

APPENDIX: SUPPLEMENTARY FIGURES

Figure S1. Scatter plots of six trace gases (O_3 , CFC-113, CH_3CN , CH_4 , CO_2 , C_2H_6) vs. CO during ARCTAS-A. The background air is shown in black and the individual air masses are highlighted in color: stratosphere (purple), stratosphere-troposphere mixed (lilac), anthropogenic pollution (blue) and biomass burning (green). These scatter plots show that tracer-tracer correlations offer valuable information in determining the threshold levels of markers for distinguishing air in the stratosphere, air associated with recent STE, biomass burning and anthropogenic emissions. In particular, the anthropogenic pollution and biomass burning plumes have very different CH_4/CO , CO_2/CO , and C_2H_6/CO ratio in addition to the differences in the CH_3CN mixing ratios.

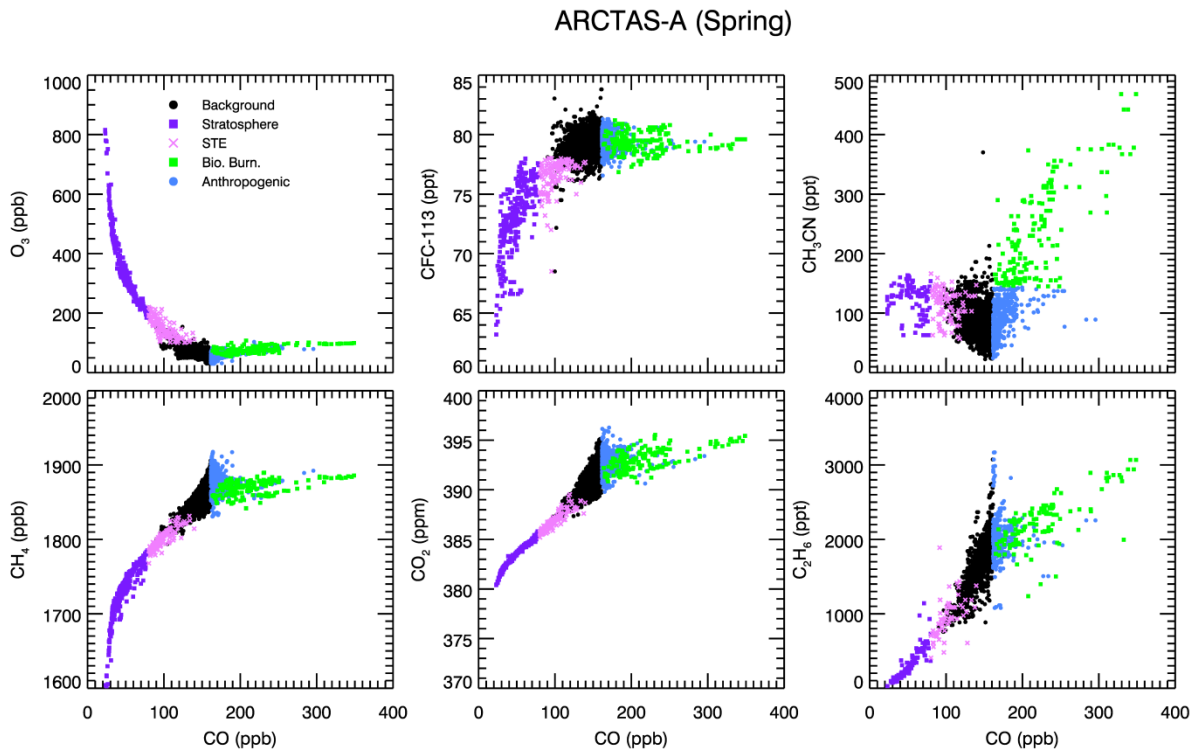


Figure S2. Similar to Figure S1 but for ARCTAS-B.

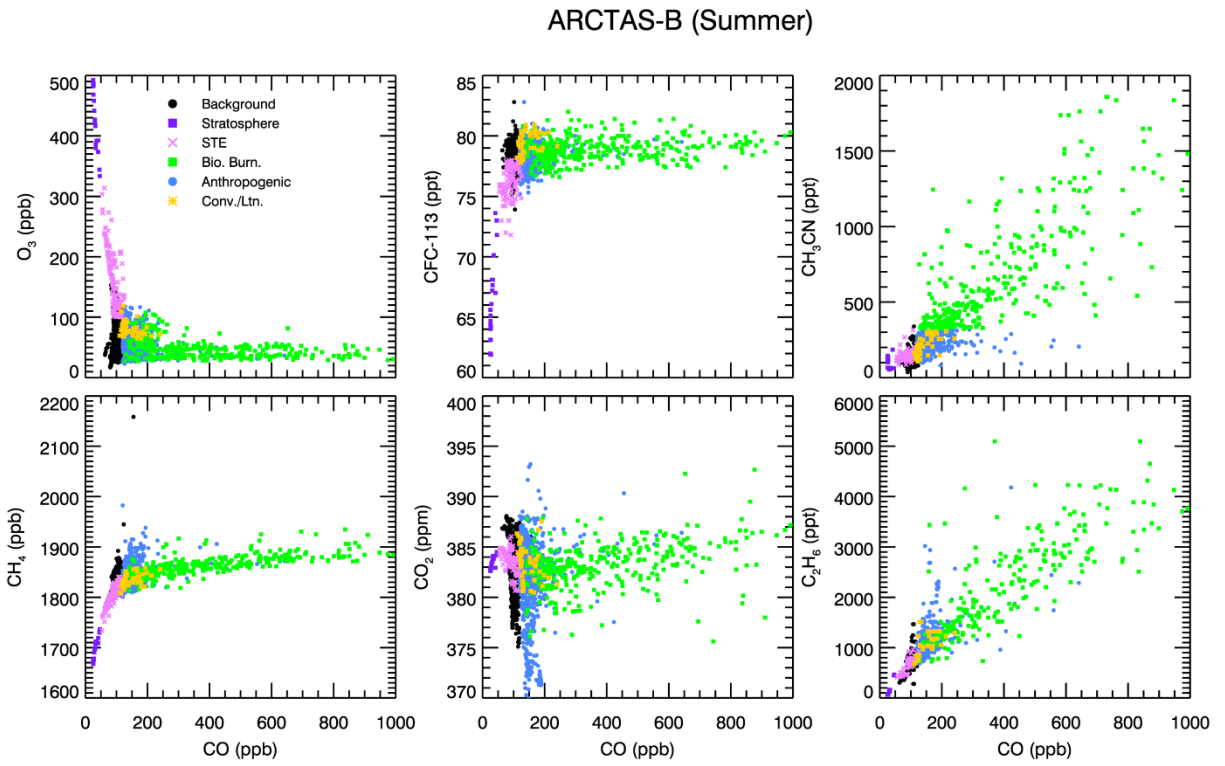


Figure S3. Time dependent calculation of NO_x , HNO_3 , and PAN in mean condition stratosphere-troposphere mixed air masses. The results are calculated using a simple box model that only includes C_2H_6 , $\text{C}_2\text{H}_5\text{O}_2$, CH_3CHO , $\text{CH}_3\text{C}(\text{O})\text{O}_2$, NO , NO_2 , PAN, HNO_3 chemistry and is constrained with the observed temperature, OH , HO_2 , NO/NO_2 ratio and photolysis rates sampled in the STE plumes during ARCTAS-B. The box model calculation is initialized with the same C_2H_6 , NO , PAN, HNO_3 as those used in the mean STE case by the Langley model (Figure 9d) with $\text{C}_2\text{H}_5\text{O}_2$, CH_3CHO , and MCO_3 initially set to zero. Note this simplified box model does not account for diurnal cycles and therefore yields PAN production rates approximately twice that calculated by the Langley box model. These results suggest that the $\text{C}_2\text{H}_6 \rightarrow \text{CH}_3\text{CHO} \rightarrow \text{CH}_3\text{C}(\text{O})\text{O}_2 \rightarrow \text{PAN}$ mechanism alone is adequate to explain the calculated PAN production in the STE plumes by the Langley box model.

