

Figure S1. Development of modeled trace gas mixing ratios in the upper tropical troposphere at 200 hPa from 2000 to 2019.

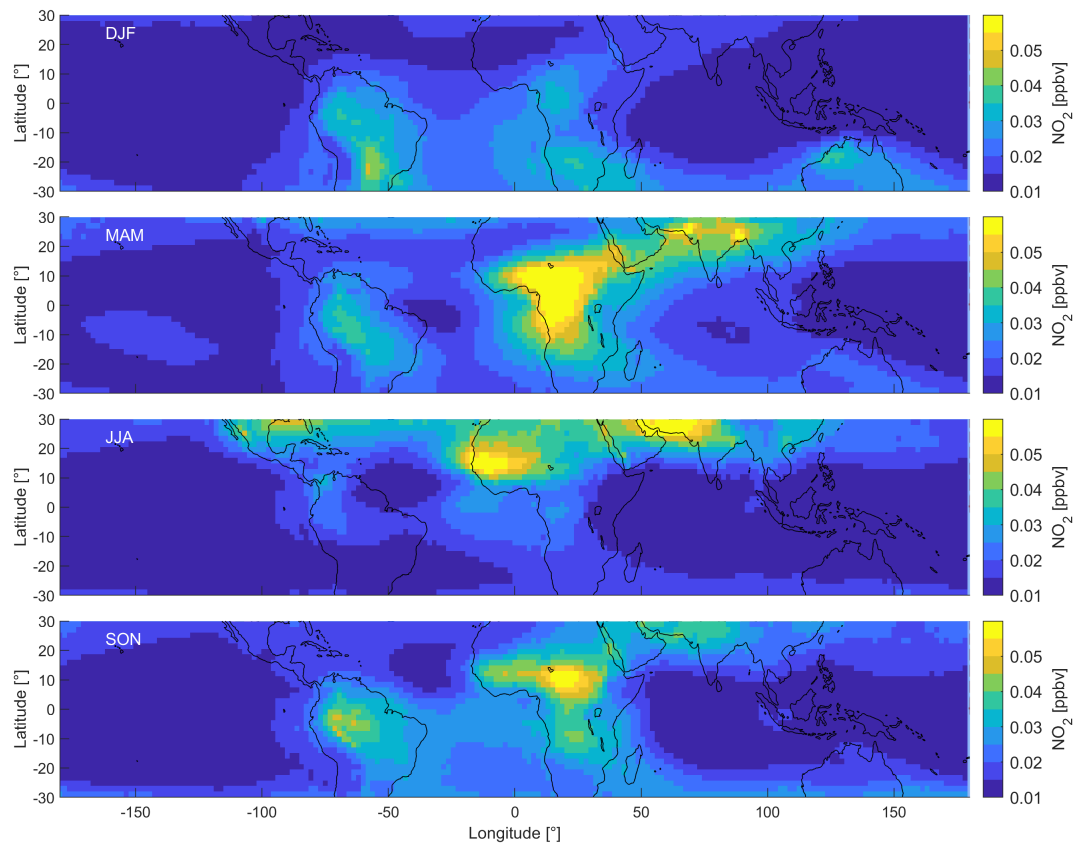


Figure S2. Distribution of NO₂ in the tropical UT between 30° S and 30° N for all periods.

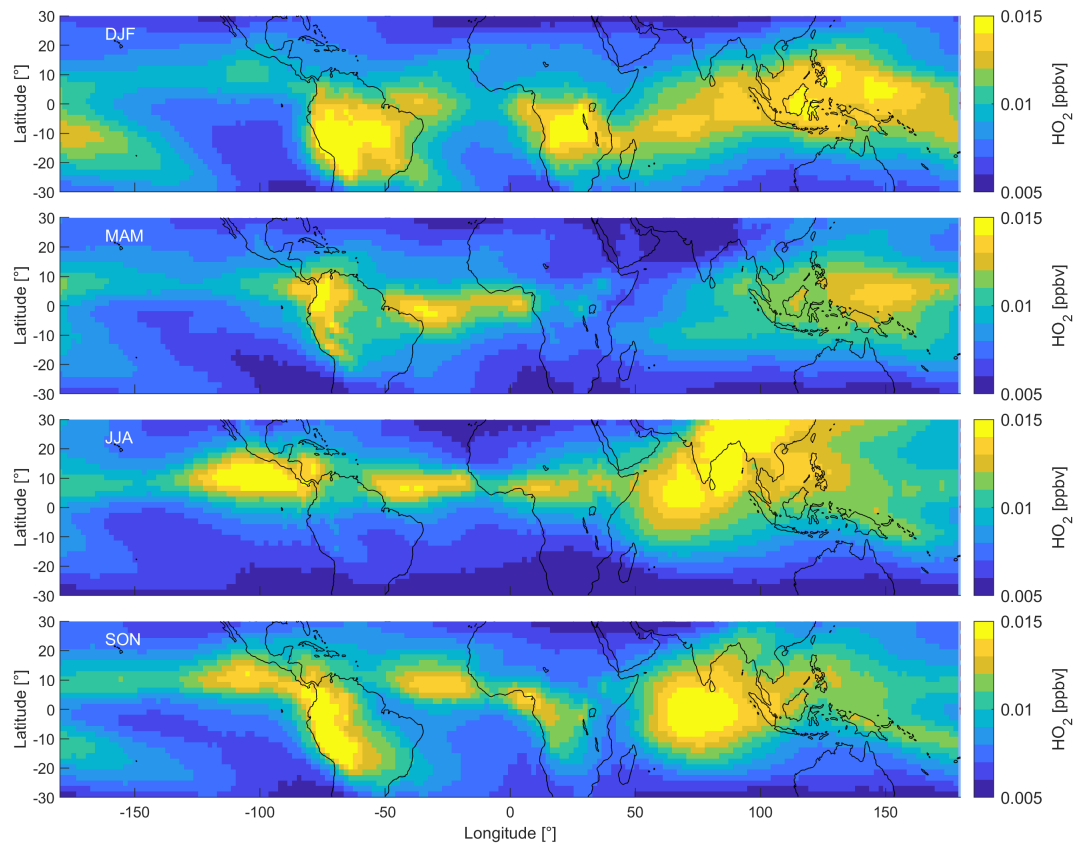


Figure S3. Distribution of HO₂ in the tropical UT between 30° S and 30° N for all periods.

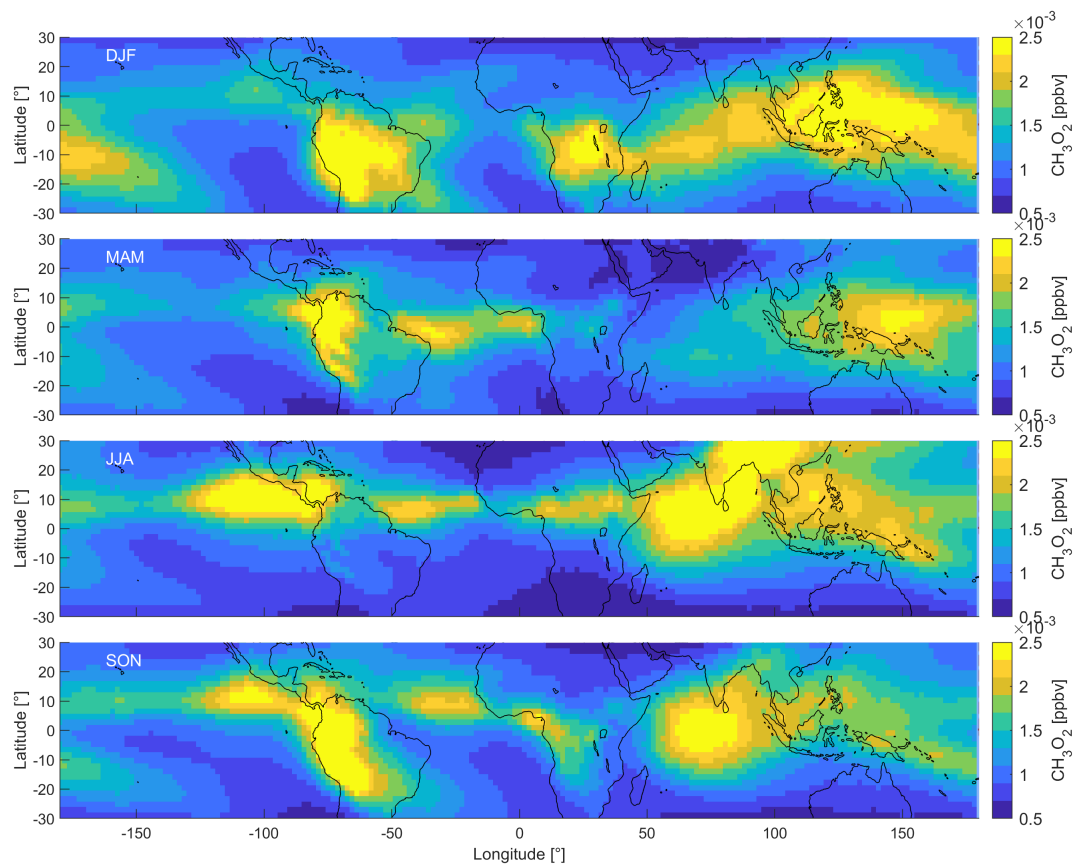


Figure S4. Distribution of CH_3O_2 in the tropical UT between 30°S and 30°N for all periods.

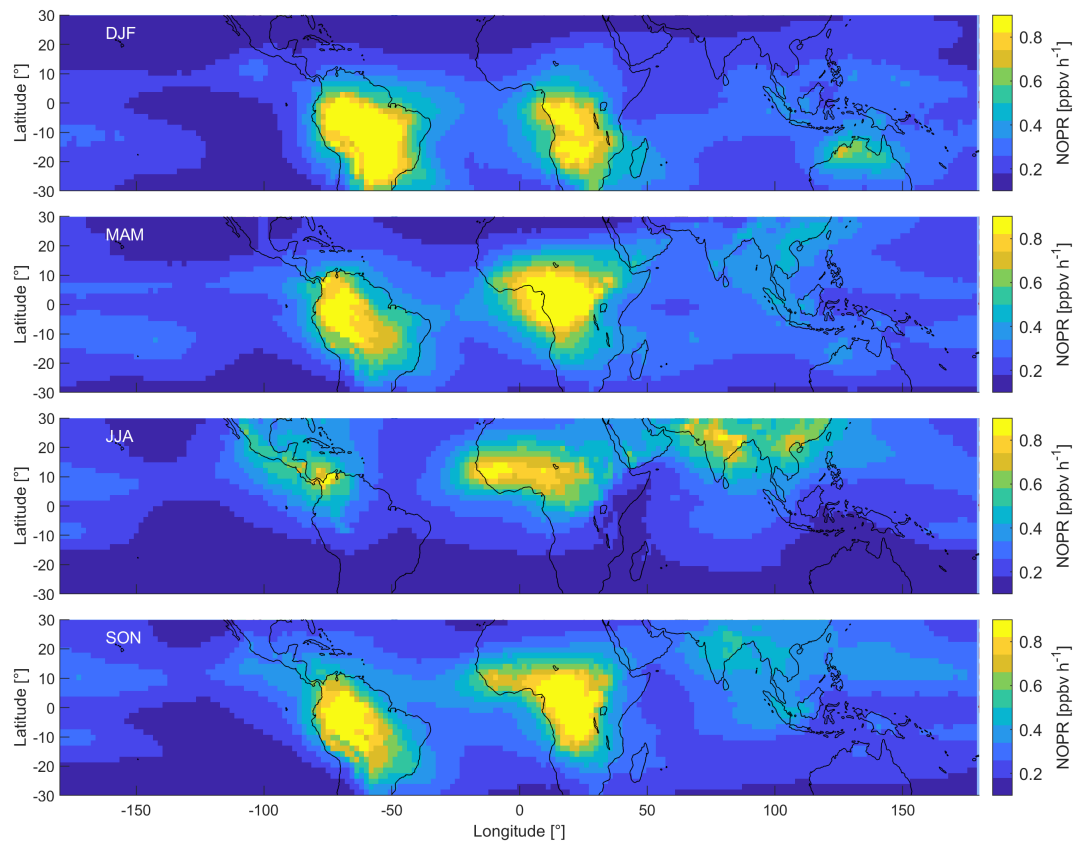


Figure S5. Distribution of NOPR in the tropical UT between 30° S and 30° N for all periods.

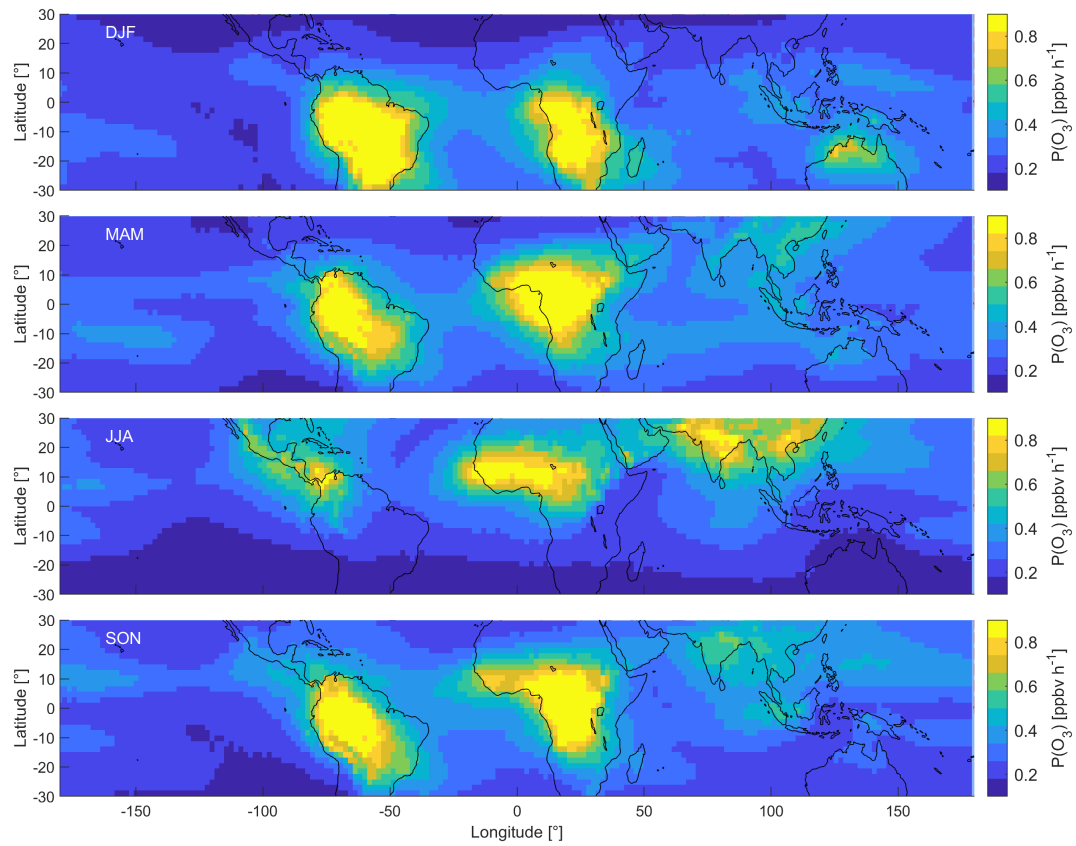


Figure S6. Distribution of $P(O_3)$ in the tropical UT between 30° S and 30° N for all periods.

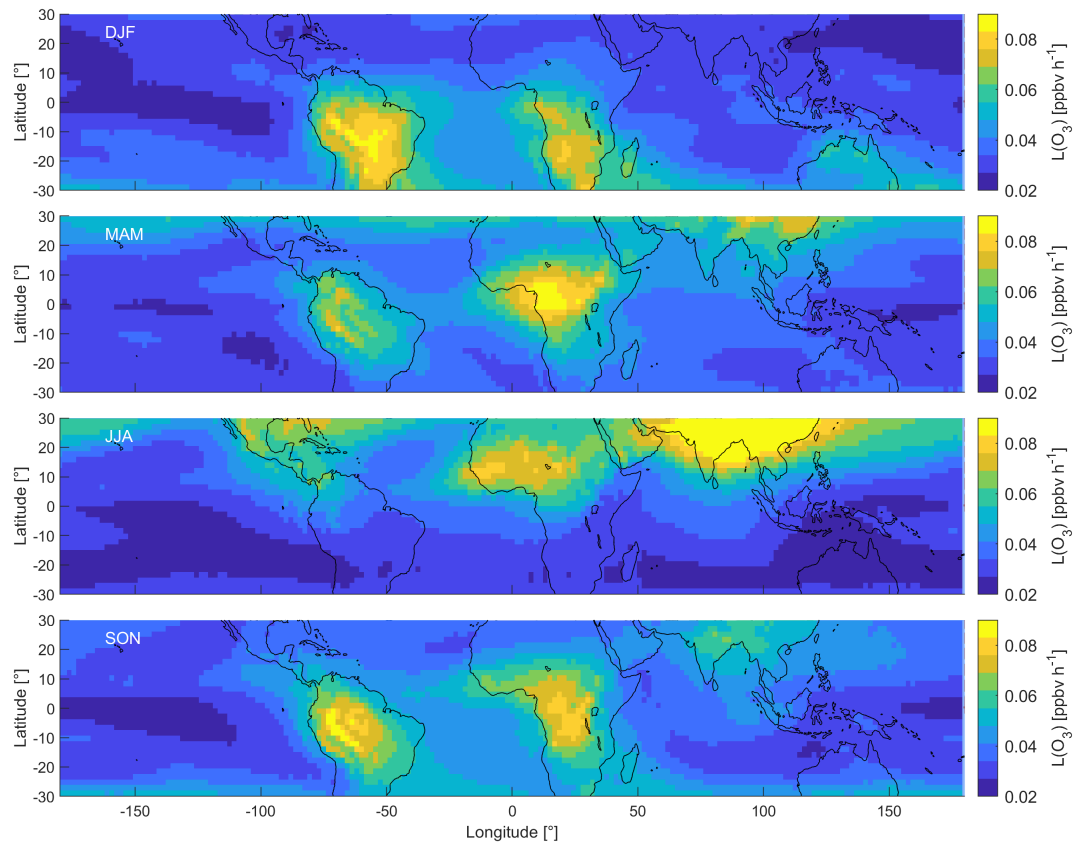


Figure S7. Distribution of $L(O_3)$ in the tropical UT between 30° S and 30° N for all periods.

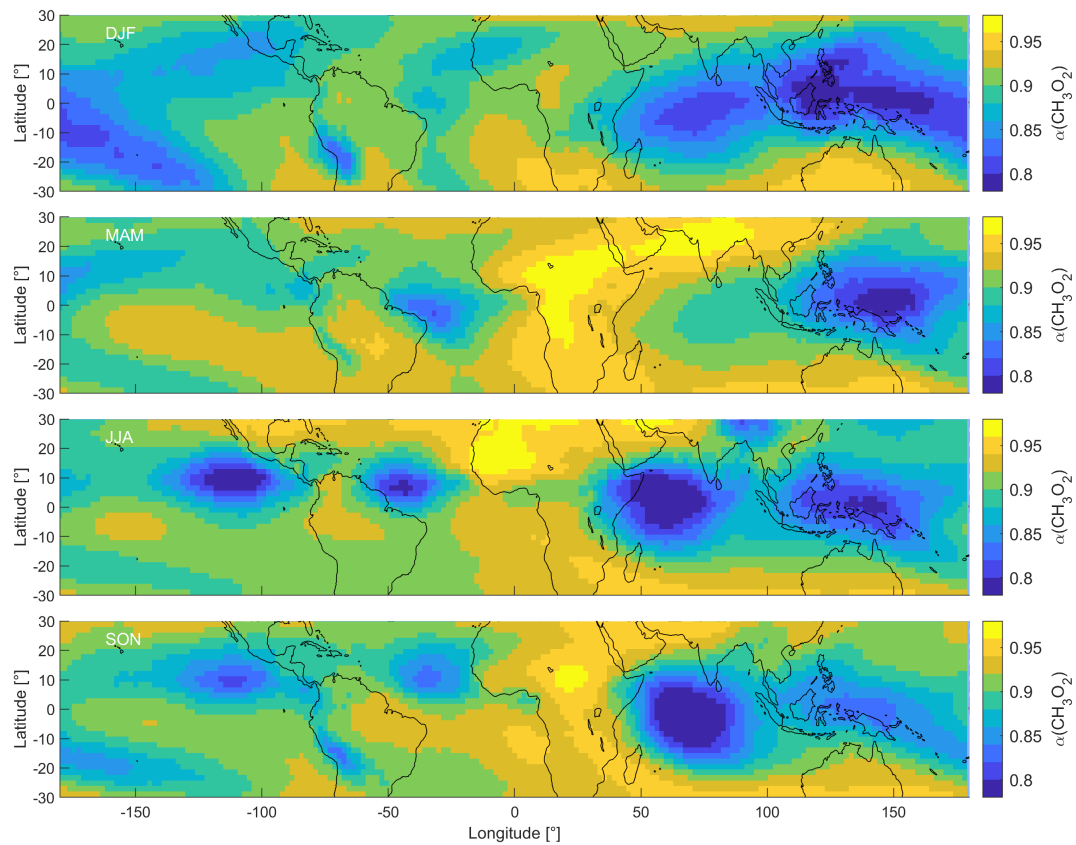


Figure S8. Distribution of $\alpha(\text{CH}_3\text{O}_2)$ in the tropical UT between 30° S and 30° N for all periods.

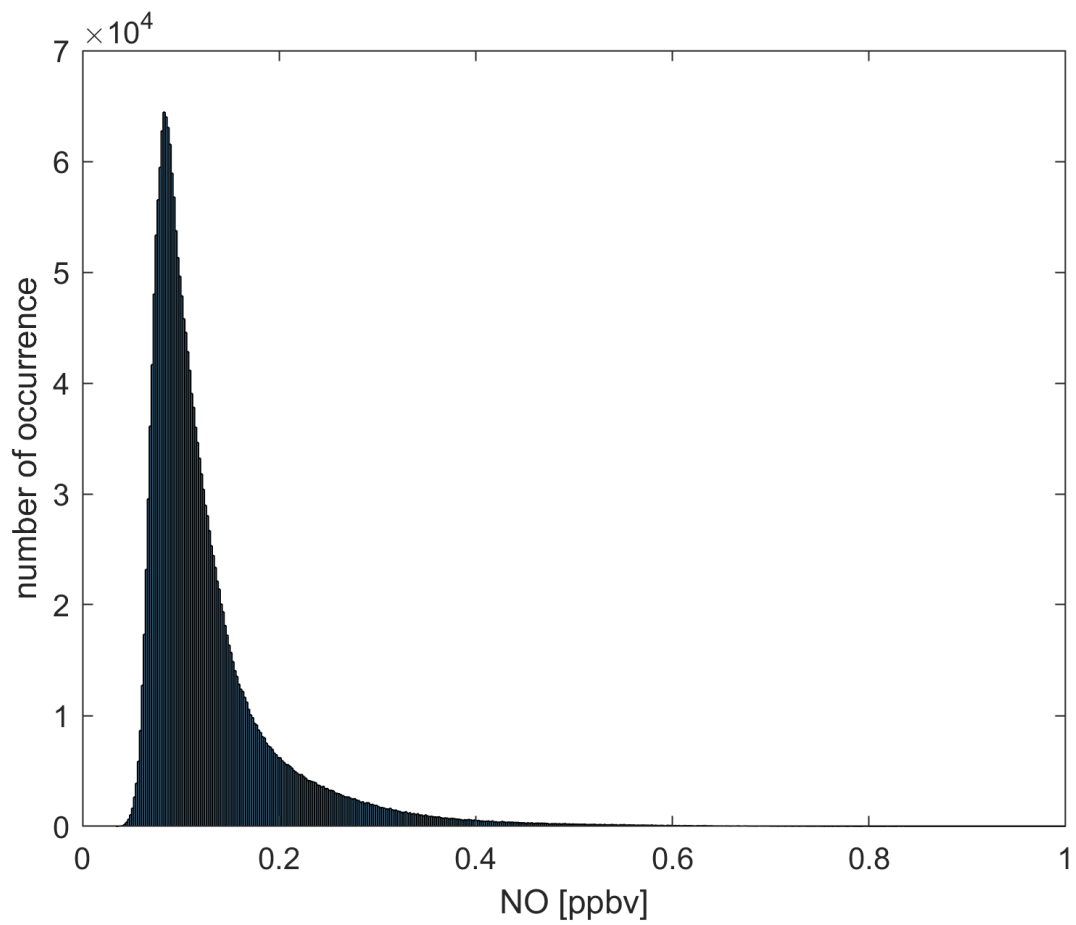


Figure S9. Frequency distribution of NO mixing ratios. 99.6 % of the data points show ≤ 0.5 ppbv NO.

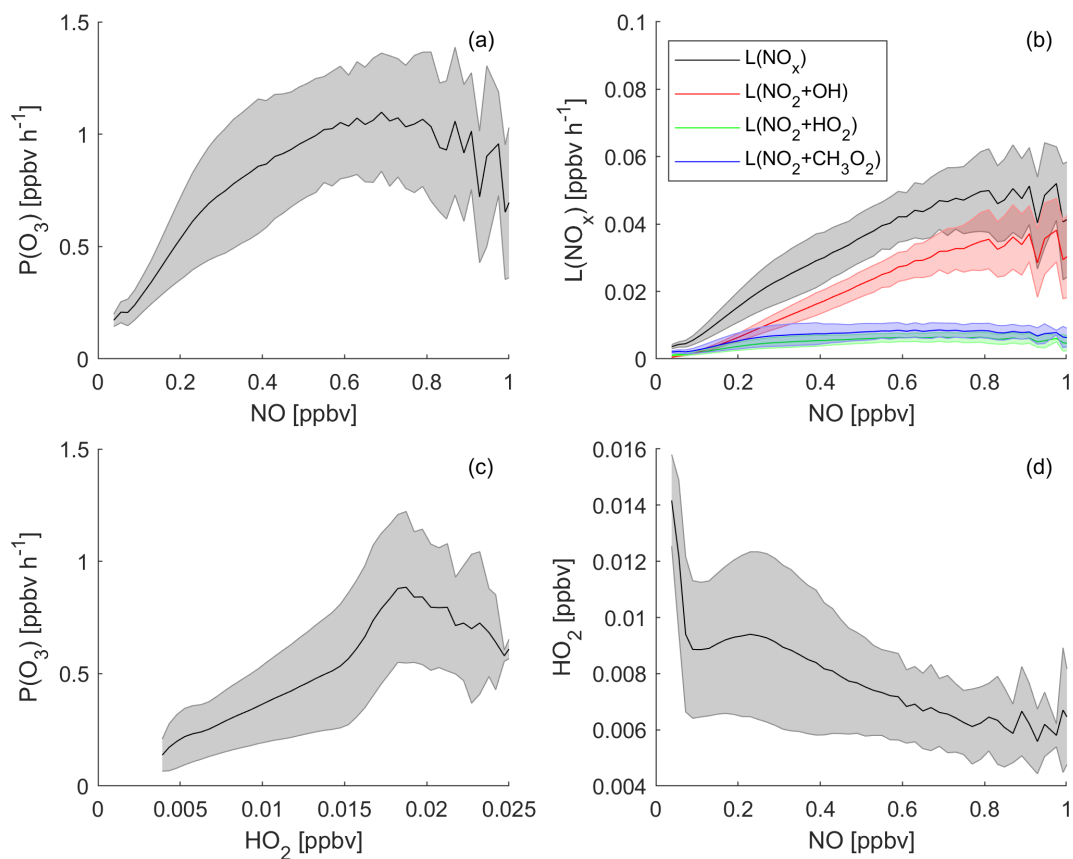


Figure S10. (a) $P(\text{O}_3)$ binned to NO mixing ratios up to 1 ppbv NO, (b) NO_x loss binned to NO mixing ratios and subdivided into loss via OH, HO_2 and CH_3O_2 , (c) $P(\text{O}_3)$ binned to HO_2 mixing ratios and (d) HO_2 binned to NO mixing ratios. Lines show averages of all data points and gray shades present the 1σ standard deviation.

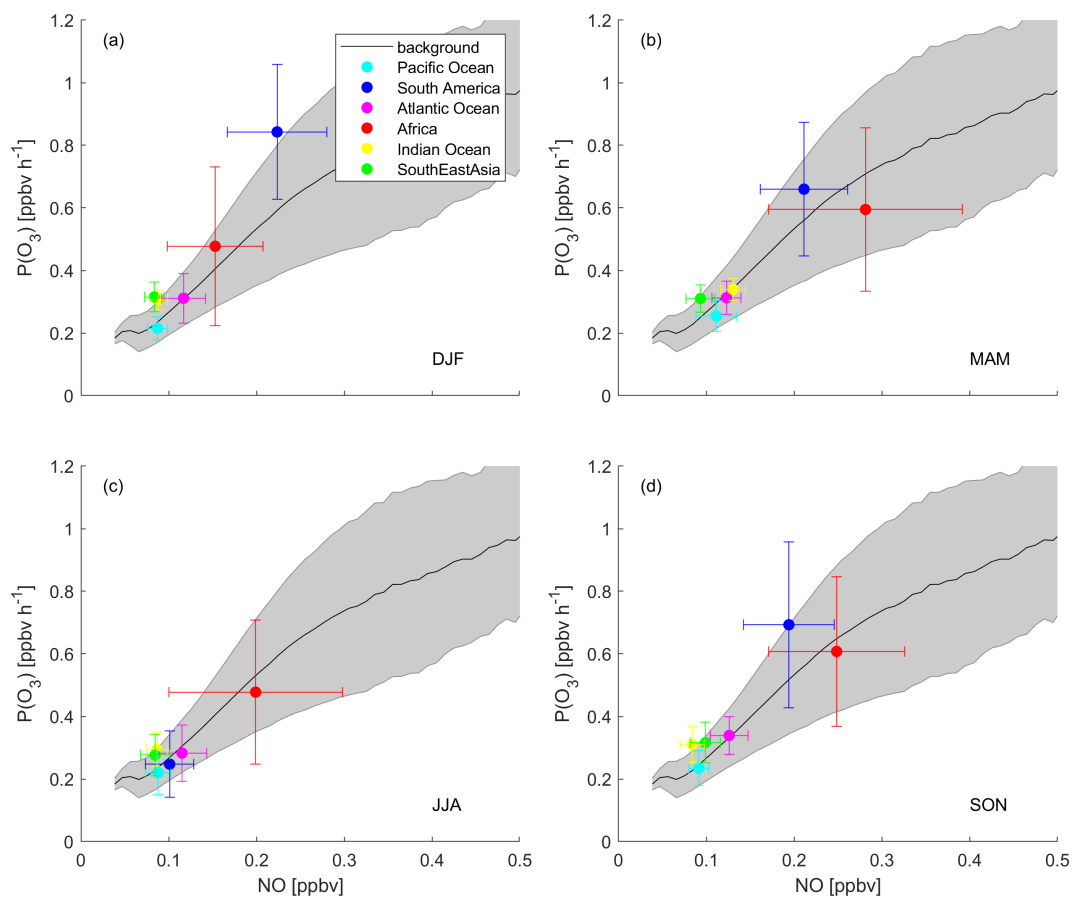


Figure S11. $P(O_3)$ binned to NO mixing ratios, distinguishing the four periods. Black lines show averages of all data points and gray shades present the 1 σ standard deviation. Colored data points show the averages for the indicated areas with the 1 σ standard deviation.

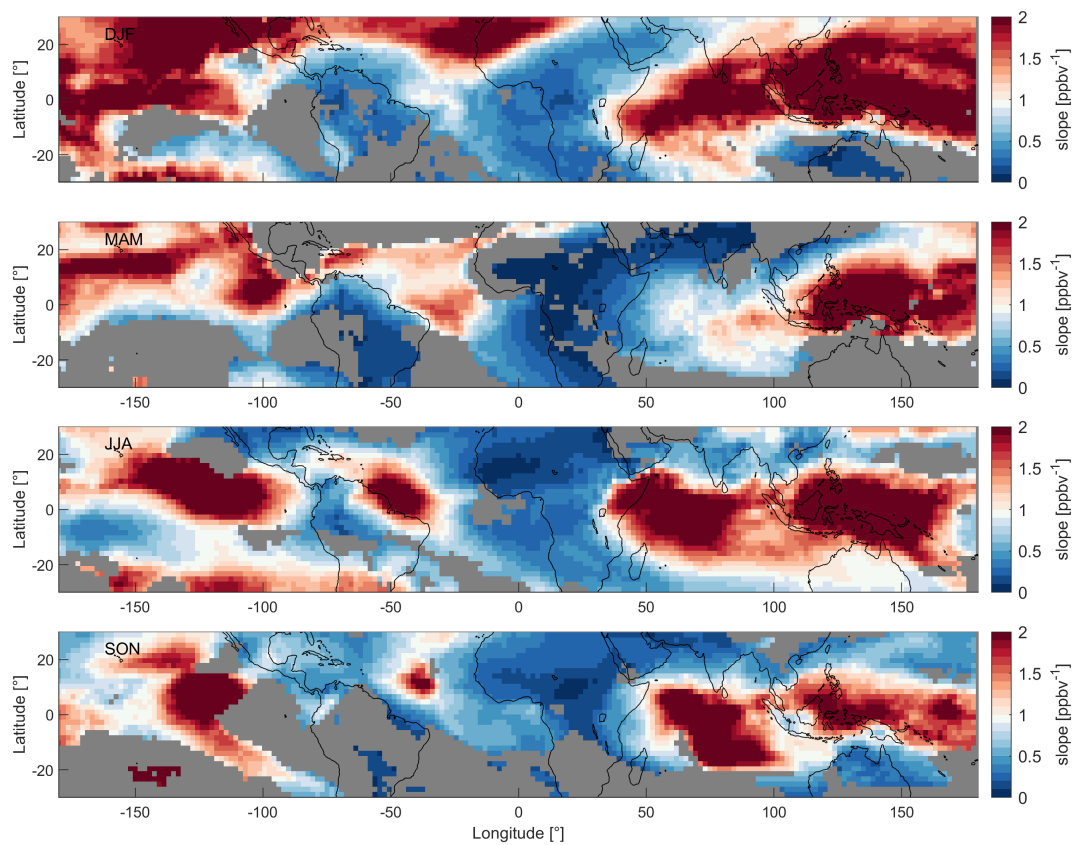


Figure S12. Map of the tropical UT between 30° S and 30° N for each period colored by the slopes of NO vs $\alpha(\text{CH}_3\text{O}_2)$ of the data in model grid regions. Red colors indicate NO_x and blue colors VOC sensitive regimes. For gray areas the R^2 of the fit is below 30 %.

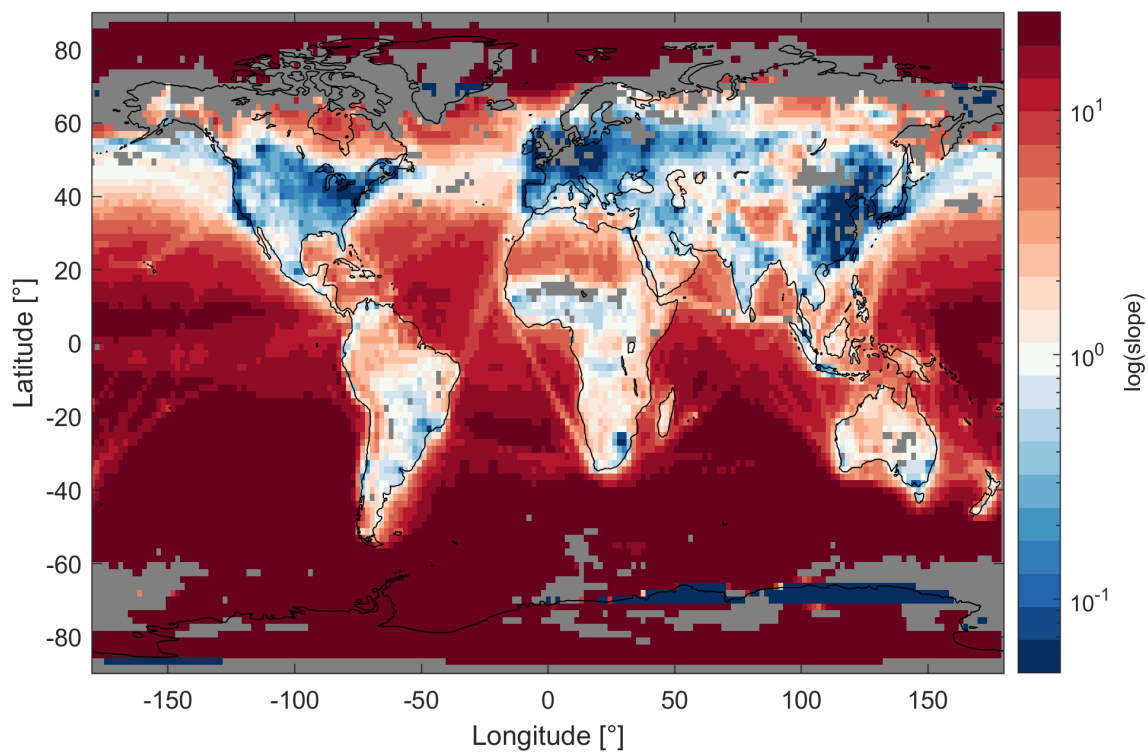


Figure S13. Determination of the dominant chemical regime at the surface for all latitudes, using the slope of NO vs $\alpha(\text{CH}_3\text{O}_2)$ as indicator – showing the annual average. Red colors indicate NO_x and blue colors VOC sensitive regimes. For gray areas the R^2 of the fit is below 30 %.

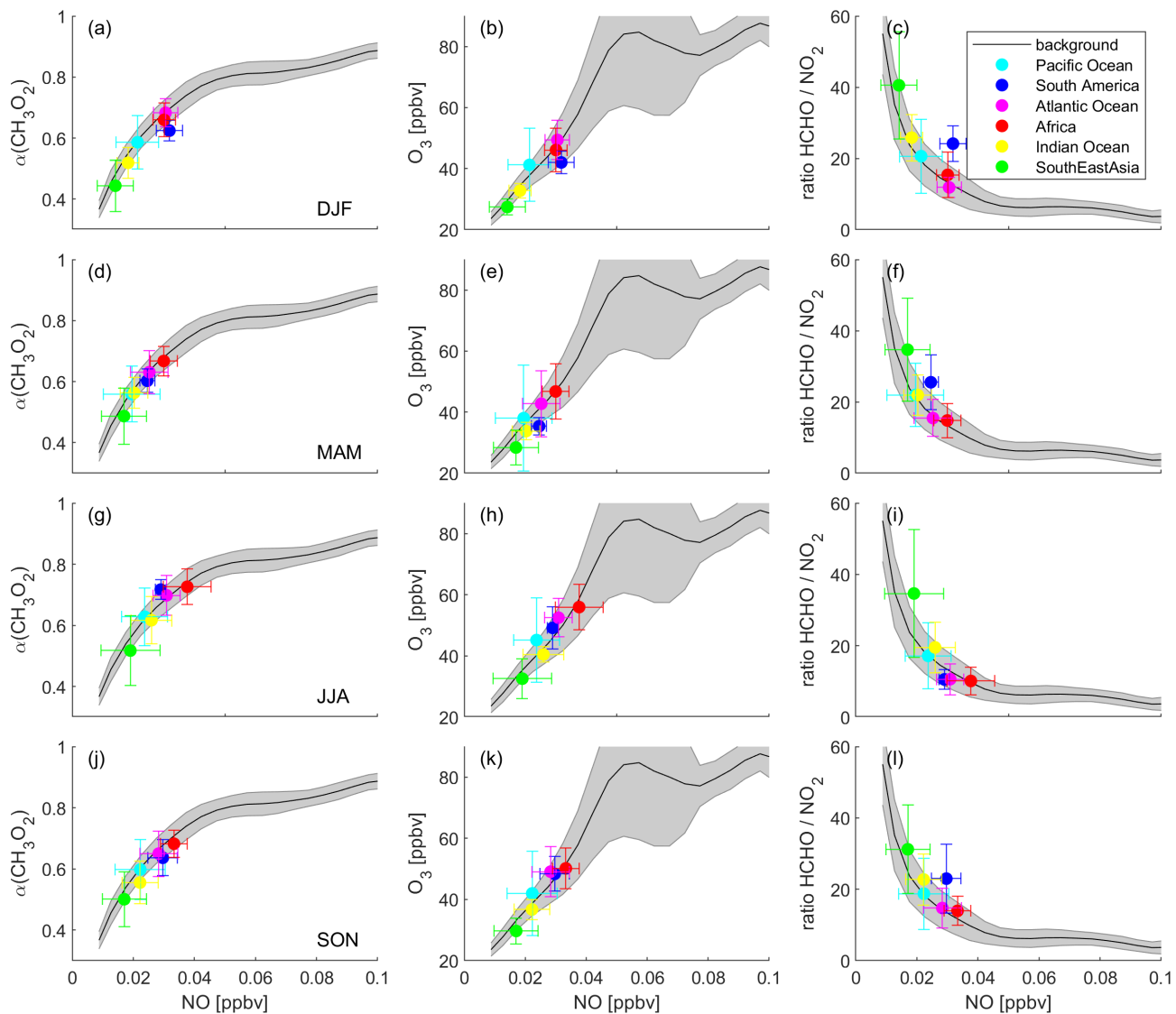


Figure S14. Determination of the dominant chemical regime in the tropical UT in the no lightning scenario via $\alpha(\text{CH}_3\text{O}_2)$, O_3 and the HCHO/NO_2 ratio binned to NO mixing ratios for the four periods. Black lines show averages of all data points and gray shades present the 1σ standard deviation. Colored data points show the averages for the indicated areas with the 1σ error.

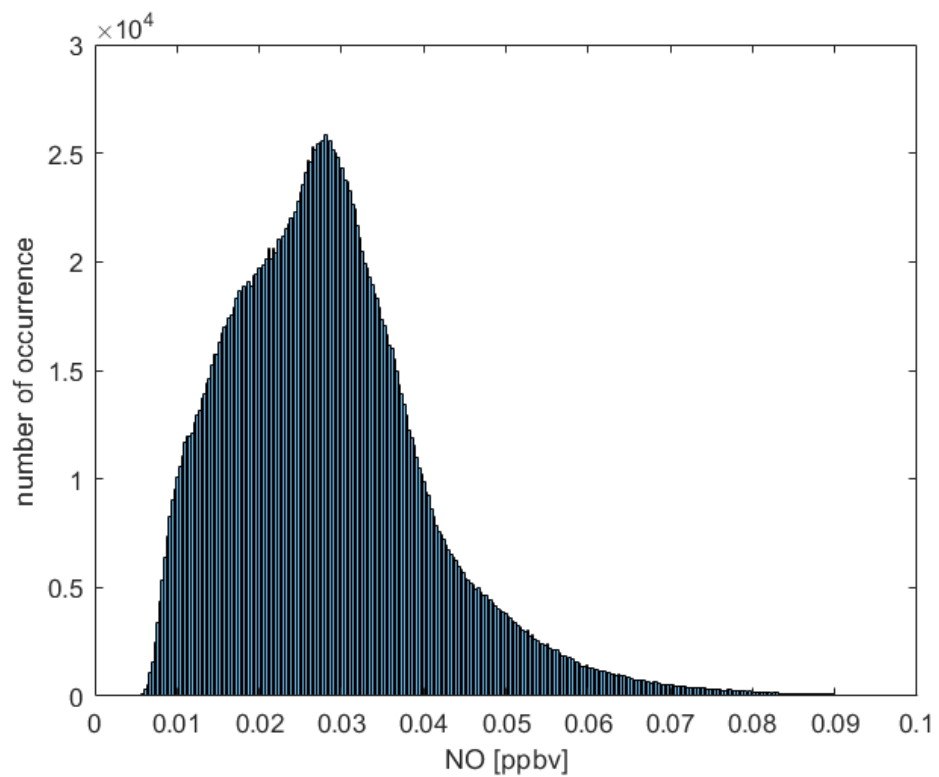


Figure S15. Frequency distribution of NO mixing ratios in the no lightning scenario.

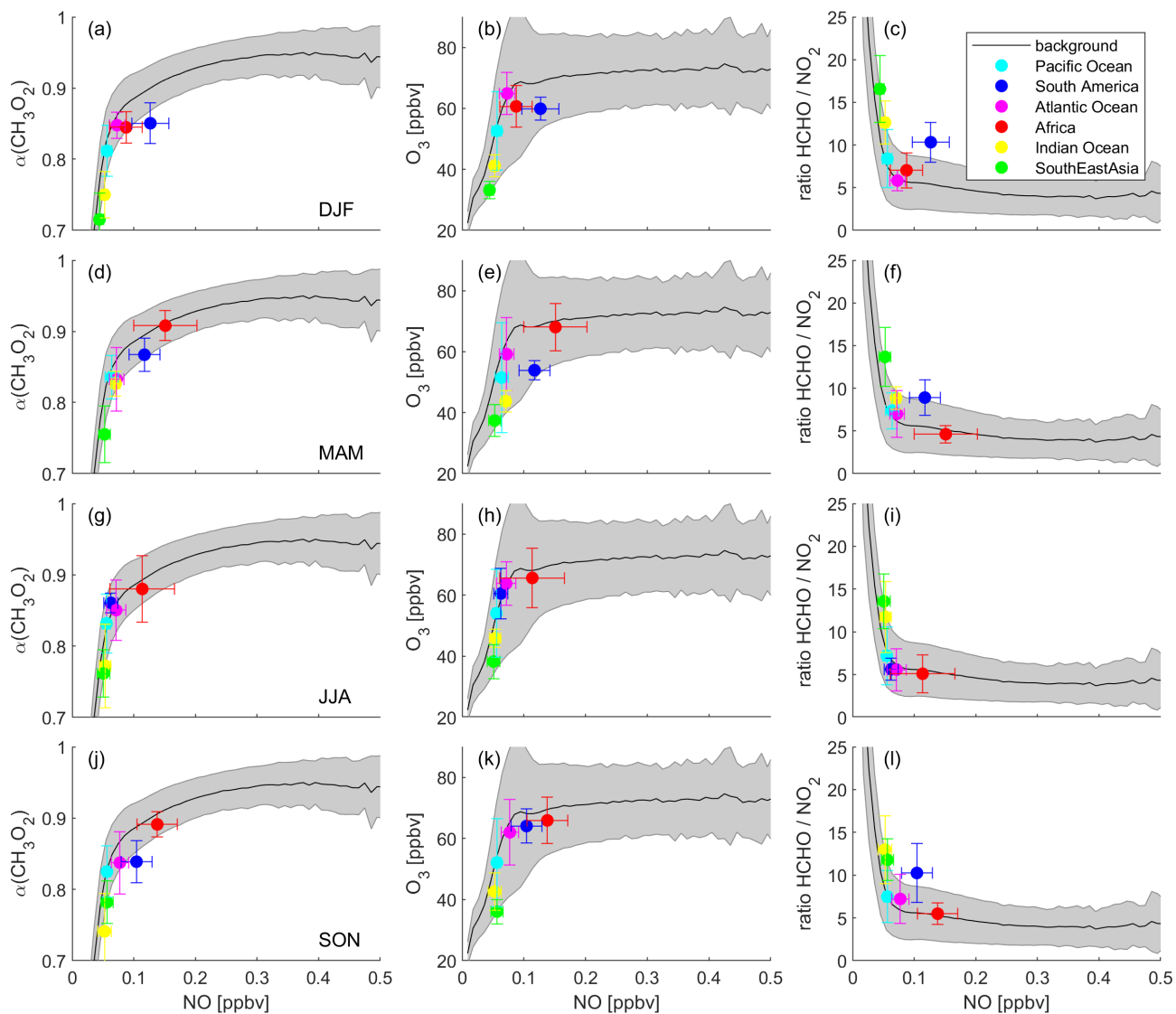


Figure S16. Determination of the dominant chemical regime in the tropical UT in the scenario with halved lightning via $\alpha(\text{CH}_3\text{O}_2)$, O_3 and the HCHO/NO_2 ratio binned to NO mixing ratios for the four periods. Black lines show averages of all data points and gray shades present the 1σ standard deviation. Colored data points show the averages for the indicated areas with the 1σ standard deviation.

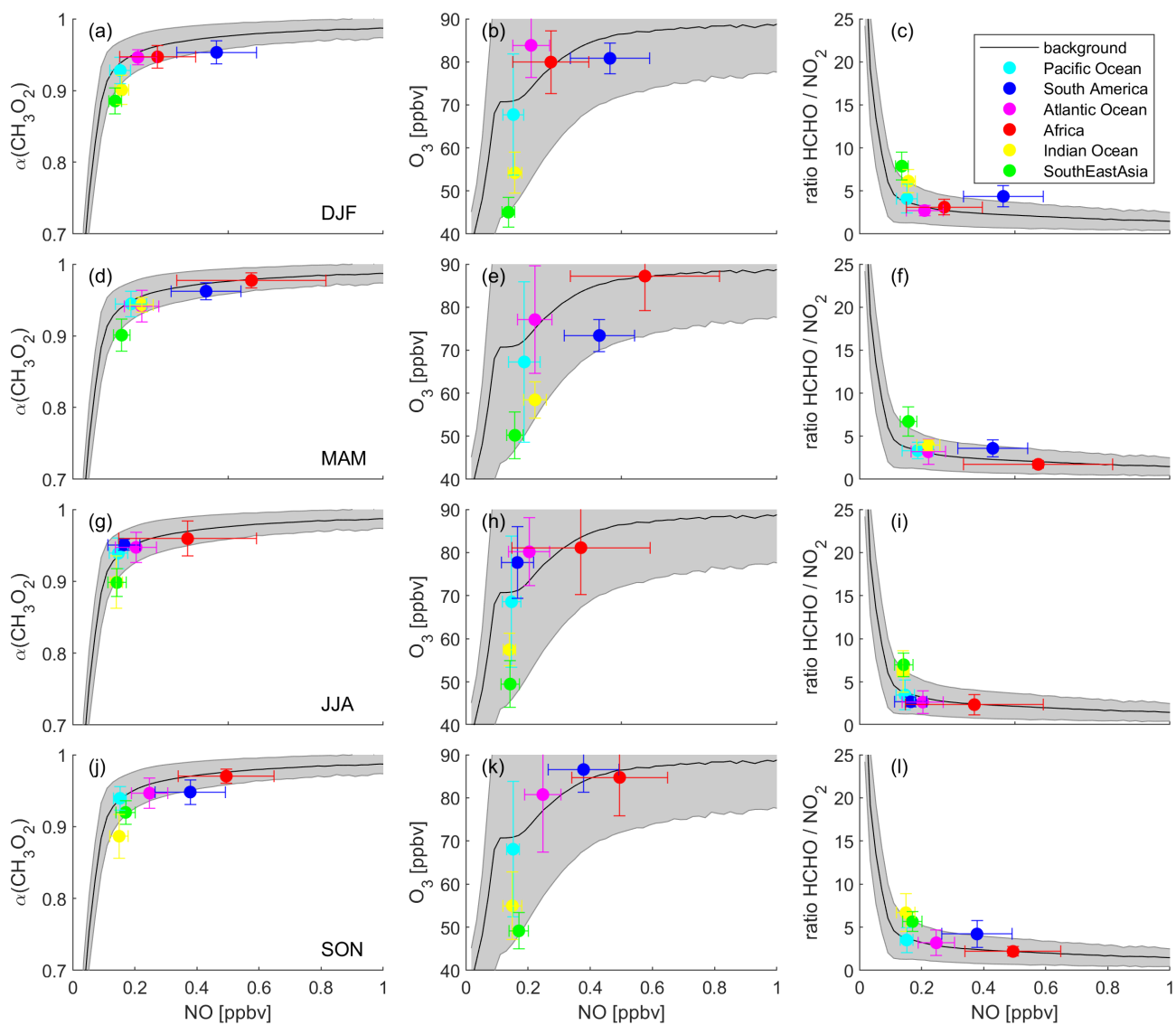


Figure S17. Determination of the dominant chemical regime in the tropical UT in the scenario with doubled lightning via $\alpha(\text{CH}_3\text{O}_2)$, O_3 and the HCHO/NO_2 ratio binned to NO mixing ratios for the four periods. Black lines show averages of all data points and gray shades present the 1σ standard deviation. Colored data points show the averages for the indicated areas with the 1σ standard deviation.

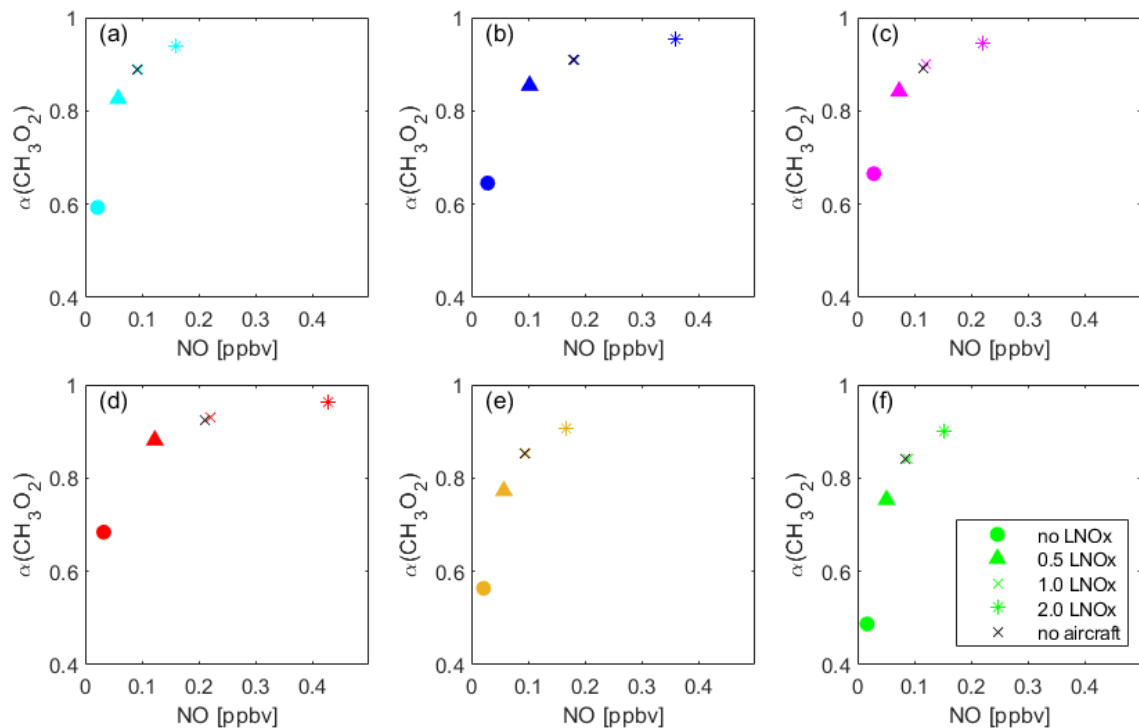


Figure S18. Overview of the impact of lightning NO_x on the dominant regime. The data points present the average NO vs $\alpha(\text{CH}_3\text{O}_2)$ of all data points located in each of the six regions (a) Pacific Ocean, (b) South America, (c) Atlantic Ocean, (d) Africa, (e) Indian Ocean, (f) South East Asia for the baseline scenario (crosses), excluding lightning (circles), halved lightning (triangles) and doubled lightning (asterisks).