



LmT ozone
monitoring from
MAGEAQ-TIR

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Monitoring the lowermost tropospheric ozone with thermal infrared observations from a geostationary platform: performance analyses for a future dedicated instrument

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Abstract

In this paper, we present performance analyses for a concept geostationary observing system called MAGEAQ (Monitoring the Atmosphere from Geostationary orbit for European Air Quality). The MAGEAQ mission is designed to include a TIR spectrometer and a broadband VIS radiometer; in this work we study only the TIR component (MAGEAQ-TIR). We have produced about 20 days of MAGEAQ-TIR tropospheric ozone pseudo-observations with a full forward and inverse radiative transfer pseudo-observations simulator. We have studied the expected sensitivity of MAGEAQ-TIR and we have found that a completely independent surface -6 km ozone column (about 1.0 DOF (degrees of freedom) and maximum sensitivity at about 3.0 km, on average), as well as a partially independent surface -3 km ozone column (about 0.6 DOF and maximum sensitivity at about 2.5 km, on average) can be achieved. Then, we have compared the tropospheric ozone profiles and the lower (surface -6 km) and lowermost (surface -3 km) tropospheric ozone column pseudo-observations to the target pseudo-reality, produced with the MOCAGE (MODèle de Chimie Atmosphérique à Grande Echelle) chemistry and transport model. We have found very small to not significant average biases ($< 1\%$ in absolute value, for the surface -6 km TOC, and about -2 to -3% , for the surface -3 km TOC) and small RMSEs (about 1.3 DU (5%), for the surface -6 km TOC, and about 1.5 DU (10%), for the surface -3 km TOC). We have tested the performances of MAGEAQ-TIR at some selected small ($0.2^\circ \times 0.2^\circ$) urban and rural locations. We have found that, while the vertical structures of the lower tropospheric ozone pseudo-reality are sometimes missed, MAGEAQ-TIR lower and lowermost column pseudo-observations follow stunningly good the MOCAGE column pseudo-reality, with correlation coefficients reaching values of 0.9 or higher. Unprecedented retrieval performances for the lowermost tropospheric ozone column are shown. In any case, our MAGEAQ-TIR pseudo-observations are only partially able to replicate the MOCAGE pseudo-reality variability and temporal cycle at the very lowest layers (surface and 1 km altitude), especially at Southern European urban locations, where the photochemistry signal is partially

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missed or shifted at higher altitudes. Temporal artifact on the daily cycle are sometimes observed. Stratospheric-to-tropospheric exchanges during short time periods (of the order of 1 day) are detected by the MAGEAQ-TIR pseudo-observations.

1 Introduction

The ozone is an important tropospheric constituent, due of its threefold effect. First, it is a greenhouse gas in the upper troposphere (Shindell et al., 2009). Second, it is a precursor of the hydroxyl radical, and thus it can modulate the oxidizing capacity of the troposphere (Fuglested et al., 2011). Finally, it is a secondary pollutant at the lowest altitudes, where it can adversely interact with the biosphere and human health (Amann et al., 2005). Tropospheric ozone is formed with photochemical reactions involving carbon monoxide, methane and other volatile organic compounds in the presence of nitrogen oxides and sunlight (Derwent et al., 1996) or it is imported from the stratosphere due to stratosphere–troposphere exchanges (STEs) (Hsu and Prather, 2009). Due to its role as a pollutant, the ozone concentration in the lower troposphere is one of the most important atmospheric variables for air quality (AQ) monitoring (WMO, 2006).

The short term variability (of the order of hours) of the ozone concentrations in the lowermost (here defined as altitudes lower than 3 km) troposphere can be significant, due to the heterogeneity of the sources and sinks of ozone precursors at the lowest altitudes, as well as transportation and mixing processes. Satellite observations of tropospheric ozone provide a valuable complement to in situ measurements and atmospheric modelling to draw a more comprehensive picture of pollution processes that can have a relevant impact on the biosphere (The Integrated Global Atmospheric Chemistry Observation Theme Team, 2004). Monitoring AQ-relevant variables, as the lower tropospheric ozone concentrations, from space is of a great importance (Martin, 2008). Currently, the tropospheric ozone information is derived from nadir satellite instruments flying in a low Earth orbit (LEO), exploiting different spectral bands and multi-spectral approaches, like the ultraviolet (e.g. Fishman and Larsen, 1987; Munro

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et al., 1998; Ziemke et al., 2003; Liu et al., 2005, 2010; Schoeberl et al., 2007; Sellitto et al., 2011; Di Noia et al., 2012), the synergy ultraviolet/visible (UV/VIS) (Sellitto et al., 2012a, b), the thermal infrared (TIR) (e.g. Worden et al., 2007a; Eremenko et al., 2008), the synergy UV/TIR (Fu et al., 2013; Cuesta et al., 2013). The complementarity of the observations from space, with respect to in situ measurements, lies in their dense spatial sampling. The drawbacks are a generally scarce sensitivity at the lowest altitudes and a limited revisit time. Satellite instruments flying in a LEO orbit and operating in the TIR spectral range, like IASI (Infrared Atmospheric Sounder Interferometer) (Eremenko et al., 2008; Boynard et al., 2009; Dufour et al., 2010, 2012) or TES (Tropospheric Emission Spectrometer) (Bowman et al., 2006; Worden et al., 2007a; Osterman et al., 2008) are regarded as more sensitive to lower tropospheric ozone than present UV/VIS instruments. They are able to provide the tropospheric ozone information with up to about 1–2 degrees of freedom (DOF) and a total error of 8–14 % in the troposphere, and up to 0.6 DOF and a total error of 10–16 % in the lower troposphere (surface –6 km) (Dufour et al., 2012). The consequence is that it is not possible, in general, to obtain an independent information on the lower tropospheric ozone, except when in favourable conditions, e.g. with high positive thermal contrasts (Dufour et al., 2010). In addition, a LEO orbit cannot provide a sufficient time sampling to observe phenomena occurring at sub-daily to hourly time scales. To this aim, a better choice would be a geostationary Earth orbit (GEO) observing system, whose revisit time is of the order of hours. Space agencies worldwide have recently started the planning of GEO missions dedicated to AQ monitoring, using both TIR and UV/VIS instruments. The final goal is to have a constellation of GEO observing systems, to monitor both the local to continental, and the transcontinental processes, with a high temporal sampling, in a coordinated manner (Atmospheric Composition Constellation, 2011). The GEO-CAPE (GEOstationary Coastal and Air Pollution Events) (Natraj et al., 2011; Fishman et al., 2012), which has recently been recommended for launch in an intermediate time-frame by the US National Research Council, is planned to observe lowermost tropospheric ozone over North America. In the concept of GEO-CAPE, the

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band near 10 μm , MAGEAQ-TIR is expected to be well adapted to retrieve the lower tropospheric ozone information, and then to better complement with the MTG-UVN, with respect to IRS. A first comparison of the MAGEAQ-TIR and an IRS-like instrument performances for lower tropospheric ozone monitoring has been performed by Claeysman et al. (2011b). In comparison with Claeysman et al. (2011b), our study is based on a full direct and inverse radiative transfer modelling, based on a state-of-the-art inversion algorithm, optimized for lower tropospheric ozone retrievals (Eremenko et al., 2008). In addition, we have done a punctual simulation of the observation geometry of MAGEAQ-TIR. We provide a statistical characterization of the lower tropospheric ozone sensitivity of the MAGEAQ-TIR and of the a-posteriori retrieval error, and a height-resolved analysis of the MAGEAQ-TIR performances.

In Sect. 2 we describe the dataset produced for this study and the simulator used to generate the MAGEAQ-TIR pseudo-observations. In Sects. 3 and 4 we assess the vertical sensitivity and the accuracy of the MAGEAQ-TIR pseudo-observations. In Sect. 5 we study how MAGEAQ-TIR can reproduce the temporal variations of the ozone columns and in Sect. 6 we investigate on how it is able to reproduce the vertical distribution of the ozone pseudo-reality. Section 7 gives conclusions.

2 Datasets description

2.1 Instrument configuration: the MAGEAQ-TIR instrument

The pseudo-observations simulator described by Sellitto et al. (2013a) is adapted to simulate the MAGEAQ observing system. MAGEAQ is a concept multi-spectral geostationary observing system, which has been proposed in response to the Call for Proposal for Earth Explorer Opportunity Mission EE-8 (Peuch et al., 2010; Claeysman et al., 2011b,a; Lahoz et al., 2012). MAGEAQ is a GEO payload, specifically designed for AQ monitoring over Europe. Its primary scientific goal is to provide high frequency observations of tropospheric ozone and carbon monoxide, with a significant sensitiv-

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ity to the lowermost troposphere. The lowermost tropospheric ozone is observed by means of a TIR Fourier transform spectrometer and a broadband VIS radiometer. This latter is intended to complement the TIR spectrometer by giving surface information. In our work, only the TIR component of the MAGEAQ mission is considered. We consider the outcomes of the Phase 0 study carried out by EADS (European Aeronautic Defence and Space Company)-Astrium and the MAGEAQ Science Team (Peuch et al., 2010), and we set-up our simulations accordingly. A high spectral resolution, with spectral sampling interval of 0.05 cm^{-1} , and a small radiometric noise, with noise equivalent spectral radiance of $6.04 \text{ nW (cm}^2 \text{ sr cm)}^{-1}$ (3 times better than, e.g. IASI) are here considered. It has been evaluated that this instrumental configuration is well adapted to AQ monitoring in terms of the lowermost tropospheric ozone (Claeyman et al., 2011b,a). Table 1 displays the main specifications of the Phase 0 threshold and target configuration of MAGEAQ-TIR (from Peuch et al., 2010). As for the observation geometry, we have considered the Phase 0 threshold configuration of MAGEAQ-TIR, i.e. a horizontal resolution of $15 \text{ km} \times 15 \text{ km}$ at sub-satellite point, a sub-satellite point at latitude = 0 and longitude = 0, and a field of regard of (15° W – 35° E , 35° N – 65° N). It should be noted that previous performance assessments of MAGEAQ-TIR have been performed on the coarser grid of the nature run (Claeyman et al., 2011b,a). We have simulated a hourly revisit time (so, the Phase 0 target configuration of MAGEAQ-TIR), for the period 5–28 August 2009. Please note that, in the subsequent analyses, some hourly data are missing due to failure in our data processing system.

2.2 Pseudo-observations simulator

We have produced MAGEAQ-TIR pseudo-observations with the modular simulator used by Sellitto et al. (2013a) to simulate the IASI and IASI-NG (New Generation) observing systems. The only change with respect to the set-up described in this work is the definition of the observing system technical specification, observing geometry and revisit time. Our pseudo-observations simulator provides full forward and inverse

radiative transfer calculations, based on the KOPRA (Karlsruhe Optimized and Precise Radiative transfer Algorithm) radiative transfer model (RTM) (Stiller et al., 2002).

The pseudo-reality (or nature run) is produced by means of the MOCAGE (MODèle de Chimie Atmosphérique à Grande Echelle) chemistry and transport model (CTM) (Dufour et al., 2005). The MOCAGE CTM simulates physical and chemical processes affecting the main chemical species in the troposphere and the stratosphere. MOCAGE uses the RACMOBUS chemical scheme, which is a combination of the REPROBUS (Lefèvre et al., 1994) and the RACM scheme (Stockwell et al., 1997). The horizontal grid of the MOCAGE simulations is $0.2^\circ \times 0.2^\circ$ over Europe. It produces outputs as trace gases vertical concentration profiles at 47 hybrid vertical levels (from surface up to about 35 km), with a vertical resolution of about 200 m into the lower troposphere, and up to about 1 km into the stratosphere. We have used a run of MOCAGE for the period 5–28 August 2009.

The outputs of MOCAGE are then resampled at a coarser vertical grid, 1 km in the troposphere and lower stratosphere, up to 2 to 5 km at higher altitudes, as required for the subsequent radiative transfer calculation. The forward radiative transfer is simulated by means of the KOPRA RTM, which takes as inputs the MOCAGE pseudo-reality and gives as outputs the radiance spectra as observed by the selected observing system. The simulated spectra are finally inverted by means of the KOPRAfit module (Hoepfner et al., 2001), to obtain the ozone profile pseudo-observations. Our inversion scheme is based on an altitude-dependent Tikhonov–Phillips regularization method, which, as well, uses the KOPRA RTM. The inversion algorithm, which is aimed to the maximization of the sensitivity and accuracy in the lower troposphere, is thoroughly described by Eremenko et al. (2008). In our simulations, we have used the same spectral micro-windows used by, e.g. Eremenko et al. (2008) with real IASI measurements. The inversion algorithm operates at seven spectral micro-windows, in the region of $975\text{--}1100\text{ cm}^{-1}$, for avoiding carbon dioxide and water vapour impact on ozone retrievals. The KOPRAfit also provides the averaging kernels (AKs), that are used to characterize the vertical sensitivity of the retrievals. McPeters climatology (McPeters et al., 2007) is

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used for the a priori information. As done by Sellitto et al. (2013a), we have used two different ozone a priori profiles, depending on the tropopause altitude, to limit numerical instability and aberrant oscillations in the solutions. We have considered tropopauses higher than 14 km as a proxy for tropical airmasses. Consequently, the ozone a priori profile, in these cases, has been chosen as a tropical a priori (yearly climatological profile 20–30° N, from McPeters climatology). For pixels with tropopauses lower than 14 km, a mid-latitude a priori is used (summer climatological profile 30–60° N, from McPeters climatology).

Finally, more than 20 days of MAGEAQ-TIR pseudo-observations, with 1 h revisit time, are produced in the region of interest. The number of the processed pixels is about 15 millions. The complete direct and inverse radiative transfer calculations, which need a significant computation effort, have been performed by means of the European Grid Infrastructure (EGI)-France Grilles supercomputing platform (Eremenko et al., 2012). As done by Sellitto et al. (2013a), the effect of clouds is not considered in the present study. This choice has been made to maximize the statistical population with the available simulated dataset. Please note that in Claeysman et al. (2011b,a) a cloud mask has been used.

3 Vertical resolution and lowermost tropospheric ozone sensitivity

Two useful diagnostic parameters for the evaluation of the vertical sensitivity of satellite retrievals are the degrees of freedom (DOF) and the altitude of the maximum sensitivity. Both the two parameters can be calculated from the averaging kernel (**AK**) matrix. The DOF are the number of independent pieces of information that can be obtained from an observation. The DOF for a partial column are calculated as the trace of the **AK** matrix, up to the top height of the column. The altitude of the maximum sensitivity of a partial column observation can be estimated by calculating the altitude of the maximum of the integrated **AK** for that partial column. Here we concentrate on the tropospheric ozone

partial columns up to 6 and 3 km, hereafter referred to as surface –3 km TOC and surface –6 km TOC.

Figure 1 shows the histograms of the DOF surface –6 km and surface –3 km, and of the altitudes of the maximum sensitivity of the surface –6 km and the surface –3 km TOCs. The histograms are obtained by considering all observations in our dataset. The mean value and the standard deviation of the distributions are also reported in the figure. Our simulations show that an instrument like MAGEAQ-TIR would be able to retrieve the surface –6 km ozone column with more than 1.0 DOF, on average, and a maximum sensitivity at about 3.0 km, so at the centre of the nominal column, which indeed means that the information on the lower tropospheric ozone would be independent on ozone concentrations at higher altitudes. In addition, these results show that the MAGEAQ-TIR measurements have also a significant sensitivity to the surface –3 km TOC, with 0.57 DOF on average, and a peak of sensitivity at about 2.5 km.

To further investigate on the sensitivity of the MAGEAQ-TIR observations, we have studied the DOFs and the peaks of sensitivity, for a number of different situations. We have then partitioned our dataset into different subsets: day and night observations, and land and sea pixels. Observations during daytime hours (hereafter referred to as the DT subset) are those taken in the interval 08:00–19:00 UTC and, conversely, observations during nighttime hours (hereafter referred to as the NT subset) are those taken in the interval 20:00–07:00 UTC. Tables 2 and 3 summarize the mean values of the DOFs surface –6 km and surface –3 km, and the altitude of the peak of the sensitivity for the surface –6 km and surface –3 km TOCs, respectively, for all possible combinations of land/sea and DT/NT. The most marked differences of DT/NT DOFs are for the column surface –3 km, with nearly 20 % higher values, on average, of daytime DOFs, reaching mean values as high as 0.61 for the mean daytime surface –3 km TOC. The DOFs surface –6 km are less sensitive to DT/NT differences, with mean values of 1.19 (+6 %) and 1.12 for the DT and NT subsets. This can be linked to the more important enhancement of the sensitivity at the lowest altitudes, due to the higher thermal contrasts during daytime. The altitude of the maximum sensitivity is at altitudes

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about 400 m lower at DT, for both the surface –6 km and the surface –3 km TOCs. The land/sea differences exhibit a similar behaviour, with a more marked difference for the DOF surface –3 km (+11% for the DT with respect to the NT subset, for the DOF surface –3 km, and +4% for the DT with respect to the NT subset, for the DOF surface –6 km) and a similar small difference, about 200 m, for the altitudes of the maximum sensitivity. It can be noticed that the sensitivity during nighttime over land surface pixels is similar to the sensitivity during daytime and nighttime over sea surface pixels, for both the TOCs surface –6 km and surface –3 km. In those cases, we have found similar mean DOFs (1.11–1.14 and 0.49–0.57, for the DOFs surface –6 km and the DOFs surface –3 km, respectively) and sensitivity only a bit lower (250–300 m and 100–200 m difference, on average, for the TOC surface –6 km and surface –3 km, respectively) for the DT/sea with respect to NT/land and NT/sea. On the contrary, the most important difference of sensitivity is between DT and NT surface –3 km TOCs pseudo-observations taken over land surface pixels. The average DT/land DOF surface –3 km reaches values as high as 0.71, which is about 24 % higher than DT/sea, more than 30 % higher than NT/land and about 45 % higher than NT/sea. The sensitivity of the DT/land surface –3 km TOC pseudo-observations peaks at about 2.1 km, which is about 500, 600 and 700 m lower than DT/sea, NT/land and NT/sea. The differences of sensitivity for the surface –6 km TOC pseudo-observations are less marked, with the average DT/land DOF surface –6 km reaching values of 1.27, which is about 11 % higher than DT/sea, 11 % higher than NT/land and about 14 % higher than NT/sea, and the sensitivity peaking at about 2.7 km, which is about 200, 400 and 500 m lower than DT/sea, NT/land and NT/sea. Following these results we are inclined to consider the DT/NT difference as more important than land/sea surface difference in the determination of the vertical sensitivity of our MAGEAQ-TIR pseudo-observations. This impact is markedly more evident on smaller and lower columns, like the surface –3 km TOC.

To further investigate the vertical sensitivity, especially in terms of the surface –3 km TOC, Fig. 2 shows the integrated AKs for the surface –3 km, 4–6 km and surface –6 km TOCs, averaged over all pixels of our dataset having thermal contrast between 0.0 and

1.0K. The integrated AKs for the surface –3 km and the 4–6 km TOCs are partially overlapped, thus indicating that, on average, the MAGEAQ-TIR would not be able to completely separate the surface –3 km column information. In any case the AK are not completely overlapped, and then it is possible to retrieve a partially independent information. This result is coherent with the average DOF of 0.57, as shown from Fig. 1. The AKs for strongly positive thermal contrast (> 5 K) are only a bit more separated.

4 General statistical analysis of the retrieval accuracy

In the present section, we characterize the accuracy of the MAGEAQ-TIR retrievals, with respect to the MOCAGE pseudo-reality. Tables 4 and 5 report the mean absolute and percent biases and RMSEs of the surface –6 km, and the surface –3 km TOC pseudo-observations, respectively. For both columns, the statistical parameters of the comparison are calculated for the whole ensemble of the data, and, as done previously for the vertical sensitivity, for DT/NT and land/sea surface subsets, with all different possible combinations. The MAGEAQ-TIR surface –6 km TOC pseudo-observations show biases generally smaller than 1 % in magnitude. Also for the MAGEAQ-TIR surface –3 km TOC pseudo-observations the mean biases are small, with an average under-estimation of –2% over the whole dataset. The mean bias of the surface –3 km TOC is mostly driven by the surface type, with values between –1.4 and –1.9 % over land, and between –2.7 and –3.1 % over sea. The RMSEs have a value of 1.28 DU (5.3 %), for the column surface –6 km, and 1.53 DU (10.4 %), for the column surface –3 km. The RMSEs for both columns are mostly driven by the time at which observations are done. We observe no big DT/NT differences over sea, with slightly better performances of the NT subset. On the contrary, we observe very large DT/NT differences over land, with DT RMSEs smaller of 45 and 30 %, for the surface –6 km and surface –3 km TOCs. The smallest values of the RMSEs are found for the DT/land subset. The average DT/land RMSE for the surface –6 km TOC reaches values as low as 0.92 DU (3.8 %), which is about 35 % smaller than DT/sea, about 45 % smaller than NT/land and about 32 %

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smaller than NT/sea. The average DT/land RMSE for the surface –3 km TOC reaches values as low as 1.25 DU (8.8 %), which is about 20 % smaller than DT/sea, about 30 % smaller than NT/land and about 15 % smaller than NT/sea.

To characterize the global dataset in terms of the spatial distribution, Figs. 3 and 4 show the average surface –6 km and surface –3 km TOCs, for the whole dataset, over Europe. The figures display the MOCAGE pseudo-reality, MOCAGE smoothed with the MAGEAQ-TIR **AK**, the MAGAEQ pseudo-observations and the difference between MAGEAQ-TIR pseudo-observations and MOCAGE pseudo-reality, for the whole dataset and for DT-only and NT-only subsets. The average distributions of lower and lowermost tropospheric ozone are in general well caught by the MAGEAQ-TIR pseudo-observations. The MAGEAQ-MOCAGE differences are typically in the interval ± 0.5 DU for the surface –6 km TOC, and ± 1.0 DU for the surface –3 km TOC. For both surface –6 km and surface –3 km, and at both DT and NT, a relatively marked underestimation in the Mediterranean basin is found. Typical underestimations in that area are of the order of 2.0–2.5 DU (~ 7 –8 %) for the surface –6 km TOC, and 3.0–3.5 DU (~ 15 –18 %) for the surface –3 km TOC. This underestimation area touches also a Southern European continental portion during NT.

5 Time series of the lower tropospheric ozone columns

In the present section, we study the capability of the MAGEAQ-TIR pseudo-observations to follow the temporal evolutions of lower (up to 6 km) and lowermost (up to 3 km) tropospheric ozone columns. We concentrate on the local scale and then we select very small regions, of the order of $0.2^\circ \times 0.2^\circ$. To study both urban and remote environments, we have selected six locations: Milan (Italy), Paris (France), Berlin (Germany), Amsterdam (the Netherlands), one marine location West of Norway coast (55° N/ 8° E), Barcelona (Spain). Paris, Berlin and Amsterdam, and Milan and Barcelona, have been chosen to study the performances of MAGEAQ-TIR at Northern and at Southern European urban locations. The marine location West of Norway has

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been selected to study one particular remote marine location, affected by the transport of ozone-polluted airmasses in the period of study. In fact, Forêt et al. (2013), Sellitto et al. (2013a,b) have shown that a peculiar ozone plume phenomena affected this area, during August 2009. The strong values of ozone observed in the boundary layer over the South of France, due to photochemical production, are transported to higher altitudes (up to 1–2 km altitude) and then transported eastwards and northwards. The plume then reached the mentioned marine location on the 20 August after having passed over Paris and, almost at the same time, Amsterdam. Afterwards, the plume is observed over Berlin. Then, to choose these locations gives us the possibility to further analyse the capability of MAGEAQ-TIR pseudo-observations to observe this transport phenomenon. Figures 5 and 6 display the time series of MOCAGE pseudo-reality and MAGEAQ-TIR pseudo-observations of the surface –6 km and surface –3 km TOCs, respectively, over these locations. As a complement to these figures, in Fig. 7 we show the time series for the DOF surface –6 km and surface –3 km at the same locations. The general features of the pseudo-observations time series are stunningly coherent with the pseudo-reality, in particular for the surface –6 km TOC, except for Milan, and, to a lesser extent, Barcelona. This evidences how the retrieval is more problematic over Southern European urban locations. More details on this aspect will be given in Sect. 6, where we show how our pseudo-observations fail to describe the daily cycle at Milan on a height-resolved basis, due to a smaller sensitivity to the lowest layers, where this cycle is more pronounced. This behaviour has a marked effect also on the surface –3 km and, to a lesser extent on the surface –6 km TOCs. For Milan (first panel of Fig. 7), the DOF surface –6 km and surface –3 km has a marked daily cycle, with a maximum/minimum at about 12:00/24:00 UTC. The differences between the maxima and minima can be of the order of the 40 % (minimum 1.0 DOF, maximum 1.4 DOF), for the surface –6 km column, and can reach 100 % (minimum 0.4 DOF, maximum 0.8 DOF), for the surface –3 km column. While the maximum of sensitivity is at about 12:00 UTC, the maximum of the pseudo-reality ozone concentrations at the lowest altitudes is at about 15:00 UTC. This generates a quite strong

artifact cycle at Milan, with a maximum which is shifted back of some hours. This effect is also present in the surface -3 km TOC time series of Barcelona, e.g. for the days 10, 11, 26 and 27 August. The other urban locations has pseudo-reality time series less driven by the daily cycle, and then they are less sensitive to this effect, which is only sporadically observed on the surface -3 km TOCs time series. We quantify the performances of our MAGEAQ-TIR synthetic observations by means of the mean biases, RMSEs and the Pearson correlation coefficients, with respect to the pseudo-reality, at the six locations. These quantities are summarized in Table 6, for the surface -6 km TOCs, and in Table 7, for the surface -3 km TOCs. For the surface -6 km TOC pseudo-observations, the values of the correlation coefficients are in the range 0.93–0.97 for Paris, Berlin, Amsterdam and the North Sea, while it decreases to 0.89 and 0.82 for Barcelona and Milan. The biases are very small ($< 0.35\%$ in magnitude) at the Northern European urban locations, while a more significant systematic underestimation is observed over the North Sea, -0.46 DU (-1.54%), and higher values at the Southern European urban locations, reaching the value of -1.29 DU (-4.25%) at Milan. The near zero bias at the northern urban locations, and the small underestimation over the North Sea, are consistent with the average land/sea biases shown in Table 4. The negative biases at the two Southern European urban locations, in particular at Milan, are a marked anomaly in our dataset. Also the RMSEs show this behaviour, with values in the range 1.08–1.19 DU (4.09–4.62%), except for Milan, which has a mean value of 1.96 DU (6.43%). As for the surface -3 km TOC, the Pearson coefficients have values of 0.83–0.91 over Northern European urban locations, 0.73 at Barcelona and 0.55 at Milan. There are stronger systematic underestimations than for the general characterisation of the dataset, of the order of -3 to -5% , and even higher values for Barcelona (-6.5%) and Milan (-10.82%). The RMSEs are of a similar magnitude as for the general characterisation (1.4–1.7 DU, i.e. 9–10%), except for Milan which exhibits higher values, reaching 2.7 DU (13.5%).

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6 Vertical distribution

Here, we evaluate if the synthetic MAGEAQ-TIR observations can detect different phenomena occurring at and influencing different altitude ranges. In particular, we try to understand to which extent MAGEAQ-TIR may be able to detect enhanced ozone values near the surface and then the photochemistry ozone production signal. Studying only the surface –6 km or even the surface –3 km ozone columns is not sufficient to answer this question, because these columns can be only partially influenced by the phenomena occurring in the boundary layer and this effect may be difficult to sort out from other concurring phenomena, like STEs or transport into the free troposphere.

As for Sect. 5, we focus our analysis on the same small regions: Milan, Paris, Berlin, Amsterdam, the marine location West of Norway coast, Barcelona.

Figure 8 shows the tropospheric ozone concentration profiles pseudo-observations and the pseudo-reality for each site, as well as the mean ozone profile pseudo-observations and pseudo-reality. The MAGEAQ-TIR synthetic observations have very consistent variability and mean value, with respect to MOCAGE pseudo-reality, in the interval 2–7 km. On the contrary, the MAGEAQ-TIR synthetic observations cannot replicate the variability of the pseudo-reality at the lowest two levels, especially at the surface. There, the pseudo-reality standard deviations are in the interval 9–23 ppb, while the pseudo-observation standard deviations are in the interval 3–5 ppb. The underestimation of the surface variability ranges from 55 % at the marine location over the North Sea (MOCAGE standard deviation 9 ppb, MAGEAQ-TIR standard deviation 4 ppb) to 87 % at Barcelona (MOCAGE standard deviation 23 ppb, MAGEAQ-TIR standard deviation 3 ppb). The urban locations have larger pseudo-reality variability and larger underestimations of the pseudo-observations variability, in particular at the Southern Europe subsection (87 and 85 % smaller standard deviations at Barcelona and Milan; 64, 70 and 67 % smaller standard deviations at Paris, Berlin and Amsterdam). The small variability of the MAGEAQ-TIR synthetic observations at surface is easily attributable to the scarce sensitivity at that altitude. Then, the spectral information has not the

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we concentrate on Milan but these considerations can be, to some extent, extended to the other urban locations. Higher and smaller values of the ozone concentration alternate at surface, for the MOCAGE pseudo-reality, depending on the hour. A typical diurnal cycle is characterized by values of 10–30 ppb for the nighttime minimum and 70–90 ppb for the daytime maximum. The airmasses enriched in ozone during the day are then mixed to higher levels airmasses and, during the night, values of typically 70–90 ppb are found in the height interval 1–3 km. This trend is found during weekdays, but not during the weekends of 8–9, 15–16 and 22–23 August 2009. This is due to the limited emission of ozone photochemical precursors during weekends at urban or industrial locations. This complex urban lowermost tropospheric ozone patterns are a natural choice as a test for MAGEAQ-TIR pseudo-observations. The MAGEAQ-TIR retrievals almost completely miss this daily/weekly evolution at the lowest levels. The signature of these enhancements at the lowest levels are spread onto higher altitudes, and the information content is redistributed accordingly. This indicates that a potential sensitivity exists that might be exploited by tuning the inversion schemes. In any case, with the present configuration, day/night cycle at surface and the lifting of enhanced ozone airmasses during the night are not detected. An attenuated day/night cycle is detected at 1 km altitude, but it is independent on the weekday. Such kind of structures are not detected also at Paris and Amsterdam. We are then inclined to conclude that a dedicated observing system like MAGEAQ-TIR may have potential to partially detect the photochemistry signal at the lowest atmospheric layers, which occurs at time scales of the order of the hours.

Another phenomenon which is not accurately detected is the one occurring over the North Sea on 20–21 August. The unusually high values of the ozone concentrations at surface and 1 km can be related to the ozone pollution event described by Forêt et al. (2013). During the period 19–21 August 2009, high values of lower tropospheric ozone, which are formed in the boundary layer over the South of France, are lifted to the free troposphere and then transported by a frontal motion, northwards and eastwards. The plume crosses this North Sea location during the days 20 and 21 August.

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It can be seen that MAGEAQ-TIR is not able to detect the peculiar vertical distribution at the lowest layers associated to this phenomenon. The same plume is observed on the 20 August over Amsterdam. Even if also here the vertical distribution of this phenomenon is missed, we can see enhanced values of ozone concentration at highest altitudes, i.e. between 2 and 7 km, maybe due to smoothing errors of the retrieval. This behaviour, at Amsterdam, gives the possibility to detect the signal of this ozone plume in the ozone columns time series (see Sect. 5), even if the height resolved structures are not detected. Phenomena occurring at different altitude ranges, where the retrievals are more sensitive, and/or at different time scales, are better retrieved. One possible example are the enhanced values of ozone concentrations extending from the upper troposphere, and possible also from the tropopause and the stratosphere, to the lower troposphere. Some examples are in the period 7–11 August at Milan, and 24–25 August at Paris and Amsterdam. These phenomena might be attributed to STEs. The enhanced values of the ozone concentration extend down to 4 km altitude. The phenomenon of 24–25 August has also the particularity of occurring in a very short time interval (1 day). These structures are very well resolved by the MAGEAQ-TIR, and we found very limited signature in the Hovmöller diagrams of the differences. Other well resolved structures are the peculiar low values of ozone concentration extending from surface to 5 to 8 km in the time intervals 11–14 August and 21–23 August at Paris, and 12–15 August, 16–19 August and 21–24 August at Amsterdam and over the North Sea.

7 Conclusions

We have shown some performance analyses for a future GEO instrument operating in the TIR spectral range, based on the instrumental specifications of MAGEAQ-TIR, following its Phase 0 study (Peuch et al., 2010). The instrumental design of MAGEAQ-TIR is thought to be adapted to lower and lowermost ozone monitoring. We have extended the study of Claeys et al. (2011b), to test and specify the performances of MAGEAQ-

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TIR over a bigger dataset, with a variety of different situations, and to test its capability to observe height-resolved phenomena. This has allowed, as well, the identification of the causes of poor performances. In addition, the synthetic data have been analysed at urban and rural locations, and the performances in presence of different phenomena occurring at different time-scales, e.g. STEs, photochemistry, horizontal transport, have been tested. The period 5–28 August 2009, on 1 h revisit time basis, has been considered.

First, we have characterized the vertical sensitivity and the retrieval accuracy of MAGEAQ-TIR pseudo-observations, in a global manner. By analysing the dataset as a whole, we have found that the MAGEAQ-TIR would give, on average, independent surface –6 km TOC observations, with mean DOF of about 1.0 and a maximum sensitivity at about 3.0 km, so at the centre of the column. There would be also an unprecedented sensitivity to the surface –3 km TOC, with about 0.6 DOF and maximum sensitivity at about 2.5 km, on average. Even if, MAGEAQ-TIR cannot completely separate the surface –3 km TOC information from the information coming from upper altitudes, the DOF surface –3 km can reach values of 0.8 in presence of higher thermal contrasts. The DT/NT differences drive the sensitivity more than the land/sea underlying surface differences, especially for smaller and lower columns. The sensitivity of the surface –3 km TOC pseudo-observations, in terms of their DOFs, is about 20 % better at DT than NT, and 11 % better over land than over sea. Both differences are less important for the surface –6 km TOC. The altitude of the maximum sensitivity of the surface –3 km TOC is less affected by the difference DT/NT and land/sea, than the DOF. Then, we have estimated the retrieval accuracy by comparing the pseudo-observations to the pseudo-reality. We have found average biases < 1 % in magnitude, for the surface –6 km TOC, and of about –2 to –3 %, for the surface –3 km TOC. The average RMSE is about 1.3 DU (5 %), for the surface –6 km TOC, and about 1.5 DU (10 %), for the surface –3 km TOC. The biases are mostly driven by the pixel surface type and the RMSEs by the observation hour. DT/NT performances are similar over sea but very different over land, with DT RMSEs smaller of the 45 and 30 %, with respect to the NT

subset, for the surface –6 km and surface –3 km columns, respectively. The average RMSEs for the DT/land subset are about 0.9 DU (4 %) and 1.2 DU (9 %), for the two columns.

Then, we have tested the performances of MAGEAQ-TIR at some selected urban and rural locations. We have selected 6 very small regions ($0.2^\circ \times 0.2^\circ$), representative of Milan (Italy), Paris (France), Berlin (Germany), Amsterdam (the Netherlands), one marine location West of Norway coast ($55^\circ \text{ N}/8^\circ \text{ E}$), Barcelona (Spain). We have investigated on how MAGEAQ-TIR would be able to detect the vertical structures of typical phenomena occurring in the troposphere, and then we studied the time evolutions of the surface –6 km and surface –3 km TOCs. We have found that our MAGEAQ-TIR pseudo-observations can replicate the MOCAGE pseudo-reality variability at all altitudes, except at surface to 1 km interval. The underestimations of surface ozone variability range from 55 to 87 %, with more severe underestimations at Southern European locations. The small variability of the MAGEAQ-TIR concentration pseudo-observations at the lowest layers is attributable to a limited sensitivity at those altitudes. By studying the time series of the profile pseudo-observations, we have found that MAGEAQ-TIR synthetic observations can partially describe the complex daily and weekly cycles at urban polluted sites. It seems that our MAGEAQ-TIR pseudo-observations have the potential to partly detect the photochemistry signal at the lowest altitudes, but the information content is redistributed and spread to higher altitudes. This signal seems generally very well detected in the columns time series at Northern European urban locations. In any case, we observe artifacts in the columns time series, especially for the surface –3 km TOC, at Southern European urban locations. At Milan, for example, a marked daily cycle of the MOCAGE TOC pseudo-reality, with a maximum at about 15:00 UTC is observed. At the same time, the sensitivity of the MAGEAQ-TIR pseudo-observations, in terms of the DOF, shows a marked cycle, with up to 100 % differences between maximum and minimum, with the maximum occurring at 12:00 UTC. In fact, at Southern European urban location, the MAGEAQ-TIR sensitivity is mostly driven by the thermal contrast. The result is that the MAGEAQ-TIR column pseudo-observations, in

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particular for the surface –3 km TOCs, show an artifact maximum shifted back of several hours with respect to the target pseudo-reality. This artifact is sometimes observed also at Barcelona, so we argue that it is important where the sensitivity of the observations are more strongly driven by the thermal contrast. Other kind of phenomena occurring at altitudes lower than 2 km, like the transport pattern of the 20 August 2009 at a marine location, are not detected on a height-resolved basis. However, even if the vertical structure of these transport phenomenon is missed at all sites, its signal can still be seen on the surface –6 km and surface –3 km columns, at some locations. MAGEAQ-TIR seems capable to detect the vertical structures of phenomena occurring at altitudes higher than 2 km, like deep STEs, even if characterised by particularly short time evolutions (of the order of 1 day). In summary, while the vertical structures of the lowermost tropospheric ozone pseudo-reality are sometimes missed or spread onto larger vertical intervals, MAGEAQ-TIR column pseudo-observations follow stunningly good the MOCAGE column pseudo-reality. For the comparison MAGEAQ-TIR and MOCAGE TOCs, very high correlation coefficients (0.93–0.97 and 0.83–0.91), low biases (<1.6 and <5.0 %, in magnitude) and low RMSEs (4.20–4.60 % and 8.9–9.8 %) are found for the surface –6 km and the surface –3 km TOCs, at northern European urban locations and at the marine location. Worse performances are found at Southern European locations, e.g. at Milan, correlations of 0.82 and 0.55, biases of 4.2 and 10.8 %, RMSEs of 6.4 and 13.5 %, for the surface –6 km and the surface –3 km TOCs.

These analyses, although pointing out the limitations of an AQ-dedicated GEO instrument like MAGEAQ-TIR, show that it would be a great step forward to gain a more solid monitoring capability of short term pollution phenomena at the local and continental scale in Europe. A future step would be the analysis of the performances of a multi-spectral approach, e.g. by including in our analyses the VIS component of MAGEAQ and/or MTG-UVN observations.

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Table 1. Selected Phase 0 threshold and target technical specifications of MAGEAQ-TIR (adapted from Peuch et al., 2010). In the table, SSP = sub-satellite point.

	Threshold	Target
Lifetime	5 yr	
Field of regard	15° W–35° E, 35° N–65° N	
Spatial sampling at SSP	15 km × 15 km	10 km × 10 km
Repeat cycle	2 h	1 h
DOF in the troposphere	2	3

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Table 2. Mean DOFs surface –6 km and surface –3 km for land/day, land/night, sea/day and sea/night pixels, calculated over the whole dataset, on a 1h revisit time basis.

	Day		Night	
	DOF surf –6 km	DOF surf –3 km	DOF surf –6 km	DOF surf –3 km
Land	1.27	0.71	1.14	0.54
Sea	1.14	0.57	1.11	0.49

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Table 3. Mean altitudes of the maximum of the integrated AK for the surface –6 km TOC and surface –3 km TOC for land/day, land/night, sea/day and sea/night pixels, calculated over the whole dataset, on a 1h revisit time basis.

	Daytime		Nighttime	
	Max sens surf –6 km	Max sens surf –3 km	Max sens surf –6 km	Max sens surf –3 km
Land	2.67 km	2.09 km	3.10 km	2.70 km
Sea	2.85 km	2.60 km	3.15 km	2.82 km

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Table 4. Mean biases and RMSEs of MAGEAQ-TIR surface –6 km TOC pseudo-observations, with respect to MOCAGE pseudo-reality. Results are reported for: all data, land (sea) only, DT (NT) only, and all various combinations. Percent values of biases and RMSEs are reported in parentheses.

	Land + sea		Land		Sea	
	Bias [DU (%)]	RMSE [DU (%)]	Bias [DU (%)]	RMSE [DU (%)]	Bias [DU (%)]	RMSE [DU (%)]
Daily	–0.16 (–0.30%)	1.28 (5.28%)	–0.01 (–0.27%)	1.17 (4.81%)	–0.32 (–0.93%)	1.38 (5.68%)
DT	–0.12 (–0.16%)	1.06 (4.35%)	+0.10 (+0.62%)	0.92 (3.82%)	–0.35 (–1.02%)	1.41 (5.73%)
NT	–0.20 (–0.43%)	1.47 (6.05%)	–0.11 (–0.62%)	1.61 (6.13%)	–0.30 (–0.84%)	1.36 (5.52%)

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Table 5. Same as Table 4 but for surface –3 km TOC pseudo-observations.

	Land + sea		Land		Sea	
	Bias [DU (%)]	RMSE [DU (%)]	Bias [DU (%)]	RMSE [DU (%)]	Bias [DU (%)]	RMSE [DU (%)]
Daily	–0.47 (–2.24 %)	1.53 (10.39 %)	–0.36 (–1.65 %)	1.51 (10.29 %)	–0.60 (–2.88 %)	1.55 (10.53 %)
DT	–0.46 (–2.19 %)	1.42 (9.62 %)	–0.30 (–1.40 %)	1.25 (8.78 %)	–0.63 (–3.06 %)	1.59 (10.35 %)
NT	–0.49 (–2.28 %)	1.62 (11.14 %)	–0.42 (–1.90 %)	1.73 (12.26 %)	–0.56 (–2.71 %)	1.51 (9.95 %)

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Table 6. Mean biases, RMSEs and Pearson correlation coefficients of MAGEAQ-TIR surface –6 km TOC pseudo-observations, with respect to MOCAGE pseudo-reality, at six selected locations. Results are reported for: Milan, Paris, Berlin, Amsterdam, the marine location west of Norway coast, Barcelona. Percent values of biases and RMSEs are reported in parentheses.

	Milan	Paris	Berlin	Amsterdam	North Sea	Barcelona
Bias	–1.29 (–4.25%)	–0.15 (–0.34%)	–0.29 (–0.99%)	–0.06 (–0.00%)	–0.46 (–1.54%)	–0.57 (–1.93%)
RMSE	1.96 (6.43%)	1.08 (4.23%)	1.08 (4.22%)	1.03 (4.20%)	1.15 (4.62%)	1.19 (4.09%)
Pearson	0.82	0.95	0.93	0.96	0.97	0.89

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Table 7. Same as Fig. 6 but for the surface –3 km TOCs.

	Milan	Paris	Berlin	Amsterdam	North Sea	Barcelona
Bias	–2.17 (–10.82%)	–0.82 (–4.39%)	–0.85 (–4.99%)	–0.60 (–3.11%)	–0.89 (–4.91%)	–1.22 (–6.56%)
RMSE	2.67 (13.54%)	1.54 (9.70%)	1.41 (8.92%)	1.37 (8.93%)	1.54 (9.77%)	1.67 (9.10%)
Pearson	0.55	0.87	0.83	0.91	0.91	0.73

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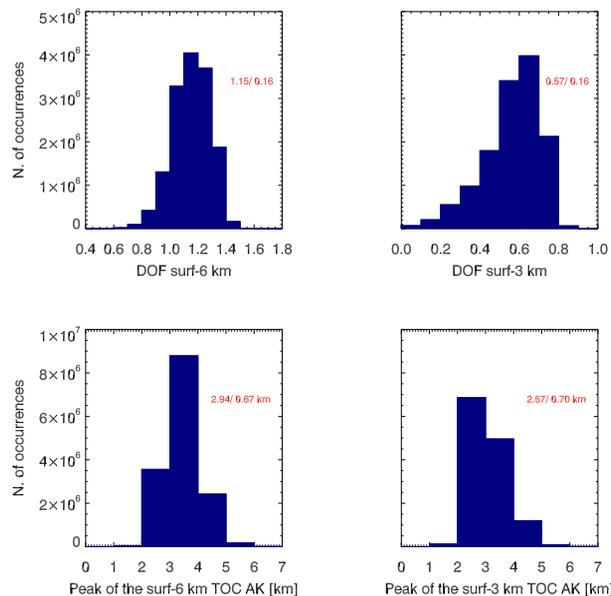


Fig. 1. Histograms of the DOF surface –6 km (top left), the DOF surface –3 km (top right), the altitudes of the maximum of the integrated AK for the surface –6 km TOC (bottom left), and the altitudes of the maximum of the integrated AK for the surface –3 km TOC (bottom right), for MAGEAQ-TIR. Mean values and standard deviations are also reported in red (mean/std). The data cover the whole period of study, on a 1 h revisit time basis.

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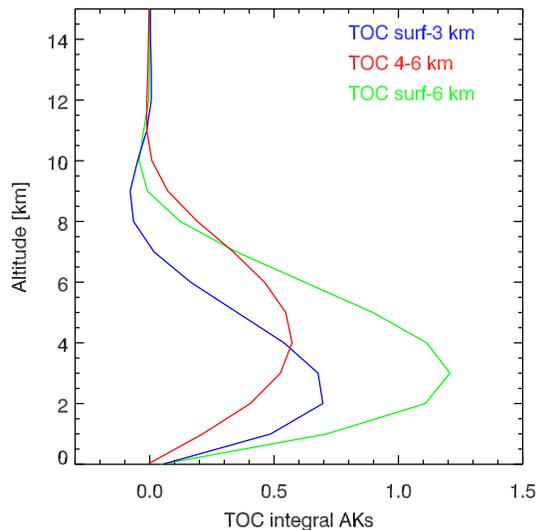


Fig. 2. Mean integral averaging kernel functions for the surface –3 km (blue), 4–6 km (red) and surface –6 km (green) TOCs, for all pixels in the dataset having thermal contrast between 0 and 1.0 K.

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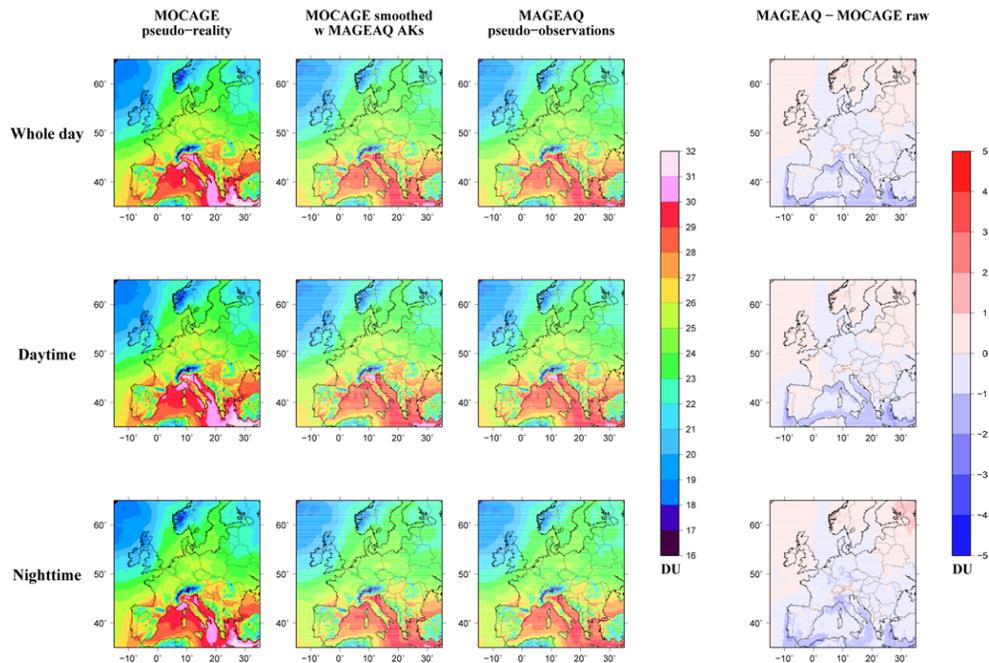


Fig. 3. Average surface –6km TOC for 5–28 August 2009. From left to right: raw and **AK**-smoothed MOCAGE pseudo-reality, MAGEAQ-TIR pseudo-observations, differences of MAGEAQ-TIR pseudo-observations with respect to raw MOCAGE pseudo-reality. The average TOC are reported for the whole day (top row), daytime (central row) and nighttime acquisitions (bottom row).

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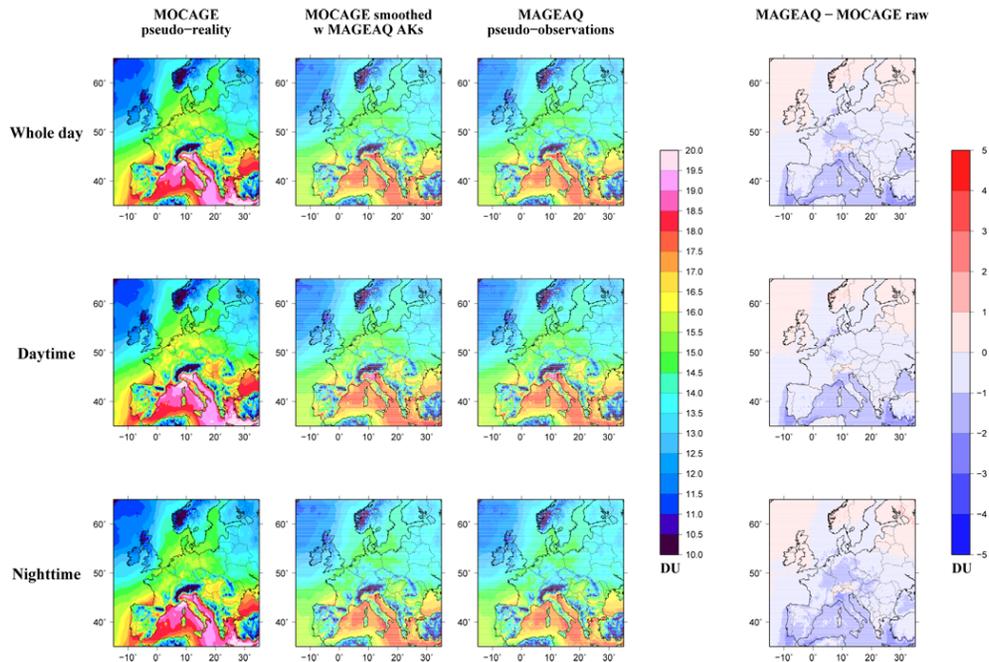


Fig. 4. Same as Fig. 3 but for surface -3 km TOC.

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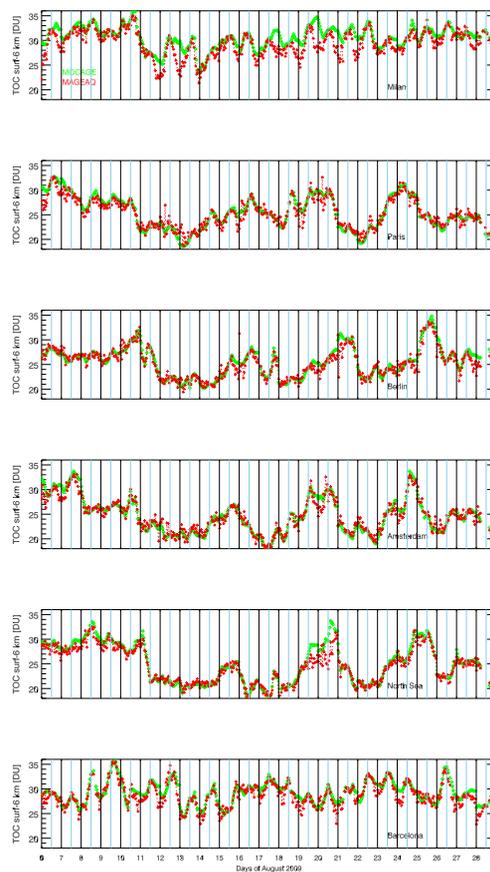


Fig. 5. Time series of MOCAGE surface –6 km TOC pseudo-reality (green), and MAGEAQ-TIR surface –6 km TOC pseudo-observations (red) over Milan, Paris, Berlin, Amsterdam, the marine location West of Norway coast, Barcelona.

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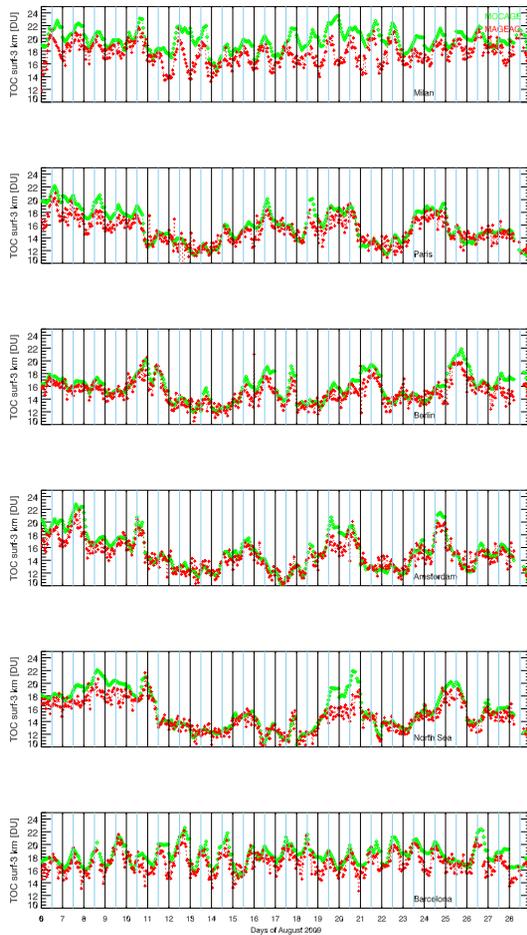


Fig. 6. Same as Fig. 5 but for surface –3 km TOC.

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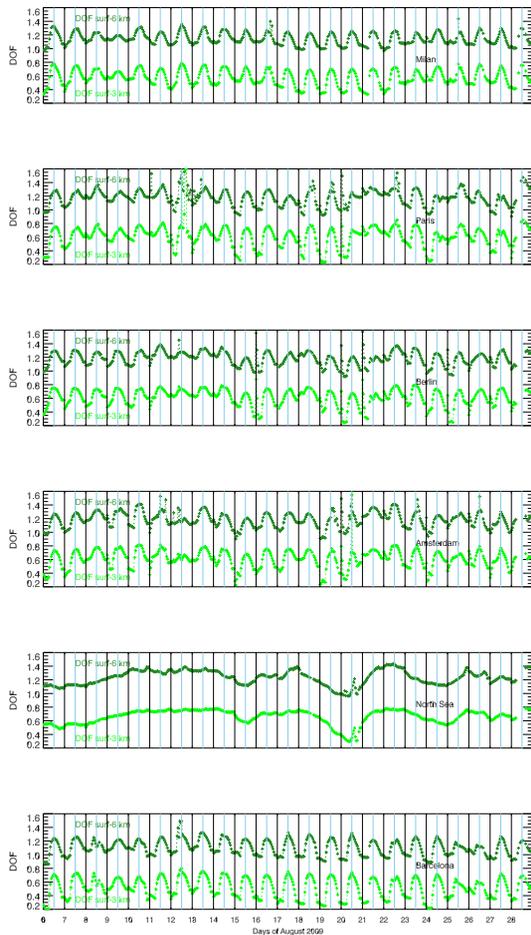


Fig. 7. Time series of the DOF surface –6 km (dark green) and surface –3 km (light green) over Milan, Paris, Berlin, Amsterdam, the marine location West of Norway coast, Barcelona.

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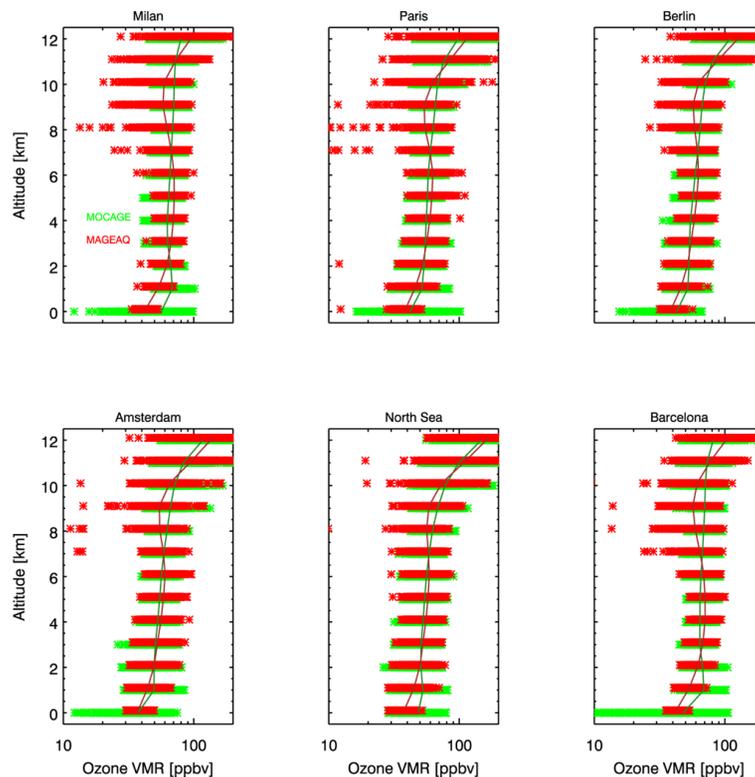


Fig. 8. Tropospheric ozone concentration profiles values for all pixels: MOCAGE pseudo-reality (green) and MAGEAQ-TIR pseudo-observation (red), at Milan (Italy), Paris (France), Berlin (Germany), Amsterdam (the Netherlands), one marine location West of Norway coast (55° N/ 8° E), Barcelona (Spain). The mean tropospheric ozone concentration profiles for both MOCAGE (green) and MAGEAQ-TIR (red) are also shown.

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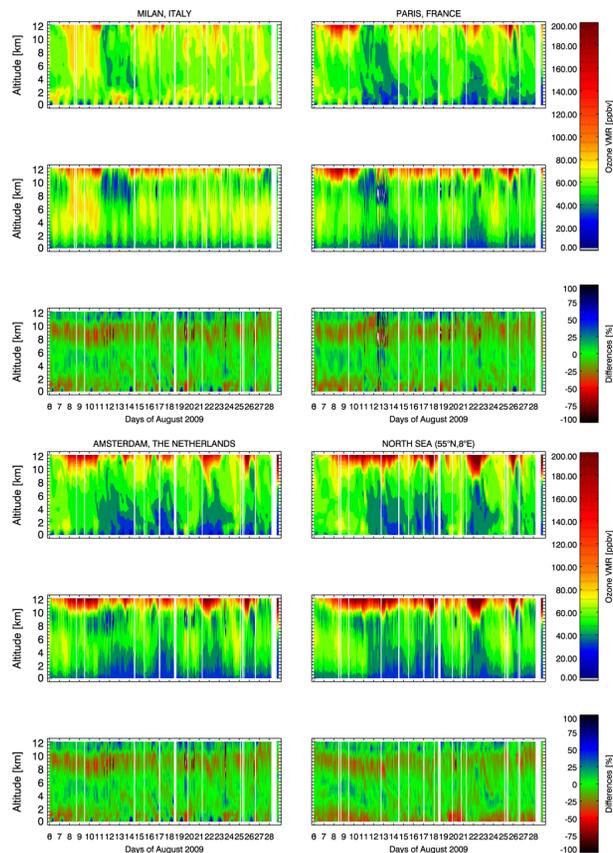


Fig. 9. Hovmöller diagrams showing the MOCAGE pseudo-reality (lines 1 and 4), MAGEAQ-TIR pseudo-observations (lines 2 and 5) and the percent differences (lines 3 and 6) for the tropospheric ozone concentration profiles over Milan, Paris, Amsterdam, the marine location West of Norway coast, for the whole time period of our study.

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