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Harmonisation and diagnostics of MIPAS ESA CH₄ and N₂O profiles using data assimilation

Quentin Errera¹, Simone Ceccherini², Yves Christophe¹, Simon Chabrillat¹, Michaela I. Hegglin³, Alyn Lambert⁴, Richard Ménard⁵, Piera Raspollini⁴, Sergey Skachko¹, Michiel van Weele⁶, and Kaley A. Walker^{7,8}

¹Belgian Institute for Space Aeronomy (BIRA-IASB), Brussels, Belgium

²Istituto di Fisica Applicata “N. Carrara” (IFAC) del Consiglio Nazionale delle Ricerche (CNR), Firenze, Italy

³University of Reading, Department of Meteorology, Reading, UK

⁴Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California, USA

⁵Air Quality Research Division, Environment and Climate Change Canada, Dorval, Canada

⁶Royal Netherlands Meteorological Institute (KNMI), De Bilt, the Netherlands

⁷University of Toronto, Department of Physics, Toronto, Canada

⁸University of Waterloo, Department of Chemistry, Waterloo, Canada

Correspondence to: Quentin Errera (quentin@oma.be)

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Abstract. This paper discusses assimilation experiments of methane (CH₄) and nitrous oxide (N₂O) profiles retrieved from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS). Here we focus on data versions 6 and 7 provided by the ESA processor. These data sets have been assimilated by the Belgian Assimilation System for Chemical Observations (BASCOE). The CH₄ and N₂O retrieved profiles can oscillate, especially in the tropical lower stratosphere. Using the averaging kernels of the observations and a background error covariance matrix, which has previously been 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 from ACE–FTS (CH₄ and N₂O) and MLS (N₂O), demonstrating the general good quality of CH₄ and N₂O retrievals provided by MIPAS ESA. Nevertheless, this study also identifies two issues in these data sets. First, time series of the observations show unexpected discontinuities due to an abrupt change in the gain of MIPAS band B, generally occurring after the instrument decontamination. Since the calibration is performed weekly, the abrupt change in the gain affects the measurements until the subsequent calibration is performed. 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 CH₄ and N₂O retrievals with caution.

1 Introduction

Carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O) 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 CH₄ concentration has been observed and the radiative forcing from CH₄ is now larger than all of the combined halocarbons. The concentration of N₂O is also increasing such that, due to the decrease of chlorofluorocarbons 12 (CFC-12), N₂O is now the third largest WMGHG contributor to radiative forcing.

While CH₄ and N₂O are considered well-mixed greenhouse gases, both gases exhibit a vertical profile and significant spatio-temporal 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 re-

sults 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. Therefore, the change in the stratosphere in response to anthropogenic climate forcing requires an accurate representation of (past and future) stratospheric composition changes. Moreover, some studies have also 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).

NWP models, global climate models and earth system models include dynamics, 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) in addition to detailed chemistry modules (e.g. Huijnen et al., 2016). Linearisation of the stratospheric chemistry (Hsu and Prather, 2010; Monge-Sanz et al., 2013) could be a computationally attractive approach, though their implementation would require further studies as well as a full reanalysis, or at least a chemically 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 CH₄ and N₂O as presented in this work provides a 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 synthesised in a numerical model (Lahoz et al., 2010). These methods have been applied to both the physical and chemical states 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, Simmons et al. (2014) 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 characterise changes in the observation errors.

CH₄ and N₂O are both emitted 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 the retrievals of these two species, we have a clean assimilation of information content that allows us to compare these two data sets and validate the assimilation methodology. In practice, as we will see, the assimilation of MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) CH₄ and N₂O shows a number of issues that need to be addressed. These issues are specific to the retrievals that use spectral points in MI-

PAS band B, mainly CH₄ and N₂O. 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 their study, CH₄ profiles 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 CH₄ and N₂O. In order to preserve the correlations, an a posteriori correction of the analysis was applied to every modelled species which are correlated with CH₄. 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. All the assimilation experiments performed in this study have been done with the chemical scheme of BASCOE turned off. This is possible since the chemical processes that affect CH₄ and N₂O occur on timescales which are much longer than the length of the assimilated window used in this study.

This paper is organised 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 set-up are presented in Sect. 3. Section 4 compares the assimilation experiments of MIPAS CH₄ and N₂O and Sect. 5 presents their comparison with independent data. Finally, Sect. 6 summarises the results of this paper.

2 Observations

2.1 MIPAS

The assimilated CH₄ and N₂O profiles 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 CH₄ and N₂O (Fischer et al., 2008). The MIPAS mission is divided in two phases: the full-resolution (FR) phase from 2002 to 2004 and the optimised-resolution (OR) phase from 2005 to 2012. The latter period is characterised by 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

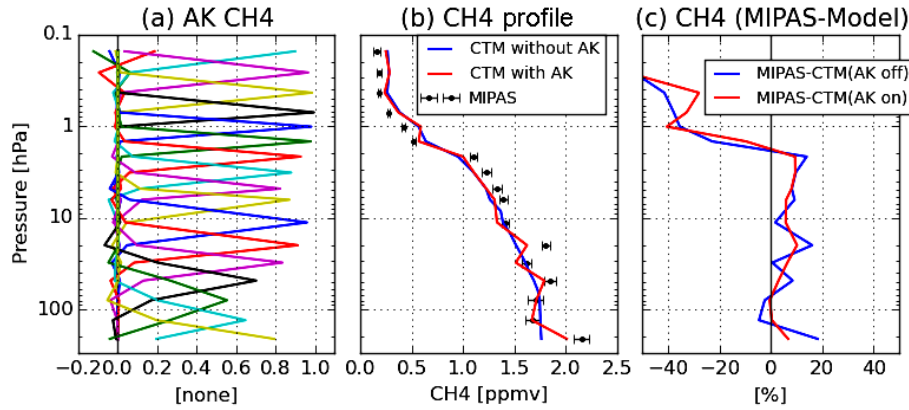


Figure 1. Examples of MIPAS AKs (a), MIPAS and BASCOE profiles (b) and profile differences MIPAS–BASCOE (c) for CH₄ on 4 April 2008 in the equatorial region. Profiles are given for MIPAS (black circles with error bars) and for two BASCOE CTM profiles interpolated at the MIPAS geolocation. Blue line is BASCOE interpolated at the observation location without using the AKs while red line shows results when AKs are used. Difference between MIPAS and BASCOE is given when AKs are (red line) and are not used (blue line).

different modes of observations, for example, the nominal mode (NOM) with altitude soundings between 7 and 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 data set is considered in this study. Note that in OR measurements, NOM mode is characterised by a floating-altitude measurement grid. This means that the limb sounding grid is shifted rigidly with the lowest measured altitude, which depends on the latitude. The floating-altitude sampling grid is meant to roughly follow the tropopause height along the orbit with the requirement to collect at least one spectrum within the troposphere while avoiding too many spectra affected by clouds.

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 exhibit some unphysical oscillations in individual CH₄ and N₂O profiles caused by the processing algorithm (which used almost no regularisation).

ML2PP v6 profiles of CH₄ and N₂O (full and optimised 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 the instruments above 20 km altitude, within the estimated uncertainty limits. This good agreement is also observed below 20 km for CH₄ while MIPAS N₂O underestimated BONBON by around 20 ppbv at 15 km.

Vertical averaging kernels (AKs) of MIPAS data, which are provided for each profile, have been used in this study. In data assimilation, the vertical AKs are used to interpolate the

model fields to the vertical grid of the retrieved profiles, as follows (Rodgers and Connor, 2003):

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

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 “ \sim ” 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. The interpolation is done along the logarithm of the pressure and $\tilde{\mathbf{A}}$ is normalised in order to have $\sum_j A_{ij} = \sum_j \tilde{A}_{ij}$ where i and j denote, respectively, the row and line of \mathbf{A} or $\tilde{\mathbf{A}}$.

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 denotes 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 AKs 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 AKs when interpolating BASCOE fields to MIPAS observation locations is illustrated in Fig. 1 for a MIPAS CH₄ profile taken in the equatorial region. Figure 1a shows the AK profiles that correspond to the CH₄ profile displayed in Fig. 1b. The AKs 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 AKs. 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 AKs is much smoother. This is expected since there is no source or sink of CH₄ in the lower stratosphere. On the other hand, the BASCOE profile interpolated with the use of the AKs 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. When the AKs are not used, the increment can oscillate such that undesirable oscillations are introduced in the model. On the other hand, the increment based on the use of the AKs is much more regular such that the BASCOE analyses will also be more regular vertically.

Note that horizontal AKs 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 AKs (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 L1v7 (used by the L2v7 processor) and L2v7 products, as well as the auxiliary data, including 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 the L1 processor, changes in the handling of the retrieval of continua, etc. In general, v7 CH₄ and N₂O products show higher concentrations than in v6, mainly in the tropics at very low altitudes. Also, v7 OR measurements use a stronger regularisation that should provide fewer oscillating profiles of CH₄ and N₂O.

2.2 ACE-FTS

The BASCOE analyses are validated by comparison with two independent data sets. 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-optimised resolution. This instrument is based on the solar occultation measurement technique which provides around 28 profiles per day (Boone et al., 2005, 2013). Here, version 3.5 of N₂O and CH₄ retrievals has been used where a data quality flag has been implemented since previous versions.

ACE-FTS v3.5 N₂O has been compared against profiles from MIPAS and MLS (Sheese et al., 2016). The coincidence criteria used in that study required observations to be made within 3 h and 350 km of each other. This provides around 100 collocated pairs of profiles, 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 data sets,

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 ± 10 %.

For CH₄, only ACE-FTS v2.2 has been validated against independent observations. An agreement within ± 10 % was found in the upper troposphere/lower stratosphere and within ± 25 % in the middle and higher stratosphere up to the lower mesosphere (below 60 km, De Mazière et al., 2008).

2.3 Aura MLS

The second data set 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 N₂O 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 to the recommendation 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 N₂O precision is $24-14$ ppbv ($9-41$ %) and the accuracy is to be $70-3$ ppbv ($9-25$ %) (Livesey et al., 2011). Although vertical AKs 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 AKs for N₂O has been tested and was found to be very small, mainly because the AK profiles are peaked at the tangent point (not shown). Note that in the case of MLS, the AKs 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 chemistry 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 parameterisation of the physico-chemical processes due to polar stratospheric clouds (PSCs). However, in this study, only the advection of CH₄ and N₂O 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 N₂O and CH₄ when using an assimilation window of 1 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 $3.75^\circ \times 2.5^\circ$ 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.

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 minimising 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 the 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). Background error correlations between species are not taken into account. Spatial correlations have 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 data sets in which 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 CH₄ and N₂O to which a random noise has been added, 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 the experiments allows one to estimate the background error covariance matrix. Figure 2 displays the calibrated standard deviation of the \mathbf{B} matrix for N₂O and CH₄. 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} (which accounts only for the random noise errors) is given

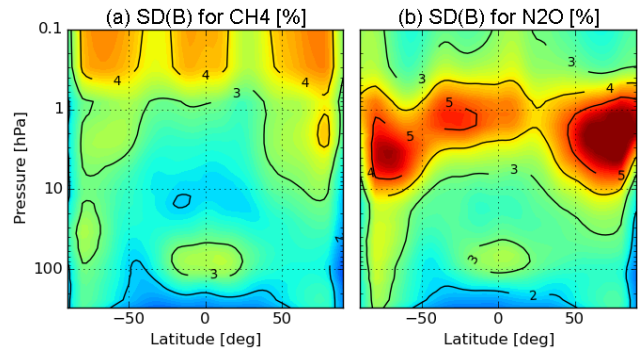


Figure 2. Standard deviation of the \mathbf{B} matrix estimated by the ensemble method for CH₄ (left) and N₂O (right).

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.

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

$$(\mathbf{y} - H(\mathbf{x}_b))_{ij}^2 > \alpha (\sigma_{ij}^o + \sigma_{ij}^b), \quad (4)$$

where i and j denote the number of the profile and the altitude index, and σ^o and σ^b denote the error variance of the observations and of the background state, the latter being interpolated from the \mathbf{B} matrix at the observation location. 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 realised for this paper aim to evaluate the impact of (1) the use of the AKs, (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 data sets. These experiments are summarised in Table 1. Experiment CTRL is a CTM run without data assimilation. Experiment RAW is a chemistry data assimilation (CDA) run in which AKs and vertical correlations in \mathbf{R} are not used and \mathbf{B} is not calibrated. Experiment BASELINE is as RAW except that AKs are used. Experiment ENS is like BASELINE but the covariances of \mathbf{B} have been estimated by the ensemble method (see Sect. 3). Experiment ENS-CR is like ENS but the vertical correlations in \mathbf{R} are used. Experiment BASEv7 is like 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

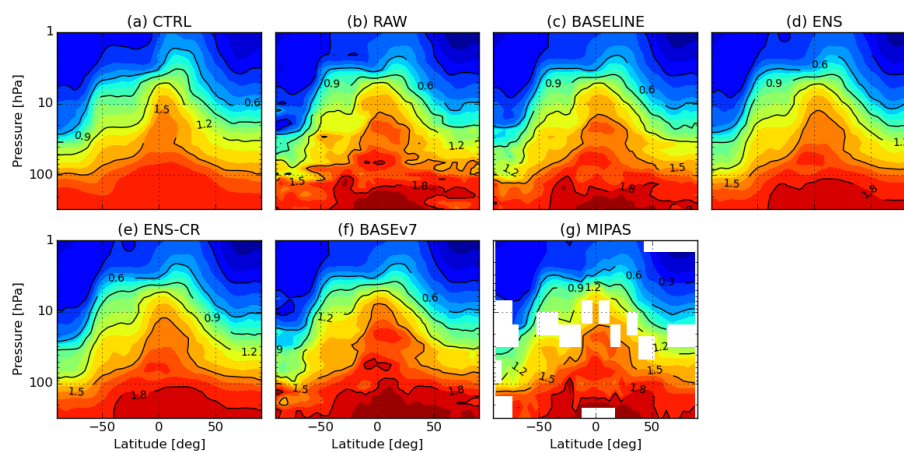


Figure 3. Zonal mean of CH₄ (ppmv) from six BASCOE experiments on 15 September 2008 at 12:00 UT and from MIPAS measured on the same date (see text for details). Here MIPAS data are binned on a 5° latitude grid and a pressure grid with 6 levels per decade of pressure.

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

Name	CTM/CDA ^a	MIPAS version	Use of AKs	Use of corr(R) ^b	Calibrated B	Period
CTRL	CTM	n/a	n/a	n/a	n/a	Apr–Nov 2008
RAW	CDA	v6	No	No	No	Apr–Nov 2008
BASELINE	CDA	v6	Yes	No	No	Apr–Nov 2008
ENS	CDA	v6	Yes	No	Yes	Apr–Nov 2008
ENS-CR	CDA	v6	Yes	Yes	Yes	Apr–Nov 2008
BASEv7	CDA	v7	Yes	No	No	Apr–Nov 2008
REAN	CDA	v6	Yes	No	Yes	Mar 2007–Apr 2012

^a Free chemistry transport model (CTM) run or chemical data assimilation (CDA) run. ^b Use of vertical correlation in **R**. n/a: not applicable.

2007–April 2012 and has been set up as ENS. Experiments with uncalibrated **B** use the following set-up 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 N₂O–CH₄. Some results in this section are shown only for CH₄ or N₂O but the same conclusions are found for the other species.

Figure 3 shows the zonal mean analysis of CH₄ from six BASCOE experiments on 1 May 2008 at 12:00 UT which is representative of other dates. The MIPAS CH₄ zonal mean is also shown in the figure in order to allow comparison with the assimilated data. The CTM run (CTRL) shows the well-known features of the CH₄ 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’s surface and has a lifetime of 10 years (Ko et al., 2013). In the stratosphere, methane is destroyed by oxidation with OH, O¹D and Cl (Brasseur and Solomon, 2005). Methane has no source in the atmosphere.

In those conditions, no physical or chemical processes can explain the small structures in the zonal mean of CH₄ in experiment RAW (e.g. local maxima around 80 hPa in the tropics or at 30 hPa and 50° S). These structures are smoothed when the assimilation system uses the averaging kernels (exp. BASELINE) and a calibrated **B** matrix (exp. ENS). The use of the vertical correlations in the **R** matrix apparently has a very small impact when comparing experiments ENS and ENS-CR. BASEv7 shows larger CH₄ volume mixing ratios than BASELINE, between 5 and 10% depending on the region. It is also found that the small structures in BASELINE are still present in BASEv7. This suggests that MIPAS v7 does not provide more regular profiles of CH₄ 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 N₂O (not shown).

Figure 4 shows the CH₄ O–F residuals between BASCOE and MIPAS for September–October 2008. CTRL results in

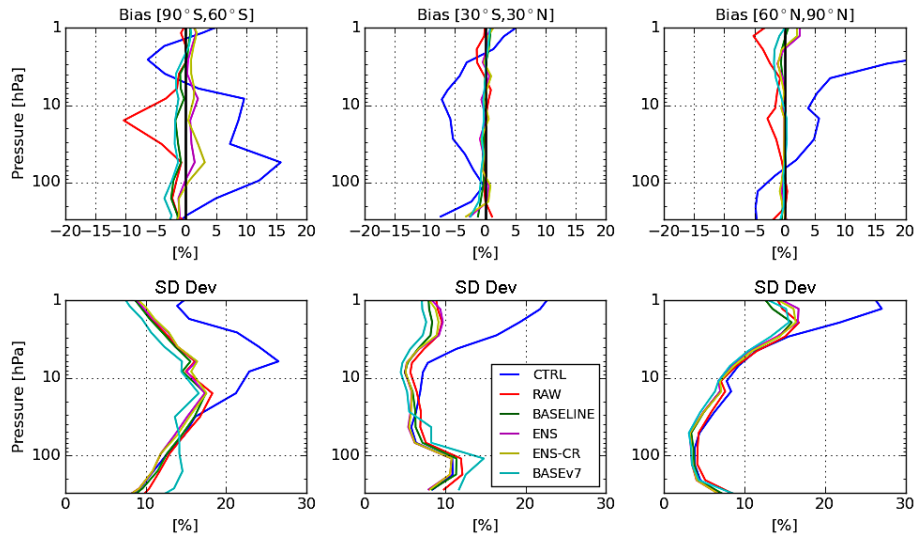


Figure 4. Mean (top) and standard deviation (bottom) of (BASCOE – MIPAS) / MIPAS for CH₄, for six BASCOE experiments (see text for details) and for the period September–October 2008. Left corresponds to South Pole (90–60° S), centre corresponds to the tropics (30° S–30° N) and right corresponds to North Pole (60–90° N).

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. From careful observation, 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 other experiments above 10 hPa but larger in the lower stratosphere.

The N₂O–CH₄ scatter plots 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. Data are shown only for the 30° S–30° N latitude band in order to provide a more compact scatter plot and better illustrate the different results obtained by each assimilation experiment. 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 scatter plot 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 perform much better when the system uses additional information from the AKs ($r = 17.5$ for BASELINE) and from the calibration of **B** ($r = 6.8$ for ENS and ENS-CR). Again, the use of the correlations in the **R** matrix does not improve the

results (same rms for ENS and ENS-CR). BASEv7 has a better scatter plot ($r = 12.7$) than BASELINE (not shown).

Figure 6 shows the evolution of the cost function at the analysis point $J(x_a)$ 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 $J(x_a)/p$ 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. BASEv7 has values of $J(x_a)/p$ which are larger than those from BASELINE and even larger than 1/2. ENS-CR shows a much larger deviation from theoretical values. This suggests that the observational error covariance matrices provided by the MIPAS ML2PP retrieval have not been used optimally by BASCOE. In particular, the observational error variance of ML2PP retrieval probably needs to be tuned and inflated to account for the additional weight of the observations due to their correlation. This has not been done in this study.

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 x_b and the variances of **B** which are different in each experiment. In particular, the standard deviation of **B** used in ENS and ENS-CR is much smaller than the 20 % used in 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 CH₄ and N₂O on 20 August 2008. These histograms are computed separately

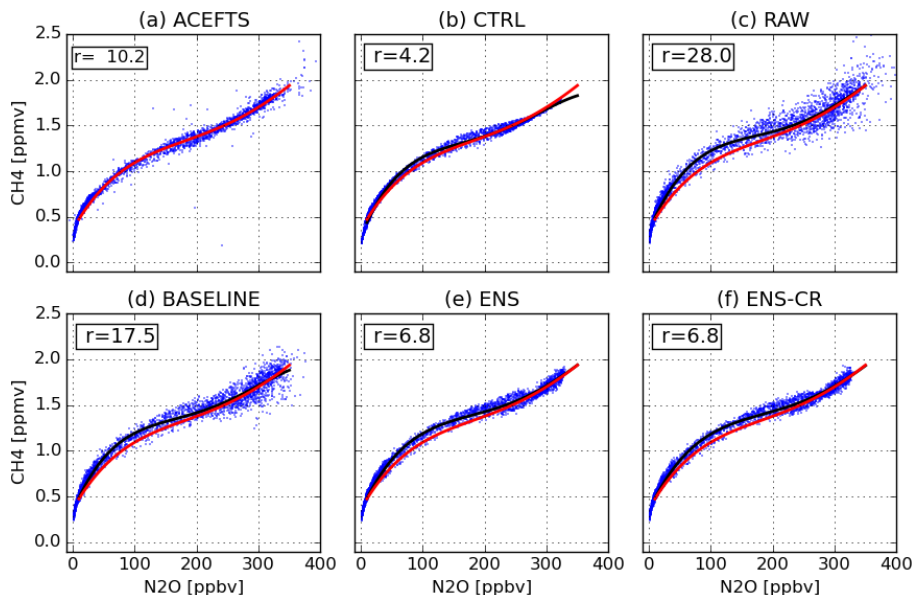


Figure 5. N₂O–CH₄ scatter plots between 30° S–30° N as observed by ACE–FTS (a) and for five BASCOE experiments interpolated to the ACE–FTS observation space (b–f). The correlations are shown for the period April–October 2008. The result of a four-degree polynomial fit is also shown (red line for ACE–FTS, black lines for BASCOE experiments) with the root mean square of the residual between the fitted curve and the observations printed in the upper left of each plot. The correlation curve of ACE–FTS (red curve) is reported in the correlation plots of the BASCOE experiments (b–f).

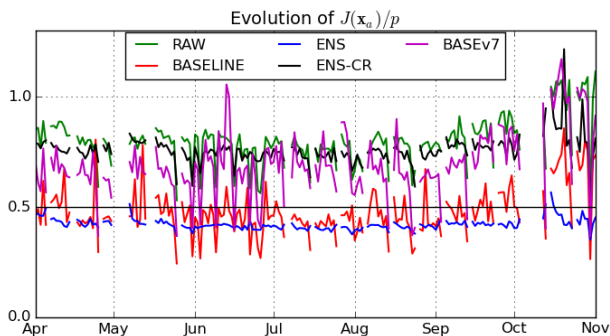


Figure 6. Time series of $J(x_a)/p$ for the BASCOE experiments RAW, BASELINE, ENS, ENS-CR and BASEv7.

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 is a Gaussian distribution, the number of observations accepted in BASELINE is too high at large differences. The use of the calibrated **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 question now remains 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, BASELINE and RAW. ENS provides the best results as it benefits from the use of the AKs, a calibrated **B** matrix and an optimal use of the BgQC filter. Experiment ENS-CR has been excluded from the ranking. As mentioned above, this experiment needs further tests to take into account the additional weight of the observations due to their vertical correlations.

A comparison between BASELINE and BASEv7 also allows us to compare MIPAS v6 and v7. Zonal means are comparable (Fig. 3), O–F and $J(x_a)/p$ are better with BASELINE (Figs. 4 and 6), and the N₂O–CH₄ scatter plot is better with BASEv7. 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 CH₄ and N₂O.

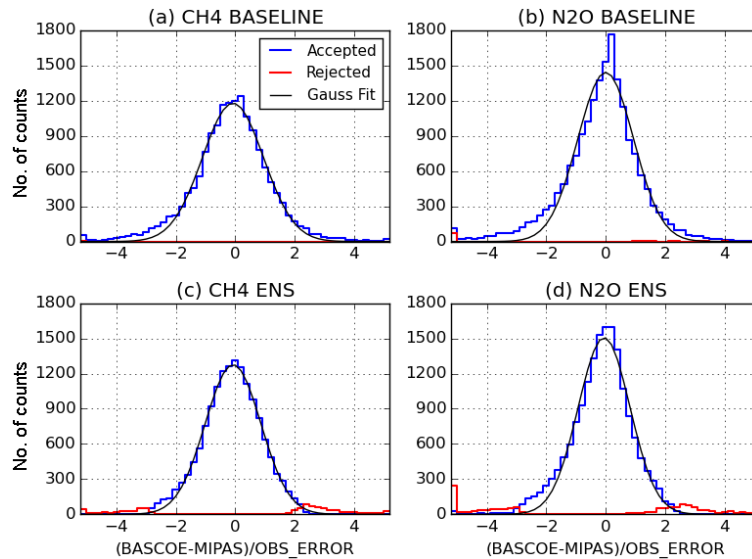


Figure 7. Histograms of counts of (BASCOE-MIPAS)/MIPAS_ERROR on 20 August 2008 for BASELINE (top row) and ENS (bottom row) experiments and for CH₄ (left column) and N₂O (right column). Blue and red lines correspond to MIPAS observations accepted and rejected by the BgQC respectively. Black line is a Gaussian fit of the blue line.

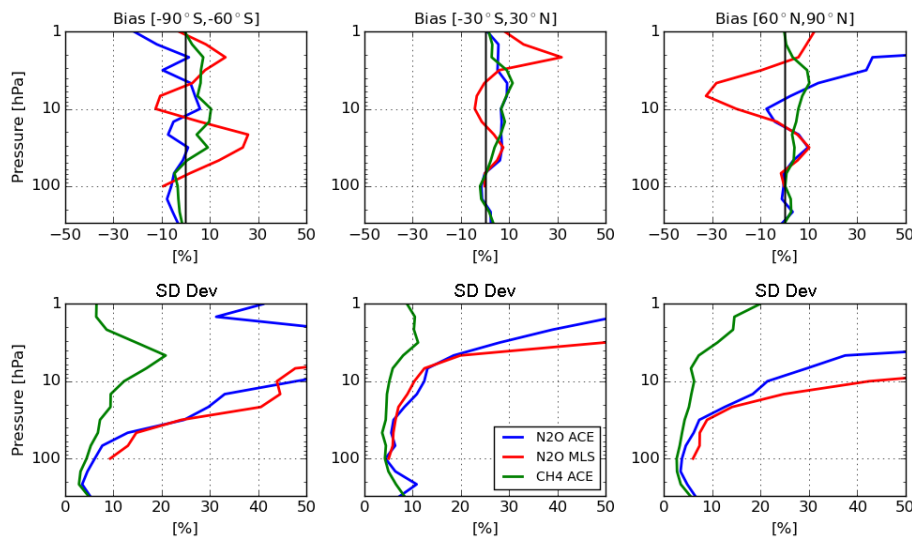


Figure 8. Mean (top) and standard deviation (bottom) of $(ENS - independent_data) / independent_data$ where independent_data is ACE-FTS N₂O (blue line), ACE-FTS CH₄ (green line) or MLS N₂O (red line) for the period September–October 2008 and for three latitude bands (from left to right: South Pole, tropics and North Pole).

5 Comparison with independent observations

Experiment ENS is evaluated against independent profiles from MLS and ACE-FTS for N₂O and from ACE-FTS for CH₄. 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. The sampling error is then minimised since the maximum difference of time between the observed profile and the BASCOE time step is 15 min.

Figure 8 shows the O–F residuals between ENS and these three independent data sets for the period September–October 2008 in three regions: the south polar cap, the tropics and the north polar cap. For CH₄, ENS and ACE-FTS are in good agreement: the mean differences and the associated standard deviations are mostly below $\pm 10\%$. This is in the range of comparison between ACE-FTS CH₄ and independent observations (De Mazière et al., 2008). For the comparison of N₂O with ACE-FTS, there is also a good agreement. The mean differences are usually below $\pm 10\%$. The stan-

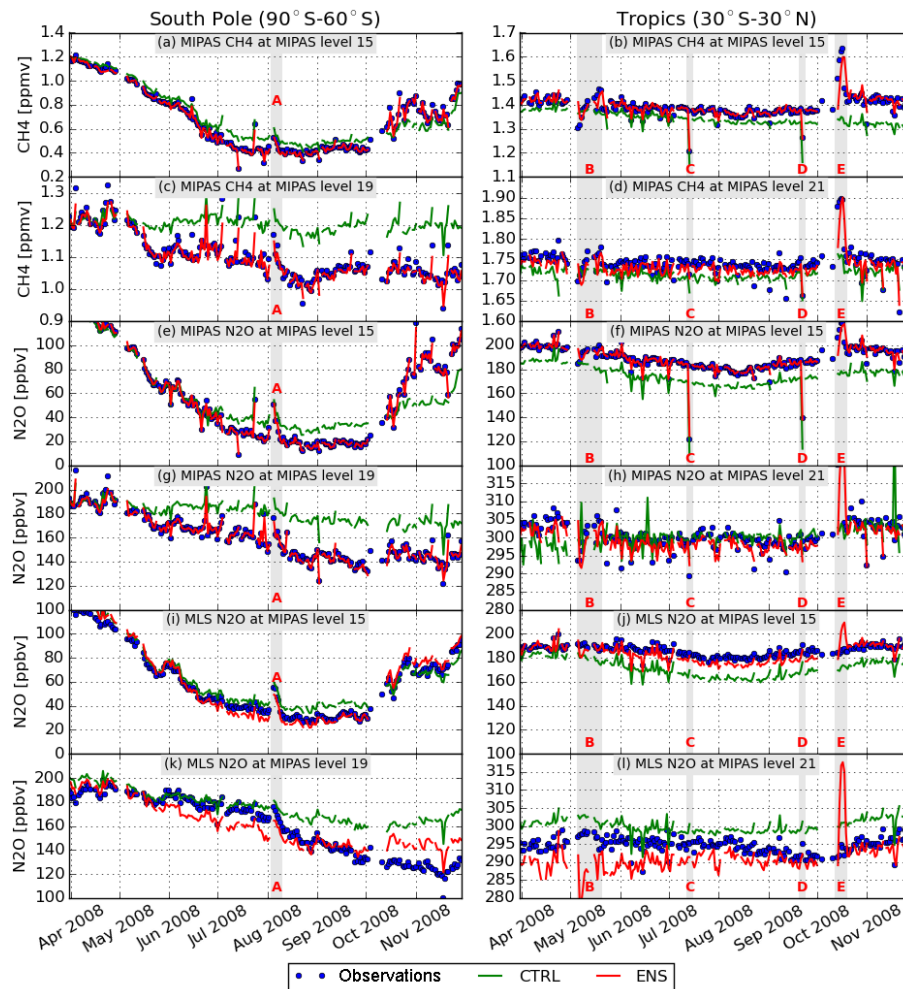


Figure 9. Time series between April and November 2008 for CH₄ and N₂O daily averaged in two latitude bands: between 90–60° S (left, southern polar region) and between 30° S–30° N (right, tropics). Above the southern polar region, values are shown at MIPAS levels 15 and 19 which correspond approximately 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 MIPAS CH₄, MIPAS N₂O and MLS N₂O (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.

dard deviation is relatively small ($< 10\%$) in regions where N₂O 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 N₂O 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 in validation paper (see Sect. 2.2), except in the South Pole region. Below 4.61 hPa, the mean difference between ENS and MLS N₂O 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 Fig. 9e and g) and ACE–FTS, these differences suggest that MLS underes-

timates N₂O concentration in the polar vortex (in agreement with Sheese et al., 2016). Overall, ENS and independent data agree within the uncertainties of each independent data set (see Sect. 2.2 and 2.3).

Figure 9 shows the time series of CH₄ and N₂O between April and November 2008 above the south polar region and in the tropics at two MIPAS levels (the middle stratosphere and lower stratosphere) for MIPAS, ENS and CTRL and MLS (N₂O only). ENS and CTRL values are obtained using the averaging kernels of MIPAS. Five periods are shaded and labelled by the letters A, B, C, D and E, and are discussed below. As expected, MIPAS and ENS are in good agreement for CH₄ and N₂O. 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

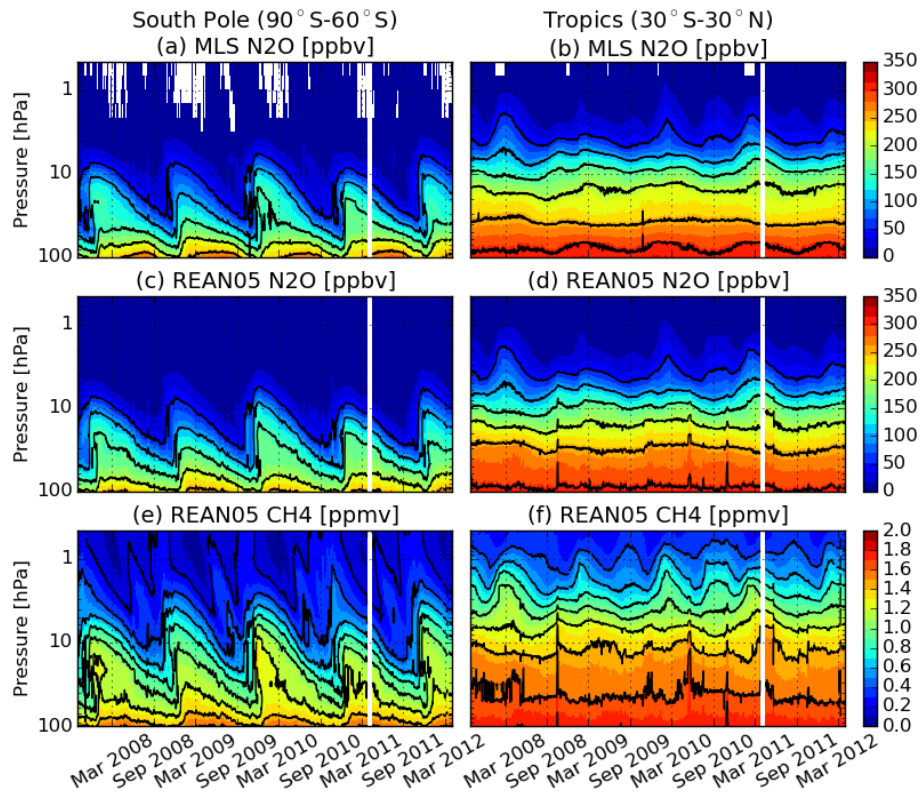


Figure 10. Top row: time series of daily average MLS N₂O between 90–60° S (left) and 30° S–30° N (right) for the period October–April 2012. Middle row: as first row but for REAN N₂O. Bottom row: as first row but for REAN CH₄.

– in particular the number of profiles per day can vary from 0 to ~ 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 show an increase in CH₄ and N₂O 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 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.

These discontinuities are due to the combination of two effects. The first one is due to the decontamination procedure of the instrument (i.e. a warming-up of the instrument to remove the ice) which is operated once or twice per year. Sometimes after the decontamination an abrupt change, as high as 2 %, is observed in the radiometric gain of band B where CH₄ and N₂O are retrieved. The second effect is due to the calibration of the L1 data, which is done once a week. Since the change in the gain occurs in the time frame of 1–2 days, and since the calibration is not performed at the corre-

sponding times, a discontinuity in CH₄ and N₂O time series is introduced. This issue should be resolved in the future version 8 of MIPAS by doing daily calibration of L1 data. Days for which L1 data are not properly calibrated are reported in De Laurentis and Raspollini (2016, see Table 5).

Figure 10 shows the time series of daily averaged MLS N₂O above the south polar region and the tropics for a period of 5 years (April 2007–April 2012). Seasonal variations of N₂O are clearly visible at South Pole but also in the tropics, in particular in the lower stratosphere. In addition, the corresponding time series of N₂O and CH₄ 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, like ENS, REAN agrees very well with MIPAS. Hence the differences between MLS N₂O and REAN N₂O highlight differences between MLS and MIPAS. The agreement between the data sets is very good overall. Discontinuities due to the abrupt change in the radiometric gain can be seen in REAN analyses, especially in the tropics. In that region, REAN analyses are relatively noisy and the N₂O seasonal variations observed by MLS are not reproduced by the reanalysis of MIPAS N₂O and CH₄. Note that seasonal variations observed by MLS are hardly seen in the MIPAS raw data (not shown).

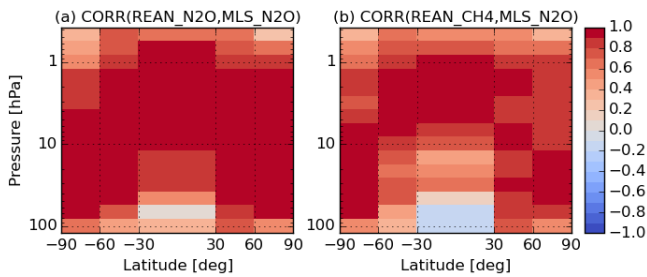


Figure 11. Correlation coefficients between (a) MLS N₂O and REAN N₂O, and (b) MLS N₂O and REAN CH₄ as a function of latitude and pressure for the period October 2007–April 2012.

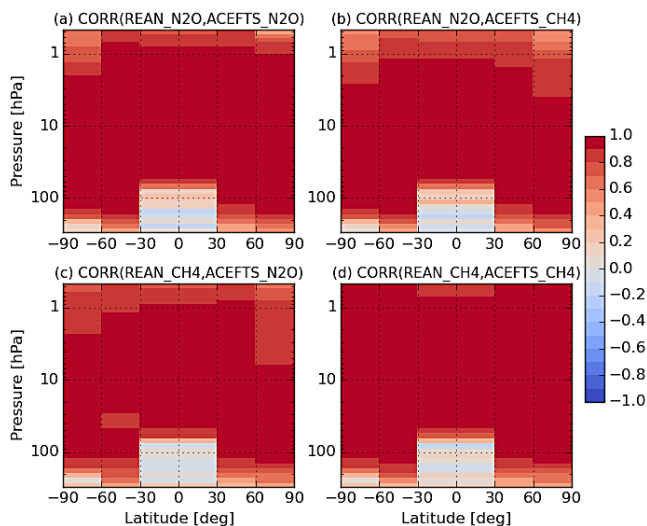


Figure 12. Correlation coefficients between (a) ACE–FTS N₂O and REAN N₂O, (b) ACE–FTS CH₄ and REAN N₂O, (c) ACE–FTS CH₄ and REAN N₂O, (d) ACE–FTS CH₄ and REAN CH₄ as a function of latitude and pressure for the period October 2007–April 2012.

This result is confirmed in Figs. 11 and 12 which show the correlation coefficients between MLS (N₂O) and REAN (N₂O and CH₄), and between ACE–FTS (N₂O and CH₄) and REAN (N₂O and CH₄) for the period October 2007–April 2012. These correlations are calculated on latitude bins space by 30° (60° at the Equator) and on a pressure grid with 6 levels per decade of pressure. 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 dates where L1 data are not properly calibrated 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 N₂O and CH₄ 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 tropical lower stratosphere. On the other hand, Sheese et al. (2016) based their comparison on 100 collocated profiles between ACE–FTS and MIPAS although most profiles are located at mid-latitudes 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 characterisation of MIPAS, we recommend carrying out a specific validation of MIPAS in the tropics. According to the diagnostics that have been set up for this study, there is not a clear indication that MIPAS v7 improves the quality of CH₄ and N₂O with respect to v6.

6 Conclusions

This paper presents assimilation experiments of MIPAS ESA version 6 N₂O and CH₄ profiles using the BASCOE system. For these two constituents, MIPAS profiles can oscillate, and this study shows that assimilation can help to harmonise these data sets. This is possible because assimilation systems can use additional information to the raw profiles: (1) the averaging kernels associated with each profile and (2) the background error covariance matrix which was calibrated using an ensemble method. Using this set-up, the BASCOE analyses are in good agreement with independent observations measured by MLS and ACE–FTS, usually within ±10%. This is due to the generally good quality of MIPAS profiles.

Nevertheless, this study also diagnoses two issues in MIPAS CH₄ and N₂O profiles. First, time series of MIPAS profiles show unexpected discontinuities which are due to the abrupt change in the radiometric gain of the instrument. A daily calibration might resolve this issue. While identified in this paper, this issue could have been found by data analysis method other than data assimilation. 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. This second issue was not identified in previous validation studies of MIPAS. One possible reason is that the method used to compare satellite observations usually depends on a coincidence criteria that limits the size of the sample. This is not the case any more if one compares data assimilated fields from one

satellite instrument with observations from another satellite instrument as done in this paper. These two issues are also present in MIPAS version 7 and will be addressed in the future, in version 8.

Overall, this study recommends using MIPAS ESA CH₄ and N₂O v6 and v7 profiles with caution in the tropical lower stratosphere. In order to improve the characterisation of MIPAS, we also recommend carrying out a specific validation of MIPAS in the tropics, which was not the case in previous validation studies. We recommend that ESA improves the data quality document, which should come with any release of a new version of the L2 product, as 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. It would greatly help members of the scientific community who work with ESA products. This will be done for the MIPAS L2 v7 release in which all issues highlighted in this paper and other known issues will be mentioned.

7 Data availability

The BASCOE analyses produced for this paper can be obtained by making a request to quentin@oma.be.

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