PREMIER – Quantification of Atmospheric Pollution and Climate Aspects

Executive Summary

ESA contract number 4000101294/10/NL/CBi

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Contents

Chapter 1

Introduction

PREMIER will be the first mission to generate atmospheric trace gas fields at a resolution high enough to study stratosphere-troposphere exchange, tropical convection, the Indian monsoon, pyroconvection, long-range transport of air pollution (and associated chemical conversions) and signatures of mesoscale dynamics including gravity waves. Through the use of limb imaging spectrometry in the infrared, PREMIER will observe 3-D fields of atmospheric composition in the upper troposphere and lower stratosphere (UTLS) with unprecedented spatial resolution and coverage. While the importance of the UTLS for dynamical, chemical, and radiative processes and their interactions affecting climate change and air quality is generally accepted, there is a lack of measurements of global atmospheric composition observations in this height range. Existing data are either sporadic (such as aircraft and balloon data) or lack the needed vertical and horizontal resolution (space data). The candidate Earth Explorer mission PREMIER is the first space mission dedicated to and optimised for the observation of this crucial layer of the atmosphere. The overall aims of this study are:

1. to demonstrate the capability of the Earth explorer candidate mission PREMIER to quantify key processes controlling global atmospheric composition in the mid/upper troposphere and lower stratosphere (5–25 km height range), which is a region of particular importance for climate change
2. to demonstrate the added value of PREMIER-type observations for validation of chemistry-climate models (CCM) and data assimilation systems used for numerical weather prediction (NWP)

The study was performed on behalf of a consortium led by **Research Centre Jülich** in collaboration with eight partner institutes **Centre national de la recherche scientifique (CNRS-GAME and CNRS-LISA)**, **German Aerospace Center (DLR) Oberpfaffenhofen**, **Environment Canada**, **Karlsruhe Institute of Technology (KIT)**, **Royal Netherlands Meteorological Institute (KNMI)**, **York University (Toronto)**, and **WxPrime Corporation (Toronto)**, together with a group of scientific experts who specialize in specific areas of relevance to this study, who was involved at consultancy level: **Prof. Piers Forster**, **Dr. Michaela Hegglin**, **Dr. Brian J. Kerridge**, and **Prof. Paul Palmer**.

The study demonstrates that the high vertical resolution and the 3-dimensional sampling of composition data obtained from PREMIER will enable small scale processes in the atmosphere to be disentangled as highlighted in the scientific objectives of PREMIER. The prime objective of this study is to investigate the possible impact of PREMIER data on the research on several atmospheric processes important to air quality, climate change and their interaction. The study is split into four corresponding tasks:

- **Task-1:** Quantify the impact of biomass burning on atmospheric composition in the UTLS and on the atmospheric radiation balance;
- **Task-2:** determine quantitatively the transport processes involved in stratosphere-troposphere exchange at high spatial resolution and investigate the impact of resolution on the calculated radiative forcing patterns;
- **Task-3:** add value to data assimilation systems such as used for numerical weather prediction or environmental prediction;
- **Task-4:** contribute to the validation of chemistry-climate models.

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Chapter 2

Task-1: Impact of pyroconvection on UTLS composition and radiative balance

Pyroconvection can be loosely described as the vertical transport of gases and aerosols as a result of the burning of biomass in the tropics and in boreal forests. The energy to sustain the vertical transport can be fuelled by unstable meteorology or by the thermal energy released by the burning process, or a combination of these processes. Sometimes the term is restricted to transport into the upper troposphere and lower stratosphere (UTLS). As a result of the admixture of VOCs and NOx from the burning, water vapour and UV sunlight there can be significant perturbation of the ozone budget leading to an impact on the radiative balance in the UTLS. Species other than VOCs can be emitted that can serve as important diagnostic markers of biomass burning (BB): HCN, CH3CN, and CO are, perhaps, the most important of these "marker" gases but certain halogenated-compounds (e.g., CH3Cl) and sulphur compounds (e.g. SO2) are also produced.

Pyroconvection events tend to be localised and so after the gases reach the maximum vertical level they will often form a compact plume in the mid-troposphere and UTLS. Characterisation of the extent of the plume, together with meteorological information, and knowledge of the location of the burned surface area can yield detailed knowledge of the processes that occur in the burning and convective processes.

The scope of **Task-1** is to study the role of high spatial and temporal measurements of trace gases provided by the PREMIER mission to advance understanding of the pyroconvection required to improve its parametrisation in large-scale models. This study is also intended to provide a rough assessment of how PREMIER measurements might advance understanding of both the chemical and radiative impact on the UTLS. This Task was broken down into 4 Work Packages (WPs), WP1100 "Generation of fire emission scenarios", WP1200 "Construction of 3-D atmospheric composition field scenarios", WP 1300 "Sampling of atmospheric fields with limb-sounding instruments" and WP1400 "Quantification of the perturbation of the radiation balance".

In **WP1100** the data and methods used to quantify surface fire emissions were documented. Three fires or fire periods that resulted in significant vertical transport of BB products into the UTLS (pyroconvection) were chosen. The three cases selected for which there is substantial evidence of vertical transport of emissions to the UTLS were (a) Victoria, Australia (Kilmore East), February 2009, (b) Siberia, July 2006, and (c) Borneo, October 2006. We quantified emissions for 23 species and sensible heat release for these fires by using biome-specific emission factors and fuel consumption in combination with MODIS 500-m burned area product for Siberia, MODIS daily active fire product and GFEDv3 monthly burned area and carbon emissions for Borneo, and PHOENIX Rapid-Fire fire spread model output for Victoria. These three emission scenarios were then used as input for global variable-grid high-resolution (~2×2 km2 horizontal resolution in the uniform core centered on the fire region) non-hydrostatic GEM-AQ runs, and the output from these high resolution runs were then used as input to global variable-grid medium-resolution (~50×50 km2 in the uniform core) hydrostatic GEM-AQ runs.

During the preparation of the fire data for WP1100 it was noted that the use of the MODIS 500-m burned area product for the Kilmore East and Borneo fires would have led to a substantial underestimation of burnt area, based on comparison with ground data (for Kilmore East) and GFEDv3 burnt area product (for Borneo).

In **WP1200** fire simulations are provided at three different scales using the input from WP1100 and other global data. The simulations are (a) high-resolution non-hydrostatic, (b) medium-resolution and (c) global hydrostatic simulations. The first two types of simulations use GEM in global variable mode (GV) where an interior domain is surrounded by an expanding resolution grid. The data from these runs have been processed as they would be seen by the PREMIER and MIPAS instruments (WP1300) and the radiative forcing (RF) has been estimated for the specific cases and the results used to project the estimated RF for a year for similar fires.

The distribution of a comprehensive suite of chemicals is calculated but the analysis was focused on PREMIER related species while a more complete suite of species has been stored for possible further analysis.

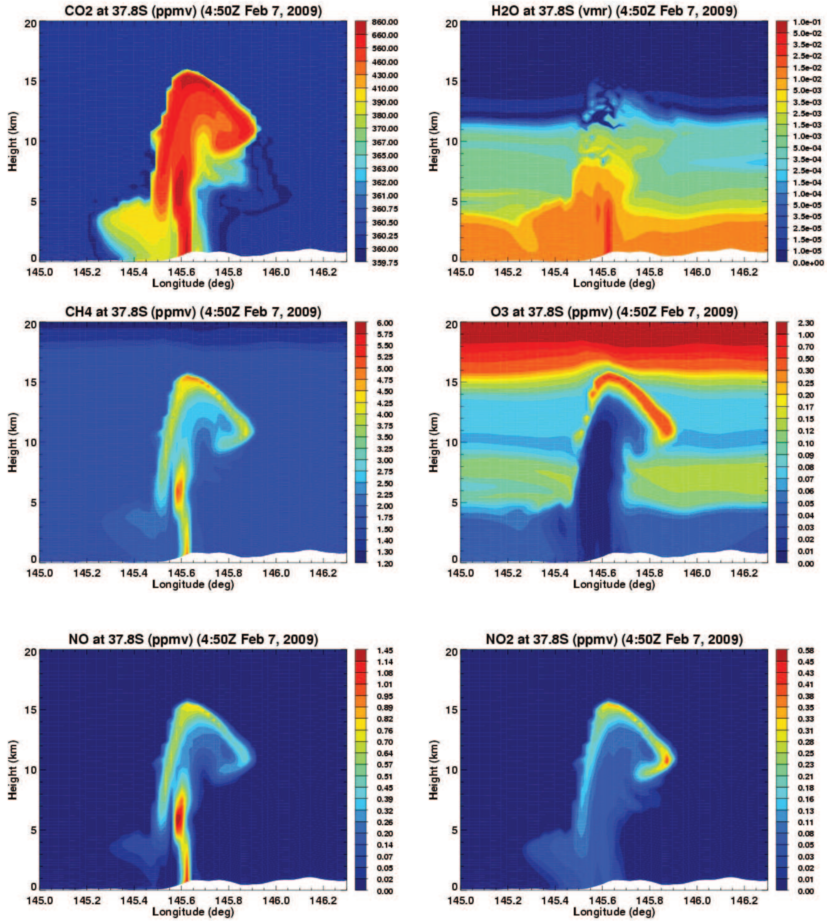

Figure 2.1: An altitude-longitude slice through the plume which has reached above 15 km is shown for 4:50 GMT or 2:50 PM local time during the Kilmore East fire. A characteristic convective plume structure is evident with a narrow central core and a mushroom shaped cap between 5 and 15 km. This snapshot shows the mixing ratios of CO2, CH4, H2O, O3, NO, and NO2 after the onset of convection.

From the analysis of the high resolution Kilmore East fire simulation it was estimated that the pyroconvection was driven by the heat from the fire – the meteorological situation prior to the start of the fire was stable against vertical convection. There were some limitations with the high resolution simulation attributed to the 1D radiative transfer code so that insufficient energy was radiated laterally from the vertical plume; a semi-empirical solution was found based on horizontal diffusion of temperature. The simulation launched material in the UTLS although the final height was quite sensitive to the details of the initial conditions varying by a few kilometers.

During the < 1 day simulation there was substantial generation of ozone at the cap of the plume and below this the ozone was titrated by the NO produced in the fire. Copious amounts of HCN, CO, CH4, CO2, and other species were produced by the fire (see Figure 2.1). In the high resolution runs the UT H2O vapour distribution was greatly disturbed from the background as a result of the induced convection. We found that the Kilmore East fire is a case of purely fire driven convection. Without the heat generated by the fire no convective tower would have formed on this day due to dry surface conditions and a statically stable temperature profile.

There were two medium resolution simulations for the dispersion of the Kilmore East fire emission products, one with the majority of the BB products injected at ~ 8 km and one with the products injected at ~ 12 km using the output from high resolution simulations. The levels of the species as noted above were well above background and were transported around in the globe in a westerly direction in about 2 weeks in each scenario (cf. Figure 2.2); the higher injection height gave a better comparison with MLS and MIPAS.

Resolution issues were addressed for **WP1300**. The medium resolution simulation was examined carefully for

WP1300 and it was clearly demonstrated that many features of the plume would be "washed out" by the horizontal and vertical resolution associated with current instruments such as AURA/MLS and ENVISAT/MIPAS. On the other hand it was clearly demonstrated that the spatial variations that resulted from the pyroconvective events could be resolved by the PREMIER instruments (cf. Figure 2.3). As the plume stretched out over the two week period simulated that PREMIER would also be able to characterise the structure of both the air quality and RF gases for these intense fires.

For the high resolution simulations of fires in Siberia any pyroconvective vertical plumes that developed were quite perturbed in the sense that the vertical transport could not really be described as being a plume or chimney-like. However, for the fires in Borneo very narrow vertical plumes developed and appeared to be triggered by the topography as much as the heat input. However, it was found in the medium resolution runs that the overall meteorological situation during the period of the fires for Borneo resulted in rapid vertical transport to 15-20 km with subsequent rapid horizontal transport. The mixing ratios of species such as HCN, CO etc. generated in the fire and transported to the UT were such a magnitude that they were well above background levels and thus the plumes would be readily detectable by PREMIER.

As part of **WP1200** a set of global biomass burning emissions based on GFEDv3 was generated for year 2008. Two one-year global simulations (with and without biomass burning emissions) on a 1.5° global uniform grid were run for year 2008. And as part of WP1400 the radiative forcing (RF) was calculated and it was found that it was significant ~ 46 mW m-2.

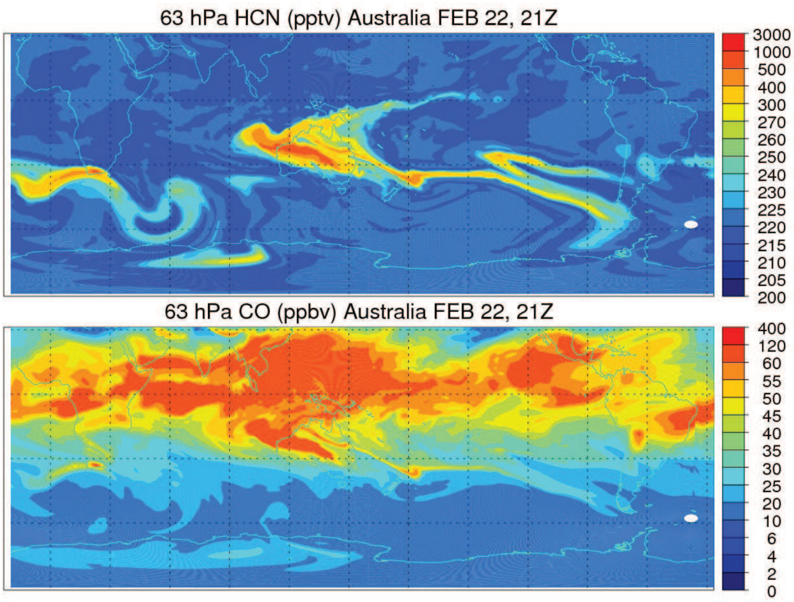

Figure 2.2: HCN (a) and CO (b) at ~ 19 km for Kilmore East Fire for February 22, 2009 at 21:00Z. The scales are fixed except at the upper end in order to accommodate the maximum as it is diluted.

The scope of **WP1300** is to demonstrate the capability of the instruments aboard PREMIER to measure atmospheric trace gas fields with a much higher spatial resolution than current spaceborne limb sounders such as MIPAS on ENVISAT or MLS on EOS. For this purpose the GEM-AQ model field for Kilmore East fire was sampled and smoothed according to the sampling rate and spatial resolution of PREMIER IRLS, PREMIER STEAMR, MIPAS and MLS. The model field has a horizontal resolution of 50 km over an area of ~ 4,000x6000 km2 containing the biomass burning, outside this domain it has a coarser horizontal resolution over the rest of the world on an irregularly spaced grid. The spatial sampling of the PREMIER instruments used was appropriate for the dynamics and chemistry modes while that of MIPAS/ENVISAT was 4.0° along track, 1.6 km in the vertical and that of MLS 3.0° along track and 1.2 km in the vertical. As noted above this analysis indicated that the spatial and temporal resolution of PREMIER's instruments would allow characterisation of the horizontal and vertical details of the plumes that ended up in the UT.

For **WP1400** the RF was estimated for the high resolution simulations averaged over the interior uniform grid, ~ 600x600 km2 with and without averaging appropriate to several contemporary instruments and also PREMIER and it was shown that there were significant differences in the estimated RF that were resolution dependent.

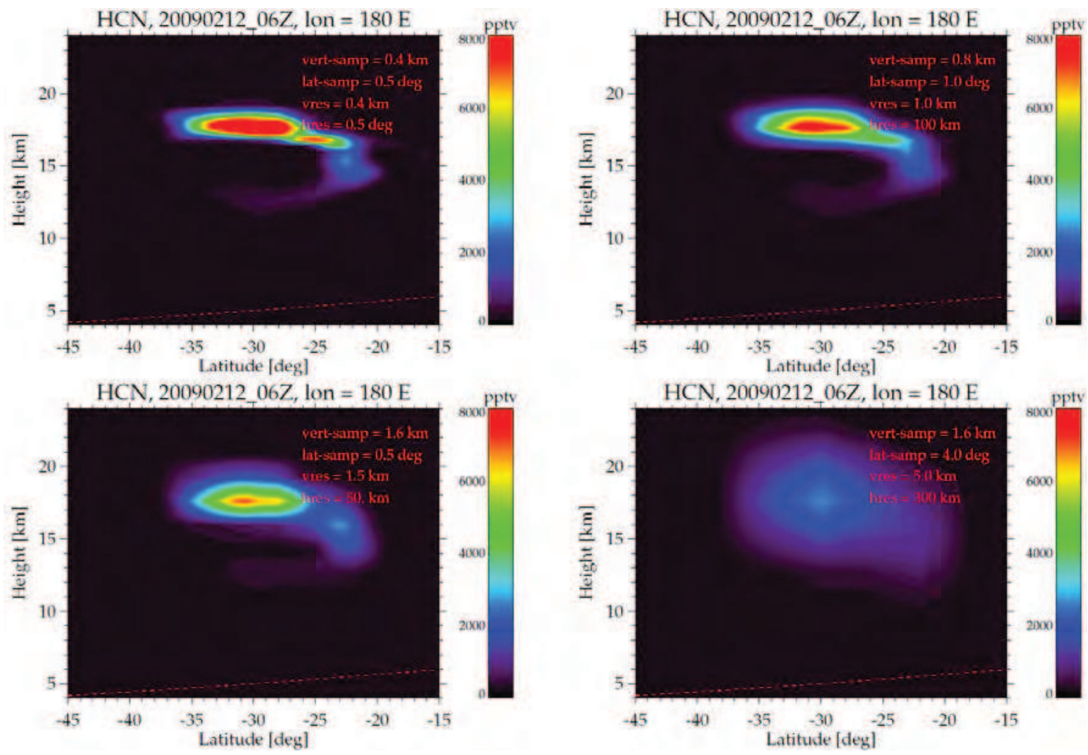

Figure 2.3: Latitudinal cross sections for 12 February 2009, 6 UT, at 180°E across the Australian biomass burning plume. Original HCN distribution modelled by GEM-AQ (top left), and HCN distribution degraded to the sampling and resolution of PREMIER IRLS in chemistry mode (top right), of PREMIER STEAMR (bottom left) and of MIPAS/ENVISAT (bottom right). Vertical and horizontal sampling and resolution are noted in the plots.

These results showed that there was substantial local RF of ~ 2 Wm-2, (local, i.e. averaged over the interior grid). This was used to project the RF from pyroconvective events that reached above 5 km. The data used to estimate this number for pyroconvective events came from a variety of sources going back ~ 30 years in some cases, viz MODIS fire data, MISR plume height data, TOMS and use of the aerosol index (AI). For example, using the MISR smoke data base indicates a large yearly variability.

It was also noted that ~ 5-8% of BB emissions are lofted above 5 km and ~ 2-4% above ~ 10 km, of which the latter is mostly in the tropics which would likely be in the UT as opposed to the LS. It was estimated that ~ 14 pyroevents per year is representative of fire events lofting material above ~ 5 km. Projecting the Kilmore East fire we estimated a RF of ~ 4.2 mWm-2 for a global and annual average (this assumed a two week lifetime for the plume/fire perturbations). This estimate is probably an overestimate since the 1 day RF includes water vapour effects which will likely dissipate more rapidly since the H2O is, to first order, under thermodynamic control. This can be compared to the global annual RF estimate for BB of ~ 46 mWm-2 due largely to ozone with methane as a minor player. The global estimates for RF do not include a water vapour impact since there is no interaction between the fires and the meteorology in the global run discussed above. If we compare this estimate with our estimate of the instantaneous RF due to pyroconvection events (which do, however, include water vapour), i.e. the "instantaneous" RF = 4 mWm-2 we see that the local estimate is much smaller. This suggests that the secondary effects of BB are much larger, i.e. the increase in ozone, increase in CO and OH over a period of a few months. Even allowing for the uncertainty in fire count the conclusion is that the majority of the RF is associated with longer time scale events such as the oxidation of VOCs and CO to produce ozone which itself has a lifetime of ~ 1 month or more in the UT.

Our overall conclusions are that the PREMIER experiment as compared to current instruments, by virtue of its spatial and temporal resolution and coverage would be able to make measurements that would permit a better understanding of the processes involved in pyroconvection and also the efficiency of the biomass burning process. This would lead to an improvement of models. Also with an improved knowledge of the transport of material to the upper troposphere and lower stratosphere (UTLS) PREMIER would be able to better assess the radiative forcing (RF) due to pyroconvection.

Chapter 3

Task-2: Quantification of transport in the UTLS using high resolution data

A quantitative understanding of transport processes in the upper troposphere and lower stratosphere (UTLS) region and its connection to climate change is one of the most challenging tasks for the PREMIER mission, because changes in the UTLS have a strong impact on radiative forcing (RF) and surface parameters like temperature, pressure, and precipitation. Transport processes at the tropopause determine the concentrations of water vapor, ozone, and other chemical species and thus the radiative properties in this sensitive region. In particular, two-way mixing across the extra-tropical (ExTL) tropopause layer and vertical mixing in the tropical tropopause layer (TTL) determine the spatiotemporal distribution of trace gases in the UTLS. Further, the region of the Asian monsoon appears to be a very important pathway for the transport of water vapor and pollutants from the troposphere into the stratosphere (via the TTL).

Projections of future climate change are subject to uncertainties in the representation of UTLS temperature and composition and their changes in a changing climate. In general, the representation of composition changes in the UTLS involves two sources of uncertainty, one is the defining emission scenarios for anthropogenic greenhouse gases such as carbon dioxide and methane, the other is the representation of dynamical processes (and associated transport), chemical, and micro-physical processes determining the structure and composition of the UTLS. The quality of the representation of the impact of well-mixed gases such as carbon dioxide relies to a large degree on the quality of emission scenarios. For natural greenhouse gases such as water vapor (the dominant greenhouse gas) and ozone, which are characterized by steep vertical and horizontal gradients in the UTLS, the situation is quite different. For water vapor and ozone the most important uncertainties arise from an inadequate model representation of atmospheric processes such as transport. Consequently, multi-model inter-comparisons show significant differences e.g. in UTLS water vapor and ozone that often depend on how physical, chemical and micro-physical processes in the UTLS are represented in the individual models (references from e.g. SPARC CCMVal).

Stenke et al. (2008) demonstrated that a failing water vapour distribution in the UTLS region (e.g. insufficient representation of the strong vertical gradient with high mixing ratios in the troposphere and low mixing ratios in the stratosphere) is to a large part responsible for the cold bias in the polar tropopause region and the cold pole problem in the polar lower stratosphere, a feature which is found in many CCMs, AGCMs, and climate models. Moreover, Stenke et al. (2009) showed that using a fully Lagrangian advection scheme (E39C-A, Stenke et al., 2009) instead of a semi-Lagrangian scheme also improves very much the overall skill of the corresponding CCM (E39C, Dameris et al., 2005) with regard to dynamical and chemical processes in the Earth stratosphere and troposphere (at the horizontal resolution of this CCM).

In **WP2500**, we used the results derived from two long-term, transient simulations (i.e. variable boundary conditions representing the period from 1960 to 2000) with the CCMs E39C and E39C-A to identify and quantify differences in the spatial distributions of radiatively active gases in the UTLS region (e.g. water vapour and ozone). It could be shown that the application of different advection schemes has obvious not only influences on the regional distribution of trace gases, but also on the seasonal cycles and the long-term (i.e. decadal) behavior. The impact on surface climate is identified by looking at differences in horizontal distribution of representative quantities, like precipitation and near surface temperature, which are derived from the same decadal simulations performed with E39C and E39C-A. The differences obtained this way highlight the link between uncertainties in transport / mixing and the evolution of surface climate. It could be demonstrated that climatological differences (e.g. absolute values, variability, and gradients from different model simulations) must be resolved by respective observations in the UTLS region, i.e. the accuracy of measurements should be sufficient to evaluate differences between model results and to rate the importance of model uncertainties with respect to prognostic studies of climate change. A detailed description of the findings is given in the WP2500 section of the final report.

From comparisons with zonal mean water vapour distributions measured by HALOE, it is obvious that the fully

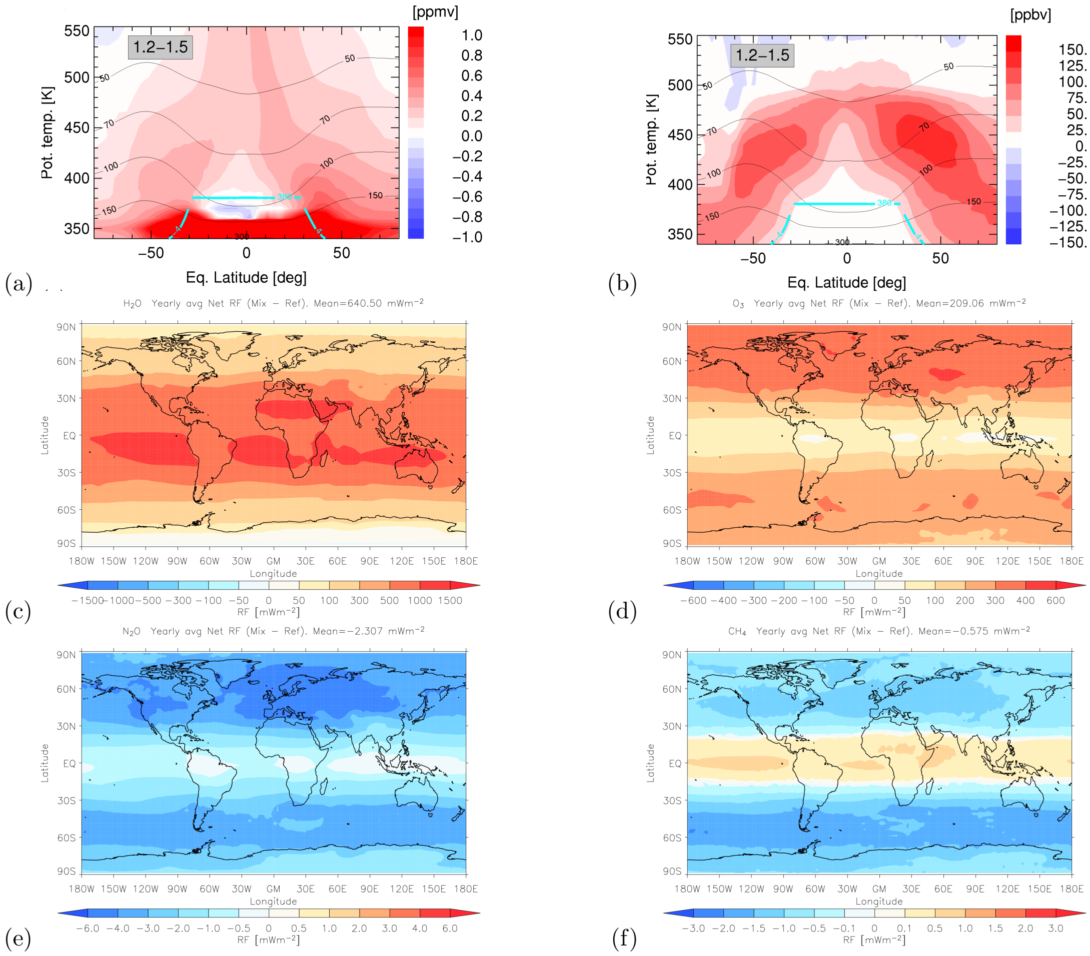

Figure 3.1: Difference in water vapor (a) and ozone (b) between the enhanced mixing and the reference CLaMS run, depicted as zonal mean for the year 2003. Radiative forcing differences between enhanced mixing run minus reference run for H2O (c), O3 (d), N2O (e), and CH4 (f).

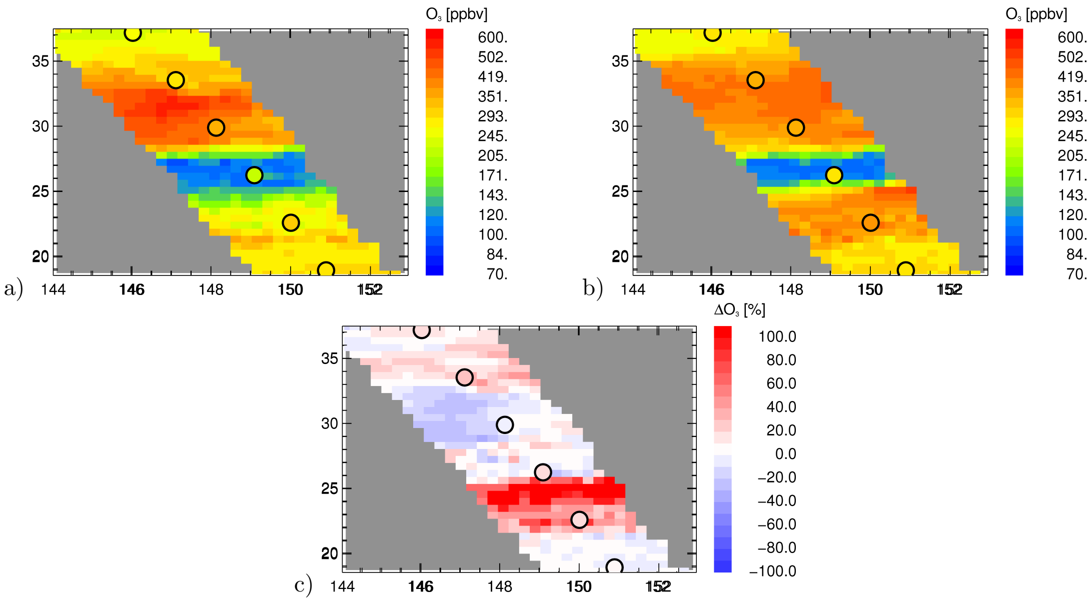

Figure 3.2: Ozone along a selected PREMIER orbit in the Asian monsoon region at 18km on 8 August 2003 from the high resolution (25km) reference run (top left), the enhanced mixing run (top right) and the difference enhanced mixing-reference run (bottom). Filled circles show the corresponding MIPAS observations for comparison.

Lagrangian scheme (E39C-A) provides a much more realistic water vapour distribution. The results of this simulation can therefore be regarded as the reference case, while the E39C results can be regarded as a “disturbed” case demonstrating the sensitivity of UTLS composition (and consequently surface climate) on major shortcomings of the advection code. This should not be interpreted as a weakness of a semi-Lagrangian (SL) representation in general, because the quality of a SL representation could be drastically improved by increasing the horizontal resolution (from the coarse value of 4 degrees used by E39C). However, even for the best Lagrangian and Semi-Lagrangian schemes (at high spatial resolution), there will remain uncertainties concerning the influence of irreversible mixing. This influence is therefore investigated on the basis of the Lagrangian model CLaMS.

The Chemical Lagrangian Model of the Stratosphere (CLaMS) is the first Chemistry Transport Model (CTM) based on Lagrangian transport where a physically meaningful concept of a deformation-induced mixing was successfully realized both in 3 dimensions (McKenna et al., 2002; Konopka et al., 2004). In CLaMS, the irreversible part of transport, i.e. mixing, is controlled by the local horizontal strain and vertical shear rates with mixing parameters (e. g. critical Lyaponov exponent) deduced from observations. Variations of the critical Lyaponov exponent allow to assess the impact of uncertainties in mixing on UTLS composition and associated radiative forcings (**WP2100, WP2200**). Global and high resolution (~ 100 km / 400m – horizontal/vertical resolution) simulations cover both the troposphere and the stratosphere were performed.

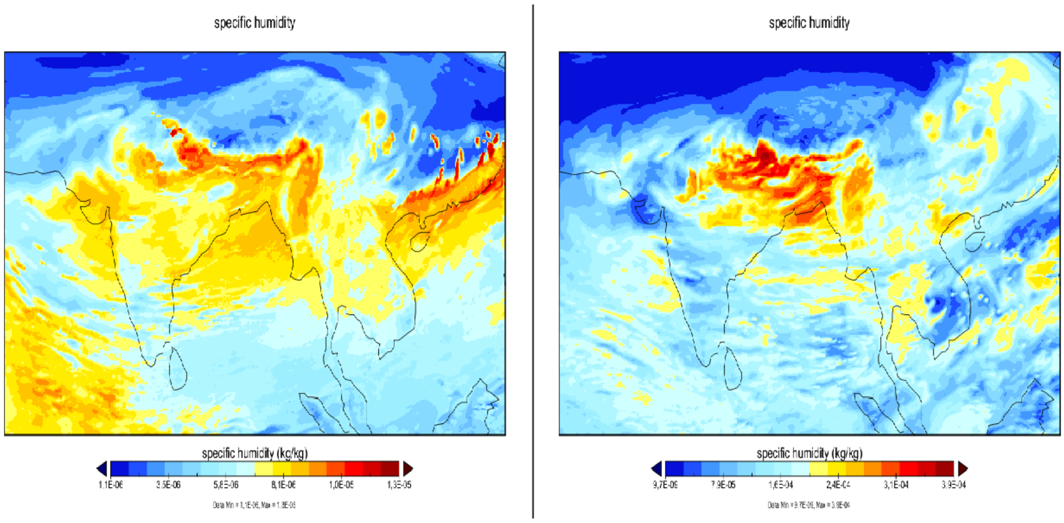

Figure 3.3: Geographical trace gas distribution of water vapor for the monsoon scenario at altitudes of 16km (left) and 12km (right).

To determine the sensitivity of the trace gas fields on the mixing strength, we have increased the mixing parameter λ (critical Lyapunov exponent) from the reference value of 1,5 to 1,2 (enhanced mixing case). These particular values of the mixing parameter λ appear to be well in the uncertainty range related to mixing (compare e.g., Konopka et al., 2003; Khosrawi et al., 2005; Konopka et al., 2005). Zonally averaged water vapor and ozone mixing ratio differences are shown for the reference run in Figures 3.1a and 3.1b, respectively. As a result of enhanced mixing, an increase of water vapor concentrations in the extra-tropical lowermost stratosphere and a significant increase of ozone throughout the lower stratosphere are found.

Differences between the CLaMS simulation with increased mixing and the reference run are interpreted in terms of radiative forcing (RF) (Leeds model by Piers Forster). They are shown in Figures 3.1c to 3.1f for the radiative active species H2O, O3, N2O, and CH4, respectively. The results impressively demonstrate that simulated radiative forcings of atmospheric species with strong spatial gradients in the UTLS (e. g. water vapour and ozone) are rather sensitive to uncertainties in the mixing scheme. For water vapour and for ozone, large differences of the yearly averaged global forcing for 2003 are found (0.64Wm2 and 0.2 Wm2, respectively). On the other hand, radiative forcings of trace gases that are rather well mixed in the UTLS such as nitrous oxides (N2O) and methane are rather insensitive to changes in the mixing strengths. For these gases, uncertainties in their tropospheric sources appear to be much more important.

In **WP 2300**, we investigated the capability of PREMIER to constrain uncertainties in mixing parameters. For this purpose, we used highly resolved snapshots in the region of the Asian monsoon, generated from CLaMS high resolution short-term runs (25km horizontal resolution) for the reference case and for the case with enhanced mixing. The top row of Figure 3.2 shows ozone mixing ratios along a selected PREMIER orbit in the Asian monsoon region at 18km on 8 August 2003 (reference run on the left, run with enhanced mixing on the right). This presentation already includes the PREMIER IRLS averaging kernel. For both cases, a pronounced filament of ozone-poor air can be seen. Filled circles show the corresponding MIPAS observations for comparison. While the PREMIER IRLS reproduces the differences of the simulated fields within a few percent, MIPAS fails to pick up ozone differences (bottom) that are of the order of 50% in the range of the ozone-poor filament (that means well above target and threshold errors for ozone in this region). In conclusion, PREMIER is well suited to resolve differences in structures resulting from different mixing strengths, while MIPAS-type instruments provide do not provide sufficient resolution.

The negative impact of too strong smoothing by MIPAS-type instruments are highlhigted in some results of WP2300. As a “test case” snap shots in the area of the Asian Monsoon were used (see Figure 3.3). The effect of instrument smoothing on calculated “instantaneous” radiative forcings for this area is most pronounced for water vapor as a result of its steep vertical gradients. Radiative forcing values obtained with the MIPAS-smoothing deviate from values obtained from the high-resolution model output by 3.5 Watt/m2. For PREMIER (IRLS dynamic mode), deviations are about an order of magnitude smaller (for more details see **WP2400** final report).

Conclusions:

PREMIER will be able to narrow down uncertainties in transport schemes (e.g. mixing) that have large impact on globally averaged radiative forcings of water vapour and ozone. This is a result of PREMIER’s capability to resolve small-scale structures in the UTLS such as filaments that are strongly affected by mixing processes. In contrast, currently flying limb-sounders like MIPAS are not able to provide information at the necessary spatial resolution.

Further, Task-2 and corresponding findings from other recent investigations clearly show that changes in the trace gas distribution in the UTLS region (and associated changes in radiative forcings) will feed back to the troposphere and therefore contribute to climate change. In particular, surface parameter like temperature and precipitation at high latitudes turn out to be very sensitive on changes in the UTLS. PREMIER will help to close gaps in our understanding of the interaction of the UTLS region with climate change. The mission will therefore significantly improve current state-of-the-art atmospheric and chemistry-climate models.

Chapter 4

Task-3: Impact of UTLS observations on data assimilation

This task aims at documenting how PREMIER data could benefit in different research and application areas, where assimilation of remote-sensing data is the key to final output quality. The first application (**WP3100**) area is Numerical Weather Prediction (NWP). The second part (**WP3200**) concerns ozone tropospheric contents. Last, **WP3300** is a sensitivity analysis targeted at estimating the added value of taking into account stratospheric methane variability for surface flux inversion.

4.1 WP3100: Assessment of impact from PREMIER in Numerical Weather Prediction

The purpose of this work package was to provide insight on the possible value added impact of including the assimilation of PREMIER temperature, water vapour, and ozone data in global numerical weather (and ozone) prediction using Observation System Simulation Experiments (OSSEs).

The preparatory phase was devoted mostly to producing assimilation-ready synthetic (simulated) observations, in the planning and setting of the various assimilation and forecasting experiments, and in conducting a random error calibration for the control observation set. The synthetic observations were produced from the output of the existing ECMWF Joint OSSE T511NR free model nature run serving as virtual truth. The simulated observation sets consisted of (a) a control dataset, (b) retrieved profiles of temperature, water vapour and ozone based on data characteristics of the Aura Microwave Limb Sounder (MLS-Aura), and (c) retrieved profiles as would be available from the PREMIER InfraRed Limb Sounder (IRLS) and Millimetre Limb Sounder (MWLS) instruments. The control dataset relied on characteristics of an existing observing system for operational meteorological assimilation and ozone partial column measurements from the Solar Backscatter Ultraviolet Spectrometer-2 (SBUV/2). Use of the MLS-type data served as an additional benchmark for the evaluation of the impact of the PREMIER-type data. The observation simulations, relying on the nature run (NR) as truth, were produced using the existing infrastructure and the observation operators already established for assimilation at Environment Canada (EC).

Global assimilation and 10-day forecasting experiments (14 and 8, respectively) were conducted with the EC Global Environmental Multi-scale (GEM) forecast model with the addition of the LINOZ linearized ozone chemistry parameterization. The model grid consisted of an 80-level hybrid η vertical coordinate extending from the surface to 0.1 hPa and a uniform 800x600 longitude-latitude grid. Incremental assimilations were conducted with the 3D-Var FGAT scheme with a T108 spectral resolution for the 2.5 month periods covering Summer 2005 and Winter 2006. The assessment of the relative impact of IRLS, MWLS, and MLS-type data on assimilation analyses, 6hr forecasts, and medium range forecasts relied mainly on examining monthly mean root-mean-square (RMS) errors, differences and ratios of RMS errors, time mean errors and differences, and anomaly correlation coefficients. A few issues limiting the performance of the assimilations were identified when evaluating the assimilation output; their potential consequences were discussed.

While the assimilation setup was furthest from being optimal for IRLS, the presented results indicate that IRLS, and MWLS to a lesser but significant degree, should benefit numerical weather and ozone predictions. Greater improvements than those generated from the addition of MLS-type data would also certainly be attained for ozone and, to a lesser degree, water vapour. This does not reflect consequences of any biases or differences in error covariances that might be present in the real observations.

The separate additions of IRLS and MLS-type temperatures were found to typically reduce temperature and wind zonal RMS 6hr forecast errors in the troposphere and lower stratosphere by less than 0.1-0.3 K and 0.2 m/s with a slight advantage to IRLS. Largest temperature improvements in the troposphere are seen in the polar regions with

RMS and time mean error reductions of about 1 K from IRLS and about half a degree from MLS; there are also improvements of about half a degree from IRLS in parts of the mid-latitude regions . For winds, improvements in the troposphere are less than 0.5 m/s from IRLS and half that from MLS. The medium range forecasts showed similar to less or no improvements relative to the control.

The largest beneficial impact on analyses and forecasts from the addition of IRLS as compared to MLS-type data and to the control case were found for ozone and water vapour in the troposphere and UTLS, e.g. see figures 4.1 and 4.2. MWLS provides weaker improvements over MLS in these same regions. The initial analysis benefits for water vapour and ozone can partially persist over the entire 10-day forecasts depending on the vertical level, this being most notable for the UTLS. In the stratosphere where the control case was already close to the nature run in various areas due to the choice of initial conditions, the impact benefit of the additional IRLS, MWLS, and MLS-type data was comparatively weaker (except in the Winter pole for ozone) or even negative. The latter resulted from the observation noise increasing RMS errors.

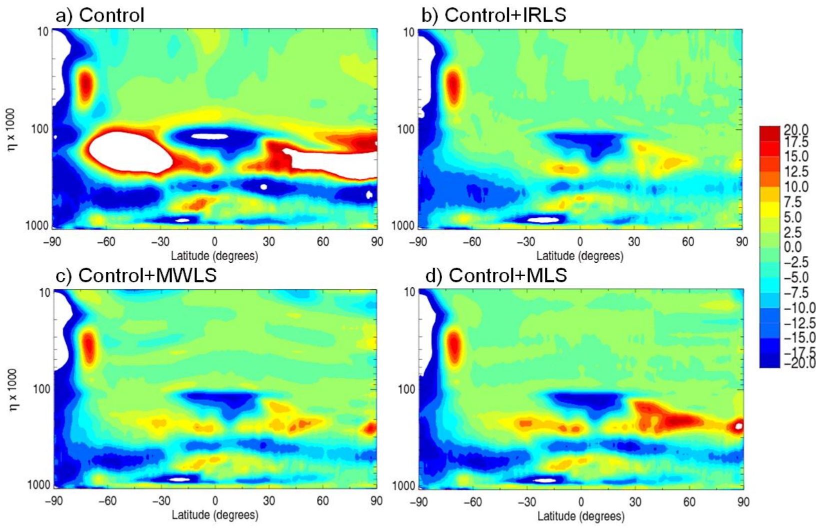

Figure 4.1: Zonally averaged time mean 6hr forecast errors for water vapour (in 100x δln(q);~% for mixing ratios) for different assimilation experiments relative to the nature run for July 2005.

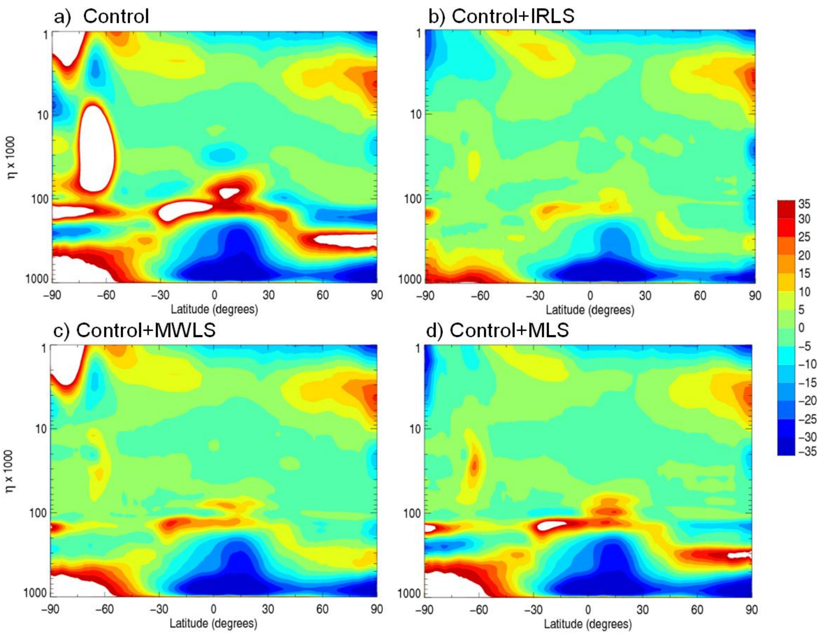

Figure 4.2: Zonally averaged time mean 6hr forecast errors for ozone (%) for different assimilation experiments relative to the nature run for July 2005.

4.2 WP3200: Tandem assimilation of MIPAS+IASI ozone observations)

The aim of this study was to demonstrate the potential of assimilating simultaneously limb and nadir ozone satellite observations for improving ozone distribution in the troposphere, in support of the PREMIER mission and of its requirement of flying in formation with MetOp. Today, observations of ozone in the troposphere are scarce, while ozone plays a major role there, contributing to the greenhouse effect, to modifying the oxidising capacity of the atmosphere and to Air Quality. Constraining the ozone budget in the troposphere by assimilating remote-sensing observations would thus bring considerable societal and scientific benefits.

PREMIER is a limb-sounding instrument that has unprecedented characteristics, particularly regarding horizontal resolution. Rather than taking an approach of OSSEs (Observing System Simulation Experiments) as in WP3100 for instance, we decided to use the MIPAS and MLS ozone limb data currently available as proxies for future PREMIER observations. The drawback of this choice is that we don't directly get an estimate of how PREMIER will perform, but we know that the results we obtain with the current instruments provide an underestimation of the benefits to expect with PREMIER. We opted OSSEs out because there is no other source of tropospheric information to assimilate currently and, thus, the "nature" and "control" runs could only be (raw) model outputs, that are possibly quite far from reality in terms of vertical and horizontal scales in tropospheric ozone distributions and of its temporal variability.

We use for this study the MOCAGE-PALM chemistry-transport model and assimilation suite that has already been successfully applied to the assimilation of various kinds of remote-sensing ozone data. Assimilation has been performed using a variational 3d-FGAT (First-Guess-At-Appropriate-Time) algorithm. Statistical tests have been performed in order to check the consistency between the errors provided for the observations and the background on the one hand and the results obtained on the other hand. We have carried out a total of 5 experiments over a period of four months, May to August 2009 (May serving as a spin-up period): FREE (a free-running MOCAGE model simulation), MIPAS (assimilation of MIPAS ozone data, from **WP3210**), MLS (assimilation of MLS ozone data), IASI (assimilation of IASI tropospheric ozone data, from **WP3220**) and SYN (tandem assimilation of MIPAS and IASI ozone data).

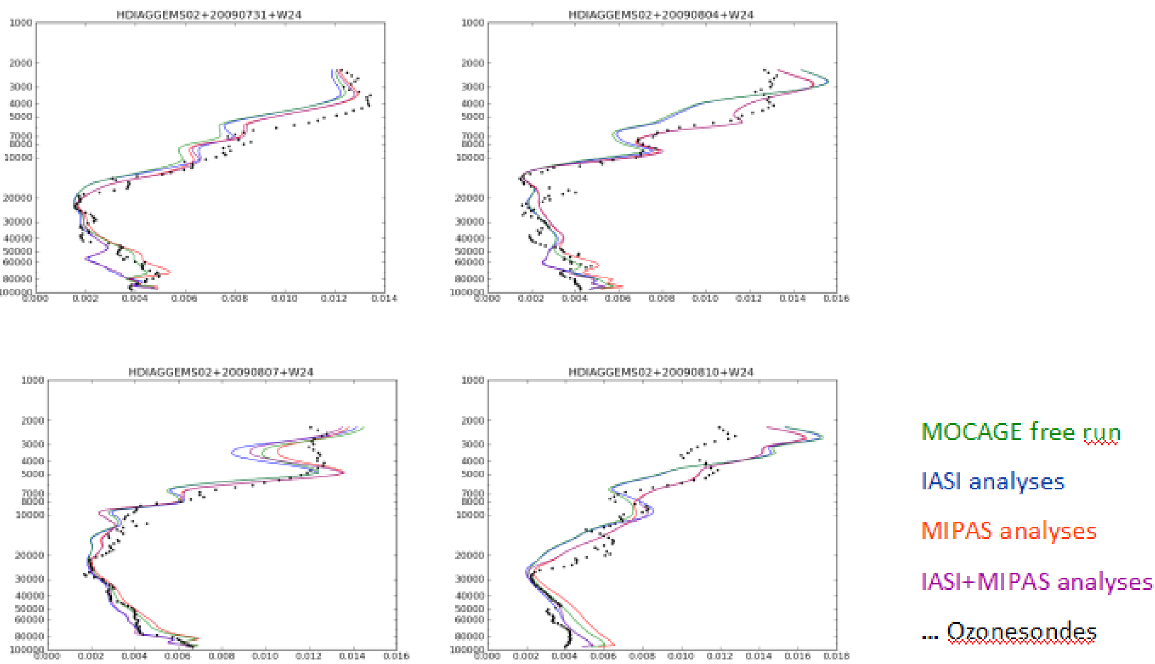

Figure 4.3: Ozone sonde at Payerne, Switzerland on August 7th 2009 (dots); FREE (green) ; MIPAS (red); IASI (blue); SYN (purple). SYN provides similar results as IASI in the troposphere and as MIPAS in the stratosphere, in both cases with best agreement with the independent observation

We show that limb instruments (MLS, MIPAS) are able to constrain effectively ozone in the region of the UTLS

(Upper Troposphere and Lower Stratosphere). We have studied a case of stratospheric intrusions in mid-July 2009, for which we had independent in-situ observations (a MOZAIC aircraft flight and an ozone sonde). Provided the chemistry-transport model resolution is sufficiently fine, assimilation of ozone limb data allows bringing realistic ozone structures down to the lowermost troposphere. This is a major asset regarding applications and in particular Air Quality forecasting. However such intrusion events are not very frequent and the current tropospheric ozone observing system is poor: conclusions can just be drawn on the basis of case studies, as there are not enough cases that can be captured in observations to impact on statistical skill scores of analyses or forecasts. Also, we stress that effects on model skill scores can be flawed by the possibility of error compensation: in our case, the model system has a positive ozone bias in the troposphere, mainly due to too active photochemistry; when assimilating limb observations only, the better description of the UTLS region leads to an increase in the stratospheric flux, which has unfortunately a detrimental impact on scores as it adds up more ozone -when this is mainly due to the fact that another term in the ozone budget is wrong.

The assimilation of IASI data brings a strong constraint on tropospheric and UT ozone. The IASI product used in this study is the one developed at LISA (**WP3220**); it is the retrieval with the lowest-peaking Averaging Kernel for each profile. Its assimilation in MOCAGE leads to a reduction of the strong model bias during summer 2009 over Europe and to improve surface air quality hindcasts. Regarding statistical skill scores against the European surface Air Quality database Airbase, the bias and rmse are improved, while correlation is unchanged. We have checked that these retrievals of IASI data are actually bringing this bias reduction, and that it is not just an effect from the a priori that is used. However, when we compare the IASI analyses with MOZAIC independent observed profiles (ascents and descents at Frankfurt airport), we find that an erroneously high variability is introduced by IASI retrievals in the middle troposphere -at least for a part of the period studied (June 2009). We suggest that this could be linked to issues in the IASI retrievals, which has to be investigated further by LISA.

The tandem assimilation of MIPAS and IASI data allows cumulating the benefits of the two types of information, with the stratosphere principally constrained by the limb instrument and the troposphere by the nadir one (see figure above). The SYN analyses present the best overall performance against independent ozone sondes (only available at Payerne for the domain and period considered). However, as previously mentioned, it is currently impossible to highlight the benefits of the synergy between the two instruments in statistical terms, because of the low frequency of UTLS events influencing the lower troposphere and of the scarcity in independent observations.

We conclude that today's state-of-the-art Air Quality assimilation and forecast models like MOCAGE, which is one of the systems used in the context of GMES atmospheric services [Hollingsworth et al. 2008], are effectively able to assimilate simultaneously limb and nadir ozone information. This allows constraining and improving ozone profiles in both the troposphere and stratosphere. The data coverage of PREMIER, as well as its horizontal resolution, will allow characterising the stratospheric influx of ozone in the analyses, which has an important influence on the variability of tropospheric ozone. Used in conjunction with IASI, this will improve the background ozone distributions and thus it will contribute to improve Air Quality forecasts, as well as estimates of the oxidising capacity and of the radiative forcings due to tropospheric ozone.

4.3 WP3300: Sensitivity of constraining stratospheric methane for surface fluxes inversion

The aim of this work package was to assess the potential improvements through CH4 observations in the UTLS to better constrain surface emission inversions based on nadir CH4 total column observations. A more general purpose of this work package for PREMIER was to expand on the potential applications in the lower troposphere using the proposed nadir-limb combination. State-of-the-art CH4 global surface emission inversions are constrained by nadir total column mixing ratio (CMR) observations in combination with surface networks (e.g. Bergamaschi et al., JGR, 2009). The inverse system gets its information from minor variations in the total CMR. However, certainly not all of the observed variations in total CMR can be related to surface emissions. Atmospheric transport, chemical oxidation, and mixing processes at all altitudes also contribute to variations in total CMR. So far these atmospheric variations are captured by the forward models in emission inversions, without any observational constraints in the UTLS region.

The important questions to answer in this work were:

- What is the contribution of atmospheric processes in the UTLS region to total CMR variations?
- To what extend are these variations captured by a state-of-the-art chemistry-transport model?
- What would be the observational requirements for CH4 profile observations in the UTLS region to better constrain present-day CH4 emission inversions?

Two TM5 chemistry-transport model simulations were performed. In the BASE simulation the surface boundary condition for CH4 was set to the observed zonal-mean climatological background values, while in the much more-sophisticated CH4-HYBRID simulation the CH4 emission distribution was prescribed in combination with a nudging in the background atmosphere to surface observations. Comparison between the BASE and CH4-HYBRID simulation sampled at the time and location of in situ aircraft observations reveals the contribution of surface emissions to the observed variability in the UTLS region.

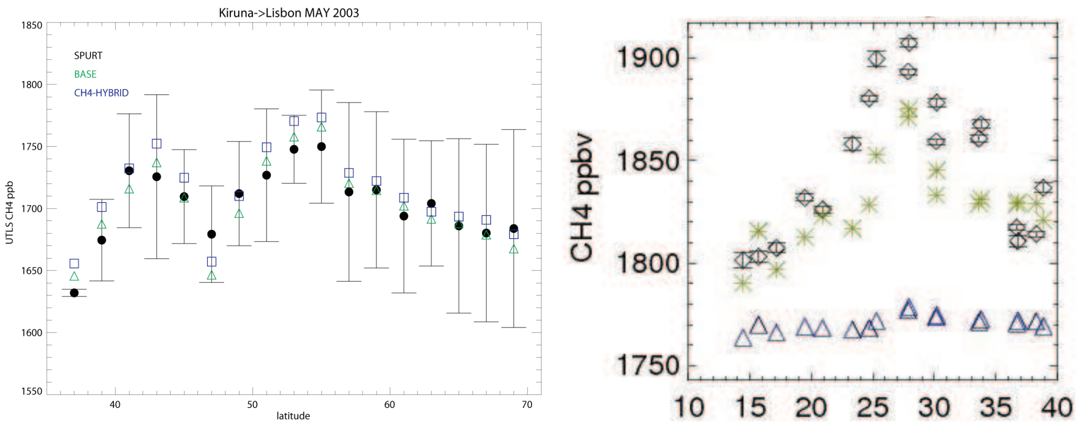

Figure 4.4: (left) Comparison of the BASE and CH4-HYBRID simulation with observations of the SPURT (Engel et al., ACP, 2006) campaign from North to South over Europe in May 2003; (right) Comparison of the BASE (triangles) and CH4-HYBRID (asterix) simulation with observations of two return flights by CARIBIC from Frankfurt to Chennai (India) v.v. in August 2008 (Schuck et al., ACP, 2010).

In winter (February 2003) over Europe the UTLS variability is only marginally affected by the European CH4 emissions given the close correspondence between both simulations (Figure 4.4, left). The latitudinal variation which is seen in both the observations and simulations is in this case dominated by long-range transport, tropopause height variations and stratosphere-troposphere exchange. In contrast, in northern summer (August 2008) over the Asian monsoon region a large difference is simulated between BASE and CH4-HYBRID, while CH4-HYBRID shows much a better comparison with the observed peak in CH4 concentrations (Figure 4.4, right). This peak in the UTLS region is caused by convective uplift of the large regional emissions over Asia in this time of the year. In both situations the model is able to follow the observed variability to some extent, but the differences with the observations may exceed several tens of ppb or a few percent.

The CH4 variations in the UTLS region contribute significantly to the variations in the total column mixing ratio (CMR). The mixing ratio at the 150 hPa level varies between ~1400-1850 ppb in the CH4-HYBRID simulation. At the 500 hPa level the mixing ratio varies between ~1730-1880 ppb. Although the relative variations are smaller at the lower altitude, the contribution to total CMR variations is somewhat larger because of the increased air density. To illustrate this effect the contribution of CH4 variability to the total CMR was calculated for the atmosphere above the 150 hPa level and above the 500 hPa level. The column above 150 hPa contributes ~12% to the total column and the spatial variability, as diagnosed from the zonal-mean anomalies in a daily global field, is ~0.3%. The column above the 500-hPa level contributes about half (48%) to the total column while the spatial variability is about ~0.5%. These percentages are significant compared to the observed variability in the total CMR. The left panel in Figure 4.5 shows the total CMR as retrieved from SCIAMACHY. The right shows the tropospheric column mixing from the surface up to 500 hPa (CMR-T500). This product has been produced by subtracting from the observed total CMR the simulated column above 500 hPa from CH4-HYBRID. The averaging kernel of the SCIAMACHY observations was taken into account. The different distribution of CMR-T500 compared to total CMR demonstrates that the UTLS contributes significantly to the variations in total CMR. The CMR-T500 distribution better resembles the surface emission distribution.

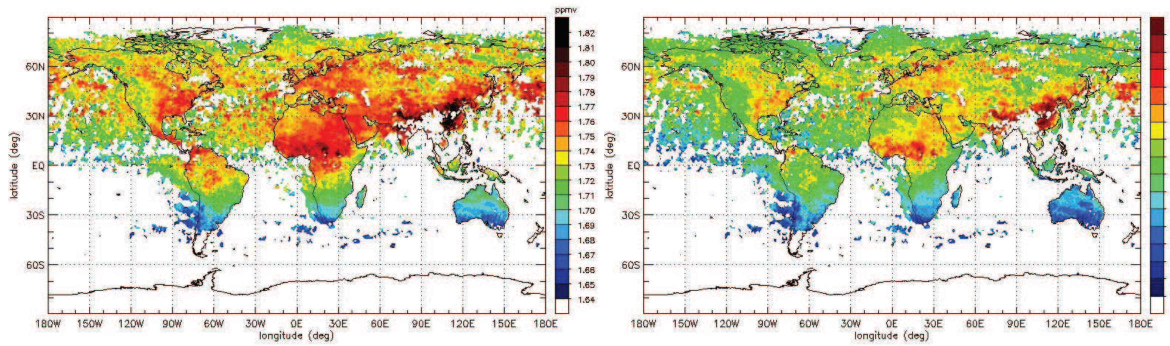

Figure 4.5: The observed total CMR from SCIAMACHY for the month of August 2004 (left) and CMR-T500 (right)as derived by subtracting the TM5 CH4-HYBRID simulated column above the 500 hPa level from the SCIAMACHY total CMR observations taking into account the averaging kernel. The color scales are not the same.

Link to PREMIER and observational requirements for the CH4 profile Observations down to ~150 hPa level would capture a large part of the variability in total column mixing ratio (CMR) due to stratospheric transport, mixing and chemistry, tropopause height variations, and stratosphere-troposphere exchange. However observations down to ~500 hPa would be much better because these capture in addition most of the variability caused by convection and long-range transport in the upper troposphere. The larger air density of the layer between 150 and 500 hPa lead to larger absolute contribution to variations in the total CMR below 150 hPa even though the relative variations are largest in the stratosphere. The observational requirements on CH4 in the UTLS region are stringent because of the limited variability in CH4 concentrations. The PREMIER threshold requirement of about 50 ppbv or ~3% for CH4 observations below the tropopause in order to capture the upper tropospheric variability in CH4 concentrations is confirmed by this study. The threshold vertical resolution would be the retrieval of a partial column of CH4 in the UTLS region. It is recommended to investigate if the combination of IASI CH4 observations and limb CH4 observations by the PREMIER IRLS instrument could provide the required observational constraints in the UTLS region on the total CMR variations.

Chapter 5

Task-4: Impact of UTLS composition data on validation of chemistry-climate models

Task-4 of the PREMIER impact study investigated the impact of PREMIER satellite measurements on chemistry-climate model (CCM) validation. One of the largest uncertainties introduced in CCM validation up-to-date is the lack of knowledge about the representativeness of atmospheric observations and therefore how much we can trust diagnostics derived from those observations. Inhomogeneous and temporally and spatially restricted sampling patterns of instruments can lead to biases in calculated climatologies. This is especially true in the upper troposphere/lower stratosphere (UTLS), where measurements are sparse and natural variability is high. This issue has been expressed in chapter 7 - The Upper Troposphere and Lower Stratosphere - in the SPARC CCMVal report (2010) as follows:

The UTLS is still relatively sparsely sampled by observations, which limits confidence in the quantitative evaluation of model performance in the UTLS.

The aim of **Task-4** therefore was to obtain information on the improvement of PREMIER's representativeness over some hitherto available data sets, and to quantify in turn the impact this improved representativeness would have on CCM validation. For this purpose, an idealized experiment has been designed using the Canadian Middle Atmosphere Model (CMAM). The CMAM has been chosen, since it is a fully coupled CCM, which had been thoroughly validated within the CCMVal activity and which had turned out to be one of the best models representing transport and dynamical processes in the UTLS.

For the idealized experiment, instrument errors and biases, as well as averaging kernels and sampling patterns for different state-of-the art instruments (which included SHADOZ ozone sondes, ACE-FTS, MIPAS, SAGE, and MLS) as well as PREMIER (for which the focus has been on the IRLS-instrument) had to be compiled. The high-resolution ozone fields from CMAM then were sub-sampled based on the sampling pattern of these instruments. Climatologies based on the sub-sampled data are then compared to those calculated with the full data set, which is defined as the truth since it includes all available data points. The differences in the sample and full climatologies can be attributed solely to sampling biases. In a second step, known instrument errors, biases as well as averaging kernels have been applied to the sub-sampled data, the climatologies have been re-calculated, and compared to the full model data set. The differences in these fields now comprise the full uncertainty of a given instrument's climatology and therefore the uncertainty that would be introduced to CCM validation. For some of the instruments, the errors, biases or averaging kernels were also used separately in order to quantify their contributions to the full uncertainty of a given instrument's climatology.

Our evaluations with full and sub-sampled fields included climatological zonal mean cross-sections, seasonal cycles, and vertical profiles in the tropics. In general it can be said that the most accurate current instruments tend to have large sampling errors (these instruments include the ACE-FTS and the SHADOZ ozone sonde data). On the other hand, current instruments with good sampling have large measurement errors (this was seen especially for MIPAS and MLS). SAGE II has been proven to be the best current instrument, however, its sampling focuses on the tropics and it is highly likely that a similar evaluation in the extratropics would not yield as favouring results. The climatologies derived using the PREMIER sampling and measurement characteristics are always very close to the climatologies derived from the full data set. The results of our evaluations have shown that PREMIER exhibits sampling characteristics that minimize sampling errors to virtually zero compared to other instruments, in particular to solar occultation measurements such as SAGE or ACE-FTS. Another key point is that the high vertical resolution of PREMIER prevents strong smearing across regions where strong tracer gradients are found in the atmosphere, leading to improvements especially over comparisons using MIPAS and MLS. In addition, the large number of measurement profiles (which are in reality sampling the model's sub-grid domain) is beating down PREMIER's rather large measurement errors. Finally, another key result of this study has been, that due

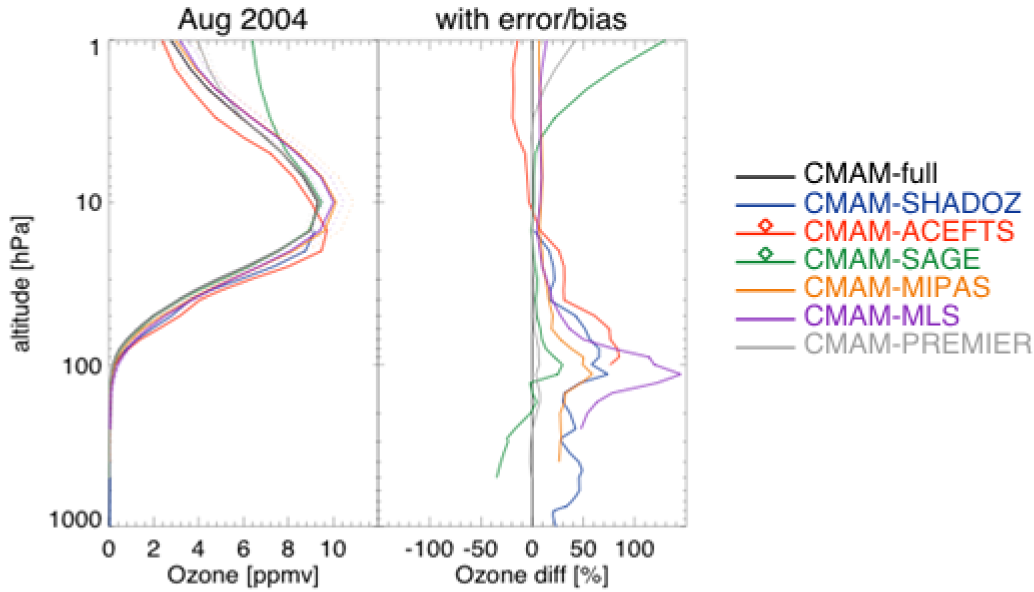

Figure 5.1: This figure quantifies how well PREMIER reproduces a climatological atmospheric ozone profile in the tropics in comparison with climatologies derived from hitherto available instruments. The study was performed using simulated ozone profiles produced by the CMAM chemistry-climate model, which were sub-sampled according to the different instruments' sampling and measurement characteristics, including measurement errors and averaging kernels. The errors in the estimated climatologies therefore result from a combination of sampling, vertical resolution, and measurement error. The left panel shows the vertical profiles of the mean tropical ozone concentrations, where CMAM-full represents the "truth". The right panel shows the percentage errors of the climatological profiles produced by the different instruments. In the critical UTLS region below about 80 hPa (approximately 18 km), PREMIER is seen to reduce the errors to about 5%, compared with the errors ranging between about 20-100% from the other instruments.*

to the sampling restrictions and measurement errors of current ozone instruments, it is hard to determine natural variability. However, this would be a key diagnostic to add for improving current CCM validation activities in the future.

The impact of PREMIER on CCM validation efforts can be quantified by how close PREMIER got to the true model field compared to the other instruments. It has been found that at particular levels in the UTLS, PREMIER will improve our knowledge of up to 30% when compared to other instruments, and will therefore substantially increase our confidence in future model-measurement validation results in this particular region. Finally, the results of this study provide evidence that PREMIER's sampling and measurement characteristics are likely to yield insight into sub-seasonal, inter-seasonal, and interannual variability in the vertical ozone distribution with much higher confidence than hitherto possible.

In conclusion, PREMIER would help improving CCM validation especially in the UTLS, where atmospheric observations are still sparse and dynamical variability often introduces large errors when trying to compile climatologies for validation purposes. The UTLS will remain a major focus in CCM validation, since the new generation of models will include a more sophisticated representation of the troposphere. PREMIER's measurements would help defining climatological mean variables without having to care about sampling biases, and also yield insight into natural variability on shorter time and length scales as hitherto available. The high spatial and temporal resolution observations are necessary to develop process-oriented diagnostics, which are key to successful CCM validation and deeper understanding of what physical and chemical processes govern tracer distributions in the UTLS. The high spatial and temporal resolution measurements would in particular help to investigate the relation between the Brewer-Dobson circulation strength and its impact on the vertical ozone distribution on different time and length-scales, and to study the associated variability.

Chapter 6

Conclusions and Recommendations

The prime objective of this study was to investigate the possible impact of PREMIER data on the research on several atmospheric processes important to air quality, climate change and their interaction. The study was split into four corresponding tasks, with the following contents:

Task-1: Quantify the impact of biomass burning on atmospheric composition in the UTLS and on the atmospheric radiation balance

Task-2: Determine quantitatively the transport processes involved in stratosphere-troposphere exchange at high spatial resolution and investigate the impact of resolution on the calculated radiative forcing patterns

Task-3: Show added value to data assimilation systems such as used for numerical weather prediction or environmental prediction

Task-4: Show contributions to the validation of chemistry-climate models

The results of all four tasks show impressingly the large impact that the PREMIER mission would have. This can be summarized as follows:

The results obtained in **Task-1** show that biomass burning makes a significant contribution to the overall atmospheric radiative forcing (RF). They also highlight that PREMIER as compared to current instruments, by virtue of its spatial and temporal resolution and coverage would make measurements that permit a better understanding of the detailed processes involved in pyroconvection and also the efficiency of the biomass burning process. This would lead to an improvement of models and their representation of radiative forcings. Also with an improved knowledge of the transport of material to the UTLS PREMIER would be able to better assess the RF due to pyroconvection.

In **Task-2**, it could shown that PREMIER would be able to narrow down uncertainties in transport schemes (e.g. mixing) that have large impact on globally averaged radiative forcings of water vapour and ozone. This is a result of PREMIER's capability to resolve small-scale structures in the UTLS such as filaments that are strongly affected by mixing processes. In contrast, currently flying limb-sounders like MIPAS are not able to provide information at the necessary spatial resolution. Further, Task-2 (WP2500) and corresponding findings from other recent investigations clearly show that changes in the trace gas distribution in the UTLS region (and associated changes in radiative forcings) will feed back to the troposphere and therefore contribute to climate change. In particular, surface parameter like temperature and precipitation at high latitudes turn out to be very sensitive on changes in the UTLS. PREMIER will help to close gaps in our understanding of the interaction of the UTLS region with climate change. The mission will therefore significantly improve current state-of-the-art atmospheric and chemistry-climate models.

Task-3 shows that PREMIER data can benefit in different research and application areas, where assimilation of remote-sensing data is the key to final output quality. The assimilation of PREMIER temperature, water vapour, and ozone data (**WP3100**) in global numerical weather (and ozone) prediction using Observation System Simulation Experiments (OSSEs) showed that the largest beneficial impact on analyses and forecasts from the addition of IRLS as compared to MLS-type data and to the control case were found for ozone and water vapour in the troposphere and UTLS.

State-of-the-art Air Quality assimilation and forecast models like MOCAGE are effectively able to assimilate simultaneously limb and nadir ozone information (**WP3200**). This allows constraining and improving ozone profiles in both the troposphere and stratosphere. The data coverage of PREMIER, as well as its horizontal resolution, will allow characterising the stratospheric influx of ozone in the analyses and therefore will contribute to improve Air Quality forecasts, as well as estimates of the oxidising capacity and of the radiative forcings due to tropospheric ozone.

Link to PREMIER and observational requirements for the CH4 profile observations down to ~150 hPa level would capture a large part of the variability in total column mixing ratio (CMR) due to stratospheric transport, mixing and chemistry, tropopause height variations, and stratosphere-troposphere exchange (**WP3300**). However observations down to ~500 hPa would be much better because these capture in addition most of the variability caused by convection and long-range transport in the upper troposphere. It is recommended to investigate if the combination of IASI CH4 observations and limb CH4 observations by the PREMIER IRLS instrument could provide the required observational constraints in the UTLS region on the total CMR variations.

Task-4 showed that PREMIER would help improving CCM validation especially in the UTLS, where atmospheric observations are still sparse and dynamical variability often introduces large errors when trying to compile climatologies for validation purposes. PREMIER's measurements would help defining climatological mean variables without having to care about sampling biases, and also yield insight into natural variability on shorter time and length scales as hitherto available. The high spatial and temporal resolution observations are necessary to develop process-oriented diagnostics, which are key to successful CCM validation and deeper understanding of what physical and chemical processes govern tracer distributions in the UTLS. The high spatial and temporal resolution measurements would in particular help to investigate the relation between the Brewer-Dobson circulation strength and its impact on the vertical ozone distribution on different time and length-scales, and to study the associated variability.

In conclusion, in this study it was demonstrated

1. that PREMIER is able to quantify key processes controlling global atmospheric composition (e. g. transport, mixing, radiative forcing) in the mid/upper troposphere and lower stratosphere (5–25 km height range), which is a region of particular importance for climate change,

2. that PREMIER has an added value for or validation of chemistry-climate models (CCM) and data assimilation systems used for numerical weather prediction (NWP).