



## Supplement of

## Decadal changes in global surface $NO_x$ emissions from multi-constituent satellite data assimilation

Kazuyuki Miyazaki et al.

Correspondence to: K. Miyazaki (kmiyazaki@jamstec.go.jp)

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Table S1: Model minus observation comparisons of the mean  $O_3$  concentrations between the MIROC simulations and TES ver. 6 special observations during 2011-2014. The results are shown for MIROC simulations using the optimized surface NOx emissions and the a priori surface NOx emissions (in brackets)

Location	Bias [ppb]	RMSE [ppb]
Fort MacKay	-5.3 (-5.6)	7.1 (7.6)
Paris	-7.8 (-8.7)	9.7 (10.4)
Trinidad Head	-5.1 (-5.7)	6.8 (7.3)
New York	-8.1 (-8.9)	13.8 (14.2)
Istanbul	-8.2 (-9.7)	10.1 (11.8)
Beijing	-7.0 (-8.9)	8.5 (10.1)
Seoul	-4.3 (-5.2)	6.5 (7.2)
Tokyo	-5.4 (-6.4)	8.5 (9.3)
Los Angeles	-9.5 (-12.2)	11.7 (14.0)
Houston	-0.6 (-2.4)	5.2(5.7)
Mexico city	-4.4 (-6.3)	4.9 (7.3)
Shenzhen	0.2 (-1.0)	4.6 (5.1)
Mumbai	0.8 (-1.7)	7.8 (8.1)
Bangkok	1.6 (-0.6)	6.8 (6.4)
Lagos	-1.7 (-3.8)	4.1 (5.5)
Sao Paulo	-0.1 (-1.5)	3.3(3.2)



Figure S1a: Time series of the monthly and regional mean error reduction of tropospheric  $NO_2$  column by data assimilation (black line, in %), relative observation error (red line in %), and the mean number of observations (blue line, per month per super observation grid) for OMI. The relative error was estimated by dividing the mean observation error by the mean observation concentration for each super-observation.



Figure S1b: Same as in Fig. S1a, but for SCIAMACHY.



Figure S1c: Same as in Fig. S1a, but for GOME-2.



Figure S2: Global distribution of OH concentration differences (in  $10^6$  molecules cm<sup>-3</sup>) between multiple species data assimilation runs and a NO<sub>2</sub>-only data assimilation in June 2008, at 850 hPa (left) and 500 hPa (right). The results are shown for the multiple-species assimilation using (top) all the non-NO<sub>2</sub> measurements (TES O<sub>3</sub>, MLS O<sub>3</sub>/HNO<sub>3</sub>, and MOPITT CO) + NO<sub>2</sub> measurements, (middle) TES O<sub>3</sub> + NO<sub>2</sub> measurements, and (bottom) MOPITT CO + NO<sub>2</sub> measurements.