Referee #1

** General comments **

The authors use CAM6-Chem to explore regional trends in organic aerosol concentrations over China. In addition to quantifying the trends, they discuss reasons for the trends.

Following minor revisions, the paper could be a useful contribution to the scientific literature.

** Specific comments **

1. In the data availability statement, I have been unable to identify where the authors' CAM6-Chem model data can be accessed. Should the relevant model data not be published to comply with ACP's data policy and support reproducibility? Would it also be possible to publish the analysis code used to produce the figures?

Response: Thanks for the comment. The long-term CAM6-Chem model data is too large to be published through public FTP or cloud drives. The code and data used to generate the figures will be made available upon reasonable request.

2. The clarity of some of the figures could be improved. Ideally, each figure should be understandable without referring to the caption. To improve readability, I offer the following suggestions:

(i) give each subfigure panel a short title (as has been done in e.g. Fig. 2, but not Figs. 1 & 7);

(ii) label colorbars with units;

(iii) explain the difference between the solid and dashed lines in the Fig. 1e legend (not just the caption);

(iv) share colorbars across subfigure panels where possible (as has been done in e.g. Fig. 3a–c, but not Fig. 2a–c);

(v) use the same y-axis limits in Fig. 9a and 9b.

Response: Thank you for your helpful suggestions on improving the clarity and readability of the figures. We have carefully considered each of your points and made the following revisions to the manuscript:

(i) We have added short titles to each subfigure panel in Figs. 1 and 7, similar to what was done in Fig. 2, to enhance the overall clarity and make each panel easier to interpret without referring to the caption.

(ii) We have labeled the colorbars with appropriate units in Figs. 1,2,3 and 7 to improve clarity and ensure that the data presentation is fully self-explanatory.

(iii) We have revised the legend in Fig. 1e to explicitly explain the difference between the solid and dashed lines, so that the distinction is clear without needing to refer to the caption.

(iv) We have shared colorbars across subfigure panels in Fig 2, in line with the approach used in Fig. 3a–c, to enhance consistency and make the figures easier to interpret.

(v) We have adjusted the y-axis limits in Figs. 9a and 9b to ensure consistency between the two figures, as per your suggestion.

We believe these changes significantly improve the readability of the figures, and we appreciate your careful review.

3. The precision of the numbers in the Fig. 1c colorbar (-1.80 to 1.80) differs from that in Fig. 1f (-1.8 to 1.8). Should they not be consistent?

Response: Thank you for pointing out the inconsistency in the precision of the numbers in the colorbars of Fig. 1c and Fig. 1f. We have revised the colorbar labels in both figures to ensure that the precision is consistent.
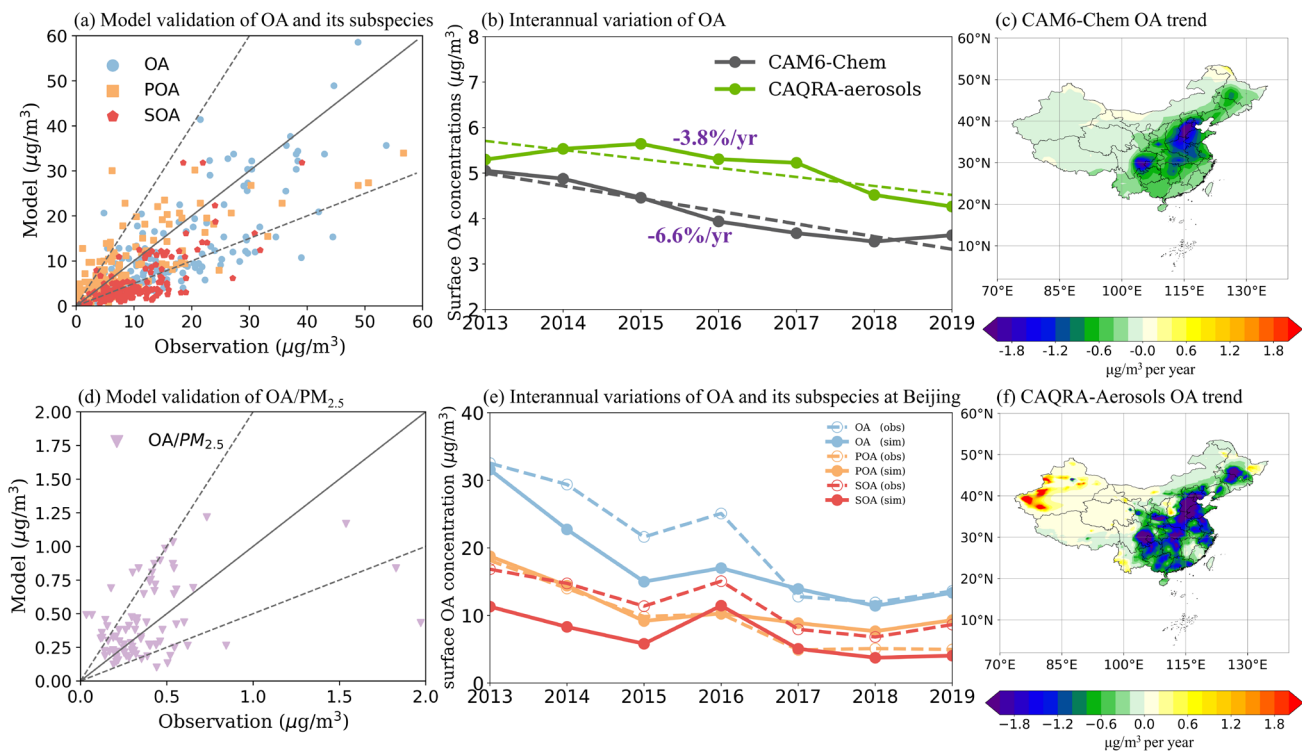
Figure 1: (a) Validation of modelled organic aerosols (OA), primary organic aerosols (POA), and secondary organic aerosols (SOA) based on ground-based measurements compiled by Miao et al. (2021) and Chen et al. (2024) (unit: μg m⁻³). (b) 2013 to 2019 interannual variation of average surface organic aerosol (OA) concentrations in CAM6-Chem (dark grey) and the CAQRA-aerosols dataset (green) (unit: μg m⁻³). The trend lines (dotted line) are based on linear regression fitting. (c) 2013 to 2019 CAM6-Chem modelled annual long-term trend of surface OA concentrations (unit: μg m⁻³ per year). The trend is calculated by linear regression on an annual scale over 1990-2019. (d) Validation of modelled OA/PM2.5 based on OA measurements compiled by Miao et al. (2021) and Chen et al. (2024) and PM2.5 observations from the National Urban Air Quality Real-time Release Platform of China Environmental Monitoring Station. (e) Interannual variation of average surface OA, POA and SOA concentrations (μg m⁻³) at Beijing site from 2013 to 2019 in CAM6-Chem (solid line; sim) and ground-based measurements compiled by Miao et al. (2021) and Chen et al. (2024) (dashed line; obs). (f) 2013 to 2019 annual long-term trend of surface OA concentrations (unit: μg m⁻³ per year) in the CAQRA-aerosols dataset. The trend is calculated by linear regression on an annual scale over 1990-2019.

4. In Fig. 4, why are normal distributions fitted? The normal distribution does not provide a good approximation to the underlying empirical distributions. The exponential distribution may be more appropriate.

Response: Thanks for your suggestion. The original curve fitting was intended to provide a more intuitive representation of the concentration differences between the two time periods for each species, in addition to the bar chart. Compared to the normal distribution, the exponential distribution does indeed better approximate the basic empirical distribution. However, the results from the exponential distribution fitting did not show significant differences between the two periods, and the normal distribution did not closely match the bar chart distribution. Therefore, we switched to using a logarithmic scale for the bar chart, and it is now unnecessary to use fitting curve. The results (Fig. 4) more clearly illustrate the differences between the two time periods.
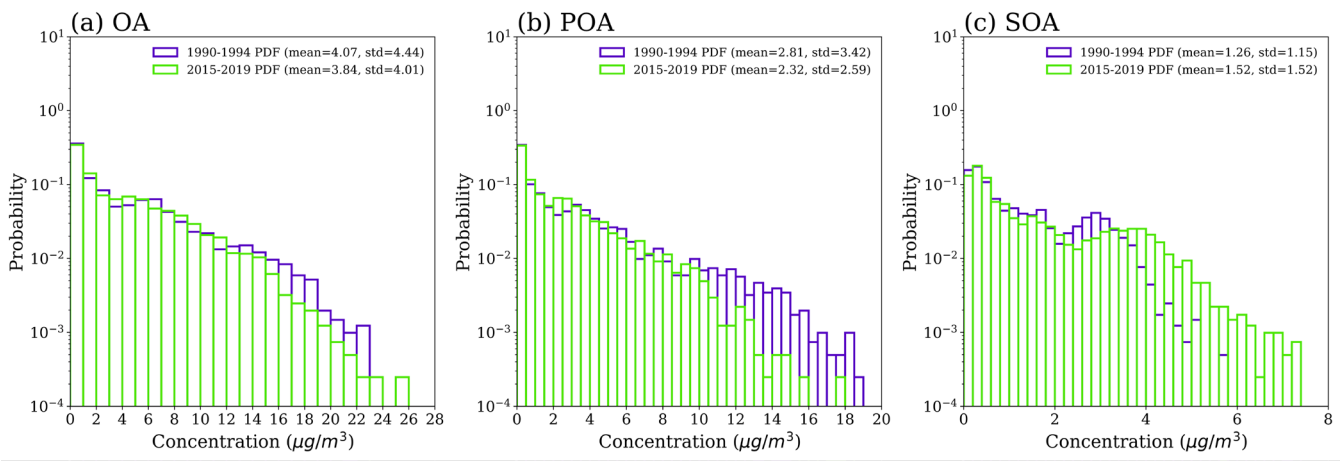
Figure 4: Probability density function (PDF) distributions of simulated five-year average concentrations of surface organic aerosols (OA; a), primary organic aerosols (POA; b), and secondary organic aerosols (SOA; c) for the periods 1990-1994 (purple) and 2015-2019 (green), along with the annual mean values and standard deviations for each species in different time periods.

5. The number of acronymns may be overwhelming to many readers. Is it possible to use fewer acronyms? For example, could "PRD" be replaced by "Pearl River Delta" throughout the paper?

Response: Thank you for your valuable suggestion regarding the use of acronyms. To improve the readability of the paper and reduce the potential for confusion, we have replaced all region-related acronyms with their full names

throughout the manuscript. For example, "PRD," "NCP," "SBA," and "YRD" have been replaced by "Pearl River Delta," "North China Plain," "Sichuan Basin Area," and "Yangtze River Delta," respectively.

Referee #2

** General comments **

The paper titled "Understanding the Long-term Trend of Organic Aerosol and the Influences from Anthropogenic Emission and Regional Climate Change in China" uses the CAM6-Chem model to analyze trends in organic aerosol (OA) in China from 1990 to 2019, identifying the roles of anthropogenic emissions and climate factors. The findings highlight a modest OA increase due to a rise in secondary organic aerosols (SOA) and a decrease in primary organic aerosols (POA), influenced by emission changes and warming. This work enhances our understanding of how emission controls and climate change have shaped OA dynamics over the past three decades. Generally, the paper is well-organized and demonstrates significant effort. However, the following questions need to be addressed prior to publication:

1. As the paper spans 30 years, significant changes in land use (due to deforestation, increased green space in urban areas, etc.) could have occurred. Does the work consider the effect of land use change? If not, it would be helpful to add a few lines discussing the impact of land use on biogenic emissions and resulting SOA concentrations.

Response: Thanks for your suggestion. Our study considered the impact of land use and land cover change (LUCC). The model captured the influence of LUCC through changes in the Leaf Area Index (LAI), which served as a key input parameter to represent the dynamic variations in LUCC (Nemani et al., 1996). BVOC emissions were further simulated online with the MEGAN model (Emmerson et al., 2018; Emmons et al., 2020; Guenther et al., 2012; Wang et al., 2022) incorporated within the CAM6-Chem. LUCC affected vegetation density, which in turn has significant impacts on plant transpiration, carbon absorption capacity and BVOC emissions (Fu and Liao, 2014). In the model, changes in LAI represent the impact of LUCC (such as deforestation, afforestation and urban greening

expansion) on BVOC emissions. The simulation results (Fig. 3(f)) showed an increasing trend in BVOC emissions over the study period. This increase may partly attributed to a series of ecological restoration and conservation policies (Guo et al., 2022), such as the Grain-for-Green Program and Urban Ecological Civilization Construction (Yin et al., 2018). These policies effectively promoted the restoration and expansion of vegetation while driving the continuous increase in urban greening areas. In addition, the warming trend (see Fig. 8(a)) may also enhanced the biosynthesis and emission of BVOC. These changes together contributed to the upward trend in SOA concentrations. We have added additional explanations between lines 380 and 390.
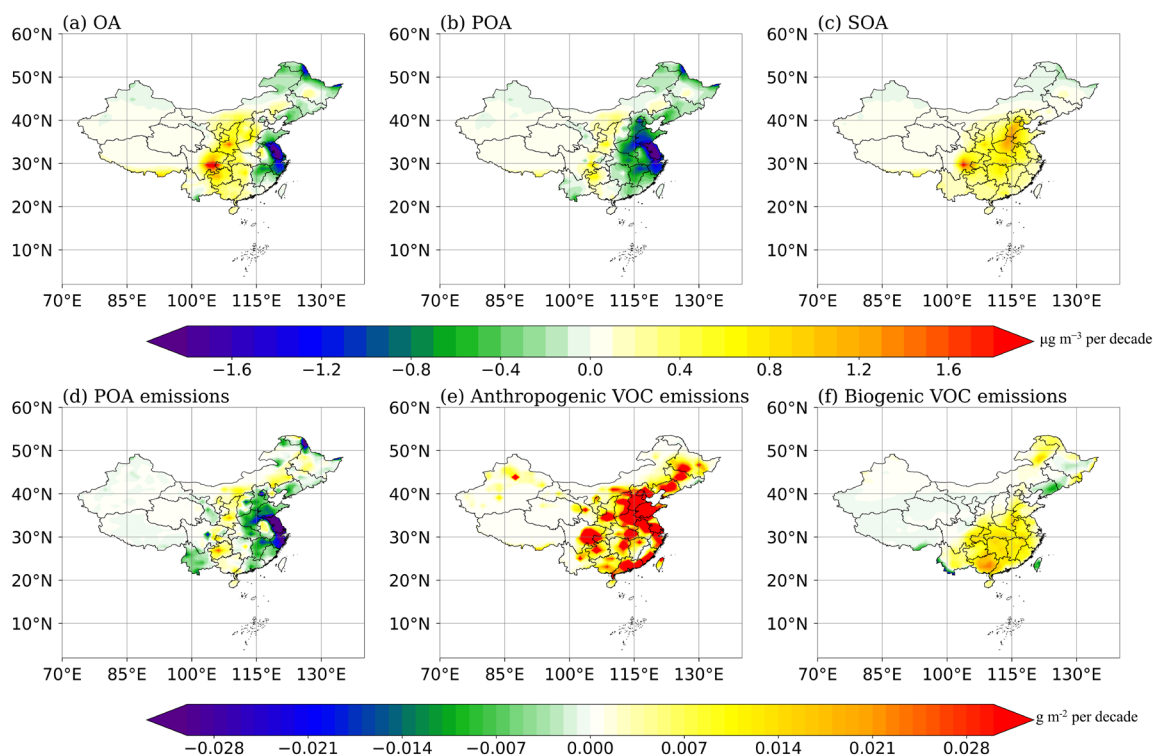
Figure 3: 1990 to 2019 annual average long-term trend of surface organic aerosols (OA; a; unit: μg m⁻³ per decade), primary organic aerosols (POA; b; unit: μg m⁻³ per decade), secondary organic aerosols (SOA; c; unit: μg m⁻³ per decade), primary organic aerosols emissions (d; unit: g m⁻² per decade), anthropogenic volatile organic compounds emissions (e; unit: g m⁻² per decade), biogenic volatile organic compounds emissions (f; unit: g m⁻² per decade).
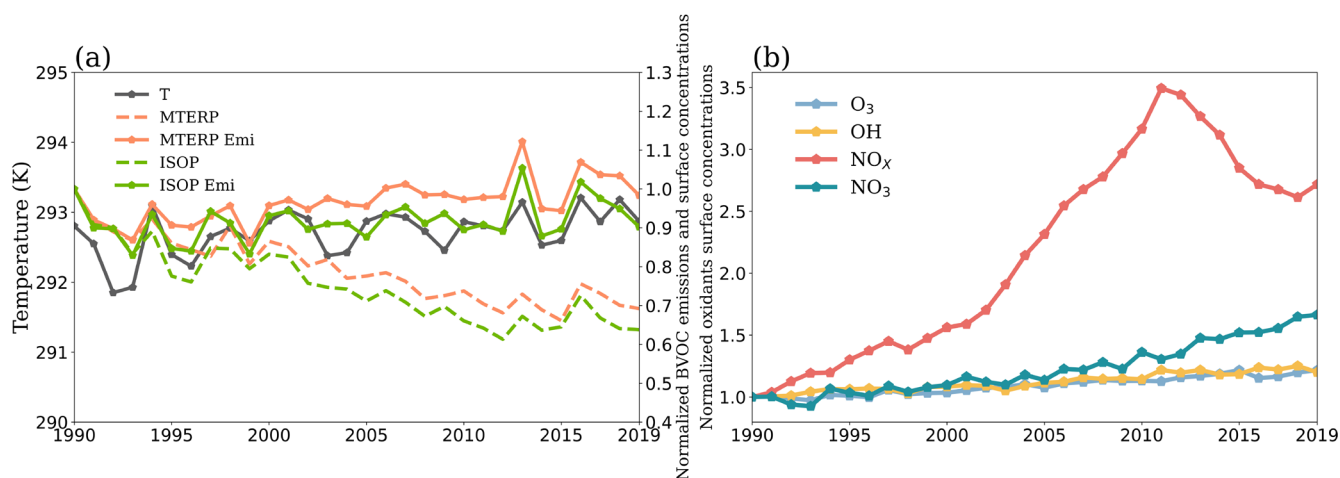
Figure 8: (a) 1990 to 2019 JJA time series of surface temperature (dark gray solid line; left Y axis; unit: K), relative changing ratio of surface concentrations for monoterpenes (orange dashed line; right Y axis; MTERP), isoprene (green dashed line; right Y axis; ISOP), monoterpenes emissions (orange solid line; right Y axis; MTERP Emi) and isoprene emissions (green solid line; right Y axis; ISOP Emi). (b) 1990 to 2019 JJA time series of relative changing ratio of surface concentrations for nitrogen oxidizes (NOx; red), ozone (O₃; pale blue), hydroxyl radical (OH; yellow), and nitrate radical (NO₃; cyan). All relative changing ratios are calculated as the concentration in each year divided by the concentration in 1990.

2. The paper shows the trend of VOC and S/IVOC emissions. However, these organic compounds have substantially different SOA formation potentials. As a result, it is unclear which of these compounds dominate the trend of ASOA abundance. A sensitivity analysis showing the contributions of these precursors to SOA would be helpful.

Response: Thanks for the suggestion. To determine which compound dominated the trend in ASOA, we conducted two sensitivity simulations for year 1990 and 2019 by turning off anthropogenic IVOC and SVOC emissions, respectively. The simulation results showed that in 1990, SVOC accounted for 51.9% of the total ASOA, dominating its formation (Fig. S10(c)). The contribution gradually dropped to 31.8% by 2019 (Fig. S10(d)) due to emission reduction (Fig. 6(b)). Over the study period, both the absolute concentration and relative contribution of IVOC increased. Notably, the significant increase in AVOC emissions during the study period also enhanced its

contribution to SOA (Fig. 6(a)). Therefore, even though SVOC emissions decreased, the rise in AVOC emissions was able to offset this decline and continued to drive the upward trend in ASOA.
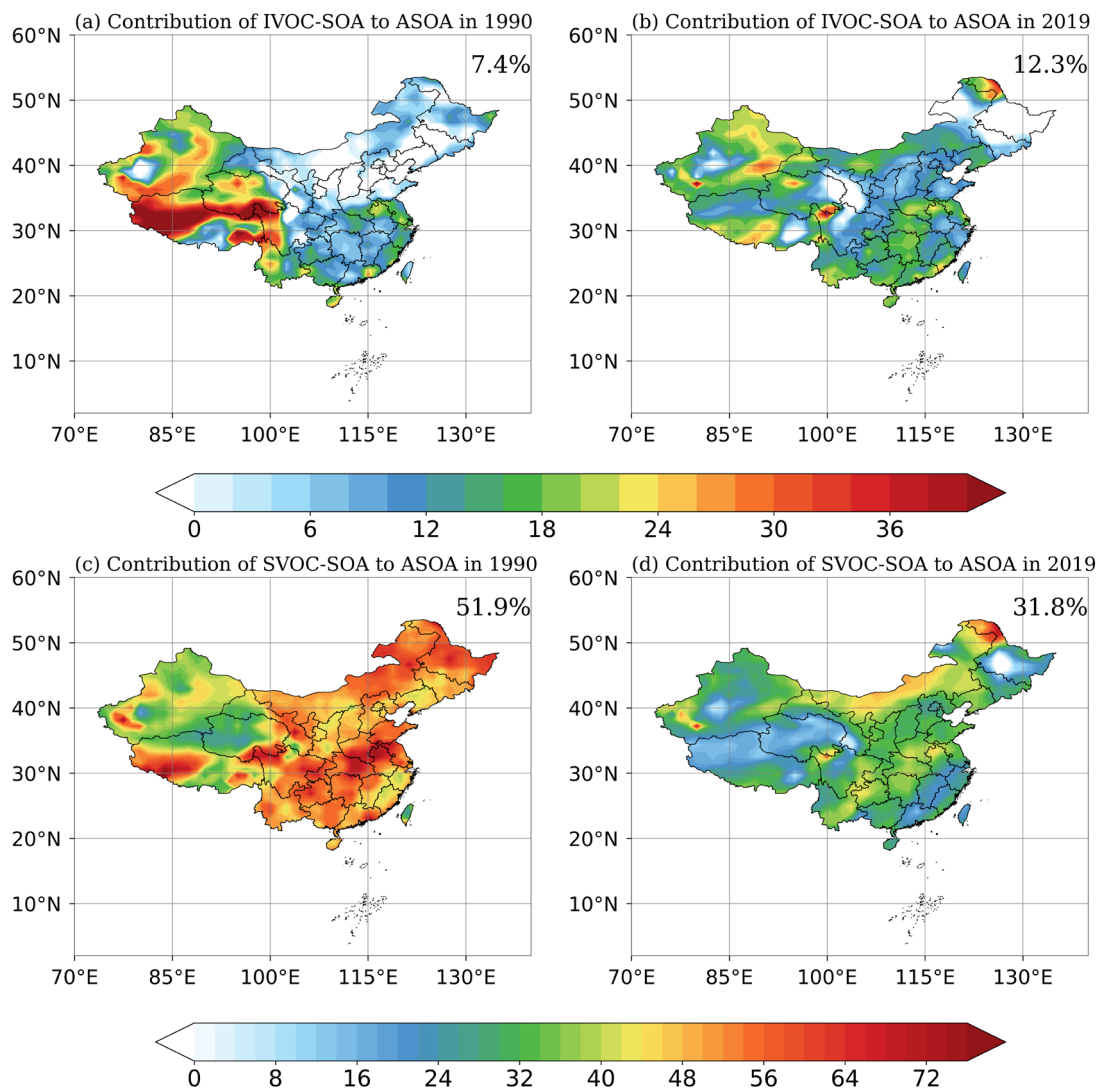
Figure S10. (a-b) Contribution of secondary organic aerosols derived from intermediate-volatile organic compounds (IVOC-SOA) to secondary organic aerosols from anthropogenic sources (ASOA) in 1990 and 2019 (unit: %). (c-d) Contribution of secondary organic aerosols derived from semi-volatile organic compounds (SVOC-SOA) to secondary organic aerosols from anthropogenic sources (ASOA) in 1990 and 2019 (unit: %). Spatial averages are shown in the upper right corner.
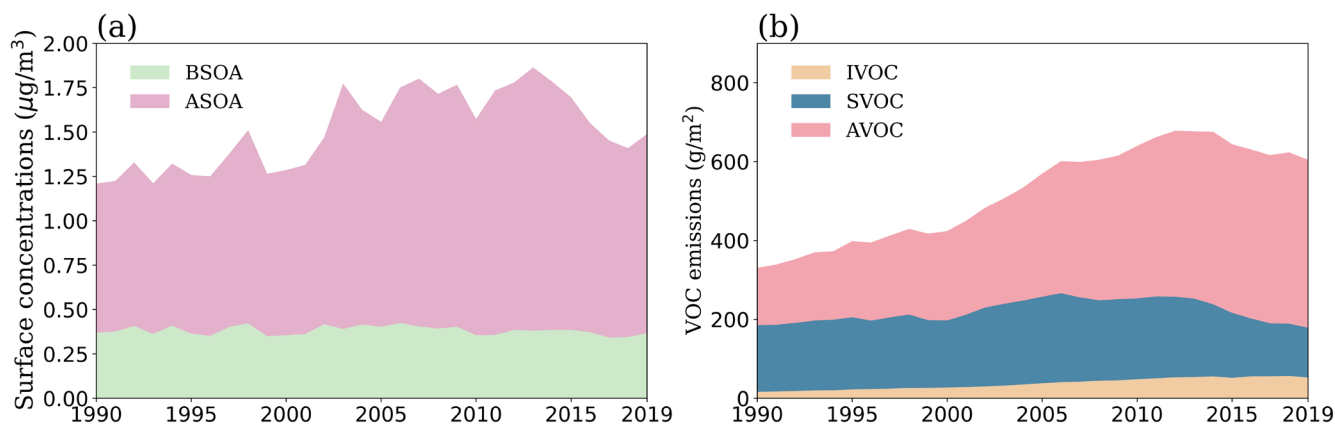
Figure 6: (a) Interannual variations in modelled average surface concentrations of secondary organic aerosols from anthropogenic sources (ASOA) and biogenic sources (BSOA) (unit: μg m⁻³). (b) Interannual variations in emissions of aromatics (AVOC), semi-volatile organic compounds (SVOC), and intermediate-volatile organic compounds (IVOC) (unit: g m⁻² mon⁻¹).
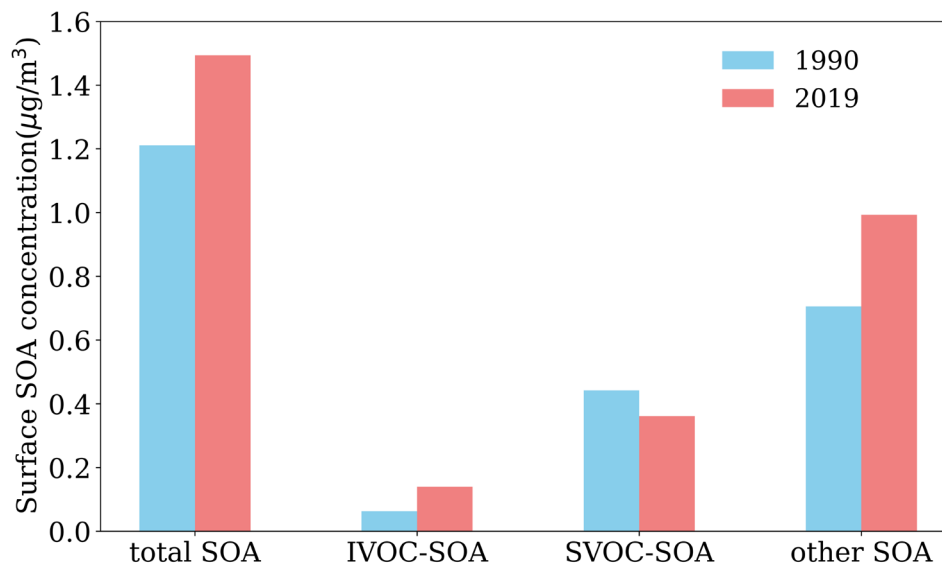
Figure S12. Annual surface secondary organic aerosols (SOA) concentrations (unit: μg m⁻³) for total SOA, secondary organic aerosols derived from intermediate-volatile organic compounds (IVOC-SOA), secondary organic aerosols derived from semi-volatile organic compounds (SVOC-SOA), and other SOA in 1990 (blue) and 2019 (red).

3. Figure 8b shows that O3 in China did not increase significantly, which seems contrary to my understanding. Is this due to the nation-wide average vs. urban areas, where monitoring sites are located?

Response: Thanks for the comment. Figure 8b shows the normalized oxidant mixing ratios instead of the absolute concentrations. Due to the large differences in the absolute concentrations of the four oxidants, directly plotting them on the same graph may compromise the comparison. Therefore, we used the ratio calculated as the concentration in each year divided by the concentration in 1990 to demonstrate the relative changes. This allows for a clearer presentation of the trends in the different oxidants and facilitates a more effective comparison. If the analysis were based on absolute concentrations, such as examining the summer average O_3 concentrations from 2000 to 2019 (Fig.X), a significant increase can be identified. We have rephrased the caption of Fig.8b to avoid misunderstanding.
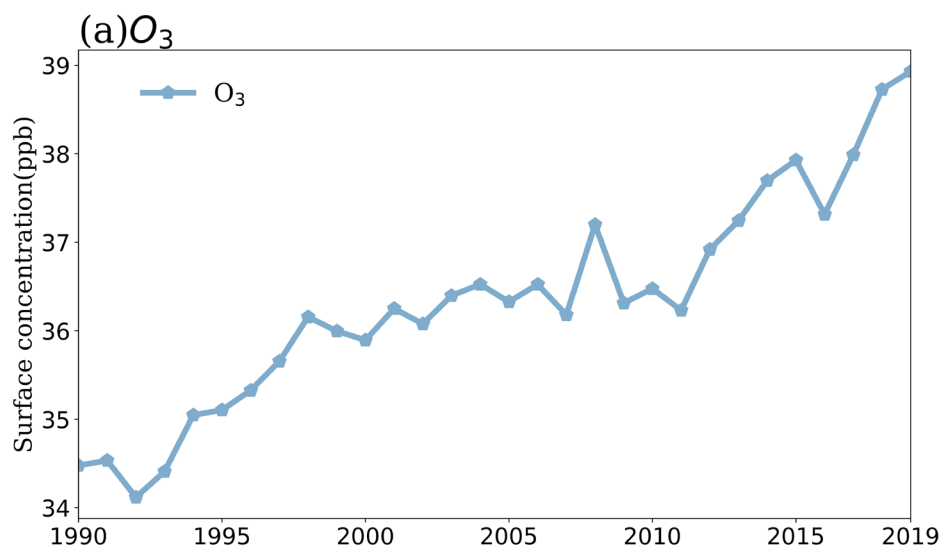
Figure X. 1990 to 2019 interannual variation of average surface ozone (O_3) (unit: ppb).

The O3 concentrations in Fig. S20 appear unreasonably high—please verify the results.

Response: Thank you for pointing this out. There was indeed a bug in plotting the O_3 concentrations. We have fixed it and replot the figure.
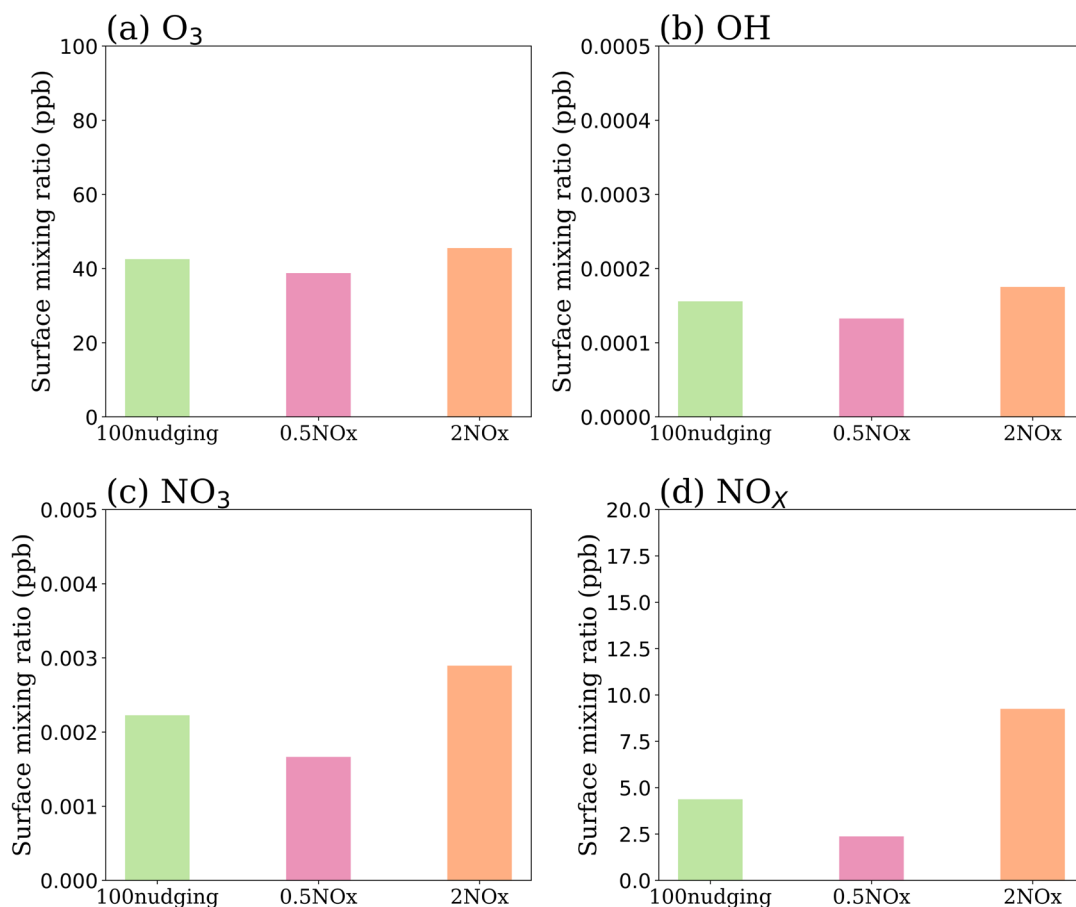
Figure S22(This figure was originally Figure S20). Surface concentrations (unit: ppb) of (a) O₃, (b) OH, (c) NO₃ and (d) NOx for July 2013 from the NOx sensitivity experiments named 100nudging (green bar), 0.5NOx (pink bar) and 2NOx (orange bar).

Additionally, Fig. 9b shows that SOA contributions from different oxidants do not change with the oxidant. For instance, all oxidants decrease in the 0.5NOx case, while SOAMT_O3 and SOAMT_NO3 increase, and SOAMT_OH decreases. Could you clarify this?

Response: Thank you for pointing this out. Fig.9(b) was mis-labelled for SOAMT_NO3 and SOAMT_OH (low NOx). We have replaced them with the correct plots (as shown below) and corrected the description in Line399-

Line406. In the 0.5NOx case, atmospheric concentrations of major oxidants, including O3, OH radicals, NO3 radicals, and NOx, show corresponding reductions with decreased NOx emissions (Fig.S22), which leads to an increase in isoprene and monoterpenes concentrations (Fig.S21). As shown in Fig. 9(b) and Fig. S21, SOAMT_O3 and SOAMT_OH (low NOx) concentrations show consistent changes with monoterpenes concentrations. However, SOAMT_NO3 and SOAMT_OH (high NOx) concentrations are more strongly influenced by the decrease in NOx emissions. In addition, decreases in SOAMT_NO3 and SOAMT_OH (high NOx) concentrations were offset by increases in SOAMT_O3 and SOAMT_OH (low NOx) concentrations, which resulted in a slight increase in total surface SOAMT concentrations in response to decreasing NOx emissions (Fig.S23(c)).
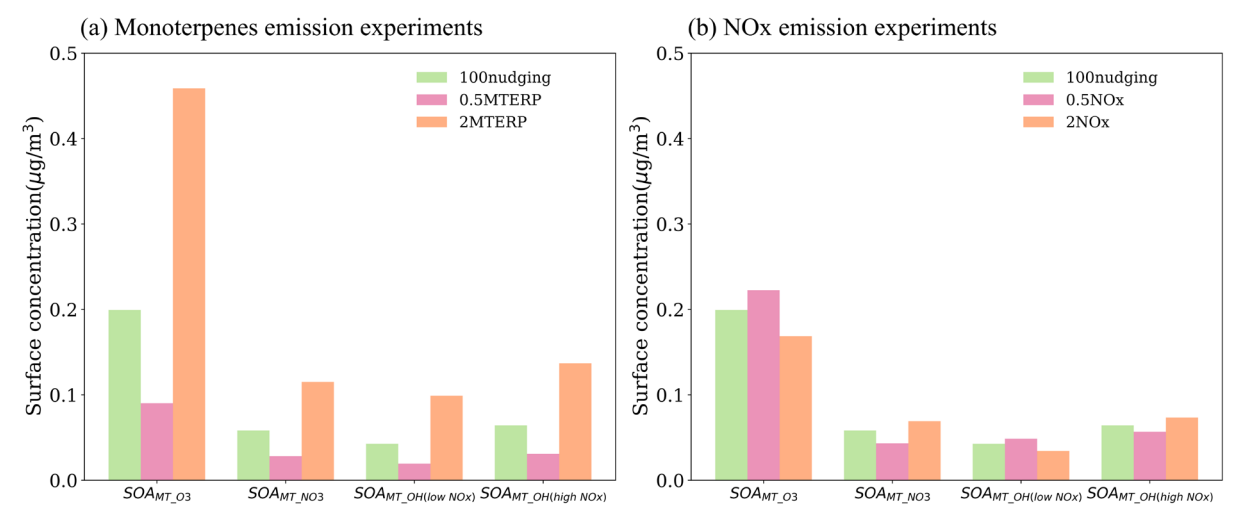
Figure 9: Surface concentrations (unit: μg m⁻³) of monoterpene-derived secondary organic aerosols (SOА$_{MT}$) compositions for July 2013 from the monoterpenes (a) and NOx (b) sensitivity experiments named 100nudging (green bar), 0.5MTERP/NOx (pink bar), and 2MTERP/NOx (orange bar).
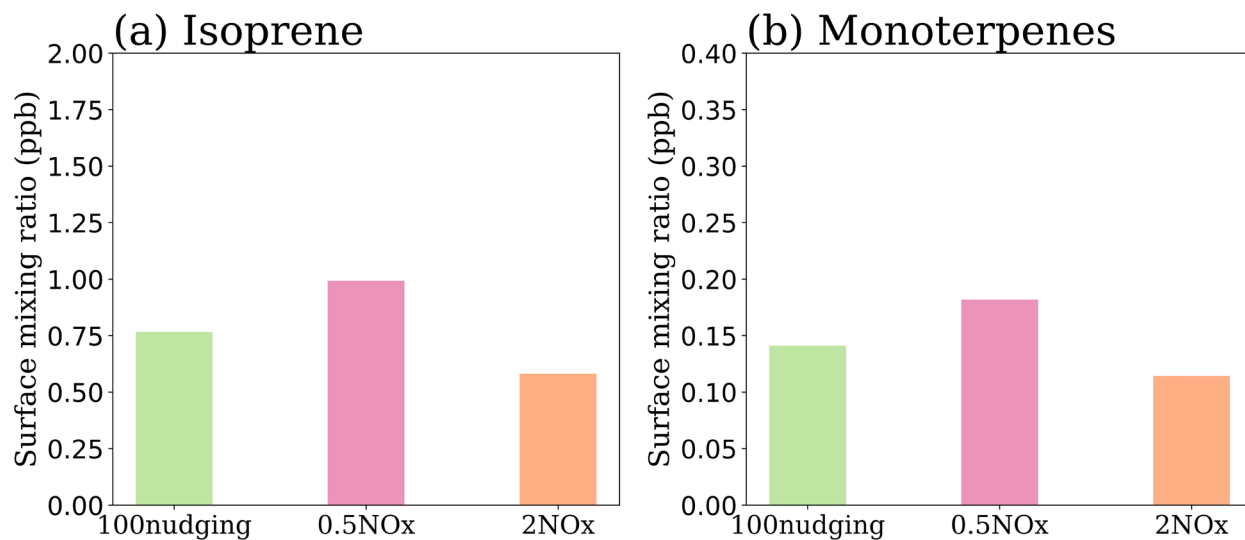
Figure S21. Surface concentrations (unit: ppb) of (a) isoprene and (b) monoterpenes for July 2013 from the NOx sensitivity experiments named 100nudging (green bar), 0.5NOx (pink bar) and 2NOx (orange bar).
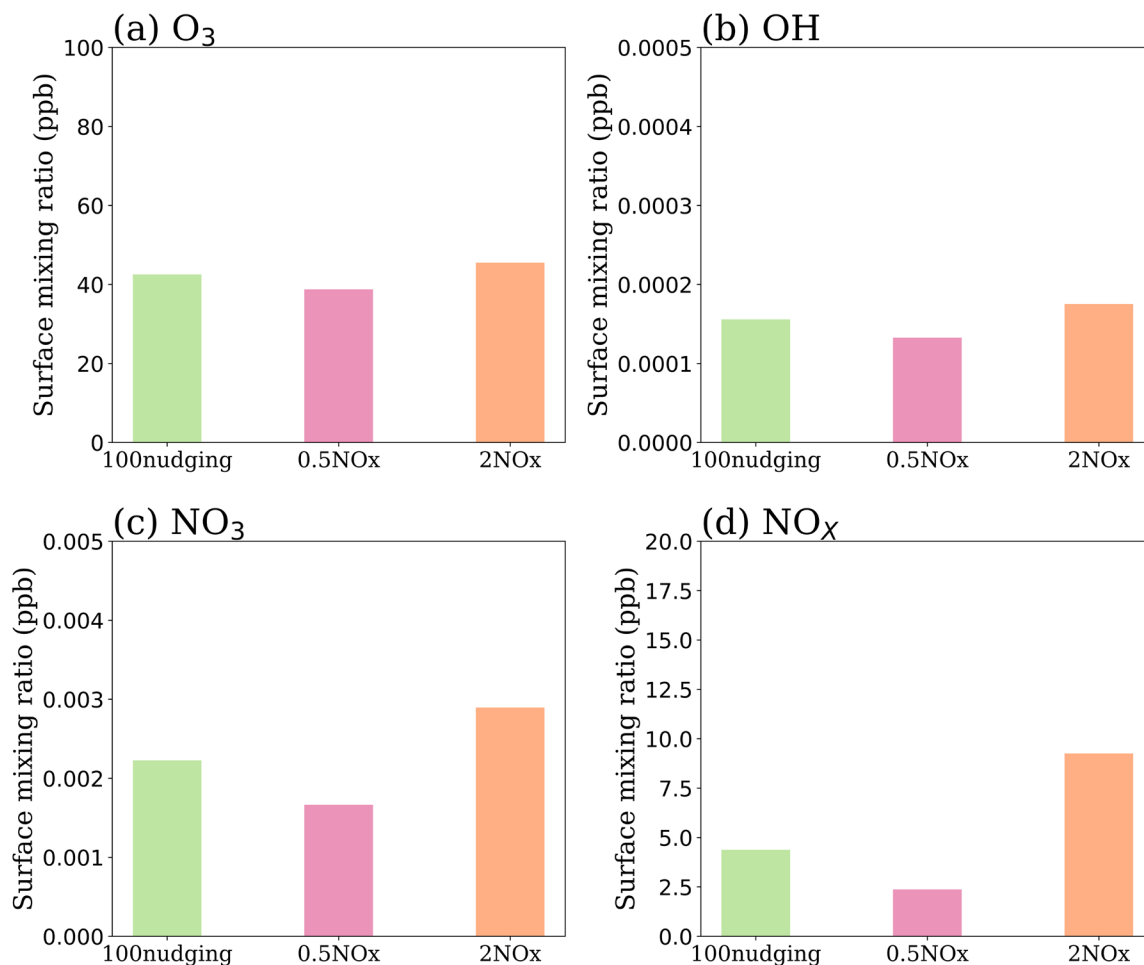
Figure S22. Surface concentrations (unit: ppb) of (a) O₃, (b) OH, (c) NO₃ and (d) NOx for July 2013 from the NOx sensitivity experiments named 100nudging (green bar), 0.5NOx (pink bar) and 2NOx (orange bar).

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