SUPPLEMENT to





Figure S1: Analogous to Figure 2 the above graphs compare statistics of MOZART (red) and WRF-Chem simulations ("Variable BC": blue; "Mean BC": green) to aircraft measurements (black) of CO, O_3 and PAN for the DC-8 flight leg on 22 June 2008 over ocean. Shown are mean (filled symbol), median (open symbols), 10th and 90th percentiles (bars) and extremes (lines). The number of data points per 1-km wide altitude bin is shown next to the graphs.

Results from the "Variable BC" simulation are overall similar to the MOZART results as can be expected since the flight track is located off the coast of California and mostly impacted by inflow, i.e. boundary conditions. However, some discrepancies are seen reflecting that also in a region that is mostly impacted by lateral boundaries the different model physics, dynamics and resolution alter the characteristics of the incoming airmasses. "Mean BC" statistics are overall lower compared to the other model statistics and show clearly reduced variability which is explained by (1) the above average pollution inflow on the flight day and (2) the reduced variability in the lateral boundary conditions.



Figure S2: Analogous to Figure 3 the above graphs show O₃-CO and O₃-PAN relationships for the flight track over ocean on 22 June 2008 from observations, MOZART, and WRF-Chem "Variable BC" and "Mean BC simulations". Data are color-coded by altitude. For illustration purposes, a 2-sided linear regression line to the flight track data is added to the graphs. The thin line in the model plots is a repeat of the fit to the aircraft observations.

Similar to Figure S1, the results from the "Variable BC" simulation overall agree with the MOZART results since the flight track is mostly influenced by inflow and the small differences reflect that different model physics, dynamics and resolution. Statistics from the "Mean BC" WRF-Chem simulation show smaller concentrations and a clearly reduced variability. The 2-sided linear regression lines for "Mean BC" also deviate significantly more from the observations compared to the "Variable BC" or MOZART-4 simulation.



Figure S3: As Figure S1 (and similar to Figure 5) but for observed and modeled flight track data over land. Fire influenced data have been omitted.

Comparable to the comparison of the ocean data, the "Variable BC" and MOZART-4 results are overall comparable in the free troposphere with smaller differences explained by the different model configurations. Towards the lower altitudes the differences between the models increase indicating a stronger influence of local processes where the different model resolution and also configurations (e.g. daily versus hourly anthropogenic emissions, different boundary layer schemes,) gain in importance and the influence of lateral boundary conditions is reduced. This is also reflected when comparing "Variable BC" to "Mean BC" results resulting in larger differences in the free troposphere and smaller differences at the lower altitudes.