**1** Supplementary Material

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3 Figures S1-S9 show CAM4 and CAM5 simulated HNO<sub>3</sub>, NO, NO<sub>2</sub>, NO<sub>x</sub>, and OH compared against observations by several campaigns. Vertical profiles of observations (box and whisker 4 5 plot) and CAM4 (blue) and CAM5 (red) model results are plotted as a function of height (km). 6 The numbers along the right vertical axis represent the number of observations at each vertical 7 height. The box represents the central 50% and 90% of observations while the star represents 8 the mean. For OH, model results are evaluated using results from Spivakovsky et al. (2010). 9 OH values are contoured with bright colors (red) representing higher values and darker colors 10 (blue) representing lower values.

11 The comparison of the CAM5 and CAM4 simulated results to observations provided here provides a general perspective of how the models compare to observations and also to compare 12 the performance of CAM5 to CAM4. The detailed analyses of how CAM4 compares to 13 observations on a finer scale is provided in Lamarque et al. (2012). Overall, the models are in 14 15 agreement with each other and with the results obtained by the detailed comparison of CAM4 16 to observations in Lamarque et al. (2012). Although both models agree well with observations 17 in the mid-troposphere (4-8 km), for some species and locations, CAM5 agrees better with observations. 18

Figure S1 shows the model representation of HNO<sub>3</sub>. As shown in Figure S1, the models' 19 20 simulated HNO<sub>3</sub> agrees well with observations, but there are some differences between the 21 models' results below and above 6 Km. HNO<sub>3</sub> concentrations are larger in CAM4 below 6 km, however, above 6 km HNO<sub>3</sub> concentrations are larger in CAM5. This indicates a lower 22 conversion of HNO<sub>3</sub> to its aerosol phase at cruise altitudes and a higher conversion of HNO<sub>3</sub> to 23 its aerosol phase near the surface in CAM5. This behavior agrees well with the lower 24 conversion of NO<sub>v</sub> to its aerosols phase at cruise altitudes and higher conversion of NO<sub>v</sub> to its 25 26 aerosols phase near the surface in CAM5 as seen in Table 1 and 2.

Figure S2 shows the model representation of NO. As shown in Figure S2, for NO both models are in good agreement with each other, but compared to observations both models underestimate NO throughout the troposphere. Figure S3 shows the model representation of NO<sub>2</sub>. Both models agree well with observations in
the mid-troposphere, but they overestimate values of NO<sub>2</sub> at higher altitudes, with the positive
bias consistently greater in CAM4 than in CAM5.

Figure S4 shows the model representation of NOx CAM5 and CAM4 simulated NOx agrees
very well with the results found in Lamarque et al (2012). As in Figure S4, NOx is well
produced by both models throughout the troposphere. The models overestimate NOx for
several locations in the tropics with CAM4 giving slightly more NOx than CAM5.

Figure S5a and S5b show the model representation of OH. For most seasons, both models are accurate at resolving OH in the UTLS over 30-60N. As shown in Figure S5b, CAM5 overestimates OH at low latitudes for all months verified against the Spivakovsky et al. (2000) climatology. The disparities are more pronounced close to the surface and in the upper troposphere during the NH spring and summer. As shown in Figure S5a, CAM4 also overestimates OH for low latitudes during the NH spring and summer, though not as much as CAM5. For NH fall and winter months, CAM4 underestimates OH.

15 Overall, in the mid-troposphere (4-8 km) where the majority of the observations were taken,

both CAM4 and CAM5 simulated concentrations and distributions of these gases are within the

17 central 50% and 90% of the available observations. Therefore, both models agree well with the

18 available observations for the majority of stations in the mid-troposphere.



Figure S1. Vertical profiles of HNO<sub>3</sub> observations (box and whisker plot) and CAM4 (blue) and CAM5
(red) model. The numbers along the right vertical axis represent the number of observations at each
vertical height. The box represents the central 50% and 90% of observations while the star represents
the mean.





Figure S1. Continued.



23 Figure S1. Continued.







Figure S1. Continued.





Figure S2. Same as Figure S1, but for NO.







Figure S2. Continued.





Figure S2. Continued.



Figure S2. Continued.





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Figure S2. Continued.



3 Figure S3. Same as Figure S1, but for NO<sub>2</sub>.



Figure S3. Continued.



Figure S4. Same as Figure S1, but for NOx.



Figure S4. Continued.





Figure S4. Continued.



3 Figure S4. Continued.



Figure S5a. Vertical profile of CAM4 simulated OH (left) and observations from the Spivakovsky et al.
(2010) climatology for four months (Jan, Apr, Jul, Oct). Bright colors (red) represent high values of
OH, dull colors (blue) represent low values of OH.





23 Figure S5b. Same as Figure S5a, but with CAM5.