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Data assimilation of satellite retrieved ozone, carbon monoxide and nitrogen dioxide with ECMWF's Composition-IFS

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1 Validation datasets

Tropospheric CO data from the experiments are validated with profiles from the MOZAIC (Measurement of Ozone, Water Vapour, Carbon Monoxide and Nitrogen Oxides by Airbus In-serviceAircraft) programme (Marenco et al., 1998; Nedelec et al., 2003) taken during aircraft ascents and descents at various airports. The MOZAIC CO analyser is based on the Gas Filter Correlation principle of infrared absorption by the 4.67 μm CO band. MOZAIC CO data have a total uncertainty of ±5 parts per billion volume (ppbv), a precision of ±5 %, and a detection limit of 10 ppbv (Nedelec et al., 2003). We use MOZAIC profiles from Frankfurt (837 profiles) and Windhoek (323 profiles).

Tropospheric CO profiles and columns are further validated against Network for the Detection of Atmospheric Composition Change (NDACC) ground-based Fourier Transform Infrared spectrometer (FTIR) measurements (see http://ndacc.org). NDACC FTIR data are acquired according to formal measurement protocols, ensuring their traceability. The median random uncertainty of the FTIR data is 2-5 % for tropospheric columns and about 10-25 % at individual profile levels. They have the largest sensitivity in the mid and upper troposphere (and in the lower stratosphere which is not evaluated here). The model profiles are smoothed with the FTIR vertical averaging kernels and a-priori profile using Rodgers formula (Rodgers, 2000). For column comparisons, the model tropospheric vertical column between the NDACC station altitude and 10 km in molecules/cm^2 is obtained by integrating the smoothed model volume mixing ratio (VMR) profile over the pressure differences. The methodology was developed in the EU FP7 project NORS (Demonstration Network Of ground-based Remote Sensing Observations in support of the Copernicus Atmospheric Service, nors.aeronomie.be) and relies on validation methods described in Dils et al. (2006) and de de Laat et al. (2010). The NORS co-location and smoothing algorithms are described by Langerock et al. (2014). A list of the selected NDACC FTIR stations is shown in Table S2.

Surface O_3 and CO mixing ratios are compared against WMO Global Atmosphere Watch (GAW) observations at selected background stations (e.g., Oltmans and Levy, 1994; Novelli and Masarie, 2014). The GAW observations represent the global background away from the main polluted areas. Detailed information on GAW and GAW related O_3 and CO measurements can be found in GAW reports No. 209 (2013) and No. 192 (2010) respectively (http://www.wmo.int/pages/prog/arep/gaw/gaw_home_en.html). For detection of long-term trends and year-to-year variability, the data quality objectives (DQOs) for CO in GAW measurements can reach a maximum uncertainty between ± 2 ppbv and ± 5 ppbv for marine boundary layer sites and continental sites that are influenced by regional pollution (WMO (2010). For surface ozone an average uncertainty of ± 1 ppbv is quoted in WMO (2013). The stations used for the CO validations are listed in Table S3. The CO model values are interpolated in time to the instantaneous measurements and then averaged on a monthly basis. The procedure described in Flemming et al. (2009b) is applied to determine the model level used to compare the model field with GAW surface observations. This method is based on the difference between a high resolution orography and the actual station height. For O_3 3-hourly surface observations at 60 GAW stations (see Table S4) are used to calculate modified normalized mean biases (MNMB) and correlation coefficients from daily mean values.

Total column O_3 (TCO3) is validated against KNMI's multi sensor reanalysis (MSR, van der A et al., 2010) which is based on SBUV/2, GOME, TOMS, SCIAMACHY and OMI observations. All satellite retrieval products as used in the MSR were bias-corrected with respect to Brewer and Dobson Spectrophotometers to remove discrepancies between the different satellite data sets. The uncertainty in the product, as quantified by the bias of the observation-minus-analysis statistics, is of the order of 1 DU (van der A et al., 2010).

Stratospheric ozone fields are validated with version 3.0 retrievals of the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS, Dupuy et al., 2009). ACE-FTS observes the limb using the solar occultation technique, delivering up to 24 profiles per day. The previous version of these retrievals (V2.2) was extensively validated against 11 other satellite instruments, ozonesondes and several types of ground-based instruments (Dupuy et al., 2009). This validation found a slight positive bias with mean relative differences of about 5% between 15 and 45 km and reported that with version 3.0 this slight positive bias in the stratosphere had been removed. With respect to precision, the same study found that the de-biased standard deviation of the mean relative differences between ACE-FTS V2.2 and ozonesondes fell within 12 to 15% (17 to 30%) above (below) 20 km.

We use for further validation the MIPAS ozone profiles retrieved by version 6 of the operational ESA processor (Raspollini et al., 2013). MIPAS is a limb-viewing high-resolution Fourier-transform spectrometer that measured atmospheric emissions in the near to mid-infrared part of the spectrum (4.15 microns to 14.6 microns), allowing the retrieval of concentration profiles of O_3 and other trace gases between about 0.1 to 200 hPa. The random and systematic errors for O_3 are between 5 and 10% for large parts of the profiles, but larger near the boundaries of the retrieval range. Even though MIPAS profiles are assimilated in CIFS-AN and therefore not an independent data set, they are used for validation too, because the good consistency between the ACE and MIPAS data give extra credibility to the validation results.

Ozonesondes are used to validate stratospheric and tropospheric ozone from the experiments. The ozonesonde data used for the validation are acquired according to WMO-recommended standard operation procedures (SOP) and archived in a variety of data centres: World Ozone and ULTaviolet Radiation Data Centre (WOUDC), Southern Hemisphere ADditional OZonesondes (SHADOZ), Network for the Detection of Atmospheric Composition Change (NDACC), and campaigns for the Determination of Stratospheric Polar Ozone Losses (MATCH). The precision of electrochemical concentration cell (ECC) ozonesondes is on the order of ±5% in the range between 200 and 10 hPa, between −14% and +6% above 10 hPa, and between −7%and +17%below 200 hPa (Komhyr et al., 1995). Larger errors are found in the presence of steep gradients and where the ozone amount is low. The same order of precision was found by Steinbrecht et al. (1998) for Brewer– Mast sondes. We average the available sondes in the areas: Arctic, North America, Europe, East Asia, Tropics, Antarctica (see Table S5 for more details about the sonde locations and numbers).

Tropospheric column NO_2 (TRCNO2) data from the experiments are compared with data retrieved from the GOME-2 instrument which measures in the ultra-violet/visible and near infrared part of the spectrum. The retrieval is based on the Differential Optical Absorption Spectroscopy (DOAS; Platt and Stutz, 2008) method using a 425 to 497 nm wavelength window (Richter et al., 2011) and the reference sector approach (e.g., Richter and Burrows 2002; Martin et al. 2002) applied by IUP-Bremen. Uncertainties in NO_2 satellite retrievals are large and depend on the region and season. The largest errors are usually found in winter at mid and high latitudes, in regions affected by the Polar vortex due to the nature of the reference sector approach. As a rough estimate, systematic uncertainties in regions with strong pollution are on the order of ±20-30%. To allow a meaningful comparison to GOME-2 data, the model data are vertically integrated to TRCNO2, interpolated to satellite observation time and then sampled to match the location of cloud free satellite data. The latter have been gridded to match the model resolution. Finally, monthly averages of the daily GOME-2 and resulting model data are calculated in order to reduce any noise. Maps of TRCNO2 and timeseries for selected areas (see Table S6) are used for the validation.

Tropospheric NO_2 profiles are validated using ground-based multi-axis (MAX-) DOAS measurements performed in the Beijing city centre (39.98°N, 116.38°E). The period covered by these observations was from July 2008 to April 2009 but only data until December 2008 are included here. The retrieval tool and corresponding settings are extensively described in Hendrick et al. (2014). In brief, measured off-axis and zenith scattered light spectra are analysed using the DOAS method, providing O_4 (oxygen dimer) and NO_2 slant column densities (SCDs). In a second step, aerosol extinction coefficient and then NO_2 vertical profiles are retrieved for each MAX-DOAS scan by applying the OEM (Optimal Estimation Method)-based profiling algorithm bePRO to the corresponding sets of measured O_4 and NO_2 SCs, respectively. The retrieval of aerosol vertical profiles is needed since the light path length through the atmosphere (and thus the measured NO_2 SCDs) strongly depends on the aerosol content. The examination of the averaging kernels shows that the MAX-DOAS measurements are sensitive to the NO_2 vertical distribution up to ~1km altitude (see Hendrick et al., 2014). The validation methodology is essentially the same as for the NDACC FTIR CO.

2 Extra tables

Table S1: Main differences in CIFS-AN and REAN setup

	CIFS-AN	**REAN**
Model	C-IFS CB05	MOZART
Chemistry	In built chemistry. Tropospheric chemistry scheme. Stratospheric ozone parametrization.	CTM coupled to IFS. Tropospheric and stratospheric chemistry scheme.
Assimilated CO data	MOPITT TCCO	MOPITT TCCO IASI TCCO from Apr - Oct 2008
Assimilated O_3 data	MIPAS, MLS, OMI, SCIAMACHY, SBUV/2	MLS, OMI, SCIAMACHY, SBUV/2
Assimilated NO_2 data	OMI TRCNO2	SCIAMACHY TRCNO2
Data assimilation	NO_2 control variable Modified vertical correlations of O_3 background errors New CO background errors	NOx control variable
Fire emissions	GFAS v1.0	GFED 3
Anthropogenic emissions	MACCITY with enhancement factors of traffic CO over North America and Europe following Stein et al. (2014)	MACCITY

Table S2: List of NDACC FTIR stations used for validation in this paper

Station	**Region**	**PI**	**Latitude, Longitude [º,º]**	**Altitude [m]**
Eureka	Canada	UT	80.0, -86.2	610
Jungfraujoch	Switzerland	ULG	46.5, 8.0	3580
Izaña	Tenerife	FZK	28.3, -16.5	370
Lauder	New Zealand	NIWA	-45.0 169.7	370

Table S3: List of GAW CO stations used for validation in this paper

Station	**Latitude, Longitude [º,º]**
Alert	82.5, -62.5
Mace Head	53.3, -9.9
Key Biscayne	25.7, -80.2
Ascencion Island	-7.9, -14.4
Samoa	-14.2, -170.6
South Pole	-90.0, -24.8

Table S4: GAW stations used for the validation of surface O_3 data

Station Num.	GAW_id	Model Level	Latitude [º]	Longtitude [º]	region
1	alt	60	82.45	-62.52	Arctic
2	sum	57	72.57	-38.48	Arctic
3	brw	59	71.32	-156.61	Arctic
4	pal	55	67.97	24.12	Arctic
5	vdl	59	64.25	19.76	Arctic
6	ice	60	63.4	-20.28	Arctic
7	wes	60	54.93	8.32	NH-ML
8	zgt	60	54.43	12.73	NH-ML
9	mhd	60	53.33	-9.90	NH-ML
10	kmw	60	53.33	6.28	NH-ML
11	ngl	59	53.17	13.03	NH-ML
12	lgb	60	52.8	10.77	NH-ML
13	est	60	51.67	-110.2	NH-ML
14	bra	60	50.2	-104.71	NH-ML
15	cps	60	49.82	-74.98	NH-ML
16	ela	60	49.67	-93.72	NH-ML
17	ssl	52	47.92	7.92	NH-ML
18	sat	60	48.78	123.13	NH-ML
19	zsf	48	47.42	10.98	NH-ML
20	rig	59	47.06	8.45	NH-ML
21	snb	48	47.05	12.95	NH-ML
22	alg	57	47.03	-84.38	NH-ML
23	pay	60	46.82	6.95	NH-ML
24	jfj	47	46.55	7.99	NH-ML
25	zrn	55	46.43	15.00	NH-ML
26	kvv	50	46.3	14.53	NH-ML
27	kvk	57	46.12	15.1	NH-ML
28	prs	46	45.93	7.7	NH-ML
29	puy	51	45.77	2.97	NH-ML
30	irb	58	45.57	14.87	NH-ML
31	kej	58	44.43	-65.2	NH-ML
32	egb	60	44.23	-79.78	NH-ML
33	cmn	47	44.18	10.7	NH-ML
34	pdm	47	42.94	0.14	NH-ML
35	beo	47	42.18	23.59	NH-ML
36	thd	59	41.05	-124.15	NH-ML
37	nwr	52	40.04	-105.54	NH-ML
38	ryo	57	39.03	141.82	NH-ML
39	glh	57	36.07	14.21	NH-ML
40	tkb	60	36.05	140.13	NH-ML
41	bmw	59	32.27	-64.88	Tropics
42	izo	46	28.3	-16.5	Tropics
43	pyr	48	27.96	86.81	Tropics
44	yon	60	24.47	123.02	Tropics
45	mnm	60	24.28	153.98	Tropics
46	ask	48	23.27	5.63	Tropics
47	mlo	43	19.54	-155.58	Tropics
48	cvo	60	16.85	-24.87	Tropics
49	rpb	58	13.17	-59.46	Tropics
50	smo	58	-14.23	-170.56	Tropics
51	cpt	57	-34.35	18.48	SH-ML
52	cgo	58	-40.68	144.68	SH-ML
53	bhd	60	-41.41	174.87	SH-ML
54	ldr	60	-45.04	169.68	SH-ML
55	ush	60	-54.83	-68.3	SH-ML
56	syo	60	-69	39.58	Antarctica
57	nmy	60	-70.65	-8.25	Antarctica
58	dcc	59	-75.1	123.33	Antarctica
59	arh	58	-77.8	166.78	Antarctica
60	spo	58	-90	-24.8	Antarctica

Table S5: Ozonesonde sites used for the validation in various regions

Region	Area S/W/N/E	Stations (Number of observations)
Arctic:	60/-180/90/180	Alert (52), Eureka (83), Keflavik (8), Lerwick (49), Ny-Aalesund (77), Resolute (63), Scoresbysund (54), Sodankyla (63), Summit (81), Thule (15)
North America:	30/-160/60/-50	Boulder (65), Bratts Lake (61), Churchill (61), Egbert (29), Goose Bay (47), Kelowna (72), Narragansett (7), Stony Plain (77), Trinidad Head (35), Wallops (51), Yarmouth (61)
Europe	35/-20/60/40	Ankara (23), 3Barajas (52), DeBilt (57), Hohenpeissenberg (126), Legionowo (48), Lindenberg (52), Observatoire de Haute-Provence (47), Payerne (158), Prague (49), Uccle (149), Valentia Observatory (49)
East Asia	15/100/60/150	Hong Kong Observatory (49), Naha (37), Sapporo (42), Tateno Tsukuba (49)
Tropics	25/-180/25/180	Alajuela (48), Ascension Island (32), Hanoi (22), Hilo (49), Kuala Lumpur (24), Nairobi (44), Natal (48), Paramaribo (35), Poona (13), Reunion (37), Samoa (33), San Cristobal (28), Suva (28), Thiruvananthapuram (12), Watukosek (20)
Antarctic	-90/-180/-60/180	Davis (24), Dumont d'Urville (38), Maitri (9), Marambio (66), McMurdo (18), Neumayer (72), South Pole (65), Syowa (41)

Table S6: Areas used for the validation against GOME-2 NO_2 retrievals

Area	Area S/W/N/E [º]
East-Asia	20/100/45/145
Europe	35/-15/70/35
Eastern US	30/-120/45/-65
South-Africa	-20/15/0/45

3 Extra figures

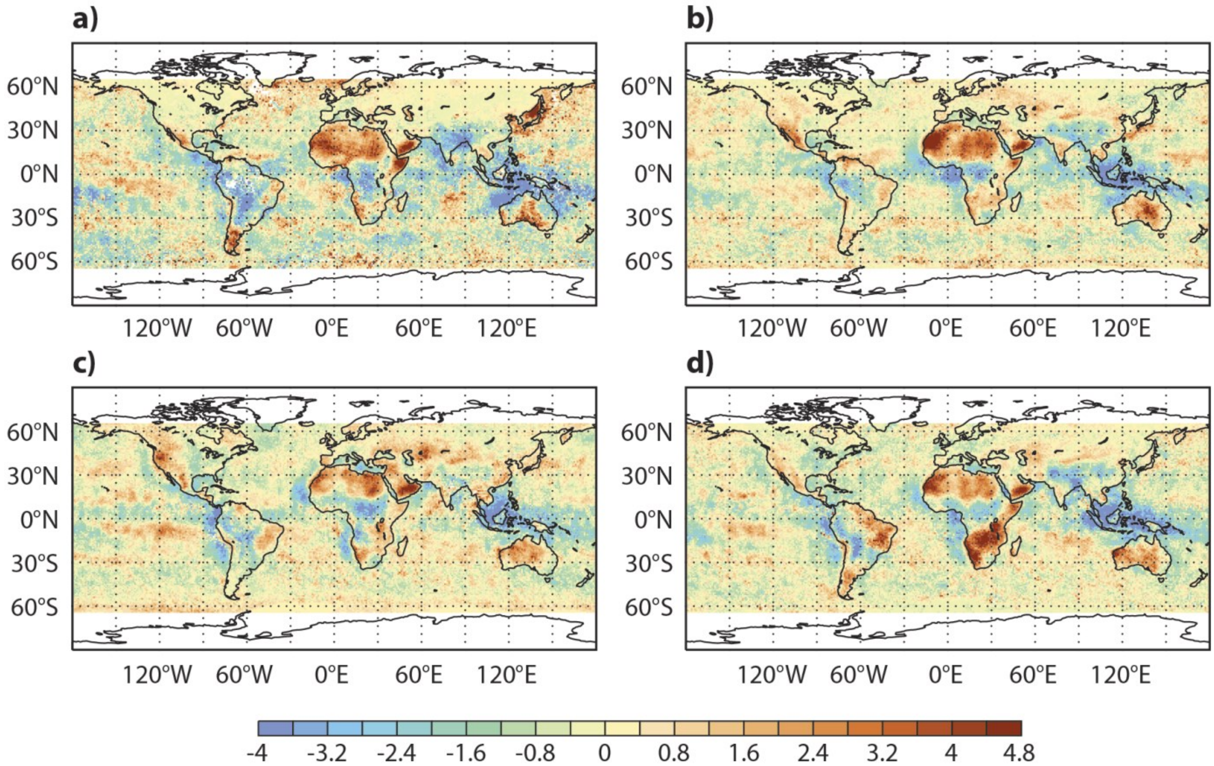

Figure S1: TCCO analysis increment (analysis minus forecast) in % from CIFS-AN averaged over (a) JF, (b) MAM, (c) JJA and (d) SON 2008. Red indicates positive values, blue negative values.

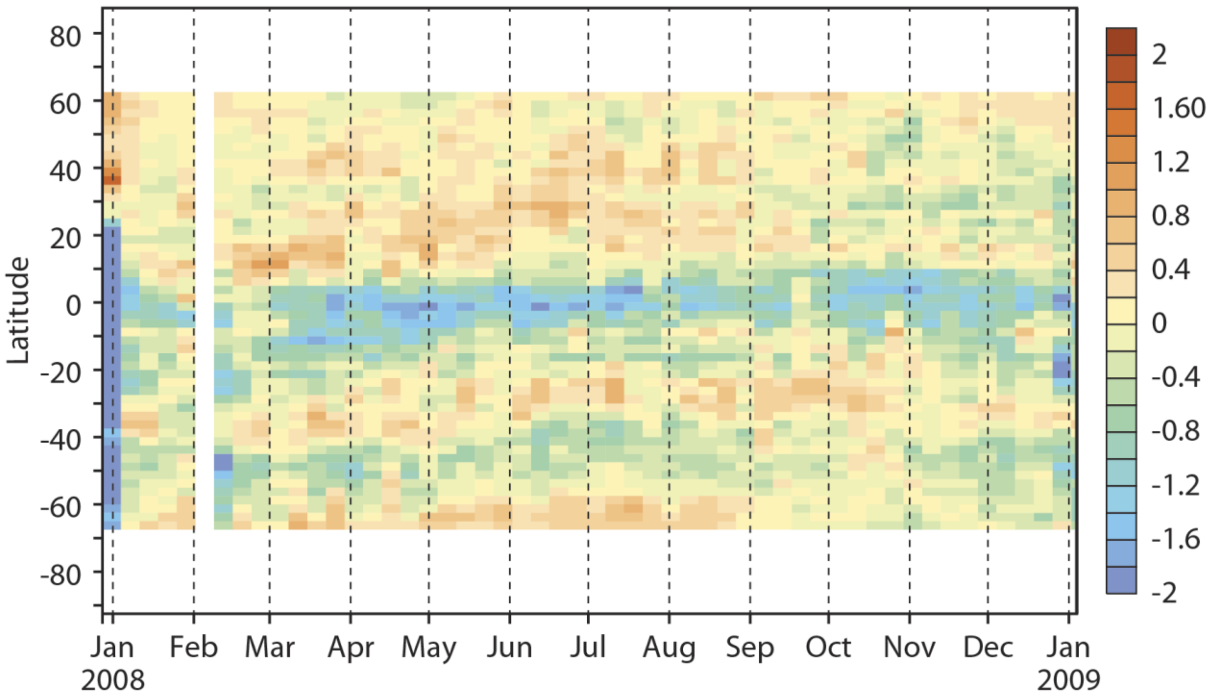

Figure S2: Timeseries of weekly averaged zonal mean MOPITT TCCO analysis increment (analysis minus forecast) in % for 2008. Red indicates positive values, blue negative values.

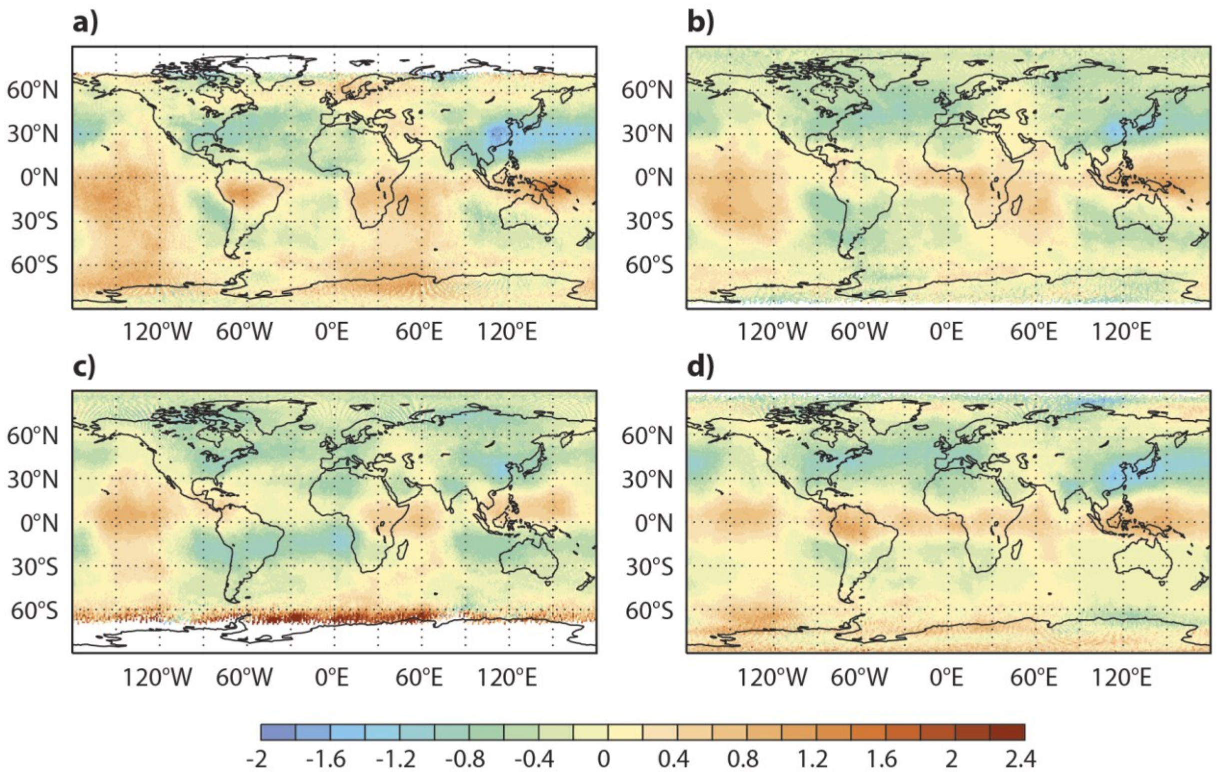

Figure S3: TCO3 analysis increment (analysis minus forecast) in % from CIFS-AN averaged over (a) JF, (b) MAM, (c) JJA and (d) SON 2008. Red indicates positive values, blue negative values.

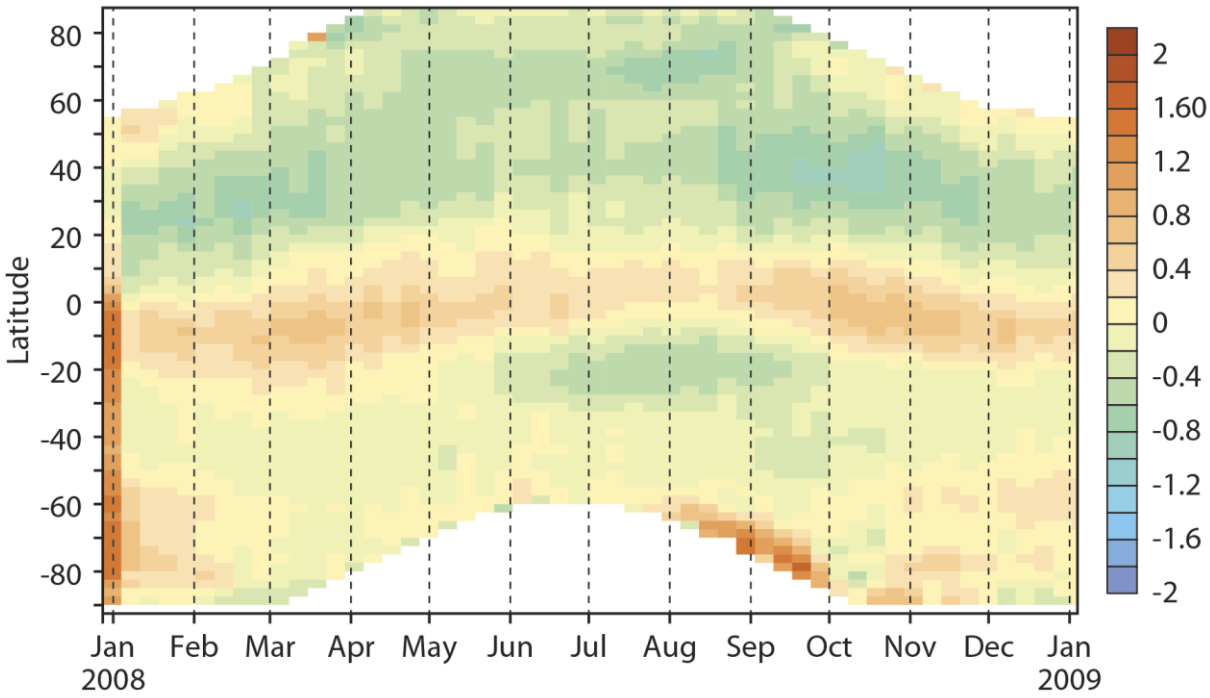

Figure S4: Timeseries of weekly averaged zonal mean OMI TCO3 analysis increment (analysis minus forecast) in % for 2008. Red indicates positive values, blue negative values.

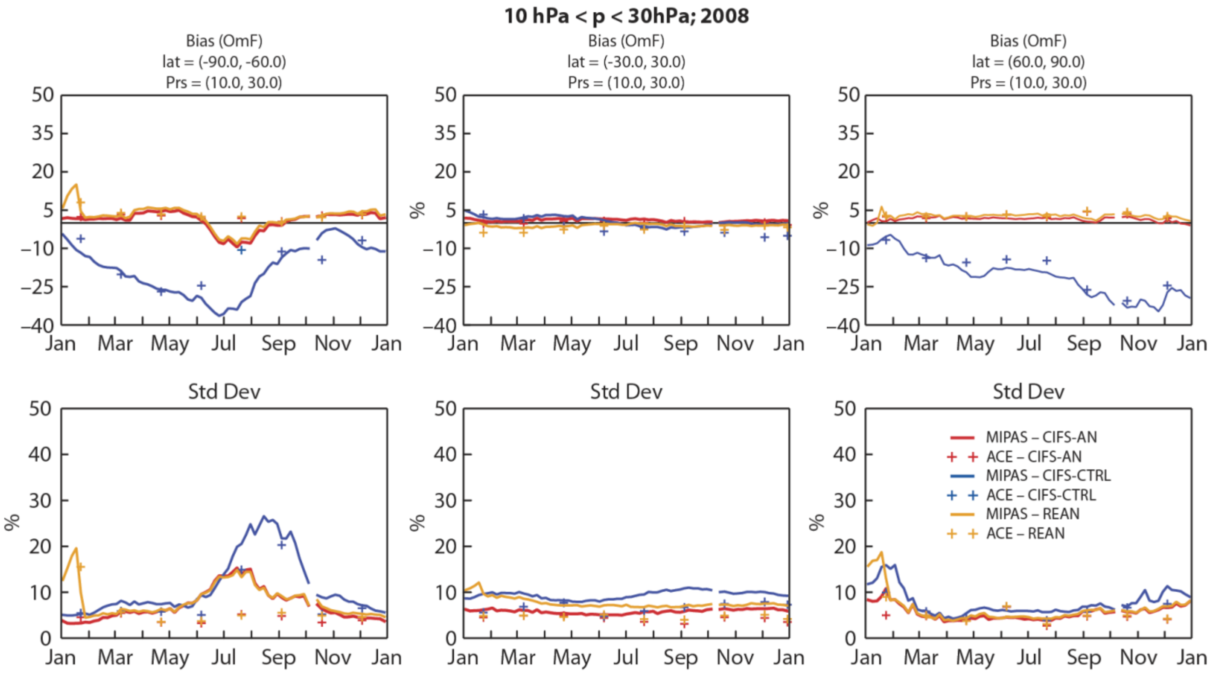

Figure S5: As in Figure 13 but for the pressure range between 10 and 30 hPa.

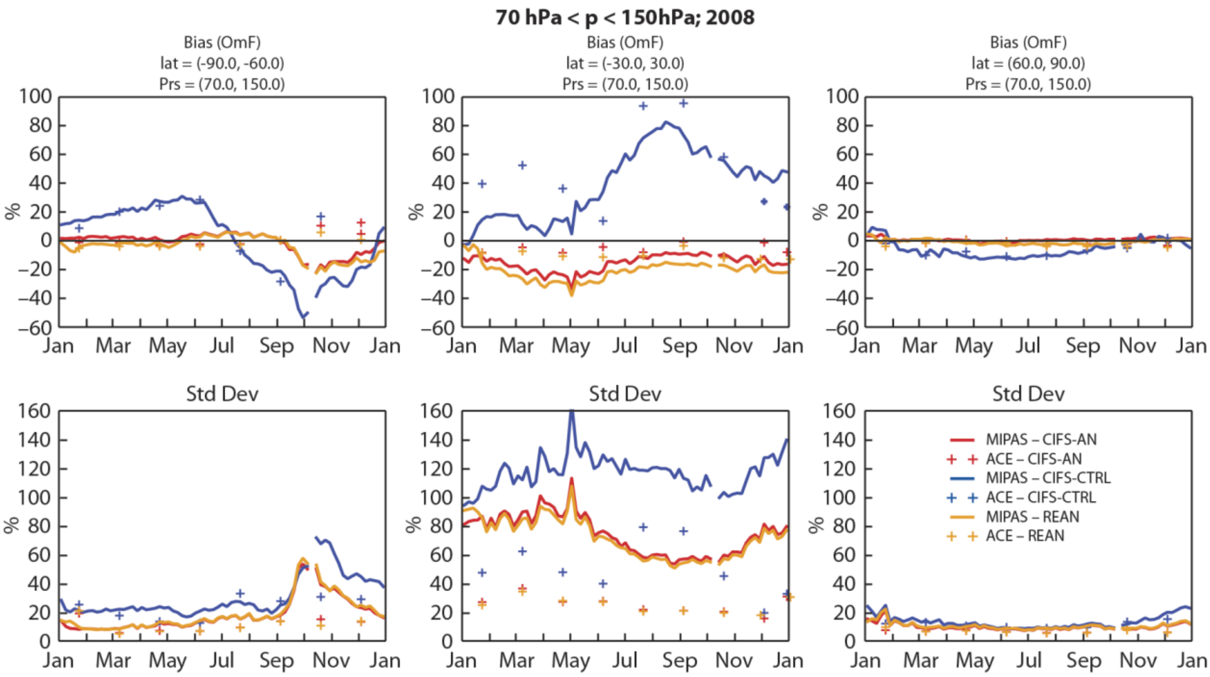

Figure S6: As in Figure 13 but for the pressure range between 70 and 150 hPa.