Dear Reviewer,

We sincerely appreciate the valuable and constructive feedback from Reviewer #1, which has significantly contributed to improving our manuscript. Below, we provide a detailed, point-by-point response to all comments. The reviewer's comments are presented in *italic black*, our responses in blue, and the corresponding revisions in the manuscript and Supporting Information are highlighted in red.

Thank you for your time and consideration.

All line numbers given in our below replies refer to the revised manuscript (color-tracked) and Supporting Information (color-tracked).

General comments

The paper analyses the effect of NOx and VOC emission reductions on surface ozone using observations and a model in Saxonia. The paper fits to the scope of ACP. There are mostly technical issues that have to be fixed before publication like missing definitions or information on input data and model version, and the need to improve figures and tables.

We thank the reviewer for his/her positive overall assessment of our manuscript.

Specific comments

R1-C1: *Line 12: Is NOx=NO+NO_2 or are also NO_3 and N_2O_5 included? Please define for clarity.*

A1: Here the term "oxides of nitrogen" (NO_x) only includes nitric oxide (NO) and nitrogen dioxide (NO_2). In the manuscript, a clarified definition of "oxides of nitrogen (NO_x = NO+NO_2)" has been added in the line 12 and now reads:

The findings indicate that despite reductions in oxides of nitrogen (NO_x = NO+NO_2) concentrations across all sites, O_3 pollution in Saxony has in fact worsened over the past decade, especially in densely populated urban areas.

According to ACP's submission requirements, abbreviations must be defined at their first occurrence in both the abstract and the main manuscript. So "= NO+NO_2" was added at line 30 of the manuscript as well, the phrase now reads:

oxides of nitrogen (NO_x = NO+NO_2)

R1-C2: *Line 16: Spell out "VOC" already here, line 29 (or line 19, hidden) is too late.*

A2: The definition of VOCs now appears earlier in the manuscript. "Volatile organic compounds (VOCs)" was added to line 17 of the manuscript, the revised sentence now reads:

To diagnose O_3 formation and the controlling effects of NO_x and volatile organic compounds (VOCs) over the past decades in this region, for the first time, detailed photochemical box modelling was performed by means of the complex MCM (Master Chemical Mechanism).

In line 31 of manuscript, the phrase of "volatile organic hydrocarbons" has been corrected to "volatile organic compounds".

The abstract must be under 250 words per submission guidelines. To meet this requirement, some words were removed or replaced in lines 10-11 of the article. The revised sentences are as follows.

Given its importance for human health, vegetation, and climate, trends in ground-level ozone (O_3) concentrations in eastern Germany were systematically analysed using the long-term O_3 data from 16 measurement stations.

R1-C3: *Line 125: I suppose "p" is probability. Please spell out since "p" can be used also for pressure.*

A3: p-values means probability. Now "(probability)" has been added in the line 127 of the manuscript and the sentence reads:

Theil-Sen function derives p-values (probability) and uncertainties by bootstrap simulations.

R1-C4: *Line 131: Is this a boxmodel on trajectories? Please more details, from the given reference that is possible. The information on meteorology provided in the following lines is too general.*

A4: SPACCIM is used in the present study as box model (box fixed at a certain location). However, SPACCIM can be also used as air parcel model with changing conditions along a predefined trajectory. In the present study, temperature, pressure, and relative humidity are fixed parameters based on observed conditions.

A sentence describing the SPACCIM model has been added to lines 133-135 of the manuscript and "box model" has been indicated in line 133, now the revised sentences are shown below.

To understand the role of photochemistry for O_3 concentration evolution in Saxony, photochemical simulations were performed with the air parcel box model SPACCIM (SPectral Aerosol Cloud Chemistry Interaction Model). SPACCIM combines a size-resolved multiphase chemistry model with a microphysical model, enabling both to function independently while accounting for their interdependencies.

Some specific information on meteorology has been included in lines 149-152 of the article, and some sentences have been revised from line 146 to line 152. They now read:

Besides emission values, other initial parameters had to be adjusted to their typical daytime and nighttime levels under rural conditions (see Table S4 for details). For meteorological parameters and trace gas concentrations (except SO_2, HONO and PAN), data was derived from measurements in Sect. 2.1. The air temperature was set to 15°C in summer and 4°C in winter. The pressure was kept constant at 1000 hPa for both seasons, while the relative humidity was maintained at 70%. Ratio of solar radiation, defined as the mean value between 10:00 and 13:00 divided by the maximum clear sky radiation value during the same period, was calculated to 0.7 in summer and 0.4 in winter.

R1-C5: *Line 133: Which version of CAPRAM? You need it on the website cited in the supplement.*

A5: In the present study, only the detailed near-explicit gas-phase chemistry mechanism MCM v3.3.1 (Master Chemical Mechanism) is applied (this information can be found in the lines 138-139 of the manuscript). No version of CAPRAM (Chemical Aqueous Phase Radical Mechanism) is used. Although the SPACCIM model framework allows for the coupling of MCM with CAPRAM to simulate multiphase chemistry, it also enables the use of each mechanism independently. Here, SPACCIM is employed solely with the gas-phase chemistry mechanism MCM v3.3.1.

To avoid misunderstanding, the revised paragraph describing the mechanisms used in lines 132-139 of the text is as follows: To understand the role of photochemistry for O_3 concentration evolution in Saxony, photochemical simulations were performed with the air parcel box model SPACCIM (SPectral Aerosol Cloud Chemistry Interaction Model). SPACCIM combines a size-resolved multiphase chemistry model with a microphysical model, enabling both to function independently while accounting for their interdependencies. Detailed descriptions of the SPACCIM model framework can be found in Wolke et al. (2005). In the present study, only the detailed near-explicit gas-phase chemistry mechanism, MCMv3.3.1, is used, which comprises 17224 reactions (http://mcm.york.ac.uk/MCM/)(Saunders et al., 2003).

Additionally, one reference (Saunders et al., 2003) has been inserted in lines 879-881 of the manuscript. The reference now reads:

Saunders, S. M., Jenkin, M. E., Derwent, R. G., and Pilling, M. J.: Protocol for the development of the Master Chemical Mechanism, MCM v3 (Part A): tropospheric degradation of non-aromatic volatile organic compounds, Atmospheric Chemistry and Physics, 3, 161-180, 2003.

R1-C6: *Line 141: After "S1" "showing NOx emissions of the main roads and urban centers".*

A6: In the manuscript, the phrase of "showing NO_x emissions of the main roads and urban centres" has been added in lines 145-146, it now reads:

(see Fig. S1 showing NO_x emissions of the main roads and urban centers)

R1-C7: *Line 147: The fixed value for HONO is not far off from typical values but it might be better to keep HONO/NOx constant which might help to get a faster increase of NO due to HONO photolysis after sunrise in Fig. 8, especially in winter (Elshorbany et al. 2012). Maybe for a sensitivity study or outlook.*

A7: Our model is initialized by setting the pollutant concentrations. Usually, we do not constrain concentrations based on measured time series. This artificial temporal fixation of the model is not done in our group because we want also to understand current model limitations which gets mask by constraining certain key species. Moreover, with the current model framework, it is difficult to maintain a constant HONO/NO_x ratio throughout the simulation. Nevertheless, we think the idea to keep one compound such as HONO constant might be a way to identify missing sources/sinks and could be a valuable task for future sensitivity studies.

The authors acknowledge that this is a very interesting point, and we will keep it in mind for further model development.

R1-C8: *Line 166: Does table S4 refer to the pre-runs?*

A8: No, Table S4 refers to the prepared initial concentrations for further 24-hour simulations, which are defined as the base case simulations. Besides, as a result of adding one new table (Table S3) showing the emission data (see R1-C14), the numbering of original Table S4 now is revised to Table S5.

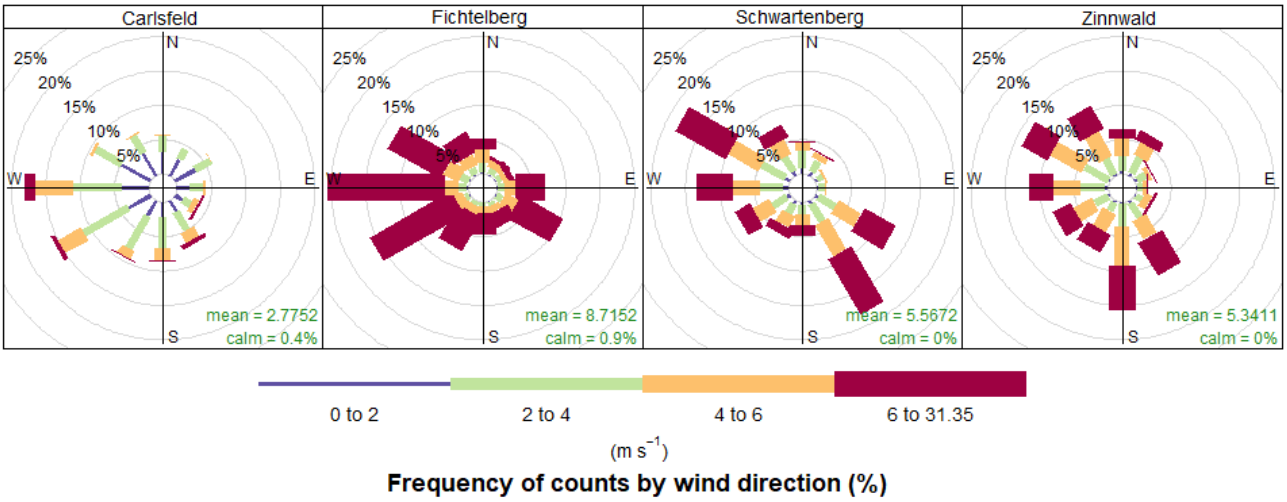
The revised text in lines 174-176 of the article is now:

The output concentrations from the last day of the pre-run simulations were used as the initial and boundary chemical data for the final 24-hour modelling. Dominant initial concentrations are given in Table S5 in the Supporting Information.

R1-C9: *Line 255: Isn't Fichtelberg due to the prevailing wind direction less affected by air masses from polluted regions than the other mountain stations?*

A9: We thank the reviewer for this insightful comment to make us gain a better understanding of ozone trends at Fichtelberg.

The following figure presents wind rose plots at different mountain sites over the past 10 year (2011-2020), generated using the *windRose* function of the package *openair* (Carslaw and Ropkins, 2012) in R (R core Team, 2020). At Fichtelberg, wind direction is predominantly westerly, accompanied by relatively strong wind speeds. A similar prevailing wind direction is observed at Carlsfeld, though with lower wind speeds. Both sites are indeed less affected by polluted air masses than the other two mountain sites Schwartenberg and Zinnwald, which have higher frequencies of air masses from the South and South-East (Czech Republic). However, in contrast to Carlsfeld, with a similar wind rose, only Fichtelberg observed the strongly decreasing trend over the past 10 years. We therefore believe -as already discussed in the original manuscript- that it might be the higher impact of free tropospheric air masses at Fichtelberg, which makes its trend differ from the lower-altitude sites in the region.



R1-C10: *Figure 4, caption: Better write "differences" as in the text.*

A10: The caption of Fig. 4 has been revised. See lines 275-277 in the article:

Figure 4: Mean yearly O_3 concentration differences, i.e. decrements, between different station types. Yellow point lines represent decrements from mean mountain to mean rural stations concentrations, blue point lines represent rural to urban, and red point lines represent urban to traffic stations decrements.

R1-C11: *Line 266: From Fig. 4 it can be also said that the difference between rural and urban stays about constant if the first 2 points are included. Please improve text in this paragraph.*

A11: We thank the reviewer for this useful comment. The reviewer is right. The difference between rural and urban indeed stays about constant if the first 2 points in 1997-1998 are considered.

Now the text in this paragraph (at lines 281-294) has been revised, the paragraph now reads:

The urban decrement, i.e. rural to urban background difference, remained rather stagnant around 7 µg m^{-3} in all years except for the period 1999-2003. The traffic decrement from the urban background to the one long-term O_3 traffic station DD-Nord also decreased from about 15 to about 5 - 7 µg m^{-3} and is now at a similar level as the urban decrement. If the O_3 trends mentioned above continue in a similar way in the future, it can be expected that the typical O_3 concentrations at traffic stations will become more similar to those in the urban background and rural background concentrations will become increasingly similar to those at the mountain sites. The Air Quality Expert Group (2021) reported a gradual convergence of urban and rural O_3 levels in the UK from 2000 to 2019, which is in contrast to the stable difference in O_3 concentration between urban and rural background observed since 2004 in the present study. The reason for this is the similarly increasing O_3 trends at urban and rural background sites in Saxony (Fig. 3), keeping the urban to rural decrement roughly similar. In any case, due to the typical settlement densities, these trends would mean higher O_3 exposures for the vast majority of the population.

R1-C12: *Lines 340f: These numbers do not agree with Fig. 5, 100, 50 and 0 percentiles. Typos or misunderstanding? Please correct or clarify.*

A12: The reviewer is right, and these were incorrect values. We much appreciate his/her attention to the details! The main issue was in the R code the authors used to calculate the averages. we mistakenly filtered the data using “==” instead of “%in%.” This led to discrepancies in the averaged results across different site types and percentiles, even when some values appeared similar. We had confused the two operators in the code.

The authors have updated the data in the text by recalculating the averages using the correct code. The corrected data can be found in lines 334 through 373, as shown below:

The traffic-influenced site shows ozone increases for all concentrations (0.00 - 1.46 µg m^{-3} $year^{-1}$) and the strongest trends at median concentrations around the 50^{th} percentile combined with nearly no increases at the very low and very high percentiles. This behaviour leaded to the depicted bell-shaped curves. The longer the considered time frame, the smaller is the dynamic range of the ozone trends from median against more extreme concentration regimes. During the recent decade, the traffic site has exhibited the most pronounced increases across the entire concentration range.

Urban and rural background sites

For the urban and rural background there are quite similarly increasing trends (0.00 - 0.77 µg m^{-3} $year^{-1}$) for nearly all concentration levels with the important exception that very high O_3 concentrations actually decrease (-0.55 - -0.00 µg m^{-3} $year^{-1}$) and the strongest increases were seen already at the 10^{th} and 25^{th} percentiles, respectively and not only at the 50^{th}. Each O_3 concentration level has also shown a greater increase over the past decade or a more pronounced decrease at the highest concentrations compared to the other two periods. Additionally, the range of differences between peak values in the, again, bell-shaped curves during the recent decade, is dampened against the traffic site and amounts to about 1 µg m^{-3} $year^{-1}$.

Mountain sites

For all the mountain sites a monotonous decrease in the O_3 is seen with increasing concentration percentiles and only for the smallest percentiles O_3 increasing trends are seen which switch to O_3 decreasing trends at the 5^{th}, 25^{th} and 50^{th} percentiles for the all years, 15 years and 10 years, respectively. There is no bell-shape anymore but a generally linear change.

Quantitatively, there are increases (0.00 - 0.66 µg m^{-3} $year^{-1}$) in percentiles from the minimum to the 10^{th} turning into decreases (-1.08 - -0.28 µg m^{-3} $year^{-1}$) in the higher percentiles (90^{th} - 100^{th}). The shorter the time frame considered, the more pronounced is the stated trend.

Overall, during recent 10 years, stagnant or even downward trends in the O_3 90^{th}, 95^{th}, 99^{th}, and maximum of O_3 in traffic, urban, rural and mountain stations had occurred, while at the lower percentiles (minimum, 1^{st}, 5^{th} and 10^{th}), O_3 concentrations across all stations seemingly continued to increase (with a mean trend ranging from 0.04 - 0.90 µg m^{-3} $year^{-1}$). In the range of medium percentiles (from 25^{th} to 75^{th}), all traffic, urban and rural sites showed statistically significant increasing trends (ranging from 0.40 to 1.46 µg m^{-3} $year^{-1}$), whilst at the mountain sites, they were decreasing or stagnant (-0.30 - 0.03 µg m^{-3} $year^{-1}$).

Quite recently, Finch and Palmer (2020) similarly reported no statistically discernible decreasing trend (-0.49 µg m^{-3} $year^{-1}$) in annual maximum O_3 on average, while mean concentrations and minimum concentrations increased with 0.41 and 0.09 µg m^{-3} $year^{-1}$, respectively, across rural, suburban and urban background sites in the UK over the period 1999 - 2019. These findings can, at best, be compared to panels A2 and A3 of Fig. 5 in illustrating O_3 trends in the similar period from 1997 - 2020, with -0.15 µg m^{-3} $year^{-1}$ in maximum O_3, 0.25 µg m^{-3} $year^{-1}$ in median concentration and 0.03 µg m^{-3} $year^{-1}$ at minimum concentration (as averaged values over panels A2 and A3 in Fig. 5). The results of another study in the UK (The Air Quality

Expert Group, 2021) also revealed that the 50th and 25th percentiles concentrations of O_3 have clearly increased (from 0 to 0.94 µg m^{-3} $year^{-1}$) at urban background sites over the period 2000 - 2019 (cf. panel A2 of Fig. 5 with 0.28 - 0.29 µg m^{-3} $year^{-1}$ in the similar period from 1997 - 2020), whilst corresponding increases (from 0 to 0.66) at rural sites (cf. panel A3 of Fig. 5 with 0.18- 0.24 µg m^{-3} $year^{-1}$) are in general smaller (and in many cases not statistically significant). In addition, there have been stagnant or significant downward trends (-2.05 to 0.64 µg m^{-3} $year^{-1}$, despite reported positive values are non-statistically significant) in the upper percentiles (99th and 99.9th percentiles) across all 27 sites examined, similar with our result of -0.16 - -0.07 µg m^{-3} $year^{-1}$ in 99th and 100th (as averaged values over panels A2 and A3 in Fig. 5).

R1-C13: *Line 362: Insert "of the latest period"*

A13: The phrase "of the latest period" has been added to lines 386-387 of the article. Now the sentence read:

For the traffic site, increasing trends are observed in all seasons over all time periods, but strongest in summer or spring of the latest period.

R1-C14: *Line 431: A table with emissions for this base case for Saxony would be useful here or earlier (line 141). Or refer at least to Figs. 11 and S9.*

A14: A summary table, Table S3, presenting the annual emission data in 2019 for the entire Saxony region in summer and winter simulation scenarios, has been added at line 27 of the Supporting Information. Given the large space needed to include this additional information, we now give the Table as a separate Excel file and include only the Table header in the Supporting information pdf file. See below:

Table S3. The annual emission data in 2019 for the entire Saxony region in summer and winter simulation scenarios. Each species is given by its SMILES string, and its IUPAC name generated using the PubChemPy package in Python (https://pubchempy.readthedocs.io/en/latest/), along with the corresponding chemical structure. It is noted that for some compounds, IUPAC names could not be retrieved from the PubChem database by the tool and they are therefore left blank.

See attached Excel file named *Table S3_The annual emission data in 2019 for the entire Saxony region.*

As a result of adding one new table, the numbering of subsequent tables in the Supporting Information (starting from Table S3) has been updated accordingly.

Additionally, the phrase of "(Table S3)" has been inserted in line 143 of the manuscript. The sentence now reads:

The summer and winter emission data (Table S3) was based on anthropogenic and biogenic emission inventories in 2019 from the German Environment Agency (UBA) for Germany and Thürkow et al. (2024), and derived for the whole Saxony area (50.9° N latitude and 14.3° E longitude) (see Fig. S1 showing NO_x emissions of the main roads and urban centres).

R1-C15: *Line 478: "of the base cases for Saxony (Sect 3.3.1)", right? If yes please insert.*

A15: Yes, right. The phrase of "of the base cases for Saxony (Sect 3.3.1)" has been inserted in line 517 of the article, as shown below:

These simulations of the base cases for Saxony (Sect 3.3.1) aimed to elucidate the O_3 production rate with regards to the ambient concentrations of NO_x and TNMVOC, which is depicted in the ozone isopleths of Fig. 10.

R1-C16: *Line 498: "for the station types"? Insert if yes, if no please explain already earlier in this subsection.*

A16: Yes, the phrase of "for the station types" has been inserted in line 557 of the article. The sentence, incorporating some other modifications in response to comments R2-C9, has been revised at line 556-557 of the manuscript. It now read:

For further clarification, in Tables S9 and S10, these TNMVOC estimates are shown together with a comparison of measured and modelled NO_x and dO_3/dt for the station types.

R1-C17: *Line 503: Mention the mean values also here, then table S11 is not needed.*

A17: Mean values have now been added in lines 560-563 of the article, and Table S11 in the Supporting Information has been deleted. The revised sentence in the article is shown below:

Indeed, unpublished data for a range of NMVOCs observed throughout the year 2022 in Borna, a city south of Leipzig, exhibited remarkably low concentrations of 66 NMVOCs species (Table S11) at a near-road measurement site, with hourly mean and maximum total mixing ratios of 3.6 ppb and 29.7 ppb in summer and 6.0 ppb and 204.6 ppb in winter, respectively.

Note that in the revised sentences above, that Table S11 corresponds to the original Table S10. Due to the addition of a new table (Table S3) showing the emission data (see R1-C14), the numbering of subsequent tables in the Supporting Information (starting from Table S3) has been updated accordingly.

Technical corrections

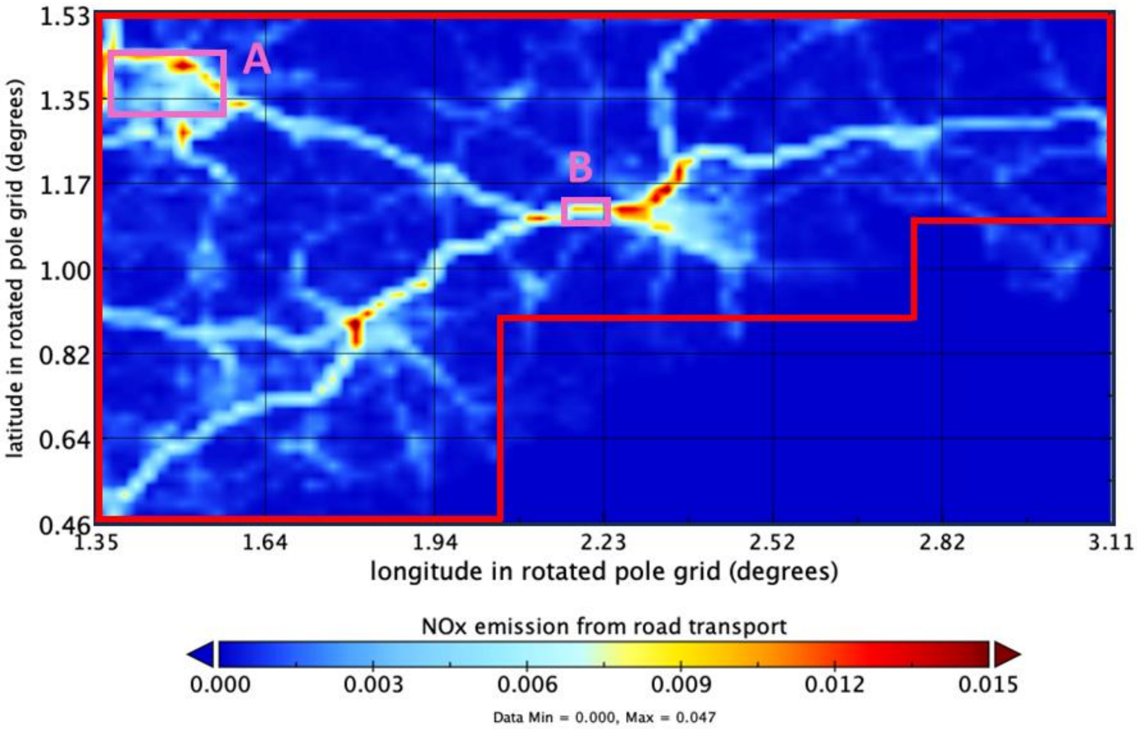
R1-C18: *Line 148: Better new paragraph.*

A18: Due to the added meteorology descriptions in lines 149-152 of the manuscript, the authors believe it would be better to start the new paragraph from "The initial SO_2..." instead of "CO and CH_4…".

So, a new paragraph starting with "The initial SO_2..." has already been added at line 153 of the article.

R1-C19: *Fig. 1: Include the frame of Fig. S1 for convenience.*

A19: A newly updated map with a frame that approximates the spatial extent of the state of Saxony (Fig. S1) has been added to line 18 of the Supporting Information. Additionally, some modifications have been made based on R1-C25. See the updated Fig. S1 below:

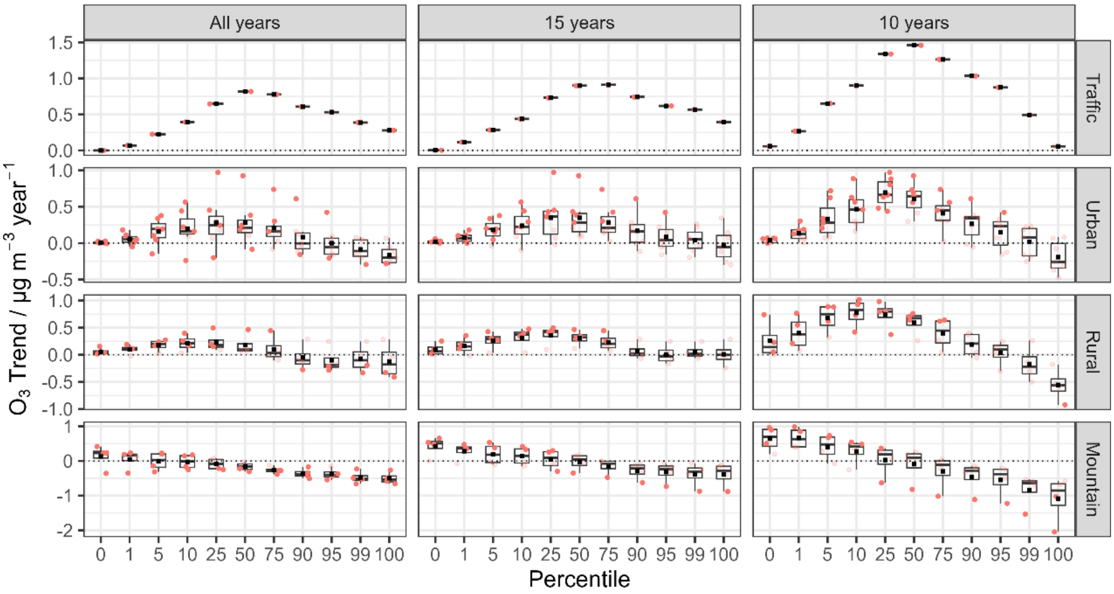


R1-C20: *Fig. 2: Include in caption after "1997" "or later".*

A20: The phrase "or later" has been added after "1997" has been added in line 223 of the article. Now the revised caption read: Figure 2: Boxplots of O_3 concentrations at individual monitoring stations, coloured according to their station type. The middle yellow point and the black horizontal line indicate the mean and median, respectively. The ends of the box indicate the lower and upper quartiles, the antennas the 1.5-fold interquartile range (IQR) and individual points are extreme values outside the IQR. Data per station from 1997 or later onward to 2020. The detailed station data are summarised in Table S1 in the Supporting Information.

R1-C21: *Fig. 5: Here colors superfluous.*

A21: Although the colors are somewhat excessive, the authors prefer the original figure. There is also a version that uses a single color to represent dots at different percentiles, but dots appear a bit crowded, making it difficult to distinguish between percentiles. See below:



R1-C22: *Lines 665, 673, 700: Please provide DOI or URL for technical reports.*

A22: URL links have been added in lines 730-731, 740, and 767 of the article. Now the references read:

Colette, A., Beauchamp, M., Malherbe, L., and Solberg, S.: Air quality trends in AIRBASE in the context of the LRTAP Convention, ETC/ACM Technical Paper, 4, 2015, https://www.eionet.europa.eu/etcs/etc-atni/products/etc-atni-reports/ETCACM_TP_2015_4_AQtrends, 2015b.

Corsmeier, U., Kalthoff, N., Vogel, B., Hammer, M.-U., Fiedler, F., Kottmeier, C., Volz-Thomas, A., Konrad, S., Glaser, K., and Neininger, B.: Ozone and PAN formation inside and outside of the Berlin plume—Process analysis and numerical process simulation, Tropospheric Chemistry: Results of the German Tropospheric Chemistry Programme, 289-321, https://doi.org/10.1007/978-94-010-0399-5_13, 2002.

EEA: European Union emission inventory report 1990-2021 — Under the UNECE Convention on Long-range Transboundary Air Pollution (Air Convention), Report 04/2023, https://data.europa.eu/doi/10.2800/68478, 2023.

R1-C23: *Lines 712, 717, 747, 789, 794: Same journal? If yes use the same abbreviation.*

A23: Yes. The journal name “Elementa: Science of the Anthropocene” has been corrected in lines 779 and 784 of the article. Now the references read:

Fleming, Z. L., Doherty, R. M., Von Schneidemesser, E., Malley, C. S., Cooper, O. R., Pinto, J. P., Colette, A., Xu, X., Simpson, D., and Schultz, M. G.: Tropospheric Ozone Assessment Report: Present-day ozone distribution and trends relevant to human health, Elementa: Science of the Anthropocene, 6, 12, 2018.

Gaudel, A., Cooper, O. R., Ancellet, G., Barret, B., Boynard, A., Burrows, J. P., Clerbaux, C., Coheur, P.-F., Cuesta, J., and Cuevas, E.: Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation, Elementa: Science of the Anthropocene, 6, 2018.

R1-C24: *Line 772: Final revised version available?*

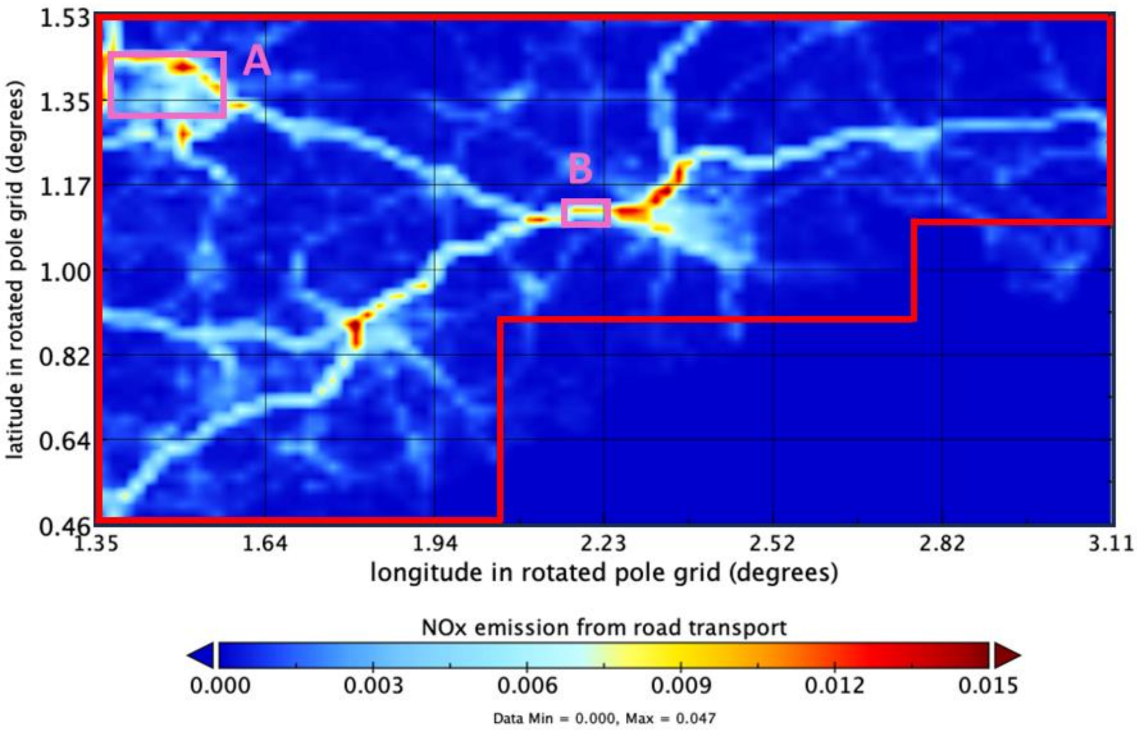
A24: Yes, the final revised version is available. The reference for the final revised version has been updated in lines 849-852 of the manuscript. The reference now reads:

Nussbaumer, C. M., Crowley, J. N., Schuladen, J., Williams, J., Hafermann, S., Reiffs, A., Axinte, R., Harder, H., Ernest, C., Novelli, A., Sala, K., Martinez, M., Mallik, C., Tomsche, L., Plass-Dülmer, C., Bohn, B., Lelieveld, J., and Fischer, H.: Measurement report: Photochemical production and loss rates of formaldehyde and ozone across Europe, Atmospheric Chemistry and Physics, 21, 18413-18432, 2021.

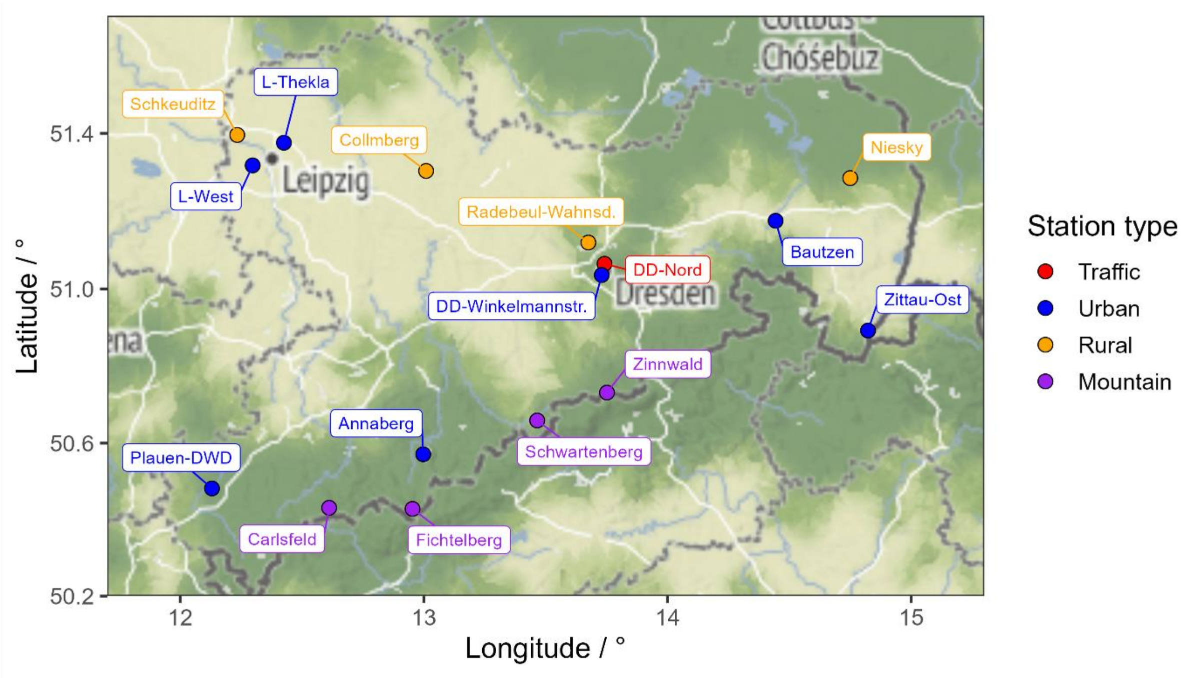
R1-C25: *Supplement: Fig. S1: Please define grid and its relation to the one in Fig. 1. It appears to be a subset of Fig. 1. Meaning or purpose of red rectangles not clear (is the sum of all 3 used in Fig. S9 which should be referred to?)? Mark the location of the extreme maximum which is almost one order of magnitude out of scale. Mark also the reference point mentioned in main text. Maybe a logarithmic color bar for the emissions would be useful.*

A25: In the original Fig. S1, three red rectangles in Fig. S1 approximated the entire Saxony area. However, it was three rectangular areas merely for technical reasons, as the averaged emission data could only be calculated for rectangles with the tool at hand.

To avoid confusion, an updated Fig. S1 is provided at line 18 of the Supporting Information, in which the approximate boundary of Saxony is marked only by using a continuous red frame instead of three separate rectangles. This red frame indicates the simulated entire Saxony area. See the updated Fig. S1 below.



To clarify the relationship between Fig. 1 and Fig. S1, the boundary of Saxony has also been outlined in the updated Fig. 1. The updated Fig. 1 has been added at line 110 of the manuscript, as shown below.



By comparing the updated Fig. 1 and Fig. S1, we can see the outlined Saxony region (red line) in Fig. S1 roughly corresponds to the shape of the Saxony boundary shown in Fig. 1.

The emissions data in each year, illustrated in Fig. 11 in the manuscript and Fig. S9 in the Supporting Information, are obtained by averaging the emissions values based on the red-outlined entire Saxony area in the updated Fig. S1.

Besides, the caption of Fig. 11 has been revised at line 644-646 of the manuscript, as shown below:

Figure 11: Anthropogenic emissions of NMVOCs in Saxony for five years from 2000-2019. Emission data were obtained by averaging values across the approximated area of the entire state of Saxony (see Fig. S1). Details of the emission categories and corresponding emission sectors can be found in Table S13.

The caption of Fig. S9 has been revised at line 138-140 of the Supporting Information, Due to the addition of two new figures (as Fig. S6 and S7, referring to R2-C9), the numbering of subsequent figures in the Supporting Information (starting from Fig. S6) has been updated accordingly. So the numbering of Fig. S9 should be Fig. S11. See the revised caption of Fig. S11 below.

Figure S11: Anthropogenic emissions of NO_x in Saxony for five years from 2000-2019. Emission data were obtained by averaging values across the approximated area of the entire state of Saxony (see Fig. S1). Details of the emission categories and corresponding emission sectors can be found in Table S13.

About the reviewer's comment on the maximum and minimum values in the original Fig. S1, now the authors make the following response to explain.

The maximum and minimum values initially reflected extreme high and low values in whole Germany in the original Fig. S1. The extreme low and high values were located in other regions of Germany rather than Saxony. In the updated Fig. S1, the authors have adjusted the color pie chart to show only the data range for Saxony in a non-logarithmic scale. It is clear from the updated Fig. S1 where the location of the maximum value (~0.015 kg m^{-2}) for the state of saxony is located.

In the updated Fig. S1, except for the red-outlined entire Saxony area, two specific rectangles in rose pink, A and B, represent the representative urban area (city of Leipzig) and traffic-dominated area (highway close to Dresden) used for the simulation, respectively. Now the caption of Fig. S1 has been revised in the Supporting Information. See below:

Figure S1: The NO_x emission (in kg m^{-2}) from road transport in the entire Saxony state region in 2019. A continuous red frame indicates the entire simulated Saxony area. Two specific rectangles in rose pink, A and B, represent the representative urban area (city of Leipzig) (0.13° × 0.20°) and traffic-dominated area (highway close to Dresden) (0.07° × 0.10°) used for the simulation, respectively.

One sentence has been revised in lines 143-146 of the manuscript, now it reads:

The summer and winter emission data (Table S3) was based on anthropogenic and biogenic emission inventories in 2019 from the German Environment Agency (UBA) for Germany and Thürkow et al. (2024), and derived for the whole Saxony area (50.9° N latitude and 14.3° E longitude) (see Fig. S1 showing NO_x emissions of the main roads and urban centres).

R1-C26: *Table S3: "Dry deposition", right? It might be useful to include conversion factors to ppbv for the species not listed with this unit.*

A26: Yes, the reviewer is right. "Deposition rate" has now been revised to "Dry deposition velocity".

The concentration unit in the updated table is uniformly set to ppb.

Additionally, a typo has been corrected, "HSO_4" is now "H_2SO_4". The concentrations of SO_2 and CH_4 were incorrect due to an error in the conversion from molec cm^{-3} to μg m^{-3}, caused by an incorrect conversion factor, which has now been corrected. The concentrations used in the modelling, expressed in molec cm^{-3}, are provided in Table S5 (former Table S4) for reference. The concentration of SO_2 should be 3 μg m^{-3} (1.1 ppb) in summer and 5 μg m^{-3} (1.9 ppb) in winter, instead of the previously written 54 μg m^{-3} in summer and 3 μg m^{-3} in summer. The concentration of CH_4 in summer and winter should be 2018.6 μg m^{-3} (1700 ppb) instead of 1155.8 μg m^{-3}. The originally cited reference Schaefer (2019) has been replaced with Herrmann et al. (2000).

The definition of ratio of solar radiation has been revised for clarity and accuracy. As a result of adding one new table (Table S3) showing the emission data (see R1-C14), the numbering of original Table S3 now is revised to Table S4. Now Table S4 shows as:

Model setting	**Unit**	**Summer**	**Winter**	**Reference**
Initial time		00:00 CET, 14 July, 2019	00:00CET, 14 January, 2019	
Meteorological conditions				
Temperature	°C	15	4	Measured
Pressure	hPa	1000	1000	Measured
Relative humidity	%	70	70	Measured
Ratio of Solar radiation*		0.7	0.4	Measured
Dry deposition velocity				
NO_2	cm s^{-1}	0.3	3	Rondón et al. (1993)
N_2O_5	cm s^{-1}	100	2	Hoffmann et al. (2019)
O_3	cm s^{-1}	0.8	0.08	Clifton et al. (2020)
NO	cm s^{-1}	0.05	0.05	Zhu et al. (2020)
HNO_3	cm s^{-1}	3.5	3.5	Zhu et al. (2020)

H_2O_2	cm s^{-1}	1	1	Zhu et al. (2020)
CO	cm s^{-1}	0.1	0.1	Zhu et al. (2020)
HCl	cm s^{-1}	1	1	Zhu et al. (2020)
NH_3	cm s^{-1}	1	1	Zhu et al. (2020)
SO_2	cm s^{-1}	1	1	Zhu et al. (2020)
H_2SO_4	cm s^{-1}	2	2	Zhu et al. (2020)
HCHO	cm s^{-1}	1	1	Zhu et al. (2020)
CH_3OH	cm s^{-1}	1	1	Zhu et al. (2020)
CH_3CH_2OH	cm s^{-1}	0.5	0.5	Zhu et al. (2020)
PANs	cm s^{-1}	0.7	0.7	Wu et al. (2012)
CHClO	cm s^{-1}	0.2	0.2	Hoffmann et al. (2019)
CHBrO	cm s^{-1}	0.2	0.2	Hoffmann et al. (2019)
Boundary layer heights (BLHs)				
Daytime BLHs	m	500	2000	2
Nighttime BLHs	m	250	1000	
Measured Chemical data				
O_3	ppb	31.7	21.6	Measured
NO_2	ppb	3.4	7.0	Measured
SO_2	ppb	1.1	1.9	UBA website
CO	ppb	153.4	153.4	Zellweger et al. (2009)
CH_4	ppb	1700	1700	Herrmann et al. (2000)
HONO	ppb	0.5	0.5	Stieger et al. (2018)
PAN	ppb	0.5	0.5	Pandey Deolal et al. (2014)

* Ratio of solar radiation, defined as the mean value between 10:00 and 13:00 divided by the maximum clear sky radiation value during the same period.

In the lines 153-155 of the manuscript, the sentences have been revised. Now they read:

The initial SO_2 concentrations in summer and winter were obtained from UBA (https://www.umweltbundesamt.de/daten). CO and CH_4 were set with 153.4 and 1700 ppb, respectively, referred the measured data from Zellweger et al. (2009) and Herrmann et al. (2000).

R1-C27: *Table S4: Your compound strings are often inconsistent with the MCM-notation and could not be found in the CAPRAM-link. Instead of listing them twice it would be useful to replace one column by a column with the full chemical notation or the compound names as in Table S10. Also some compound strings change from summer to winter, typos?*

A27: Yes, some SMILES strings are available on the MCM website, while others are not. Additionally, there is no publicly accessible webpage in the CAPRAM link for referencing these SMILES strings - they are only accessible to CAPRAM users.

Instead of listing the SMILSE strings twice, one column with the full SMILSE strings, one column with their IUPAC names and one column with their chemical structures are presented in the updated table.

The following table shows the difference between summer and winter species in the last 3 or 4 rows of the original Table S4. There are typos. The first three SMILES strings (shaded in gray) under the summer category should be removed in the updated table, as they are followed by four species listed for the winter.

Compound string	**Unit**	**Compound string**	**Unit**
Summer	**molec cm^{-3}**	**Winter**	**molec cm^{-3}**
CC(=O)C(OO)C(O)(C)C	2.31E+00	CC(=O)CC(O)(C)CO	7.43E+03
CC(=O)CC(O)(C)COO	1.08E+00	CC(=O)CC(O)(C)C=O	8.51E+02
CC(=O)CC(O)(C)CON(=O)=O	4.82E+00	CC(O)(C)CC(=O)COO	7.98E+05
CC(=O)CC(O)(C)CO	9.24E+03	CC(O)(C)CC(=O)CO	3.28E+04

As a result of adding one new table (Table S3) showing the emission data (see R1-C14), the numbering of original Table S4 now is revised to Table S5. The revised caption of Table S5 has been updated in lines 38-43 of the Supporting Information and now reads:

Table S5. Dominant initial gas-phase concentrations applied in the final 24-hour simulations for summer and winter scenarios. Each species is given by its SMILES string, and its IUPAC name generated using the PubChemPy package in Python (https://pubchempy.readthedocs.io/en/latest/), along with the corresponding chemical structure. It is noted that for some compounds, IUPAC names could not be retrieved from the PubChem database by the tool and they are therefore left blank.

See attached Excel file named *Table S5_Dominant initial gas-phase concentrations.*

R1-C28: *Table S5: Add in caption or text, line 176, "i.e. each combination is considered."*

A28: The original Table S5 may have caused some misunderstanding due to a lack of clarity regarding the changing emission factor for summer and winter simulations. To improve clarity, the authors have revised both the table and its caption.

The phrase "considering each combination" has been added to the caption of Table S6 (former S5) at lines 46-48 in the Supporting Information. Table S6 now reads:

Table S6. Changing the emission multiplier for NO_x and TNMVOC in the simulations. Three batches were conducted, considering each combination, with each batch comprising 400 model runs. The total number of simulations is 800 for summer and 1200 for winter.

Batch 1 (Summer/Winter)			**Batch 2 (Summer/Winter)**			**Batch 3 (Winter)**		
Number	**Emission factor**		**Number**	**Emission factor**		**Number**	**Emission factor**	
	NO_x	**TNMVOC**		**NO_x**	**TNMVOC**		**NO_x**	**TNMVOC**

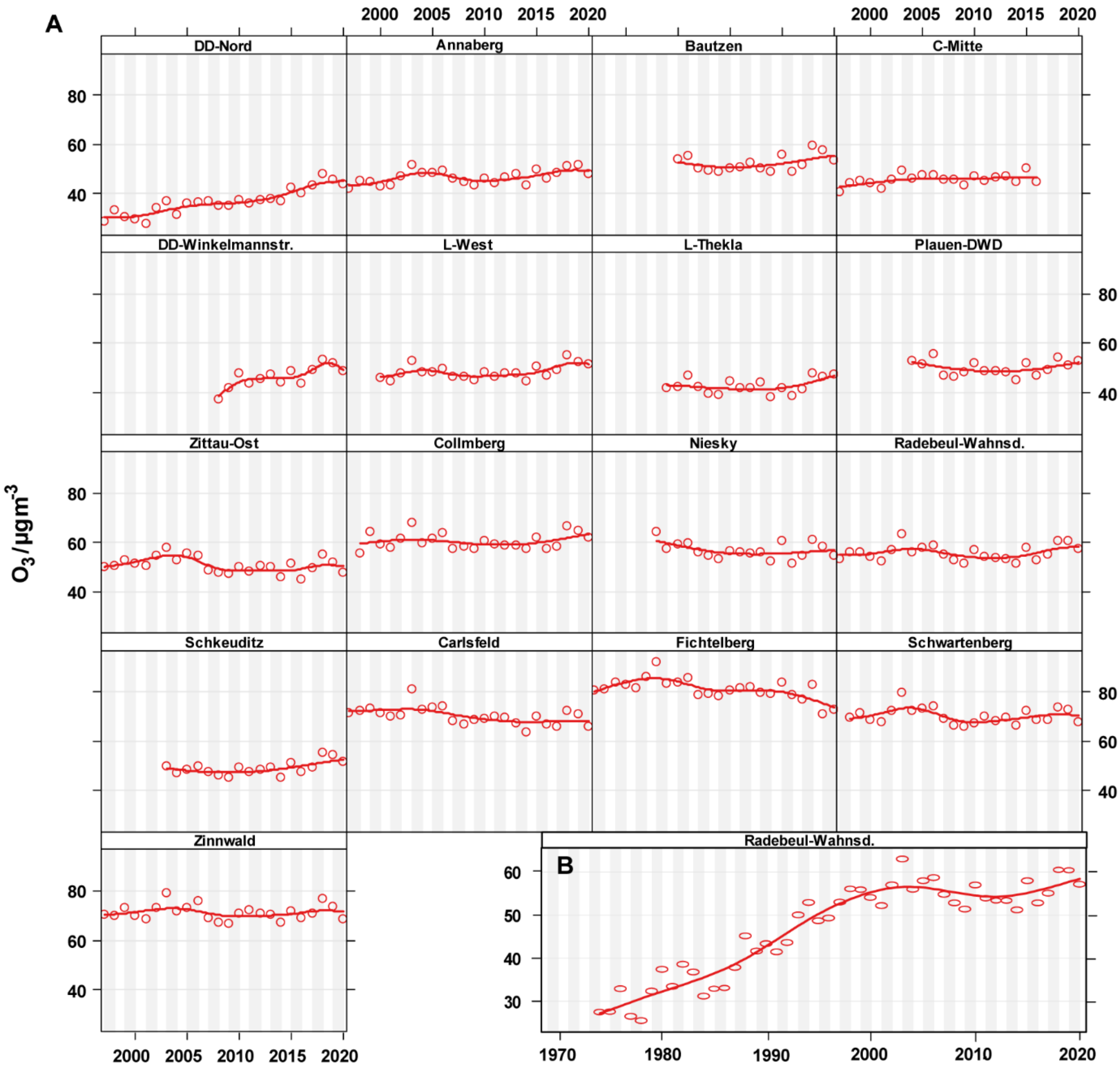
1	0.001	0.001	**1**	1.5	1.5	**1**	1.5	13
2	0.005	0.005	**2**	2	2	**2**	2	14
3	0.01	0.01	**3**	2.5	2.5	**3**	2.5	16
4	0.05	0.05	**4**	3	3	**4**	3	17
5	0.1	0.1	**5**	3.5	3.5	**5**	3.5	18
6	0.5	0.5	**6**	4	4	**6**	4	19
7	1	1	**7**	4.5	4.5	**7**	4.5	21
8	5	5	**8**	5.5	5.5	**8**	5.5	22
9	10	10	**9**	6	6	**9**	6	23
10	15	15	**10**	6.5	6.5	**10**	6.5	24
11	20	20	**11**	7	7	**11**	7	26
12	25	25	**12**	7.5	7.5	**12**	7.5	27
13	30	30	**13**	8	8	**13**	8	28
14	35	35	**14**	8.5	8.5	**14**	8.5	29
15	40	40	**15**	9	9	**15**	9	30
16	45	45	**16**	9.5	9.5	**16**	9.5	31
17	50	50	**17**	10.5	10.5	**17**	10.5	32
18	60	60	**18**	11	11	**18**	11	33
19	70	70	**19**	11.5	11.5	**19**	11.5	34
20	80	80	**20**	12	12	**20**	12	35
Total simulation number	**400**		**400**			**400**		

One sentence has been revised in lines of 186-189 in the manuscript, now it reads:

The sensitivity simulations were done by scaling the base case emissions of TNMVOC and NO_x 20 times in each of three batch runs, i.e. each combination is considered (Table S6). Three batches were performed to achieve a sensible range of resulting TNMVOC and NO_x concentrations in the total of 800 and 1200 model runs for summer and winter, respectively.

R1-C29: *Fig. S2: There is enough space to keep the time axis in the last frame on scale.*

A29: The time axis in the last frame has been adjusted in the updated Fig. S2. See below:



R1-C30: *Fig. S9 to S13 and 11: It is often difficult to attribute the legends with source categories to the color bars in the figures since the order differs from figure to figure. Please stay to the order in the legends in every figure. Captions too short or misleading (e.g. traffic dominated by solvents in Fig. S10).*

A30: To enhance readability, the legends (in Fig. 11 in the manuscript and Figs. S9-S13 in the Supporting Information) have been arranged in descending order of emissions percentage.

Since each emission category includes lots of emission sectors, it is impractical to write all the emission sectors clearly in the caption of the figures. Here a new table (as Table S13) has been added at line 183 of the Supporting Information to show the emission categories and emission sectors in detail. Now the caption of Table S13 reads:

Table S13: Emission categories and corresponding emission sectors (Schneider et al., 2016).

See attached Excel file named *Table S13_Emission categories and corresponding emission sectors.*

Here is an explanation regarding the reviewer's comment on the misleading aspects of Fig. S10. Although the selected traffic area (indicated by the rectangular B in Fig. S1) is close to the roadside, if you zoom in the B area, you will see that there are also residents along roadside, and the solvents emitted are likely associated with domestic solvent use.

Anyway, the revised figure captions and figures are shown below. It is noted that, due to the addition of two new figures (as Fig. S6 and S7, referring to R2-C9), the numbering of subsequent figures in the Supporting Information (starting from Fig. S6) has been updated accordingly. As a result, the numbering of Fig. S9 has been changed to Fig. S11, and the numbering of all following figures has been adjusted accordingly.

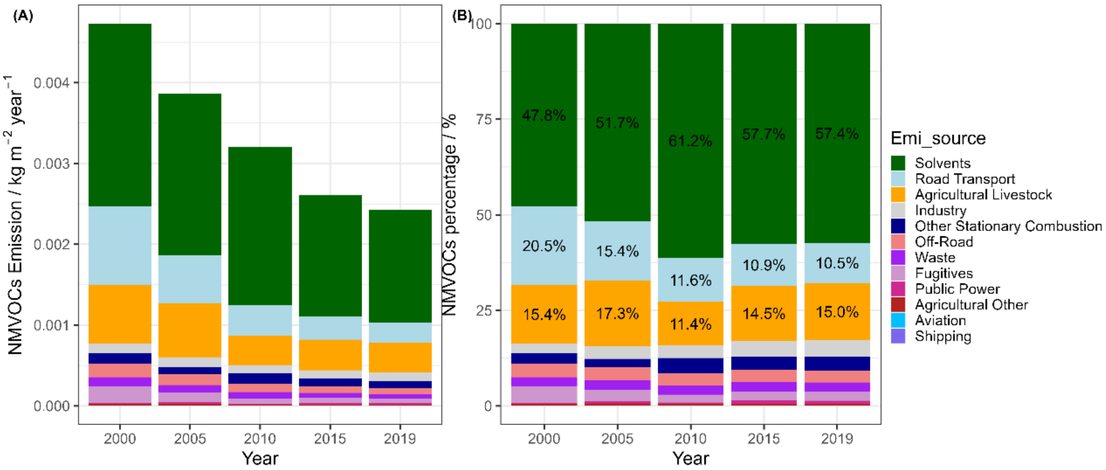


Figure 11: Anthropogenic emissions of NMVOCs in Saxony for five years from 2000-2019. Emission data were obtained by averaging values across the approximated area of the entire state of Saxony (see Fig. S1). Details of the emission categories and corresponding emission sectors can be found in Table S13.

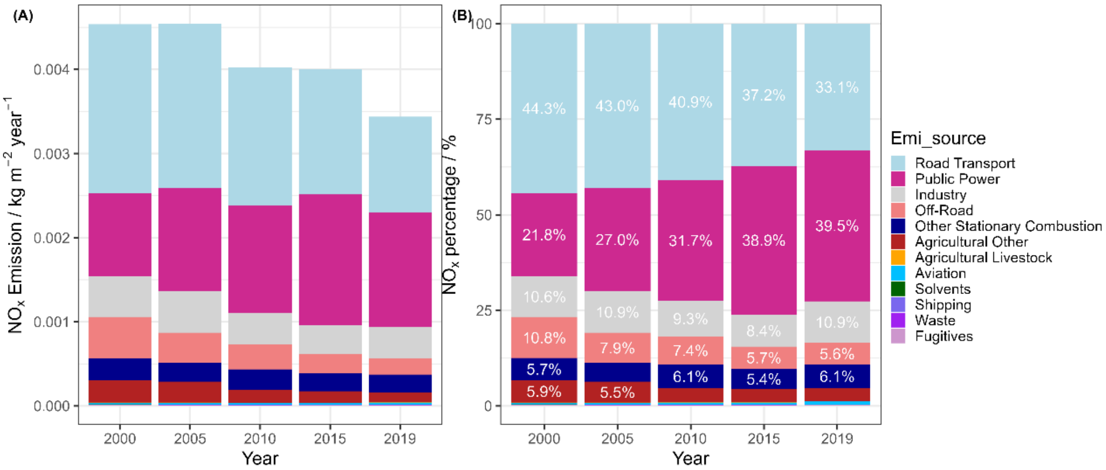


Figure S11: Anthropogenic emissions of NO_x in Saxony for five years from 2000-2019. Emission data were obtained by averaging values across the approximated area of the entire state of Saxony (see Fig. S1). Details of the emission categories and corresponding emission sectors can be found in Table S13.

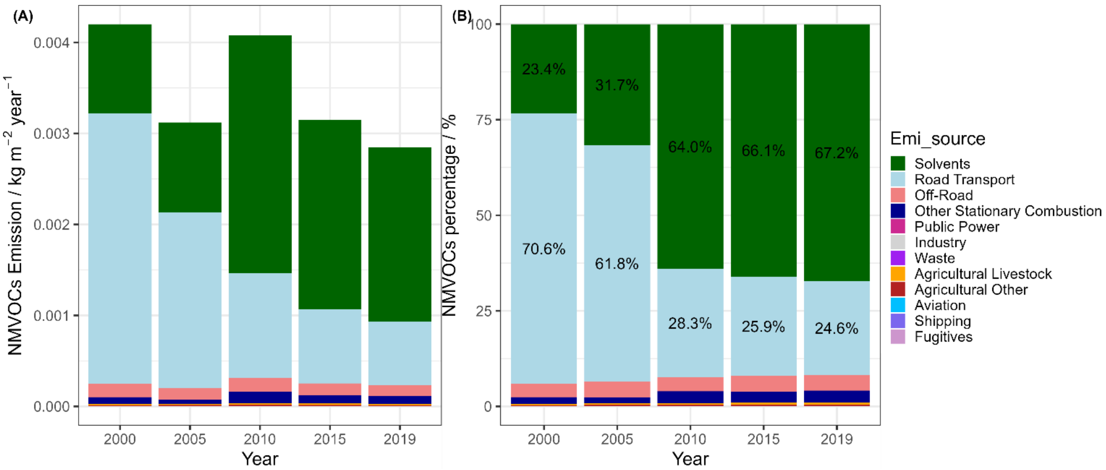


Figure S12: Anthropogenic emissions of NMVOCs in traffic area for five years from 2000-2019. Emission data were obtained by averaging values from the selected traffic-dominated area (see Fig. S1). Details of the emission categories and corresponding emission sectors can be found in Table S13.

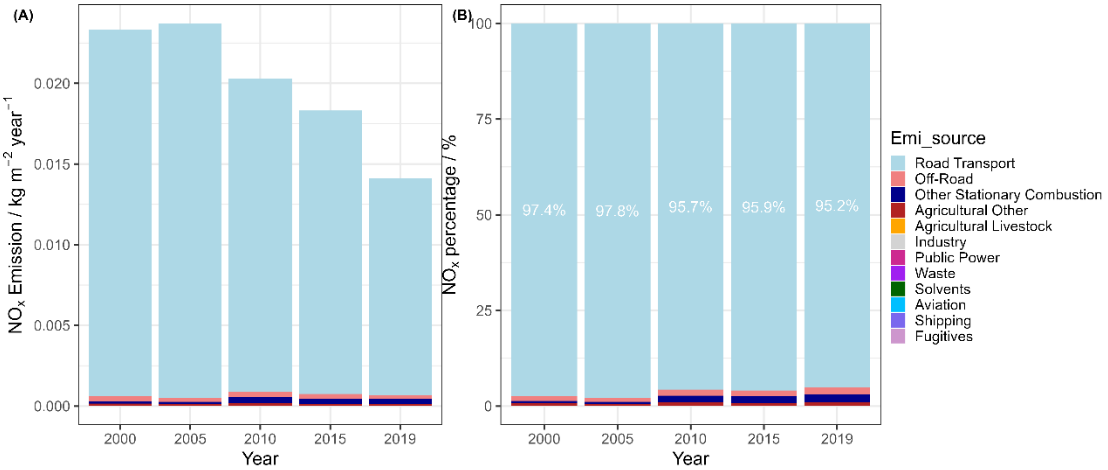


Figure S13: Anthropogenic emission of NO_x in traffic area for five years from 2000-2019. Emission data were obtained by averaging values from the selected traffic-dominated area (see Fig. S1). Details of the emission categories and corresponding emission sectors can be found in Table S13.

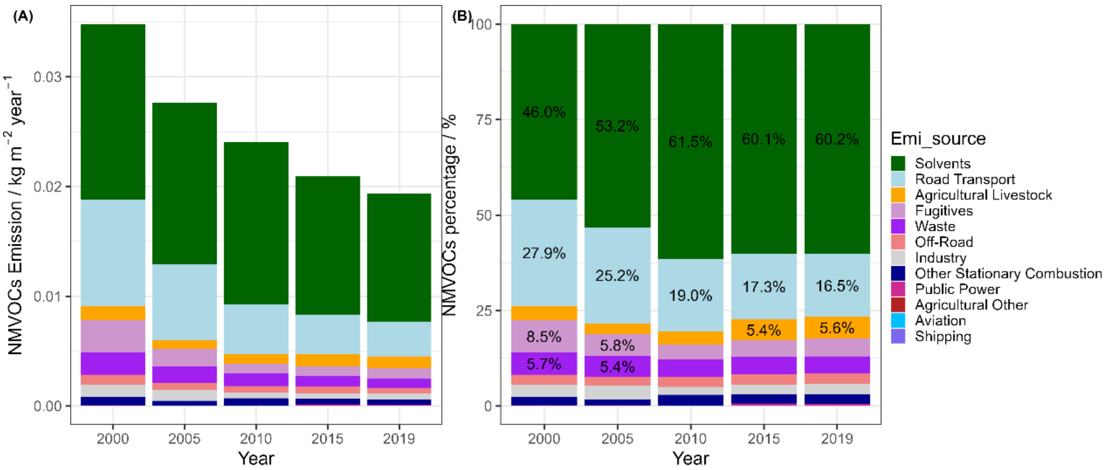


Figure S14: Anthropogenic emission of NMVOCs in urban area for five years from 2000-2019. Emission data were obtained by averaging values from the selected urban area (city of Leipzig) (see Fig. S1). Details of the emission categories and corresponding emission sectors can be found in Table S13.

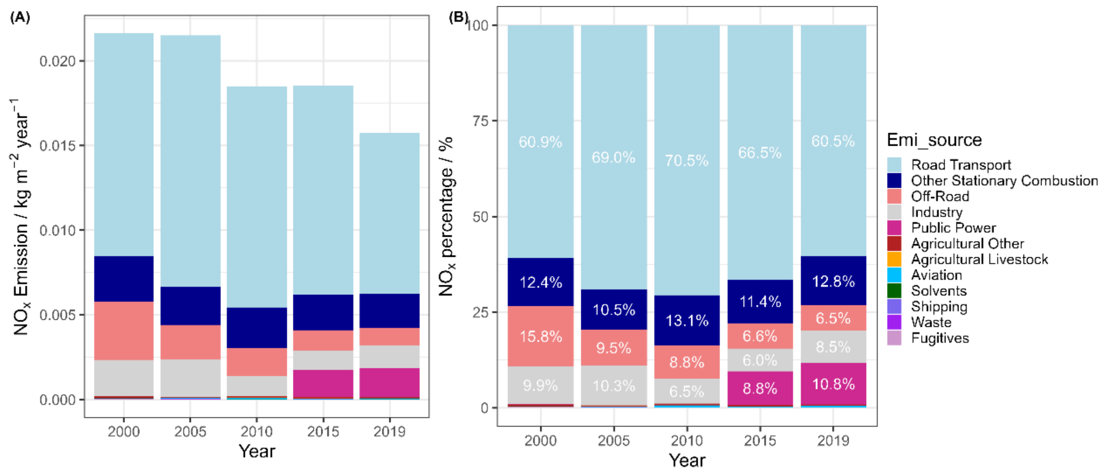


Figure S15: Anthropogenic emission NO_x in urban area for five years from 2000-2019. Emission data were obtained by averaging values from the selected urban area (city of Leipzig) (see Fig. S1). Details of the emission categories and corresponding emission sectors can be found in Table S13.

R1-C31: *Fig. S13 and S14: Better convert to a table.*

A31: The authors assume the reviewer are referring to Figs. S14 and S15. A new table (Table S12) summarizing the data from Figs. S14 and S15 has been added at line 181 of the Supporting Information. See below:

Table S12. A summary of the emission data from Figs. S16 (former Fig. S14) and S17 (former Fig. S15).

Emission species	**Emission source**	**Saxony**	**Leipzig-urban**
		Emission / kg m^{-2} $year^{-1}$	
Alpha-pinene	Biogenic emissions	0.00098	0.00008
Isoprene	Biogenic emissions	0.00041	0.00005
Limonene	Anthropogenic and biogenic emissions	0.00073	0.00528

Additionally, the captions of Figs. S16 (former Fig. S14) and S17 (former Fig. S15) have been revised accordingly at lines 168-172 and 175-179 of the Supporting Information. They now read:

Figure S16: Biogenic emissions of isoprene (ISO) and alpha-pinene (API) in Saxony for the year 2019, along with anthropogenic and biogenic emission data for limonene (LIM) in the same year. Emission data were obtained by averaging values across the approximated area of the entire state of Saxony (see Fig. S1). The emission data source is from the German Environment Agency (UBA) for Germany and Thürkow et al. (2024). Detailed data can be referred to Table S12.

Figure S17: Biogenic emissions of isoprene (ISO) and alpha-pinene (API) in urban area for the year 2019, along with anthropogenic and biogenic emission data for limonene (LIM) in the same year. Emission data were obtained by averaging values from the selected urban area (city of Leipzig) (see Fig. S1). The emission data source is from the German Environment Agency (UBA) for Germany and Thürkow et al. (2024). Detailed data can be referred to Table S12.

And the phrase "and Table S12" has been added to lines 623-626 of the manuscript. The revised sentences now read:

Although the biogenic emissions data of year 2019 in overall Saxony (Fig. S16 and Table S12) are comparable to anthropogenic NMVOCs in same year (Fig. 11A), biogenic emissions of isoprene and alpha-pinene in selected urban stations (Fig. S17 and Table S12) are indeed several orders of magnitude smaller than the anthropogenic emissions data in these areas (Fig. S14A).

R1-C32: *Please no page break directly after the table head, and no line break in a number.*

A32: OK, in the manuscript (black version) and Supporting Information (black version), there is no page break directly after the table header, and numbers are kept intact without line breaks. Please check.

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