Global scale remote sensing of water isotopologues in the troposphere: representation of first-order isotope effects

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Abstract

Over the last decade, global scale datasets of atmospheric water vapor isotopologues (HDO) have become available from different remote-sensing instruments. Due to the observational geometry and the spectral ranges that are used, only few satellites sample water isotopologues in the lower troposphere, where the bulk of hydrological processes within the atmosphere take place. Here, we compare three satellite HDO datasets, two from the Tropospheric Emission Spectrometer (TES retrieval version 4 and 5) and one from SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY), with results from the atmospheric global circulation model ECHAM4 (European Center HAMburg 4). We examine a list of known isotopologue effects to qualitatively benchmark the various observational datasets. TES version 5 (TES\textsubscript{V5}), TES version 4 (TES\textsubscript{V4}), SCIAMACHY, ECHAM, and ECHAM convoluted with averaging kernel of TES version 5 (ECHAM\textsubscript{AK5}) successfully reproduced a number of established isotopologue effects such as the latitude effect, the amount effect, and the continental effect, but to different extent. The improvement of TES version 5 over version 4 was confirmed by the steeper latitudinal gradient at higher latitudes in agreement with SCIAMACHY. Other features of the water isotopologue cycle such as the seasonally varying signal in the tropics due to the movement of the Inter Tropical Convergence Zone (ICTZ) are captured in TES\textsubscript{V5} and SCIAMACHY. We suggest that the qualitative and quantitative tests carried out in this study could become benchmark tests for evaluation of future satellite isotopologue datasets.

1 Introduction

Heavy isotopologues of atmospheric water (principally HDO and H\textsubscript{2}\textsuperscript{18}O) are important tracers that are widely used to derive information on moisture recycling, cloud physics, troposphere–stratosphere exchange, climate change, and paleoclimate (Jouzel et al., 1997; Worden et al., 2006; Herbin et al., 2007; Uemura et al., 2008; Frankenberg et al.,...
Equilibrium and kinetic isotope effects in the hydrological cycle, associated mainly with evaporation, condensation, and diffusion can be measured (and modeled) with high precision.

Due to the high potential of these measurements, the Global Network of Isotopes in Precipitation (GNIP) has been operational for several decades (Aggarwal et al., 2007). Compared to this global scale international activity directed at precipitation, only very few measurements were directed at measuring water vapor, because of the logistical effort required for sampling water vapor using mechanical cold trap devices (e.g. Ehhalt et al., 1989; Franz and Röckmann, 2005). Nevertheless, the bulk of water in the atmosphere is in the vapor phase and the liquid fraction of atmospheric water amounts only to a very small percentage of the total water.

With the development of faster and more robust in-situ measurement methods for water vapor isotopologues, the number of measurements has been strongly increasing in the last years. Available techniques include tunable diode laser (TDL) absorption spectroscopy (Lee et al., 2005), in-situ FTIR (Fourier Transform Infrared) (Griffith et al., 2006), cavity ringdown spectroscopy (Gupta et al., 2009) and integrated cavity output spectroscopy (Wang et al., 2009; Sturm and Knohl, 2010). These techniques are now operational at several ground sites and has been installed on mobile platforms like balloons, ships, and aircraft. In addition, the ground-based FTIR remote sensing observations are made within MUSICA/NDACC (MUltiplatform remote Sensing of Isotopologues for investigating the Cycle of Atmospheric water/Network for the Detection of Atmospheric Composition Change, Schneider et al., 2006, 2012) and TCCON (Total Carbon Column Observing Network, Wunch et al., 2010) networks.

In addition, global water isotopologue data have become available using remote sensing techniques installed on-board satellite platforms: the Interferometric Monitor for Greenhouse Gases (IMG) on ADEOS (Zakharov et al., 2004; Herbin et al., 2007), the Tropospheric Emission Spectrometer (TES) on Aura (Worden et al., 2006), the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on Envisat (Payne et al., 2007; Steinwagner et al., 2007, 2010; Lossow et al., 2011), the SCanning Imaging
Absorption spectroMeter for Atmospheric CHartography (SCIAMACHY) on Envisat (Frankenberg et al., 2009; Scheepmaker et al., 2013), the Infrared Atmospheric Sounding Interferometer (IASI) on Metop (Herbin et al., 2009; Lacour et al., 2012; Schneider and Hase, 2011; Wiegele et al., 2014), and the Greenhouse gases Observing SATellite (GOSAT) launched by the Japanese Space Agency in January 2009 (Boesch et al., 2013; Frankenberg et al., 2013). These instruments are sensitive to different parts of the atmosphere. For example, MIPAS has high sensitivity in the upper troposphere and stratosphere as it is a limb thermal infrared sounder, TES is a nadir-looking thermal infrared sounder with high sensitivity from 850 hPa to 500 hPa (version 4) or from 900 to 425 hPa (version 5), while SCIAMACHY has high sensitivity throughout the column down to the surface as it is a nadir looking short-wave infrared (SWIR) sensor. The sensitivity of the retrieval to the true state of the atmosphere of these sensors is quantified by the averaging kernel (AK).

Adequate tools to investigate and use these global scale water isotopologue measurements are isotope-enabled atmospheric general circulation models (Iso-AGCMs), such as: ECHAM (Hoffmann et al., 1998), GISS-E (Goddard Institute for Space Studies, Schmidt et al., 2005), MUAGCM (Melbourne University General Circulation Model, Brown et al., 2006), IsoGSM (Isotopes-incorporated Global Spectral Model, Yoshimura et al., 2008), and LMDZ iso-GCM (the Laboratoire de Météorologie Dynamique atmospheric general circulation model with Zooming capability, Risi et al., 2010). These models integrate the well-known fractionation physics in the model's hydrological cycle. The main objective of water isotope studies is to test the parameterization of the hydrological cycle in AGCMs with isotope data as an independent and sensitive tracer of the model’s hydrology. Several studies demonstrated that these climate models could simulate the isotopic composition of meteoric water quite realistically compared with measurements in precipitation from the GNIP network (Hoffmann et al., 1998; Noone and Simmonds, 2002; Schmidt et al., 2007; Yoshimura et al., 2008).

In recent years, several studies have been carried out to investigate the use of new datasets of water isotopes retrieve from satellite measurements. Frankenberg
et al. (2009) compared the SCIAMACHY HDO measurements with the IsoGSM model. Yoshimura et al. (2011) extensively compared the HDO measurements from SCIAMACHY, and TES version 4 with IsoGSM results, Risi et al. (2012a) conducted an inter-comparison study between models and observations, both ground-based and from satellites. Recently Frankenberg et al. (2013) compared GOSAT with LMDZ model outputs.

When comparing satellite data with model results, the sensitivity of the satellite sensor to the different layers of the atmosphere has to be taken into account. For example, the TES\textsubscript{V4} dataset is sensitive only to a limited altitude range (mid troposphere, 850–500 hPa), in order to reduce the impact of non-linearities within the retrieval process (Worden et al., 2006). Therefore TES\textsubscript{V4} is not sensitive to humidity and isotopologues in the lower troposphere. A low sensitivity means that the measured signal receives a low weight in the retrieval compared with the a priori assumed profile.

The retrieval process provides us with a quantitative measure of how much the a priori profiles have been modified by the actual satellite observations, i.e. the averaging kernel (AK). For a meaningful satellite-model comparison, the retrieval’s AK must be applied to the model. The principle of applying an AK to the model output can be formulated as: $X_{\text{New GCM}} = X_a + A[X_{\text{GCM}} - X_a]$, where $A$ is the averaging kernel vector obtained from the satellite retrieval, $X_a$ is a priori information that is used for the satellite retrieval and $X_{\text{GCM}}$ is the original model field. If the satellite retrieval represents the atmospheric conditions perfectly ($AK = 1$), applying the AK has no effect on the model results. On the other hand, if the sensitivity of the satellite retrieval is small and thus the AK is low, applying the AK to the model will yield the a priori profile (as does the measurement in this case). Here, we compare the TES version 4 and 5 datasets to the ECHAM4 model output, which was convoluted with the respective AK of the two retrieval versions. Moreover, the results are compared with data from SCIAMACHY, which has high sensitivity near the surface. The focus of this paper is to establish to what degree the different datasets reproduce several well-established isotope signals (temporal and spatial variations).
The paper is organized as follows. In Sect. 2 we describe the SCIAMACHY, TES version 4 and 5 datasets, and the ECHAM4 model. The SCIAMACHY and TES $\delta D$ products are fundamentally different. For SCIAMACHY, the $\delta D$ total column is calculated from the retrieved HDO and H$_2$O columns while for TES, the $\delta D$ is very close to an optimal estimation product calculated from the spectral radiances. In Sect. 3 we describe several well-known isotopic effects of water vapor. In Sect. 4 we compare the remote sensing $\delta D$ datasets with each other and with the model results. In particular we discuss the consequences of applying the AK from both TES versions to the ECHAM4 model output. Conclusions are presented in Sect. 5.

2 Instruments and methods

2.1 SCIAMACHY data

The SCIAMACHY instrument aboard the European Space Agency (ESA) environmental research satellite ENVISAT measured between 2003 and 2012 near short-wave infrared spectra, which allows the retrieval of total column abundances of H$_2$O and HDO (Frankenberg et al., 2009; Scheepmaker et al., 2013). SCIAMACHY has high sensitivity throughout the column down to the surface. The SCIAMACHY spectrometer has relatively high spectral resolution (0.2 nm to 0.5 nm) and covers a wide wavelength range (240 nm to 1700 nm and 2000 nm to 2400 nm) with an apodized spectral resolution of 0.85 cm$^{-1}$, which enables SCIAMACHY to detect many different gases, clouds and aerosols. In addition, SCIAMACHY has three different viewing geometries, which are nadir, limb and sun/moon occultation. HDO data were retrieved from nadir measurements using a narrow wavelength interval from 2355 to 2375 nm (Frankenberg et al., 2009). The footprint area of an individual HDO measurement is 120 km $\times$ 30 km. HDO data used in this study are temporally averaged from 2003 to 2005. Detailed information about the retrieval procedure and data processing can be found in Frankenberg et al. (2009) and Scheepmaker et al. (2013).
2.2 TES data

The Tropospheric Emission Spectrometer (TES) aboard the Aura satellite is an infrared Fourier Transform Spectrometer (FTS), which measures the spectral infrared (IR) radiances between 650 and 3050 cm\(^{-1}\) in the limb and a nadir viewing mode. HDO and H\(_2\)O abundances were obtained from TES thermal radiances between 1200 and 1350 cm\(^{-1}\) (7400 to 8300 nm in wavelength) with an apodized spectral resolution of 0.1 cm\(^{-1}\) for the nadir view. The footprint is 5.3 km \(\times\) 8.4 km in the nadir-viewing mode. In this configuration, TES provides vertical information of abundant atmospheric species, such as O\(_3\), CO, CH\(_4\), H\(_2\)O and HDO (Worden et al., 2006). For the version 4 dataset, weighted mean values of the isotopic composition were provided for the height interval 500 to 850 hPa, where the HDO measurements have the highest sensitivity. Detailed information about TES data in general and the TES water isotopologue dataset can be found in Worden et al. (2004, 2006, 2007).

The TES retrieval version 5 has an improved sensitivity near the surface and covers the altitude range from 900 to 425 hPa (Worden et al., 2012). The TES version 5 data product is different from version 4 in many aspects, for example the aggregation of data by month, the reduced data size, the application of known corrections through validation campaigns, and the combination of all necessary meta-data in ancillary and multiple TES product files. The data used in this study are TES data measured in 2006 for both versions when TES had good spatial and temporal resolution. Filtering procedures have been applied to both versions with the same criteria, such as Degree Of Freedom signal, DOF > 0.5 and Species Retrieval Quality, SRQ = 1, in order to have an equal comparison.

2.3 The ECHAM4 model

The ECHAM atmospheric GCM was developed at the Max-Planck Institute for Meteorology in Hamburg. The ECHAM4 version used in this study was run with a spatial resolution of 2.8° by 2.8° (spectral resolution T42), with a vertical sigma-pressure hybrid
resolution of 19 layers. The ECHAM model uses a semi-Lagrangian advection scheme for both active tracers (e.g. moisture and cloud liquid water) and passive tracers (e.g. moisture and cloud water isotopologues). A detailed description of the incorporated water isotope physics can be found in Hoffmann et al. (1998). We note that the model considers fractionation effects at the surface only during evaporation from interception water and snow, however not from bare soil. Like other Iso-AGCMs, ECHAM distinguishes two types of fractionation processes: equilibrium and non-equilibrium fractionation. Equilibrium evaporation/condensation is a result of the different partial pressures of the water isotopologues and its description in the model is straightforward. Non-equilibrium effects play a major role during evaporation from open water (i.e. the oceans), during evaporation from falling raindrops below the cloud base and during ice crystal formation in an oversaturated environment. Its parameterization within the model is based on the different molecular diffusivities.

In the ECHAM results discussed here, the model wind fields were nudged to observational data (ERA40, Uppala et al., 2005). The ECHAM data used in this study correspond to the year 2001. Details of the satellite data and the model are summarized in Table 1.

2.4 Data processing

2.4.1 Unit

All HDO data from observations and model are presented as deviation (delta deuterium, $\delta D$) from the isotopic composition of the international standard Vienna Standard Mean Ocean Water (VSMOW, Craig, 1961) expressed in per mill (‰):

$$\delta = \frac{R_{\text{sample}}}{R_{\text{VSMOW}}} - 1 \quad (1)$$

$R_x$ refers to the $D/H$ ratio of the sample and of the reference material, respectively. Due to the low abundance of $D$, this is very similar to the observed and modeled
isotopologue ratio HDO/H₂O, which is retrieved by the satellite and modeled in the model, and the differences between the two are negligible for our study. Positive δ values indicate an enrichment of deuterium in the sample compared to the VSMOW standard, while negative values indicate depletion of D.

Since the water vapor mixing ratio and δD decrease with altitude, the total column value of δD is calculated as a weighted mean:

$$\delta D = \sum_{i=0}^{n} (\delta D_n \cdot H_2O_n) / \sum_{i=0}^{n} H_2O_n$$  \hspace{1cm} (2)

Where n is the number of layers. All results are given as weighted means ($\delta D$).

2.4.2 Application of satellite averaging kernel to model results

In order to compare the model results to satellite observations, the ECHAM model output was convoluted with the averaging kernel from the TES satellite datasets. Unlike for real atmospheric observations, in the model the “true state” is explicitly available. By applying the AK to the model results we mimic the way in which the satellite observes the atmosphere, to allow a meaningful comparison with the model.

The TES data were interpolated on the horizontal grid of the model, while in the vertical the model was interpolated on the TES layers. One should note that the AK convolution must account for the cross correlations in the joint HDO/H₂O profile retrieval (Yoshimura et al., 2011; Worden et al., 2006, 2011). The basic application of the AK to the model is presented in Eqs. (3) and (4). These equations can be re-formulated into Eq. (5) for the HDO/H₂O ratio, and to Eq. (6) for H₂O (see also Supplement of Risi...
ETEC = \( X_a + A_{TES}[X_m - X_a] \)  

\[ A_{TES} = \begin{bmatrix} A_{DD} & A_{DH} \\ A_{HD} & A_{HH} \end{bmatrix} \]  

\[ \ln \left( ECHAM^R_{AK} \right) = \ln (R_a) \]  

\[ + \left( (A_{DD} - A_{HD}) \cdot \ln \left( \frac{R_m}{R_a} \right) + (A_{DD} - A_{HD} - A_{HH} + A_{DH}) \cdot \ln \left( \frac{q_m}{q_a} \right) \right) \]  

\[ ECHAM^H_{AK} = X^H_a + A_{HH} \left( X^H_m - X^H_a \right) + A_{HD} \left( X^D_m - X^D_a \right) \]  

D and H stand for HDO and H\(_2\)O, respectively while m and a stand for the model field and the a priori field, respectively. \( R \) is the ratio of volume mixing ratios of HDO and H\(_2\)O (i.e. \( R_a = (\text{HDO}/\text{H}_2\text{O}) \) of TES a priori). \( q \) is the specific humidity; \( A_{DD} \) and \( A_{HH} \) are the averaging kernel sub-matrices for HDO and H\(_2\)O; \( A_{HD} \) and \( A_{DH} \) are the cross correlation AK matrices between H\(_2\)O and HDO and the reverse (see also Worden et al., 2006; Yoshimura et al., 2011; Risi et al., 2013). \( ECHAM^R_{AK} \) is the new HDO/H\(_2\)O ratio \( R \) as a function of pressure grid for the convoluted ECHAM.

The application of the averaging kernel to the model output has two components (see Eq. 5). The first depends on the difference between the a priori HDO/H\(_2\)O ratio profile and the model HDO/H\(_2\)O ratio profile. The second depends on the difference between the a priori humidity profile and the model humidity profile. If there is a model bias in humidity, the difference between model humidity and a priori humidity will affect the model-data comparison of \( \delta D \). The model humidity bias commonly occurs in the upper troposphere, also in the ECHAM model. This bias may then be propagated to the lower-tropospheric \( \delta D \) through the averaging kernels (Worden et al., 2012; Risi et al., 2013). The effect of the model humidity bias can be eliminated by assuming that the model captures correctly the satellite a priori humidity profile. Based on this
In the case of SCIAMACHY, we do not apply the AK’s to the model, since the AK for SCIAMACHY measurements is close to unity throughout the entire column and does not vary in time and space (Frankenberg et al., 2009; Yoshimura et al., 2011).

2.4.3 Bias correction

A bias of ~ 5% on average and ~ 15% maximum in the TES HDO vapor data has been attributed to uncertainties in the spectroscopic line strengths (Worden et al., 2006, 2007, 2011). A correction for this bias is already included in the version 5 dataset (Worden et al., 2012) but not in version 4. The sensitivity of the measurements must be accountd for in the application of the bias correction to the TES<sub>V4</sub> data (Lee et al., 2011; Risi et al., 2013) according to:

\[ X_{\text{HDO corrected}} = X_{\text{HDO original}} - A_{\text{DD}}(\delta_{\text{bias}}) \] (8)

\( X_{\text{HDO corrected}} \) is the logarithm of the volume mixing ratio of the HDO profile after bias correction, \( X_{\text{HDO original}} \) is the logarithm of the original volume mixing ratio of the model or satellite,
and $\delta_{\text{bias}}$ is a column vector of the same length as $X_{\text{original}}^{\text{HDO}}$ that contains the bias correction values. Note, that this correction is only applied to HDO and not to H$_2$O. We applied the bias corrections of 5% to TES$_{V4}$ (Worden et al., 2006) and no bias correction was applied to TES$_{V5}$ since it has already been corrected by 6.5% (Worden et al., 2012). In order to have a good agreement amongst the data in the tropic, bias corrections of 3% and 5‰ were applied to ECHAM$_{AK4}$ and SCIAMACHY.

3 The isotopic effects

Before presenting the results of our study, we briefly summarize the most important spatial and temporal isotope features and their underlying mechanisms. We focus in the following on $\delta D$; similar arguments hold for $\delta^{18}O$.

Since the start of water isotopologue studies in the 60s (Dansgaard, 1964) a number of empirical relationships between the isotopic composition of precipitation and several geographical or climatological parameters have been established. The principal “isotopic effects” such as the temperature effect, the amount effect, the altitude effect and the latitudinal effect will be used in the following to make a first order evaluation of the satellite data and the model results. The temperature effect denotes the spatial relationship between annual (or monthly or seasonal) mean temperatures and $\delta D$ of the respective precipitation (i.e. annual or monthly or seasonal mean). A linear relationship holds over a wide temperature range. The altitude effect and the latitudinal effect denote linear relationships between $\delta D$ and these geographical quantities. Since both quantities, i.e. altitude and latitude, correlate strongly with temperature, both effects are partly a direct consequence of the spatial temperature effect. However, there are some mechanisms involved that are independent of temperature. For instance, the triggering of strong convective activity next to orographic obstacles contributes to the altitude effect.

In the tropics (latitudes $< \pm 15^\circ$), the linear relation between surface temperatures and $\delta D$ becomes less apparent. More frequent convectively formed precipitation and
thus more vertical air movement seem to disturb the control of the temperature effect. However, in particular in tropical and subtropical regions, a relation between the amount of precipitation and $\delta D$ (the amount effect) appears in the data for which a full explanation is still under debate (Dansgaard, 1964; Aggarwal et al., 2007; Risi et al., 2008). Another geographical isotope effect is the continental effect. Along their trajectory across large continental land masses air is becoming disconnected from the oceanic water supply to compensate for the successive rain events. This leads again to a distillation effect with the heavier isotopes being progressively removed from these travelling air masses. The continental effect is therefore also related to the temperature effect, but adds an additional mechanism to the purely temperature controlled rainout processes. It is particularly pronounced along principal air mass trajectories, e.g. from the Gulf of Mexico into the Southwest of the United States or from the North Atlantic into Western and Central Europe.

4 Results and discussion

4.1 Spatial isotope distribution

The annual mean $\delta D$ isotope distributions of the different satellite and model datasets and the TES prior are shown in Fig. 1. The TES prior profiles as an initial guess for HDO and H$_2$O used to constrain the HDO/H$_2$O estimation does not show a latitudinal effect (Fig. 1a). The TES prior for HDO ($\text{HDO}_{\text{prior}}$) was calculated based on an estimated atmospheric H$_2$O profile from re-analysis data, multiplied by a single a priori profile of the HDO/H$_2$O ratio, which was obtained from a run of the National Center for Atmospheric Research (NCAR) Community Atmosphere Model (CAM), augmented with an isotope physics approach developed by Noone and Simmonds (2002) (Worden et al., 2006, 2012; Zhang et al., 2010). This prior HDO profile is representative for the tropical HDO/H$_2$O ratio and as a result, the TES $\delta D$ prior is strongly biased high at high latitudes. The TES prior does include an altitude effect (Fig. 1a), which is clearly...
visible in the Himalayas, the Andes, the Rocky Mountains, and Greenland. The prior for TES version 5 is not significantly different from version 4.

The SCIAMACHY prior shows the same pattern as the TES prior (Fig. 1i) but generally lower $\delta D$ values of $-140$ to $-160 \permil$. It does not include a latitudinal gradient and altitudinal effect. The sensitivity to the weak HDO absorber is close to unity throughout the column (Frankenberg et al., 2009; Scheepmaker et al., 2013). Unlike TES, the SCIAMACHY prior was constructed from ECMWF water vapor profiles and a fixed prior depletion profile. We assume $-100 \permil$ at the lowest layer and it increases to $-500 \permil$ at the highest layer. This results the prior total column $\delta D$ value of approximately $-150 \permil$.

Figure 1b–h and j presents modeled and observed patterns (isoscapes) of total column $\delta D$ of atmospheric water vapor. The first order isotope feature shown in all figures is the significant difference in $\delta D$ between low, mid and high latitudes roughly following global temperatures. The latitude effect is stronger in the observed global isotope pattern of the TES version 5 (TES$_{V5}$) and of the SCIAMACHY dataset than in the TES version 4 (TES$_{V4}$). It is also present in the modeled results of ECHAM. ECHAM results convoluted with the AK of TES version 4 (ECHAM$_{AK4}$) shows a smaller latitude gradient than for version 5 (ECHAM$_{AK5}$). Another feature, visible in these global isoscapes, is the tropical and sub-tropical zones with enriched values are wider in the ECHAM model results (Fig. 1b, e, f, h, j) compared to the TES$_{V5}$ and SCIAMACHY satellite observations (Fig. 1d and g).

A strong latitude effect of the order of $\sim 150 \permil$ for $\delta D$ between the tropics and the cold and isotopically depleted polar regions of both hemispheres is well established in the literature. Isoscapes based on an interpolated multi-variable regression of GNIP precipitation data show strong latitudinal gradients (Bowen and Wilkinson, 2002). Also the few existing near surface vapor measurements (Uemura et al., 2008) indicate a strong isotopic gradient between low and high latitudes. This principal geographical pattern is better represented in TES$_{V5}$ compared to TES$_{V4}$, which has a stronger influence from the a priori field at higher latitudes. It clearly indicates a substantial improvement of the version 5 retrieval over version 4.
The altitude effect is apparent in all datasets. Major mountain chains such as the Andes, Rocky Mountains or the Himalayas together with the Tibetan Plateau are easily recognizable by lower $\delta D$ values. $\text{ECHAM}_\text{AK5}$, however, is marked by unrealistically high $\delta D$ values over the Tibetan plateau (Himalayas; Fig. 1f). This unrealistic pattern is due to a high bias in the model humidity profile as discussed in Sect. 2.4 (Risi et al., 2012b, 2013) and the effect of limited vertical resolution and a priori constraints on retrieved $\delta D$ over the Himalaya region. Therefore, this problem disappears when we leave out the humidity term in the application of the AK (Eq. 7), and the resulting $\text{ECHAM}_\text{AK5BCorr}$ shows a local isotope minimum over the Tibetan Plateau again (Fig. 1h). Thus, the humidity bias can have a large effect on the isotope results of a model convoluted with the averaging kernel of a satellite instrument.

The unrealistically high $\delta D$ values over the Tibetan plateau also disappear if we perform the a posteriori analysis suggested by Schneider et al. (2012); Pommier et al. (2014) (Eqs. 16 and 17) (Fig. 1j). The a posteriori data treatment results $\delta D$ and H$_2$O profiles that are sensitive to the same atmospheric airmass. As a result, the AK application to the model with applied a posteriori analysis produces a similar result as the humidity correction (Fig. 1h and j). Latitudinal profiles from both $\text{ECHAM}_\text{AK5Corr}$ and $\text{ECHAM}_\text{AK5Pos}$ show very similar $\delta D$ values in the tropics and only 2–5‰ differences at the higher altitudes (figure not shown). Both correction procedures thus show that humidity biases have to be taken into account when the satellite AKs are applied to model results, either by a humidity correction or an a posteriori processing. Since the results from $\text{ECHAM}_\text{AK5Corr}$ and $\text{ECHAM}_\text{AK5Pos}$ are similar, we only plotted the results of $\text{ECHAM}_\text{AK5Corr}$ for the rest figures.

The isotopically most enriched regions of the atmosphere are situated over tropical South America and tropical Africa and are associated with the Amazon and Congo River basins. The location and extension of these maxima are relatively robust throughout all datasets. Since these large areas with $\delta D$ between −95 and −80‰ are not apparent in the TES and SCIAMACHY a priori fields (Fig. 1a and i) but clearly appear in the final product (Fig. 1c, d, and g), they are a robust result of the added information
from the satellite measurements and not an artificial product of the retrieval procedure. The intense recycling and very strong evapo-transpiration over these rainforest regions is responsible for this pattern (Worden et al., 2007; Yoshimura et al., 2011).

In the scientific literature, many examples of the continental effect are documented, e.g. the Southwestern US or Western/Central Europe, based on measurements in ground waters (Rozanski, 1985) and precipitation (Aggarwal et al., 2007). The continental effect appears in all datasets with varying intensity, except for ECHAM convoluted with AK version 4. TES\textsubscript{V4}, TES\textsubscript{V5}, SCIAMACHY, ECHAM, ECHAM\textsubscript{AK5}, ECHAM\textsubscript{AK5Corr}, and ECHAM\textsubscript{AK5Pos} results clearly show the continental effect (white arrows in Fig. 1d).

The Hadley-Walker Circulation defines the seasonally varying zones of strong convective activity in the tropics. Three rising branches of the meridional Walker circulation are situated over tropical South America, tropical Africa and the Western Pacific Warm Pool (Oort and Yienger, 1996). As described above, the strong enrichment over the continental parts of the Walker circulation is due to the intense recycling of continental water there. However, over the Pacific warm pool one recognizes a zone with slightly more depleted $\delta D$ (ellipses in Fig. 1b, d, f, g, and h). This area of very high Sea Surface Temperature (SST), strong convection and rainfall and more depleted vapor extends over the Inner Tropics and is surrounded by a zone of descending air, less rainfall (Oort and Yienger, 1996; Jo et al., 2014) and more enriched vapor. We consider this pattern as a manifestation of the isotopic amount effect since the isotopic pattern anti-correlates with regional rainfall (Brown et al., 2008; Dansgaard, 1964; Kurita et al., 2011; Lee and Fung, 2007; Lee et al., 2011; Risi et al., 2008; Worden et al., 2007). Apparently, TES\textsubscript{V4} and ECHAM\textsubscript{AK4} (Fig. 1c and e) do not pick up this important feature of the tropical water cycle.

Figure 2 presents a comparison of zonal mean $\delta D$ values for all datasets. As discussed above, $\delta D$ decreases to $-200$‰ in the TES\textsubscript{V4} product at high northern latitudes, whereas it decreases to $-250$‰ for TES\textsubscript{V5}, the latter in agreement with SCIAMACHY, and with other independent observations (Uemura et al., 2008). TES\textsubscript{V4}
displays much weaker $\delta D$ gradients at mid latitudes compared to SCIAMACHY and TES$_{V5}$. As mentioned earlier, the tropical and sub-tropical isotope maximum in the ECHAM model is significantly wider than for SCIAMACHY and TES$_{V5}$ (Fig. 2b). This issue is aggravated when the ECHAM results are convoluted with the TES averaging kernels. The ECHAM$_{AK4}$ product shows the smallest latitude gradient and highest $\delta D$ values at high latitude of all datasets. The small AK values of version 4 enhance the influence of the a priori field, which consequently leads to more enriched $\delta D$ values. This high-latitude problem has been improved in the version 5 datasets (Fig. 2b), but still the ECHAM$_{AK5}$ and ECHAM$_{AK5Corr}$ results are significantly higher than the model state of ECHAM at northern high latitudes. TES$_{V5}$ is in good agreement with SCIAMACHY.

A main reason for the improvement of TES$_{V5}$ latitude gradient is the greatly increased in sensitivity. The increased number of radiance measurements used for retrieval and the changed of both the hard constraints (e.g. retrieval levels and mapping matrices) and the soft constraints (e.g. constraint matrix) in the version 5 dataset improve the TES$_{V5}$ sensitivity (Worden et al., 2012). A good indicator for retrieval sensitivity is the DOF (Degree Of Freedom) satellite as it shows how well the satellite captures the range of variability of the true distribution. There are many version 5 datasets at higher latitudes that pass the applied DOF criteria (see Fig. 5 in Worden et al., 2012). Therefore, version 5 represents better the true natural variability of atmosphere at higher latitudes than version 4.

Although, there are significantly discrepancies at mid and high latitudes, all data products agree fairly well in a tropical/subtropical band between 30° N and 30° S with values around $-100$ ‰, similar to Webster and Heymsfield (2003), Lawrence et al. (2004) and Zakharov et al. (2004). It seems therefore that different remote sensing datasets and model results (with AK applied) coincide in the tropics, which mean that the isotope measurements there can be exploited to examine smaller scale effects (see below).
4.2 Temporal isotope distribution

Above we analyzed the spatial structure of the dominant annual average isotope patterns. In addition, both satellite data and model simulations allow us to study the seasonality of $\delta{}D$ patterns and to identify the leading processes on this time scale. Zonal means of all datasets are computed for the mean winter (DJF) and summer season (JJA) of the respective annual time period (Fig. 3). $\text{TES}_V^5$ shows similar results as SCIAMACHY during summer and winter in the Northern Hemisphere and both datasets show a clear and consistent seasonal isotope difference between the two seasons in the Northern Hemisphere. In the Southern Hemisphere, however, the difference between the two seasons is much smaller for both datasets. In addition, in the Southern Hemisphere there appears to be a significant difference between the instruments. $\text{TES}_V^5$ $\delta{}D$ values are consistently lower compared to SCIAMACHY (Fig. 3a).

Due to the high solar zenith angle (low sun), SCIAMACHY data are seasonally scarce at mid and high latitudes. Therefore, there are not many measurements above 50° N or 50° S in the respective winter season (Fig. 3a). Also, large parts of the oceans are not covered due to the low albedo of the ocean; therefore the signals from oceanic regions are primarily caused by reflection on low-level clouds. In the SCIAMACHY retrieval, a data filter is applied, which only accepts water isotopologue measurements if the $\text{H}_2\text{O}$ total column corresponds to at least 70% of the ECMWF total water column (Frankenberg et al., 2009). This constraint excludes profiles with high clouds, but accepts profiles with low clouds (up to 1 km). Thus all SCIAMACHY measurements are biased towards clear sky or low cloud conditions. The $\delta{}D$ seasonality of SCIAMACHY shown in Fig. 3a (especially in the Southern Hemisphere during the Northern Hemisphere Summer) is therefore neither temporally nor spatially fully representative of the mean state of the atmosphere. These issues are not applicable to TES data. However, TES measurements are also less sensitive at higher latitudes, especially the version 4 dataset.
The wider tropical maximum of ECHAM is seen in both seasons (Fig. 3b). It is apparent that in the summer season the ECHAM_{AK5} results show considerably higher δD values than TES_{V5} in the Northern Hemisphere and it is vice versa for the winter season in the Southern Hemisphere. This shows that the annual bias in ECHAM as discussed above originates to a large degree from the summer season. The mid latitudinal bias found in the ECHAM model is a common bias that is present in many GCM models. In general, most GCM models tend to overestimate humidity in the tropics and subtropics due to the inadequate representation of cloud processes or of the large-scale circulation, or to excessive diffusion during the transport of water vapor (Risi et al., 2012a, b). The overestimation of humidity in the subtropic affects the enrichment of δD in the mid and high troposphere.

Whereas there are still some discrepancies between the different data products at mid and high latitudes, isotope data within a zonal band from 30° N to −30° N are roughly consistent, both for the annual average and the summer and winter profiles. An interesting feature of all datasets except for TES_{V4} (not shown) is a seasonal seesaw behavior of the latitudinal δD profiles in the inner tropics: in both seasons, the δD values close to the equator are lower in the respective summer hemisphere than in the winter hemisphere. The absolute δD variations are small and extend over different ranges in the respective hemispheres, but the δD latitude profiles for the two seasons (solid and dashed lines of each color) intersect very close to the equator.

This seasonal variation of δD in the tropics is a robust feature of the TES_{V5} dataset and a consequence of the seasonal displacements of the ITCZ (Inter Tropical Convergence Zone). Tropical rainfall bands and corresponding convective activity closely follow the maxima of insolation. The ITCZ is displaced towards the north during Northern Hemisphere summer and towards the south during Southern Hemisphere summer. Due to the isotopic amount effect, we expect the convectively active regions within the ITCZ to be associated with lower δD values. Therefore, areas north (south) of the equator are isotopically more (less) depleted during NH summer, and vice versa during NH winter. The isotopic amount effect therefore leads to the seesaw behavior of the zonal
δD means where the latitudinal profiles in the two seasons have their lower values in the summer hemisphere (see the crossing lines in Fig. 4). This seesaw behavior has already been recognized in precipitation data (Waliser and Gautier, 2010; Wu et al., 2003; Back and Bretherton, 2005).

SCIAMACHY and TESV5, show this isotope feature with varying intensity (Fig. 4a). The amount effect due to the movement of ICTZ is more pronounced in SCIAMACHY than in TESV5. The original ECHAM model and ECHAM_{AK5Corr} also show this seesaw pattern (Fig. 4b).

A possible reason why SCIAMACHY shows this seesaw pattern more clearly is SCIAMACHY’s higher sensitivity at lower altitudes where many processes contributing to the amount effect occur (such as re-evaporation of raindrops in more/less humid air, etc). In order to investigate this further, we separated ECHAM, TESV5, and ECHAM_{AK5Corr} into two layers (Fig. 5). Figure 5b confirms that seesaw pattern in the TESV5 originates from the lower layer. This is qualitatively in line with the fact that SCIAMACHY is sensitive down to the surface and the seesaw pattern nicely shows up in the SCIAMACHY dataset. We note that this was not necessarily expected since the seesaw phenomenon is largely produced by displacements of the ITCZ over the oceans, where the SCIAMACHY coverage is relatively low (due to the requirement of low-level clouds) compared to the coverage over land. In contrast, ECHAM and ECHAM_{AK5Corr} show the seesaw pattern throughout the entire atmospheric column, both at the lower and high layers (Fig. 5a and c). It seems that the model overestimates the correlation between lower and higher layers than the observations. It was speculated that GCMs in general show strong coherence between processes at lower altitudes (such as Sea Surface Temperature variations) and associated features at high altitudes (such as high convective cloud formation). In this case the common isotope seasonality at low and high altitudes in the model might be a further consequence of these known model problems (Risi et al., 2012a; Conroy et al., 2013). The representation of this “fine structure” is an important feature, which needs further investigation in remote sensing datasets of water isotopologues. It should be noted that the cloud height issue might cause the
seesaw pattern seen in the satellite datasets. Satellites in general measure the $\delta D$ values above the clouds, which means during convection the satellite data will represent $\delta D$ at high altitudes where $\delta D$ is lower.

### 4.3 Relation of water vapor and $\delta D$

Already in 1964, Dansgaard (1964) described the global water cycle as a Rayleigh-type distillation model. Each condensation process extracts a certain quantity of water from an air mass, when it moves from its tropical/subtropical source regions to higher latitudes. Assuming that this process is exclusively temperature dependent and controlled by a moist adiabatic lapse rate one can simply quantify the isotopic temperature effect over a wide temperature range. It is instructive to check to what degree the atmosphere corresponds to this simple temperature dependent scheme: how Rayleigh-like is the earth’s atmosphere?

Decreasing temperatures from the surface to higher altitudes dry the corresponding air and lead to both lower total precipitable water (TPW), and via the Rayleigh mechanism, to more depleted isotope values. In a pure Rayleigh distillation system, $\ln(\text{HDO}/\text{H}_2\text{O})$ is linearly related to $\ln(\text{TPW})$ and the slope corresponds to the effective equilibrium fractionation, $\alpha_{\text{eff}} - 1$ (Frankenberg et al., 2009; Schneider et al., 2010; Yoshimura et al., 2011). Here, $\alpha_{\text{eff}}$ refers to a mean fractionation over the entire distillation process with an “effective fractionation temperature” (Frankenberg et al., 2009; Yoshimura et al., 2011).

In the following we investigate whether the atmosphere follows such a Rayleigh distillation model. Globally, a $\ln(\text{HDO}/\text{H}_2\text{O})$ vs. $\ln(\text{TPW})$ plot (see Fig. 6a and b) is dominated by the latitudinal gradient of both quantities (i.e. the latitude effect). Lower $\ln(\text{TPW})$ values and more depleted isotopic values are largely organized along a gradient from lower to higher latitudes. However, in the correlation plot also some clear deviations occur. At high $\ln(\text{TPW})$ (> 3), the different datasets agree rather well, but below 3, significant differences become apparent. Between 2 and 3 $\ln(\text{TPW})$, SCIAMACHY appears to have lower $\ln(\text{HDO}/\text{H}_2\text{O})$ values than ECHAM and the TES. Below
2 ln(TPW), SCIAMACHY also displays a group of very high \(\ln(\text{HDO}/\text{H}_2\text{O})\) values, which show a larger scatter and deviate strongly from linear correlation. The ECHAM model shows two separated branches at the lower end of the global distillation chain, which corresponds to air-masses over Antarctica and the Arctic (Fig. 6b). This feature is not reproduced by any available observational dataset since observations are scarce in these regions.

It is evident that these structures influence the linear trend lines that are fit to the datasets and shown in Fig. 6. Steeper overall slopes are simulated for the original ECHAM model than observed by the satellite, because the model results include strongly depleted water vapor at higher latitudes, especially at Polar Regions, where no satellite data are available. TES\textsubscript{V5} data results in a steeper slope than TES\textsubscript{V4}, which again reflects the smaller latitude gradient in TES\textsubscript{V4}, and convolution with the version 4 AK leads to a similarly low slope for ECHAM\textsubscript{AK4}. The slope for the ECHAM\textsubscript{AK5} dataset is lower than TES\textsubscript{V5}. However, both TES products disagree with ECHAM and SCIAMACHY for \(\ln(\text{TPW})\) lower than 2.5. The differences between datasets for lower \(\ln(\text{TPW})\) reflect the differences discussed in Sect. 4.1 for the latitudinal \(\delta D\) gradients at mid and high latitudes. In the following we focus on the tropical region (high \(\ln(\text{TPW})\)) where the latitudinal profiles agreed well.

In the tropics, the differences between the datasets in the \(\ln(\text{HDO}/\text{H}_2\text{O})\) vs. \(\ln(\text{TPW})\) correlation plot are smaller, but the correlation between \(\ln(\text{TPW})\) and the isotopes and the respective slope is lower also (Fig. 6c and d). This implies unrealistically high condensation temperatures for the satellite datasets (TES\textsubscript{V5} and SCIAMACHY) and even more for the ECHAM model (up to almost 60°C). Apparently, a description of the relatively small isotopic variance by a simple overall distillation process is hardly possible. This is likely due to the dominance of convective activity there, which affects the isotopic composition by many large scale (e.g. low level humidity confluence) and sub-scale processes (e.g. entrainment/detrainment of vapor in convective systems) that are not sufficiently described by Rayleigh fractionation. The isotope amount effect in
the tropics is another factor contributing to the relatively flat slope because it leads to lower δ values at higher humidity (Fig. 6d).

Figure 6e and f show ln(HDO/H2O) vs. ln(TPW) from satellites and model simulations over the Sahel region (0–10° E, 15–30° N). The Sahel has been chosen because SCIAMACHY shows the best performance here and has recorded most measurements (> 6000 measurements, see Frankenberg et al., 2009). The slopes from all datasets in the Sahel are relatively flat except for SCIAMACHY. This may again be related to the vertical sensitivity. Based on a network of FTIR HDO/H2O observations, Schneider et al. (2010) conclude that ln(HDO/H2O) vs. ln(TPW) slopes are steeper in air masses close to the surface and decrease progressively with height. In our observations, the slope is highest for SCIAMACHY, with the highest sensitivity close to the surface. The slope is high for TESV4 and higher for TESV5, which has the lowest sensitivity in the lower troposphere. Interestingly, the steeper slope from SCIAMACHY (0.093) is in the range of a typical Rayleigh distillation process slope, which is between 0.08–0.15 (+20 ~ −20°C) depending on evaporation/condensation temperatures (Majoube, 1971a, b; Yoshimura et al., 2011). The SCIAMACHY slope would correspond to an effective condensation temperature of 282 ± 7 K. The slopes from TESV5, ECHAM and ECHAMAK5 and ECHAMAK5Corr correspond to 314 ± 12 K, 318 ± 5 K, 314 ± 10 K, and 325 ± 5, respectively. These unrealistically high condensation temperatures imply that the atmosphere as a whole cannot be considered as a pure Rayleigh distillation column. In the atmosphere, there are many factors influencing this process such as mixing of the air parcel, evaporation of condensate water, kinetic effect, active convection (Gedzelman, 1988; Smith, 1992; Moyer et al., 1996; Keith, 2000; Galewsky et al., 2007). Apparently these processes have an important influence on the isotope-concentration correlations that overrule the condensation temperature-induced correlation.
5 Conclusions

Over the last years, water isotope retrievals have become available from different global satellite datasets. Here we compared the HDO/H$_2$O data from TES version 4, TES version 5, and SCIAMACHY with each other and with large-scale isotope patterns from the ECHAM model. We systematically assessed how first-order water isotope effects (temperature-, latitude-, altitude-, continental-, and amount-effect) are represented in the respective remote sensing datasets.

The geographical and temporal patterns in the respective water isotopologue fields reproduced the different “classical” large-scale isotope effects to a varying degree. Our analysis confirmed the improvement of TES$_{V5}$ compared to TES$_{V4}$ for the first-order global isotope signals investigated. Similarly, when the model results are convoluted with the AK, ECHAM$_{AK5}$ outperforms ECHAM$_{AK4}$. Nevertheless, we identified a problem with the ECHAM$_{AK5}$ results over the Himalayas region. The large positive isotope anomalies were shown to be caused by a high bias in humidity in ECHAM. A humidity correction or an a posteriori processing is necessary for model-data comparison because of cross dependence between H$_2$O and HDO in the application of the AK to the model. Furthermore, ECHAM overestimates $\delta D$ values at mid latitudes compared to SCIAMACHY and TES$_{V5}$ and the tropical and sub-tropical band of high $\delta D$ values is wider in the model than in the satellite datasets. This is a common problem in many GCM models since the models tend to have a high moist bias in the tropical and sub-tropical regions associated with errors in cloud processes, large-scale circulation and diffusion during water vapor transport.

When examining the seasonally varying $\delta D$ signal in the tropics associated to the movement of the ITCZ, SCIAMACHY, TES$_{V5}$ and ECHAM showed the expected seasonal co-variation of the latitudinal $\delta D$ minima and maxima in water vapor with insolation and rainfall. Some of the isotopologue effects are difficult to identify in SCIAMACHY because of its limited coverage of large parts of the respective winter hemispheres and oceans.
We also tested to what extent the atmosphere in the different datasets can be described as a Rayleigh distillation system. The results show that the atmosphere as a whole cannot be considered as a pure Rayleigh distillation column. For small region, e.g. in the Sahel, TES\(_{V5}\) and SCIAMACHY imply more Rayleigh type atmospheric processes than ECHAM. However, none of the effective condensation temperatures deduced from the observed slopes reflect pure Rayleigh condensation temperatures. This comes not as a surprise since many processes influence this condensation process, such as mixing of the air parcel, evaporation of condensate water, kinetic effect, and active convection, which are not controlled by a single effective condensation temperature.

One central application of existing and future water isotopologue datasets will be the evaluation of global observations and models. We suggest to use the qualitative and quantitative tests carried out in this study as a benchmark for the different data products and to evaluate their strengths and weaknesses.

Acknowledgements. This study was funded by NWO project number ALW-GO-AO/10-11 (the Netherlands Organization for Scientific Research). We thank M. Schneider for a very insightful and helpful review.

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### Table 1. Comparison of satellite instruments and model used in this study.

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<thead>
<tr>
<th>Comparison</th>
<th>TES</th>
<th>SCIAMACHY</th>
<th>ECHAM</th>
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<tr>
<td>Wavelength</td>
<td>650–3050 cm(^{-1})</td>
<td>2355–2375 nm</td>
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<td>Mode</td>
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<td>Limb, Nadir, Sun/Moon Occultations</td>
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Figure 1. TES prior version 4 (A), ECHAM model input (B), $\delta D$ product from TES$_V4$ (C), TES$_V5$ (D), ECHAM$_{AK4}$ (E), ECHAM$_{AK5}$ (F), SCIAMACHY (G) and ECHAM$_{AK5Corr}$ (H), SCIAMACHY prior (I), and ECHAM$_{AK5Pos}$ plot (J). The figures of TES prior, TES$_V4$, and ECHAM$_{AK4}$ are averaged between 850–500 hPa and TES$_V5$, ECHAM$_{AK5}$, ECHAM$_{AK5Corr}$, and ECHAM$_{AK5Pos}$ between 900–425 hPa. The results are bias corrected by 5%, 3%, and $-5\%$ for TES$_V4$, ECHAM$_{AK4}$, and SCIAMACHY, respectively. Arrows in (D) point out the continental effect and ellipses in (B), (D), (F), (G), (H) show the region with a strong isotope amount effect around Indonesia. All plots are annual average $\delta D$. 

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Figure 2. Annual average latitude profile of δD in water vapor from TES, ECHAM, ECHAM AK, ECHAM AK5Corr, and SCIAMACHY. In (A) the TES for version 4 datasets are shown and in (B) the TES version 5 datasets.
Figure 3. Seasonal comparison of zonal means of $\delta D$ from TES prior, TES$_{V5}$ and SCIAMACHY (A), and from TES$_{V5}$, ECHAM$_{AK5}$, and ECHAM$_{AK5Corr}$ (B). Solid lines show NH summer profiles and dashed lines NH winter profiles.
Figure 4. Latitudinal $\delta D$ profiles between 30° N and 30° S in northern summer (JJA, solid lines) and northern winter (DJF, dashed lines) of TES$_{V5}$ and SCIAMACHY (A); ECHAM and ECHAM$_{AK5Corr}$ (B).
Figure 5. Latitudinal profiles between 30° N and 30° S in northern summer (JJA, solid lines) and northern winter (DJF, dashed lines) of ECHAM from 900 to 680 hPa and from 618 to 425 hPa (A), of TESV5 from 900 to 680 hPa and from 618 to 425 hPa (B), and of ECHAM\textsubscript{AK5Corr} from 900 to 680 hPa and from 618 to 425 hPa (C).
Figure 6. Correlation plot of ln (TPW) (Total Precipitable Water derived from specific humidity) vs. model simulated and satellite retrieved ln(HDO/H2O). (A) and (B) global correlations. (C) and (D) Correlations in the tropics (30° N–30° S). (E) and (F) Correlations in the Sahel region. Panels (A), (C), and (E) show the version 4 TES datasets and SCIAMACHY. Panels (B), (D), (F) show the version 5 TES datasets, both together with ECHAMAK. ECHAM plot at the left side is averaged from 850–500 hPa and ECHAM plot at the right side is averaged from 900–425 hPa.