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Summary

Taking into consideration the key role played by the BASCOE assimilation system as a validation product in the preparation phases of the operational Copernicus Atmosphere Monitoring Service (CAMS), this report critically assesses the performance of BASCOE as used in MACC-III. BASCOE is operated by BIRA-IASB and delivers, principally, MLS ozone assimilated abundances with errors of less than 3% in the lower stratosphere and less than 5% at the middle stratosphere in its current 04 version. Such very good achievements and the fact that the present operational C-IFS model uses a parametrization of the stratosphere instead of a real model, gave rise to the ongoing development for the merged version C-IFS-TM5-BASCOE. This fully coupled troposphere-stratosphere model system is expected to be available in the near future.

The performance of BASCOE data assimilation is discussed in this deliverable report by comparing timeseries and vertical profiles of ozone with satellite MLS, OMPS and OSIRIS retrievals as well as total columns maps to other MACC-III products taken from our stratospheric service pages (http://www.copernicus-stratosphere.eu/. The MLS assimilated results from other species: H_2O, HNO_3, HCl, ClO and N_2O are also addressed, in particular in relation to the Antarctic ozone hole event produced every year between July and November.

For this report, we assess that the system has "good performance" if the relative biases of the analyses with respect to the assimilated Aura-MLS observations are smaller than 5%. This is always the case for ozone (O_3) and water vapor (H_2O) between 3hPa and 150hPa. For nitric acid (HNO_3) the performance is not as good, especially in the upper-middle stratosphere (3-10hPa) where the analyses underestimate the observations by 10% in the Tropics and up to 40% in the polar regions. For HCl the system performs well except in South Pole ozone hole conditions, where the analyses can underestimate the observations by up to 50%. For the short-lived species ClO, the assimilation basically failed with large biases in many regions. Finally for N_2O the performance is good except in the upper-middle (3-10Pa) stratospheric polar night where the analyses underestimate the observations by up to 40%.

Contents

1 Introduction

BASCOE (Belgian Assimilation System for Chemical ObsErvations) (Errera et al., 2008; Viscardy et al., 2010; Errera and Ménard, 2012) is a Data Assimilation System principally designed to provide analyses of the chemical composition in the stratospheric layer. As explained in detail by Lefever et al. (2015), BASCOE has been contributing since 2009 to the validation tasks of the MACC projects through comparison with models based on the Integrated Forecasting System (IFS) as well as SACADA (Elbern et al.,2010) and TM3DAM (Eskes et al.,2003; van der A et al.,2010). This MACC stratospheric ozone service (http://www.copernicus-stratosphere.eu) is one of the preparatory activities for the future Copernicus Atmosphere Monitoring Service (CAMS). During these pre-operational phases BASCOE has been contributing with MLS assimilated global analysis of stratospheric ozone abundances to the Near Real Time (NRT) production. Besides, a combination of the TM5 (Huijnen et al., 2010) model (basically a model for tropospheric chemistry) and BASCOE is under development in MACC-III. BASCOE assimilation results are additionally used for comparison with other data products in the WMO Antarctic Ozone Bulletins for Global Atmospheric Watch (GAW) research.

Taking into account the extensive contribution of BASCOE in the mentioned activities, it appears very important to explicitly present in this deliverable a specific validation of ozone assimilation and of other species related to it: H_2O, HNO_3, HCl, ClO and N_2O.

2 Description of the BASCOE DAS version used for MACC-III

2.1 Synthetic description of the system

The most recent description of the system is provided by Lefever et al. (2015). Here we provide a brief overview. BASCOE is a 4D-Var system developed at the Belgian Institute for Space Aeronomy, BIRA-IASB, and based on a stratospheric Chemistry Transport Model (CTM). For the MACC stratospheric ozone service BASCOE assimilates satellite retrievals (version 3.3) of O3, H2O, HNO3, HCl, HOCl, N2O and CO of the Aura-MLS instrument. It must be noted that the Aura-MLS retrievals (i.e. level 2 products) are distributed in two forms:

- the Near-Real Time dataset, which includes only O3, CO, H2O, N2O, HNO3 and SO2 and has a latency of 2-4 hours[1]. This is the dataset chosen for ozone assimilation in the MACC o-suite, due to its short latency. As explained by Lefever et al. (2015, section 6.2), this choice had a large and negative impact on the data quality of the MACC o-suite ozone analyses until 7 January 2013[2] because before this date the MACC o-suite assimilated Aura-MLS NRT v2 ozone data which could not be used below 68hPa.
- The offline dataset which includes all available species and has a typical latency of 3-4 days[3] . This is the dataset chosen for assimilation in BASCOE, due to the added availability of chlorine-containing species, i.e. HCl and ClO.

The assimilation window of BASCOE is 24 h, and it is currently configured to produce gridded output every 3 h. The BASCOE DAS is driven by the ECMWF operational 6-hourly analyses (winds, temperature and surface pressure). Presently BASCOE is run at a horizontal resolution of 3.75 degrees longitude by 2.5 degrees latitude and uses a vertical hybrid-pressure grid comprising 137 levels, most of them in the troposphere. Just like the driving meteorological analyses, this vertical grid extends from 0.01 hPa down to the surface. BASCOE does not include any tropospheric processes and calls its photochemical module only at gridpoints above 400 hPa. It is therefore not expected to produce a realistic chemical composition below the tropopause, resulting in larger systematic error biases for the total columns and in the lower stratosphere.

The CTM includes 57 species that interact using 143 gas-phase reactions, 48 photolysis reactions and 9 heterogeneous reactions. Heterogeneous reactions on the surface of polar stratospheric cloud (PSC) particles are explicitly taken into account. The BASCOE version used here adopts a simple cold-point temperature parameterization to represent the surface area available for these reactions: type I (Nitric Acid Trihydrate). PSCs are set to appear at temperatures between 186 and 194 K with a surface area density of 10^{-7} cm^2 cm^{-3}.

When the BASCOE forward CTM is run with no constraining observations, the stratospheric ozone fields become less realistic after a few weeks or months, depending on the region. These results are similar to those found with IFS-MOZART by Flemming et al. (2011). In the case of the BASCOE CTM, this is due to the absence of tropospheric processes and surface emissions, which prevents proper exchanges with the troposphere; and to the parameterisation of PSC surface area density, which lacks any memory of the coldness experienced by polar air masses. This last issue was discussed by Lindenmaier et al. (2011) using the coupled model GEM-BACH that inherited its photochemistry and PSC parameterisation from BASCOE.

The accompanying deliverable D_G-RG_20.2 (Summary report on progress and issues in GRG BASCOE data assimilation) explains in detail the current set-up for filtering the input MLS data, and its consequences.

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[1] http://mls.jpl.nasa.gov/data/nrt.php
[2] Entry 20130107 in http://www.copernicus-atmosphere.eu/about/project_structure/global/g_idas/g_idas_2/log_fnyp/ entry o
[3] http://mls.jpl.nasa.gov/cal/opsCalDisplay_v3.php#2015

2.2 Technical specifications

BASCOE is written in fortran 2003. The source code described here (version 04.03) includes ~50,000 lines and can be compiled only with intel compiler (ifort) version 11; compilation with gfortran should require only slight modifications of the Makefile. This software requires the chemical pre-processor KPP (http://people.cs.vt.edu/~asandu/Software/Kpp/) and the following libraries:

- HDF 4 to read/write the data files (http://www.hdfgroup.org/)
- Shtools, a package to manipulate spherical harmonics (http://shtools.ipgp.fr/)
- LAPACK, a linear algebra package (http://www.netlib.org/lapack/). We currently use the version optimized for the intel compiler (https://software.intel.com/en-us/intel-mkl/)

The code is parallelized using OpenMP. It is run daily on 24 Itanium cores (1.66GHz). The assimilation of one day of obervations, on the grid described above, takes ~10 hours of wallclock time and uses ~20 Gb of RAM.

2.3 Changelog of the system run in NRT for MACC-II and MACC-III

BASCOE delivers analyses with a latency of 3 to 4 days, i.e. the latency time of the Aura-MLS offline dataset plus the 10 hours necessary for processing. We are now running BASCOE version 04.03, a major upgrade introduced on 2014-02-01. Here is the changelog of all version changes since version q02.05, i.e. the version described in detail by Lefever et al. (2015) and used between 2009-07-01 and 2012-10-20:

2012-10-20 Assimilated dataset switches from Aura-MLS offline v2.2 to v3.3. See the EOS MLS Data Documentation (http://mls.jpl.nasa.gov/data/datadocs.php)

2013-01-01 Major upgrade: switching from version q02.05 to 03.08.

- The previous operational version (q02.05) suffered from aliasing errors in the input wind fields, leading to erroneous noise in the horizontal distribution of chemical tracers (e.g. ozone at 50 hPa). The new version generates input wind fields from the operational analyses of vorticity and divergence, retrieved in spectral space at the correct truncation (T31).
- This new pre-processing of input wind fields is based on the algorithm of Segers (Bregman et al., 2003) : the horizontal fluxes are adjusted to ensure mass conservation despite the temporal variations of surface pressure in the meteorological analyses.
- The vertical grid of the model is improved and extended, from 37 levels (model top at 0.1 hPa) to 91 levels (model top at 0.01 hPa). This new vertical grid comes from the IFS configuration used operationally for meteorological deterministic forecasts (since 1 February 2006). It has a much finer resolution in the Upper Troposphere-Lower Stratosphere (UTLS) region.
- The new version nudges tropospheric ozone towards a climatology based on ozone soundings for the period 1966-1996 (Logan, JGR, 1999). Note that ozone at tropospheric levels should still not be used: its distribution is irrealistic due to the absence of any surface emissions, tropospheric processes and NMVOC species. Nudging to a climatology is only meant to improve the quality of total ozone columns.
- Photochemical reaction rates were updated from JPL-2006 to JPL-2009, except for the photolysis cross-section of Cl2O2 which was updated to JPL-2011. This last change has an important impact on chlorine radicals (ClOx, not assimilated) in ozone hole conditions (Papanastasiou et al., 2009).
- Format standardization for the dataset distributed in NRT: the BASCOE analyses were written in HDF-4 format using ad-hoc conventions. The new version writes the output in netCDF-3 format compliant with CF-1.0 conventions.

2013-01-12 Version 03.09 : H2O replaced by ECMWF meteo analysis between surface and 2 km above tropopause.

2013-06-25 Version 03.11 : Vertical grid upgraded from 91 to 137 hybrid-pressure levels.

2013-08-19 ClO is included in the list of species actively assimilated by BASCOE. ClO is part of the Aura-MLS offline dataset v3.3 but was pure model output until now

2014-02-01 Major upgrade: switching from version 03.11 to 04.03.

- All previous operational versions used a diagonal Background Error Covariance Matrix (BECM). Version 4 uses a non-diagonal BECM where spatial correlations are based on an (isotropic) spectral representation as described in Errera and Ménard (2012).
- In the current configuration, these BECM spatial correlations are set up using a gaussian model, both in the horizontal (correlation length scale=600km) and in the vertical (correlation length scale=1 model level).
- The processing of the following species has changed:
    - Ozone: the Aura-MLS observations above 3 hPa are now rejected. This allows a much better assimilation of the other species (especially H2O and HCl) in the upper stratosphere. The only drawback w.r.t. ozone analysis is seen between 3hPa and 1hPa above the winter pole, where the bias (underestimation by 24-h forecast against assimilated observations) increases from ~5% to ~15%.
    - CO: this species was not usable in any previous operational version as its only source in the middle atmosphere, i.e. the photolysis of CO2, had been forgotten from the chemical scheme. This has been corrected and since CO is retrieved from Aura-MLS, it has been added to the list of assimilated species. Caution: this product is very preliminary and in evaluation phase; it is based on the assimilation of Aura-MLS retrievals of CO which require caution themselves (according to the MLS Version 3 data quality document).
- The results for all other species are similar or slightly improved. Slightly better results were noted for ozone in the tropical UTLS and in the polar lower stratosphere (thanks to the non-diagonal BECM) and for H2O, HCl in the upper stratosphere (thanks to the blacklisting of ozone observations above 3 hPa).

3 System performance for production of ozone analyses

Nearly 90% of the atmospheric ozone mass resides between 10 and 50 km at the stratosphere playing a beneficial role for life by absorbing ultraviolet radiation. The distribution of stratospheric ozone is basically regulated by the Chapman cycle (Chapman, 1930) involving photolysis and reactions with odd and molecular oxygen to create and destruct ozone. In addition, ozone is also removed by catalytic cycles with the NOx, HOx, or ClOx families composed of highly reactive species as well as to the effect of other ozone depleting substances (ODS): the anthropogenic chlorofluorocarbons (CFC's), methane, water vapor, nitrous oxide or still stratospheric aerosol particles that contribute to the ozone depletion particularly at the south pole.

In **Figure 1** the timeseries of relative differences between BASCOE and MLS are plotted for the middle stratosphere (layers 3-10hPa, top) and the low stratosphere (layers 30-70hPa, bottom) at the polar and tropical regions for the year 2014. Everywhere the biases do not exceed 5% showing a global underestimation of satellite values. Indeed, in the middle stratosphere the errors are very small at the poles (generally less than 2%), with the exception of the Antarctic ozone hole season where we obtain a maximum bias peak of -4.5% in September. At the tropics the underestimation increases up to 3%. The biases are still smaller in the low stratosphere of around -0.5% everywhere and again a peak of -3% during the Antarctic ozone hole season. The reason of such slight different biases for such season could perhaps be explained by a deficient abundance evaluation of ozone depletion substances assimilated or not by BASCOE.

Further comparisons are displayed in **Figure 2** for a year ranging from December 2013 to December 2014. In this case time series from daily means of the MACC models and BASCOE are contrasted with MLS, OMPS and OSIRIS observations at two specific altitudes (20 and 33 Km). It must be noticed that while all observational instruments are also sampled to daily means, the models outputs were not interpolated to the location of the observations, allowing only a crude comparison between models and observations. Some general comments can be established from the present plots concerning the satellite data. Firstly, at the lower stratosphere (20 Km) OMPS yields lower observed ozone abundances than MLS up to about 30% at all latitude bands. We can identify such an underestimation for OSIRIS data in the Tropics. Secondly, at the middle stratosphere (33 Km) where the ozone maximum is observed by MLS, the other instruments seem to be in global agreement at the Poles with MLS data, but a general overestimation of about 1 ppm in average all over the period is obtained at the Tropics. These comparisons show that the analyses may still disagree with independent observations, such as OSIRIS and OMPS. The latter instrument is expected to become of particular importance for the future validation and operation of Copernicus models.

From the vertical profiles of **Figure 3**, it can be pointed out a very good performance of BASCOE throughout the stratosphere with the smaller biases of at most 3% between 100 and 10 hPa. With respect to the standard deviation of observations (plotted in grey), it is interesting to note that the dispersion is about half a ppmv nearly all along the whole vertical range during October, which indicates a larger incertitude of measures during the ozone hole episode.

An intercomparison of ozone total column maps between MACC_products MACC_osuite, SACADA, TM3DAM and SACADA is displayed in **Figure 4** for December 2014. BASCOE shows features very similar to the other systems.

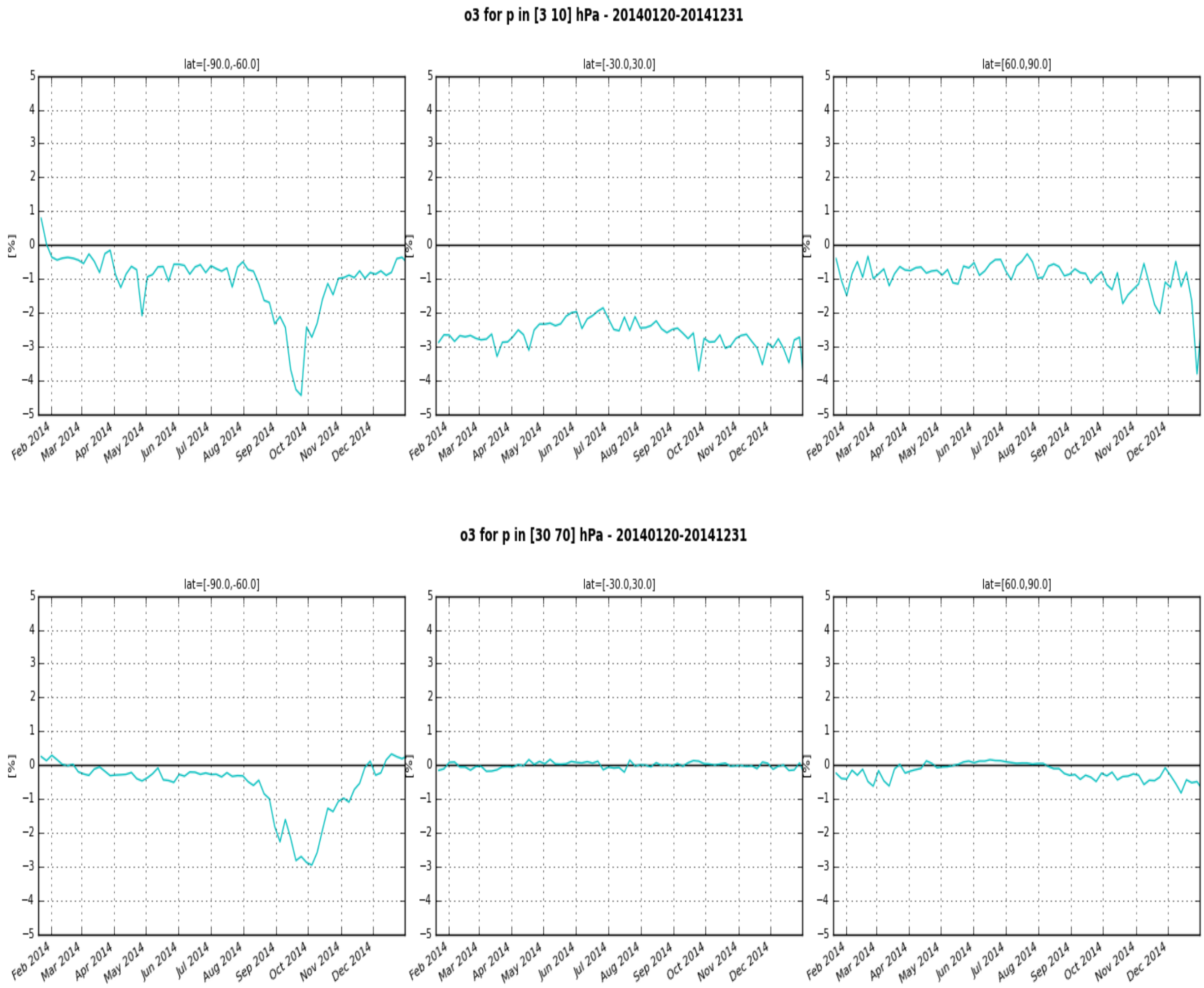

Figure 1: Verification of ozone (O3) analyses by BASCOE: time series of the relative differences between BASCOE and the assimilated data (i.e. MLS) for the layers 3-10hPa (top) and 30-70 hPa (bottom) at the Tropics and the poles for the year 2014.

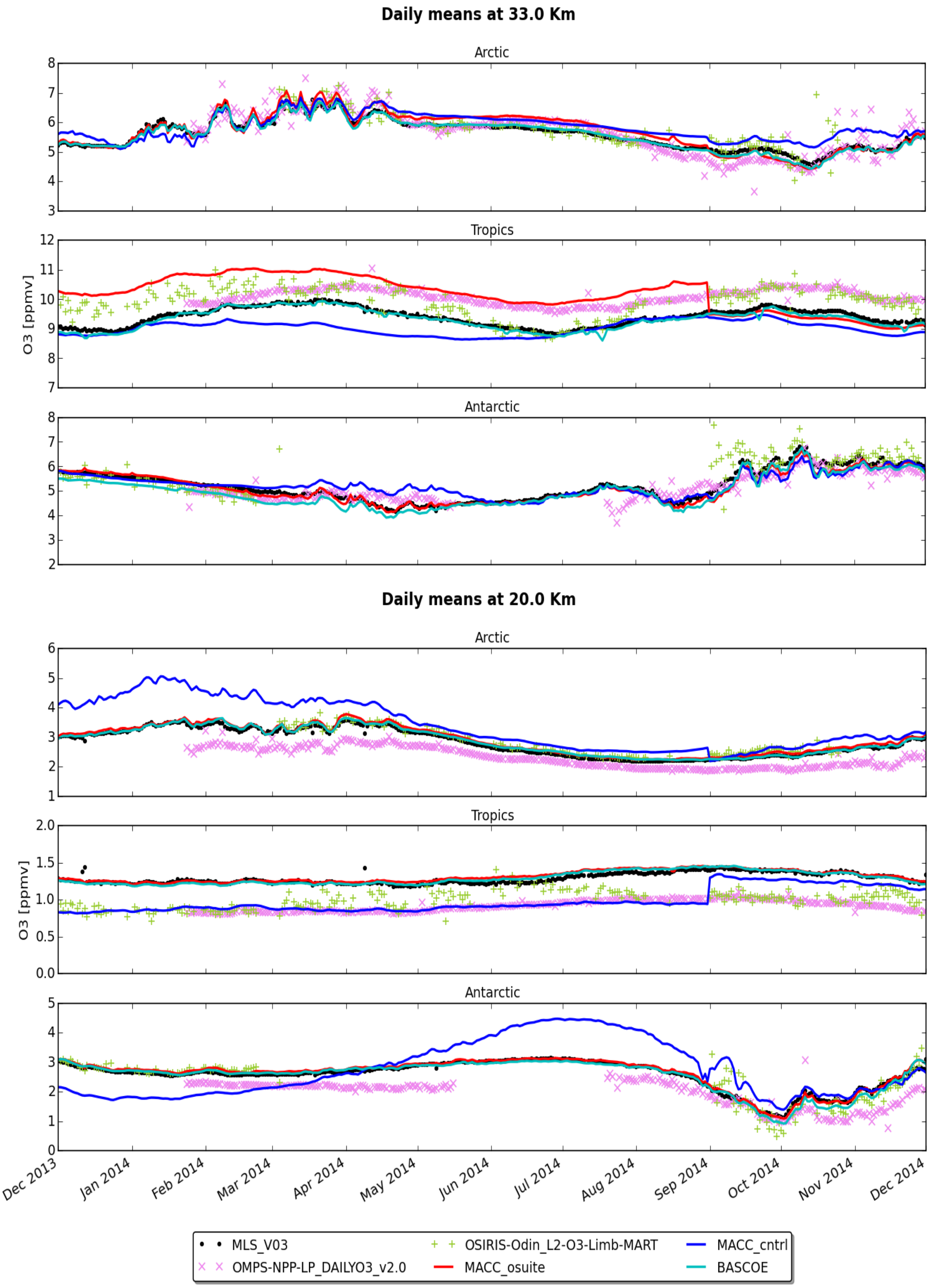

Figure 2: Preliminary evaluation of the ozone analyses delivered by BASCOE and te MACC o-suite: daily mean time series of ozone abundances from MACC_osuite (red line), MACC_cntrl (i.e. with no assimilation of ozone; blue line), and BASCOE (cyan line) with satellite observations by Aura-MLS (i.e. assimilated dataset), and independent OMPS and OSIRIS observations for the period 2013/12/01-2014/12/01 interpolated at altitudes 20 Km (bottom) and 33 Km (top).

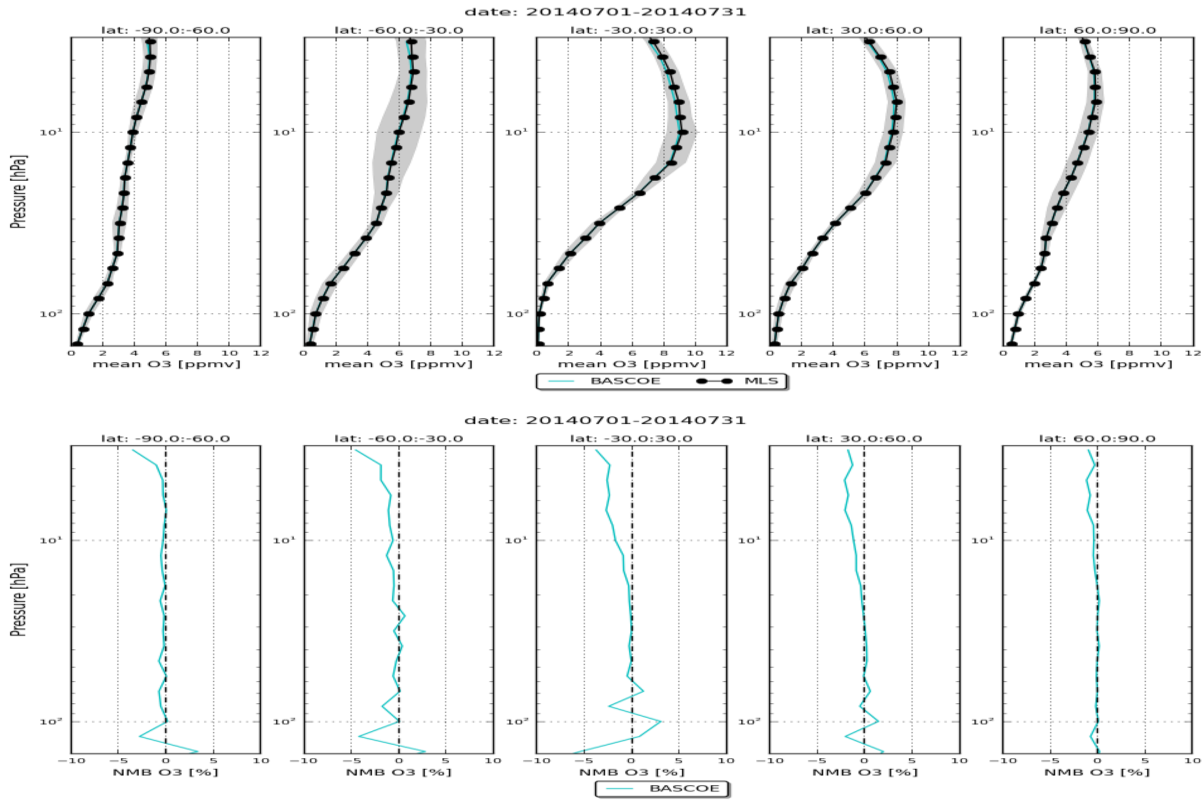

Figure 3: Normalized mean ozone profiles (top) and bias (bottom) of the ozone profile between BASCOE and MLS observations for July 2014.

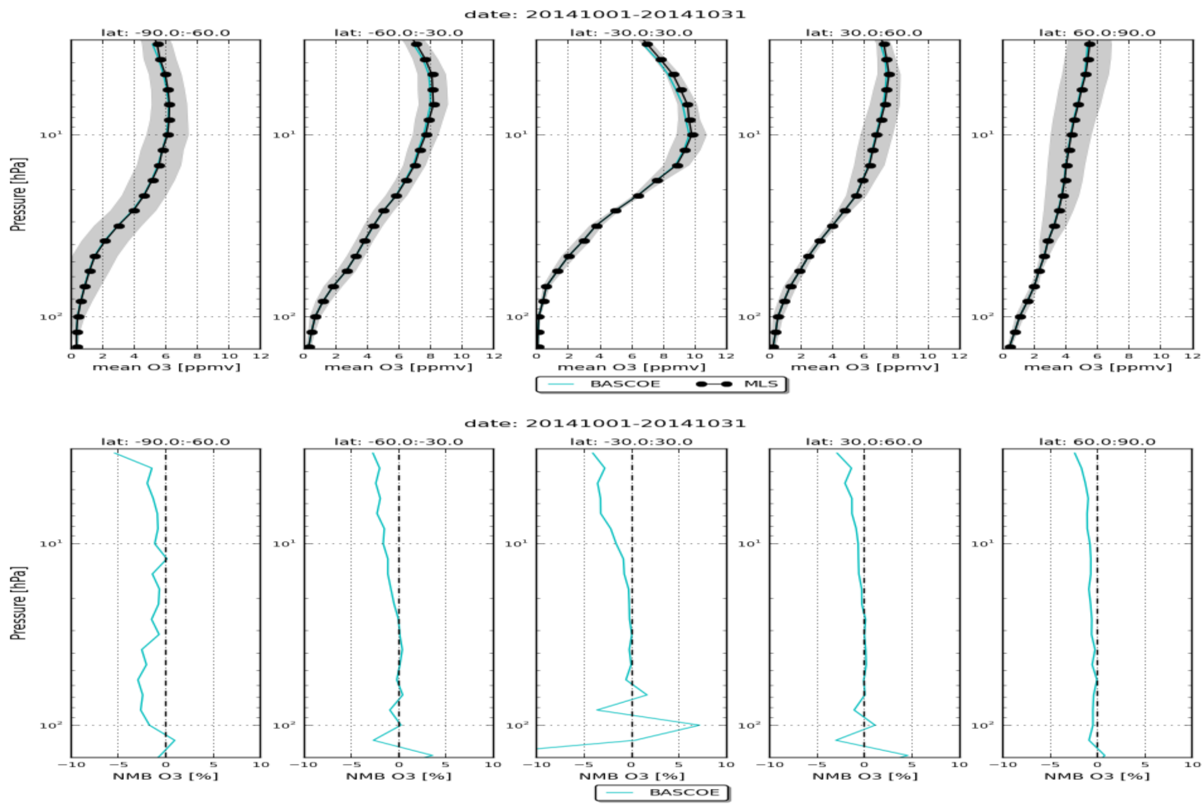

Figure 4: Normalized mean ozone profiles (top) and bias (bottom) of the ozone profile between BASCOE and MLS observations for October 2014.

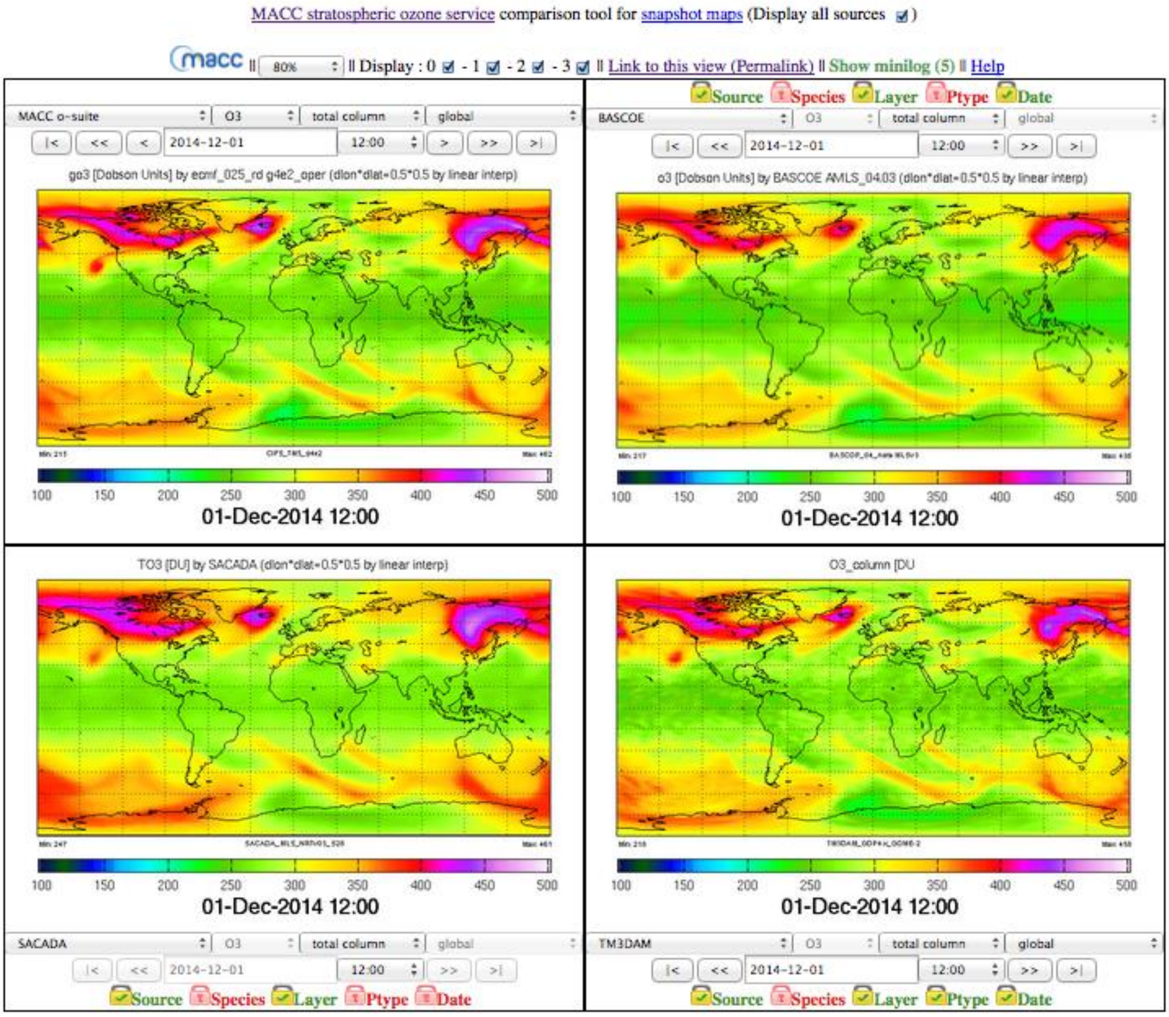

Figure 5: Ozone total columns maps comparison between MACC_osuite, BASCOE, SACADA and TM3DAM from the intercomparison tool of the MACC stratospheric service on December 1st 2014.

4 System performance w.r.t. stratospheric H_2O, HNO_3, HCl, ClO, N_2O

The deep decrease of temperatures to values below -85°C during the austral polar winter induces the formation of big particles of nitrid acid trihydrate (NAT) that contain H2O and HNO3 species and constribute to the ozone depletion during the spring season. Several halogenated species like chlorofluorocarbons, HCl, HBr contributes to the activation of ClO and BrO. For each short-lived molecule of ClO thousands of ozone molecules can be destroyed. In addition, N2O is a very important tracer in the stratosphere and its increasing concentrations can also modify the halogen-ozone and the NOy-ozone chemistry contributing to ozone depletion.

In order to correlate the biases obtained for ozone in the last section - implying particularly the Antarctic ozone hole season- with BASCOE performance for the other species assimilated, the time series during 2014 and vertical profiles of H2O, HNO3, HCl, ClO and N2O for the low stratosphere (30-70 hPa) and the middle stratosphere (3-10hPa) are examined below in **Figures 5 to 14**.

Figures 5 and **6** demonstrate the very good performance of BASCOE assimilation for water vapor. The biases at the lower stratosphere do not exced 2%, with underestimations at the South pole during the ozone hole event and overestimations at the tropics between March and November. Elsewhere the differences are less than 0.5%.

HNO3 assimilation (**Figure 7** and **8**) is slightly worse at the lower stratosphere not exceding 6%, overestimating in winter and underestimating in spring. On the other hand, the perfomance is still worse at the polar middle stratosphere, where the differences can attain -30% in the Antarctica and -45% in the Artics. At tropical regions the biases attain -12%. The dispersion of the observational values is particularly substantial at the South pole and the tropics for July and October with deviations of the order of the mean values.

HCl present very small deviations at the middle stratosphere of at most 1%, but it delivers underestimations up to 30% during the Antarctic ozone hole event and 12% at the North pole during winter (see **Figures 9** and **10**).

ClO exists in low quantities in the stratosphere (see **Figures 11** and **12**). At the low stratosphere There are sudden marked changes in the biases at the poles that turn around -70% during summer an fall and +/-20% in winter and spring. At the middle stratosphere the biases changes are more gradual passing from 5% in average during summer and fall to a maximum of -30% in winter. A regular underestimation of about 60% is observed at the tropical low stratosphere, whereas a slight overestimation of about 5% is obtained for the middle stratosphere.

N2O BASCOE assimilation performance is rather deficient in the middle and upper stratosperic layers of the polar night region, as can be observed in **Figures 13** and **14**. An increasing negative deviation begins in autumn; it passes through a maximum of 40% in winter and decreases to negligible values at the end of the season. At the lower stratosphere an overestimation of about 4% occurs during October and November. Elsewhere the biases are at most of 1%.

In summary, for all the five species investigated and closely related to the Antarctic ozone hole season, BASCOE presents some deficiencies in simulating their abundances. Their assimilations can influence the ozone assimilated results. Both HCl and ClO, for example, display underestimations that rise to 25% during the hole event, but they cannot explain an underestimation of ozone during the hole episode. HNO3, on the other hand, shows an overestimation of around 6% during the time of formation of PSC's, but later it is underestimated during fall, which could has an indirect influence on the ozone deficit. Indeed, HNO3 is a recervoir of NO2, which regulates the ClO-ozone cycle depletion. At the end, all these differences result to be rather small and other species not assimilated like CFC's substances could also play a role.

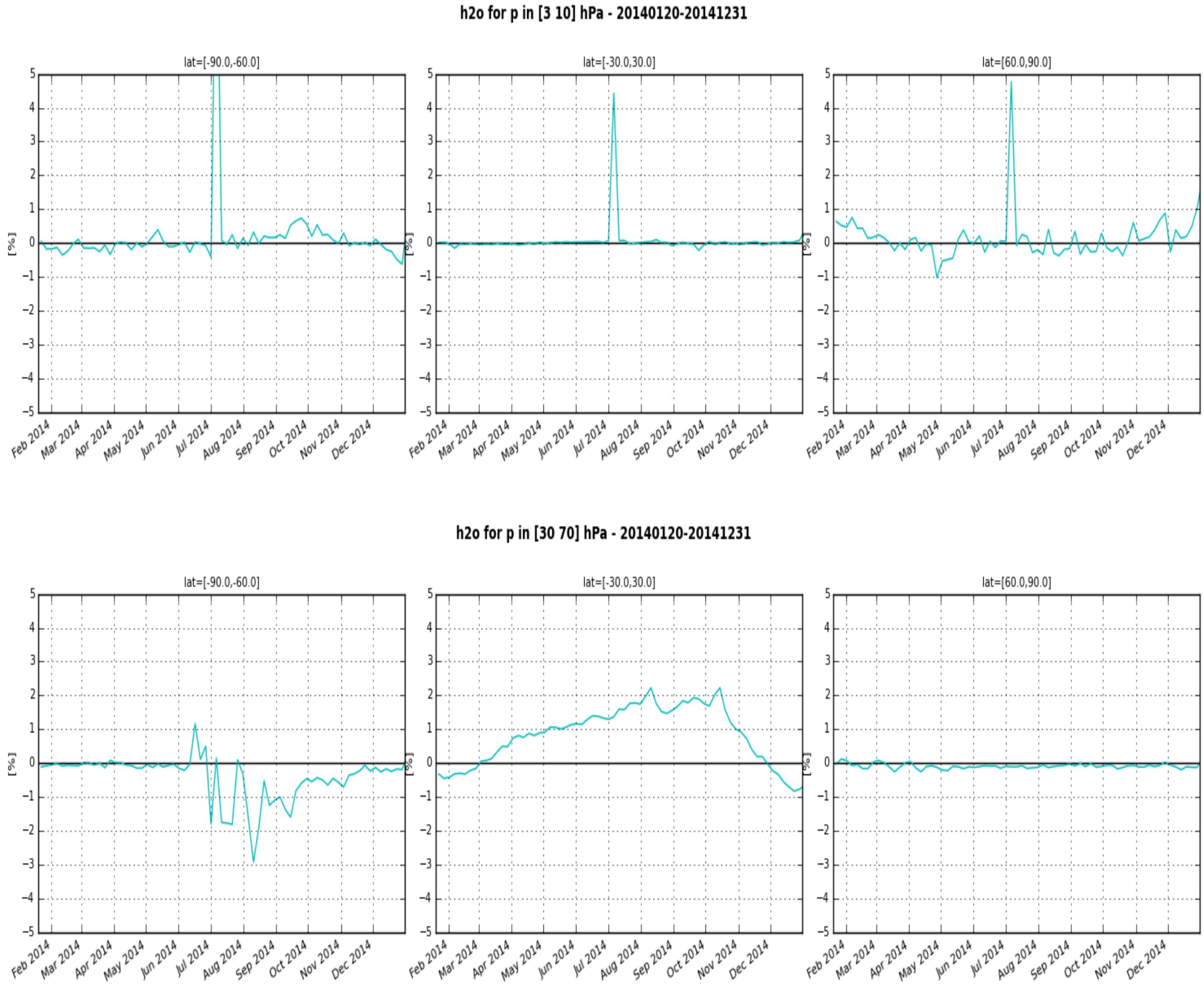

Figure 6: Verification of water vapor (H2O) analyses by BASCOE: time series of the relative differences between BASCOE and the assimilated data (i.e. MLS) for the layers 3-10hPa (top) and 30-70 hPa (bottom) at the Tropics and the poles for the year 2014.

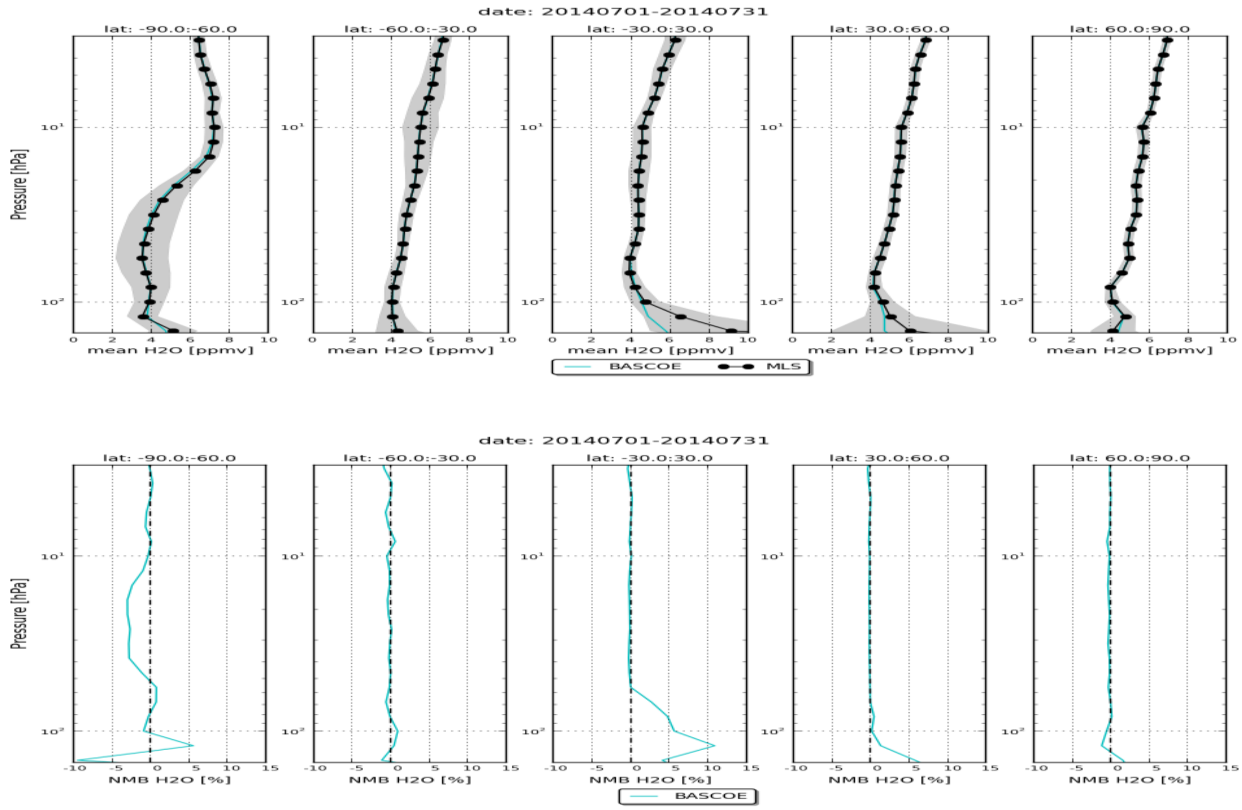

Figure 7: Normalized mean H2O profiles (top) and bias (bottom) between BASCOE and MLS observations for July 2014.

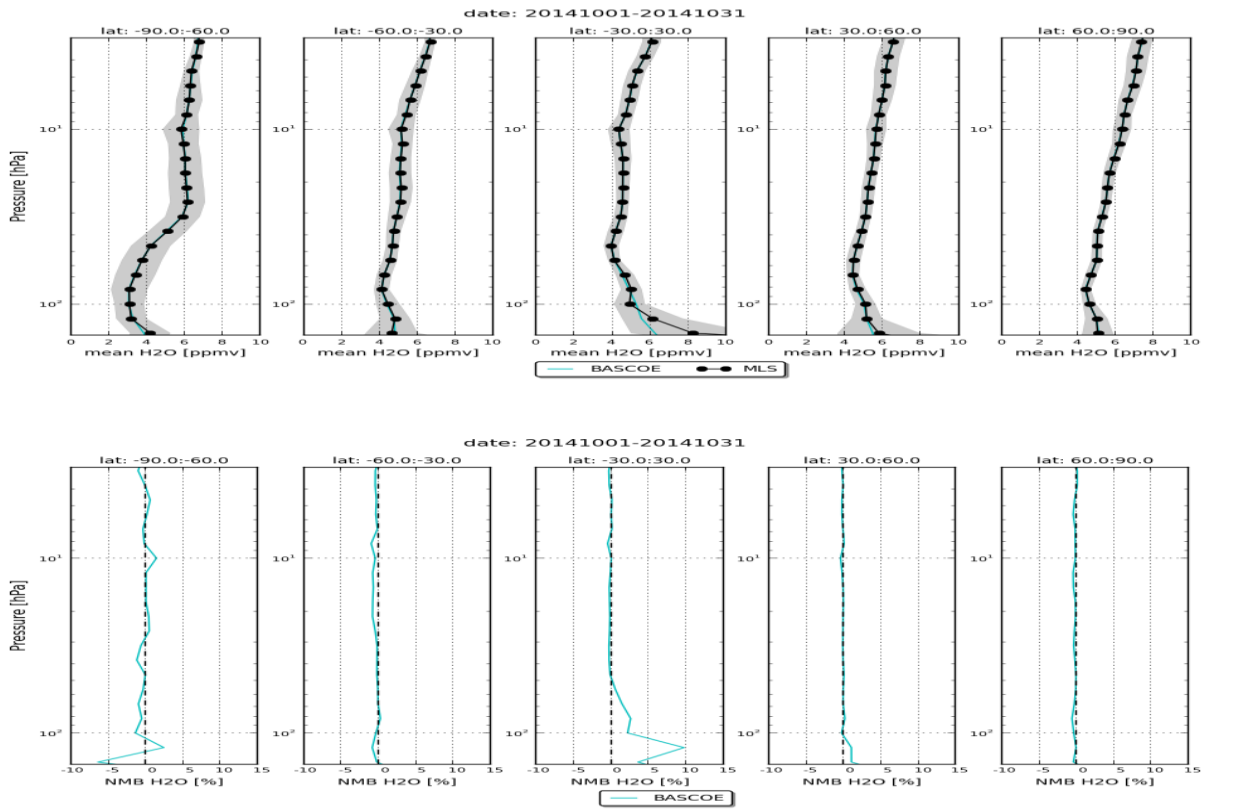

Figure 8: Normalized mean H2O profiles (top) and bias (bottom) between BASCOE and MLS observations for October 2014.

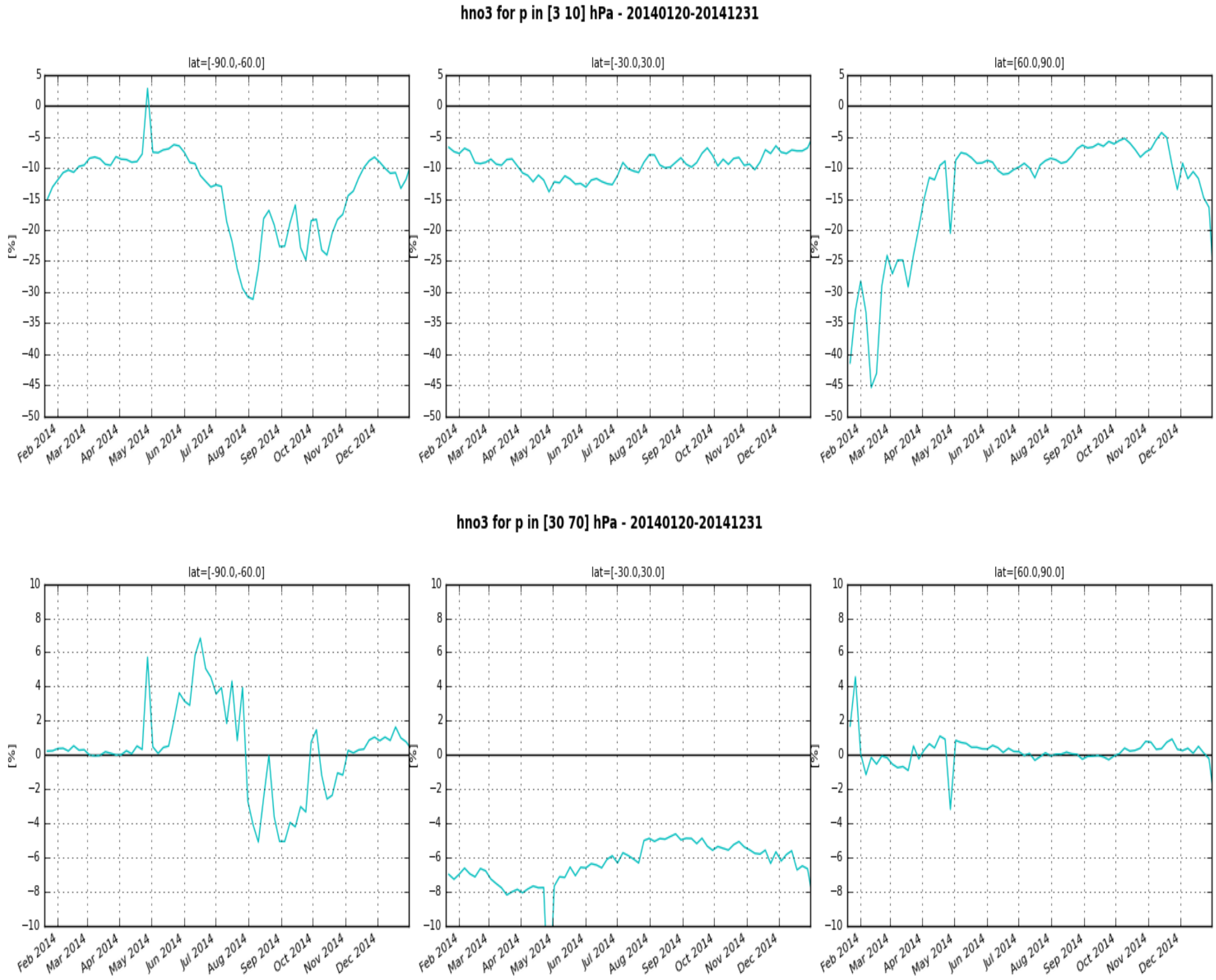

Figure 9: Verification of nitric acid (HNO3) analyses by BASCOE: time series of the relative differences between BASCOE and the assimilated data (i.e. MLS) for the layers 3-10hPa (top) and 30-70 hPa (bottom) at the Tropics and the poles for the year 2014.

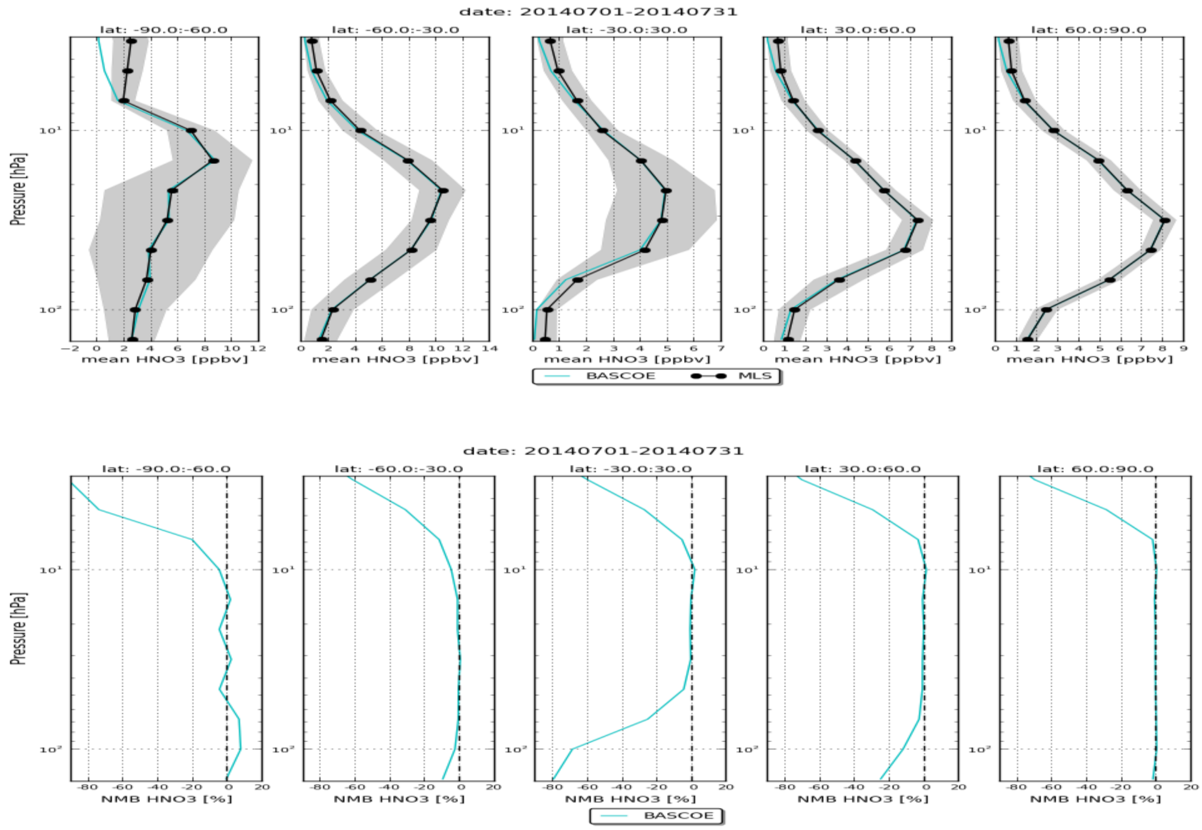

Figure 10: Normalized mean HNO3 profiles (top) and bias (bottom) between BASCOE and MLS observations for July 2014.

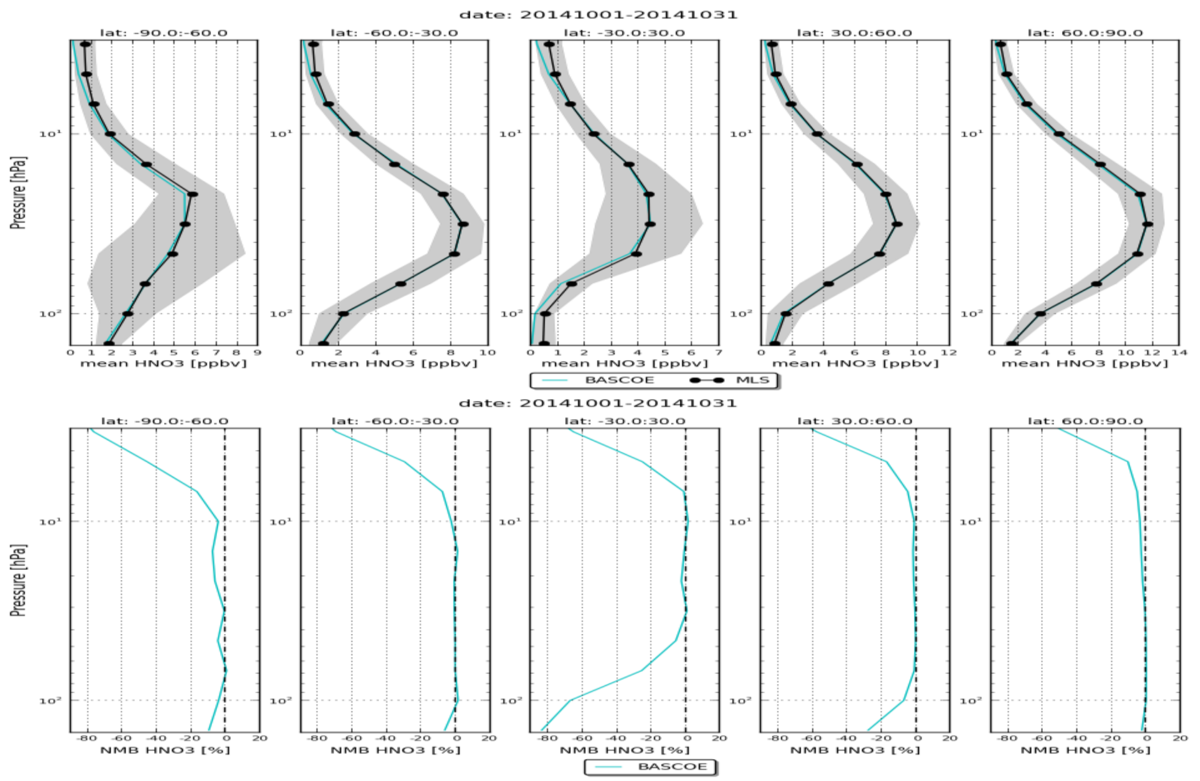

Figure 11: Normalized mean HNO3 profiles (left) and bias (right) of the ozone profile between BASCOE and MLS observations for July (top) and October (bottom) 2014.

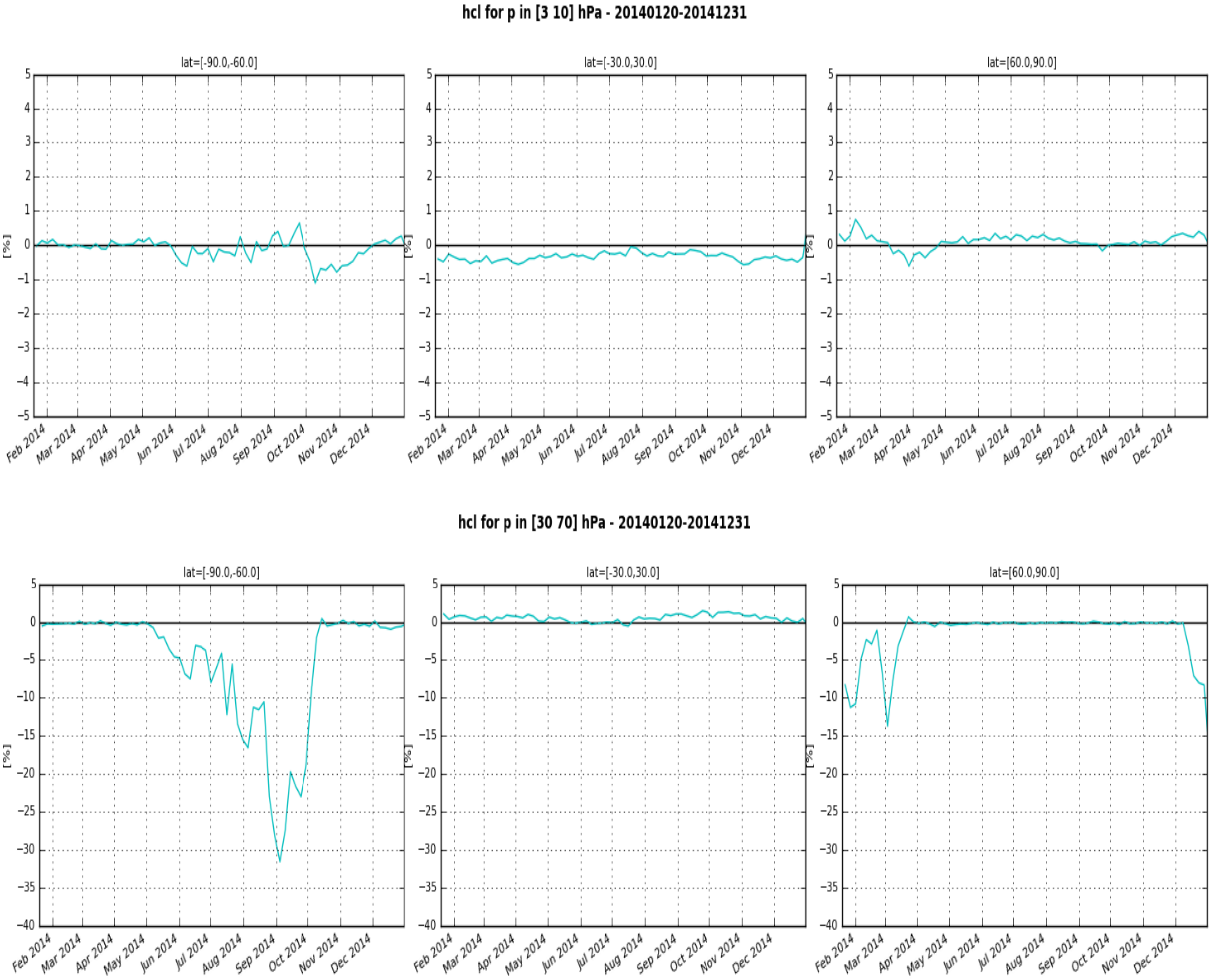

Figure 12: HCl time series of the relative differences between BASCOE and MLS for the layers 3-10hPa (top) and 10-30 hPa (bottom) at the tropics and the poles for the year 2014.

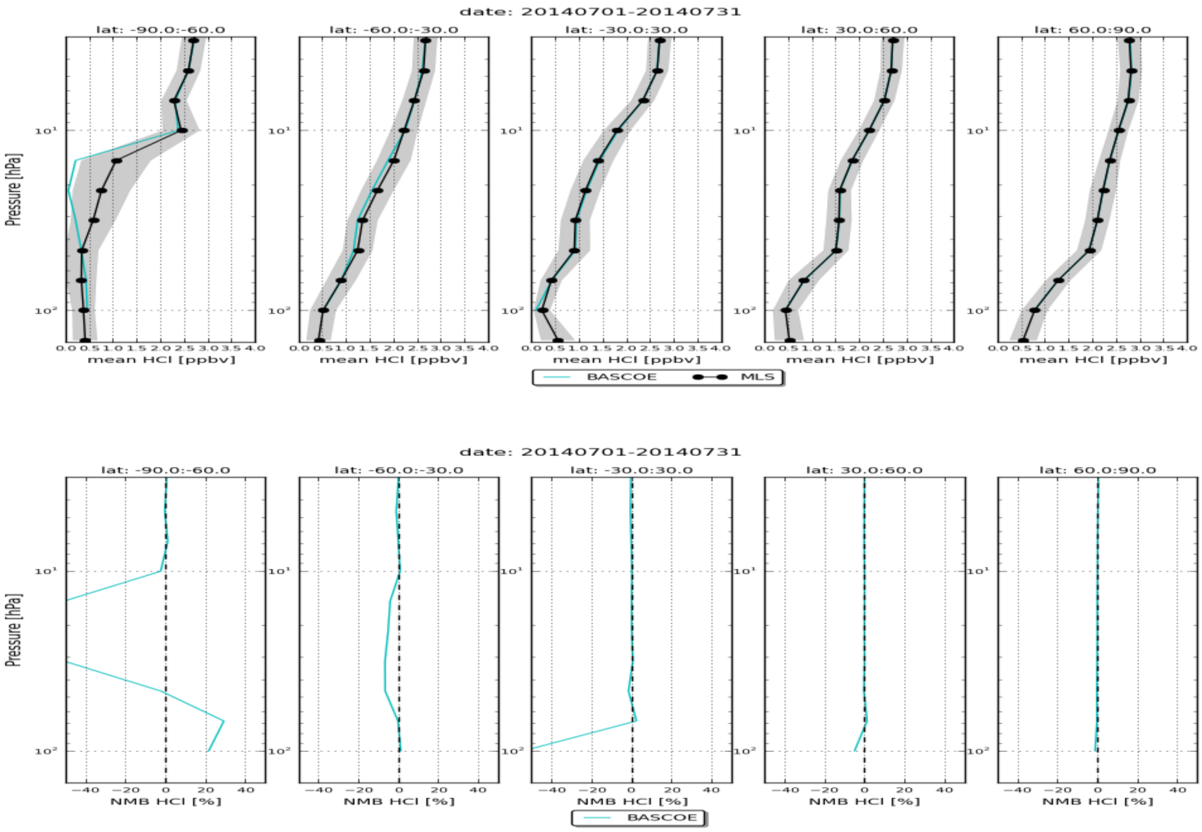

Figure 13: Normalized mean HCl profiles (top) and bias (bottom) between BASCOE and MLS observations for July 2014.

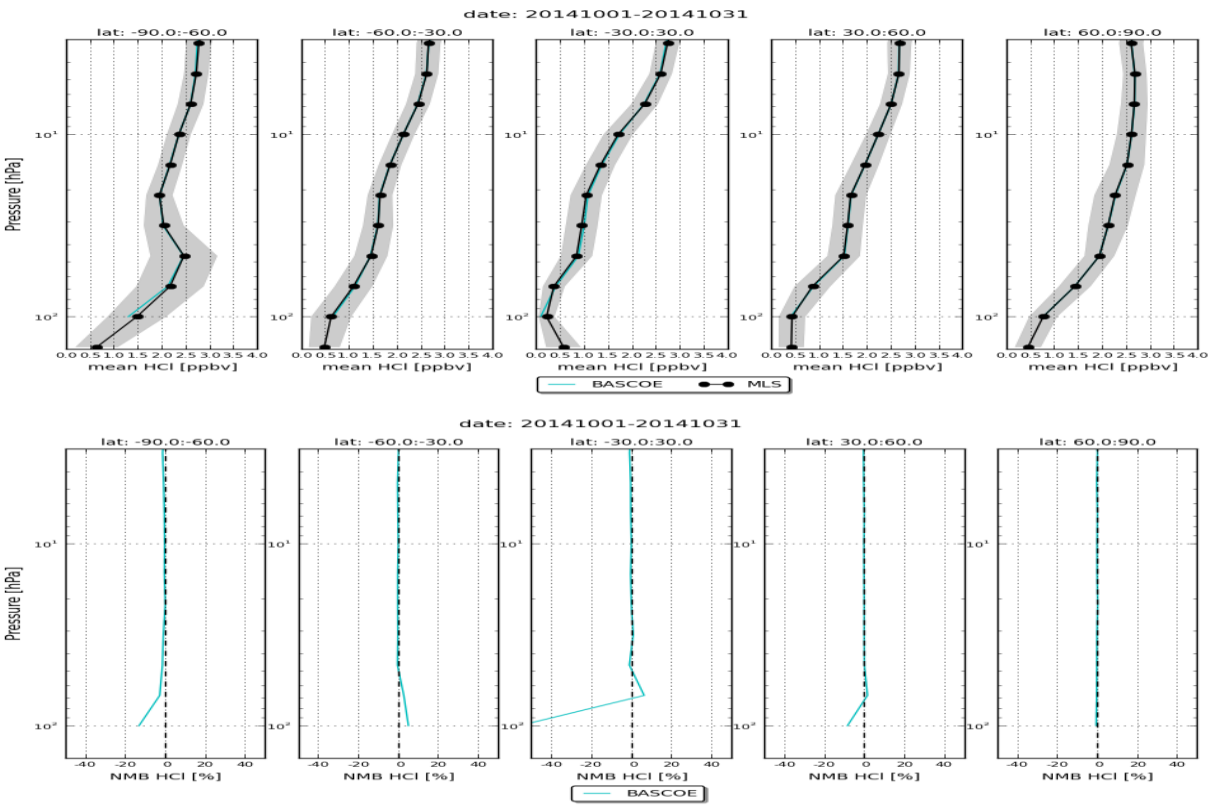

Figure 14: Normalized mean HCl profiles (top) and bias (bottom) of the ozone profile between BASCOE and MLS observations for October 2014.

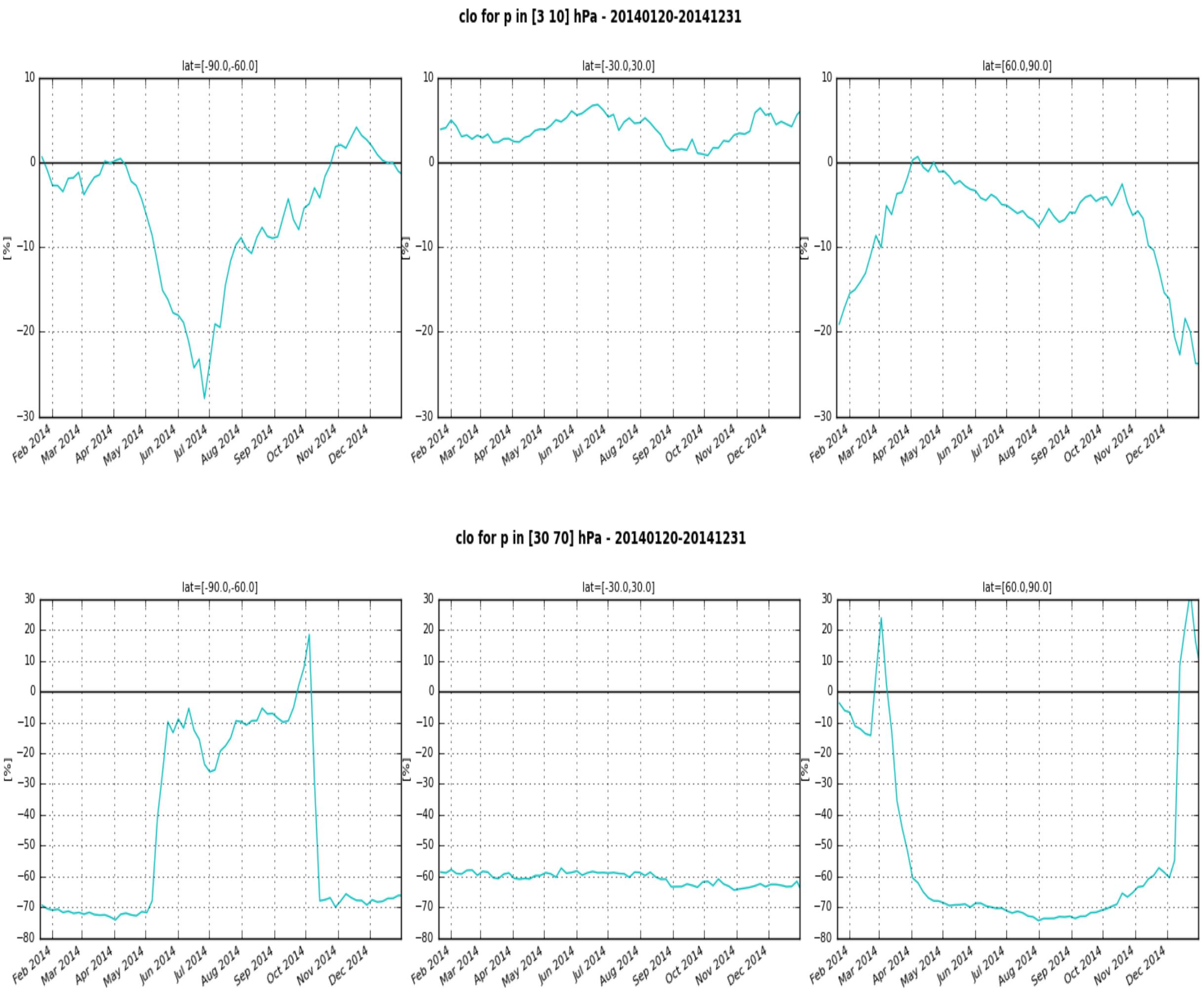

Figure 15: Verification of chlorine monoxide (ClO) analyses by BASCOE: time series of the relative differences between BASCOE and the assimilated data (i.e. MLS) for the layers 3-10hPa (top) and 30-70 hPa (bottom) at the Tropics and the poles for the year 2014.

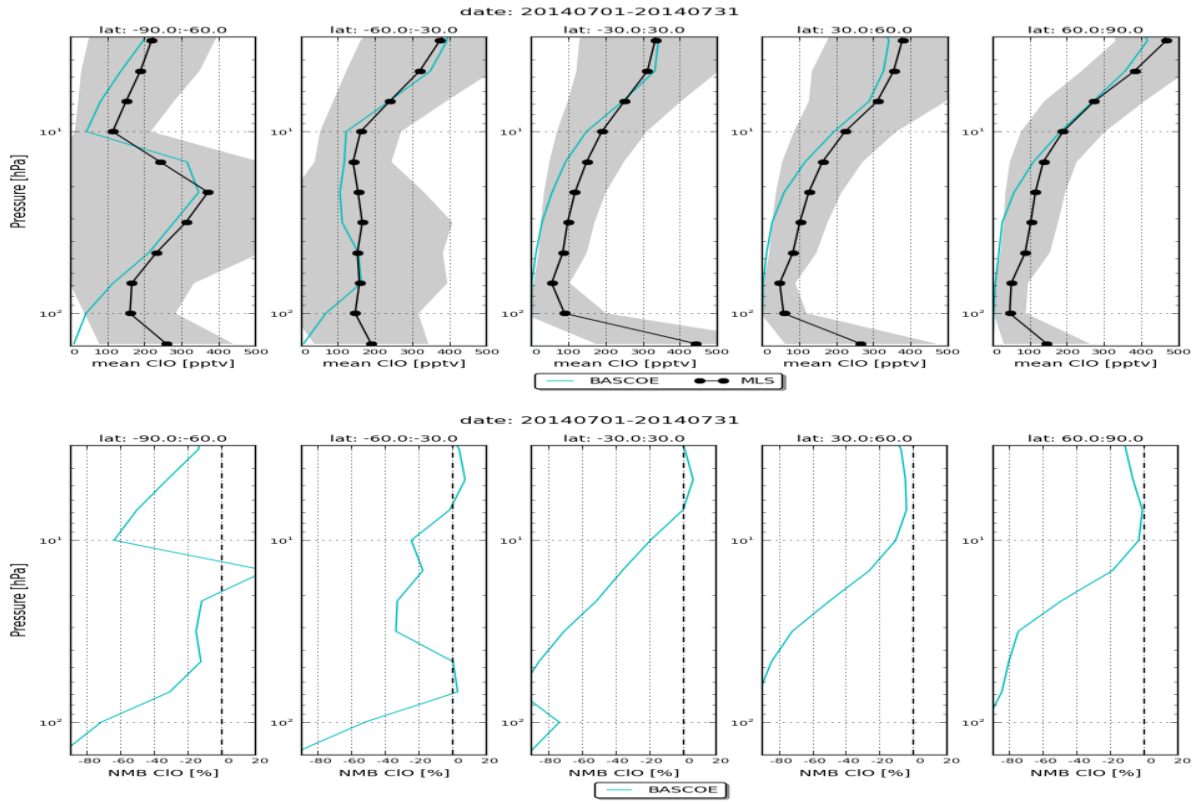

Figure 16: Normalized mean ClO profiles (top) and bias (bottom) between BASCOE and MLS observations for July 2014.

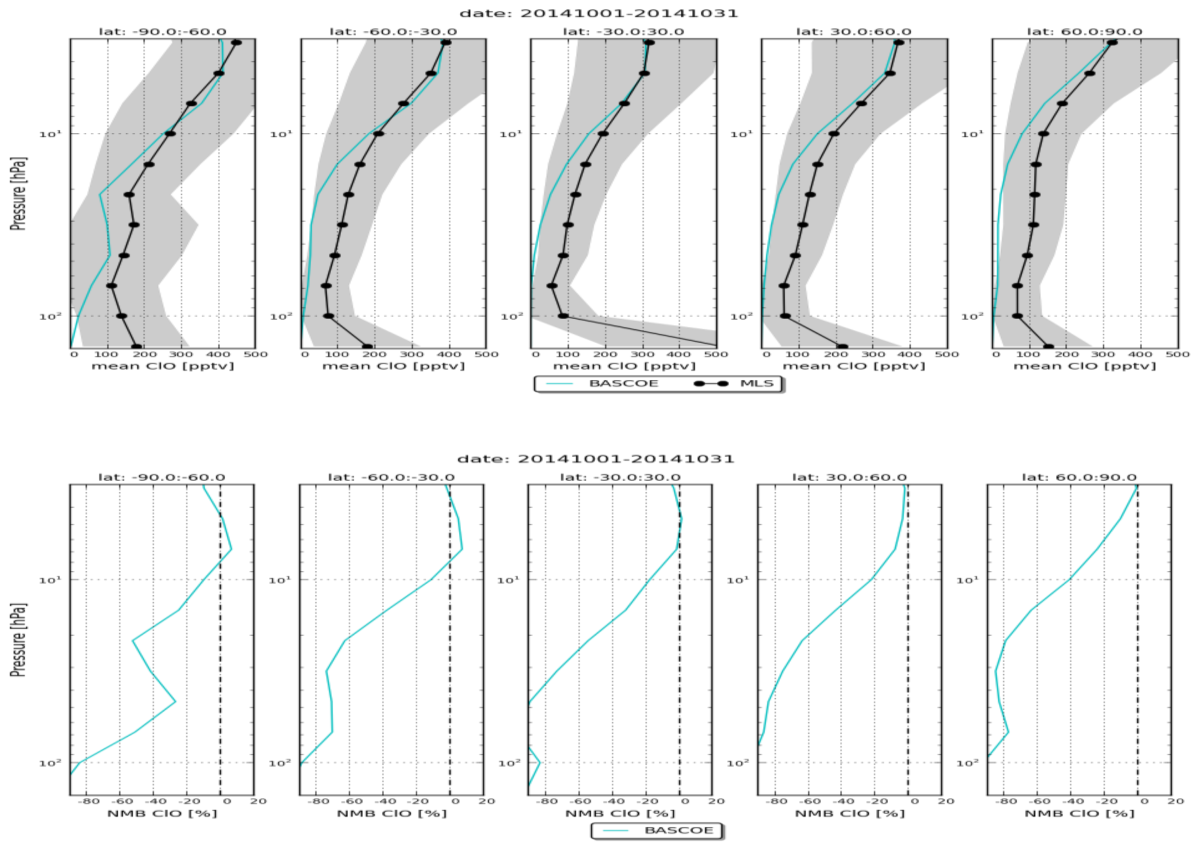

Figure 17: Normalized mean ClO profiles (top) and bias (bottom) between BASCOE and MLS observations for October 2014.

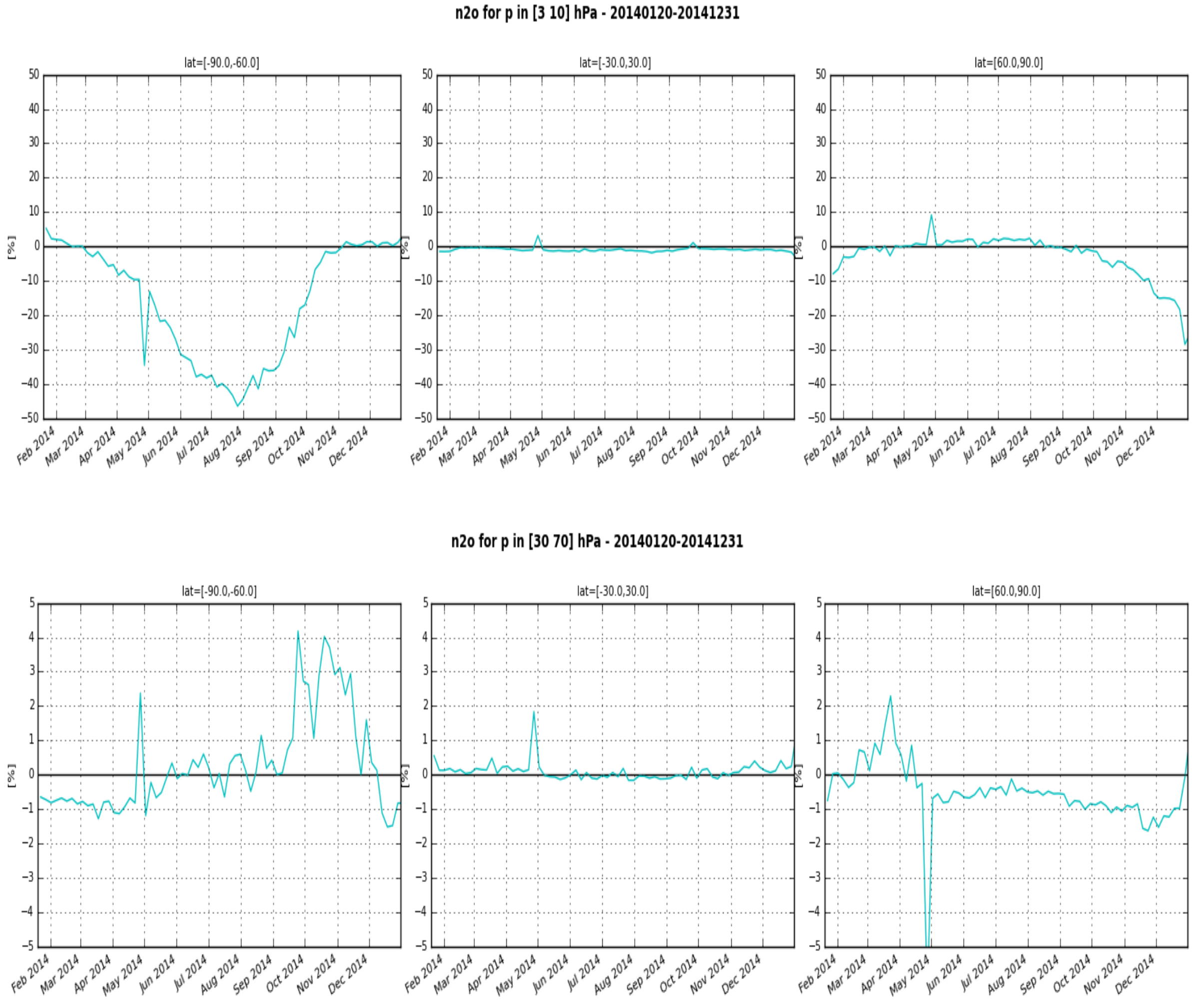

Figure 18: Verification of N2O analyses by BASCOE: time series of the relative differences between BASCOE and the assimilated data (i.e. MLS) for the layers 3-10hPa (top) and 30-70 hPa (bottom) at the Tropics and the poles for the year 2014.

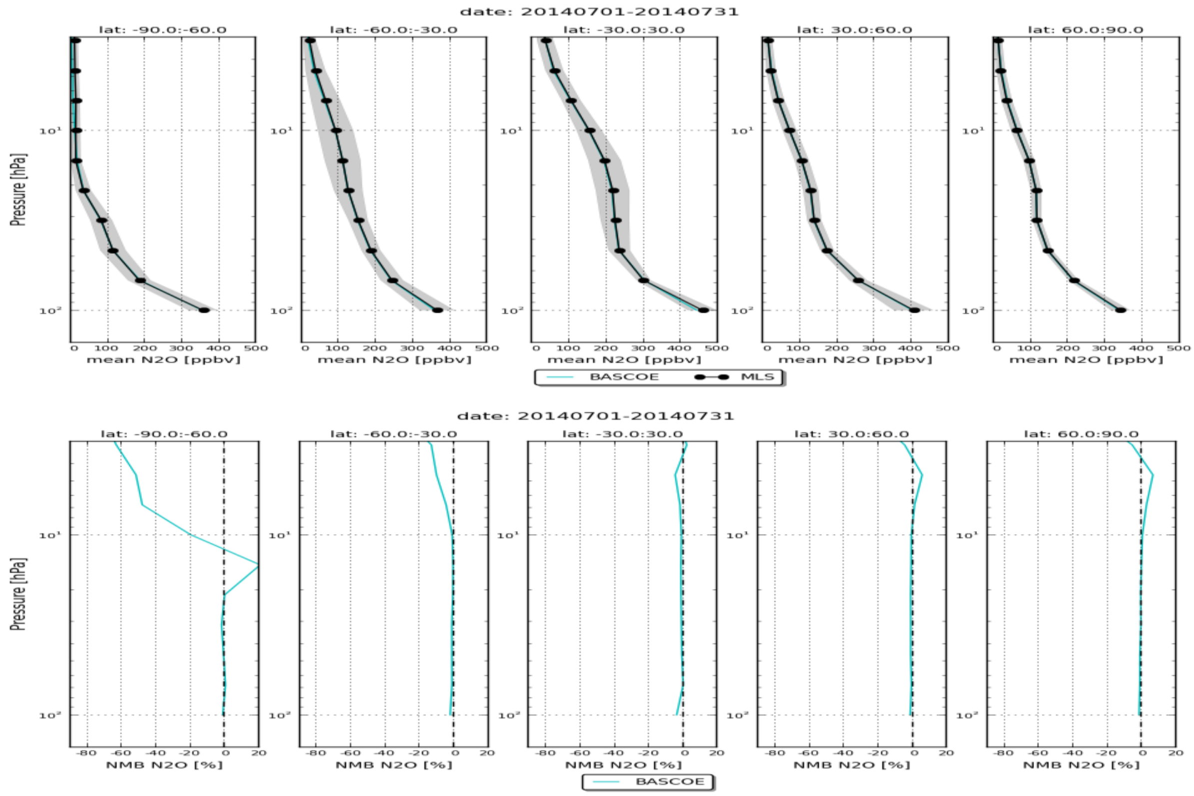

Figure 19: Normalized mean N2O profiles (top) and bias (bottom) between BASCOE and MLS observations for July 2014.

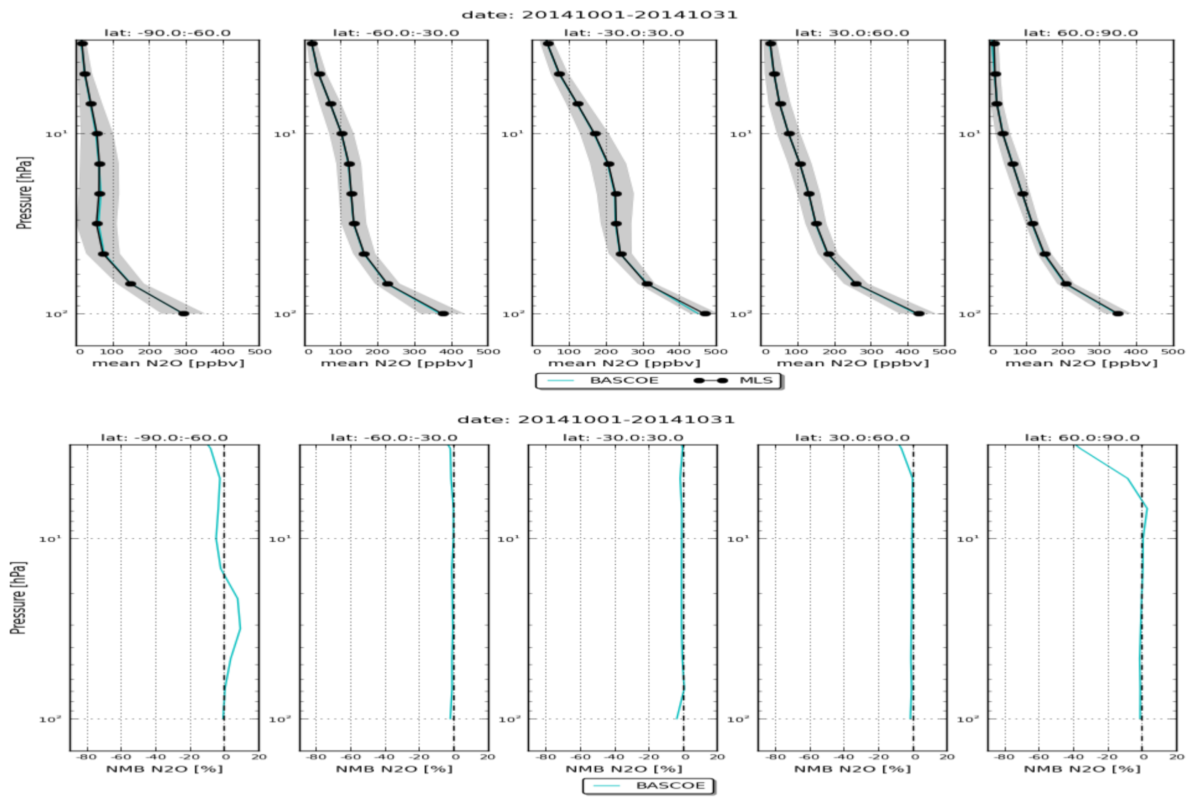

Figure 20: Normalized mean N2O profiles (top) and bias (bottom) between BASCOE and MLS observations for October 2014.

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