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Harmonization and Diagnostics of MIPAS ESA CH_4 and N_2O Profiles Using Data Assimilation

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Abstract. This paper discusses assimilation experiments of methane (CH_4) and nitrous oxide (N_2O) profiles observed by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS). Here we focus on data versions 6 and 7 retrieved by the ESA processor. These datasets have been assimilated by the Belgian Assimilation System for Chemical ObsErvations (BASCOE). The CH_4 and N_2O profiles can be noisy especially in the tropical lower stratosphere. Using the averaging kernels of the observations and a background error covariance matrix – the $\mathbf{B}$ matrix, which has been previously calibrated, allows the system to partly remedy this issue and provide assimilated fields that are more regular vertically. In general, there is a good agreement between the BASCOE analyses and independent observations demonstrating the general good quality of these two retrievals provided by MIPAS ESA. Nevertheless, this study also identifies two issues in these datasets. First, time-series of the observations show unexpected discontinuities, due to the calibration method used for the level-1 data. Second, the correlations between BASCOE analyses and independent observations are poor in the lower stratosphere, especially in the tropics, probably due to the presence of outliers in the assimilated data. In this region, we recommend using MIPAS CH_4 and N_2O observations with caution.

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1 Introduction

Carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) are the three most important well-mixed greenhouse gases (WMGHG, Stocker et al., 2013). Their contributions in enhancing radiative forcing are, respectively, 1.82, 0.48 and 0.17 Wm^{-2}. After a decade of near stability, the increase of CH_4 concentration has been observed and the radiative forcing from CH_4 is now larger than all of the combined halocarbons. The concentration of N_2O is also increasing such that, due to the decrease of chlorofluorocarbons 12 (CFC-12), N_2O is now the third largest WMGHG contributor to radiative forcing.

While CH_4 and N_2O are considered well-mixed greenhouse gases, both gases exhibit a vertical profile and significant spatiotemporal variability in the stratosphere through the combination of prevailing long transport timescales in the stratosphere and a shorter chemical lifetime than in the troposphere. In general, the traditional assumption in climate modelling that both gases are homogeneously distributed results in minor errors in their global mean forcing, less than 2% (Freckleton et al., 1998). However, local heating rates and thus the dynamics in the stratosphere, are sensitive to atmospheric composition changes. An accurate representation of the role of stratospheric changes in response to anthropogenic climate forcing therefore requires an accurate representation of (past and future) stratospheric composition changes. Moreover, some studies also have acknowledged the impact of the representation of the stratosphere in Numerical Weather Prediction (NWP) systems (Monge-Sanz et al., 2013; de Grandpré et al., 2009).

The computing time to resolve state-of-the art chemical equation systems for the stratosphere is much too expensive for NWP models, and the same issue arises for present-day Global Climate Model (GCM) and Earth-System Model (ESM) baseline simulations. These models also include processes to resolve tropospheric climate and chemistry, land-surface, sea-ice and ocean processes, each with some degrees of complexity. For this reason, simplified chemical schemes are being investigated (e.g. Baumgaertner et al., 2010). Linearization of the stratospheric chemistry (Hsu and Prather, 2010; Monge-Sanz et al., 2013) could be a computationally attractive alternative in some of the complex NWP and climate models, though implementation would require both further studies as well as a full reanalysis, or at least a chemical-consistent representation of the key gases of interest in the stratosphere, which is however not available for present-day conditions. The development of 3-D assimilated fields – i.e. the analyses – of CH_4 and N_2O as presented in this work provides useful starting point for a full present-day stratospheric composition reanalysis.

Data assimilation methods aim at estimating the true state of the atmosphere by combining the information from sparse observations, the a-priori state of the atmosphere and the theoretical knowledge synthesized in a numerical model (Lahoz et al., 2010). These methods have been applied to both the physical and chemical state of the atmosphere. Data assimilation procedures thereby yield information on departures between the estimated model fields and the observations. This information can be exploited for the assessment of the quality of the assimilated observations. For example,

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Simmons et al. (2014) have identified drifts in different temperature data records going into the European Centre for Medium-range Weather Forecast (ECMWF) systems for the ERA-Interim reanalysis. Furthermore, Stajner et al. (2004) have used ozone analyses of Solar Backscatter Ultra Violet/2 (SBUV/2) to detect and characterize changes in the observation errors.

CH_4 and N_2O are both produced at the Earth's surface and are long-lived species over the troposphere and stratosphere, where they are not directly chemically coupled. Nevertheless, they show a strong and robust tracer-tracer correlation due to transport and rapid mixing along isentropic surfaces (Plumb, 2007). By simultaneously assimilating these retrievals we have a clean assimilation of information content that allows one to compare these two sets of observations and validate the assimilation methodology. In practice, as we will see, the assimilation of MIPAS CH_4 and N_2O shows a number of issues that need to be addressed. The lesson learned here establishes some of the milestones that should be addressed for an effective multi-species chemical data assimilation system.

The property of tracer-tracer correlation between atmospheric constituents has been used in data assimilation by Chipperfield et al. (2002). In that study, CH_4 observations from the HALogen Occultation Experiment (HALOE) were assimilated using a suboptimal Kalman filter. Their assimilation gave rise to noisy tracer-tracer correlations, in particular between CH_4 and N_2O. In order to preserve the correlations, an a posteriori correction of the analysis was applied to every modelled species which are correlated with CH_4. In our study, both species are assimilated with the Belgian Assimilation System for Chemical ObsErvation (BASCOE) which is based on the four dimensional variational (4D-Var) method. Our experiments usually focus on MIPAS version 6 but the case of version 7 has also been considered.

This paper is organized as follows. Section 2 presents the observations used in this paper: the assimilated observations from MIPAS and independent observations from the Atmospheric Chemistry Experiment - Fourier Transform Spectrometer (ACE-FTS) and the Microwave Limb Sounder (MLS). The BASCOE system and its setup are presented in Sect. 3. Section 4 compares the assimilation experiments of MIPAS CH_4 and N_2O and Sect. 5 presents their validation using independent data. Finally, Sect. 6 summarizes the results of this paper.

2 Observations

2.1 MIPAS

The assimilated CH_4 and N_2O data are retrieved from the limb Fourier transform spectrometer MIPAS on board the Envisat platform which operated between 2002 and 2012. Measuring in the infrared, limb spectra are inverted to provide profiles of numerous trace gases, including CH_4 and N_2O (Fischer et al., 2008). The MIPAS mission is divided in two phases: the full resolution (FR) phase, from 2002 to 2004, and the optimized resolution (OR) phase, from 2005 to 2012. This later

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period is characterized by a finer vertical and horizontal sampling attained through a reduction of the spectral resolution. This study focuses on the second phase of MIPAS i.e. from 2005 to 2012.

Several level-2 retrievals have been developed for MIPAS (Raspollini et al., 2014). In this study, profiles delivered by the MIPAS Level-2 profile Prototype Processor (ML2PP) version 6 are used (Raspollini et al., 2013). MIPAS also has different modes of observations, in particular, the nominal mode (NOM) with altitude soundings between 7-72 km, the middle atmosphere mode (MA, 18-102 km) and the upper atmosphere mode (UA, 42-172 km). Most of the MIPAS profiles have been measured with the NOM mode and only this dataset is considered in this study.

Full resolution ML2PP v4.61 of methane and nitrous oxide have been validated by Payan et al. (2009), during the first phase of MIPAS. In the middle stratosphere, no significant bias is observed between ML2PP profiles and correlative measurements. In the lower stratosphere/upper troposphere (UT/LS), it was reported that the ML2PP data v4.61 exhibits some unphysical oscillations in individual CH_4 and N_2O profiles caused by the processing algorithm (which used almost no regularization).

ML2PP v6 profiles of CH_4 and N_2O (full and optimized resolution) have also been compared with measurements obtained by the balloon-borne cryogenic whole air sampler BONBON (Engel et al., 2016). Based on seven flights of BONBON, a good agreement is found between both instruments above 20 km altitude, within the estimated uncertainty limits. This good agreement is also observed below 20 km for CH_4 while MIPAS N_2O underestimated BONBON by around 20 ppbv at 15 km.

Vertical averaging kernels (AK) of MIPAS data, which are provided for each profile, have been used in this study. In data assimilation, the vertical AK are used to interpolate the model fields to the vertical grid of the observations, as follows (Rodgers, 2000):

$$\mathbf{x} = \mathbf{y}_0 + \tilde{\mathbf{A}}[\tilde{\mathbf{x}} - \tilde{\mathbf{y}}_0] \quad (1)$$

where $\mathbf{y}_0$ is the a priori profile used in the retrieval, $\mathbf{x}$ is the model state vector projected in the observation space by an observation operator. $\mathbf{A}$ is the AK matrix corresponding to the measured profile. The "˜" sign indicates that the value is provided on the model vertical grid, i.e. that $\tilde{\mathbf{y}}_0$ and the rows of $\tilde{\mathbf{A}}$ are interpolated to the model vertical grid.

This equation is valid for retrieval methods which use an a priori profile which is not the case for ML2PP algorithms (Raspollini et al., 2013). In that case, $\mathbf{y}_0$ should be replaced by $\mathbf{y}_{k-1}$ in Eq. (1), where k denote the second to last iteration of the retrieval (Ridolfi et al., 2011). Since the convergence criteria adopted in the retrieval are rather conservative and since $\mathbf{y}_{k-1}$ is not provided in the MIPAS data files, the retrieval team recommends using $\mathbf{y}_k$ such that the use of the AK has been implemented as follows:

$$\mathbf{x} = \mathbf{y}_k + \tilde{\mathbf{A}}[\tilde{\mathbf{x}} - \tilde{\mathbf{y}}_k]. \quad (2)$$

The impact of the AK when interpolating BASCOE fields to MIPAS observation locations is illustrated in Fig. 1 for a MIPAS CH_4 profile taken in the equatorial region. Figure 1a shows the

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AK profiles that correspond to the CH_4 profile displayed in Fig. 1b. The AK are relatively peaked between the top level and 20 hPa and more elongated below 20 hPa. Figure 1b also shows two BASCOE modelled (no assimilation) profiles interpolated at the observation location, with and without the use of the AK. The MIPAS profile exhibits oscillations below 10 hPa showing that this issue has not been resolved since the release of the full resolution v4.61. The BASCOE profile interpolated without the use of the AK is much smoother. This is expected since there is no source or sink of CH_4 in the lower stratosphere. On the other hand, the BASCOE profile interpolated with the use of the AK presents vertical oscillations. This is due to the use of an oscillating MIPAS profile in Eq. (2). Figure 1c shows the difference between the MIPAS profile and the two BASCOE profiles. These profiles, approximately, represent the analysis increment that will correct the model field. In the case where the AK are not used, the increment is oscillating such that these undesirable oscillations will be introduced in the model. On the other hand, the increment based on the use of the AK is much more regular such that the BASCOE analyses will also be more regular vertically.

Note that horizontal AK have been derived for the ML2PP retrieval (von Clarmann et al., 2009) but they have not been used in this study. However, the latitude and longitude of the tangent point are used in the observation operator instead of the average latitude and longitude of the profile. This provides most of the correction given by the horizontal AK (von Clarmann et al., 2009).

Finally, ESA recently produced a new version 7 of MIPAS ML2PP. Both level-1 (L1) and level-2 (L2) processors used for generating respectively L1v7 (used by the L2v7 processor) and L2v7 products, as well as the auxiliary data, include significant improvements with respect to v6. For OR measurements no species-dependent changes were performed, the changes being the results of changes in temperature due to the new non-linearity correction implemented in L1 processor, changes in the handling of the retrieval of continua, etc. In general, v7 CH_4 and N_2O products show higher concentrations than in v6, mainly in the tropics at very low altitudes. Also, v7 OR measurements use a stronger regularization that should provide less oscillating profiles of CH_4 and N_2O.

2.2 ACE-FTS

The BASCOE analyses are validated by comparison with two independent datasets. The first one is from the ACE-FTS instrument (Bernath et al., 2005) which was launched in 2003 and was operating during the period of MIPAS optimized resolution. This instrument is based on the solar occultation measurement technique providing around 28 profiles per day (Boone et al., 2005, 2013). Here, the version 3.5 of N_2O and CH_4 retrievals has been used where a data quality flag has been implemented since previous versions.

ACE-FTS v3.5 N_2O has been compared against observations from MIPAS and MLS (Sheese et al., 2016). The coincidence criteria used in that study required observations being made within 3 hours and within 350 km of each other. This provides around one hundred collocated pairs of profiles,

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almost all observed at mid-latitudes and in the polar regions. Below 35 km, MIPAS and MLS are reported to agree reasonably well with ACE-FTS, yielding a strong correlation between the three datasets, and MIPAS typically agrees with ACE-FTS within -9 and +7%, with standard deviations in the range of 7-45%. In the 30-45 km region, ACE-FTS and MIPAS typically agree within $\pm$10%. ACE-FTS

For CH_4, only ACE-FTS v2.2 has been validated against independent observations. An agreement within $\pm$10% was found in the upper troposphere/lower stratosphere, and within $\pm$25% in the middle and higher stratosphere up to the lower mesosphere (below 60km, De Mazière et al., 2008).

2.3 Aura MLS

The second dataset used to validate the BASCOE analyses is provided by the MLS instrument on board the Aura satellite (Waters et al., 2006; Livesey et al., 2006). MLS was launched in 2004 and was also in operation during the second phase of MIPAS. Here, only N_2O measurements are available since there is no spectral signal of methane in the microwave. The data version used here is 3.3 and profiles are filtered according the recommandation of Livesey et al. (2011). Nitrous oxide v2.2 has been validated by Lambert et al. (2007). In the pressure range 100-4.6 hPa, MLS N_2O precision is 24-14 ppbv (9-41%) and the accuracy is to be 70-3 ppbv (9-25%) (Livesey et al., 2011). Although vertical AK of MLS are available, as well as the a priori profile of the retrieval, they have not been used in this study. The impact of the MLS AK for N_2O has been tested and was found very small, mainly because the AK profiles are peaked at the tangent point (not shown). Note that in the case of MLS, the AK must be used with Eq. (1) and not with Eq. (2).

3 The BASCOE system

This study is based on numerical experiments made with the Belgian Assimilation System for Chemical ObsErvations (BASCOE, Errera et al., 2008; Errera and Ménard, 2012). Based on a Chemitry Transport Model (CTM), this system usually considers 57 stratospheric species advected by the Flux-Form Semi-Lagrangian scheme (Lin and Rood, 1996), 200 chemical reactions and a parameterization of the physico-chemical processes due to Polar Stratospheric Clouds (PSCs). However, in this study, only the advection of CH_4 and N_2O is considered (i.e. the chemical and PSC schemes have been turned off) in order to reduce the CPU time. Hence, we assume that these species behave like an inert tracer, a fair assumption for N_2O and CH_4 when using an assimilation window of one day. In the experiments performed for this paper, the dynamical fields are provided by the European Centre for Medium-range Weather Forecasts (ECMWF) ERA-Interim reanalyses (Dee et al., 2011). The horizontal resolution is set to 3.75°x2.5°longitude/latitude grid. The vertical grid is represented by 37 vertical levels from the surface to 0.1 hPa, a subset of the ERA-Interim 60 levels.

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While an Ensemble Kalman filter version of BASCOE is now available (Skachko et al., 2014, 2016), the BASCOE system used in this study is still based on the four dimensional variational (4D-Var) assimilation method. The 4D-Var method aims at minimizing the following cost function:

$$J(\mathbf{x}) = \frac{1}{2}[\mathbf{x}-\mathbf{x}_b]^T\mathbf{B}^{-1}[\mathbf{x}-\mathbf{x}_b] + \frac{1}{2}[\mathbf{y}-H(\mathbf{x})]^T\mathbf{R}^{-1}[\mathbf{y}-H(\mathbf{x})] \quad (3)$$

where $\mathbf{x}_b$ is the model background state, $\mathbf{B}$ is the background error covariance matrix and $\mathbf{R}$ is the observational error covariance matrix. H is the observation operator that maps the model state vector in the observation space. In notation used, it is assumed that H contains a model operator that projects $\mathbf{x}$ from the initial time of the assimilation window to the time of the observation. The other variables have been defined in Sect. 2.

The $\mathbf{B}$ matrix is defined on a spherical harmonic basis and assumes homogeneous and isotropic spatial correlations as described in Errera and Ménard (2012). It has been calibrated by an ensemble method as proposed by Fisher (2003) and discussed by Bannister (2008) and Massart et al. (2012), in the following way. Two assimilation experiments are conducted in parallel between February 2009 and March 2010. They are based on two MIPAS datasets where Gaussian noise is added to the retrieved measurements, the variance of the noise being equal to the MIPAS error variance. For each experiment, the $\mathbf{B}$ matrix assumes a standard deviation of 20% and Gaussian spatial correlations with horizontal and vertical length scales of 400 km and 0.5 levels, respectively. The initial conditions of both experiments are based on a BASCOE free CTM run of CH_4 and N_2O where a random noise has been added, this noise having the same statistical properties - in variances and correlations - as the initial $\mathbf{B}$ matrix used for these two experiments. So, the two parallel experiments differ only in the noise added to the MIPAS data and to the initial conditions. According to Bannister (2008), the ensemble of the difference between the short-term forecasts of both experiments allows one to estimate the background error covariance matrix. Figure 2 displays the calibrated standard deviation of the $\mathbf{B}$ matrix for N_2O and CH_4. In both cases, the standard deviations are relatively small, between 2 and 5%, where small values are found in the tropical lower stratosphere and high values are found in the upper stratosphere and in the polar regions. The impact of the calibration of the $\mathbf{B}$ matrix will be evaluated in Sect. 4.

The $\mathbf{R}$ matrix in Eq. (3) corresponds to the observational error covariance matrix as seen by the data assimilation system. It should theoretically also account for the representativeness error of the operator H. In this paper, the variance of $\mathbf{R}$ is given by the ML2PP retrieval with a minimal threshold of 5% as in Inness et al. (2013). The ML2PP retrieval also provides the off-diagonal terms of $\mathbf{R}$ for the vertical correlations (successive profiles are supposed to be uncorrelated). The impact of correlations in $\mathbf{R}$ on the BASCOE analyses is evaluated in Sect. 4.

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Finally, MIPAS ML2PP outliers are rejected by a background quality check (BgQC, Anderson and Järvinen, 1999). This filter rejects any value if:

$$(\mathbf{y} - H(\mathbf{x}_b))^2 > \alpha(\sigma_o + \sigma_b) \quad (4)$$

where σ_o and σ_b denote, respectively, the error variance of the observations and of the background state. The value of α has been set to 5 such that the BgQC will only reject outlying profiles. It should be noted that the BgQC filter depends on the variance of $\mathbf{B}$. The impact of this filter will be discussed in Sect. 4.

4 Comparison of the Assimilation Experiments

The experiments realized for this paper aim to evaluate the impact of (1) the use of the AK, (2) the use of a calibrated $\mathbf{B}$ matrix, (3) the use of the vertical correlation in the $\mathbf{R}$ matrix and (4) the use of MIPAS v6 or v7 dataset. These experiments are summarized in Table 1. Experiment CTRL is a CTM run without data assimilation. Experiment RAW is a Chemistry Data Assimilation (CDA) run where AK and vertical correlations in $\mathbf{R}$ are not used and where $\mathbf{B}$ is not calibrated. Experiment BASELINE is as RAW except that AK are used. Experiment ENS is as BASELINE but the covariances of $\mathbf{B}$ have been estimated by the ensemble method (see Sect. 3). Experiment ENS-CR is as ENS but the vertical correlations in $\mathbf{R}$ are used. Experiment BASEv7 is as BASELINE except that MIPAS v7 is assimilated instead of v6. All these experiments cover the period between April and November 2008. A final experiment, REAN, covers the period April 2007-April 2012, and has been set up as ENS. Experiments with uncalibrated $\mathbf{B}$ use the following setup for the background error covariance matrix: a standard deviation error of 20% of the background field and Gaussian spatial correlations with length scales of 1 model level vertically and 800 km horizontally.

The BgQC filter has been used in all these experiments. Its impact will also be evaluated in this section. The evaluation of the different experiments is based on an inspection of the experiment analyses, several data assimilation diagnostics like the Observation minus Forecast (O-F) residuals or the value of the cost function, and the tracer-tracer correlations between N_2O-CH_4. Some results in this section are shown only for CH_4 or N_2O but the same conclusions are found for the other species.

Figure 3 shows the zonal mean analysis of CH_4 from six BASCOE experiments on 1 May 2008 at 12 UT which is representative of other dates. The CTM run (CTRL) shows the well known features of the CH_4 distribution: high values in the lower stratosphere and in the tropics and lower values at higher altitude and latitudes. Methane is emitted at the Earth surface and has a life time of 10 years (Ko et al., 2013). In the stratosphere, methane is destroyed by oxidation with OH, O^1D and Cl (Brasseur and Solomon, 2005). Methane has no source in the atmosphere. In those conditions, no physical or chemical processes can explain the noisy zonal mean of CH_4 displayed by experiment RAW. This noise is reduced when the assimilation system uses the averaging kernels (exp. BASE-

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LINE) and a calibrated $\mathbf{B}$ matrix (exp. ENS). The use of the vertical correlations in the $\mathbf{R}$ matrix apparently has a very small impact when comparing experiments ENS versus ENS-CR. BASEv7 shows larger CH_4 volume mixing ratios than BASELINE, between 5 and 10% depending on the region. It is also found that the noisy structure in BASELINE is also present in BASEv7. This suggests that MIPAS v7 does not provide more regular profiles of CH_4 than v6. These vertical oscillations in the tropics were discussed for MIPAS v4.61 (Payan et al., 2009) and are still present in v6 and v7. Although less pronounced, the same issues appear for N_2O (not shown).

Figure 4 shows the CH_4 O-F residuals between BASCOE and MIPAS for September-October 2008. CTRL results in larger biases and standard deviations since MIPAS data are not assimilated in this experiment. This is due to imperfection of the model and initial conditions. The figure also highlights the larger bias for RAW in the polar regions. BASELINE, ENS and ENS-CR display very similar O-F residuals. Looking carefully, it can be seen that among the experiments, ENS and ENS-CR always delivered the lowest standard deviations of the differences. Biases from BASEv7 are similar to those from BASELINE, COVR and ENS except at the South Pole where they are slightly larger. The standard deviation from BASEv7 is generally smaller than those other experiments above 10 hPa but larger in the lower stratosphere.

The N_2O-CH_4 correlations for five experiments have been compared with those from ACE-FTS, for the period May-October 2008 and between 30°S-30°N (Fig. 5) where BASCOE values are interpolated in the ACE-FTS observation space with the observation operator. The result of a four-degree polynomial fit is also shown with the root mean square (rms) of the residual between the fitted curve and the observations. ACE-FTS shows a compact tracer-tracer correlation with a rms r=10.2. CTRL exhibits the lowest rms from the different BASCOE experiments (r=4.2). The rms for the RAW experiment is relatively large compared to ACE-FTS (r=28) while the assimilation experiments perfom much better when the system uses additional information from the AK (r=17.5 for BASELINE) and from the calibration of $\mathbf{B}$ (r=6.8 for ENS and ENS-CR). Again, the use of the correlations in the $\mathbf{R}$ matrix does not improve the results (same rms for ENS and ENS-CR). BASEv7 has a better correlation (r=12.7) than BASELINE (not shown).

Fig. 6 shows the evolution of the cost function at the analysis point $J(\mathbf{x}_a)$ weighted by the number of observations p, for the period April-November 2008. If the error statistics used in the assimilation system are consistent with the O-F residuals then the value of $J(\mathbf{x}_a)/p$ should be close to 1⁄2 (Talagrand, 2010). Note that this is a necessary but not sufficient condition. The best results are from ENS which shows values close to 1/2 with small day-to-day variations. BASELINE also displays cost values close to 1/2 but their daily variability is much higher. ENS-CR shows a much larger deviation from theoretical values and with high variability. This suggests that the observational error covariance matrices provided by the MIPAS ML2PP retrieval are not optimal for data assimilation. This issue has not been further investigated in this study. BASEv7 has values of $J(\mathbf{x}_a)/p$ which are higher than those from BASELINE and further larger to 1/2.

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All experiments have a common BgQC set-up ($\alpha = 5$). However, the observations rejected by the BgQC differ between the different experiments. Indeed, Eq. (4) depends on the background state $\mathbf{x}_b$ and the variances of $\mathbf{B}$ which are different in each experiment. In particular, the standard deviation of $\mathbf{B}$ used in ENS and ENS-CR is much smaller than the 20% used by the other experiments. Figure 7 shows histograms of the difference between BASCOE and MIPAS values weighted by the observational errors for experiments BASELINE and ENS, and for CH_4 and N_2O on 20 August 2008. These histograms are computed separately for accepted and rejected MIPAS observations. The 4D-Var method assumes that the distribution of the observational errors is Gaussian which should lead to O-F residuals displaying a Gaussian shape. Hence, Fig. 7 also shows a Gaussian fit of the histograms for the accepted observations.

Due to the large background error variances used in BASELINE, very few observations are rejected by the BgQC. Assuming the histograms for accepted observations should be a Gaussian distribution, the number of observations accepted in BASELINE is too high at large differences. The use of the calibrated $\mathbf{B}$ matrix in ENS partly remedies this issue. In that case, many more observations are rejected with large departures and the histogram of accepted data is much closer to a Gaussian distribution than in the BASELINE case. Nevertheless, the number of accepted observations is still too high, especially for differences between values -2 and -3. The questions now remains open of whether the BgQC filter should be supplemented by a more sophisticated filter such as a variational quality control filter (VarQC) as done at ECMWF (Anderson and Järvinen, 1999). Note that the impact of the BgQC on experiment BASEv7 has been checked to evaluate any improvement in MIPAS v7 and results similar to BASELINE have been found (not shown).

Based on the results already discussed in this section, assimilation experiments based on MIPAS v6 can be sorted according to their quality from best to worse: ENS, ENS-CR, BASELINE and RAW. ENS provides the best results as it benefits from the use of the AK, a calibrated $\mathbf{B}$ matrix and an optimal use of the BgQC filter. While providing analyses very similar to ENS, experiment ENS-CR is not as good as ENS since it provides values of $J(\mathbf{x}_a)/p$ much higher than the expected value of 1/2. The fact that ENS-CR is worse than ENS while using the full $\mathbf{R}$ matrix has not been further investigated.

A comparison between BASELINE and BASEv7 also allows us to compare MIPAS v6 and v7. Zonal means are comparable (Fig. 3), O-F and $J(\mathbf{x}_a)/p$ are better with BASELINE (Figs. 4 and 6), and the N_2O-CH_4 correlation is better with BASEv7 (Fig. 5). O-F with ACE-FTS also shows a general better agreement with BASELINE than BASEv7 (not shown). According to the diagnostics that have been set up for this study, MIPAS v7 does not improve the quality of v6 for CH_4 and N_2O.

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5 Validation Against Independent Observations

Experiment ENS is evaluated against independent observations from MLS and ACE-FTS for N_2O, and from ACE-FTS for CH_4. To do this, assimilated fields are interpolated at the geolocation of MLS and ACE-FTS profiles during the assimilation procedure using the observation operator of BASCOE. Sampling error is then minimized since the maximum difference of time between observed profile and the BASCOE time step is 15 minutes.

Figure 8 shows the O-F residuals between ENS and these three independent datasets for the period September-October 2008 in three regions: the south polar cap, the tropics and the north polar cap. For CH_4, the agreement between ENS and ACE-FTS are in good agreement: the mean differences and the associated standard deviations are mostly below $\pm 10\%$, i.e. within the ranges found by De Mazière et al. (2008). For the comparison of N_2O with ACE-FTS, there is also a good agreement. The mean differences are usually below $\pm 10\%$. The standard deviation is relatively small (<10%) in regions where N_2O is abundant, i.e. in the tropical lower stratosphere (below 10 hPa) and in the UT/LS in the polar regions. When the amount of N_2O is relatively small, in the upper stratosphere and in the south polar vortex, relative differences can lead to large values and absolute differences are more suited. The standard deviation is usually less than 5 ppbv in the upper stratosphere and less than 15 ppbv in the south polar vortex (not shown). These values are in agreement with those found by Sheese et al. (2016), except in the South Pole region. Below 4.61 hPa, the mean difference between ENS and MLS N_2O is usually below $\pm 10\%$ except in the South Pole region where values around 20% are found. Since ENS agrees very well with MIPAS (see below in Figs. 9e and g) and ACE-FTS, these differences suggest that MLS underestimates N_2O concentration in the polar vortex (in agreement with Sheese et al., 2016). Overall, ENS and independent data agree within the uncertainties of each independent dataset.

Figure 9 shows the time series of CH_4 and N_2O between April and November 2008 above south polar region and in the tropics at two MIPAS levels (in the middle stratosphere and in the lower stratosphere) for MIPAS, ENS and CTRL and MLS (N_2O only). Five periods are shaded and labelled by the letters A, B, C, D and E, and are discussed below. The agreement between MIPAS and ENS is very good for CH_4 and N_2O. MIPAS time series are relatively noisy, much more than those from MLS. CTRL is shown in order to check whether this noise is due to the sampling of MIPAS - in particular the number of profiles per day can vary from 0 to $\sim$1500 - or to a more fundamental issue in MIPAS. Usually, CTRL follows the variation of MIPAS very well. For example, in the south pole region in early August (see A in Fig. 9), MIPAS, MLS, ENS and CTRL all show an increase in CH_4 and N_2O abundances which are due to an elongation of the vortex across the South Pole (not shown). There are also daily discontinuities (see C and D) in MIPAS that are well captured by CTRL and correspond to days with poor sampling by MIPAS. The origin of two other discontinuities in MIPAS in early May and mid October (see B and E) are more problematic. They occur immediately after an

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interruption of MIPAS Nominal mode and last a few days. They are not related to the sampling of MIPAS because the numbers of profiles for these days are higher than 1000.

The discontinuities are related to the calibration procedure of the MIPAS L1 data as explained in the following. During the mission, the instrument was contaminated by ice which was removed several times per year by a warm-up of the instrument. After a warm-up (i.e. decontamination) an abrupt change in the signal measured in the spectral region where CH_4 and N_2O are retrieved may occur. The calibration of L1 data is operated weekly. Since this is not always after a decontamination, discontinuities in the retrieved profiles may occur.

Such discontinuities are relatively frequent in the whole period of MIPAS optimal resolution as illustrated in Fig. 10. This figure shows the time series of daily averaged MLS N_2O above the south polar region and the tropics for a period of five years (April 2007-April 2012). Seasonal variations of N_2O are clearly visible at South Pole but also in the tropics, in particular in the lower stratosphere. In addition, the corresponding time series of N_2O and CH_4 from the assimilation experiment REAN interpolated in the MLS observation space are shown on the figure. REAN is based on the same BASCOE configuration as ENS and, as ENS, REAN agrees very well with MIPAS. Hence the differences between MLS N_2O and REAN N_2O highlight differences between MLS and MIPAS. The agreement between both datasets is very good overall. Discontinuities due to the weekly calibration of L1 data can be seen in REAN analyses, especially in the tropics. In that region, REAN analyses are relatively noisy and the N_2O seasonal variations observed by MLS are not reproduced by the reanalysis of MIPAS N_2O and CH_4.

This result is confirmed in Fig. 11 and 12 which show the correlation coefficients between MLS (N_2O) and REAN (N_2O and CH_4), and between ACE-FTS (N_2O and CH_4) and REAN (N_2O and CH_4), as a function of altitude and latitude and for the period October 2007-April 2012. The correlation coefficients are calculated only for days that have more than 1000 MIPAS profiles to exclude the possibility that REAN is not representative of MIPAS during an unobserved period. The MIPAS retrieval team have also identified dates where L1 data are not properly calibrated just after decontamination. These dates are also excluded from the calculation of the correlation coefficients. Correlations are usually larger than 0.9 except in the lower stratosphere, especially in the tropics where the correlation coefficients are close to 0 or even negative. In the tropical lower stratosphere, MIPAS retrieval may be affected by clouds. Some of these profiles have been flagged out by the retrieval team (and are not assimilated by BASCOE) but this study shows that a significant number of outliers have not been filtered out by the retrieval code nor by the BgQC filter. Thus, we suggest using MIPAS profiles of N_2O and CH_4 with caution in the tropical lower stratosphere. This study will also help the retrieval team to improve the identification of outliers in this region.

This conclusion contrasts with other validation studies that did not mention any issues in that region (Engel et al., 2016; Sheese et al., 2016). However, Engel et al. (2016) compared only two collocated profiles between BONBON and MIPAS which may be too small to assess MIPAS in the

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tropical lower stratosphere. On the other hand, Sheese et al. (2016) based their comparison on one hundred collocated profiles between ACE-FTS and MIPAS although most profiles are located at mid and polar latitudes. As a consequence, the weight of the tropics in their statistics is too small to provide insight into this region. In our study, all available profiles of independent data have been used which delivers more robust statistics. In order to improve the characterization of MIPAS, we recommend to perform a specific validation of MIPAS in the tropics.

6 Conclusions

This paper presents assimilation experiments of MIPAS ESA version 6 N_2O and CH_4 profiles by the BASCOE system. For these two constituents, MIPAS profiles can be noisy and this study shows that assimilation can help to harmonize these observations. This is possible because assimilation systems can use additional information to the raw observations: (1) the averaging kernels associated with each profile and (2) the $\mathbf{B}$ matrix which was calibrated using an ensemble method. Using this setup, the BASCOE analyses are in good agreement with independent observations measured by MLS and ACE-FTS. This is due to the generally good quality of MIPAS profiles.

Nevertheless, this study also diagnoses several issues in MIPAS CH_4 and N_2O profiles. First, time series of MIPAS profiles show unexpected discontinuities which are to be due to the weekly calibration of L1 data. A daily calibration might resolve this issue. Second, the correlations between BASCOE analyses and independent observations from MLS and ACE-FTS are poor in the tropical lower stratosphere. This is due to outlier profiles which are not flagged out in the presence of clouds. These issues, also present in MIPAS version 7, will be addressed in the future version 8.

Overall, this study recommends using MIPAS ESA CH_4 and N_2O v6 and v7 profiles with caution in the tropical lower stratosphere. In order to improve the characterization of MIPAS, we also recommend performing specific validation of MIPAS in the tropics, which was not the case in previous validation studies. We recommend ESA to improve the data quality document that should come with any release of a new version of the L2 product, as in the case for MLS (Livesey et al., 2011). This document would describe the region of scientific validity for each product and their potential issues. This information is not always available in peer reviewed publications especially when a new version of the retrieval is released. This would greatly help the scientific community that works with ESA products. This will be done for the MIPAS L2 v7 release where all issues highlighted in this paper and other known issues will be mentioned. Finally, this study also shows that data assimilation can be considered a useful validation tool for geoscientific datasets.

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Table 1. List of BASCOE experiments discussed in this paper.

Name	CTM/CDA[1]	MIPAS version	Use of AK	Use of corr($\mathbf{R}$)[2]	Calibrated $\mathbf{B}$	Period
CTRL	CTM	n/a[3]	n/a	n/a	n/a	Apr 2008 - Nov 2008
RAW	CDA	v6	No	No	No	Apr 2008 - Nov 2008
BASELINE	CDA	v6	Yes	No	No	Apr 2008 - Nov 2008
ENS	CDA	v6	Yes	No	Yes	Apr 2008 - Nov 2008
ENS-CR	CDA	v6	Yes	Yes	Yes	Apr 2008 - Nov 2008
BASEv7	CDA	v7	Yes	No	No	Apr 2008 - Nov 2008
REAN	CDA	v6	Yes	No	Yes	Mar 2007 - Apr 2012

[1]: Free Chemistry Transport Model (CTM) run or chemical data assimilation (CDA) run
[2]: Use of vertical correlation in $\mathbf{R}$
[3]: n/a = not applicable

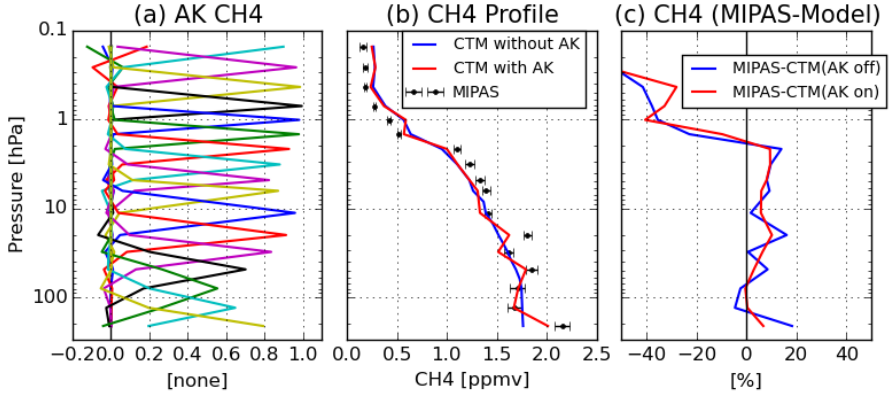

Figure 1. Example of MIPAS AK (left); MIPAS and BASCOE profiles (center); and profile differences MIPAS-BASCOE (right); for CH_4 on April 4, 2008 in the equatorial region. Profiles are given for MIPAS (black circles with error bars) and for two BASCOE CTM profiles interpolated at the MIPAS geolocation. Blue line is BASCOE interpolated at the observation location without using the AK while red line shows results when AK are used. Difference between MIPAS and BASCOE is given when AK are (red line) and are not used (blue line).

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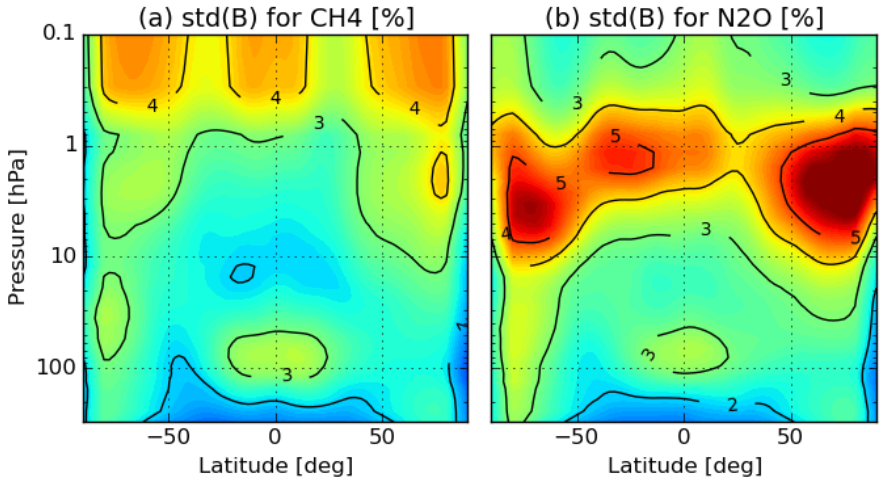

Figure 2. Standard deviation of the $\mathbf{B}$ matrix estimated by the ensemble method for CH_4 (left) and N_2O (right).

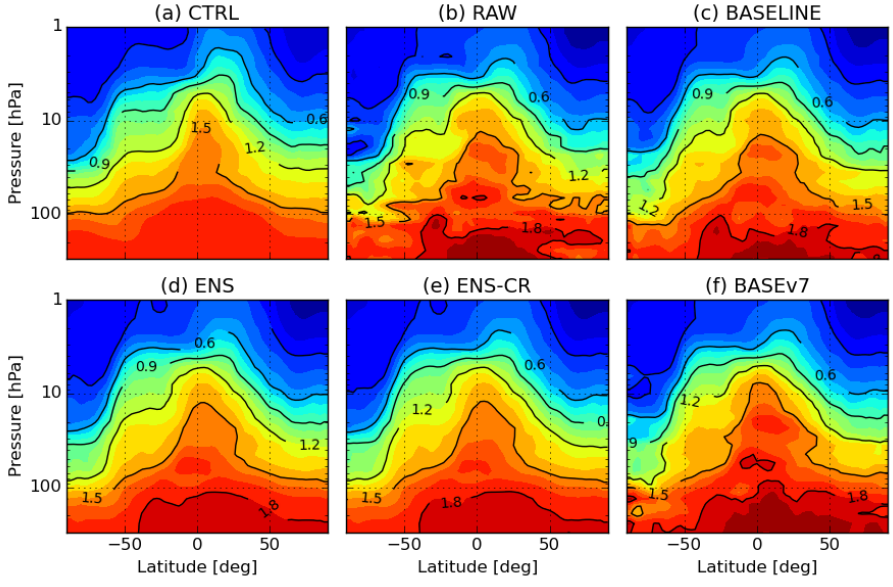

Figure 3. Zonal mean of CH_4 (ppmv) from six BASCOE experiments on September 15, 2008 at 12 UT. (see text for details).

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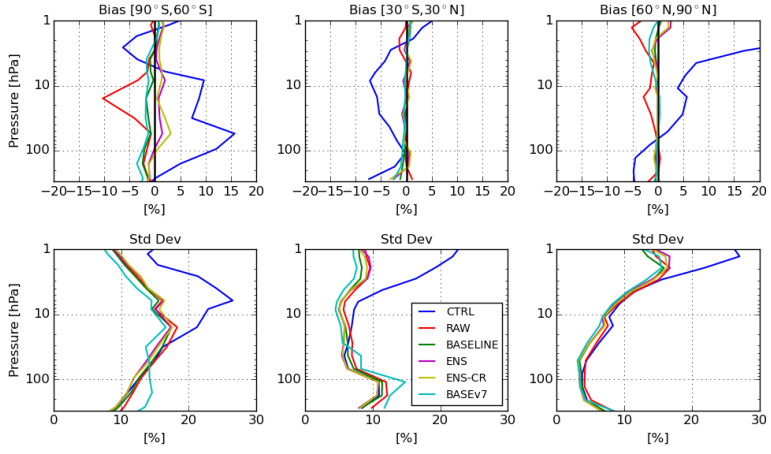

Figure 4. Mean (top) and standard deviation (bottom) of (BASCOE-MIPAS)/MIPAS for CH_4, for six BASCOE experiments (see text for details) and for the period September-October 2008. Left corresponds to South Pole (90°S-60°S), center corresponds to the tropics (30°S-30°N) and right corresponds to North Pole (60°N-90°N).

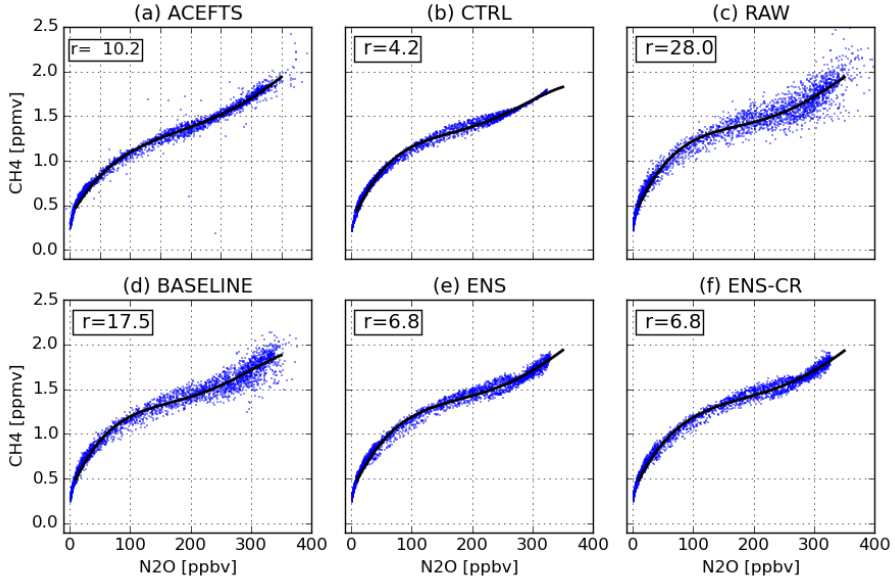

Figure 5. N_2O-CH_4 correlations between 30°S - 30°N as observed by ACE-FTS and for five BASCOE expriments interpolated to the ACE-FTS observation space (blue dots). The correlations are shown for the period April-October 2008. The result of a four-degree polynomial fit is also shown (black line) with the root mean square of the residual between the fitted curve and the observations is printed in the upper left of each plot.

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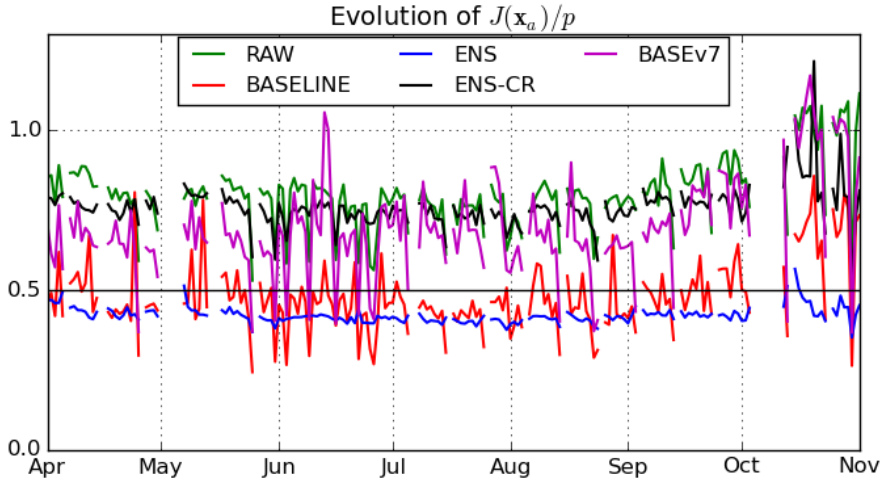

Figure 6. Time series of $J(\mathbf{x}_a)/p$ for the BASCOE experiments RAW, BASELINE, ENS, ENS-CR and BASEv7.

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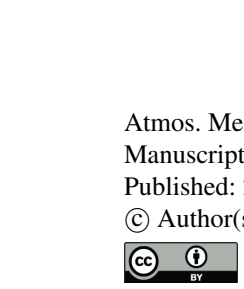

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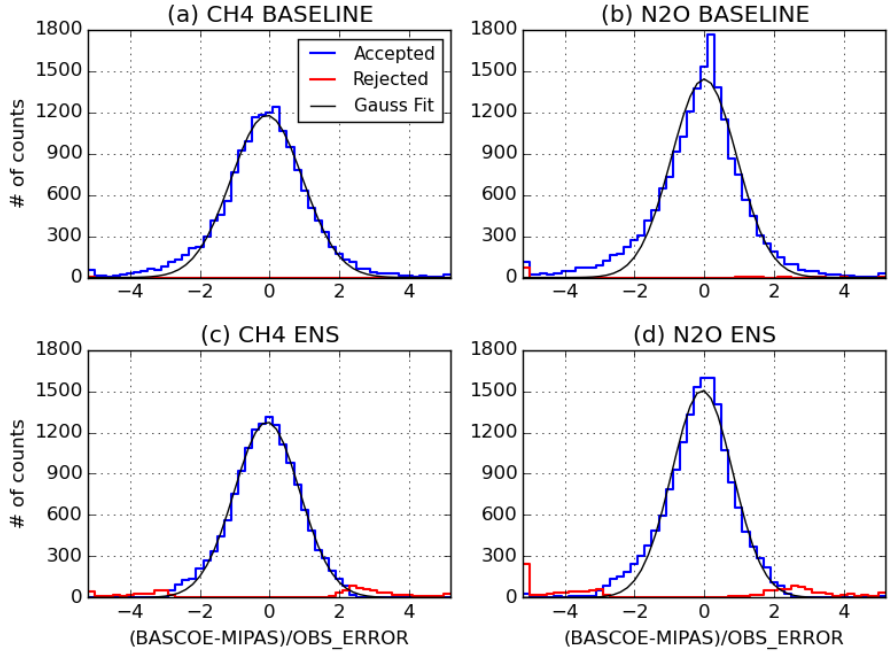

Figure 7. Histograms of counts of (BASCOE-MIPAS)/MIPAS_ERROR on August 20, 2008 for BASELINE (top row) and ENS (bottom row) experiments and for CH_4 (left column) and N_2O (right column). Blue and red lines correspond, respectively, to MIPAS observations accepted and rejected by the BgQC. Black line is a Gaussian fit of the blue line.

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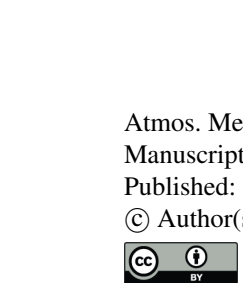

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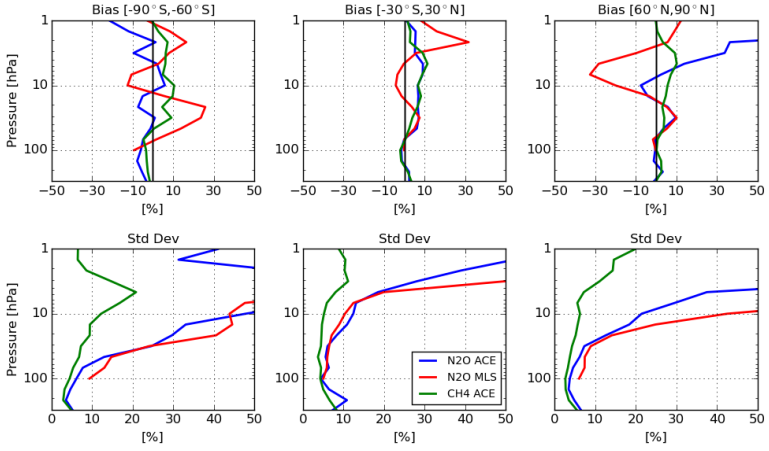

Figure 8. Mean (top) and standard deviation (bottom) of (ENS-independent_data)/independent_data where independent_data is ACE-FTS N_2O (blue line), ACE-FTS CH_4 (green line) or MLS N_2O (red line) for the period September-October 2008 and for three latitude bands (from left to right: South Pole, tropics and North Pole).

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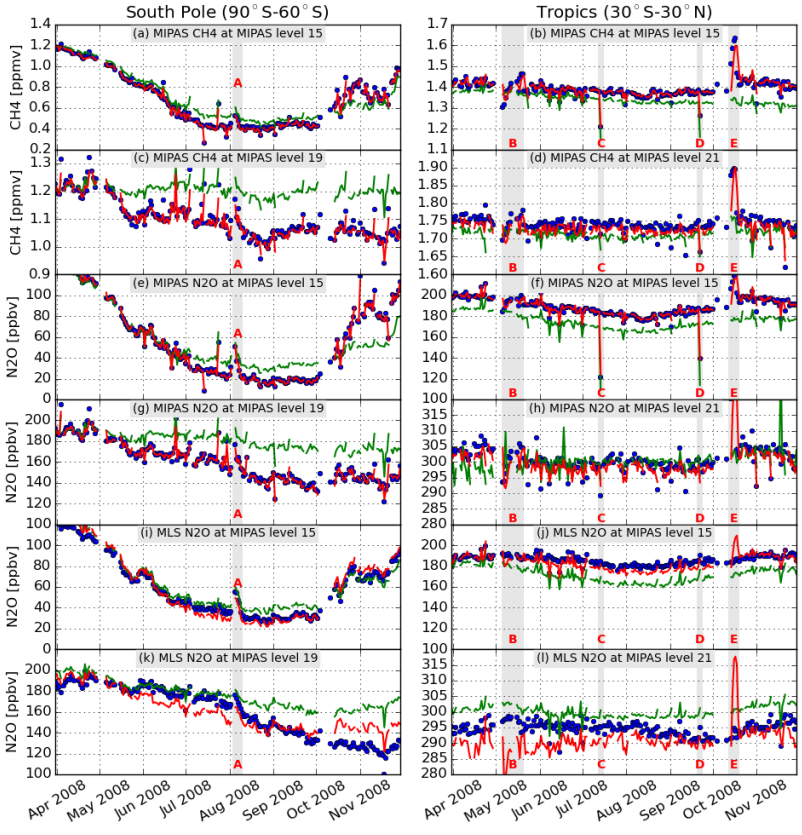

Figure 9. Time series between April and November 2008 for CH_4 and N_2O daily averaged in two latitude bands: between 90°S-60°S (left, southern polar region) and between 30°S-30°N (right, tropics). Above southern polar region, values are shown at MIPAS levels 15 and 19 which correspond approximately to 20 and 65 hPa. In the tropics, values are shown at MIPAS levels 15 and 21, around 15 and 70 hPa. Plots (a-d), (e-h) and (i-l) show respectively MIPAS CH_4, MIPAS N_2O and MLS N_2O (blue dots), and the corresponding values for ENS (red line) and CTRL (green line). In the case of MLS (plots i-l) , observations and BASCOE values are interpolated from the MLS pressure grid to the MIPAS daily average pressure. Shaded periods A, B, C, D and E are discussed in the text.

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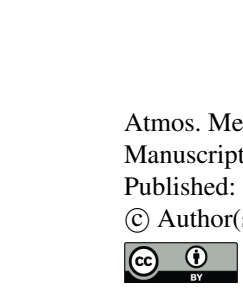

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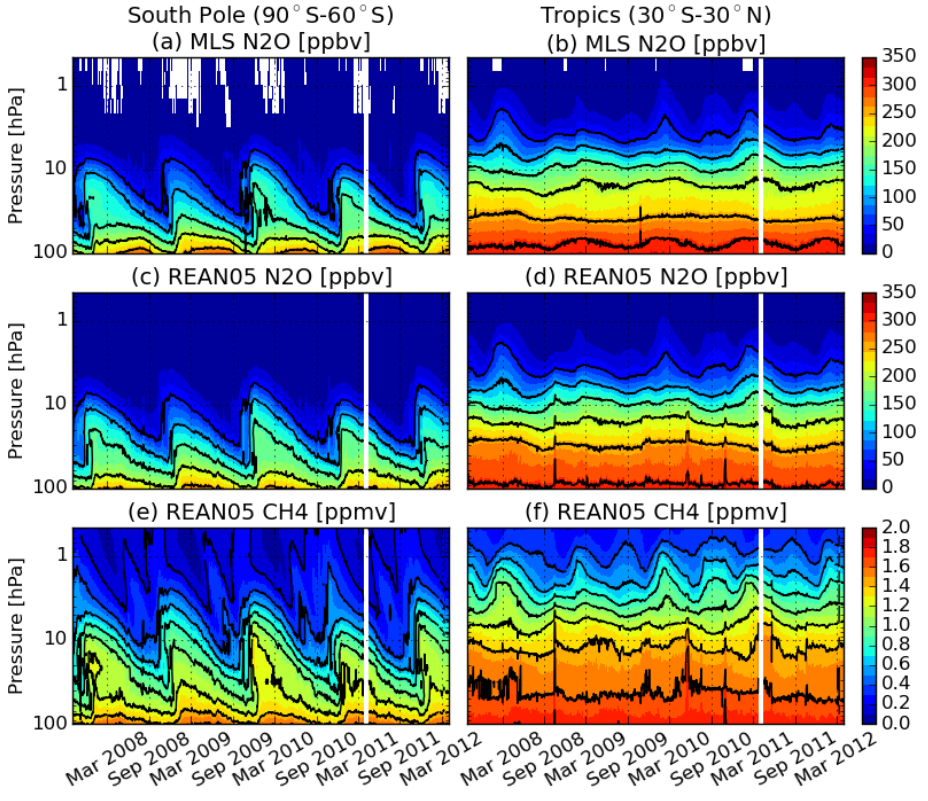

Figure 10. Top row: time series of daily average MLS N_2O between 90°S-60°S (left) and 30°S-30°N (right) for the period October 2007-April 2012. Middle row: as first row but for REAN N_2O. Bottom row: as first row but for REAN CH_4.

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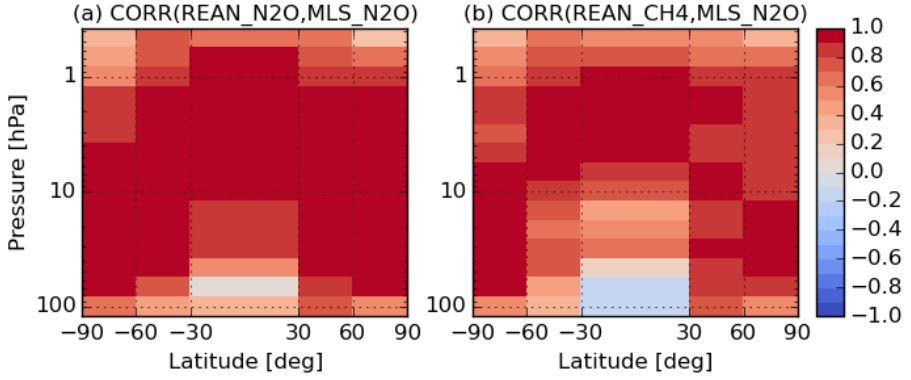

Figure 11. Correlation coefficients between (a) MLS N_2O and REAN N_2O and (b) MLS N_2O and REAN CH_4, as a function of latitude and pressure for the period October 2007-April 2012.

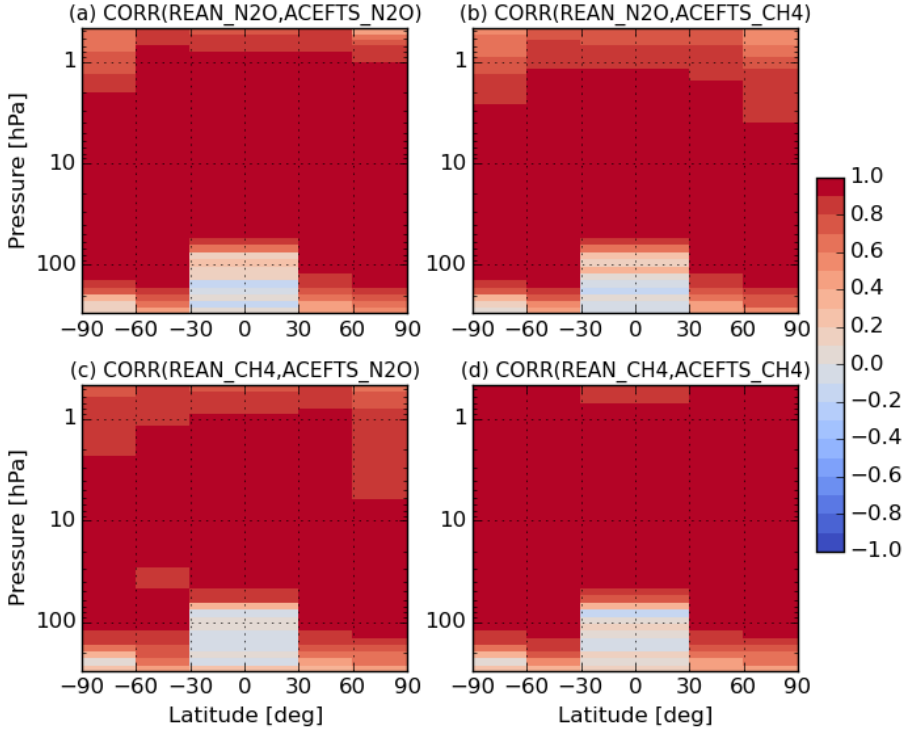

Figure 12. Correlation coefficients between (a) ACE-FTS N_2O and REAN N_2O (b) ACE-FTS CH_4 and REAN N_2O, (c) ACE-FTS CH_4 and REAN N_2O (d) ACE-FTS CH_4 and REAN CH_4, as a function of latitude and pressure for the period October 2007-April 2012.