formal analysis, with contributions from JJ and AK. PP made contributions to data processing and visualisation. Funding acquisition was led by JJ and AK. IT prepared the manuscript with contributions from JJ. All co-authors reviewed and edited the manuscript.

Competing interests

All authors declare that they have no competing interests that could have influenced the results reported in this paper.



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