# MODELLED OZONE BIAS NEAR THE STRATOPAUSE USING ESA CCI OZONE DATA

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## ABSTRACT

Photochemical models are known to underestimate the ozone in the upper stratosphere and lower mesosphere (USLM), i.e. above 45 km of altitude. In the present study, we evaluate this issue within the state-of-the-art BASCOE model. A reference BASCOE model underestimates the ozone in USLM by 30-50%. First, we discuss the impact of the vertical model grid and the corresponding temperature forcing. Second, we investigate the impact on ozone of the gas-phase chemical reaction rates and photo-dissociation cross-sections of the latest Jet Propulson Laboratory (JPL) recommendations published in 2011. Third, methods of computing the photodissociation rates (J-tables) are evaluated. Fourth, a sensitivity test to the solar irradiance spectrum is performed. Finally, the impact of the temperature field on the modeled ozone is studied. To this end, we compare the temperature field used in our model with temperature profiles provided by limb and occultation satellite data. The results of our experiments are evaluated using the ESA CCI level 2 ozone data as well as MLS, MIPAS and ACE-FTS to document the ozone underestimation issue. As a result, the BASCOE model provides essentially less biased ozone in the USLM. The mean model bias decreases to 0 - 15%.

Key words: stratospheric CTM, ozone, bias, underestimation.

## 1. INTRODUCTION

The problem of the underestimation of ozone in the upper stratosphere lower mesosphere (USLM) by chemistry transport models (CTMs) has been known since many years. For the first time, it was revealed by Prather (1981). And even recently, Siskind et al. (2013) reported an important discrepancy between their modeled ozone and the SABER data. As stated in the article, the topic has been of interest for two reasons. First, an important discrepancy between a model and data has revealed an essential lack of knowledge in our understanding of atmospheric processes. And second, the USLM ozone has been believed to be an indicator of the response of the atmosphere to variable solar ultraviolet radiation.

The present study reports a current state of ozone in the BASCOE (Belgian Assimilation System for Chemical ObsErvations) model. It assess the problem and discuss issues that may help resolving it. We begin with the impact of the vertical model grid. Then, an update of the photochemical scheme used in the model is discussed. The method of computing the photodissociation rates (J-tables) in the model is also evaluated. We also assess an impact of the solar activity on ozone. And finally, the temperature forcing applied to the model is considered.

## 2. OZONE OBSERVATIONS

In this section, we report the ozone measurements provided by MIPAS, ACE-FTS, Aura-MLS and HARMOZ used in this study. The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) (Fischer et al., 2008) has been operating since March 2002 on board of the ENVIronmental SATellite of the European Space Agency (ESA). It is a limb sounder inverting vertical distribution of many key stratospheric species including ozone. Usually there are around 1000 MIPAS profiles per day. The MIPAS ozone data have been validated for scientific applications. Between 1 to 50 hPa, the ozone bias with respect to correlative data is lower than 10%; it increases to 25% at 100 hPa. At levels above 1 hPa, the number of correlative data are too small to derive quantitative conclusions (Cortesi et al., 2007).

The EOS Aura-MLS data version 3.3 (Froidevaux et al., 2008) are also used in the present study. The observations of ozone cover the latitude range between 82 °S and 82 °N with an along-track separation of around 165 km between consecutive scans. Around 3500 vertical scans are performed every day. Ozone profiles have a vertical resolution around 3 km in the stratosphere and they are valid for scientific studies between 265 and 0.02 hPa. The accuracy of MLS ozone measurements, being 7% at 1 hPa pressure level, drops to 35% at 0.02 hPa.

Finally, the HARMonized data of Ozone profiles (HARMOZ) are used in this work. A complete report on these data may be found in (Sofieva et al., 2013). Here, we report the issues that are relevant for the present study only. The HARMOZ data set is based on limb and occultation ENVISAT (MIPAS, GOMOS and SCIAMACHY), Odin (OSIRIS and SMR) and SCISAT (ACE-FTS) satellite

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measurements. These six instruments provide long-term measurements and are involved in the European Space Agency (ESA) Climate Change Initiative (CCI). The vertical coverage of each instrument is different. But it is much larger than the region of our primary interest, from 10 to 0.01 hPa. Each profile has been interpolated onto a common pressure grid with a vertical spacing of 1 km below 20 km and 2-3 km above. All the data sets include temperature profiles at the locations of measurements. MIPAS CCI and ACE-FTS CCI contain retrieved temperatures. Other four data sets are provided with the temperature from ECMWF analyses. GOMOS CCI temperature above 1 hPa is an interpolation of MSIS-E-90 model reanalysis (V. Sofieva, personal communication). Sofieva et al. (2013) show (Fig.6) the altitude-latitude bias with respect to MIPAS. Considering the pressure range of our interest, most important biases are situated in the tropics, starting from -20% for SMR and reaching more than 10% for ACE-FTS and SCIAMACHY.

## 3. TEMPERATURE MEASUREMENTS

The reanalyses of ECMWF are of great importance for the BASCOE CTM because they are used as a model thermodynamical forcing. The model forcing is known to be one of the most important source of errors in the atmospheric models. In this section, we briefly describe the temperature products, observations and analyses, used in this study. We overview the parameters of temperature measurements, mainly the uncertainty of each product.

MLS retrieves atmospheric temperature using limb observations from bands near $O_2$ spectral lines at 118 GHz and 239 GHz that are measured with MLS radiometers R1A/B and R3, respectively. The precision of the temperature measurement is 1 K or better at altitudes below 0.316 hPa, but degrades to $\sim$ 2.2 K at 0.01 hPa (Schwartz et al., 2008). This result was obtained by comparing MLS temperatures with correlative data sets. They showed that the bias in the stratosphere was generally less than 2 K as compared with other observations, at some levels there were persistent MLS temperature biases with $\sim$ 3 K peak-to-peak vertical structure. In the mesosphere MLS temperatures are $\sim$ 0-7 K lower than most other measurements. Hoppel et al. (2008) compared MLS with SABER temperatures prior to assimilation into a global NWP model: The observation minus forecast (OmF) standard deviations for MLS and SABER are $\sim$ 2 K in the mid-stratosphere and increase monotonically to about 6 K in the upper mesosphere. The MLS data quality report provides bias estimations: from 0 to +5 K at 1 hPa, from -7 to -4 K at 0.216 hPa, from -8 to 0 K at 0.1 hPa and from -2 to 0 at 0.01 hPa.

Ridolfi et al. (2007) validated the MIPAS temperature by comparing it with radiosonde, lidar, in-situ and remote sensor measurements operated from the ground or stratospheric balloons. The mean difference between MIPAS and radiosondes is generally less than +2 K but increases monotonically starting from 30 km altitude reaching +6 K at 35 km altitude. Comparing to lidars, the difference is again around +5 K in the USLM.

Sica et al. (2008) validated ACE-FTS temperatures using ground-based and space-borne measurements. The agreement of ACE-FTS temperatures with other sensors is typically better than +2 K in the stratosphere and upper troposphere and +5 K in the lower mesosphere. There is evidence of a systematic high bias (roughly from +3 to +6 K) in the ACE-FTS temperatures in the mesosphere, and a possible systematic low bias (roughly +2 K) in ACE-FTS temperatures near 23 km.

## 4. METEOROLOGICAL ANALYSES OF WINDS AND TEMPERATURE

The BASCOE model is forced in this work by two meteorological products of the European Centre for Medium-Range Weather Forecasts (ECMWF), ERA-Interim and operational analyses (OD). ERA-Interim is a global atmospheric reanalysis (Dee et al., 2011). Gridded data products include a large variety of parameters, including 6-hourly parameters covering the troposphere and stratosphere. ERA-Interim is provided on a 60 level hybrid pressure vertical grid up to 0.1 hPa. A reanalysis deals with a forecast model and a data assimilation system. In the upper-stratosphere, observations are sparse and model errors used in the ERA-Interim model are large. Besides, the model is affected by large warm temperature biases in this region. There are not enough observations to correct this bias, so that the reanalyzed temperature in the upper-stratosphere still has a warm bias from the model (Uppala et al., 2008) that can be roughly estimated as 5-8 K.

Operational global analyses from the Integrated Forecasting System (IFS) of ECMWF provide similar information as ERA-Interim. The OD vertical grid for the year 2008, which is chosen as a period of our experiments, has 91 vertical hybrid pressure levels coming up to 0.01 hPa. A warm temperature bias is also contained in this product.

## 5. BASCOE CTM

The BASCOE CTM model configuration includes 58 chemical species with a full description of stratospheric chemistry (Errera et al., 2008). All species are advected via the Flux-Form Semi-Lagrangian scheme (Lin and Rood, 1996). The CTM is driven by winds and temperatures obtained from the ECMWF ERA-Interim reanalysis as well as ECMWF OD analysis described in the previous section. The horizontal resolution of the model grid is $3.75^\circ$ longitude by $2.5^\circ$ latitude. Vertically, the reference BASCOE model uses a subset of 37 levels of the ERA-Interim 60 levels which excludes most tropospheric levels. The vertical domain extends from 0.1 hPa down to the surface in this case. Hence the model state is described by the vector $x \in R^n$ of length $n = 96 \times 73 \times 37 \approx 2.6 \times 10^5$. When BASCOE is driven by ECMWF OD analyses, the vertical dimension is increased to 91 hybrid pressure levels up to 0.01 hPa down to the surface. Finally, the model time step is set to 30 minutes.

Solar irradiance spectrum used in the reference BASCOE simulation is derived from Lean et al. (1997). It is constant in time solar spectrum corresponding to the minimum of the solar activity. In order to evaluate the sensibility of the BASCOE model to the solar spectrum irradiance, we use daily Solar Radiation and Climate Experiment (SORCE) data. SORCE is a NASA-sponsored satellite mission providing measurements of the incoming X-ray, ultraviolet, visible, near-infrared, and total solar radiation. The SORCE spacecraft was launched on 25 January, 2003. SORCE carries four instruments, including the spectral irradiance monitor (SIM), solar stellar irradiance comparison experiment (SOLSTICE), total irradiance monitor (TIM), and the XUV photometer system (XPS). In the present study, TIM daily data are used in the BASCOE model for the period between January 1 and March 1 2008.

Photolysis rates used in the reference BASCOE simulation correspond to the Jet Propulsion Laboratory (JPL) recomendations published in 2006. We refer this scheme later as JPL2006 as compared to JPL2011, the latest recommendations published in 2011. The impact of JPL2011 chemical scheme on ozone will be evaluated in this work as well. The updated JPL2011 scheme includes modifications in the cross sections, new temperature dependance parameters. Besides, the photochemical reaction on nitrogen oxides $\mathrm{N_2O_5}$ has been modified. The JPL2006 reaction includes only one branch: $\mathrm{N_2O_5} + hv \rightarrow \mathrm{NO_3} + \mathrm{NO_2}$, whereas the JPL2011 provides recommendations for an another additional branch: $\mathrm{N_2O_5} + hv \rightarrow \mathrm{NO_3} + \mathrm{NO} + \mathrm{O}(^3\mathrm{P})$ . The photodissociation rates (J-tables) of the reference BASCOE model are precomputed. On the contrary, evolutive photodissociation rates will be computed in our numerical simulations to assess this issue.

## 6. NUMERICAL EXPERIMENTS

This section describes the strategy of our sensibility experiments in changing model parametrizations. All the experiments described in this section are performed for the same period, form 1 January to 1 March 2008. All experiments use the same initial condition from BASCOE 4D-Var system (Errera et al., 2008). Figure 1 shows the ozone OmF (Observations-minus-Forecast) statistics between MLS, MIPAS, ACE-FTS and HARMOZ ozone measurements and the BASCOE reference experiment for five different latitudal bands covering the globe. The statistics is computed for the whole period of experiments. Generally speaking, all considered data confirm the presence of an important model ozone bias. SMR CCI datasets matches better the BASCOE reference state than all others data. However, it shows an important discrepancy with the BASCOE reference model in the Northern hemisphere, where the difference between observations and the model reaches 30%. All other datasets show a similar OmF pattern with respect to BASCOE. The bias starts growing at pressure levels from approximately 3 hPa. It reaches values between 30 and more than 70% from observations. Generally, ACE-FTS and ACE-FTS CCI have the most important difference with respect to the reference BASCOE model.

Our first try to improve the ozone underestimation in the USLM is to run model with a different vertical resolution. To this end, we force the BASCOE model with the ECMWF operational data (OD). These datasets have higher vertical resolution than ERA-Interim, extend the BASCOE lid up to 0.01 hPa and introduce new additional pressure levels between 1 and 0.01 hPa. However, using OD instead of ERA-Interim forcing does not lead to any significant improvement in the ozone underestimation problem. Hence in later experiments, the ERA-Interim forcing will be used as this configuration is less computationally expensive.

Next experiment concerns the comparison of gas-phase chemical reaction rates and photo-dissociation cross-sections, compiled by the Jet Propulsion Laboratory (JPL) in 2006 and 2011. The update to the JPL2011 does not improve the ozone underestimation in the USLM. On the contrary, the latest scheme results in ozone biases that are 2-3% bigger than in case of the JPL2006 scheme (the statistics is not shown here). Thus, we will keep the JPL2006 scheme in later experiments.

The results of all following experiments are summarized on Fig. 2. The figure shows the ozone OmF mean biases between the MLS data and BASCOE experiments for five different latitudal bands covering the globe. The statistics is computed for the period between January 1 and March 1 2008. The reference experiment, referred as EXP1, represents the BASCOE model with ERA-Interim thermodynamic forcing, on a 37 hybrid pressure level grid (a subset from the 60 ERA-Interim vertical level grid), the JPL2006 chemical scheme and precomputed photodissociation rates (J-tables) that we refer as J-offline. EXP1 is shown on the figure with blue curves. The EXP2, shown by magenta curves, is carried out to compare J-offline with J-online, where the photodissociation rates are computed every time step. Generally, the evolutive photodissociation rates J-online allow one to decrease the ozone bias in the USLM by 5 - 17%. The improvements in the Northern hemisphere are bigger than in the Southern hemisphere during the winter. The most important improvement of 17% is observed in the Arctic region. Vertically, the improvements are observed at pressure levels above 10 hPa. And the maximal differences between J-offline and J-online are situated around 1 hPa. All following experiments will use J-online.

Afterwards, EXP3, in green, shows an impact of the SORCE solar irradiance daily spectra. Using these data decreases biases in every latitude band at pressure levels above 10 hPa. The decrease of bias is generally 3 -5%.

And finally, we aim to assess is the ozone sensibility to the temperature acting in the model. As explained in the Sect. 4, the accuracy of ECMWF products deteriorates in the USLM. The main obstacles here are the presence of a warm temperature bias in the ECMWF model and a lack of available radiosondes observations to constrain the model. The aim of this test is to try to correct the temperature field in the USLM using available limb observations. The ACE-FTS CCI and MIPAS CCI temperatures originate from the retrievals of ACE-FTS and MIPAS measurements, respectively. The auxilliary temperature field in SMR CCI, OSIRIS CCI, GOMOS CCI and SCIAMACHY CCI datasets contain ECMWF analysis temperatures. Hence, only the temperatures from MLS, MIPAS and ACE-FTS will be considered as three independent sources of information in the present work. Figure 3 shows the global mean difference between ERA-Interim and MIPAS, MLS and ACE-FTS temperatures. All three experiments confirm the presence of the ERA-Interim warm bias in the upper-stratosphere. The only positive difference of roughly 1K is observed between ERA-Interim and MLS at 1 hPa. However, the MLS quality paper reports a warm bias from 0 to +5 K at this pressure level.

Let us now suppose that the mean temperature bias may be computed as a mean temperature profile in the observation space. The computation should account for the uncertainties provided by each instrument and the estimations of ERA-Interim temperature uncertainties, which are generally within 5-8 K range. To this end, we apply the least squares approach. Figure 4 shows the computed profile of temperature bias (blue) and a polynomial (red) approximating the temperature bias correction to all model pressure levels.

EXP4, in red, on Fig2 shows the result of such temperature bias correction. The impact of temperature field on ozone is the most important among all issues discussed in this paper. The ozone bias decreases to 0-10% with respect to MLS data in all latitude bands and at all pressure levels. The difference between EXP4 and EXP3 reaches values of 20% and more at 1 hPa. Figure 5 shows a final state of the model which combines all corrections. Generally, the initial bias is essentially decreased. The initial bias of 30-50% is decreased to 0-15%.

## 7. CONCLUSIONS

The present study discussed the problem of the model ozone underestimation in the USLM within BASCOE CTM. The reference state of our model showed typically biases of order 30-50%. We updated the chemistry scheme according to the latest JPL recommendations published in 2011. A different vertical model resolution has also been tested. However, these issues do not have any essential impact on the ozone underestimation problem. On the contrary, we managed to decrease the initial bias to 0-15% by correcting the forcing temperature bias, applying an evolutive method of computing

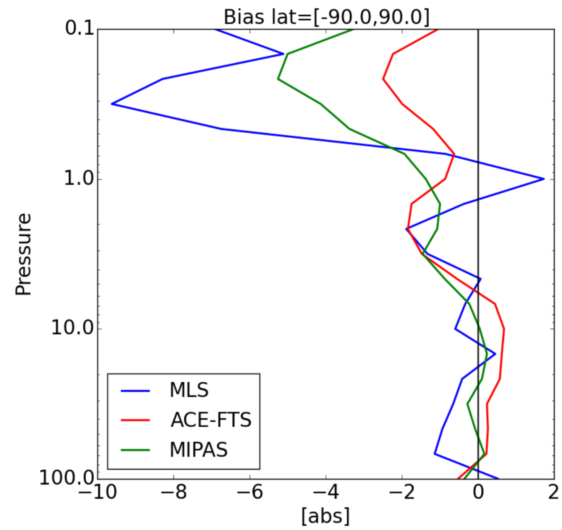


Figure 3: Two-month global mean difference, in K, between the ERA-Interim temperature and MIPAS, MLS and ACE-FTS temperatures. The statistics is computed for the period between 1 January and 1 March 2008.

the photodissociation rates and introducing the SORCE daily solar irradiance spectra to the model. The temperature bias correction plays a dominant role in decreasing the ozone bias. The perspective of this work could be a closer look at the different temperature data available in the USLM to find out how the temperature bias could be eliminated from atmospheric models and reanalyses. Methods to compute the mean temperature bias may also be investigated in more details.


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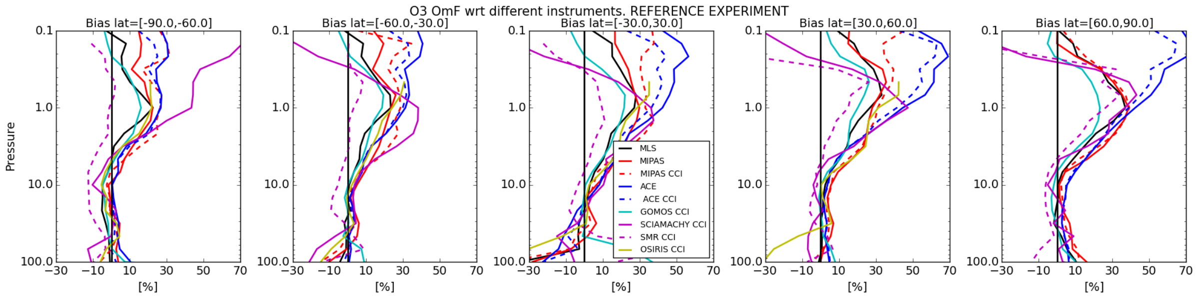


Figure 1: OmF (Observations-minus-Forecast) bias with respect to reference BASCOE simulation for all considered observations (MIPAS, MLS, ACE-FTS and HARMOZ). The statistics is computed for the period between 1 January and 1 March 2008.

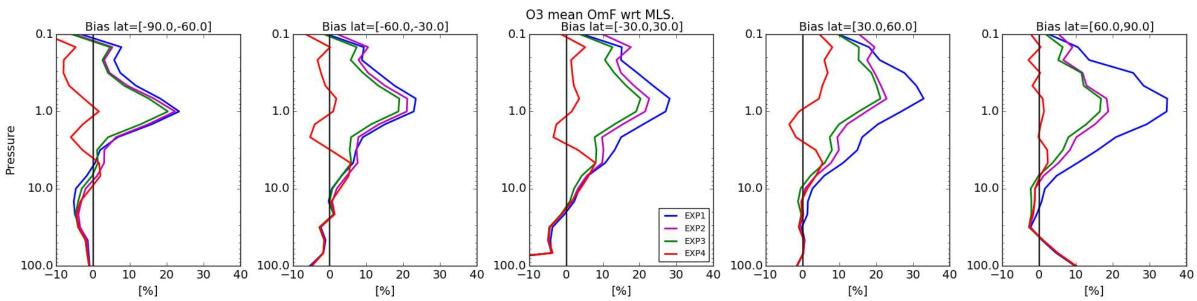


Figure 2: OmF (Observations-minus-Forecast) bias between MLS data and BASCOE experiments. EXP1 (blue): reference model (Joffline, solar irradiance spectrum at minimum of solar activity, ERA-I temperature forcing); EXP2 (magenta): as EXP1 but using Jonline; EXP3 (green): as EXP2 but using SORCE; EXP4 (red): final experiment, as EXP3 but with corrected temperature bias. The statistics is computed for the period between 1 January and 1 March 2008.

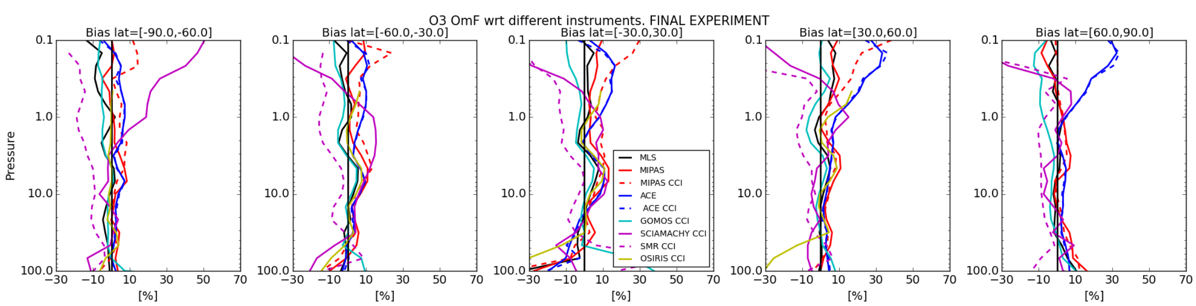


Figure 5: OmF bias with respect to final corrected BASCOE simulation for all considered observations (MIPAS, MLS, ACE-FTS and HARMOZ). The statistics is computed for the period between 1 January and 1 March 2008.

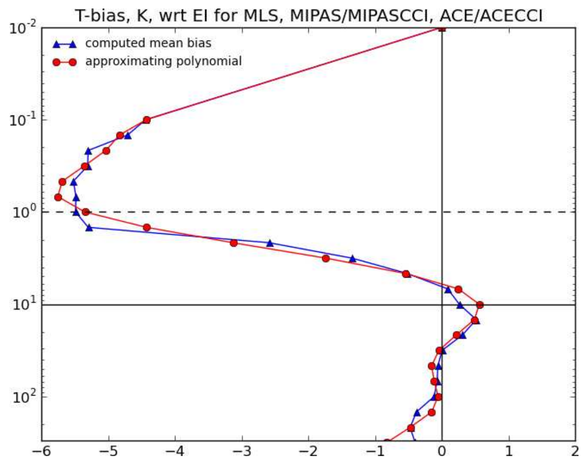


Figure 4: Two-month global mean temperature bias with respect to ERA-Interim temperature. Computed bias (blue) is approximated for all model pressure levels by a polynomial (red). The statistics is computed for the period between 1 January and 1 March 2008.


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