Technical Note: Latitude-time variations of atmospheric column-average dry air mole fractions of CO2, CH4 and N2O

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Keywords
techical, note, latitude, time, variations, atmospheric, column, average, ch4, dry, n2o, air, mole, fractions, co2

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Abstract. We present a comparison of an atmospheric general circulation model (AGCM)-based chemistry-transport model (ACTM) simulation with total column measurements of CO₂, CH₄ and N₂O from the Total Carbon Column Observing Network (TCCON). The model is able to capture observed trends, seasonal cycles and inter-hemispheric gradients at most sampled locations for all three species. The model-observation agreements are best for CO₂, because the simulation uses fossil fuel inventories and an inverse model estimate of non-fossil fuel fluxes. The ACTM captures much of the observed seasonal variability in CO₂ and N₂O total columns (∼81 % variance, R > 0.9 between ACTM and TCCON for 19 out of 22 cases). These results suggest that the transport processes in troposphere and stratosphere are well represented in ACTM. Thus the poor correlation between simulated and observed CH₄ total columns, particularly at tropical and extra-tropical sites, have been attributed to the uncertainties in surface emissions and loss by hydroxyl radicals. While the upward-looking total column measurements of CO₂ contains surface flux signals at various spatial and temporal scales, the N₂O measurements are strongly affected by the concentration variations in the upper troposphere and stratosphere.

1 Introduction

Carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O) are the major atmospheric greenhouse gases, with a substantial fraction of their emissions coming from anthropogenic activities. Due to the rapid rise in their tropospheric concentrations and significant contribution to anthropogenic radiative forcing during the period of 1750–2005 (Forster et al., 2007 and references therein), these gases are being monitored by in situ (ground-based, airborne) and remote sensing (satellite, ground-based) measurements. Understanding these measurements with the help of chemistry-transport models (CTMs) is critical for interpreting changes in surface fluxes as well as identifying processes that affect flux variations (e.g., Prinn et al., 1990; Keeling et al., 1996; Dlugokencky et al., 2009). Recently, calibrated total column measurements of CO₂, CH₄ and N₂O have become available from the ground-based Total Carbon Column Observing Network (TCCON) (Wunch et al., 2010, 2011). However, recent model-observation comparisons of total columns of CO₂ (defined as X_CO₂) have found weaker seasonal cycles for the models at continental sites, which are attributed to model transport errors within the planetary boundary layer or in...
the stratosphere or model errors in the seasonal amplitude of surface fluxes (Yang et al., 2007; Keppel-Aleks et al., 2012; Saito et al., 2011; Niwa et al., 2011). Further understanding of the contributions of the tropospheric and stratospheric partial columns to the total columns is required before these new data streams can be used in deriving surface fluxes, in particular for the reactive species (e.g., CH₄, N₂O). The reactive species exhibit a greater decrease rate of concentration with altitude in the stratosphere compared to that of the photochemically inert species, e.g., CO₂.

Parker et al. (2011) compared the TCCON and GOSAT (using the modified retrieval algorithm of the Orbiting Carbon Observatory mission) XCH₄ for a whole year within 2009–2010 with a CTM simulation and found that their simulations were lower than TCCON retrievals by ∼30 ppb. Butz et al. (2011) have found that the GOSAT retrievals, by the Netherlands Institute for Space Research (SRON) and Karlsruhe Institute of Technology (KIT), are within ±1.5 ppm (in the range of −0.27 % to +0.49 %) and ±6 ppb (range: −0.53 % to +0.22 %) of the TCCON observations for XCO₂ and XCH₄, respectively. Comparison of TCCON and GOSAT retrievals at National Institute of Environmental Studies (NIES) reveal that the GOSAT XCO₂ and XCH₄ are biased low by 8.85 ppm and 20.4, respectively. These recent inter-comparisons of total columns from remote sensing instruments and models have prompted us to evaluate the simulations of XCO₂, XCH₄ and XN₂O by the Center for Climate System Research/National Institute for Environmental Studies/Frontier Research Center for Global Change (CCSR/NIES/FRCGC) AGCM-based CTM (hereinafter, ACTM; Patra et al., 2009; Ishijima et al., 2010; Patra et al., 2011).

Here we compare the simultaneous forward ACTM simulations of XCO₂, XCH₄ and XN₂O with TCCON observations, with the main aims of understanding possible causes for the offsets found between model and observations, and the differences between the seasonal cycles among multiple species at a variety of locations. The use of multiple species, with unique properties of their sources, sinks and photochemical loss processes, are shown to be useful for disentangling errors in model total columns due to the surface fluxes and model transport. Such segregation of processes contributing to the total columns measured by remote sensing instruments are required for assimilating this set of observations for source/sink estimations.

2 Model, observation and analysis method

We use the CCSR/NIES/FRCGC AGCM-based chemistry-transport model (i.e., ACTM), which has been developed for simulating the major long-lived greenhouse gases, such as CO₂, CH₄ and N₂O (Patra et al., 2011a, b; Ishijima et al., 2010 and references therein). The ACTM simulations are conducted at T42 spectral truncations in the horizon-
Table 1. List of the TCCON sites used in this study. A comparison between the ACTM and TCCON time series presents the correlation coefficient $R$, model bias $b$, model-data difference $d$, 1-$\sigma$ standard deviation of residuals (RSD) for observations. We provide three $R$ corresponding to the full time series (TS; Figs. S1–S3 in the Supplement), fitted seasonal cycle (SC; Fig. S5–S7 in the Supplement) and residuals (RS; Figs. S8–S10 in the Supplement) for the observed and model time series.

<table>
<thead>
<tr>
<th>Site</th>
<th>Latitude</th>
<th>Longitude</th>
<th>X$_{CO_2}$, Ppm</th>
<th>X$_{CH_4}$, Ppb</th>
<th>X$_{N_2O}$, Ppb</th>
</tr>
</thead>
<tbody>
<tr>
<td>EUR</td>
<td>80.1°N</td>
<td>86.4°W</td>
<td>0.00</td>
<td>1.4</td>
<td>1.1</td>
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<tr>
<td>SOD</td>
<td>67.4°N</td>
<td>26.6°E</td>
<td>0.91</td>
<td>1.0</td>
<td>1.5</td>
</tr>
<tr>
<td>BIA</td>
<td>53.2°N</td>
<td>23.0°E</td>
<td>0.88</td>
<td>0.93</td>
<td>0.38</td>
</tr>
<tr>
<td>BRE</td>
<td>53.1°N</td>
<td>8.8°E</td>
<td>0.90</td>
<td>0.98</td>
<td>0.25</td>
</tr>
<tr>
<td>KAR</td>
<td>49.1°N</td>
<td>8.4°E</td>
<td>0.77</td>
<td>0.95</td>
<td>0.02</td>
</tr>
<tr>
<td>ORL</td>
<td>48.0°N</td>
<td>2.1°E</td>
<td>0.96</td>
<td>0.90</td>
<td>-0.03</td>
</tr>
<tr>
<td>GAR</td>
<td>47.5°N</td>
<td>11.1°E</td>
<td>0.85</td>
<td>0.96</td>
<td>0.08</td>
</tr>
<tr>
<td>LEF</td>
<td>45.9°N</td>
<td>90.3°W</td>
<td>0.95</td>
<td>0.98</td>
<td>0.02</td>
</tr>
<tr>
<td>LAM</td>
<td>36.6°N</td>
<td>97.5°W</td>
<td>0.88</td>
<td>0.97</td>
<td>0.33</td>
</tr>
<tr>
<td>TKB</td>
<td>36.1°N</td>
<td>140.1°E</td>
<td>0.86</td>
<td>0.97</td>
<td>0.31</td>
</tr>
<tr>
<td>JPL</td>
<td>34.2°N</td>
<td>118.1°W</td>
<td>0.93</td>
<td>1.00</td>
<td>0.06</td>
</tr>
<tr>
<td>KAO</td>
<td>28.3°N</td>
<td>16.5°S</td>
<td>0.96</td>
<td>0.99</td>
<td>0.20</td>
</tr>
<tr>
<td>DAR</td>
<td>12.4°S</td>
<td>130.9°E</td>
<td>0.92</td>
<td>0.58</td>
<td>0.30</td>
</tr>
<tr>
<td>WOL</td>
<td>34.4°S</td>
<td>150.9°E</td>
<td>0.87</td>
<td>0.72</td>
<td>0.40</td>
</tr>
<tr>
<td>LAU</td>
<td>45.1°S</td>
<td>169.7°W</td>
<td>0.93</td>
<td>0.63</td>
<td>-0.04</td>
</tr>
</tbody>
</table>

$^a$ X$_{N_2O}$ are not measured at these sites during the period of this analysis

$b$ time series too short for fitting, and thus no calculation of SC and RS components is presented

(RAU), Orleans (ORL), Park Falls (LEF), Sodankylä (SOD), Tsukuba (TKB) and Wollongong (WOL).

Model equivalents of the TCCON $X_y$ are derived taking the measurement column averaging kernels for the tracer $y$($CO_2$, CH$_4$ or N$_2$O) into account. ACTM-simulated profiles of the tracer mole fraction, $x_{m}(P)$, are extracted for each site. Following Rodgers and Connor (2003), the tracer mole fraction profile $x(P)$ which is integrated to derive the tracer column abundance is given by:

$$x_j = x_{a,j} + A_{j}(x_{m,j} - x_{a,j})$$ (1)

where $x_{a,j}$ is the TCCON retrieval a priori and $A_{j}$ is the TCCON column averaging kernel (a function of solar zenith angle) on the $j$-th pressure level. The tracer total column abundance is then divided by the corresponding ACTM dry air column abundance to infer the column dry-air mole fraction (DMF; Eqs. A6 and A7 in Wunch et al., 2011). All the ACTM results are adjusted by an offset for X$_{CO_2}$, X$_{CH_4}$ and X$_{N_2O}$ (ACTM-TCCON = 0 ppm, 25.6 ppb and 3.2 ppb, respectively) to match the average TCCON concentrations at Lauder. Imbalance in surface emissions and loss rates over the time of simulations lead to these offsets in CH$_4$ and N$_2$O values. This offset correction is made at the southern-most TCCON site, because the main focus of this study is to understand the seasonal and latitudinal distribution of these species for column distributions.

The partial column (PC) of DMF for the tracer $y$ are calculated as

$$X_{y,tropo} = PC_{y,tropo}/(P_s - P_t)$$ (2)

$$X_{y,strato} = PC_{y,strato}/P_t$$ (3)

where $PC_{y,tropo}$ is the partial column of the tracer from the surface ($P_s$) to the tropopause ($P_t$): $\int_{P_t}^{P_s} x(P) dP$ and $PC_{y,strato}$ is the partial column of the tracer from the tropopause ($P_t$) to the top of the atmosphere ($X_{H_2O}(P)$ assumed = 0 for simplicity) and the corresponding dry air partial columns are given by $\int_{P_{\text{max}}}^{P_t} x(P) dP$.

For quantitative evaluation of the model-observation agreement, we prepared following statistics: correlation coefficient $R$, model bias $b = [\Sigma(X_{ACTM} - X_{TCCON})]/N$; where $N$ is the number of data points in the time series, and standard deviations of model-data difference $d = [\sqrt{\Sigma(X_{ACTM} - X_{TCCON})^2}/N]}$. We use the digital filtering method described in Nakazawa et al. (1997) for decomposing the daily averaged original time series (model and observations separately) into long-term trend (periodicity...
Fig. 2. Monthly mean global distributions of the ACTM-simulated total column CO$_2$, CH$_4$ and N$_2$O (without applying averaging kernel and a priori smoothing) in January (left column) and July (right column) 2009.

During July, the average X$_{CO_2}$ values in mid-high latitudes (pole-wards of 45°) of both hemispheres show similar concentrations, with values in the tropics higher by about 4 ppm. The inter-hemispheric (IH) gradients (defined as NH minus SH) of X$_{CH_4}$ are about 100 ppb both in January and July, indicating CH$_4$ sources in the NH dominate sinks by the reaction with OH in all seasons. This is contrary to the CH$_4$ distribution near the Earth’s surface, where the CH$_4$ IH gradient is as strong as 250 ppb in January and much weaker (~150 ppb) in July (Patra et al., 2009). Despite higher CH$_4$ emissions in July, lower CH$_4$ concentrations near the surface are caused by the stronger vertical transport and loss of CH$_4$ due to its reaction with OH. Since the vertically transported CH$_4$ resides in the middle and upper troposphere, the differences in X$_{CH_4}$ with seasons are not distinct, compared to the surface concentrations. The X$_{N_2O}$ are always higher in the tropics by at least 5 ppb compared to the high latitudes (~60°) in both the hemispheres, and particularly low values are seen over the Antarctic region and Greenland reflecting...
the meridional gradient in tropopause height. During January the tropical upwelling branch of the Brewer-Dobson circulation is narrow and N$_2$O-rich air is transported deep in to the stratosphere (240 ppb isopleths reach beyond 10 mb) but not over wider tropical latitudes (Ishijima et al., 2010). Thus the peak in N$_2$O column over the equator is more flattened in July than in January.

Figure 3 compares the average values for 2009 and 2010 at each site is within ±1.0 ppm, except at Eureka (1.4 ppm), JPL (−1.4 ppm) and Izana (−1.2 ppm) (refer to Fig. 5 and Table 1 for detailed statistics).

The time series are highly correlated ($R > 0.8$), suggesting a realistic representation of transport and fluxes in ACTM for simulating X$_{CO_2}$ seasonal cycle and trends. This is despite the fact that interannual variability in non-fossil fluxes is not accounted for in ACTM, and that the fossil fuel emissions map is based on the EDGAR4 distribution for a single year (2005). The model-data differences are slightly smaller in the SH (∼1 ppm) than those at the NH continental sites (<1.8 ppm).

The goodness of model-observation agreement may also be defined with respect to the residual data variability at a particular site. The 1-σ standard deviation of residuals (RSDs) for the measured time series are given in Table 1. If model-data differences ($b$ and $d$) are less than the observed RSDs, the model-observation agreement can be considered good. We find, both $b$ and $d$ are generally smaller or similar in magnitude compared to the observed RSD at all sites for X$_{CO_2}$, X$_{CH_4}$ and X$_{N_2O}$.

Fig. 3. Observed (black) and simulated (blue) values of X$_{CO_2}$ (top), X$_{CH_4}$ (middle) and X$_{N_2O}$ (bottom) at the TCCON sites. Annual mean values for 2009 and 2010 are shown for this comparison (note that there are data gaps in time series for both years). The sites are arranged by latitude from Lauder to Eureka, located at the highest latitudes in the SH and NH, respectively.
Fig. 4. Comparisons of TCCON measurement (black dots) and ACTM-simulation smoothed by averaging kernels and a priori profiles (blue dots) for \(X_{\text{CO}_2}\), \(X_{\text{CH}_4}\), and \(X_{\text{N}_2\text{O}}\) (from left to right panels) at seven selected sites (corresponding names marked on the right for each row). The observational records at the other operational TCCON sites are short or do not cover all three species. The brown line represents the original/unsmoothed ACTM continuous time series with a 3-hourly time step. Comparisons of \(X_{\text{CO}_2}\), \(X_{\text{CH}_4}\), and \(X_{\text{N}_2\text{O}}\) from TCCON and ACTM are depicted for all measurement sites in Fig. S1, S2 and S3, respectively.

The time series of the ACTM \(X_{\text{CH}_4}\) (blue dots) compared with the subset of TCCON \(X_{\text{CH}_4}\) (black dots) is shown in the middle column panels in Fig. 4. The correlation coefficients between observed and simulated time series are generally greater than 0.3, except at Orleans and Tsukuba (Fig. 5 and Table 1). The model biases \(b\) are relatively large at Eureka (19.8 ppb) and Sodankyla (23.6 ppb), Lamont (−15.4 ppb), JPL (−10.5 ppb), Izana (−13.6 ppb), Darwin (−23.6 ppb) and Wollongong (−12.1 ppb), and less than ∼10 ppb at the other sites. Model-data comparisons of \(X_{\text{CO}_2}\) and surface \(\text{CH}_4\) concentrations (Patra et al., 2009, 2011b) would suggest ACTM inter hemispheric transport is not the source of relatively large biases in \(X_{\text{CH}_4}\). Rather, we speculate these are driven by errors in regionally varying emissions, tropospheric chemistry, and transport across the tropopause. The model-data differences \(d\) are less than 13 ppb (except at Sodankyla), which are more uniform across the sites than the more widely varying biases.

The right panels in Fig. 4 show \(X_{\text{N}_2\text{O}}\) time series of the ACTM (blue dots) and TCCON (black dots). The impact of applying averaging kernels and a priori profiles to ACTM \(X_{\text{N}_2\text{O}}\) is clearly seen in two ways: (1) the absolute values are adjusted higher and agree better with the TCCON measurements at the tropical and sub-tropical sites, and (2) the amplitude of daily and seasonal variability increases significantly (blue dots) from those without applying the averaging.
Larger bias and differences at Sodankyla for $X_{CH_4}$ and $X_{N_2O}$ are due to ACTM-TCCON differences in spring (Fig. S4 in the Supplement), which is a dynamically active period at this latitude, related to the movement of the Arctic polar vortex. The ACTM simulations show higher $N_2O$ concentration within the polar spring vortex compared to the measurements from limb-viewing satellites (Ishijima et al., 2010). The systematically high values for measured $X_{CH_4}$ and $X_{N_2O}$ compared to the simulation for sites between Wollongong and Lamont ($34.4^\circ$S to $36.6^\circ$N) may arise from higher tropical tropopause heights due to coarse horizontal resolution of the model failing to reproduce the sharp gradient in tropopause height between the equator and $30^\circ$ latitude (e.g., Patra et al., 2011b). It is also known that the $N_2O$ sources in the tropical regions as used in this simulation are underestimated (Kort et al., 2011), particularly in Southeast Asia, which can affect the $N_2O$ concentrations around Darwin significantly. However, the ACTM-simulated IH gradients for all three species are in close agreement with the TCCON measurements, e.g., the ACTM-TCCON differences in annual mean $X_{CO_2}$, $X_{CH_4}$ and $X_{N_2O}$ at all sites are generally within 1.5 ppm, 20 ppb and 2 ppb, respectively, and with no apparent seasonal bias (except at Sodankyla and Darwin).

### 3.3 Comparison of seasonal cycles and residuals

The seasonal cycles from the measured and simulated time series are obtained by removing long-term trends both from the fitted smooth curve and raw data as described in Sect. 2. Peak-to-trough amplitudes and the phase of the cycles in ACTM $X_{CO_2}$ are in very good agreement with those of TCCON $X_{CO_2}$ at the NH sites (Fig. S5 in the Supplement; $R$ for SC in Table 1). For example, no apparent mismatches can be found at the Tsukuba, JPL, Park Falls and Bremen sites for the 2007–2010 period. These results show better agreement than previous studies (Basu et al., 2011; Keppel-Aleks et al., 2012). This improvement is potentially associated with differences in the surface fluxes and transport model. At Orléans and Białystok, about a month delay in the ACTM $X_{CO_2}$ seasonal cycle phase is observed compared with that measured by TCCON. Noteworthy here is that the inverse model fluxes have only monthly time resolution and only four large regions over western Europe. European sites Bremen, Białystok and Orleans all fall in the same model region, and hence the model simulates the same phase of seasonality (in agreement with Bremen), while the TCCON measurements at those sites differ. An inversion at finer spatial and temporal resolution may help distinguish the flux heterogeneity between sites.

Among the sites consisting of more than one year of data coverage, we find the seasonal cycle amplitudes are overestimated by about 2 ppm at Izana and Lamont, unlike previous studies, which report an underestimation of $X_{CO_2}$ peak-to-trough amplitudes (e.g., Yang et al., 2007; Basu et al., 2011). In particular, when the seasonal cycle amplitude and phase

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**Fig. 5.** The model bias $b$ (black dots) and model-data difference $d$ (brown bars) for (a) $X_{CO_2}$, (b) $X_{CH_4}$, and (c) $X_{N_2O}$. Correlation coefficient $R$ (blue symbols) corresponds to the $y$-axis on right. No observations are available for $X_{N_2O}$ at Izana, Tsukuba, and Karlsruhe. Biases corresponding to four seasons are shown in Fig. S4.

[Diagram showing model bias and model-data differences for $X_{CO_2}$, $X_{CH_4}$, and $X_{N_2O}$ with correlation coefficients $R$.]
The extraction of CH$_4$ seasonal cycle information from the TCCON measurements is affected by the signal-to-noise ratio and data gaps (Fig. S6). At the SH sites, the daily and synoptic variabilities (residuals) are as large as the seasonal cycle, which affects the Darwin site most, followed by Wollongong and Lauder. It is also likely that some of the local and regional sources of CH$_4$ are not included in the ACTM simulations (e.g., Fraser et al., 2011). However, the relatively high correlation coefficients for residuals at Darwin and Wollongong indicate realistic representation of CH$_4$ emission distribution and synoptic transport in ACTM over Australia. At Park Falls and Lamont, dense data coverage leads to better seasonal cycle extraction, and we find that the ACTM and TCCON CH$_4$ time series are highly correlated. Generally, the correlation coefficient of the CH$_4$ time series between the ACTM and TCCON is lower than 0.7 at all sites, which is significantly less than for CO$_2$ or N$_2$O (Table 1).

The N$_2$O seasonal cycles are in excellent agreement ($R > 0.92$) at most sites except for NH high latitudes (Fig. S7; Table 1), suggesting the seasonal variations of N$_2$O loss rates in the stratosphere and the seasonal variation of tropopause height are fairly well represented in the ACTM. The residual variability, presumably due to synoptic variation in tropopause height and stratosphere-troposphere exchange events, is reasonably well represented at some (Bialystok, Bremen, Garmisch, Pasadena, Wollongong), but not all TCCON sites.

### 3.4 Characteristics of partial columns in the troposphere and stratosphere

Figure 6 shows the time series of modeled dry-mole fraction partial columns of CO$_2$, CH$_4$ and N$_2$O using Eqs. (2 and 3) for the period of 2007–2010. These comparisons suggest that most of the variability in the total columns of CO$_2$ and CH$_4$ is from the tropospheric column (Figs. 4 and 6). While the N$_2$O seasonal cycle amplitudes near the Earth’s surface are typically within 1 ppb, the total column abundances vary by more than 10 ppb with seasons. The larger fraction of the N$_2$O seasonal cycle amplitude is contributed by the stratospheric part of the column. The high correlation coefficients for ACTM and TCCON N$_2$O at all time scales and sites suggest that the ACTM is capable of simulating the variations in stratospheric photochemical loss (mostly seasonally varying) and daily-synoptic scale variability in transport in the upper troposphere and lower stratosphere (UT/LS) region. Note here that the N$_2$O averaging kernels are more sensitive to the UT/LS height region than in the troposphere, and that the
derived XN2O are sensitive to the stratosphere-troposphere exchange (STE) processes.

4 Conclusions

The ACTM-simulated dry-air column-averaged mole fractions of CO2, CH4 and N2O are compared with observed time series at 15 TCCON sites. TCCON measurement averaging kernels and retrieval a priori are taken into account to derive model equivalents of the TCCON observations. Weighting by the TCCON column averaging kernels has the largest impact on XN2O and its variability, compared with that for XCH4 and CO2. The model fairly successfully captures the seasonal cycle amplitude and phase as well as the inter-latitude gradients between most sites for all three species. The correlation coefficients for ACTM and TCCON XCO2 and XN2O are mostly over 0.9 for seasonal cycles. The model-observation differences (b and d) are mostly found to be below the observed residual variability in the observed time series. Our results suggest the measured XCO2 is sensitive to the surface flux heterogeneity between continental sites in Europe and North America and should have implications for inverse estimations of regional sources/sinks.

However, the model-observation comparisons of XN2O do not provide conclusive evidence of a surface emission signal in total column observations, which largely contain the signal of N2O variability due to tropopause altitude variability. The underestimation of ACTM XCH4 in comparison with TCCON observations clearly indicate a need for greater emissions at the Earth’s surface or reduction in tropospheric loss between the Wollongong (34°S) and Lamont (37°N) sites. The role of model transport uncertainty as the cause of this XCH4 underestimation is ruled out because ACTM can simulate fairly well the observed XN2O variations associated with the UT/LS and altitudes above. Thus the stratospheric contribution to the tracer column DMFs must be accounted for accurately if total column data are to provide useful constraints on the surface fluxes of trace gases with stratospheric photochemical sinks. Some test cases are available for separating the tropospheric partial column of CH4 using hydrogen fluoride (HF) stratospheric columns (Washenfelder et al., 2003). Similarly, another conserved quantity, the ‘age’ of stratospheric air, may also be used for estimating stratospheric partial columns (e.g., Saito et al., 2011).

Supplementary material related to this article is available online at: http://www.atmos-chem-phys.net/12/7767/2012/acp-12-7767-2012-supplement.pdf.

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