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Supplementary material to "Long-term changes of tropospheric NO_2 over megacities derived from multiple satellite instruments"

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Abstract. This document contains supplementary material to the manuscript *Long-term changes of tropospheric* NO_2 *over megacities derived from multiple satellite instruments* (Hilboll et al., 2013).

1 The Bremen 3d CTM

The Bremen 3d CTM is a combination of the "Bremen transport model" (Sinnhuber at al., 2003a) with the chemistry code of the "Bremen two-dimensional model of the stratosphere and mesosphere" (Sinnhuber et al., 2003b; Winkler et al., 2008), which evolved from SCLIMACT (Chipperfield, 1999). The model is driven by ECMWF ERA-Interim meteorological reanalysis fields (Dee et al., 2011) and runs on a combined 30 min/15 min time-scale for transport and chemistry, respectively. Vertical transport is included in the dynamics using the advection scheme developed by Prather (1986). While the horizontal resolution is constant at $2.5° \times 3.75°$, the vertical resolution varies from $\sim$1 km in the lower stratosphere to $\sim$4 km at $\sim$60 km altitude. The model runs on 28 isentropic surfaces from 330 K to 3402 K (about 10–65 km). Stratospheric profiles for each satellite measurement are interpolated in space and time using smoothing spling and linear interpolation, respectively, to yield stratospheric vertical columns VCD_{strat} and air mass factors AMF_{strat}.

2 Additional trend results

The tables shown here include results of the following trend calculations:

- Levelshift trend calculation (Eq. 3 in Hilboll et al., 2013), applied to original GOME data, for the 1996–2006 time period.
- Levelshift trend calculation (Eq. 3 in Hilboll et al., 2013), applied to resolution-corrected GOME data, for both the 1996–2011 and the 1996–2006 time period.
- Multi-instrument trend calculation with variable seasonality component (Eq. 9 in Hilboll et al., 2013), for the 1996–2011 time period.

Regarding the uncertainties of the estimator for the trend in the amplitude of the seasonality in the multi-instrument fit, $\hat{\xi}$, it must be noted that in some cases, the bootstrap leads to unreasonably large absolute replication values $\hat{\xi}^*$. This in turn leads to a very high estimated standard error. However, it is still possible that all replications are either larger or smaller than zero, leading to 95% confidence intervals which do not include zero, which in turn indicates a significant trend. Thus trends in the seasonality component might be significant in spite of extremely large standard errors.

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Table 1. Annual growth rate $\Delta VCD_{trop}NO_2$ from the levelshift model (Eq. 3 in Hilboll et al., 2013) and the multi-instrument fit (Eq. 9 in Hilboll et al., 2013) for the regions shown in Fig. 2 in Hilboll et al. (2013). The relative trends have been computed relative to the 1996 annual mean. Non-significant trends are shown *in italics*.

Region	Levelshift-model (orig. GOME data) 1996–2006		Levelshift-model (rescorr. GOME data) 1996–2011		1996–2006		Multi-instrument fit (var. seasonality) $\hat{\omega}$ (1996–2011)		$\hat{\xi}$ (1996–2011)
	[10^{14} molec cm^{-2} yr^{-1}]	[% yr^{-1}]	[10^{14} molec cm^{-2} yr^{-1}]	[% yr^{-1}]	[10^{14} molec cm^{-2} yr^{-1}]	[% yr^{-1}]	[10^{14} molec cm^{-2} yr^{-1}]	[% yr^{-1}]	[% yr^{-1}]
Continental U.S.	*+0.01 ± 0.17*	*+0.02 ± 0.38*	-0.81 ± 0.15	-1.62 ± 0.30	*+0.03 ± 0.18*	*+0.07 ± 0.39*	-0.8 ± 0.19	-1.64 ± 0.40	*-2.27 ± 0.97*
Central-Eastern U.S.	-1.54 ± 0.47	-1.63 ± 0.50	-2.94 ± 0.39	-2.92 ± 0.39	-1.54 ± 0.48	-1.61 ± 0.50	-2.62 ± 0.44	-2.67 ± 0.44	-3.29 ± 0.66
Western Europe	-3.6 ± 1.1	-3.11 ± 0.98	-2.96 ± 0.65	-2.56 ± 0.56	-3.6 ± 1.2	-3.08 ± 0.99	-2.61 ± 0.84	-2.34 ± 0.75	*-2.8 ± 1.2*
Japan	*-0.17 ± 0.23*	*-0.45 ± 0.59*	-0.49 ± 0.14	-1.17 ± 0.33	*-0.16 ± 0.24*	*-0.40 ± 0.58*	-0.49 ± 0.17	-1.23 ± 0.42	*-1.3 ± 1.1*
Middle East	+0.59 ± 0.11	+2.94 ± 0.54	+0.785 ± 0.081	+4.01 ± 0.42	+0.62 ± 0.11	+3.05 ± 0.53	+0.725 ± 0.087	+3.69 ± 0.44	+7 ± 65
East Central China	+5.85 ± 0.85	+9.2 ± 1.3	+10.1 ± 1.1	+20.3 ± 2.2	+5.86 ± 0.85	+9.2 ± 1.3	+8.6 ± 1.1	+16.1 ± 2.0	+23 ± 14
North Central India	+1.18 ± 0.43	+3.7 ± 1.3	1.08 ± 0.27	+4.6 ± 1.1	*+0.64 ± 0.36*	*+2.6 ± 1.4*	+1.01 ± 0.31	+3.08 ± 0.95	+9.0 ± 7.0

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Table 2. Annual growth rate $\Delta VCD_{trop}NO_2$ from the levelshift model (Eq. 3 in Hilboll et al., 2013) and the multi-instrument fit (Eq. 9 in Hilboll et al., 2013) for a list of megacities. The relative trends have been computed relative to the 1996 annual mean. Non-significant trends are shown *in italics*.

Region	Levelshift-model (orig. GOME data) 1996–2006		Levelshift-model (rescorr. GOME data) 1996–2011		Levelshift-model (rescorr. GOME data) 1996–2006		Multi-instrument fit (var. seasonality) $\hat{\omega}$ (1996–2011)		$\hat{\xi}$ (1996–2011)
	[10^{14} molec cm^{-2} yr^{-1}]	[% yr^{-1}]	[10^{14} molec cm^{-2} yr^{-1}]	[% yr^{-1}]	[10^{14} molec cm^{-2} yr^{-1}]	[% yr^{-1}]	[10^{14} molec cm^{-2} yr^{-1}]	[% yr^{-1}]	[% yr^{-1}]
Algiers	+0.64 ± +0.17	+3.10 ± 0.81	+0.83±0.17	+2.62±0.53	+0.87 ± 0.23	+2.75 ± 0.74	+0.60 ± 0.18	+2.89 ± 0.87	*-1.7 ± 4.2*
Athens	*+1.15 ± 0.84*	*+2.6 ± 1.9*	-2.10±0.87	-1.72±0.71	*+2.0 ± 1.2*	*+1.9 ± 1.1*	-2.08 ± 0.82	-3.7 ± 1.5	*-2.8 ± 1.6*
Baghdad	+1.81 ± 0.35	+7.9 ± 1.5	+3.91±0.34	+10.12±0.87	+2.82 ± 0.45	+6.7 ± 1.1	+3.23 ± 0.38	+17.9 ± 2.1	+17 ± 75
Beijing	+8.8 ± 3.6	+6.7 ± 2.8	+9.4±2.7	+4.8±1.4	+10.2 ± 4.0	+5.3 ± 2.1	+9.4 ± 2.7	+7.2 ± 2.1	+7.4 ± 6.3
Buenos Aires	*+0.06 ± 0.73*	*+0.2 ± 2.2*	*+0.85±0.73*	*+1.11±0.94*	*-0.7 ± 1.3*	*-0.8 ± 1.5*	*+0.55 ± 0.51*	*+1.7 ± 1.6*	*-1.2 ± 6.7*
Cairo	+1.46 ± 0.41	+5.2 ± 1.5	+2.12±0.27	+4.35±0.55	+2.00 ± 0.46	+4.08 ± 0.93	+1.72 ± 0.28	+6.4 ± 1.0	+10 ± 94
Chicago	*-3.8 ± 3.0*	*-2.5 ± 2.0*	-7.0±2.0	-2.46±0.68	*-5.0 ± 3.4*	*-1.8 ± 1.2*	-6.3 ± 2.2	-3.9 ± 1.4	*+0 ± 23*
Dhaka	+1.39 ± 0.48	+6.5 ± 2.2	+3.94±0.50	+12.7±1.6	+2.01 ± 0.55	+5.3 ± 1.5	+3.35 ± 0.53	+23.0 ± 3.6	+22 ± 61
Damascus	+2.93 ± 0.63	+8.3 ± 1.8	+3.90±0.60	+6.03±0.92	+4.12 ± 0.78	+6.4 ± 1.2	+2.60 ± 0.51	+7.1 ± 1.4	+13 ± 78
Guangzhou	+7.8 ± 2.7	+10.6 ± 3.6	+3.0±2.6	+1.6±1.4	+13.6 ± 3.8	+9.2 ± 2.6	*+0.3 ± 2.6*	*+0.3 ± 2.6*	-1 ± 12
Hong Kong	+7.0 ± 2.2	+8.7 ± 2.7	*-0.8±2.2*	*-0.4±1.2*	+12.4 ± 3.0	+8.5 ± 2.0	*-1.0 ± 2.2*	*-0.9 ± 2.0*	*-2.1 ± 1.9*
Istanbul	*+1.6 ± 1.6*	*+2.5 ± 2.5*	*-0.5±1.3*	*-0.4±1.0*	*+1.5 ± 2.1*	*+1.3 ± 1.7*	*-0.4 ± 1.1*	*-0.5 ± 1.5*	*+1 ± 16*
Jakarta	*-0.23 ± 0.63*	*-0.7 ± 1.9*	-1.57±0.55	-2.58±0.91	*-0.61 ± 0.93*	*-1.1 ± 1.6*	-1.20 ± 0.41	-3.3 ± 1.1	*+1 ± 16*
Jeddah	+0.83 ± 0.38	+2.5 ± 1.1	+1.64±0.38	+2.31±0.53	+1.71 ± 0.58	+2.42 ± 0.81	+1.41 ± 0.35	+4.5 ± 1.1	+30 ± 350
Karachi	*+0.21 ± 0.29*	*+1.3 ± 1.7*	+1.07±0.29	+3.8±1.0	*+0.56 ± 0.47*	*+1.9 ± 1.6*	+0.84 ± 0.25	+5.9 ± 1.8	+6 ± 24
Kolkata	*+0.35 ± 0.34*	*+1.3 ± 1.3*	+0.76±0.23	+2.03±0.61	*+0.39 ± 0.37*	*+1.00 ± 0.94*	+0.80 ± 0.25	+3.19 ± 0.99	*+2.9 ± 2.2*
Lagos	*+0.03 ± 0.15*	*+0.2 ± 1.1*	+0.40±0.11	+2.63±0.73	*-0.03 ± 0.18*	*-0.2 ± 1.1*	+0.33 ± 0.12	+2.67 ± 0.96	*+0.4 ± 2.6*
Lima	*+0.63 ± 0.55*	*+4.2 ± 3.7*	+1.22±0.48	+3.0±1.2	*+1.27 ± 0.74*	*+3.1 ± 1.8*	+1.06 ± 0.36	+7.9 ± 2.7	+0 ± 190
London	*-2.9 ± 2.1*	*-1.6 ± 1.2*	-4.5±1.4	-2.24±0.68	*-3.2 ± 2.3*	*-1.6 ± 1.2*	*-3.0 ± 1.6*	*-1.68 ± 0.89*	*+7.1 ± 7.7*
Los Angeles	*-3.9 ± 2.4*	*-2.0 ± 1.2*	-14.1±1.9	-3.14±0.43	*-4.7 ± 3.1*	*-1.13 ± 0.76*	-13.1 ± 2.6	-5.8 ± 1.1	*-3.0 ± 1.3*
Manila	-0.99 ± 0.30	-4.8 ± 1.5	-1.43±0.31	-3.56±0.76	-1.74 ± 0.57	-4.2 ± 1.4	-1.04 ± 0.21	-5.0 ± 1.0	*-4.0 ± 9.8*
Mexico City	*+0.8 ± 1.2*	*+1.6 ± 2.4*	*-0.30±0.98*	*-0.22±0.72*	*+1.7 ± 1.6*	*+1.3 ± 1.2*	*+0.51 ± 0.82*	*+1.0 ± 1.6*	*+0.1 ± 7.4*
Moscow	*+2.1 ± 2.2*	*+3.1 ± 3.2*	*+0.3±1.7*	*+0.2±1.2*	*+3.8 ± 2.5*	*+3.4 ± 2.2*	*-1.4 ± 1.6*	*-1.7 ± 1.9*	*-2 ± 16*
Mumbai	+0.48 ± 0.23	+2.4 ± 1.1	+0.91±0.20	+2.46±0.53	+0.69 ± 0.30	+1.84 ± 0.81	+0.68 ± 0.21	+3.5 ± 1.0	*+2.1 ± 1.7*
New Delhi	+2.67 ± 0.76	+7.9 ± 2.2	+3.07±0.56	+5.5±1.0	+2.80 ± 0.87	+5.0 ± 1.6	+2.47 ± 0.55	+6.9 ± 1.6	+24 ± 62
New York	*+2.0 ± 2.2*	*+1.0 ± 1.1*	-5.2±2.3	-1.45±0.64	*+2.3 ± 3.5*	*+0.7 ± 1.1*	-5.7 ± 2.3	-2.6 ± 1.0	*+1.1 ± 5.1*
Osaka	*+0.1 ± 1.3*	*+0.1 ± 1.2*	-2.1±1.1	-1.23±0.61	*-0.2 ± 1.5*	*-0.13 ± 0.90*	-2.52 ± 0.98	-2.21 ± 0.86	*-1.7 ± 2.3*
Paris	*-4.3 ± 3.5*	*-2.8 ± 2.2*	-5.8±2.6	-2.3±1.0	*-7.7 ± 4.6*	*-3.0 ± 1.8*	-5.1 ± 2.4	-3.3 ± 1.6	*-2.0 ± 3.6*
Riyadh	+1.29 ± 0.45	4.0 ± 1.4	+2.51±0.49	+3.49±0.68	+2.60 ± 0.75	+3.6 ± 1.0	+2.06 ± 0.39	+7.0 ± 1.3	*-3.3 ± 1.9*
São Paolo	*+0.08 ± 0.65*	*+0.2 ± 1.6*	*+0.53±0.58*	*+0.62±0.69*	*+0.16 ± 0.93*	*+0.2 ± 1.1*	*+0.36 ± 0.51*	*+0.9 ± 1.3*	-3.54 ± 0.92
Seoul	*-1.3 ± 2.6*	*-0.8 ± 1.6*	*+0.6±2.1*	*+0.21±0.72*	*-1.4 ± 3.5*	*-0.5 ± 1.2*	*+1.0 ± 1.8*	*+0.7 ± 1.2*	*+2.3 ± 3.4*
Shanghai	+10.0 ± 3.9	+9.9 ± 3.8	+12.8±3.2	+9.5±2.4	+12.1 ± 4.0	+8.8 ± 2.9	+9.3 ± 2.9	+9.1 ± 2.8	+12 ± 57
Shenzhen	*+3.2 ± 2.0*	*+3.0 ± 1.9*	-1.8±1.7	-1.2±1.1	+4.9 ± 2.2	+3.6 ± 1.7	*-2.2 ± 1.7*	*-1.7 ± 1.3*	*-2.6 ± 2.2*
Tehran	+1.94 ± 0.82	+5.2 ± 2.2	+2.49±0.74	+2.84±0.84	+3.05 ± 0.95	+3.6 ± 1.1	+2.66 ± 0.92	+7.7 ± 2.7	*+1.6 ± 6.3*
Tokyo	*-0.9 ± 1.7*	*-0.7 ± 1.3*	-5.5±1.4	-2.21±0.54	*-1.9 ± 2.3*	*-0.78 ± 0.96*	-5.3 ± 1.3	-3.72 ± 0.93	-2.97 ± 0.97