Ozone trends and drivers at a Southern Hemisphere background site in Chile

Supplementary Material

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Population and infrastructure around Tololo

The population of the La Serena and Coquimbo conurbation has more than doubled between the census 1992 and 2024 (https://www.ine.gob.cl/), reaching today roughly 543 k inhabitants (https://www.bcn.cl). Smaller towns of more than 10 k inhabitants are situated nearby: Vicuña (20 km NE), and Andacollo (30 km SW). Vicuña grew 41% from 1992 to 2024 having today ca. 31 k inhabitants, and Andacollo decreased by 3% over the same period, having today ca. 12 k inhabitants. Finally, Ovalle (60 km SW) grew by 46% between 1992 and 2024, and today has a population of ca. 124 k inhabitants. Also, the number of motor vehicles has substantially grown between 1992 and present, reaching today ca. 298 k units in La Serena and Coquimbo. However, due to the implementation of standards and technological changes in Chile, vehicles emissions are estimated to be similar (NOx) or much lower (CO, VOC) between 1992 and 2020 (Osses et al., 2022). Figure S 1 shows population density as well as infrastructure in the surroundings of Tololo. Available air quality monitoring stations run by the Ministry of Environment at Coquimbo-La Serena and Andacollo only measure particles mass concentrations (https://sinca.mma.gob.cl/).

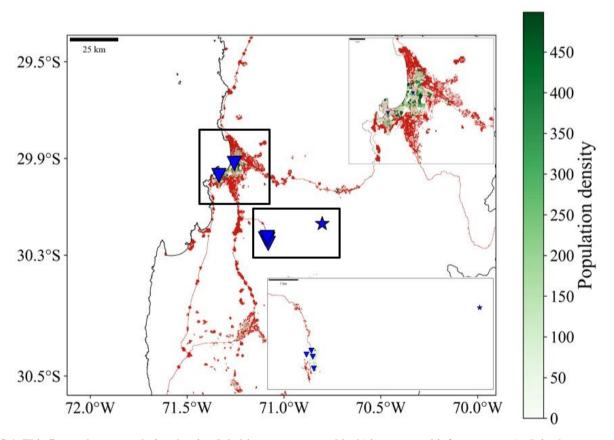


Figure S 1. This figure shows population density (inhabitants per census block) in green and infrastructure (red) in the surroundings of Tololo, which is indicated by the star. Triangles indicate the location of air quality monitoring stations run by the

Bias correction of ERA 5 data

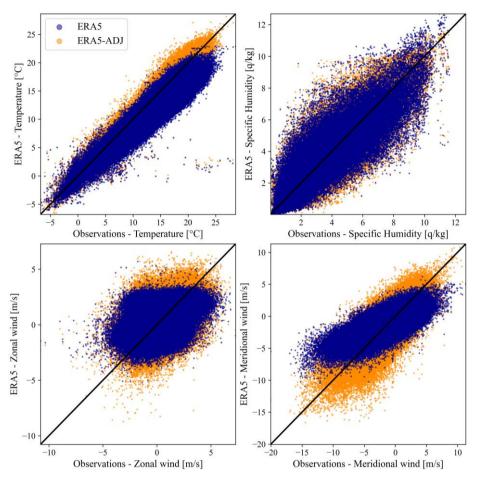


Figure S 2. Scatterplots between observed meteorological time series, and corresponding time series obtained from ERA5 and adjusted ERA5 time series. Adjusted ERA5 time series were obtained according to the bias correction described in section 2.3. Meteorological variables shown are: temperature, specific humidity, zonal wind and meridional wind.

Methane at Rapa Nui and Tololo (Charlie)

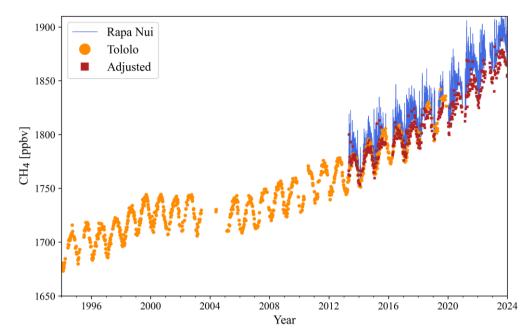


Figure S 3. Time series of observed methane (CH₄) in Rapa Nui (orange), Tololo (blue) and adjusted methane in Rapa Nui (red). The adjusted methane time series was obtained according to a simple linear regression described in section 2.4.

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Figure S 4. Upper panel shows observed (Obs, blue) and simulated (Mod, black) carbon monoxide (CO) mixing ratios (blue and black respectively). The grey shading indicates the overlap period between measurements and simulations. The second panel corresponds to a zoom over the overlap period before the bias correction. The third panel shows the simulated (Mod, black) and adjusted (Adj, bias corrected, red) time series. The lower most panel shos the simulated and adjusted curves for the estimated biomass burning (Cobb) signal. The middle panels also show error statistics (R: Pearson correlation; RMS: Root Mean Square Error; IA: Index of Agreement; Std. Dev: Normalized standard deviation as described in (Brasseur and Jacob, 2017)).

Hourly ozone distribution over time

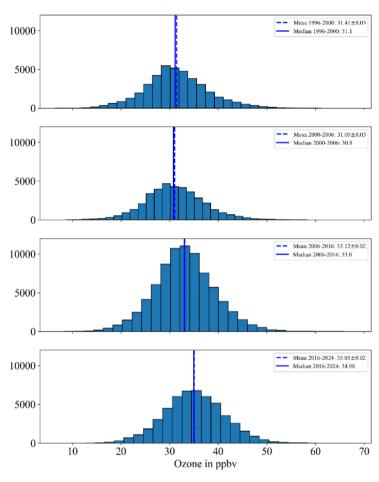


Figure S 5. Ozone hourly distributions over four periods, chosen according to change points in the methane time series. Each panel shows in the vertical axis the frequency, and in the horizontal the ozone values. Also indicated are the distribution's mean (dashed line) and median (solid line). In the corresponding legends one can read the mean \pm the standard error, and the median.

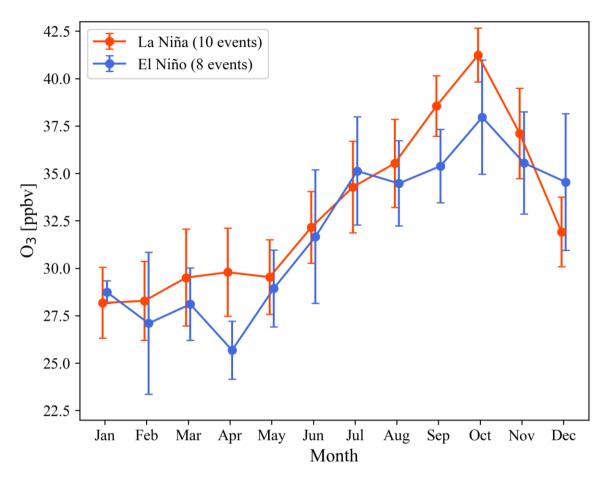


Figure S 6. Annual cycle of ozone in Tololo during El Niño and La Niña conditions. El Niño (La Niña) conditions were defined as consecutive months (at least 3 months) when MEI index was greater (less) than 0.5 (-0.5). In parentheses is indicated the number of periods with El Niño/La Niña conditions between November 1995 and 2023.

Tololo.

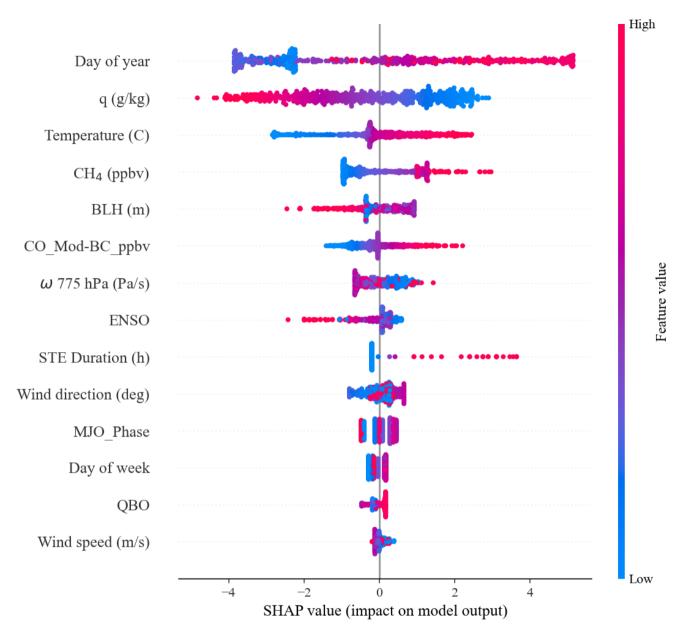


Figure S 7. Results from the SHAP methodology estimating the relative importance of explanatory variables to ozone at