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## FRENCH/BELGIAN SCIENTIFIC CONTRIBUTION TO TROPOSPHERIC STUDIES USING THE METOP SENSORS

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### Abstract

We propose to combine the tropospheric measurements provided by the IASI and GOME2 instruments aboard METOP, together with data from ground-based stations, airborne and spaceborne remote sensors, along with atmospheric chemistry models (CTMs) in order to improve our knowledge of processes constraining the chemical composition of the troposphere and to study the regional and global scale air quality. The measurements of ozone (O<sub>3</sub>), carbon monoxide (CO), nitrogen dioxide (NO<sub>2</sub>), formaldehyde (CH<sub>2</sub>O), and methane (CH<sub>4</sub>) will be used in conjunction with the 3D tropospheric LMDz-INCA, MOZART, IMAGES and CHIMERE CTM models, using data assimilation and inversion modeling techniques, to

study the global distribution of these species and to derive improved emissions estimates.

Before the METOP launch, all the tools developed in the framework of this project will be used to analyse the AURA TES and OMI tropospheric products. After the launch of METOP, the data provided by both satellites will then be analyzed with the CTMs. The resulting global distributions will be used to perform detailed studies of the role of biomass burning in the budget of tropospheric species, the emissions of ozone precursors, the role of convective transport of pollutants, and its consequences in terms of air quality.

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## 1. Analysis of atmospheric spectra

### 1.1. Trace gases measurements

In the framework of the IASI mission, we have developed processing tools to analyze Level 1 data (geolocated radiance spectra) to retrieve Level 2 products (trace gas contents at the measurement locations) and enhanced Level 3 products (global distributions). We have now three different retrieval codes available (SA-NN, LARA, Atmosphit), which were optimized on different criteria (processing time, number of species retrieved at the same time, vertical resolution). Each code was validated against aircraft and balloon-borne [Té *et al.*, 2002] atmospheric spectra, and a detailed intercomparison exercise was organized using the IMG/ADEOS radiance measurements [Clerbaux *et al.*, 2002]. The IMG data were used to evaluate the performance of the retrieval codes developed for IASI and allowed to derive global distributions for CO [Barret *et al.*, 2005], O<sub>3</sub> [Coheur *et al.*, 2005], and CH<sub>4</sub> [Clerbaux *et al.*, 2003].

The SA-NN retrieval scheme, based on a multilayer neural network that uses a gradient descent back-propagation, provides near real time total column contents for ozone, CO and methane [Turquety *et al.*, 2004]. The LPMA-LARA retrieval algorithm, composed of a radiative transfer model and a Levenberg-Marquardt minimization algorithm, allows simultaneous retrieval of the column amounts of more than 8 species, surface temperature, surface emissivity, and instrumental spectral response function. The ULB-Atmosphit code relies on the optimal estimation method and allows retrieving the vertical profiles of molecules with a significant degree of vertical resolution, and with a full characterization in terms of error and information content analyses [Coheur *et al.*, 2005].

All three algorithms will be applied to the analysis of IASI spectra in order to retrieve O<sub>3</sub>, CO, CH<sub>4</sub> and HNO<sub>3</sub> from the radiance spectra, and to provide the required information in terms of vertical sensitivity and accuracy. When the AURA/TES spectra will be available, a direct comparison of Level 1 radiance spectra for co-located measurements will be possible, and the retrieved products from both instrument using a common algorithm will be undertaken. The differences with the Level 2 IASI official products will be analyzed.

### 1.2 Detection of tropospheric aerosol.

Occurrences of significant aerosol concentrations such as Sahara dust, biomass burning or fresh

volcanic aerosol affect nadir infrared radiance measurements. Previous studies at IASB-BISA, and retrieval algorithm developments for IASI have focused on the effects of boundary layer aerosols above the sea. This work will be continued to take into account aerosol located at higher altitudes in the free troposphere and above land, including the impact of the land surface emissivity. The aerosol retrieval algorithm will also retrieve surface emissivity and surface temperature. It will be applied to the IASI spectral radiances, using IASI L2 data for molecular species and METOP meteorological data.

### 1.3 Detection of SO<sub>2</sub> volcano plumes

The emissions of gases and particles associated with intense volcanic eruptions can ascend up to the stratosphere where they modify the mean temperature and affect the transport and the chemistry. Studies have already shown the contribution of the satellite observations for the early detection and the characterization of the volcanic eruptions. The method currently developed to monitor the volcanic emission plumes using the IASI radiances relies in part on the measurement of the increase of SO<sub>2</sub> and OCS concentrations. Further analysis includes the characterization of the emission plume in terms of nature and quantity of the emitted gases and aerosols and the follow-up of the plume with respect of chemistry and dynamics.

## 2. Tropospheric chemistry

### 2.1 Chemical composition

The photochemical production of ozone is intense in the tropics due to the combination of high insolation and large emissions of ozone precursors (CO, NO<sub>x</sub>, hydrocarbons) resulting from biomass burning, biogenic emissions and lightning. Due to rapid population and economic growth in the tropical regions, future changes in ozone are mainly predicted in the tropics. It is therefore crucial to provide a better understanding of the budget of ozone and related species in order to quantify the impact of human activities in the tropics. Important but currently unresolved science issues include the role of biomass burning, the role of convective clouds, and the photochemistry leading to ozone production, in particular the influence of nonmethane hydrocarbons.

After evaluation against METOP measurements, the LMDz-INCA model [Hauglustaine *et al.*, 2004] will be constrained by measured concentrations of ozone precursors through data assimilation techniques, in

order to investigate the photochemical budget of ozone in the tropics and quantify the production and destruction terms as a function of altitude, latitude and season. In the assimilation procedure, IASI and GOME2 observations will be fed into the LMDz-INCA model (using the ECMWF reanalysis) and properly weighted with model-calculated values in order to obtain an improved description of the ozone precursors (and ozone itself) in the tropics. As the model describes the time evolution of the fields, the assimilation procedure will result in global synoptic maps of chemical species at any specific time [Clerbaux *et al.*, 2001]. Benefiting from the simultaneous measurements of different species by METOP, the concentrations of these species will be photo-chemically consistent with each other. The procedure will thus provide additional information about chemical species which are not directly observed (OH, photochemical budgets) or on important physical processes for the tropics parameterized in the model (convection, biomass burning and biogenic emissions, lightning emissions).

## 2.2 Emission estimates

In this task, we will take advantage of the simultaneous measurements of several important tropospheric ozone precursors by the METOP platform in order to derive improved emissions estimates for these compounds, using inverse modeling techniques. The measurements of interest for that purpose are the CO, CH<sub>4</sub> distributions (IASI), and the NO<sub>2</sub> distributions (GOME2). Finally, the tropospheric ozone profiles measured by IASI will be used to evaluate the improvement in the CTM-calculated ozone distributions brought by the optimization of the ozone precursors emissions.

For the optimization of the ozone precursors emissions, several techniques will be implemented and compared. The IMAGES model and its adjoint will be used in a 4d variational approach. This technique has already been applied to ground-based network measurements, as well as to the GOME NO<sub>2</sub> and MOPITT CO vertical columns. Muller and Stavrou [2005] have shown that this technique makes it possible to simultaneously optimize the emissions of several compounds, while taking their chemical interactions into account. The MOZART model will be used in an optimal interpolation setup. Petron *et al.* [2004] have successfully applied this method to derive optimized CO surface emissions for the globe using the MOPITT CO. The results of both studies have been validated by comparisons with independent observations provided by aircraft campaigns and in situ surface measurements. Inverse modeling techniques also provide a posteriori

uncertainties on the emission estimates. All available observations will be used to further constrain chemical species budgets and to infer seasonal and interannual changes in regional emissions.

## 3. Air quality studies

The unprecedented horizontal and vertical resolution of IASI observations and the continuity in observations from GOME-2 make satellite data from METOP extremely useful for air quality studies. The conjoint use of satellite observations and model simulations through assimilation and inverse modelling techniques should lead to a significant improvement in air quality modelling. We will use the CHIMERE transport-chemistry model [Vautard *et al.*, 2005], already used for operational continental scale air quality forecast (<http://www.prevoir.org>). Proposed work proposed is organised along 4 lines: intensive observation - simulation comparisons (1), inverse emission modelling (2), evaluation of model uncertainties (3) and improvement of forecast skill through assimilation of METOP observations (4).

### 3.1 Intensive observation - model comparisons

We will perform intensive comparisons between METOP observations and CHIMERE simulations. A specific version of CHIMERE coherent with the horizontal resolution of satellite observations and extended up to the troposphere will be set-up in order to optimally take advantage of the full range of information available in METOP observations. This also implies to apply a specific parameterization for the convective processes. These adaptations will be checked by comparing the output of the model over Western Europe with surface observations (O<sub>3</sub>) and airborne measurements from the MOZAIC campaign (O<sub>3</sub>, CO).

### 3.2 Inverse emissions modelling

We will use METOP observations to derive emissions for CO, NO<sub>x</sub> and eventually hydrocarbons (from HCHO observations) for several target regions (Europe, East Asia, etc.) and more specifically large urban agglomerations in these areas. Variational techniques will be applied for that task, but also more simple and robust iterative methods. In particular, the continuity in GOME and GOME-2 NO<sub>2</sub> data will allow to derive the temporal evolution for regions with rapidly evolving emissions (Eastern Europe, East Asia).

### 3.3 Evaluation of model uncertainties

This action will consist in using METOP data to constrain the model uncertainties [Beekmann and Derognat, 2004] and in particular uncertainties in the simulation of emission reduction scenarios. Bayesian Monte Carlo analysis (BMC) will be applied in order to assess the overall uncertainty in model output due to the uncertainty in model input parameters (emissions, meteorology, rate constants, etc.) and model processes and will make use of combined observations of primary (CO, NO<sub>2</sub>) and secondary pollutants (O<sub>3</sub>, HCHO).

### 3.4 Improvement of forecast

We will quantify the improvement of forecast skill (3 days in advance) of CHIMERE through the use of assimilated METOP observations as initial conditions. Recent studies have already shown that assimilating surface ozone data significantly improves ozone short-term forecasts [Blond and Vautard, 2004]. The added value of METOP products in terms of performance will be measured by comparison with ground based data from air quality survey networks. Different methods will be applied to combine observations and simulations (Optimal Interpolation, Kriging, 3D-Var).

## 4. Integration of different instruments and satellite data

We propose to extend the analysis by integrating satellite data (METOP and AURA) with observations from ground-based stations and airborne instruments, as well as global and regional model analysis. Such coupled analysis would allow an optimal use of the available data, taking advantage of the good precision and vertical resolution of the in situ data, and of the good spatial and temporal coverage of the satellite observations.

We will perform data validation of different satellite data retrieval and comparison with ground-based and in situ observations, using atmospheric chemistry and transport models to couple the data from different platforms taking into account the different characteristics of the instruments. This aims to provide an optimal use of the available information for tropospheric chemistry study, to improve our knowledge and evaluate of the current emission inventories, and to understand the intercontinental transport of pollution and its impact on air quality.

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