Supplementary Material:

S1: Baylor Aztec Observations on 31 August 2006.

During a morning flight on 31 August 2006, the Baylor Aztec aircraft repeatedly sampled a plume downwind of the HSC. Given the unique character of this plume and the suggestion by *Olaguer et al.* [2009] that this plume may contain a substantial contribution of primary CH_2O emissions, the measurements within the plume deserve a more detailed analysis. Figure S1 shows measurement data from a 12-minute flight segment that includes one of the three transects of the plume. This plume contained CH_2O concentrations higher than the instrument could quantify (≈9 ppbv), as well as high concentrations of O_3 and a variety of primary species. (There are differences between this figure and Fig. 5 of *Olaguer et al.* [2009] because the archived data set available from the TERC website has significantly higher O_3 and significantly lower CH_2O concentrations than shown by *Olaguer et al.* [2009]; the reason for these differences is unknown to the authors, but they do not significantly affect the conclusions reached here.) This plume is clearly a complicated situation with separate parts of the plume showing markedly different ratios of the primary pollutants NOx, CO and SO_2. Further, it is evident that relatively fresh emissions (air with high NOx/NOy ratio) were mixing with aged pollution (air with high O_3 concentrations approaching 200 ppbv, which is the highest observed by the Baylor Aztec during 2006).

Figure S1 shows two clear problems with the data and their interpretation. First, the measured CH_2O concentrations were above the levels that the instrument could quantify during the entire plume transect. Second, and more importantly, the instrument evidently has a long time constant (approaching 1 minute), while the character of the plume varies on a time scale that is short with respect to even the 5-s averaging time upon which the data were reported. Although the CH_2O data were recorded as 5-s averages, it is clear that they effectively represent much longer time averages. Thus, Fig. S1 provides no indication whether CH_2O correlates with O_3 (as expected for secondary CH_2O formation) or with any of the primary pollutants (as would be expected for primary CH_2O emission), since the instrument does not have the time response necessary to follow rapidly changing CH_2O concentrations that may have followed variations of O_3 or primary pollutant concentrations.

In summary, the plume sampled on 31 August 2006 by the Baylor Aztec aircraft was unique, with no comparable plume ever encountered by the much more extensive flights of the Electra

and WP-3D aircraft. However, the Baylor Aztec data are adequately explained by mixing of air parcels of different histories. The high observed O_3 concentrations (approaching 200 ppbv) indicate that air parcels with large concentrations of secondary pollutants were included, and the expected concentrations of CH_2O expected to be present with the O_3 are certainly high enough to account for CH_2O > 9 ppbv. For example, *Wert et al.* [2003] report a plume with O_3 approaching 200 ppbv and CH_2O > 25 ppbv. When the time and signal response of the Baylor Aztec instrumentation is properly considered, there is no direct evidence for primary emissions of CH_2O in this sampled plume.

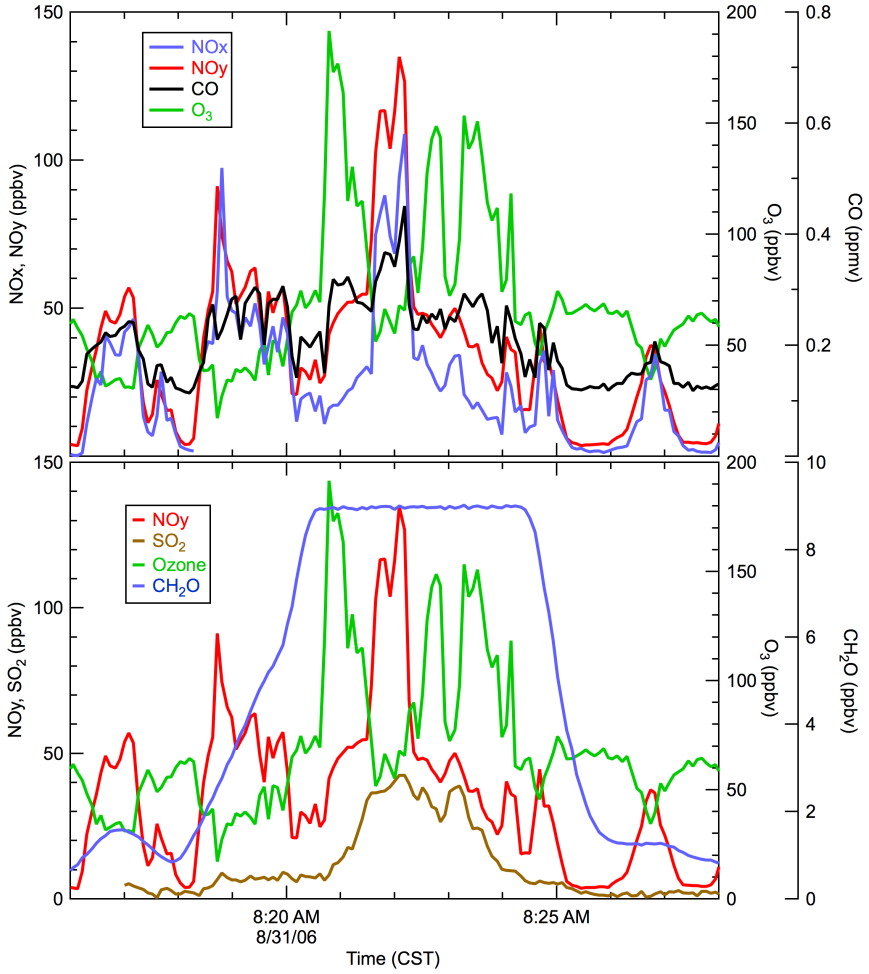


Figure S1: Time series of Baylor Aztec data during a 12-minute period that included a plume transect downwind of HSC. Both panels include NOy and ozone for ease of comparison of the behavior of different species. Data downloaded from the TERC website (http://projects.tercairquality.org/AQR/H063) on 18 January 2011.

S2: Surface Observations on 27 September 2006.

Eom et al. [2008] report an observation of a CH_2O plume during the morning of 27 September 2006 at the Lynchburg Ferry USEPA site in Baytown, TX. This plume reached a maximum concentration of 52 ppbv. A definitive examination of the sources of CH_2O in this (or any other) plume requires consideration of the recent transport of the sampled air parcel. Figure S2 shows two different representations of the histories of the air parcels impacting the HGB area on 27 September 2006. Both indicate that the air from the HGB region on 26 September was transported south over Galveston Bay and returned to the HGB area on 27 September. In Fig. S2b the air located at the asterisk at 2:00 pm on 27 September at an altitude of 500 m (i.e. within the CBL) had originated from nearly that identical location 24 hours earlier (indicated by the numeral "1", where the numerals give the transport time in days). In Fig. 2a, the trajectories during the previous 24 hrs varied markedly with altitudes below 1000m; this indicates that the flow was strongly stratified during the night preceding the observation of the CH_2O plume on 27 September. Given the pronounced recirculation and the strongly stratified flow characteristic of this period, simple analyses based upon local wind direction will certainly be fraught with uncertainty.

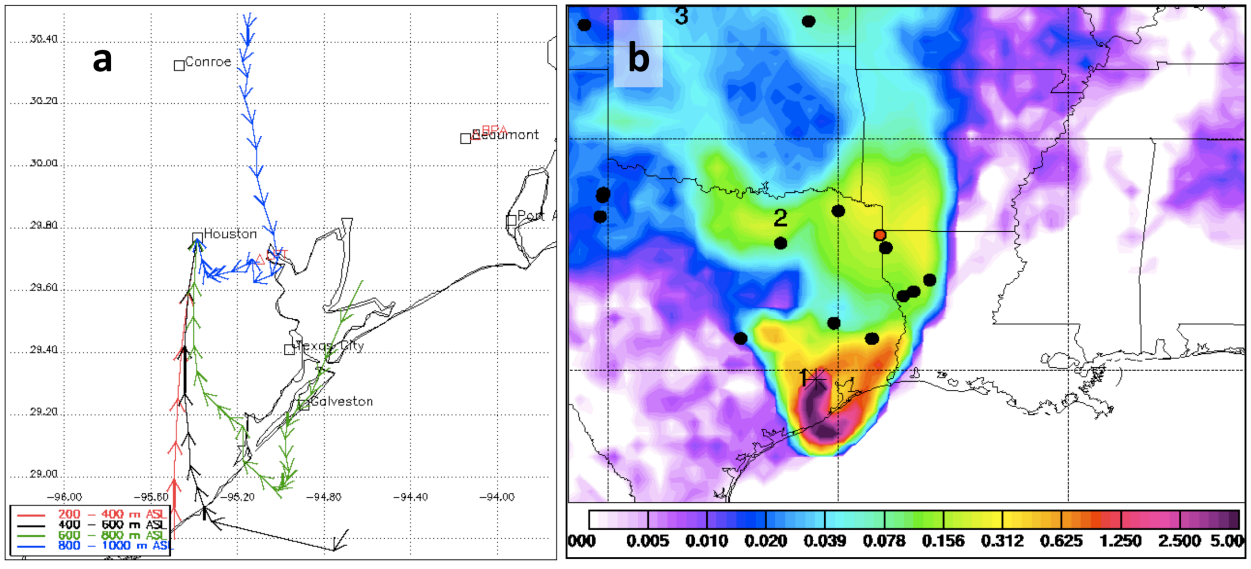
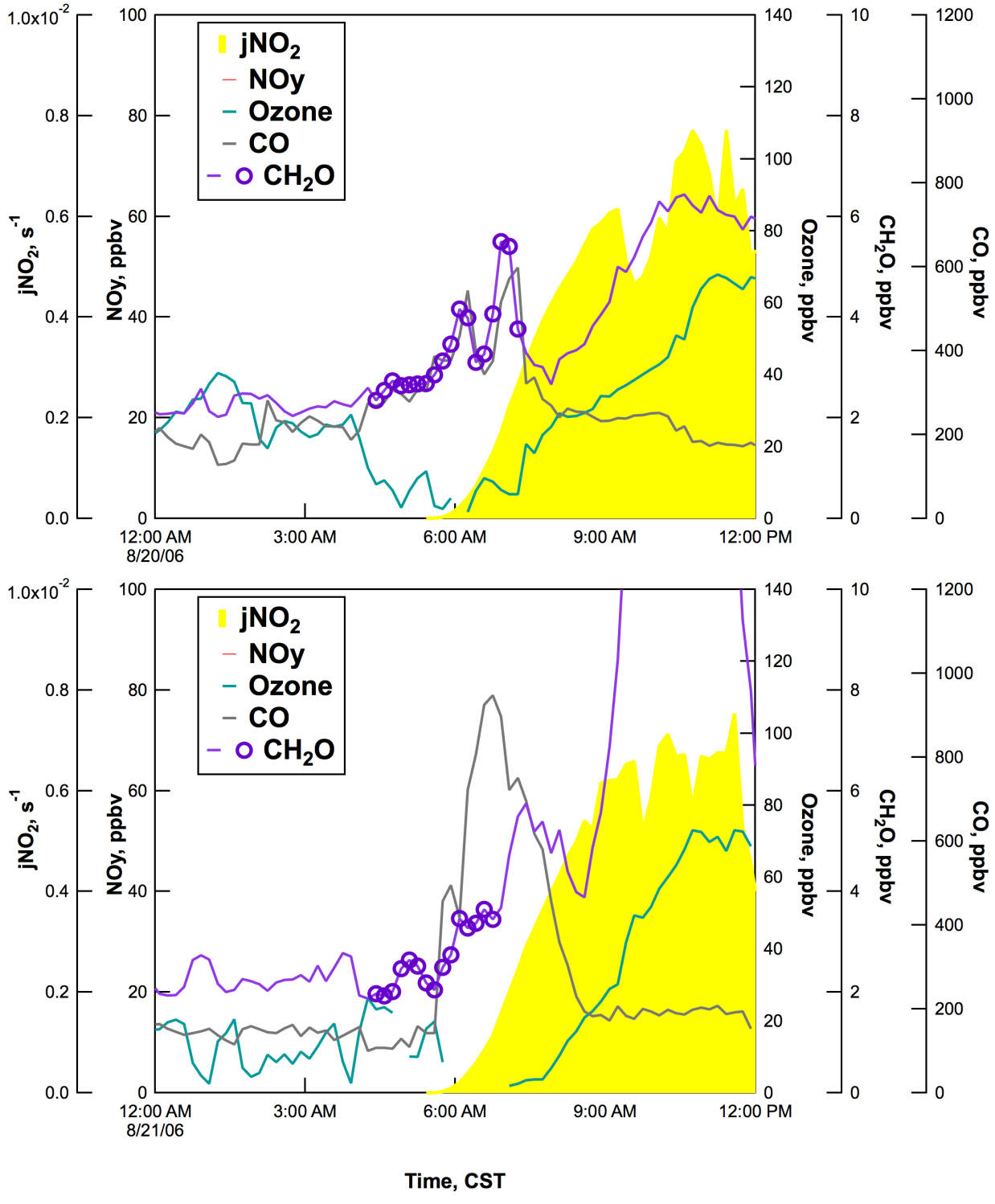
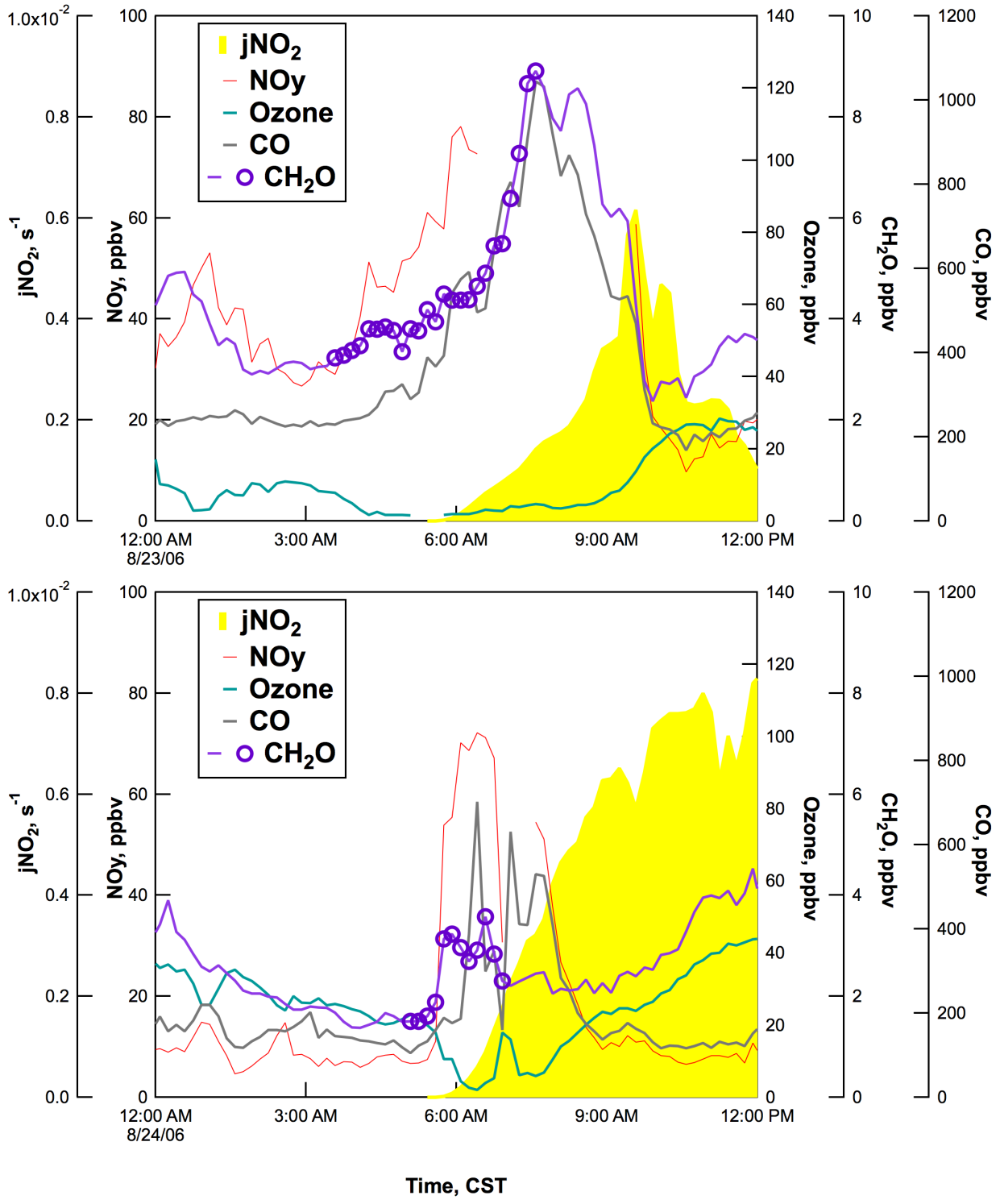


Figure S2: Trajectory analysis of air reaching the HGB area on the morning of 27 September 2006. **a)** 24-hr back trajectories calculated by the NOAA/ESRL/PSD TexAQS Boundary Layer Profiler Trajectory Calculator (http://www.esrl.noaa.gov/psd/programs/2006/texaqs/traj//) at 4 different heights above the ground. Arrival time of the trajectory is 8:00am local standard time. **b)** Footprint emission sensitivity plot for air arriving at asterisk at 960 hPa altitude calculated from FLEXPART; note that the scale is logarithmic in arbitrary units. Arrival time is 2:00pm local standard time.

S3. Plots of atmospheric species measured each day at Moody Tower during TRAMP in the same format as Figs. 5 and 6 of the text.

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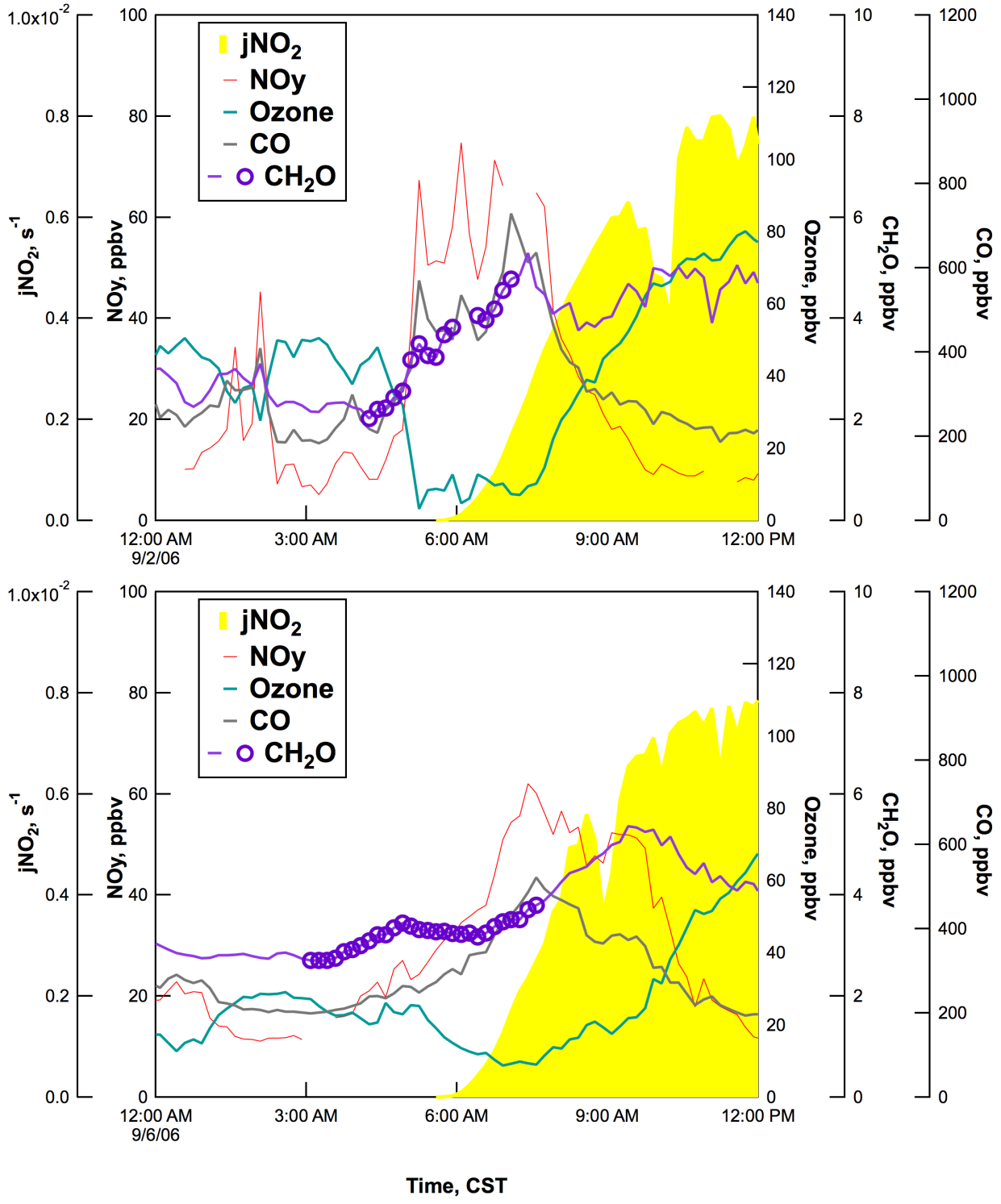
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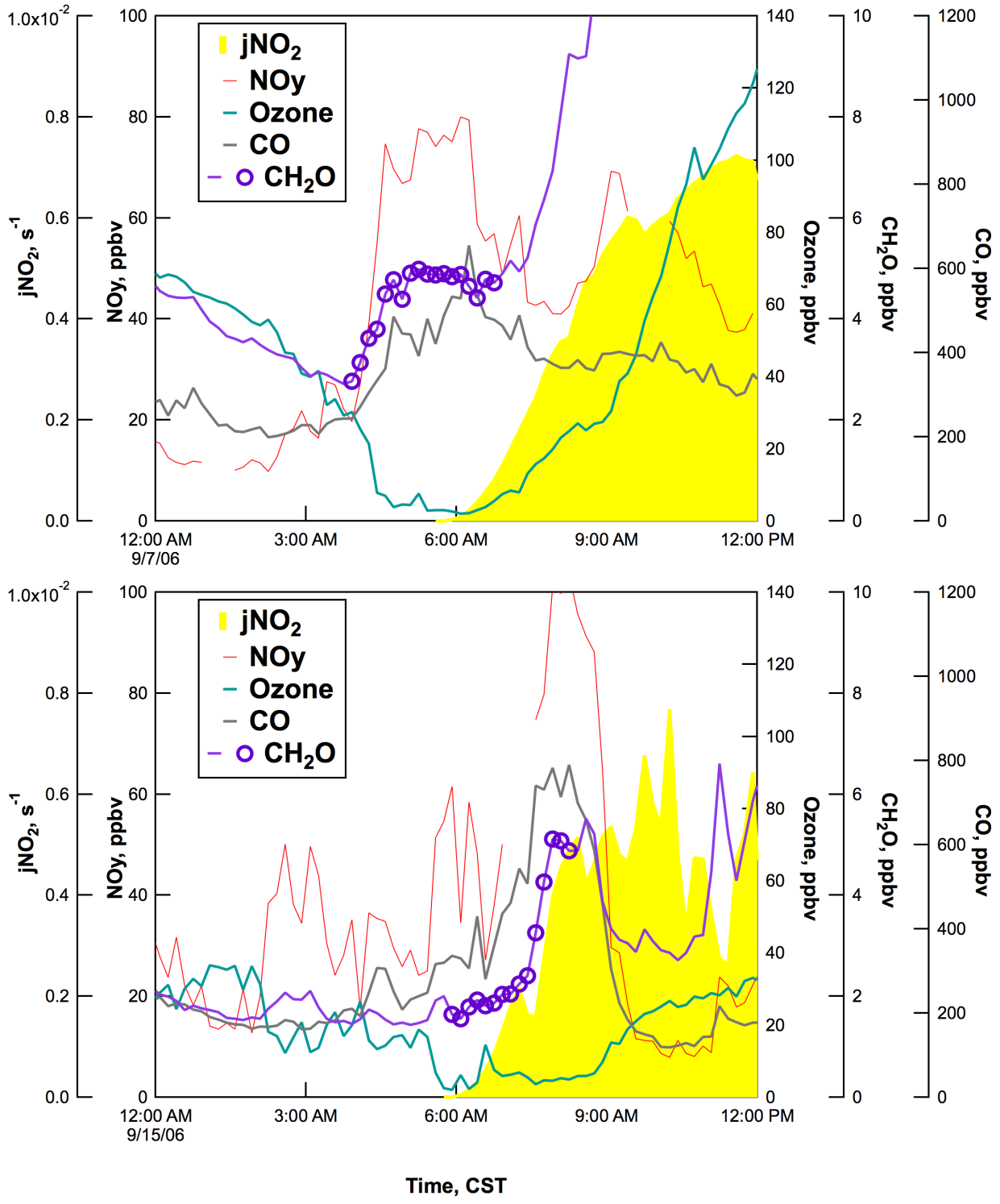
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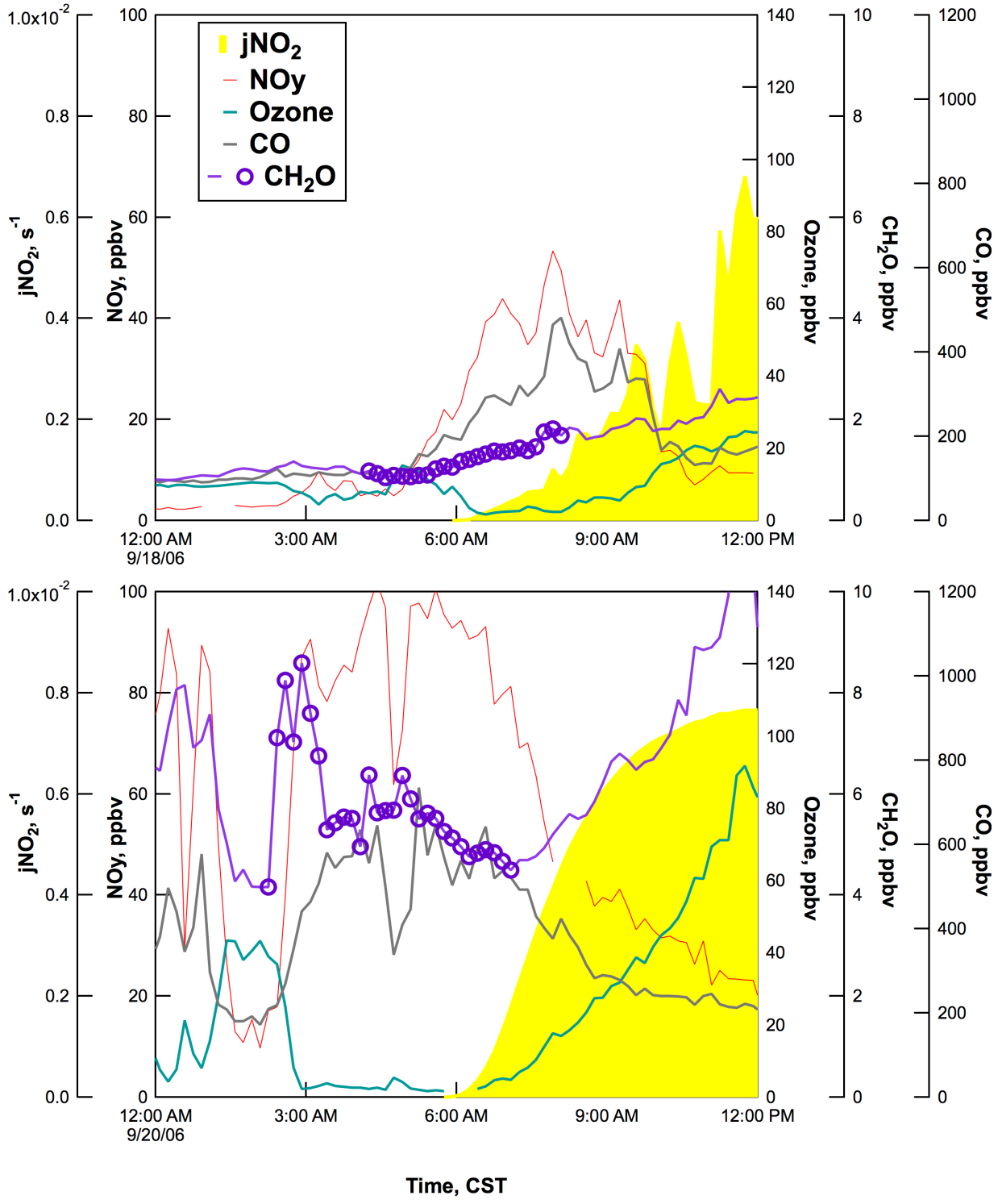
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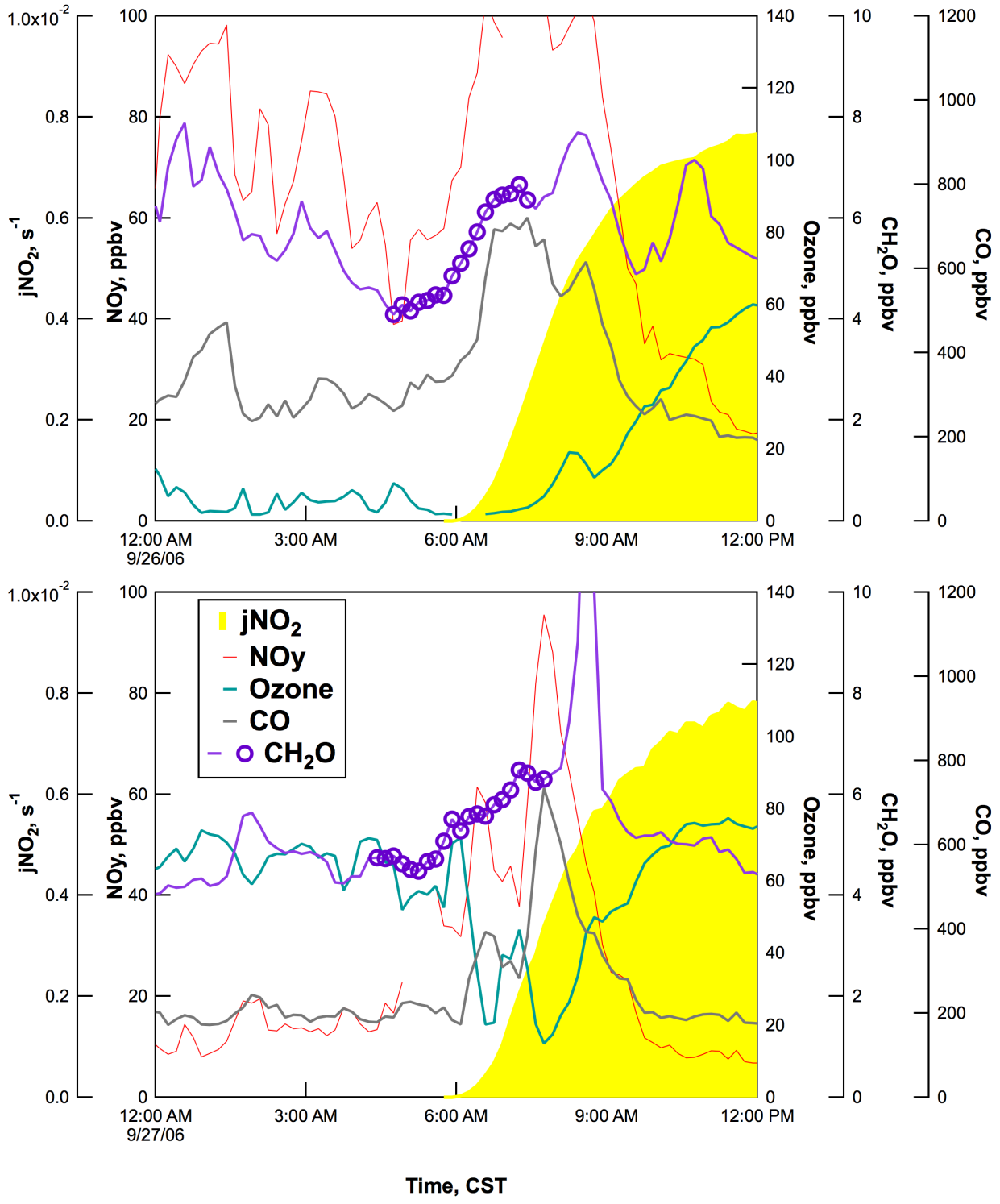
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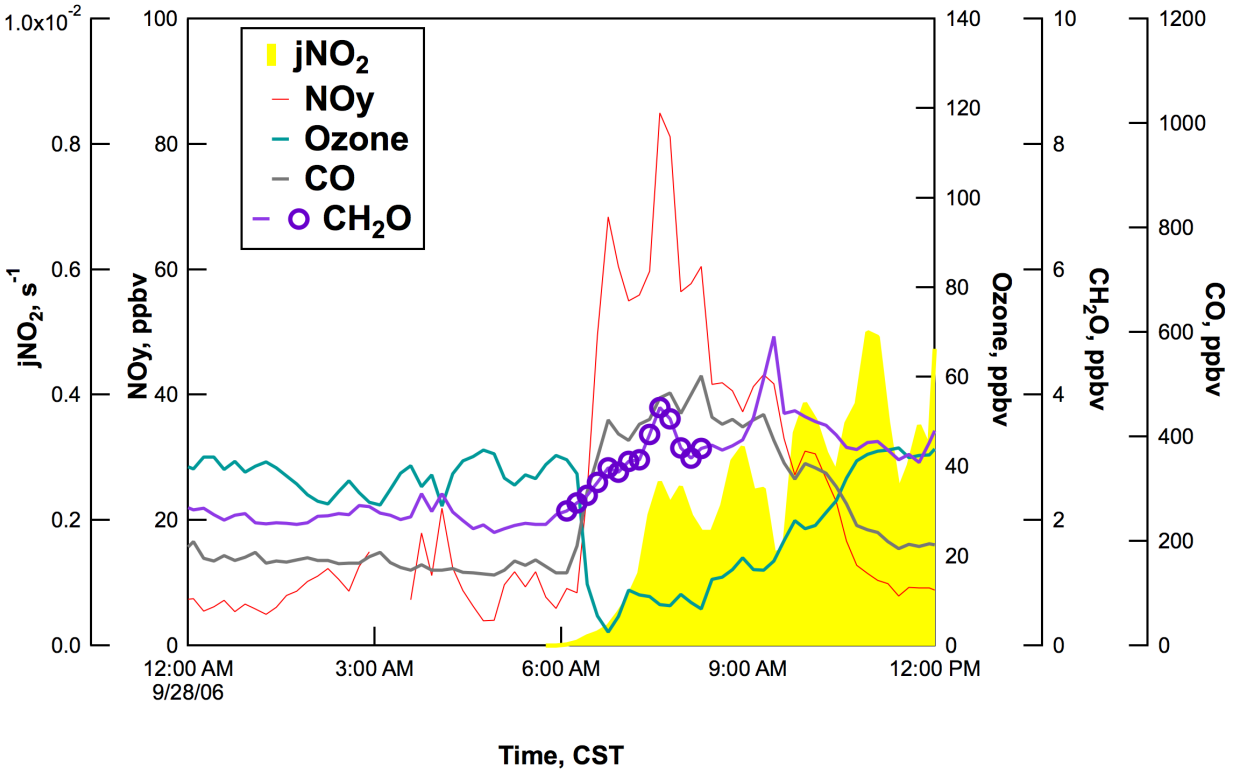
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