

Ozone trends and their sensitivity in global megacities under the warming climate

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The topic and results are of interest. Overall, the paper is timely, well-written, robust, and reports informative results. I suggest revising your paper, in particular by: correcting the English/writing throughout the text (some typos), developing your introduction further by highlighting the significance and novelty of your work, and updating the state-of-the-art.

Abstract - The current version is not informative, i.e., missing quantified results (trend magnitude), missing information about the applied approach (e.g., for trend analysis), etc.

Introduction

Line 30: ref - In 2016, 54% of the world's population lived in urban areas (UN, 2018) where ground-level ozone, PM2.5, and NO2 are the most harmful air pollutants for human health.

Line 39: not only – ozone exposure leads to premature deaths & hospital admissions for cardiovascular and respiratory diseases (e.g., 10.1016/j.atmosenv.2023.119926).

In 2019, the chronic ozone exposure led to 147,100 deaths in 13,160 urban areas with more than 50,000 inhabitants in cities in 2019 worldwide (Malashock et al., 2022).

Line 40: Ozone is a threat to biodiversity (10.1126/sciadv.abc1176; 10.1007/s11676-023-01645-y).

Global exposure of urban population to ozone increased, due to lower O3 titration by NO. The summertime O3 MDA8 rose at 74% of cities worldwide (10.1016/j.scitotenv.2022.160064).

Ozone is rising also by night (10.1126/science.adm7628).

Line 45: in general, ozone levels are rising in cities and remote areas, and declining in rural areas (10.1016/j.coesh.2020.100226)

Line 76-77: add a reference

Results

Table 1: be careful with the number of digits. I suggest adding the trend magnitude for ozone, or from literature, for instance the TOAR database (<https://join.fz-juelich.de>) or the Global Reanalysis Datasets for all cities worldwide at a fine spatial resolution from 2000 to 2019.

Discussion

In addition, with the future vehicle electrification, fostered by the ban of thermal engines, is expected to have an important

effect on urban air quality, with improvements in NO₂ pollution and increases in ground-level ozone concentrations (10.1016/j.envres.2023.115835).

Line 324-325 & Line 401-402 (implication) - Hence, low VOCs emitting tree species should be selected for greening and re-naturing programs (10.1126/science.add973).

Reviewer #2

(Remarks to the Author)

This manuscript analyzed the long-term trends of ozone sensitivity using a satellite dataset. Taking into account the journal's high-quality standard, this study should be rejected. I found the following three flaws in this study. First of all, what's new in this study? The methodologies and approaches themselves have been well-used in so many other studies. Even though the target areas extended into the tropics and/or global south, such analyses could be found in other literature. The simple application to other areas would not meet the journal's scope and criteria. Second, although the manuscript (and title itself) emphasized the relation between ozone to warming climate, the analysis could be misleading. For example, as stated "according to NASA, 2019 was the second warmest year since records", Fig. 6 used the difference in the year 2019 and just imposed the high-temperature biases. However, temperature can be varied year-to-year and the fact will not be so simple. In addition, the ozone regime was diagnosed based on the column data whereas temperature and SWR are surface datasets. I do not fully understand such confusing data analyses. Finally, the satellite data of HCHO and NO₂ column can be a proxy of emissions, but the bottom-up emission inventories need to be analyzed, and discussed the relation between column concentration and emission if the authors suggest the policy regulation.

Reviewer #3

(Remarks to the Author)

This manuscript evaluates the trends of OMI HCHO and NO₂ in 41 megacities worldwide, explores ozone sensitivity through their ratios, and analyses their correlation with temperature. The authors provide a global perspective on the study of ozone and precursor changes. However, the current manuscript leaves something to be desired. I suggest that the manuscript be further refined before it is considered for publication.

1.Ozone sensitivity: The authors do not explicitly provide ozone sensitivity, let alone "ozone sensitivity trends", but only FNR values and qualitative analyses, which is insufficient for a study on the topic of Ozone sensitivity.

2.Ozone data: It is recommended that authors use ground-level ozone data to analyze trends, even for just a few stations or cities. This will allow for specific analyses rather than just speculation when exploring the influence of precursors or meteorological factors on ozone changes, and will be more persuasive for the conclusion than just that "the observed trends are contrary to what is required to reduce O₃ levels".

3.Due to the long time scales of the study, there may be exceptions to the non-significant trends in ozone precursors, such as the rapid decline in NO₂ in Beijing from 2013, as shown in Figure 5, which is the case for many cities in China. This change in trend helps to study the role of precursor changes under ozone sensitivity.

4.Increase in temperature: The authors attempt to explore the effect of "temperature change" on ozone concentrations, but do not give quantitative results for temperature change. Figure 6 only shows the situation in 2019. In addition, the term "warming climate" is not appropriate for this scale of temperature change.

5.Clustering analysis: The authors classify the cities into four clusters according to FNR values, but the significance of this needs further consideration. In fact, they can be regarded as different ozone generation sensitivity classified by FNR. As the authors state, emission trends differed among the grouped megacities, so what is the usefulness of cluster analysis? Authors explain the different roles of meteorological factors in terms of whether they are developed regions or not, which in itself has no relevance to the results of the clustering analysis.

6.Figures and Table: The time scale and units of the data should be given in Table 1. The meaning of the colours should be indicated in Figure 3. How was the trend calculated in Figure 5?

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors replied to open issues satisfactorily. The requested changes and suggestions were taken into account, and any missing information added. The revised version is improved, and ready for publication.

Reviewer #2

(Remarks to the Author)

This manuscript is the revised version that was previously submitted to this journal. I sincerely appreciate the authors' revisions to address my comments. After the clarification from the authors, I reread this carefully, but I still have concerns about considering it as a possible publication from this journal's high-quality standard. My biggest concerns were the lack of appropriate references and relevant discussions. Especially, the added discussion using O₃ reanalysis data has been severely lacking in discussion with previous research and strongly relates to the analysis using their data.

Major points:

The visualization and the relevant discussion will still be difficult to understand. In Fig. 2., HCHO and NO₂ columns were shown from 1.0E15 to 17.0E15 mol.cm⁻², so I guess that these white colors will indicate lower concentration (less than 1.0E15), right? Thus, the white color in the FNR map corresponds to a higher value rather than 6, right? However, I am confused "When mapping the data, masks for land-ocean, HCHO values below 7×10¹⁵ mol cm⁻², NO₂ values below 1×10¹⁵ mol cm⁻², and FNR values above 10 were applied to facilitate data visualisation." in line 501-503. Could you clarify this analysis methodology? It is very interesting to see the behavior of HCHO, NO₂, and FNR in Africa as categorized into cluster no. 1. However, based on the analysis shown in Fig. 2, NO₂ column corresponded to a relatively lower region. In this sense, how could we consider the detection limit of NO₂ column? The usage of HCHO was described in line 486, but it is also required for the detailed description of NO₂. I am afraid that a high FNR has simply drawn from a low NO₂ which was just lower than the detection limit. Without clarification, the analysis would be meaningless.

The added analysis for O₃ based on the reanalysis data seems to be partly critical; however, I do not speculate the methodology of the re-gridding from a coarser resolution to a finer resolution. Although it could be easy in the re-gridding from a finer resolution to a coarser resolution as HCHO column, it seems to be difficult how to regrid into a finer resolution. Did this apply some a priori or other information? Related to this O₃ analysis, the authors claimed that the transition from the decreasing trend to the increasing trend in China. However, this result and discussion on China in lines 195-210 will be conflict with the continuously increasing O₃ trend reported in previous papers (<https://doi.org/10.5194/acp-16-10637-2016>; <https://doi.org/10.5194/acp-19-1455-2019>; <https://doi.org/10.1016/j.atmosenv.2021.118869>). The discussion was very weak in the current manuscript quality just relying on their data without referring to appropriate pieces of literature. Judging from this careless analysis without the previous studies, I am sorry, but the results and discussion on other sites will also be suspicious.

Specific points:

Line 72-78: I am confusing this explanation. Despite these discussions for FNR values, why did the authors use the method of Duncan et al. (2010)? To avoid confusion, I will recommend moving or repeating the sentence in lines 495-497. I agree with focusing on the trend analysis instead of the FNR value because the threshold values are still discussed. However, the authors compared FNR at site-to-site and discussed the ozone formation regime. If there is a difference in the threshold value site-by-site, it is not as simple as discussed.

Line 162-166: It should be carefully defined which countries are developed/developing countries. Specifically, which is categorized for China?

Line 391-392: We cannot follow NO_x emission itself from this manuscript. Need reference. Even though the measured column can be a proxy of emissions, it could not be defined as emissions.

Line 392-393: See my major concern.

Line 396-398: This sentence seems to be redundant.

Line 433: Related to the comment on lines 162-166, what was in China? Some literatures also reported the transition of ozone sensitive regime in China, but they noted that the shift from VOC-limited to a transitional or NO_x-limited regime (<https://doi.org/10.5194/acp-21-7253-2021>; <https://doi.org/10.1002/2017JD026720>)

Line 453: I am still wondering if this remark is overstated because the emissions themselves have never been analyzed.

Reviewer #3

(Remarks to the Author)

The authors have revised the manuscript extensively to emphasize its importance and novelty. I recommend further refinement of the manuscript before considering it for publication.

1. Based on the FNR metric it is possible to clearly indicate which areas are VOC-sensitive or NO_x-sensitive and demonstrate the changes visually. What considerations did the author fail to provide this information?

2. With regard to the effects of temperature and radiation, the authors focus mainly on the increase in biological VOC emissions and seem to neglect their contribution to the chemical reactions that lead to ozone production.

3. We already know that an increase in VOC or a decrease in NO_x alone increases ozone levels in VOC-sensitive areas, and that ozone precursor abatement measures depend on the O₃ sensitivity. The authors build on this by conducting a clustering analysis to categorize sites into four clusters. What is the significance of this? Any new insights into the relationship between ozone changes and precursors? Further clarification is needed.

4. The abstract is too succinct in its presentation of the results, simply stating "The O₃ trends ... aligning with changes in precursor emissions", which does not emphasize the novelty of this study at all.

5. The colors in Figure 3 make it difficult to distinguish between positive and negative values. In addition, the marine portion can be deducted as shown in Figure 2.

6. The time frame is not indicated in table 1 and related text.

7. Line 223-234: The sequence numbers of the clusters do not match those in the previous text and figure.

Version 2:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

This manuscript is the second revision that has been submitted to this journal. I greatly appreciate the authors' revisions to address concerns; however, I do not fully understand their response to the following Q&A.

- Regarding the color scales for NO₂, HCHO, and FNR, I can still see the white areas even in Supplementary Fig. 9 over South America. This region will correspond to Amazonia Rainforest (AMZ), one of the significant regions classified as cluster 1. Unfortunately, I cannot fully follow the discussion regarding the non-masked version.

- Even in the possible spatial difference in the threshold value of FNR, I did not catch up with the added line in Line 149-154 "Due to the inherent spatial and temporal variability of the FNR values that define ozone sensitivity regimes, the results did not aim to provide a specific threshold value for regime classification at every site, however they emphasised the observed trends across regions. Sites displaying the lowest FNR were considered as VOC-limited conditions, intermediate as transitional/NO_x-limited, and the highest FNR as NO_x-limited regions.". Under the possible difference in the threshold value of FNR as previous studies suggested, why can a relative discussion be made, and how can clustering analyses be interpreted? In addition to this concern, the added line in Line 250-257, which will be related to the other reviewer's response, is further confusing. As shown in Table S9, NYC and LAX are classified as VOC-limited and transition, respectively, based on the threshold value suggested by Jin et al.²⁷. But when we assume the threshold value based on Duncan et al. for other regions, Paris is categorized into NO_x-limited for example. Again, because of the possible regional difference in the threshold value, the discussion includes high uncertainty, and I cannot interpret the result.

- Finally, regarding the mismatch between the observational results from the related reference and the CAMS reanalysis, "Nonetheless, the coarse resolution of the used data limits the observation of local variations in Chinese megacity clusters. Li et al. (2022)⁹ reported a continuous increase from 2006 to 2019 of the O₃ 50th percentile in the Pearl River Delta urban sites, however a significant decrease in regional sites was observed. Due to the spatial resolution limitations, this study cannot resolve these local variations.", I want to disagree with this statement. As the authors replied to my previous concern, the finer resolution of the regridding process showed almost the same performance as the original dataset. I partly agree that this could be the spatial resolution; however, I think this is the limitation of the current reanalysis dataset. It is hard to rely on the reanalysis dataset without the appropriate reproducibility of the observational fact. This is just the case in China, and I am suspicious of other cases in other regions analyzed in this study. This is not a denial of the reanalysis dataset, which is a promising way to understand the air quality, but the reanalysis data should be carefully used. I believe observation-oriented analyses should be established.

Reviewer #3

(Remarks to the Author)

The authors fully responded to all suggestions and questions and made necessary adjustments, revisions, or explanations. The revised version has been improved and is ready for publication.

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Reviewer #1

Remarks to the Author: *The topic and results are of interest. Overall, the paper is timely, well-written, robust, and reports informative results. I suggest revising your paper, in particular by: correcting the English/writing throughout the text (some typos), developing your introduction further by highlighting the significance and novelty of your work, and updating the state-of-the-art.*

Dear Reviewer 1,

We appreciate your thorough review and constructive feedback, which helped greatly improve our manuscript. Below, we provide a point-by-point response to each of the recommendations and the changes made in the manuscript.

1. Abstract - *The current version is not informative, i.e., missing quantified results (trend magnitude), missing information about the applied approach (e.g., for trend analysis), etc.*

Response:

The abstract has been modified to include quantitative results and information on the approach used for the analysis. The changes are as follows:

"Ground-level ozone (O₃) reanalysis data from the Copernicus Atmosphere Monitoring Service were used to assess the changes in O₃. Changes from 2005 to 2019 and their relationship with the warming environment in 41 megacities worldwide were evaluated using seasonal Mann-Kendall, trend decomposition methods, and Pearson correlation analysis. The results revealed significant increases in HCHO, with yearly changes ranging from 0.1 to 0.31 ($\times 10^{15}$ mol.cm⁻²), whereas NO₂ varied from -0.22 to 0.07 ($\times 10^{15}$ mol.cm⁻²) depending on the region. The O₃ trends showed variations from -0.31 to 0.70 ppb year⁻¹, aligning with changes in precursor emissions."

Introduction

2. Line 30: ref - *In 2016, 54% of the world's population lived in urban areas (UN, 2018) where ground-level ozone, PM_{2.5}, and NO₂ are the most harmful air pollutants for human health.*

Response:

Based on the provided reference, the population percentage in urban areas by 2016 has been added in lines 32-34.

"Urban agglomerations have increased significantly over the last two decades. In 2016, 54% of the global population lived in urban areas¹, and by 2022, 10% of the urban population was concentrated in 44 megacities²."

3. Line 39: not only – ozone exposure leads to premature deaths & hospital admissions for cardiovascular and respiratory diseases (e.g., 10.1016/j.atmosenv.2023.119926).

Response:

The increased premature deaths caused by ozone and the reference have been included in lines 41-42.

"Ground-level ozone (O₃) is responsible for increased premature deaths and hospital admissions due to cardiovascular and respiratory diseases³"

4. In 2019, the chronic ozone exposure led to 147,100 deaths in 13,160 urban areas with more than 50,000 inhabitants in cities in 2019 worldwide (Malashock et al., 2022).

Response:

The mortality rates with the provided reference have been included in lines 42-44.

"In 2019, chronic O₃ exposure led to 147,100 deaths in urban areas worldwide, with a population exceeding 50,000⁴."

5. Line 40: Ozone is a threat to biodiversity (10.1126/sciadv.abc1176; 10.1007/s11676-023-01645-y).

Response:

Lines 44-46 have been included to discuss the adverse effects of ozone and its threat to biodiversity based on the provided references.

"The adverse effects of O₃ on crops, ecosystems, and materials have been well-documented⁵⁻⁷. O₃ also poses a threat to biodiversity^{8,9} and serves as a greenhouse gas, contributing to climate change¹⁰."

6. *Global exposure of urban population to ozone increased, due to lower O₃ titration by NO. The summertime O₃ MDA8 rose at 74% of cities worldwide(10.1016/j.scitotenv.2022.160064).*

Response:

The discussion about the changes in the O₃ MDA8 , and reduced O₃ titration in VOC-limited areas has been included in lines 49-52 with the recommended references.

"Between 2000 and 2019, summertime O₃ daily maximum 8h values (MDA8) increased in 74% of urban areas worldwide. In cities under VOC-limited conditions, mean O₃ concentrations also increased owing to reduced NO_x emissions, resulting in less titration of O₃ by NO¹¹"

7. *Ozone is rising also by night(10.1126/science.adm7628).*

Response:

The increased relevance of nocturnal ozone has been included in lines 54-57 based on the recommended reference.

"Furthermore, nocturnal O₃ pollution, which is historically considered to be low and unthreatening, is also of rising concern in several regions, such as the United States and China, owing to reduced NO_x emissions without similar reductions in the VOCs.¹²."

8. *Line 45: in general, ozone levels are rising in cities and remote areas, and declining in rural areas (10.1016/j.coesh.2020.100226)*

Response:

The trends in urban and remote areas have been included in lines 49-50 and 52-54 with the recommended references.

"Between 2000 and 2019, summertime O₃ daily maximum 8h values (MDA8) increased in 74% of urban areas worldwide."

"In remote areas, O₃ levels rose owing to increased NO_x emissions and transport of O₃ and its precursor from urban areas¹³⁻¹⁵."

9. Line 76-77: add a reference

Response:

Two references have been added to this claim in lines 95-96.

"Considering that O₃ represents a significant global concern^{14,16}, understanding its long-term changes and those from its precursor emissions is essential"

Results

10. *Table 1: be careful with the number of digits. I suggest adding the trend magnitude for ozone, or from literature, for instance the TOAR database (<https://join.fz-juelich.de>) or the Global Reanalysis Datasets for all cities worldwide at a fine spatial resolution from 2000 to 2019.*

Response:

Thank you for the insightful suggestion. We have included ground-level ozone reanalysis data from the Copernicus Atmosphere Monitoring Service (CAMS). The inclusion of the data led to significant changes in the contents of our manuscript. We list here the sections that have changed and the modifications that have been included.

Title

The title has been modified to consider the inclusion of the ozone data.

"Ozone trends and their sensitivity in global megacities under the warming climate"

Introduction

Lines 93-96 and 100-105 introduce the ground-ozone data analysis and highlight its relevance.

"Monthly O₃ reanalysis data from the Copernicus Atmosphere Monitoring Service (CAMS) were used to evaluate O₃ trend changes during the same period. Considering that O₃ represents a significant global concern^{14,16}, understanding its long-term changes and those from its precursor emissions is essential for assessing the burden of emission-driven O₃ on the increased global budget."

"The findings inform the observed changes in areas with the most significant sources of precursors and highest levels of O₃ pollution globally, which have yet to be achieved, and serve as support for establishing O₃ mitigation strategies on a global scale."

Results

Lines 139-144 present the findings of the ozone changes at a global scale.

"The O₃ levels between 2005 and 2019 showed a significant global increase (Figure 3). The yearly average anomaly for the 50th percentile revealed that in 2019 (Figure 3c), concentrations were remarkably higher than those in previous years, with certain areas reporting an increase of up to 15 ppb. Europe, the Middle East, East Asia, and Southeast Asia are among the regions with the most substantial O₃ differences, which aligns with these regions having the highest emissions of precursors.

Lines 195-214 present the results in the 45 sites based on the new ozone data added in Table 1.

"Consistent with the precursor emission trends, the Mann–Kendall test results for O₃ in Table 1 revealed increasing O₃ in 35 of the 45 studied areas, although the trends were significant in only 25 sites. Interestingly, O₃ in most megacities in China decreased throughout the analysis period. However, the O₃ seasonal trend decomposition analysis in Supplementary Figure 5 displays two periods at several sites: 2005–2011 and 2012–2019. Supplementary Table 3 presents the trend and magnitude changes (ppb year⁻¹) for 2005–2011 and 2012–2019.

From 2005 to 2011, O₃ remained relatively stable in several megacities, such as Los Angeles, London, Paris, Mexico City, and Seoul, but decreased in most megacities in China. However, from 2012 to 2019, a significant increase was observed at 29 of the analysed sites, with the greatest changes observed in megacities in Asia, such as Tehran, Seoul, and Jakarta. Chinese megacities transitioned from decreasing during 2005–2011 to a significant increasing trend from 2012, with Wuhan and the megacity clusters of the Pearl River Delta and Yangtze River Delta showing the most marked changes. Similarly, megacities in India showed decreasing values from 2005 to 2011 but significant increasing trends from 2012 to 2019.

The four remote areas also showed increased O₃ levels from 2012, with the increase being significant only in the Amazon and Congo rainforests, possibly owing to higher biogenic sources of ozone precursors than in the desert areas of the Sahara and the Great Victoria Desert."

Discussion

Lines 359-364 discuss the ozone differences in the analyzed megacities based on the precursor emissions.

"In alignment with the spatial distribution of precursor emissions, the megacities with the highest O₃ levels are also those with the highest precursor emissions, indicating that emission-driven O₃ constitutes the most relevant source of O₃ in megacities globally and will continue to do so mainly because of increased VOC and reduced NO_x emissions in VOC-limited regions and increasing NO_x emissions in regions under transitional/NO_x-limited conditions."

Lines 389-419 discuss the drivers of the ozone trends.

"Although global O₃ trends indicate a generalised increase, regional differences are apparent and are influenced by different factors. The analysis of precursor emissions and their ratios helps explain these differences. NO_x emissions in China increased from 2005 to 2012 but have since decreased. In contrast, O₃ decreased from 2005 to 2011 and increased significantly from 2012 onwards. The FNR in megacities in China is among the lowest in the analysed regions, indicating a VOC-limited regime; therefore, the increase in O₃ as NO_x decreases is evidence of VOC-limited conditions. Furthermore, reducing the precursor emissions without considering the sensitivity regime may lead to increased O₃ owing to the nonlinear relationship between O₃ and its precursors.

However, the high FNR (towards NO_x-limited conditions) observed in tropical megacities in Africa and Southeast Asia indicates that the significant increase in O₃ trends is caused by increased NO_x emissions. Megacities in the Indian region have been increasing their NO_x emissions and O₃ levels significantly since 2012, indicating a NO_x-limited chemical regime from the later period.

In certain analysed megacities such as Los Angeles, Mexico City, Paris, London, Seoul, and Osaka, non-significant changes were observed during 2005–2011 in O₃ levels. However, a steep increase has been observed since 2015, which is possibly associated with higher temperatures. According to NOAA, between 2015 and 2019, five of the warmest years on record occurred¹⁷. This temperature increase during the later years might exacerbate O₃ levels due to increased emissions of precursors of biogenic origin, particularly in areas where the potential to reduce anthropogenic emissions is decreasing¹⁸. Additionally, regional transport has influenced the increase in O₃ levels,

particularly in East and Southeast Asia^{19,20}. Furthermore, vertical transport has been reported to be a relevant factor for persistently high O₃ levels in this region^{21,22}.

The increased O₃ in remote and rural areas has been attributed to increased NO_x emissions driven by soil microbes²³. The four remote areas analysed showed a positive correlation between temperature and NO_x emissions. Hence, because O₃ formation in remote areas is strongly NO_x-limited, O₃ continues to increase with temperature."

Implications

Lines 431-437 and 444-449 summarize the observed trends and their implications.

"The analysis showed that O₃ increased in global megacities from 2005 to 2019 owing to different trends in precursor emissions. Megacities in most developed economies remain in areas with saturated NO_x emissions (towards VOC-limited conditions). Therefore, the increasing VOC and decreasing NO_x stagnated or increased the O₃ levels. In contrast, megacities experiencing rapid development, located mainly in the tropics, show the most significant increase in NO_x emissions. Owing to its transitional/NO_x-limited conditions, O₃ will likely continue to rise in the coming years."

"Urban populations are at higher risk of exposure to unhealthy O₃ levels. Although less extensive urban areas are desired, projections indicate that megacities will continue to grow, especially in fast-developing economies in the tropics and global south. Without effective mitigation efforts, O₃ will continue to be a critical issue for the public health of inhabitants in the largest urban agglomerations globally, along with the adverse effects of increasing global temperatures."

Methods

Lines 505-516 present the methods for retrieving and processing CAMS ozone data.

"Ground-level ozone data"

The Copernicus Atmosphere Monitoring Service (CAMS) provides reanalysis data of atmospheric composition (AC) for different chemical species, including ozone²⁴. The fourth-generation datasets (EAC4) cover 2003 to 2023, providing information in a globally 3-dimensional time-consistent grid with a horizontal spatial resolution of 0.75° at different vertical levels and temporal resolutions. For this analysis, global monthly averaged data at the ground level were retrieved from 2005 to 2019. A regridding process was applied to the datasets to equal the grid size to that for HCHO and NO₂

(0.25 ° x 0.25 °) using bilinear interpolation. The annual global averages were calculated from the monthly datasets. The validation of EAC4 by comparison with Global Atmosphere Watch data showed improvement compared with the previous versions of the reanalysis, with reported overestimations within 30%, particularly in northern latitudes²⁵."

Tables

Table 1 was modified to include the trends of ground-level ozone.

Supplementary Table 3 was included to discuss the ozone trends in the overall period and subdivided into two periods.

Figures

Figure 3 was included to present the global ozone anomaly in three different years.

Supplementary Figure 5 was included to present the deseasonalised ozone trend in the 45 analysed sites.

11. *In addition, with the future vehicle electrification, fostered by the ban of thermal engines, is expected to have an important effect on urban air quality, with improvements in NO₂ pollution and increases in ground-level ozone concentrations (10.1016/j.envres.2023.115835).*

Response:

Lines 344-347 have been included to discuss how vehicle electrification might worsen the O₃ issue in urban areas based on the provided references.

"Additionally, the O₃ issue might worsen in megacities under VOC-limited conditions owing to future vehicle electrification, which will cause a decrease in NO_x but increased O₃ levels caused by reduced titration^{26,27}."

12. *Line 324-325 & Line 401-402 (implication) - Hence, low VOCs emitting tree species should be selected for greening and re-naturing programs (10.1126/science.add973).*

Response:

Based on the provided reference, the importance of choosing the right species for urban greenery has been included in lines 351-353.

"Owing to the increased relevance of BVOC emissions, choosing the right species for green urban infrastructure should be one of the strategies for decreasing VOC in urban areas under VOC-limited conditions²⁸.

Reviewer #2

Remarks to the Author: *This manuscript analyzed the long-term trends of ozone sensitivity using a satellite dataset. Taking into account the journal's high-quality standard, this study should be rejected. I found the following three flaws in this study.*

Dear Reviewer 2,

We are thankful for the feedback you provided. We appreciate the concerns raised and would like to address them as follows in the point-by-point response:

1. First of all, what's new in this study? The methodologies and approaches themselves have been well-used in so many other studies. Even though the target areas extended into the tropics and/or global south, such analyses could be found in other literature. The simple application to other areas would not meet the journal's scope and criteria.

Response:

Although previous studies have examined ozone, its sensitivity, and its relationship with meteorology, our research is the first to comprehensively analyze these factors across multiple global megacities, including underrepresented regions in the tropics and the Global South. By comparing megacities with diverse socioeconomic, climatic, and geographical conditions, we provide new insights into the factors that drive ozone pollution in different urban contexts. Lines 86-90 have been modified to highlight these factors. The presented broader scope allows for a more holistic understanding of the dynamics of ozone and its precursor emissions globally.

"Nonetheless, most studies have focused on regions in northern latitudes; therefore, studies of megacities in the tropics and the global south are relatively few, highlighting the need for an extensive analysis to address this disparity, more so when the projections indicate that by 2030, megacities will be concentrated in the less developed regions or the global south¹."

Additionally, although we employed well-established methodologies, the combinations of different statistical methods helped us verify the identified trends robustly. Moreover, in the revised version of the manuscript, the trends of ground ozone

have been included, which further reinforces our analysis by showing consistency between the precursor emission trends and the ozone trends. By integrating new data sources for ozone, we provide more accurate insights into ozone trends and their sensitivity. These factors have been highlighted in lines 100-105.

"Therefore, the aim of this study was to provide a synoptic evaluation by performing a robust analysis that integrates several tools for trend estimation. The findings inform the observed changes in areas with the most significant sources of precursors and highest levels of O₃ pollution globally, which have yet to be achieved, and serve as support for establishing O₃ mitigation strategies on a global scale."

We believe these aspects collectively represent a significant advancement in the field and meet the journal's scope and criteria by providing new knowledge and useful insights for mitigating ozone pollution.

2. Second, although the manuscript (and title itself) emphasized the relation between ozone to warming climate, the analysis could be misleading. For example, as stated "according to NASA, 2019 was the second warmest year since records", Fig. 6 used the difference in the year 2019 and just imposed the high-temperature biases. However, temperature can be varied year-to-year and the fact will not be so simple. In addition, the ozone regime was diagnosed based on the column data whereas temperature and SWR are surface datasets. I do not fully understand such confusing data analyses.

Response:

We agree about the potential bias that could be introduced by selecting a single warm year. When referencing NASA data, we intended to provide context for the particular significance of 2019 as an anomalously warm year. We have further reinforced this claim by referring to the data sets provided by NOAA, where they compare temperature trends between different datasets (including those from NASA), emphasizing the period from 2005 to 2019. Figure 6 shows the anomaly for 2019 calculated over the average for 2005-2019. Although anomalies are typically calculated at averaging 30 years, the fifteen-baseline comparison in our analysis can provide a clearer picture of how temperatures have deviated in the studied period, not only between two years. We have revised the caption in Figure 6 to improve clarity and reduce potential confusion. Additionally, Supplementary Table 5 and newly added Supplementary Figure 8 provide detailed information on how Temperature and shortwave radiation changed in every site during the analyzed period. Temperature increased significantly in most sites,

giving evidence of a warming environment. Lines 258-262 and 265-269 present the above-mentioned changes.

"Figure 6 shows the temperature and SWR anomalies in 2019, relative to the average for 2005–2019. Temperatures increased between 0.5–1.5 °C in most of the planet, and according to NOAA and NASA, 2019 was the second warmest year since records, and the period spanning 2005 to 2019 held nine of the ten warmest years in records¹⁷, evidencing the increasing global temperatures due to climate change."

"Locally, in the 45 areas, the trend analysis in Supplementary Table 5 shows that the temperature increase was statistically significant in most regions, with Istanbul, Tehran, and Paris exhibiting the most significant increase. Supplementary Figure 8 shows the deseasonalised trend of temperatures at the 45 sites; an increasing trend was observed at most of the analysed sites."

Regarding the use of column data to evaluate the ozone regime, since ozone formation occurs at different levels throughout the Planet Boundary Layer (PBL), column data is useful for capturing all the variations at the different levels. These variations of ozone precursors through the PBL directly influence ground-level ozone concentrations. Therefore, OMI column data have been widely used for diagnosing ozone sensitivity at the ground level, as referenced in lines 74-75 in the introduction.

3. Finally, the satellite data of HCHO and NO₂ column can be a proxy of emissions, but the bottom-up emission inventories need to be analyzed, and discussed the relation between column concentration and emission if the authors suggest the policy regulation.

Response:

Since formaldehyde is a secondary product in the oxidation path of most VOC and NO₂ is closely related to NO_x (due to the rapid oxidation of NO to NO₂), the OMI HCHO and NO₂ have been widely used as proxies of VOC and NO_x, this is discussed in line 68 to 72 in the introduction.

"Formaldehyde (HCHO) is a secondary product of VOC oxidation, and nitrogen dioxide (NO₂) is closely related to NO_x owing to the rapid oxidation of NO to NO₂²⁹. Therefore, HCHO and NO₂ have been widely used as proxies for VOC and NO_x emissions, respectively, and the HCHO to NO₂ ratio (FNR) has been used as an indicator of O₃ formation sensitivity³⁰."

Although we do not directly compare column data with bottom-up emissions, different studies have shown that OMI data correlates well with ground-based measurements, providing confidence in its use for estimating ground-level emissions of ozone precursors. Lines 473-475 and 482-483 in the methods section discuss and provide references about it.

"Validation of the OMI HCHO products showed a strong spatial correlation with aircraft observations. However, a negative bias under high-HCHO conditions and a positive bias under low-HCHO conditions have been reported^{31,32}."

"Previous studies have shown good agreement among OMI, ground-based, and aircraft observations, with correlation factors of >0.7 ³³."

Additionally, the revised version of the manuscript includes ground-level ozone reanalysis data globally and in the 45 selected sites. We have evaluated the ozone trends during the same period as with OMI data, finding consistency in the precursor emission trends and ground-level ozone trends, further validating the usefulness of OMI observations to diagnose the ozone sensitivity at the surface level. The discussion of the ground-level ozone data based on the FNR has been added in lines 389-403.

"Although global O_3 trends indicate a generalised increase, regional differences are apparent and are influenced by different factors. The analysis of precursor emissions and their ratios helps explain these differences. NO_x emissions in China increased from 2005 to 2012 but have since decreased. In contrast, O_3 decreased from 2005 to 2011 and increased significantly from 2012 onwards. The FNR in megacities in China is among the lowest in the analysed regions, indicating a VOC-limited regime; therefore, the increase in O_3 as NO_x decreases is evidence of VOC-limited conditions. Furthermore, reducing the precursor emissions without considering the sensitivity regime may lead to increased O_3 owing to the nonlinear relationship between O_3 and its precursors.

However, the high FNR (towards NO_x -limited conditions) observed in tropical megacities in Africa and Southeast Asia indicates that the significant increase in O_3 trends is caused by increased NO_x emissions. Megacities in the Indian region have been increasing their NO_x emissions and O_3 levels significantly since 2012, indicating a NO_x -limited chemical regime from the later period."

Reviewer #3

Remarks to the Author: *This manuscript evaluates the trends of OMI HCHO and NO₂ in 41 megacities worldwide, explores ozone sensitivity through their ratios, and analyses their correlation with temperature. The authors provide a global perspective on the study of ozone and precursor changes. However, the current manuscript leaves something to be desired. I suggest that the manuscript be further refined before it is considered for publication.*

Dear Reviewer 3,

Thank you for your valuable feedback on our manuscript. We appreciate your time and effort in providing detailed comments and suggestions. Below, we address each of your points and describe our revisions to the manuscript.

1. Ozone sensitivity: *The authors do not explicitly provide ozone sensitivity, let alone "ozone sensitivity trends", but only FNR values and qualitative analyses, which is insufficient for a study on the topic of Ozone sensitivity.*

Response:

We acknowledge the need for a more explicit analysis of ozone sensitivity based on the FNR. Using the Mann-Kendall and trend decomposition, We have included a quantitative evaluation of the ozone-sensitivity trends through the FNR. Supplementary Table 2 and Supplementary Figure 4 have been included, and the results have been added in lines 184-194.

"The Mann-Kendall analysis of FNR (Supplementary Table 2) indicates that ozone sensitive trends are shifting towards transitional/NO_x-limited conditions in most of the compared sites, primarily owing to the generalized increase in HCHO trends. The greatest magnitude changes were observed in megacities in the tropics and the global south, such as Jakarta and Sao Paulo, owing to their significant elevations in HCHO and decreasing trends in NO₂. Megacities in developed economies are also moving towards higher FNR values but at a slower pace. Dhaka and Lagos were the only megacities with significant reductions in their FNR, coinciding with being the megacities with the most significant in NO₂. Supplementary Figure 4 shows the deseasonalised trends for FNR in all the analysed sites."

2. Ozone data: It is recommended that authors use ground-level ozone data to analyze trends, even for just a few stations or cities. This will allow for specific analyses rather than just speculation when exploring the influence of precursors or meteorological factors on ozone changes, and will be more persuasive for the conclusion than just that "the observed trends are contrary to what is required to reduce O₃ levels".

Response:

We appreciate your valuable comment. We have included reanalysis data of ground-level ozone from the Copernicus Atmosphere Monitoring Service (CAMS) for all the analyzed sites. The ozone data inclusion led to major changes in the contents of our paper, which are detailed below:

Title

The title has been modified to consider the inclusion of the ozone data.

"Ozone trends and their sensitivity in global megacities under the warming climate"

Introduction

Lines 93-96 and 100-105 introduce the ground-ozone data analysis and highlight its relevance.

"Monthly O₃ reanalysis data from the Copernicus Atmosphere Monitoring Service (CAMS) were used to evaluate O₃ trend changes during the same period. Considering that O₃ represents a significant global concern^{14,16}, understanding its long-term changes and those from its precursor emissions is essential for assessing the burden of emission-driven O₃ on the increased global budget."

"The findings inform the observed changes in areas with the most significant sources of precursors and highest levels of O₃ pollution globally, which have yet to be achieved, and serve as support for establishing O₃ mitigation strategies on a global scale."

Results

Lines 139-144 present the findings of the ozone changes at a global scale.

"The O₃ levels between 2005 and 2019 showed a significant global increase (Figure 3). The yearly average anomaly for the 50th percentile revealed that in 2019 (Figure 3c), concentrations were remarkably higher than those in previous years, with certain areas reporting an increase of up to 15 ppb. Europe, the Middle East, East Asia, and Southeast

Asia are among the regions with the most substantial O₃ differences, which aligns with these regions having the highest emissions of precursors.

Lines 184-214 present the results in the 45 sites based on the new ozone data added in Table 1.

"Consistent with the precursor emission trends, the Mann–Kendall test results for O₃ in Table 1 revealed increasing O₃ in 35 of the 45 studied areas, although the trends were significant in only 25 sites. Interestingly, O₃ in most megacities in China decreased throughout the analysis period. However, the O₃ seasonal trend decomposition analysis in Supplementary Figure 5 displays two periods at several sites: 2005–2011 and 2012–2019. Supplementary Table 3 presents the trend and magnitude changes (ppb year⁻¹) for 2005–2011 and 2012–2019.

From 2005 to 2011, O₃ remained relatively stable in several megacities, such as Los Angeles, London, Paris, Mexico City, and Seoul, but decreased in most megacities in China. However, from 2012 to 2019, a significant increase was observed at 29 of the analysed sites, with the greatest changes observed in megacities in Asia, such as Tehran, Seoul, and Jakarta. Chinese megacities transitioned from decreasing during 2005–2011 to a significant increasing trend from 2012, with Wuhan and the megacity clusters of the Pearl River Delta and Yangtze River Delta showing the most marked changes. Similarly, megacities in India showed decreasing values from 2005 to 2011 but significant increasing trends from 2012 to 2019.

The four remote areas also showed increased O₃ levels from 2012, with the increase being significant only in the Amazon and Congo rainforests, possibly owing to higher biogenic sources of ozone precursors than in the desert areas of the Sahara and the Great Victoria Desert."

Discussion

Lines 359-364 discuss the ozone differences in the analyzed megacities based on the precursor emissions.

"In alignment with the spatial distribution of precursor emissions, the megacities with the highest O₃ levels are also those with the highest precursor emissions, indicating that emission-driven O₃ constitutes the most relevant source of O₃ in megacities globally and will continue to do so mainly because of increased VOC and reduced NOX

emissions in VOC-limited regions and increasing NO_x emissions in regions under transitional/NO_x-limited conditions."

Lines 389-419 discuss the influence of the FNR on the observed ozone trends.

"Although global O₃ trends indicate a generalised increase, regional differences are apparent and are influenced by different factors. The analysis of precursor emissions and their ratios helps explain these differences. NO_x emissions in China increased from 2005 to 2012 but have since decreased. In contrast, O₃ decreased from 2005 to 2011 and increased significantly from 2012 onwards. The FNR in megacities in China is among the lowest in the analysed regions, indicating a VOC-limited regime; therefore, the increase in O₃ as NO_x decreases is evidence of VOC-limited conditions. Furthermore, reducing the precursor emissions without considering the sensitivity regime may lead to increased O₃ owing to the nonlinear relationship between O₃ and its precursors.

However, the high FNR (towards NO_x-limited conditions) observed in tropical megacities in Africa and Southeast Asia indicates that the significant increase in O₃ trends is caused by increased NO_x emissions. Megacities in the Indian region have been increasing their NO_x emissions and O₃ levels significantly since 2012, indicating a NO_x-limited chemical regime from the later period.

In certain analysed megacities such as Los Angeles, Mexico City, Paris, London, Seoul, and Osaka, non-significant changes were observed during 2005–2011 in O₃ levels. However, a steep increase has been observed since 2015, which is possibly associated with higher temperatures. According to NOAA, between 2015 and 2019, five of the warmest years on record occurred¹⁷. This temperature increase during the later years might exacerbate O₃ levels due to increased emissions of precursors of biogenic origin, particularly in areas where the potential to reduce anthropogenic emissions is decreasing¹⁸. Additionally, regional transport has influenced the increase in O₃ levels, particularly in East and Southeast Asia^{19,20}. Furthermore, vertical transport has been reported to be a relevant factor for persistently high O₃ levels in this region^{21,22}.

The increased O₃ in remote and rural areas has been attributed to increased NO_x emissions driven by soil microbes²³. The four remote areas analysed showed a positive correlation between temperature and NO_x emissions. Hence, because O₃ formation in remote areas is strongly NO_x-limited, O₃ continues to increase with temperature."

Implications

Lines 431-437 and 444-449 summarize the observed trends and their implications.

"The analysis showed that O₃ increased in global megacities from 2005 to 2019 owing to different trends in precursor emissions. Megacities in most developed economies remain in areas with saturated NO_x emissions (towards VOC-limited conditions). Therefore, the increasing VOC and decreasing NO_x stagnated or increased the O₃ levels. In contrast, megacities experiencing rapid development, located mainly in the tropics, show the most significant increase in NO_x emissions. Owing to its transitional/NO_x-limited conditions, O₃ will likely continue to rise in the coming years."

"Urban populations are at higher risk of exposure to unhealthy O₃ levels. Although less extensive urban areas are desired, projections indicate that megacities will continue to grow, especially in fast-developing economies in the tropics and global south. Without effective mitigation efforts, O₃ will continue to be a critical issue for the public health of inhabitants in the largest urban agglomerations globally, along with the adverse effects of increasing global temperatures."

Methods

Lines 505-516 present the methods for retrieving and processing CAMS ozone data.

"Ground-level ozone data

The Copernicus Atmosphere Monitoring Service (CAMS) provides reanalysis data of atmospheric composition (AC) for different chemical species, including ozone ²⁴. The fourth-generation datasets (EAC4) cover 2003 to 2023, providing information in a globally 3-dimensional time-consistent grid with a horizontal spatial resolution of 0.75° at different vertical levels and temporal resolutions. For this analysis, global monthly averaged data at the ground level were retrieved from 2005 to 2019. A regridding process was applied to the datasets to equal the grid size to that for HCHO and NO₂ (0.25° x 0.25°) using bilinear interpolation. The annual global averages were calculated from the monthly datasets. The validation of EAC4 by comparison with Global Atmosphere Watch data showed improvement compared with the previous versions of the reanalysis, with reported overestimations within 30%, particularly in northern latitudes ²⁵."

Tables

Table 1 was modified to include the trends of ground-level ozone.

Supplementary Table 3 was included to discuss the ozone trends in the overall period and subdivided into two periods.

Figures

Figure 3 was included to present the global ozone anomaly in three different years.

Supplementary Figure 5 was included to present the deseasonalised ozone trend in the 45 analysed sites.

3. Due to the long time scales of the study, there may be exceptions to the non-significant trends in ozone precursors, such as the rapid decline in NO₂ in Beijing from 2013, as shown in Figure 5, which is the case for many cities in China. This change in trend helps to study the role of precursor changes under ozone sensitivity.

Response:

With the inclusion of the ground-ozone data, this observation was very helpful in identifying associations in the ozone trends with those from NO₂, especially as the reviewer mentioned, in megacities in China. The results and analysis considering this aspect have been included in lines 195-201 and 389-398

"Consistent with the precursor emission trends, the Mann–Kendall test results for O₃ in Table 1 revealed increasing O₃ in 35 of the 45 studied areas, although the trends were significant in only 25 sites. Interestingly, O₃ in most megacities in China decreased throughout the analysis period. However, the O₃ seasonal trend decomposition analysis in Supplementary Figure 5 displays two periods at several sites: 2005–2011 and 2012–2019. Supplementary Table 3 presents the trend and magnitude changes (ppb year⁻¹) for 2005–2011 and 2012–2019."

"Although global O₃ trends indicate a generalised increase, regional differences are apparent and are influenced by different factors. The analysis of precursor emissions and their ratios helps explain these differences. NO_x emissions in China increased from 2005 to 2012 but have since decreased. In contrast, O₃ decreased from 2005 to 2011 and increased significantly from 2012 onwards. The FNR in megacities in China is among the lowest in the analysed regions, indicating a VOC-limited regime; therefore, the increase in O₃ as NO_x decreases is evidence of VOC-limited conditions."

Furthermore, reducing the precursor emissions without considering the sensitivity regime may lead to increased O₃ owing to the nonlinear relationship between O₃ and its precursors."

4. Increase in temperature: The authors attempt to explore the effect of "temperature change" on ozone concentrations, but do not give quantitative results for temperature change. Figure 6 only shows the situation in 2019. In addition, the term "warming climate" is not appropriate for this scale of temperature change.

Response:

The discussion about the changes in temperature and the quantitative results has been extended. Figure 6 shows the anomaly for 2019 calculated over the average for 2005-2019. We have revised the caption in Figure 6 to improve clarity and reduce potential confusion. Additionally, Supplementary Table 4 and newly added Supplementary Figure 8 provide detailed information on how Temperature and shortwave radiation changed in every site during the analyzed period. Temperature increased significantly in most sites, giving evidence of a warming environment. The results are discussed in lines 258-262 and 265-269.

"Figure 6 shows the temperature and SWR anomalies in 2019, relative to the average for 2005–2019. Temperatures increased between 0.5–1.5 °C in most of the planet, and according to NOAA and NASA, 2019 was the second warmest year since records, and the period spanning 2005 to 2019 held nine of the ten warmest years in records¹⁷, evidencing the increasing global temperatures due to climate change."

"Locally, in the 45 areas, the trend analysis in Supplementary Table 5 shows that the temperature increase was statistically significant in most regions, with Istanbul, Tehran, and Paris exhibiting the most significant increase. Supplementary Figure 8 shows the deseasonalised trend of temperatures at the 45 sites; an increasing trend was observed at most of the analysed sites."

5. Clustering analysis: The authors classify the cities into four clusters according to FNR values, but the significance of this needs further consideration. In fact, they can be regarded as different ozone generation sensitivity classified by FNR. As the authors state, emission trends differed among the grouped megacities, so what is the usefulness of cluster analysis? Authors explain the different roles of meteorological factors in terms of whether they are developed regions or not, which in itself has no relevance to the results of the clustering analysis.

Response:

We have refined the discussion to better justify the significance of clustering analysis when analyzing the ozone trends. We have explicitly linked the clusters to the ozone sensitivity regimes and discussed the ozone trends in each group based on the FNR. This discussion has been included in lines 340-344 and 354-364

"Clustered megacities with the lowest FNRs (VOC-limited) not only included regions with the most significant NO₂ emissions globally but also those with the sharpest NO₂ decrease. However, NO_x reduction is ineffective in decreasing O₃ under VOC-limited conditions, even more so when VOC emissions are also increasing, as indicated by the increasing HCHO trends."

"The most significant increase in NO₂ levels was observed in Cluster 2, which grouped areas under transitional/NO_x-limited regime. In particular, Dhaka and Bengaluru showed the highest relative differences between 2005 and 2019 (Supplementary Table 6). Similarly, Cluster 3 of megacities with the highest FNRs (strong NO_x-limited conditions) displayed increasing NO_x trends."

In alignment with the spatial distribution of precursor emissions, the megacities with the highest O₃ levels are also those with the highest precursor emissions, indicating that emission-driven O₃ constitutes the most relevant source of O₃ in megacities globally and will continue to do so mainly because of increased VOC and reduced NO_x emissions in VOC-limited regions and increasing NO_x emissions in regions under transitional/NO_x-limited conditions."

6. Figures and Table: *The time scale and units of the data should be given in Table 1. The meaning of the colours should be indicated in Figure 3. How was the trend calculated in Figure 5?*

Response:

We have revised Table 1 to include the units for each data set. The colors in Figure 4 (previously Figure 3) were not associated with any specific aspect, so the figure's color has been changed to avoid confusion. For Supplementary Figure 3 (previously Figure 5), we have added a description of the trend calculation methodology in the caption, and the Methods section provides detailed information about it in lines 563-577.

"Seasonal trend decompositions using LOESS

Seasonal trend decompositions using LOESS (STL) was performed to complement the SK test. This is a robust method for analysing time-series data with recurring temporal patterns. The method proposed in Cleveland et al. (1990) decomposes the time-series data y_t into three main components as follows:

$$y_t = T_t + \sum_{i=1}^I S_t^{(i)} + R_t \quad (3)$$

where T_t is the changing trend, $S_t^{(i)}$ is the seasonal component, and R_t is the remaining component. This method uses multivariate locally weighted regression (Loess)³⁵ for smoothing operations. STL analysis was performed using the monthly 50th percentile of all variables at the 45 sites using the Statsmodels module in Python³⁶

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Reviewer #1

Remarks to the Author: *The authors replied to open issues satisfactorily. The requested changes and suggestions were taken into account, and any missing information added. The revised version is improved, and ready for publication.*

Dear Reviewer 1,

We are grateful for your time and effort in reviewing our manuscript. Your insightful comments have been instrumental in enhancing the quality and relevance of our paper. We are pleased that the revisions we have made align with your expectations.

Reviewer #2

Remarks to the Author: *This manuscript is the revised version that was previously submitted to this journal. I sincerely appreciate the authors' revisions to address my comments. After the clarification from the authors, I reread this carefully, but I still have concerns about considering it as a possible publication from this journal's high-quality standard. My biggest concerns were the lack of appropriate references and relevant discussions. Especially, the added discussion using O3 reanalysis data has been severely lacking in discussion with previous research and strongly relates to the analysis using their data.*

Dear Reviewer 2,

We appreciate your consideration of our revised manuscript and acknowledgment of the previous revisions. We understand your concerns and have taken them into account. Below, we provide a point-by-point response to the issues raised and the changes made to the manuscript.

Major points:

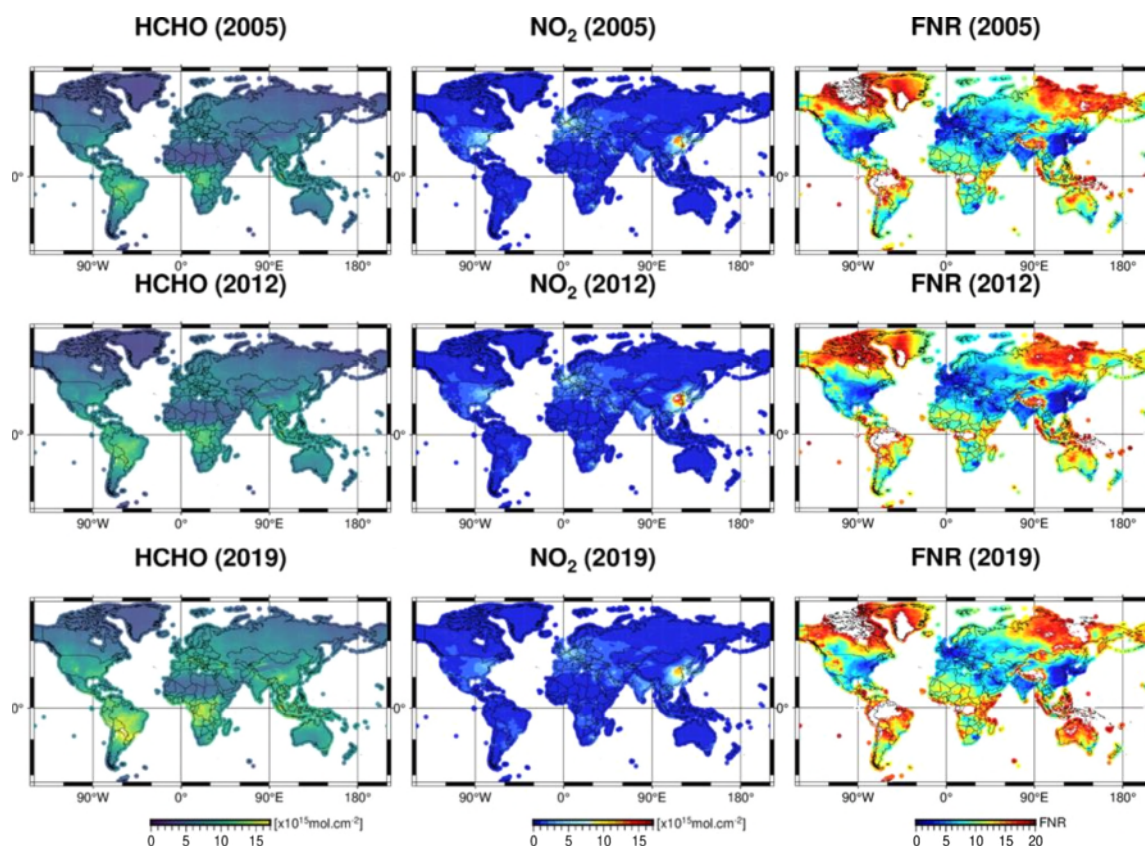
1. *The visualization and the relevant discussion will still be difficult to understand. In Fig. 2., HCHO and NO₂ columns were shown from 1.0E15 to 17.0E15 mol.cm⁻², so I guess that these white colors will indicate lower concentration (less than 1.0E15), right? Thus, the white color in the FNR map corresponds to a higher value rather than 6, right? However, I am confused "When mapping the data, masks for land-ocean, HCHO values below 7×10¹⁵ mol cm⁻², NO₂ values below 1×10¹⁵ mol cm⁻², and FNR values above*

10 were applied to facilitate data visualisation." in line 501-503. Could you clarify this analysis methodology?

Response:

We apologize for the confusion and appreciate your detailed observation. The white areas in the HCHO maps correspond with values below $7 \times 10^{15} \text{ mol.cm}^{-2}$ and for NO_2 below $1 \times 10^{15} \text{ mol.cm}^{-2}$. However, we set the color bars to start from $1 \times 10^{15} \text{ mol.cm}^{-2}$ in both cases for consistency and better color distribution. On the other hand, the FNR maps show only data for values 1-6. We set these values to drive the focus to the most polluted regions, which are the main targets of our analysis. We have included a non-masked version of the maps in supplementary figure 9 for comparative purposes. To address this comment, we have modified the description in lines 551-555, to indicate that for FNR, the mask was 6 and not 10 as erroneously mentioned earlier. We have also included this description in the Figure 2 caption for better understanding.

"When mapping the data, masks for land-ocean, HCHO values below $7 \times 10^{15} \text{ mol cm}^{-2}$, NO_2 values below $1 \times 10^{15} \text{ mol cm}^{-2}$, and FNR values above 6 were applied to facilitate data visualisation and for driving attention to the polluted areas. Maps with non-masked values are shown in Supplementary Figure 9 for comparative purposes."



Supplementary Figure 9. Global distribution of formaldehyde (HCHO), nitrogen dioxide (NO₂), and their ratios (FNR) in 2005, 2012, and 2019 with non-masked values.

2. *It is very interesting to see the behavior of HCHO, NO₂, and FNR in Africa as categorized into cluster no. 1. However, based on the analysis shown in Fig. 2, NO₂ column corresponded to a relatively lower region. In this sense, how could we consider the detection limit of NO₂ column? The usage of HCHO was described in line 486, but it is also required for the detailed description of NO₂. I am afraid that a high FNR has simply drawn from a low NO₂ which was just lower than the detection limit. Without clarification, the analysis would be meaningless.*

Response

The latest OMI NO₂ retrieval algorithm¹ set a threshold value of $0.3 \times 10^{15} \text{ mol.cm}^{-2}$ when separating the tropospheric column from the stratospheric. This value was based on Boersma et al. (2004)², who indicated that the detection limit of NO₂ columns in the troposphere is $0.3 \times 10^{15} \text{ mol.cm}^{-2}$. The NO₂ data in Figure 2 was masked to values $>1 \times 10^{15} \text{ cm}^{-2}$, which is well above the detection limit. For the specific case of the Congo

region in Africa, categorized in cluster 1, the mean NO₂ concentration was 0.48 x10¹⁵ mol.cm⁻², also above the reported detection limit. The NO₂ mean values at every site have been added to supplementary table 5, and as observed, all are above the detection limit. Therefore, the high FNR in the remote areas is due to low NO₂ levels (yet within the detection limit) and the high HCHO due to the abundant BVOC sources. The documentation for the data product version 4.0 recommends that all valid measurements, regardless of their sign, should be included to avoid biases when computing statistics³. Therefore, no filtering was applied to the NO₂ daily retrievals. This information has been included in lines 538-540.

*"For NO₂, following the recommendations in the documentation of the L3 OMNO2d v4, all daily values were included in the averaging procedure, regardless of their sign"*³

3. *The added analysis for O3 based on the reanalysis data seems to be partly critical; however, I do not speculate the methodology of the re-gridding from a coarser resolution to a finer resolution. Although it could be easy in the re-gridding from a finer resolution to a coarser resolution as HCHO column, it seems to be difficult how to regrid into a finer resolution. Did this apply some a priori or other information?*

Response

No apriori information or assumptions were used for the regridding process. Regridding or remapping involves transferring data between different grids while preserving the qualities of the original dataset. Among the most used methods are bilinear and conservative interpolations, which are monotonic operators, indicating that no new data are generated during the process⁴. We acknowledge that regridding from a finer to a coarser resolution is more straightforward. However, the same approach applies when going from a coarse to a finer resolution, the spatial detail of the original dataset remains the same, and the finer grid represents the same data with more points; the inherent resolution of the dataset does not increase. Specifically, bilinear interpolation is an adequate method for highly spatially correlated data in a regular rectilinear grid⁵, as is the case with the data used in our analysis. The Earth System Modelling Framework is a well-known tool that provides examples of regridding from a coarse to a finer resolution using bilinear interpolation⁶.

To exemplify the above for the specific case of the ground-ozone data from CAMS used in our analysis, here we provide (Figure R1) two maps for the ozone data in July 2012 with the original grid size of 0.75 degrees (a) and the regridded dataset to a grid size of

0.25 degrees. As observed, the ozone distribution is the same in both datasets. Additionally, the global median, 50th percentile, and standard deviation are shown in Table R1 for both datasets, observing that they remain the same despite the higher number of grids (n) in the 0.25-degree dataset. Thus, the regridding process does not affect or alter the data contents.

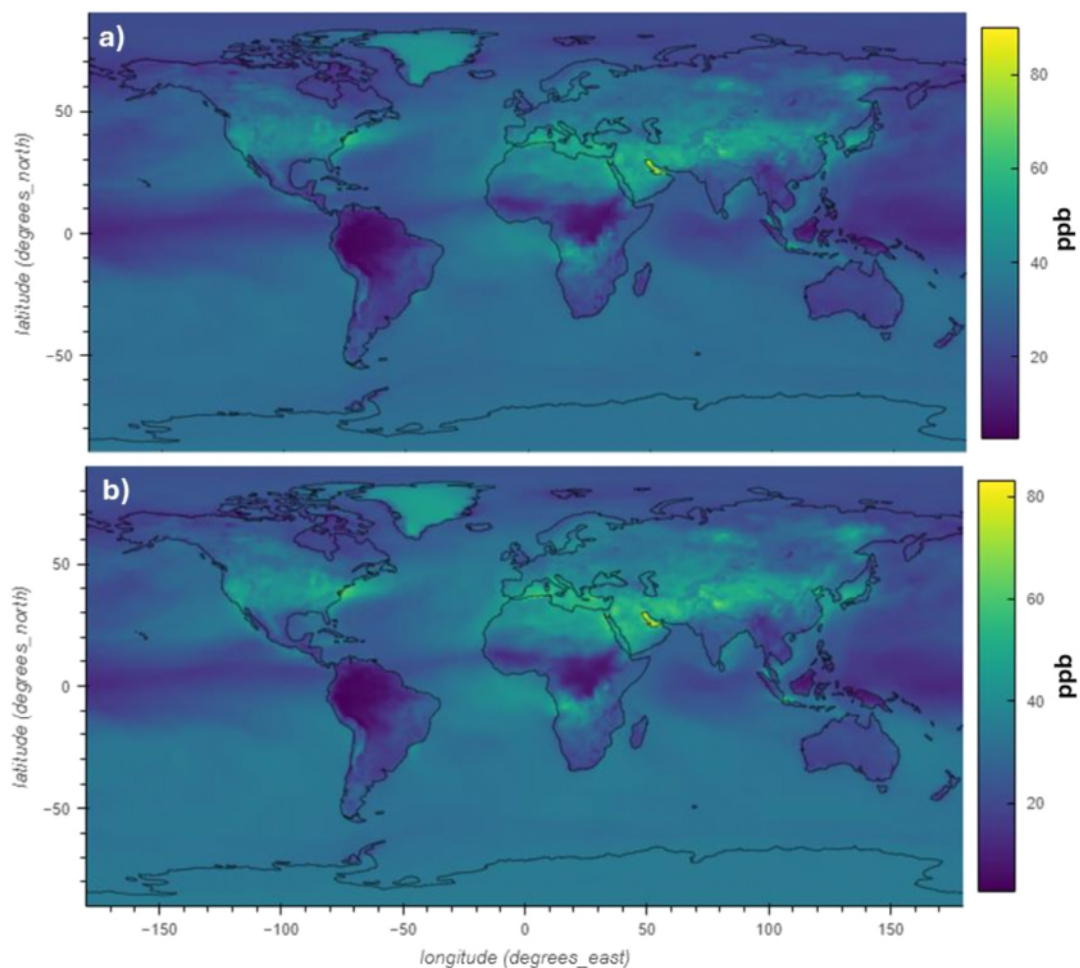


Figure R1. Ground-level ozone distribution for July 2012 at the original resolution of 0.75 degrees (a), and after regridding to 0.25 grid size (b).

Table R1. Mean, 50th percentile (50p), and standard deviation (SD) for the CAMS ground-level ozone data at its native resolution (0.75°) and after regridding it to 0.25°.

Grid size				
(°)	n	Mean	50p	SD
0.75	115680	29.28	31.92	8.12
0.25	1036800	29.28	31.92	8.17

4. Related to this O₃ analysis, the authors claimed that the transition from the decreasing trend to the increasing trend in China. However, this result and discussion on China in lines 195-210 will be conflict with the continuously increasing O₃ trend reported in previous papers (<https://doi.org/10.5194/acp-16-10637-2016>; <https://doi.org/10.5194/acp-19-1455-2019>; <https://doi.org/10.1016/j.atmosenv.2021.118869>). The discussion was very weak in the current manuscript quality just relying on their data without referring to appropriate pieces of literature. Judging from this careless analysis without the previous studies, I am sorry, but the results and discussion on other sites will also be suspicious.

Response

In our analysis, we observed a transition from a decreasing to a significantly increasing O₃ trend in specific regions of China. This finding may appear to contradict the consistently increasing trends reported in the provided references. However, we believe this apparent discrepancy can be explained by differences in the targeted regions, time periods, spatial scales, and methodologies used in our study compared to these previous works.

The article titled "Significant increase of summertime ozone at Mount Tai in Central Eastern China" by Sun et al. (2016)⁷ analyzes the increase in surface ozone (O₃) concentrations from 2003 to 2015 at Mount Tai, China. The authors report a significant increase in the O₃ levels. However, the analyzed region is not part of our targeted areas, which might explain the differences. The article "Impacts of meteorology and emissions on summertime surface ozone increases over central eastern China between 2003 and 2015" by Sun et al. (2019)⁸ analyzes the significant rise in surface ozone levels over central China using an air quality model. The analyzed region encompasses several of the included megacities in our study. While our analysis

calculated monthly time series for 2005 to 2019 using statistical methods considering seasonality, Sun et al. (2019) evaluated the O₃ differences only during the summer between 2003 and 2015. Focusing on a single season between only two years might have led to different results; additionally, 2003 was not considered in our analysis. The article "Long-term trend of ozone in southern China reveals future mitigation strategy for air pollution" by Li et al (2022)⁹. investigates the ozone trends from 2006 to 2019 in the Pearl River Delta, one of our analysis's targeted areas. The authors analyzed ozone trends at a fine scale using data from 16 monitoring stations, finding that the 50th percentile at urban sites continuously increased, but regional sites showed decreasing trends. The finer spatial resolution used in their analysis allowed them to identify local trend differences, which is one of the limitations of our analysis. The coarse resolution of the data used in our study makes it challenging to identify regional variations in the ozone trends. Furthermore, those non-accounted regional differences might explain the decreasing trends observed from 2005 to 2011 in our results. To address this issue, we have revised the manuscript to include a discussion of the limitations of our analysis based on Li et al. (2022) in lines 434-439.

"Nonetheless, the coarse resolution of the used data limits the observation of local variations in Chinese megacity clusters. Li et al. (2022)⁹ reported a continuous increase from 2006 to 2019 of the O₃ 50th percentile in the Pearl River Delta urban sites, however a significant decrease in regional sites was observed. Due to the spatial resolution limitations, this study cannot resolve these local variations."

Specific points:

Line 72-78: *I am confusing this explanation. Despite these discussions for FNR values, why did the authors use the method of Duncan et al. (2010)? To avoid confusion, I will recommend moving or repeating the sentence in lines 495-497. I agree with focusing on the trend analysis instead of the FNR value because the threshold values are still discussed. However, the authors compared FNR at site-to-site and discussed the ozone formation regime. If there is a difference in the threshold value site-by-site, it is not as simple as discussed.*

Response

Thank you for your raised concern. The main objective of lines 72-78 was to provide a robust framework for using FNR for ozone sensitivity analysis. Since the work by Duncan et al. (2010)¹⁰ was the first to propose using HCHO and NO₂ ratios from OMI,

we based the text in lines 72-78 on it. However, the uncertainties in the FNR threshold values across different sites and times were also highlighted. The results did not aim to provide a specific threshold value for regime classification site by site. As the reviewer pointed out, establishing ratios for inferring ozone sensitivity requires a specific analysis that considers the chemical characteristics of every site. We appreciate your suggestion of including that clarification in the results and not only in the methods. The site-by-site analysis (lines 181-199) was based on the observed trends without providing a specific threshold for regime classification for each megacity. To clarify this comment, we have included lines 148-153.

"Due to the inherent spatial and temporal variability of the FNR values that define ozone sensitivity regimes, the results did not aim to provide a specific threshold value for regime classification at every site, however they emphasised the observed trends across regions. Sites displaying the lowest FNR were considered as VOC-limited conditions, intermediate as transitional/NO_x-limited, and the highest FNR as NO_x-limited regions."

Line 162-166: *It should be carefully defined which countries are developed/developing countries. Specifically, which is categorized for China?*

Response

Thank you for your suggestion. To address this comment, we have used the World Bank's income classification system to categorize the country of every megacity. This system categorizes countries into low, lower-middle, upper-middle, and high income economies. Our analysis considered high income economies to be developed, while the rest of the classifications were considered developing economies. On that basis, China would be a developing economy. We have included Supplementary Table 7 and added lines 592-596 in the main text to address this comment.

"The countries of the 41 megacities were categorised as developed and developing based on the World Bank income classification system¹¹. In this study, megacities in high-income countries are considered developed, and those in upper-middle, middle-low, and low-income countries are considered developing (Supplementary Table 8)."

Line 391-392: *We cannot follow NO_x emission itself from this manuscript. Need reference. Even though the measured column can be a proxy of emissions, it could not be defined as emissions.*

Response

Thank you for pointing this out. References confirming the observed trends in our analysis have been included. The word "emissions" has been replaced by "column densities". The changes are in lines 431 – 433.

"NO₂ column densities in China increased from 2005 to 2012 but have since decreased, which has also been confirmed by other studies^{9,12,13}

Line 392-393: *See my major concern.*

Response

Thank you for your comment. We have addressed this major concern by including a discussion of the limitation of our analysis in lines 434-439.

"Nonetheless, the coarse resolution of the used data limits the observation of local variations in Chinese megacity clusters. Li et al. (2022)⁹ reported a continuous increase from 2006 to 2019 of the O₃ 50th percentile in the Pearl River Delta urban sites, however a significant decrease in regional sites was observed. Due to the spatial resolution limitations, this study cannot resolve these local variations. "

Line 396-398: *This sentence seems to be redundant.*

Response

Thank you for pointing this out. The lines have been removed.

Line 433: *Related to the comment on lines 162-166, what was in China? Some literatures also reported the transition of ozone sensitive regime in China, but they noted that the shift from VOC-limited to a transitional or NO_x-limited regime (<https://doi.org/10.5194/acp-21-7253-2021>; <https://doi.org/10.1002/2017JD026720>)*

Response

Thank you for pointing this out. In our analysis, megacities in China were among the regions with the highest NO₂ column densities, which caused them to be under NO_x-saturated (VOC-limited) conditions. However, the FNR trends showed increasing values due to decreasing NO₂ and increasing HCHO. Our findings agree with those in the provided references, although some differences exist in the analyzed time periods, and methods. Wang et al. (2020) reported that megacities in China were under VOC-limited conditions during 2016, and they reported that areas in the North China Plain,

the Yangtze River Delta, and the Pearl River Delta shifted to transitional regimes. However, the central regions of the megacities remained under VOC-limited conditions. Jin et al. (2019) performed a more detailed analysis comparing the monthly FNR in megacities in China between 2005 and 2015. Their findings showed that VOC-limited conditions prevail for 8 to 9 months per year throughout the entire analyzed period and 1.4 to 2.2 months under NO_x-saturated conditions. Thus, megacities in China remain mainly under VOC-limited conditions. This discussion with the provided references has been included in lines 440-445.

"The FNR in megacities in China is among the lowest in the analysed regions, and the increase in O₃ as NO_x decreases is evidence of VOC-limited conditions. However, the FNR showed increasing trends due to reduced NO₂ and increased HCHO, which will likely cause a shift towards transitional/ NO_x-limited conditions in the coming years. Other studies have reported this change in the ozone sensitivity regime in several regions of China ^{14,15}"

Line 453: *I am still wondering if this remark is overstated because the emissions themselves have never been analyzed.*

Response

Thank you for pointing this out. We acknowledge that direct emissions were not measured in our analysis. However, satellite observations provide valuable proxies for emissions, especially in areas where ground observations are unavailable or limited. We have revised the wording of the statement to address this comment. The changes are in lines 500-502.

"The results of this analysis support the selection of adequate strategies for reducing O₃ based on the long-term trends of its precursor, as inferred from satellite observations."

Reviewer #3

Remarks to the Author: *The authors have revised the manuscript extensively to emphasize its importance and novelty. I recommend further refinement of the manuscript before considering it for publication.*

Dear Reviewer 3,

We appreciate your continued engagement and valuable feedback, which have been fundamental in enhancing the clarity and relevance of our manuscript. Below, we provide a point-by-point response to each of the recommendations and the changes made in the manuscript.

1. Based on the FNR metric it is possible to clearly indicate which areas are VOC-sensitive or NO_x-sensitive and demonstrate the changes visually. What considerations did the author fail to provide this information?

Response

Thank you for your raised concern. We did not provide a specific regime classification for the analysed sites due to the spatial and temporal dependency of the threshold values for each ozone sensitivity regime. Studies have shown that the thresholds for regime classification need to be adjusted for every region, taking into consideration the chemistry feedback^{15,16}. A detailed, location-specific analysis would be required to accurately determine a threshold to classify each of the 45 analysed areas, which could detract from the broader insights presented in our study. Therefore, our analysis focused on analyzing the trends of the indicators, which is also helpful in informing mitigation strategies. This discussion has been included in lines 148-153 in the results section.

"Due to the inherent spatial and temporal variability of the FNR values that define ozone sensitivity regimes, the results did not aim to provide a specific threshold value for regime classification at every site, however they emphasised the observed trends across regions. Sites displaying the lowest FNR were considered as VOC-limited conditions, intermediate as transitional/NO_x-limited, and the highest FNR as NO_x-limited regions."

Nonetheless, to shed light on the regime classification, Supplementary Table 9 has been included, which shows the mean values of HCHO, NO₂, and FNR between 2005 and 2019 in each of the 45 analysed sites. A regime classification has been assigned to

every site based on the threshold values established by Jin et al. (2020). Although this classification can provide some insights into the regime for every site, it should be noted that the threshold values were determined specifically for urban areas in the US. The mean values in Supplementary Table 5 were used for the clustering analysis. Thus, the relevant discussion has been added in lines 248-255.

"Supplementary Table 5 shows the mean HCHO, NO₂, and FNR at each site used in the clustering analysis from 2005 to 2019. When comparing the FNR at each site with the threshold values established by Jin et al. (2020)¹⁷ for regime classification, where the transitional regime was at FNR 3-4, it was observed that sites in cluster 3 would be under VOC-limited conditions, while the rest were under transitional or NO_x-limited conditions. Although this classification can provide some insights into the regime of each site, it should be noted that the used threshold values were determined specifically for urban areas in the US."

2. *With regard to the effects of temperature and radiation, the authors focus mainly on the increase in biological VOC emissions and seem to neglect their contribution to the chemical reactions that lead to ozone production.*

Response

Thank you for pointing this out. We have addressed this comment by extending the discussion of the relevance of the warming environment on the photochemical processes that lead to O₃ pollution. The text has been included in lines 344-360

"Furthermore, in addition to influencing the precursor emissions, the warming environment also influences the reaction rates of the photochemical processes controlling O₃ formation. Globally, rising temperatures lead to increased BVOC emissions as well as higher O₃ formation rates owing to increased recycling of NO_x from isoprene nitrates formed from BVOC oxidation, as demonstrated by Ito et al. (2009)¹⁸. Coates et al. (2016)¹⁹ conducted a modelling study that evaluated the effects of temperature on O₃ under different NO_x conditions, finding that increased VOC oxidation reaction rates and increased peroxy nitrate decomposition rates led to higher O₃ production as temperature increased. Similarly, Meng et al. (2023)²⁰ analysed the O₃ formation under extreme temperature events in urban areas of China, combining observations with simulated data. Their findings showed that radical cycling is more effective at high temperatures. Therefore, the rate of HO₂ + NO significantly increases with temperature, leading to NO removal without O₃ consumption, which is a major

factor in causing a net O₃ accumulation. Moreover, they reported that O₃ production duration is longer under extreme heat temperatures than under cold weather.

Overall, the warming climate is causing increased emissions of ozone precursors from biogenic sources while enhancing the photochemical processes that result in O₃ production. The above emphasises the need to consider the direct and indirect effects of increasing temperatures in a continuously warming environment where extreme temperature events are expected to rise."

3. *We already know that an increase in VOC or a decrease in NO_x alone increases ozone levels in VOC-sensitive areas, and that ozone precursor abatement measures depend on the O₃ sensitivity. The authors build on this by conducting a clustering analysis to categorize sites into four clusters. What is the significance of this? Any new insights into the relationship between ozone changes and precursors? Further clarification is needed.*

Response.

Thank you for your raised concern. The relevance of the clustering analysis has been highlighted by discussing its usefulness in finding joint strategies for mitigating O₃ pollution. We have included lines 269-279.

"Through categorising the sites based on their ozone sensitivity, it is possible to find areas that could share similar approaches in their O₃ regulations. The regions with the highest O₃ levels fell within the same group owing to their similar FNR, which indicates that joint efforts to control air pollution among these regions could be an effective approach to address the issue. Additionally, the clusters highlighted that economic development influences O₃ sensitivity; O₃ formation is more dependent on the VOC emissions in developed and industrialised economies. Furthermore, their increasing FNR indicates that O₃ sensitivity would vary at different levels of development. Thus, the air quality management and policy measures of megacities in developed regions could serve as an example for those in developing economies, which are in transitional/NO_x-limited regimes."

4. *The abstract is too succinct in its presentation of the results, simply stating "The O₃ trends ... aligning with changes in precursor emissions", which does not emphasize the novelty of this study at all.*

Response

Thank you for your observation. We have revised the abstract to underscore the novelty and significance of our study, highlighting how the results contribute to understanding the O₃ trends and their relation to precursor emissions and climate change. The revised version is as follows:

"Tropospheric ozone formation is sensitive to the emissions of its main precursors, volatile organic compounds (VOC) and nitrogen oxides (NO_x). In megacities, abundant VOC and NO_x sources cause relentlessly high O₃ episodes, affecting a large proportion of the global population. In this study, formaldehyde (HCHO) and nitrogen dioxide (NO₂) column densities from the Ozone Monitoring Instrument were used as proxies for VOC and NO_x emissions, respectively, using their ratio as an indicator of ozone sensitivity. Ground-level ozone (O₃) reanalysis data from the Copernicus Atmosphere Monitoring Service were used to assess the changes in O₃. Changes from 2005 to 2019 and their relationship with the warming environment in 41 megacities worldwide were evaluated using seasonal Mann-Kendall, trend decomposition methods, and Pearson correlation analysis. The results revealed significant increases in HCHO, with yearly changes ranging from 0.1 to 0.31 ($\times 10^{15}$ mol.cm⁻²), whereas NO₂ varied from -0.22 to 0.07 ($\times 10^{15}$ mol.cm⁻²) depending on the region. The O₃ trends showed variations from -0.31 to 0.70 ppb year⁻¹, highlighting the significant influence of precursor ratios and their effect on O₃ levels. Furthermore, the strong correlation between precursor emissions and increasing temperature suggests that O₃ will continue to rise as climate change persists. This study provides support for establishing global O₃ mitigation strategies."

5. The colors in Figure 3 make it difficult to distinguish between positive and negative values. In addition, the marine portion can be deducted as shown in Figure 2.

Response

Thank you for pointing this out. We have revised Figure 3 to make the O₃ differences more straightforward. The scale of the color bar was modified to improve the color distribution, and an ocean-land mask was also included.

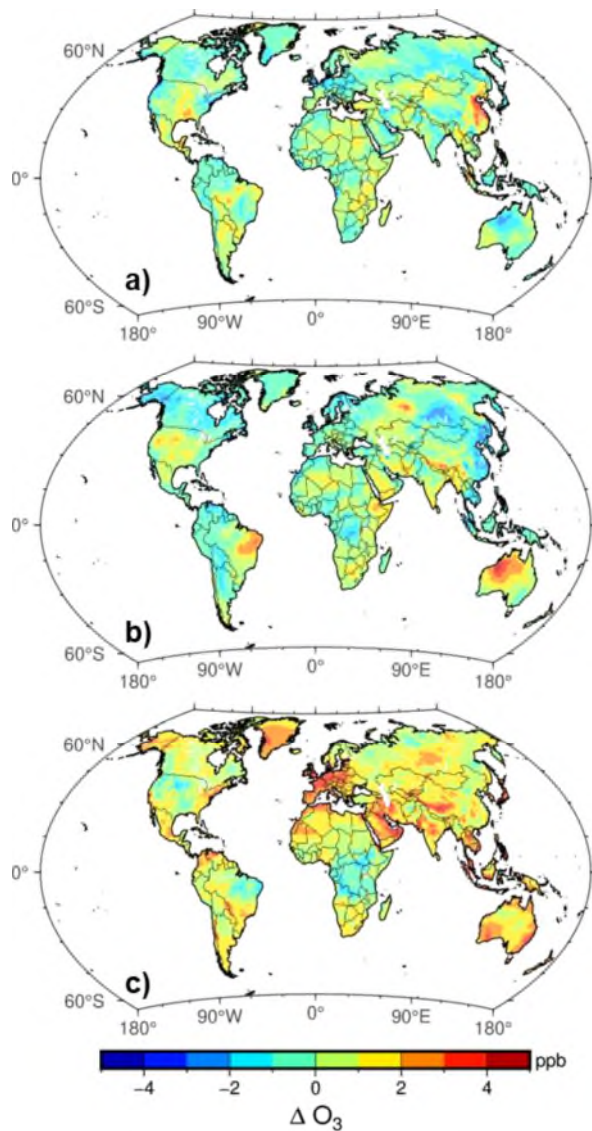


Figure 3. Global surface ozone (O_3) anomalies in 2005 (a), 2012 (b), and 2019 (c) relative to the average for 2005–2019.

6. The time frame is not indicated in table 1 and related text.

Response

Thank you for your comment. The title for Table 1 has been modified to include the time frame of the analysis. The description in the main text has also been reworded to include the timeframe; this change is in lines 145-147.

"The results of the Mann–Kendall test for trend detection and Sen's slope test for the magnitude of change between 2005 – 2019 are shown in Table 1 for HCHO, NO₂, and O₃."

7. Line 223-234: The sequence numbers of the clusters do not match those in the previous text and figure.

Response

Thank you for your observation. The cluster description has been revised to agree with those in Supplementary Figure 7 and Supplementary Table 4. The changes are in lines 226-239.

"Megacities in the most developed economies, such as New York, Paris, Moscow, Tokyo, and Osaka, were grouped into regions with high NO₂ and intermediate levels of HCHO (Cluster 0). Urban areas in India, South Africa, Iran, and Pakistan were also assigned to this group. The remaining sites had similar NO₂ emissions but were differentiated by their HCHO levels. Los Angeles, Istanbul, Cairo, and Karachi were among the regions with lower NO₂ and HCHO levels (Cluster 2). Notably, these megacities have dry climates and low vegetation. The remote areas of the Saharan and Great Victorian deserts were also assigned to this cluster. In contrast, urban areas with tropical climates, such as Jakarta, São Paulo, Bangkok, and Ho Chi Minh, were grouped as regions with lower NO₂ and high HCHO (Cluster 1). Remote areas in the Amazon and Congo rainforests were also in this cluster."

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Reviewer 2

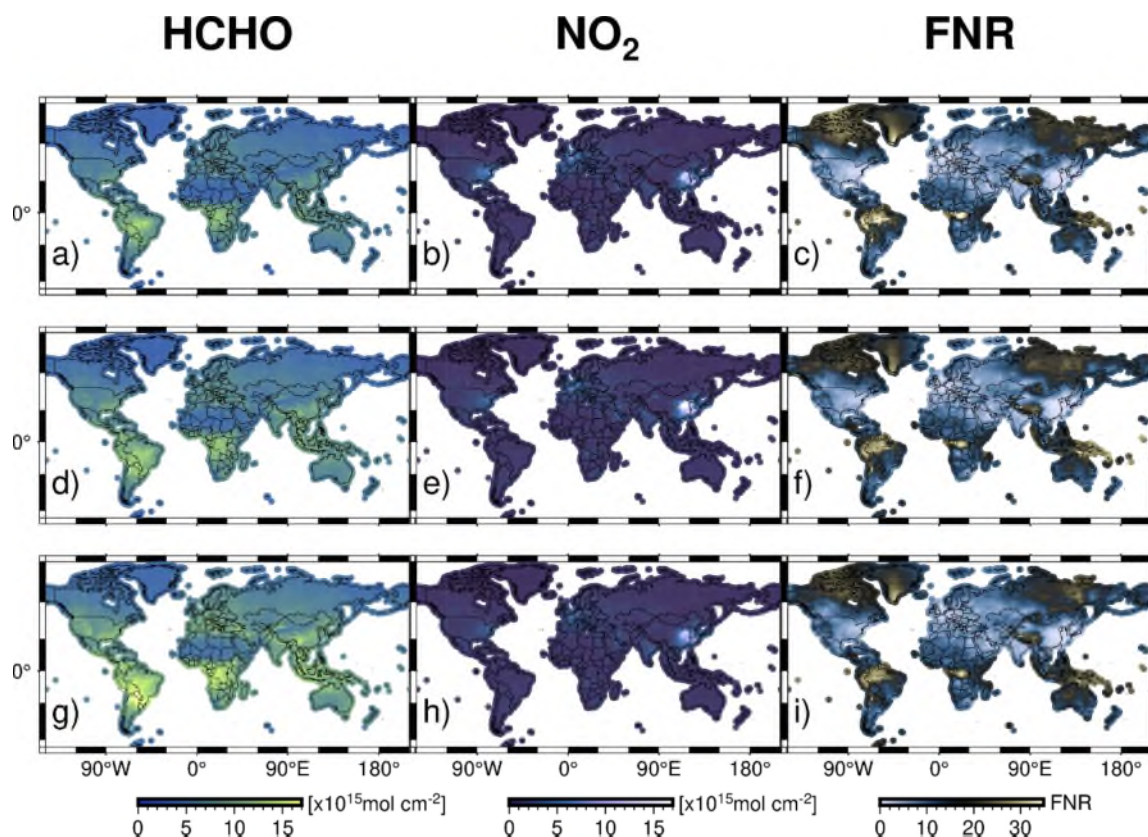
Remarks to the Author: This manuscript is the second revision that has been submitted to this journal. I greatly appreciate the authors' revisions to address concerns; however, I do not fully understand their response to the following Q&A.

Dear Reviewer 2:

We appreciate your time in carefully revising our manuscript and appreciate your raised concerns. Here, we provide a point-by-point response to the issues raised:

1. Regarding the color scales for NO₂, HCHO, and FNR, I can still see the white areas even in Supplementary Fig. 9 over South America. This region will correspond to Amazonia Rainforest (AMZ), one of the significant regions classified as cluster 1. Unfortunately, I cannot fully follow the discussion regarding the non-masked version.

Response: Thank you for your comment. As the reviewer points out, the white areas for FNR in Supplementary Figure 9 correspond to the Amazon and Congo rainforests. Since these regions have the highest HCHO column densities, their FNR is among the highest in the compared sites, with median values above 25, as shown in Supplementary Table 5. Since the scale of the non-masked map for FNR was set to 25, these regions appeared as white areas. To address this issue, we have modified the scale of the figure, extending it to 35. As observed, the pointed-out regions are not shown as white areas in the new Supplementary Figure 9. We apologize for the confusion caused.



Supplementary Figure 9. Global distribution of yearly averaged column densities from OMI for formaldehyde (HCHO), nitrogen dioxide (NO₂), and their HCHO/NO₂ ratios (FNR) in 2005 (a-c), 2012 (d-f), and 2019 (g-i) at a 0.25 × 0.25 degrees of spatial resolution with non-masked values (except for land-ocean). Units for HCHO and NO₂ are ×10¹⁵ mol cm⁻², and FNR is dimensionless. Thin black lines represent country boundaries or shorelines.

2. Even in the possible spatial difference in the threshold value of FNR, I did not catch up with the added line in Line 149-154 “Due to the inherent spatial and temporal variability of the FNR values that define ozone sensitivity regimes, the results did not aim to provide a specific threshold value for regime classification at every site, however they emphasised the observed trends across regions. Sites displaying the lowest FNR were considered as VOC-limited conditions, intermediate as transitional/NO_x-limited, and the highest FNR as NO_x-limited regions.”. Under the possible difference in the threshold value of FNR as previous studies suggested, why can a relative discussion be made, and how can clustering analyses be

interpreted? In addition to this concern, the added line in Line 250-257, which will be related to the other reviewer's response, is further confusing. As shown in Table S9, NYC and LAX are classified as VOC-limited and transition, respectively, based on the threshold value suggested by Jin et al.²⁷. But when we assume the threshold value based on Duncan et al. for other regions, Paris is categorized into NO_x-limited for example. Again, because of the possible regional difference in the threshold value, the discussion includes high uncertainty, and I cannot interpret the result.

Response: Thank you for your comment. The discussion on lines 149-154 was intended to explain that our analysis does not seek to provide a universal threshold value for regime classification. However, clustering regions with similar characteristics in their HCHO, NO₂, and FNR levels could help to identify trends across areas, making it possible to interpret regions with similar ozone sensitivities. We acknowledge that these specific lines might confuse readers due to the section where they are placed. The lines have been removed, and the explanation has been included in the clustering analysis section. The description is now in lines 213-220.

“Due to the inherent spatial and temporal variability of the FNR values that define ozone sensitivity regimes, the analysis did not aim to provide a single universal threshold for each site. However, by clustering regions with similar HCHO, NO₂, and FNR, it was possible to identify areas with similar ozone sensitivities. In the clusters, sites displaying the lowest FNR were considered to have VOC-limited conditions, sites with intermediate FNR were considered to have transitional/NO_x-limited conditions, and sites with the highest FNR were considered NO_x-limited regions.”

Regarding the classification provided in Supplementary Table 5, we agree that thresholds from different studies can lead to different classifications. This region-dependant classification has been emphasized in the revised manuscript, and the uncertainties associated with the regime classification in Supplementary Table 9 have been highlighted. The discussion has been included in lines 233-237.

“Although this classification can provide some insights into the regime of each site, it should be interpreted with caution because of the high uncertainty associated with it due to the spatial and temporal dependency of the established thresholds. In the case of Jin et al. (2020), the values were determined specifically for urban areas in the US.”

3. Finally, regarding the mismatch between the observational results from the related reference and the CAMS reanalysis, "Nonetheless, the coarse resolution of the used data limits the observation of local variations in Chinese megacity clusters. Li et al.

(2022)9 reported a continuous increase from 2006 to 2019 of the O3 50th percentile in the Pearl River Delta urban sites, however a significant decrease in regional sites was observed. Due to the spatial resolution limitations, this study cannot resolve these local variations.", I want to disagree with this statement. As the authors replied to my previous concern, the finer resolution of the regridding process showed almost the same performance as the original dataset. I partly agree that this could be the spatial resolution; however, I think this is the limitation of the current reanalysis dataset. It is hard to rely on the reanalysis dataset without the appropriate reproducibility of the observational fact. This is just the case in China, and I am suspicious of other cases in other regions analyzed in this study. This is not a denial of the reanalysis dataset, which is a promising way to understand the air quality, but the reanalysis data should be carefully used. I believe observation-oriented analyses should be established.

Response: Thank you for your insightful comment. Despite its utility in providing a broader regional understanding, we fully agree that reanalysis datasets have inherent limitations. In response, we have clarified in the manuscript that while reanalysis data offer a valuable tool for identifying large-scale trends, they may not fully replicate the variability seen in ground-based observations, especially at the local level. We have emphasized caution when interpreting reanalysis data for localized analysis and the need for observation-oriented analyses for a more precise understanding of air quality trends. The discussion has been included in lines 453-465.

"Additionally, although the CAMS EAC4 reanalysis dataset provided valuable insights into the regional ozone trends and their precursors across the sites analysed, the inherent spatial resolution was insufficient to resolve the fine-scale spatial heterogeneity observed in extensive urban areas fully. Therefore, caution is warranted when using reanalysis data for site-specific interpretations. Future studies should consider integrating higher-resolution datasets to complement the regional insights provided by reanalysis.

Lastly, satellite and reanalysis datasets comprehensively view atmospheric conditions over large areas. However, they may not fully replicate the detailed variability captured by ground-based observations. Thus, improving the monitoring infrastructure in developing regions is essential for obtaining a better-integrated analysis, which would help validate the accuracy of satellite-based and reanalysis datasets at global scales."