Results of the preparatory study "PREMIER Analysis of Campaign Data"

ELISA CASTELLI[1,*], SAMUELE DEL BIANCO[2], BIANCA M. DINELLI[1], DANIEL GERBER[3], HERMANN OELHAF[4], WOLFGANG WOIWODE[4], BÄRBEL VOGEL[5], BJÖRN-MARTIN SINNHUBER[4], ROLAND RUHNKE[4], UGO CORTESI[2]

[1]Istituto di Scienze dell'Atmosfera e del Clima del Consiglio Nazionale delle Ricerche (ISAC-CNR), Bologna, Italy,

[2]Istituto di Fisica Applicata "N. Carrara" del Consiglio Nazionale delle Ricerche (IFAC-CNR), Firenze, Italy,

[3]Rutherford Appleton Laboratory (STFC-RAL), Chilton, UK,

[4]Institute for Meteorology and Climate Research (IMK-ASF), Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany,

[5]Institute for Energy and Climate Research - Stratosphere (IEK-7), Forschungszentrum Jülich GmbH (FZJ), Jülich, Germany

e.castelli@isac.cnr.it

I. INTRODUCTION

The relevance of the UTLS (Upper Troposphere and Lower Stratosphere) region and the impact of limb emission measurements at millimetre and sub-millimetre wavelengths for investigation of chemical, dynamical and radiative processes occurring at these altitudes constitute a major focus of the atmospheric research activities supported by the European Space Agency (ESA) in the last two decades. Since the end of the 1990's, several studies were conducted on this topic, like those related to the design of the SOPRANO (Submillimetre-wave Observations of PRocesses in the Atmosphere Noteworthy for Ozone) and MASTER (Millimetre-wave Acquisitions for Stratosphere/Troposphere Exchange) instruments [Bühler, 1999], the latter originally conceived for the ESA Explorer mission ACECHEM (Atmospheric Composition Explorer for CHEM-

* Corresponding author: Elisa Castelli, Istituto di Scienze dell'Atmosfera e del Clima (ISAC-CNR), Bologna, Italy, email: e.castelli@isac.cnr.it

istry and climate interaction). Further evolution of the scientific objectives and of the measurement concept originally proposed for ACECHEM lead to the selection of the PREMIER (Process Exploration through Measurements of Infrared and millimetre-wave Emitted Radiation) mission as one of the three candidate core missions of ESA Earth Explorer 7.

Within the scientific context of PREMIER, first priority is assigned to the currently open issues on the sensitivity of surface climate to UTLS variability and to global circulation, and on the troposphere/stratosphere exchange processes. PREMIER will address these topics using atmospheric composition data obtained in 3D by an infrared limb-imaging spectrometer and by a mm-wave limb sounder [ESA, 2012] with unprecedented spatial and temporal resolution. As part of the PREMIER preparatory studies, the PACD project (PREMIER Analysis of Campaign Data) aimed at the analysis of the measurements acquired by the MARSCHALS (Millimetre wave Airborne Receivers for Spectroscopic CHaracterization in Atmospheric Limb Sounding) instrument, an airborne precursor of the millimetre-wave spectrometer of the PREMIER payload, during the field campaigns PREMIER-Ex (Kiruna, Sweden, March 2010) and ESSenCe (Kiruna, Sweden, December 2011) with the M-55 Geophysica high altitude research aircraft. The scope of the project was to demonstrate the capabilities of MARSCHALS to measure the vertical distribution of atmospheric targets (namely temperature, H_2O, O_3, HNO_3, N_2O, and CO) by the inverse processing of its limb sequences. Moreover, the study included the evaluation of the potential synergy between limb measurements in the infrared and in the millimetre-wave region and the comparison of MARSCHALS retrieval products with the state of the atmosphere simulated using two atmospheric chemical transport models: CLaMS (Chemical Lagrangian Model of the Stratosphere) (e. g., [Grooß et al., 2005], [Konopka et al., 2010], and references therein) and EMAC (ECHAM/MESSy Atmospheric Chemistry) [Jöckel et al., 2006].

In this paper, we provide an overview of the activities covered by the PACD project and present the consolidated results for the analysis of MARSCHALS measurements, for the comparison with models and for the synergy with infrared measurements, the latter obtained exploiting the measurements of MIPAS-STR, an infrared interferometer on board the same aircraft. Full details on the results from investigation of the individual aspects addressed by the PACD project are reported in dedicated papers, already published [Castelli et al., 2013] or currently in preparation.

II. THE MARSCHALS INSTRUMENT

MARSCHALS is a millimetre-wave heterodyne spectrometer that operates on board the M-55 Geophysica as a demonstrator of the millimetre-wave limb sounder for the proposed Earth Explorer 7 Mission PREMIER. It acquires atmospheric emission spectra over three spectral bands around 300 GHz (Band B), 325 GHz (Band C) and 345 GHz respectively (Band D). Limb scans consecutively iterate over single spectral bands. A description of the spectrometer can be found in Oldfield et al., [2001] and in Dinelli et al., [2009], while Castelli et al., [2013] report on the instrument upgrades performed before the PREMIER-Ex and the ESSenCe campaigns.

The MARC code (Millimetre-wave Atmospheric Retrieval Code, [Carli et al., 2007]) was designed for the analysis of MARSCHALS measurements. The code can retrieve vertically distributed quantities such as Temperature, Volume Mixing Ratios (VMRs) and external continuum (a retrievable profile used to account for continuum effects from cloud opaque in the millimetre-wave frequency range, [Dinelli et al., 2009], [Castelli et al., 2013]), as well as scalar quantities like frequency shift, pointing bias and radiative gain and offset. All spectra acquired during a limb scan are analyzed simultaneously. The following list denotes all the different species measured by MARSCHALS, grouped together in corresponding frequency bands. Band B: O_3 and N_2O; Band C: Temperature, H_2O, O_3 and HNO_3; Band D: HNO_3, O_3 and CO. Band B contains some information on HNO_3, but it is not sufficient to produce significant results. The external continuum, which is retrieved for every scan, provides information on the impact of clouds on MARSCHALS measurements. MARSCHALS has been deployed on board the M-55 Geophysica in several measurement campaigns: SCOUT-O3 (2005) in the tropics [Dinelli et al., 2009]; PREMIER-Ex Test Campaign (2009) at mid-latitudes; PREMIER-Ex Science Campaign (2010) and ESSenCe Campaign (2011), both at polar latitudes. In the following sections we describe the results obtained by MARSCHALS during the PREMIER-Ex and ESSenCe Science campaigns.

III. Measurement campaigns results

III.I PREMIER-Ex campaign: Flight on 10 March 2010 from Kiruna (Sweden)

The PREMIER-Ex campaign took place on the 10 March 2010 from Kiruna (Sweden) and aimed at the quantification of horizontal mixing processes in the UTLS. The flight track followed a triangular pattern, aiming toward the West and then turning back to Kiruna. During the flight MARSCHALS sampled air masses of different origin (polar vortex as well as mid-latitude and mixed air masses). From MARSCHALS spectra we retrieved VMR profiles of temperature, H_2O, O_3, HNO_3, N_2O, CO. In particular, the HNO_3 retrieved profiles along the flight show that the MARSCHALS instrument was able to resolve regions of vortex and filament of vortex air. The results obtained during this campaign have been thoroughly described in Castelli et al., [2013].

During the campaign the MIPAS-STR instrument was deployed aboard the M-55 Geophysica [Woiwode et al., 2012], as a demonstrator of the infrared limb sounder instrument proposed for the Earth Explorer 7. MIPAS-STR is a high resolution Fourier-Transform limb sounder with an unapodized spectral resolution of 0.036 cm^{-1}. The spectral channel 1 utilized for the retrievals considered here covers the spectral range from 725 to 990 cm^{-1}. Depending on the measurement scenario and flight altitude, typically one limb scan is obtained each 25 to 45 km along flight track. The MIPAS-STR measurements are inverted using the Tikhonov-Phillips regularization method, yielding vertical profiles of the target parameters with typical vertical resolutions between 1.0 to 2.5 km between flight altitude and the lowest tangent point (depending on the retrieval target). During the flight, MIPAS-STR detected dense clouds in the first leg below 8 km and in the final leg up to an altitude of 10 km.

H_2O, O_3 and HNO_3 profiles retrieved from MARSCHALS were then compared with MIPAS-STR results, using only the collocated results. Since the synergy between IR and mm-wave measurements is one of the main characteristic of the PREMIER mission, it was explored using the VMR profiles retrieved from the two limb sounders, using two data fusion techniques: the Level 1 plus Level 2 technique (L1+L2) and the Measurement Space Solution (MSS) method [Ceccherini et al., 2009].

Table 1: *DOFs, GI increase in % and total retrieval error reduction in % with respect to MIPAS-STR obtained from the (L1+L2) data fusion method for H_2O, O_3, and HNO_3 in each MARSCHALS Band.*

Quantifier	Band	H2O	O3	HNO3
DOFs	B	2	7	negligible
	C	8	5	2
	D	2	4	3
GI increase [%]	B	3	13	negligible
	C	11	7	8
	D	3	5	12
Total error reduction (10-20 km range) [%]	B	negligible	60	negligible
	C	50	60	30
	D	negligible	60	30

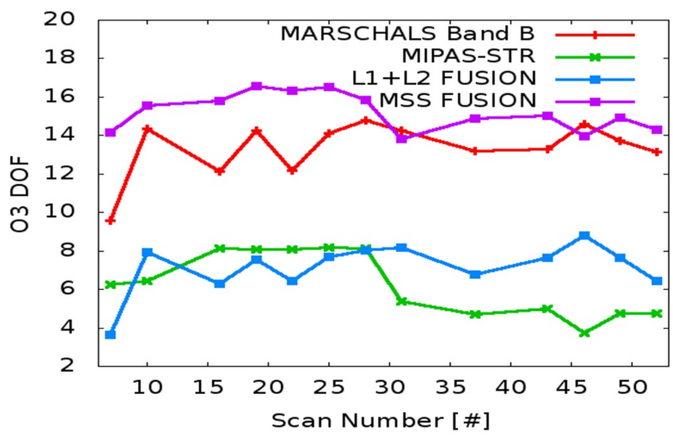

Figure 1: *Comparison of number of O_3 DOFs from individual retrievals for MARSCHALS Band B (red), MIPAS-STR (green) with the number of DOFs from synergistic retrieval performed using the L1+L2 (blue) and the MSS (purple) methods.*

III.I.I Investigation of potential synergy between mm-wave and infrared limb-sounding during the PREMIER-Ex campaign: L1+L2 and MSS methods

The potentiality of data fusion between mm-wave and infrared limb-sounding of the UTLS was investigated combining collocated data acquired by MARSCHALS and by MIPAS-STR and comparing the quality of the products from individual and synergistic retrieval of selected targets, i.e. H_2O, O_3, and HNO_3. The performance of the different methods was estimated using a set of quality quantifiers that included the Degrees of Freedom (DOF) of the retrievals, the Gain of Information (GI) (both described in Rodgers, [2000]) and the total retrieval error comprising the random and systematic components. Both data fusion methods, L1+L2 and MSS, were tested and a brief description of the results is given below.

The L1+L2 method consists in repeating the analysis of MARSCHALS Level 1 (L1) data using the Optimal Estimation (OE) method

[Rodgers, 2000] with the MIPAS-STR Level 2 (L2) products (with the associated covariances) used as a priori information. The MSS method is a post-retrieval processing and it has been thoroughly described in Ceccherini et al., [2009, 2010]. This method is based on the concept that a retrieved profile is the sum of a component belonging to the measurement space, the MSS, and a component belonging to the null space. The observations only provide information on the MSS and leave completely undetermined the component belonging to the null space. If we compute the MSS in the space that is the sum of the two independent atmospheric measurements spaces, we obtain the fused data. This new MSS includes all the information contained in the observations without any bias due to the a priori information used in the single retrievals.

The L1+L2 data fusion was performed on H_2O, O_3, and HNO_3 VMR vertical profiles retrieved from the best matching sequences of MARSCHALS and MIPAS-STR. Table 1 summarizes the results obtained for each retrieval target separated for the three MARSCHALS bands (B, C and D). Since MIPAS-STR L2 data are used as a priori, we report the improvements due to data fusion to those data. A significant improvement is found for H_2O in Band C for all the three considered quantifiers, while for the other two bands we have a slightly lower improvement for the DOF and GI. For O_3 we found an improvement (with a reduction of the total error of up to 60%) in all spectral bands with the best results obtained for the GI and DOF in Band B. In the case of HNO_3 retrievals, as expected, we did not obtain any improvement from Band B since there is negligible information on the HNO_3 in this band. However an improvement is obtained in both Bands C and D with a total error reduction in the altitude range from 10 to 20 km of up to 30%.

The evaluation of the performances of the MSS method has been performed for the O_3 VMR only. Here, we briefly report a direct comparison of the results obtained in Band B, highlight the basic differences between the two approaches and describe the potentialities of synergy between mm-wave and infrared limb-sounding measurements.

In Figure 1 the number of DOFs from individual retrievals is compared with the number of DOFs from synergistic retrieval performed using the L1+L2 and the MSS methods. We recall that the number of DOFs obtained using the L1+L2 method represents the independent information that can be retrieved from MARSCHALS measurements in addition to the a priori knowledge provided by MIPAS-STR L2 data, while the MSS number of DOFs are related to the fused data. The improvement obtained with the MSS approach is evident. This is due to the fact that the MSS approach is capable to evaluate the contribution of the measurement space component with no impact of the used external constraints, such as the Tikhonov regularization used for the MIPAS-STR retrievals [Woiwode et al., 2012], or the a priori information used in MARSCHALS retrievals.

The conclusions of this exercise are that the application of the L1+L2 method for the mm-wave and infrared measurements qualitatively demonstrates the improvements due to the synergy between the different techniques, but quantitative comparisons are difficult since the characteristics of the different retrieval methods are not fully compatible. In contrast, the MSS method allows for a more clear comparison of the individual results and the results of the data synergy, since the measured components from the different techniques are treated in a similar way and the same external constraint is applied to both. The investigation of potential synergy between mm-wave and infrared limb-sounding measurements shows that the fused products (obtained with both MSS or L1+L2 techniques) provide an improvement with respect to the single sensor retrieval. In particular, from Figure 1 the improvement given by the use mm-wave with respect to infrared measurements in case of presence of high altitude clouds is evident (last part of the flight, from MARSCHALS scan 31 onward).

III.II ESSenCe campaign: Flight on 16 December 2011 from Kiruna (Sweden)

The ESSenCe campaign consisted of two flights. The first flight was performed on the 11 December 2011 at the boundary of the Arctic polar vortex while the second flight was performed on the 16 December 2011 inside the vortex. During the second flight the M-55 followed a rectangular flight pattern. MARSCHALS spectra acquired during the second ESSenCe flight were used to retrieve VMR profiles of temperature, H_2O, O_3, HNO_3, N_2O, and CO inside the Arctic vortex. These profiles are compared with results of the chemistry transport models CLaMS and EMAC.

III.II.I Comparison between MARSCHALS and model results from CLaMS and EMAC

In order to assess the capability of the MARSCHALS instrument to resolve UTLS processes the retrieval results are compared to results from established models that allow to characterize the atmospheric conditions observed by measurements. While the primary goal of this cross-validation is to prove that atmospheric structures are represented adequately by the MARSCHALS results, these results in turn provide information to which degree these structures and the associated processes are captured by the models. The EMAC model is a numerical chemistry and climate simulation system used to investigate the past and future evolution of the atmosphere. The model includes sub-models for the description of tropospheric and middle atmosphere processes and their interaction with oceans, land and human influences. The CLaMS model, originally developed for the stratosphere, was extended to the troposphere and can be used to investigate atmospheric processes in high resolution studies. Both models are well suited for the comparisons with the MARSCHALS data and complements each other. The complementarity is due to different model resolutions and initialization, to differences in treatment of tropospheric processes and to differences in model architecture/transport scheme, which allows to identify shortcomings in the data sets. The comparison between MARSCHALS measurements and model results has been performed through the use of a common grid hereafter indicated as the Vertical and Horizontal Grid of the Comparison or VHGC. The models results were extracted on their own horizontal (2.8°x2.8° for EMAC and about 100 km for CLaMS) and vertical resolution and were interpolated considering the horizontal distribution of the MARSCHALS tangent point geolocations. The VHGC ranges from 0 km to 26 km, in steps of 0.5 km, with spatial (longitude, latitude, altitude) and temporal coordinates associated to each MARSCHALS limb scan. For each altitude level of the VHGC in the range between the lowest and highest MARSCHALS tangent points, we used the interpolated time, latitude and longitude of the tangent points immediately above and below the selected altitude, while for each altitude level of the VHGC

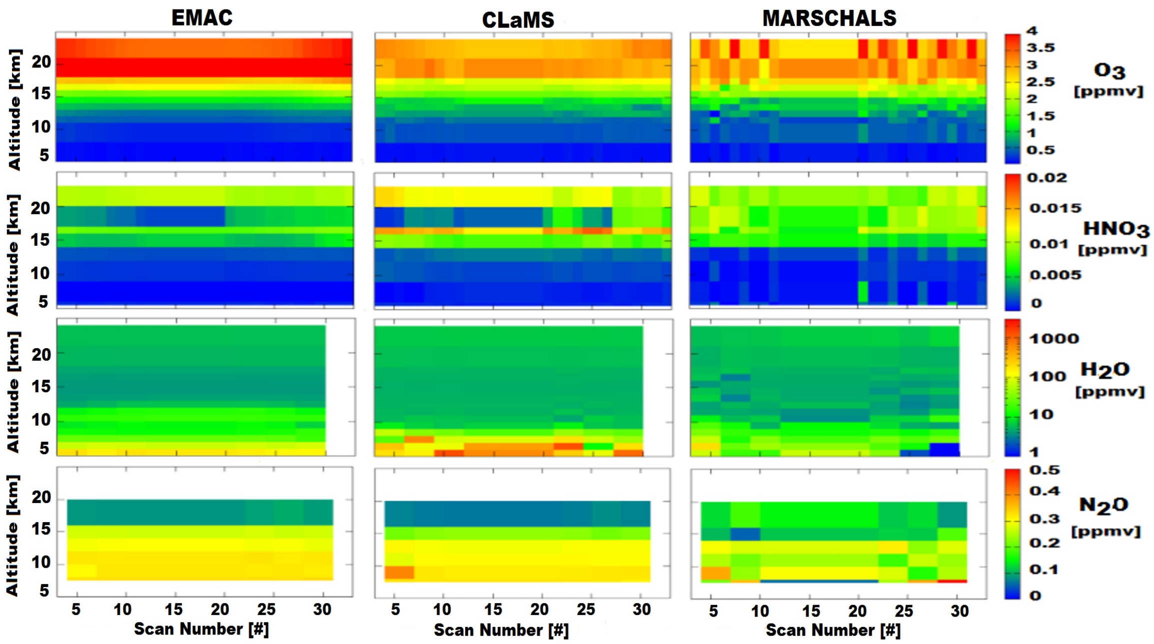

Figure 2: *From Top to bottom: EMAC (left) and CLaMS (center) results after convolution with MARSCHALS AK and MARSCHALS retrieved VMRs (right) for* O_3*,* HNO_3*,* H_2O *and* N_2O*.*

above (or below) the highest (or the lowest) MARSCHALS tangent point, we used the time, latitude and longitude values of the highest (or of the lowest) MARSCHALS tangent point. The models provide the atmospheric status on a very fine vertical grid. Retrieved data are obtained on a coarser grid, optimized for each target. In order to compare the model data with the retrieved vertical profiles we need to take into account the smoothing introduced by the retrieval procedure due to the information content of the MARSCHALS measurements. Using the method described in Ceccherini, [2012] we can take into account the retrieval effect on the model profiles using the expression reported below:

$$x_{model}=AK_h\, x_{model,h}+(I-AK)x_a+AK\ \hat{x}\ -AK_h\ \hat{x}_h \qquad (1)$$

where $\mathbf{x}_{model}$ is the vertical profile obtained by the models and reported on the vertical grid of the retrieval, $\mathbf{x}_{model,h}$ is the vertical profile provided by the models on the high resolution altitude grid, $\hat{\mathbf{x}}$ is the MARSCHALS retrieved profile on the vertical grid of the retrieval, $\hat{\mathbf{x}}_h$ is the retrieved profile interpolated on the high resolution altitude grid, $\mathbf{x}_a$ is the a priori profile represented on the vertical grid of the retrieval, **AK** is the square Averaging Kernel matrix on the vertical grid of the retrieval,is $\mathbf{AK}_h$ the rectangular high resolution Averaging Kernel matrix.

Figure 2 shows the comparisons of the convoluted EMAC (left) and CLaMS (center) model results with MARSCHALS (right) retrievals for O_3, HNO_3, H_2O and N_2O. The Figure shows that there are some differences between the two models and that in general, the agreement between models results and MARSCHALS measurements is quite good. The observed differences between the two models can be due to both the different chemical initialization (the CLaMS model was initialized using MLS data on the 1st November 2011 while EMAC was initialized with model data on the 1st January 2011) and to differences in the Polar Stratospheric Clouds (PSC) NAT particles sedimentation scheme. For ozone the models are in good agreement below 17 km, while above 17 km EMAC results are systematically larger. Within this uncertainty MARSCHALS ozone is in good agreement with the models. The HNO_3 comparison shows that below 15 km the models are in good agreement and the agreement with MARSCHALS retrievals is reasonably good. Above 15 km the differences among MARSCHALS and the models are larger; however these differences are present also between the model profiles with CLaMS results systematically larger then EMAC ones. Nevertheless both MARSCHALS and the models consistently show a HNO_3 reduction at 20 km. Models H_2O is in quite good agreement above 12 km, while below 12 km CLaMS results are systematically larger. At lower altitudes, MARSCHALS is in better agreement with EMAC results. The model results for N_2O are in good agreement, while MARSCHALS results are systematically lower than the models below the flight altitude. CO values from both models agree quite well in the stratosphere above 10 km, however CO from MARSCHALS retrievals assumes much lower values (not shown here) substantially because of the interference of CO with CH_3Cl [Castelli et al., 2013].

IV. Summary and Conclusions

In the frame of the PREMIER preparatory activities, the activities conducted for the ESA project "PREMIER Analysis of Campaign Data" aimed at demonstrating the capabilities of MARSCHALS to measure the vertical distribution of atmospheric targets (also through the comparison with chemical transport model results) and to evaluate of the potential synergy between limb measurements in the infrared and in the millimetre-wave region.

The inverse processing of MARSCHALS limb spectra, during the PREMIER-Ex 2010 and the ESSenCe 2011 campaigns, clearly demonstrated the capability of the instrument, when operating on the three spectral bands (i.e. Band B, C and D, centered respectively at 300, 325 and 345 GHz) to measure vertical profiles of temperature and VMR of Water Vapor, Ozone, HNO_3, N_2O and CO. This capability was successfully tested both in clear-sky and cloudy-sky conditions and proved the advantages of using mm-wave limb measurements to sound the UTLS altitude range in presence of clouds that are opaque to the infrared sounding. Significant results were obtained in the investigation of the potentiality of combining the information from collocated and independent data sets measured by mm-wave and infrared limb sounders. By using MARSCHALS and MIPAS-STR data on the flight of 10 March 2010, we compared the performances of individual and synergistic retrieval based on different approaches to data fusion. Both the L1+L2 and the MSS methods showed that the fused data have higher quality than the single one. The improvement was quantitatively assessed through selected quantifiers (total retrieval error, DOFs and GI). In addition, we proved that the MSS method enables an easier evaluation of the quality improvements of the data fusion products. Finally, the comparison of MARSCHALS retrieved profiles with results from CLaMS and EMAC models showed that for O_3 a generally good agreement is found between retrieved profiles and model outputs. The agreement is good also for H_2O and HNO_3, with discrepancies between model values and MARSCHALS products at some altitudes. For N_2O and CO MARSCHALS retrieved profiles are in general low compared to model simulations.

Acknowledgements

We acknowledge ECMWF for having granted direct access to their archives of atmospheric data and J. Langen for useful comments.

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