MONITORING THE SPATIAL DISTRIBUTION AND THE EVOLUTION OF TROPOSPHERIC OZONE WITH IASI IN EUROPE AND EASTERN ASIA: NEW POSSIBILITIES FOR CHEMICAL AND TRANSPORT MODEL EVALUATION

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Abstract

Tropospheric ozone plays a major role in air quality by affecting human health and causing damages to ecosystems. In this framework, our main concern is to regionally characterize major pollution events and the inter-annual evolution of tropospheric ozone. For our studies, we primarily use IASI space-borne observations for monitoring daily distributions of ozone in the lower troposphere from the regional to the continental scale. We provide a cross-analysis of the observed ozone distribution with model simulations. The ability of the models to reproduce the observed ozone distribution and its temporal variations is evaluated. The observations are used to assess the models' weaknesses and their sources. We focus on two regions: Eastern Asia and Europe. IASI observations of ozone over Eastern Asia are correlated with the population density distribution and then can be, at least partly, attributed to strong pollution in this region. Comparison with the simulations of the ECHAM model for April and May 2008 reveals differences between the observed and simulated offshore transport pathways from China. The lack of knowledge of Chinese emissions also limits the capability of the model to reproduce the magnitude of ozone over the main source regions. In Europe we consider in more details the Mediterranean basin which is a region very sensitive to the transport of pollution emitted in the North of Europe. IASI ozone observations are here compared with the regional model CHIMERE during different summers. The main features observed are a fairly good agreement in the Eastern part of the basin (near Italy) but a systematic underestimation of the model in the Western part of the basin (near Spain).

INTRODUCTION

Ozone is a key species of tropospheric chemistry affecting the troposphere's oxidative capacity and is a well-known pollutant with significant impact on health and vegetation [Seinfeld and Pandis, 1997]. Ozone is also an important greenhouse gas with large radiative forcing in the upper troposphere [Fishmann et al., 1979]. Monitoring of tropospheric ozone is essential to quantify its sources, transport, chemical transformation [Clerbaux et al., 2003] and to evaluate and improve models used for climate and pollution modelling.
Tropospheric ozone concentrations are mainly measured at surface level using national operational networks and vertical information is for the most part provided at selected sites by meteorological balloon sondes or during dedicated aircraft campaigns. In addition to these in situ measurements, satellite observations in the nadir geometry are very promising because of their large spatial coverage. However, tropospheric ozone retrieval from satellite observations is a challenging task because most of the ozone is contained in the stratospheric ozone layer. The first satellite measurements of tropospheric ozone have been obtained using ultraviolet-visible sounders [e.g. Fishmann et al., 2003, Liu et al., 2005] but they have some limitations in the mid- and high latitudes. The recent development of infrared nadir sounders allows accurate measurements of tropospheric ozone, with the advantage that measurements are also possible during the night. The first demonstration of tropospheric ozone column retrieval has been done using the IMG

(Interferometric Monitor for Greenhause gases) instrument [Turquety et al., 2002, Coheur et al., 2005]. More recently, the TES (Tropospheric Emission Spectrometer) instrument has provided measurements of tropospheric ozone [Worden et al., 2007] with applications to air quality modelling [Jones et al., 2008] and climate [Worden et al., 2008]. Finally more recently, using measurements from the IASI (Infrared Atmospheric Sounding Interferometer) instrument aboard the European Metop-A satellite (launched in October 2006), enhanced tropospheric ozone during the heat wave over Europe in summer 2007 has been observed [Eremenko et al., 2008] as well as the seasonal and daily variations of ozone at the regional scale of Chinese megacities during 2008 [Dufour et al., 2010].

THE IASI INSTRUMENT

The IASI (Infrared Atmospheric Sounding Interferometer) [Clerbaux et al., 2007] is a nadir viewing spectrometer onboard the MetOp-A satellite and was designed for operational meteorology. The MetOp-A satellite launched in October 2006 flies in a polar sun-synchronous orbit (about 800 km altitude) and crosses the equator at two fixed local solar times 9:30 am (descending mode) and 9:30 pm (ascending mode). The IASI instrument is a Fourier-transform spectrometer with a 2 cm optical path difference covering the 645-2760 cm^{-1} spectral range. The apodized spectral resolution is 0.5 cm^{-1} (Full-Width at Half-Maximum). The radiometric accuracy in noise-equivalent radiance temperature at 280 K is 0.28 K at 650 cm^{-1} and 0.47 K at 2400 cm^{-1}. IASI measures the thermal infrared radiation (TIR) emitted by the Earth's surface and the atmosphere. The instrument scans the surface perpendicular to the satellite's flight track with 15 individual views on each side of the track. The distance between two successive overpasses is 25° in longitude (i.e. 2800 km at the equator). For latitudes higher than 45° in latitude, the footprints of two successive overpasses overlap. At the nadir point, the view size is 50×50 km^2. The view is composed of 4 individual ground pixels with 12 km diameter each. The maximum scan angle of 48.3° from the nadir corresponds to coverage for one swath of about 2200 km in the direction perpendicular to the satellite's track.
In addition to meteorological products (surface temperature, temperature and humidity profiles, cloud information), the IASI instrument provides distributions of several trace gases (e.g. O_3, CO) [Eremenko et al., 2008, Turquety et al., 2009].

OZONE RETRIEVAL METHOD

The ozone products from IASI (mainly partial tropospheric columns) presented in this paper are based on profile retrieval of ozone. The retrievals are performed using the radiative transfer model KOPRA (Karlsruhe Optimised and Precise Radiative transfer Algorithm, [Stiller, 2000]) and its inversion module KOPRAFIT, both adapted to the nadir-viewing geometry. A constrained least squares fit method using an analytical altitude-dependent regularization is used. The regularization method applied as well as the error calculations are detailed in [Eremenko et al., 2008]. To summarize, the regularization matrix is a combination of zero, first and second order Tikhonov [Tikhonov, 1963] constraints with altitude-dependent coefficients. The coefficients are optimized to both maximize the degrees of freedom (DOF) of the retrieval and to minimize the total error on the retrieved profile. The analysis of IASI data is performed in three steps. First, the effective surface temperature is retrieved from selected windows between 800 and 950 cm^{-1} considering a blackbody with an emissivity equal to unity. In the second step, the atmospheric temperature profile is retrieved from CO_2 lines in the 15 μm spectral region and using the ECMWF profiles as a priori. In the third step, the ozone profiles are retrieved from seven spectral windows in the 975-1100 cm^{-1} region that avoid strong water vapor lines. The spectroscopic parameters of ozone are from the MIPAS database [Flaud et al., 2003] and from HITRAN 2004 for the other species. The a priori profile used during the retrieval is compiled from the climatology of Mcpeters et al. [2007] and is the same independently of the time and the location of the observations (in the midlatitudes). Note that before the retrieval, the IASI spectra are filtered for cloud contamination. Only spectra for clear sky conditions are considered. A quality flag is also applied to the retrieved products to discard unphysical results.
Capabilities to monitor lower tropospheric ozone for air quality concerns with the developed retrieval method have been demonstrated [Eremenko et al., 2008,Dufour et al., 2010]. Performances of the retrieved ozone product, especially in terms of vertical sensitivity, have been extensively studied [Dufour et al., 2011, Galkina et al., 2011]. Figure 1 shows the degrees of freedom (DOF) and the altitude of the maximum of sensitivity for summer midlatitudes for three ozone partial columns: the lower tropospheric column from surface up to 6 km (LT), the tropospheric column from surface up to 12 km (TROPO) and the upper tropospheric lower stratospheric column from 8 to 16 km (UTLS). The ozone product presents DOF in the LT between 0.5 and

0.7 during summer midlatitudes with a maximum of sensitivity between 3 and 4 km on average leading to the possibility to discriminate between ozone in the lower troposphere and in the upper troposphere [Dufour et al., 2010]. Moreover, a recent validation exercise [Dufour et al., 2011] using ozone sonde measurements as reference has been conducted over 2008 and shows that the biases are smaller than 2.5% in the northern midlatitudes except in the UTLS where the bias is larger (~15%). In the tropics, the ozone partial column is underestimated by about 6% in the lower troposphere and overestimated (9%) for the entire troposphere [Dufour et al., 2011].

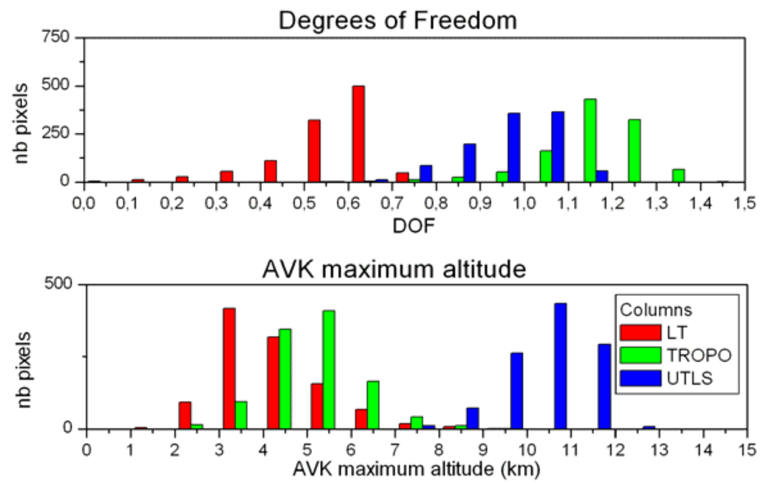

***Figure 1*: Top - Distribution of degrees of freedom for different ozone partial columns: surface-6km (LT), surface-12km (TROPO), and 8-16 km (UTLS) for summer midlatitudes. Bottom - Distribution of the altitude of the maximum of the averaging kernels for the same partial columns.**

We have additionally analyzed the impact of aerosols as a potential source of error in the ozone retrieval. Preliminary results show that urban aerosols do not affect the ozone retrieval even in case of large loading. The biomass burning aerosols start to affect the ozone retrieval only if the aerosol layer is sufficiently high (> 5 km). On the contrary, dust aerosols always affect the ozone retrieval. A joint retrieval including ozone and aerosols retrieval is necessary to overcome the introduced bias.

DO CHEMICAL TRANSPORT MODELS REPRODUCE THE TEMPORAL AND SPATIAL VARIABILITY OF OBSERVED OZONE FIELD?

In this section, we evaluate the ability of the models to reproduce the observed ozone distribution and its temporal variations. We focus on two regions: Eastern Asia and Europe.

EASTERN CHINA: IASI OBSERVATIONS AGAINST ECHAM MODEL

We compare IASI ozone partial columns in the lower troposphere (0-6km) observed in April and May 2008 over Eastern Asia with the columns simulated with the ECHAM model [Jöckel et al., 2006]. For the comparison, the model profiles are smoothed with the averaging kernels of the observations. Figure 2 displays the monthly observed and simulated ozone columns for May 2008. The IASI observations are regrided at the horizontal resolution of the model (1.1° in latitude and longitude). The model reproduces the main source regions observed with IASI: North China Plain, Yangste River region and Chengdu region. The main difference between the simulated and observed ozone arises for the transport over sea. The offshore transport is overestimated by about 10% in the midlatitudes whereas it is underestimated by about 10% in the tropics. The lack of vertical representativity of the model associated with differences in the altitude of the transported layer of ozone is one possible explanation for this discrepancy.

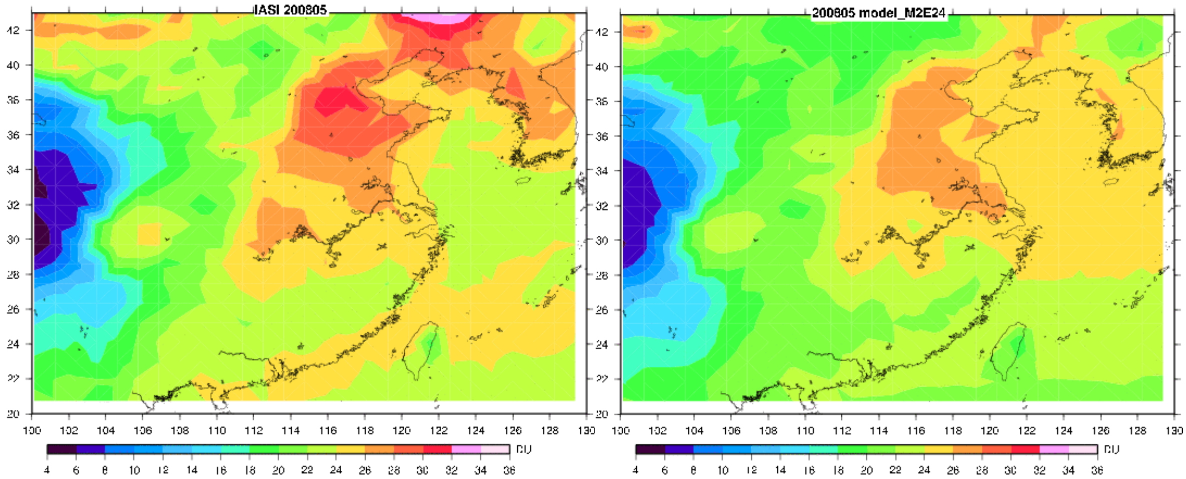

Figure 2: **Monthly ozone partial columns from surface to 6 km observed with IASI (left) and simulated with the ECHAM model (right) in May 2008 over Eastern Asia.**

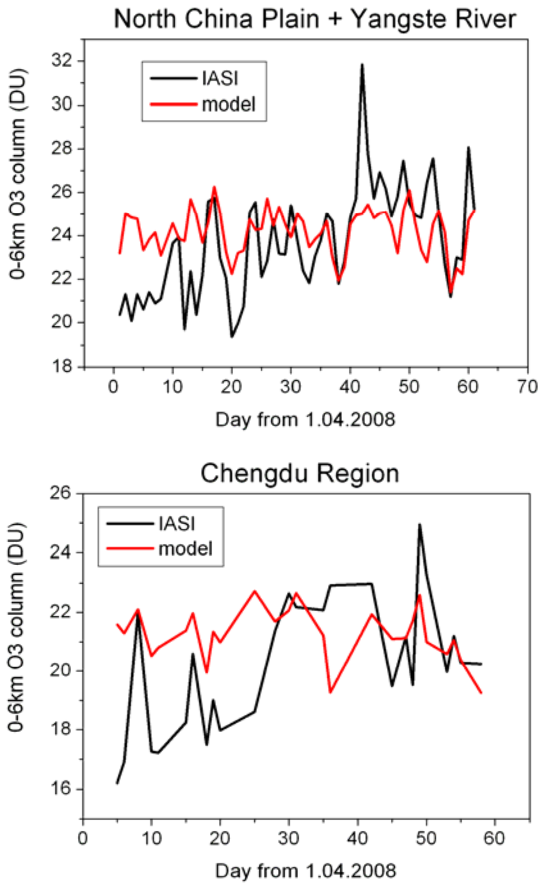

Figure 3: **Two-months timeseries of ozone partial columns observed with IASI and simulated with the ECHAM model starting from April 1st, 2008. The daily columns are spatially averaged over the North China Plain and Yangste river region (top) and over the Chengdu region (bottom)**

Figure 3 shows the daily variations of the 0-6km ozone column over the two main source regions - North China Plain and Yangste River, and the Chengdu region – from the beginning of April 2008 to the end of May 2008. On a monthly basis the model overestimates ozone in the lower troposphere in April for the two considered regions (10% and 14% respectively) whereas the difference in May is much smaller (4% and 2% respectively). The daily variations of lower tropospheric ozone observed with IASI are poorly reproduced by the model (correlation < 0.5). The differences are likely due to the uncertainty in the emissions and their temporal variations.

EUROPE: IASI OBSERVATIONS AGAINST CHIMERE MODEL

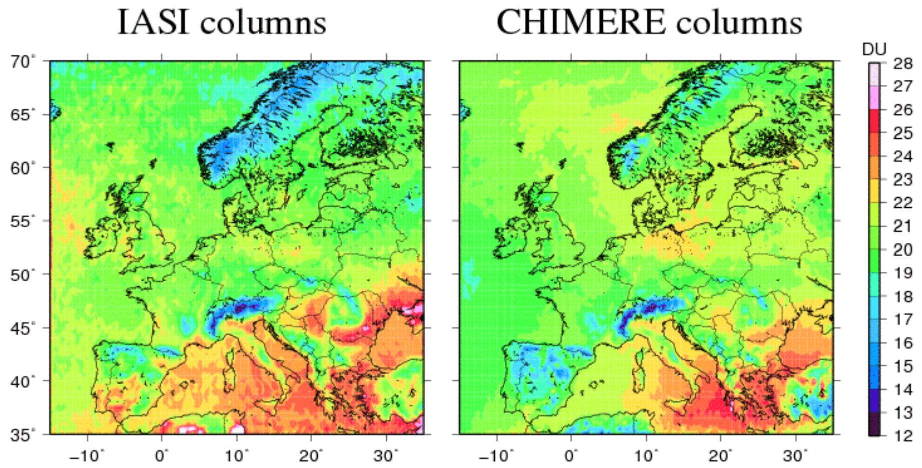

Figure 4: **Observed and simulated ozone partial columns over Europe. The columns are averaged over the three summer months of 2008.**

We compare IASI ozone partial columns in the lower troposphere (0-6km) observed during three summers (2007-2009) over Europe with the columns simulated with the CHIMERE model [Schmidt et al., 2001]. Results for summer 2008 are presented here. For the comparison, the model profiles are smoothed with the averaging kernels of the observations. Figure 4 displays the observed and simulated ozone partial columns in the lower troposphere averaged over summer 2008. Both the observations and the simulations show a North West / South East gradient with larger ozone amount in the South East of Europe. In the South of Europe and especially over the Mediterranean basin, a West/East gradient is observed and simulated. However, larger ozone columns are observed on the western part of the basin compared to the simulation. One hypothesis for this difference is the role of the boundary conditions used to force the model. We investigated the influence of using monthly or daily boundary conditions. As expected, simulations using daily boundary conditions allow a better representation of the daily variations by the model: the temporal correlation for the western part of the Mediterranean basin is for example 0.5 with monthly boundary conditions against 0.7 with daily boundary conditions. On the contrary, the use of daily boundary conditions slightly increases the bias from 6.6% to 8.5% over the western part of the basin and from 2.7% to 5.4% over the eastern part. Detailed investigations on the impact of boundary conditions, especially on the consistency of the monthly and daily conditions are necessary to clearly assess their impact.

CONCLUSION

The characterization and the detailed validation of the IASI tropospheric ozone product summarized here show the good performances of the product to investigate lower tropospheric ozone variations. We showed that the IASI ozone observations are very promising to evaluate the models ability to reproduce temporal and spatial variations of tropospheric ozone from the regional to the continental scale and also to investigate the reasons of the discrepancies.

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