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1 Objectives of the report

This deliverable is dedicated to the assessment and validation of the WP3 numerical models providing the atmospheric composition with regards to observational datasets such as satellite, ground-based and airborne measurements obtained during the 2016 field campaign. The focus is on the main atmospheric gaseous and particulate pollutants, which are targeted by air quality regulations for their detrimental impacts on health.

The gaseous species, nitrogen dioxide, carbon monoxide and ozone (hereafter NO_2 , CO and O_3) are evaluated however they are challenging to predict as they originate from many different sources (traffic, domestic stoves, vegetation fires). Ozone is particularly difficult because it is a secondary pollutant formed in the atmosphere.

For the aerosols, the total column aerosol optical depth is investigated in order to the complex spatial distribution due to the diverse aerosols sources, which are both anthropogenic (traffic, residential, industrial) and natural (dust from the Sahara desert, sea salt from the Guinean Gulf, biogenic from forests, soot from vegetation fires).

The assessment of particulate and gaseous species from the different models will allow us to point out the strengths and weaknesses of the WP3 models. We will address the aspects consistent between models and observations, and we will identify the robust modeled features. Weaknesses shared by all models will be described and future analyses will be proposed to improve the state-of-the-art air quality modeling in Southern West Africa (SWA). In order to understand if refining the spatial resolution could solve the biases between the models and observations, different spatial resolutions of the models will be compared.

The evaluation strategy is to compare all models for the same domain and time period. All modeling groups have provided simulation output data covering the period of the field campaign from the 25th June 2016 to the 14th July 2016. We aim to investigate both the temporal and the spatial variability, with ground station data that has been implemented in the framework of the DACCIWA program, with satellite data, and during airborne measurements from the three aircraft.

In Figure 1, the evaluation domain is presented going from latitude 1°S to 15°N , and longitude 12°W to 12°E . The domain includes 7 AERONET stations including two stations set up with the DACCIWA project. Moreover the CO and O_3 concentrations have been measured at the Savè ground station. The trajectories of all flights used in this report are presented in Figure 1. There were 20 flights made by the French-ATR42 aircraft, 16 by the British-Twin-Otter aircraft, and 12 by the German-Falcon aircraft.

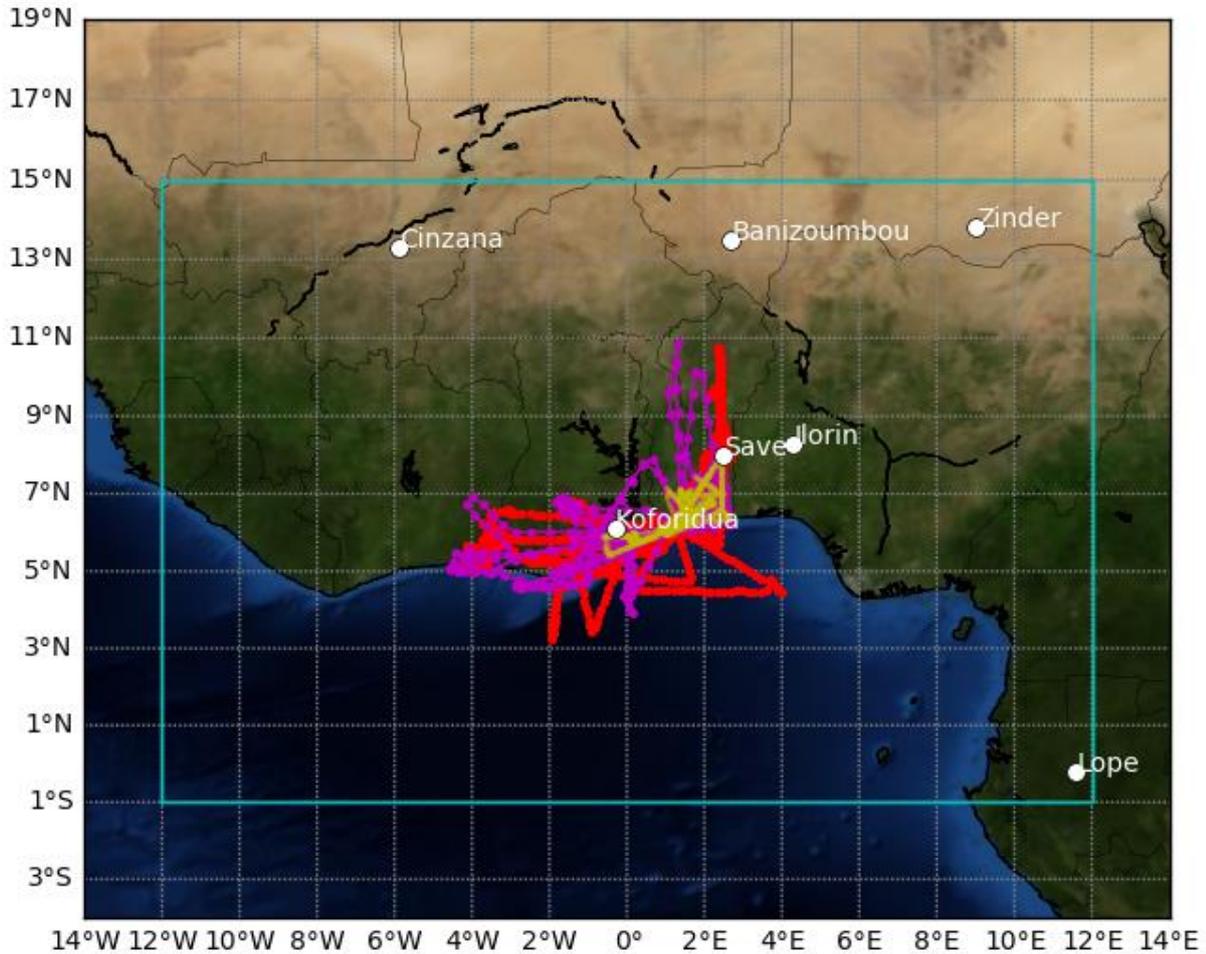


Figure 1– Model evaluation domain (light blue rectangle), flight tracks of the French ATR plane operated by the SAFIRE team (in red), of the German Falcon operated by the DLR team (in violet), of the British Twin Otter operated by the BAS team (in yellow), AERONET stations (white dots) used in the D3.2 report.

Section 2 gives a general overview of the WP3 models and the simulation characteristics run in the framework of the DACCIWA program. The results of the evaluation of the atmospheric composition concerning the aerosols are given in the Section 3. For the gaseous pollutants, the evaluation focuses on three gaseous species concentrations (CO , NO_2 and O_3) in Section 4. The report closes with an assessment summary for our region of interest in SWA as well as a discussion of the overall model performances and improvements that could be achieved within the DACCIWA project.

2 Numerical models overview

This report includes six models which are either running in research or operational mode (see Table 1). The spatial domains and resolutions of the models cover a highly broad range from the regional to global simulations.

Table 1 – Models overview

Model name	Model version used	Mode	Contact person
COSMO-ART	COSMO5.1-ART3.1	Quasi-operational	Konrad Deetz
CHIMERE	CHIMERE v2017	research	Adrien Deroubaix
GEOS-Chem	GEOS-Chem v10-01	research	Eleanor Morris
ECHAM-HAM	ECHAM6.3-HAM2.3	research	Tanja Stanelle
CAMS-IFS	CAMS-IFS	operational	Angela Benedetti

Models description and configuration for DACCIWA

The models have been extensively presented in the deliverable D4.2 (see this document for full description). In the following section, the configuration adaptations made for the DACCIWA program are detailed.

o CHIMERE model

CHIMERE is a chemistry-transport model allowing the simulation of concentrations fields of gaseous and aerosols species at a regional scale. It is an off-line model version, driven by pre-calculated meteorological fields. In this study, the version is fully described in *Menut et al. (2013a)* and updated in *Mailler et al. (2016)* is used. The simulations are performed with the same horizontal domain, the 28 vertical levels of the WRF simulations are projected onto 20 levels from the surface up to 200 hPa for CHIMERE.

Two simulations have been done with two different resolutions (0.3° and 0.1°) and meteorological fields. The highest resolution has been nested with the lowest resolution for aerosol and gas concentrations. The meteorological fields used are ECMWF (for the 0.3° resolution) and WRF (for the 0.1° resolution).

o COSMO-ART model

COSMO-ART (Consortium for Small-scale Modeling – Aerosols and Reactive Trace gases) is a comprehensive online-coupled model system (*Vogel et al., 2009*) based on the operational weather forecast model COSMO (*Baldauf et al., 2011*). COSMO-ART includes a comprehensive chemistry module to describe the gaseous composition of the atmosphere and secondary aerosol formation. It allows for feedback of the simulated aerosol particles with radiation, cloud formation and precipitation (e.g. *Stanelle et al., 2010*, *Athanasopoulou et al., 2014*; *Rieger et al., 2014*; *Walter et al., 2016*). The size distribution of aerosol within COSMO-ART is approximated by log-normal distributions (modes hereafter). Chemical reactions are calculated with RADMKa (Regional Acid Deposition Model Version Karlsruhe; *Vogel et al., 2009*), which is based on RADM2 (Regional Acid Deposition Model, *Stockwell et al., 1990*). The formation of secondary organic aerosol is

calculated by a VBS approach (volatility basis set; *Athanasopoulou et al., 2013*). COSMO-ART is able to describe the emission and atmospheric dispersion of natural and anthropogenic pollutants.

To apply COSMO-ART to the conditions of SWA, several adaptations have been realized. The global EDGAR emission database was preprocessed for COSMO-ART and the biogenic emission routine MEGAN2.1 of *Guenther et al. (2012)* was implemented into COSMO-ART. Since gas flaring seems to be a relevant source of pollution, Nigeria is with 15 billion cubic meter flared gas the second largest flaring country, we have developed a flaring emission inventory (*Deetz and Vogel, 2017*). This inventory allows for a physically based estimation of the flaring emissions of carbon dioxide, carbon monoxide, sulphur dioxide, nitrogen monoxide and nitrogen dioxide based on combustions calculations and remote sensing observations. Additionally we have further developed our mineral dust emission routine. By combining the parameterization of *Vogel et al. (2009)* and *Shao et al. (2010)* the emission routine can now be flexibly applied globally compared to the previous scheme in COSMO-ART. In summary COSMO-ART considers the following emissions within the DACCIIWA studies: mineral dust, sea salt, dimethyl sulfate, biogenic volatile organic compounds as well as anthropogenic trace gases and aerosols (including biomass burning and flaring). Not considered are NO_x emissions from lightning and soil. For anthropogenic emissions we use EDGAR HTAP V2 (*EDGAR, 2010*) and for the biomass burning emissions the CAMS Global Fire Assimilation System (*GFAS, 2016*), which is available near real-time.

For COSMO we use the tropical setup which includes among others an increased number of vertical levels up to 30km.

o GEOS-Chem model

GEOS-Chem (www.geos-chem.org) is an atmospheric chemical transport model that can be run either globally (*Bey et al. 2001a*), or in a higher resolution regional configuration (*Wang et al. 2004*). For this report, a regional simulation has been performed for the West Africa domain (latitudes 6°S–16°N, longitudes 18.125°W–26.875°E) at a horizontal resolution of 0.25° x 0.3125° and a vertical resolution of 47 levels up to 0.01hPa.

The model is driven by assimilated meteorological data from the NASA Global Modelling and Assimilation Offices (<https://gmao.gsfc.nasa.gov/>). The simulation includes HO_x-NO_x-VOC-O₃-BrO_x tropospheric chemistry as well as a mass based scheme for sulfate, nitrate, ammonium, carbonaceous, dust and sea salt aerosols. Boundary conditions for running the regional simulation for West Africa were generated from a global simulation at a resolution of 4° x 5°.

Anthropogenic emissions are from the EDGAR inventory (*European Commission, 2011*) for the year 2008, biomass burning is from GFED4 (*Giglio et al. 2013*) for the year 2014 and biogenic VOCs are derived from an online implementation of the MEGAN2.1 inventory (*Guenther et al. 2012*).

o ECHAM-HAM model

The ECHAM6-HAM2 model is global aerosol climate model. The aerosol module HAM was first implemented in the 5th generation of the atmospheric general circulation model ECHAM (*ECHAM5, Roeckner et al., 2003*) by the Max Planck Institute for Meteorology (*Stier et al., 2005*). Over the past years, the HAM module has been improved and completed with new processes (now it is called HAM2) as described in *Zhang et al. (2012)*. The HAM2 module is now coupled to the 6th generation of the ECHAM family (*Stevens et al., 2013*).

Aerosol microphysics is simulated using the M7 module (*Vignati et al., 2004*), which accounts for sulfate, black carbon, particulate organic matter, sea salt, and dust. The atmospheric aerosol population is described as a superposition of seven lognormal distributed modes for which standard deviations are prescribed. The total number concentration and masses of the different chemical components are prognostic variables in the .model. The modes are divided into soluble, internally mixed modes (containing sulfate) and insoluble, externally mixed modes, which are assigned to different size ranges. The modal diameters can vary and are calculated at each time step from the mass and number concentrations for each mode. Dust particles are considered as part of the soluble and insoluble accumulation and coarse modes. Sedimentation and dry and wet deposition are parameterized as functions of the aerosol size distribution, composition, and mixing state and depend on the ECHAM6 meteorology. The emission fluxes of dust, seasalt, and dimethyl sulfide from the oceans (DMS) are calculated online, based on the model meteorology. Anthropogenic emissions are prescribed.

After a brief evaluation of first test simulations we decided to use the ECHAM6.3-HAM2.3 version in the framework of the DACCIWA project. For the purpose of this model evaluation, we performed a nudged simulation. Therefore, we prescribed the large-scale meteorology with ERAinterim Reanalysis. Anthropogenic and biomass burning aerosol emissions are prescribed by the ACCMIP (Atmospheric Chemistry and Climate Model Intercomparison Project) emission inventory (*Lamarque et al., 2010*).

o CAMS-IFS model

The Composition-Integrated Forecast System is an extended version of the Integrated Forecast System (IFS) with capabilities for modelling and assimilation of atmospheric constituents such as gases and aerosols. It is run operationally within the Copernicus Atmosphere Monitoring Service (CAMS) managed by ECMWF. The composition modules are online within IFS, offering a comprehensive description of the state of the atmosphere. CAMS forecasts have been used operationally during the aircraft campaign. The results presented in this report are relative to those operational runs. The CAMS system is described in *Flemming et al. (2015)* for the tropospheric chemistry, and in *Morcrette et al. (2009)* for the aerosols. Data assimilation with C-IFS is described in *Inness et al. (2015)* for the chemistry and *Benedetti et al. (2009)* for the aerosols.

No specific adaptations for the DACCIWA campaign were implemented.

The chemistry and transport processing varies between each WP3 model, driving differences in the modeled pollutant concentrations. Nevertheless, the main differences may depend above all else on the emission and meteorological datasets. More information on the modeling configurations is presented in Table 2 below for each modeling group, giving clues for interpreting and assessing the models performance in Section 3 and 4.

Table 2 - Models characteristics

Model	COSMO-ART	CHIMERE	GEOS-chem	ECHAM-HAM	CAMS-IFS
Horizontal resolution	0.25°	- 0.1x0.1° - 0.3x0.3°	0.25 x 0.3125°	T63 (~1.8x1.8° at equator)	T255 – 0.8x0.8°
Vertical levels	50 (up to 30km)	20 (up to 200hPa)	47 (up to 0.01hPa)	L47 (up to 0.01hPa)	60 (up to 0.1hPa)
Output frequency	3h	1h	1h	3h	1h (3h data were delivered)
Anthropogenic emissions	HTAP 2010	HTAP 2010	EDGAR v4.2 2008	ACCMIP RCP4.5	MACCity
Biogenic	Guenther et al. (2012) (MEGAN2.1, online)	MEGAN (online)	MEGAN 2.1	prescribed	
Mineral dust	Vogel et al. (2009), further developed	AG2001 (online)	Online	Tegen et al. (2002), Cheng et al. (2008) online	Online
Sea salt	Film droplets: Mårtensson et al. (2003), jet droplets: Monahan et al. (1968), spume droplets: Smith et al. (1993)	Monahan (online)	Online	Long et al (2011) online	Online
Fires	GFAS	Apiflame	GFED4 2014	ACCMIP RCP4.5	GFAS
Aerosol distribution (bins)	Modal (12 lognormal modes)	10 bins	Dust (4 bins), organic carbon, black carbon, seasalt, sulphate, nitrate and ammonium	Modal (7 lognormal modes)	3 bins sea salt 3 bins desert dust bulk organic matter, black carbon and sulphate
Data assimilation	none	none	none	none	Yes, MODIS AOD

3 Aerosol content evaluation

The Aerosol Optical Depth (AOD) is a vertically integrated measurement, which includes several aerosol origins. The AOD gives information on the aerosols variability for the entire atmospheric column. In the first part, the modeled AOD are compared to satellite measurements, and in the second part to ground-based stations. In the third part, some case studies are detailed for two specific dates, when the three aircraft were flying with a city emission flight objective.

3.1 Spatial variability

This section is dedicated to the models' assessment against the MODIS probe, which provides AOD (at 550 nm) onboard two satellite platforms: Terra (equator crossing at 10:30) and Aqua (equator crossing at 13:30). The MODIS data have been extracted from the Giovanni data portal (available at <https://giovanni.gsfc.nasa.gov/giovanni/>). The MYD08-D3 and MOD09-D3 daily products provide an AOD at 1° resolution over dark and brilliant surfaces: the Dark-Target and the Deep-Blue products (Hsu et al. 2004, Sayer et al. 2013, Sayer et al. 2014). These two products have been merged to present AOD in a single map (Figure 2). When both products are available, they are averaged. Clouds disable the MODIS-AOD retrieval and no data is displayed when this was the case onboard the two platforms.

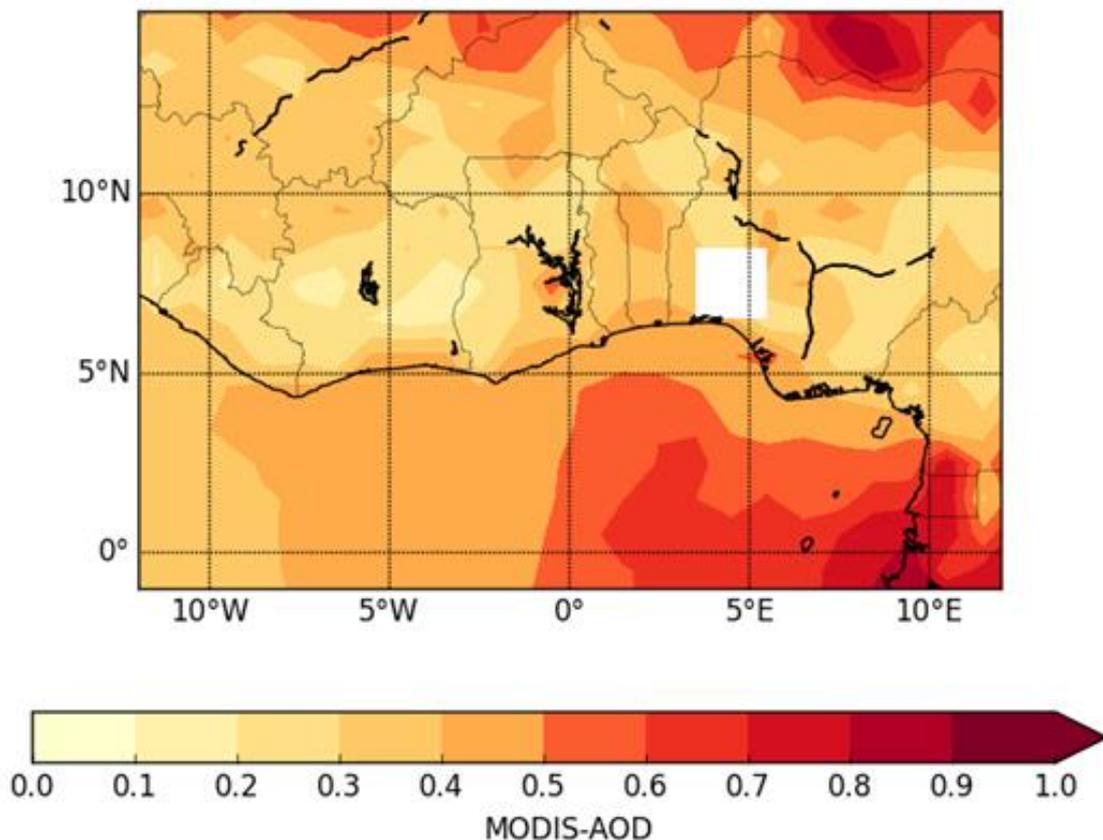


Figure 2 - MODIS-AOD averaged over the whole field campaign period from the 25th June to the 14th July 2016 for the combined Dark-Target and Deep-Blue product acquired by Aqua and Terra (average of MYD08-D3 and MOD09-D3 products when both are available).

Figure 2 presents the MODIS-AOD average, however each pixel is not associated with the same number of observations because the cloud cover is different every day. Over the DACCIIWA field campaign period, we can see that there are two areas of higher AOD in the North over the Eastern part of Niger, and in the South over Western Gabon. In the North, the high AOD area is located close to the major dust source of the Bodélé depression in Chad (e.g. Flamant et al. 2009). In the

South, the high AOD area corresponds to the biomass burning plume pathway from the vegetation fire sources in Central Africa (e.g. *Mari et al. 2007*). In our region of interest, the Guinean coastal area, we note a moderate AOD (about 0.4). There is no clear signature of higher aerosol contents over the main coastal cities, which could be linked to the coarse resolution of this level 3 product and the frequent masking through clouds.

AOD is higher above the ocean than above the continent, which is surprising given that the urban areas with industries, traffic, domestic fires are situated along the coastline. This surprising spatial feature might be linked to the biomass burning plume, which is above the marine boundary layer and almost disconnected from this layer. The daily convection catches this plume and mixes it with the continental boundary layer when the Terra and Aqua platforms overpass our evaluation domain. On the other hand, it could be an artifact due to the different number of measurements over land and ocean, and also because the AOD could be over-estimated in cloudy regions (*Remer et al. 2008*).

For the modeled AOD, an average of all modeled hours over the entire evaluation period is presented in Figure 3, thus it is not directly comparable to MODIS-AOD in Figure 2. Nevertheless, the main large scale features should be in agreement.

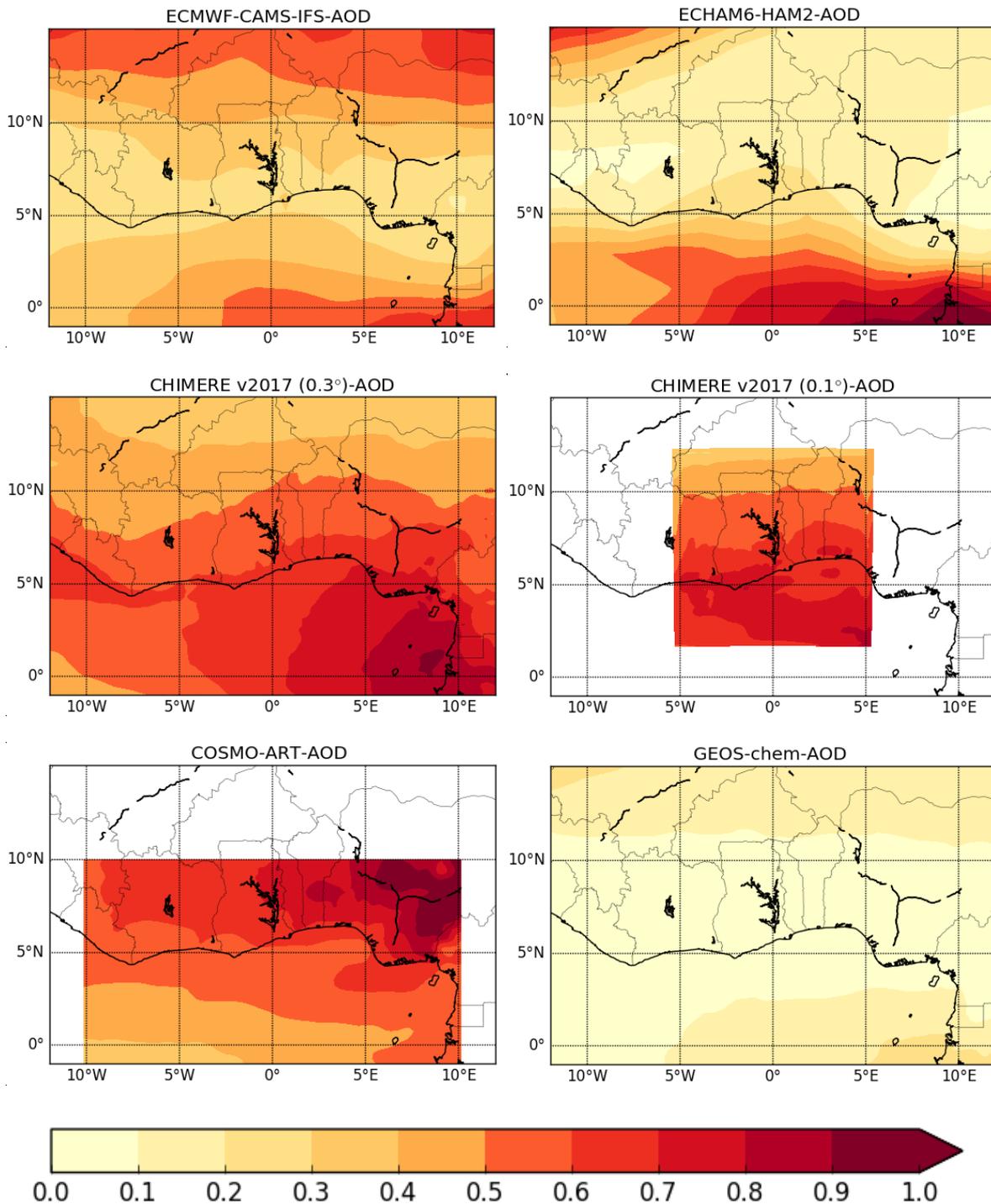


Figure 3 - AOD average of the six models (CAMS-IFS, ECHAM6-HAM2, CHIMERE 0.3° and 0.1°, COSMO-ART, GEOS-Chem) over the whole field campaign period from the 25th June to the 14th July 2016.

The comparison between MODIS-AOD and the AOD modeled by the CAMS-IFS model shows that it reproduces well the two high AOD areas. Moderate AOD are modeled over the Guinean coastal region in agreement with observations.

The comparison between MODIS-AOD and the AOD modeled by the ECHAM-HAM model shows that the biomass burning plume is well located with the same magnitude. Over South-East Niger, the high AOD area is not reproduced. Moderate AOD are modeled over the Guinean coastal region in agreement with observations.

The comparison between MODIS-AOD and the AOD modeled by the CHIMERE model at 0.3° shows that it reproduces well the high AOD area in the South of the evaluation domain. The high AOD associated with dust is not reproduced. An AOD over-estimation is modeled over the Guinean coastal region (about 0.2).

For CHIMERE at 0.1°, the same lack of high AOD in the South-East Niger is observed. It is worth to note that the highest resolution, which has been run with different meteorological fields, leads to a different AOD spatial structure over the Guinean gulf. An AOD over-estimation is modeled over the Guinean coastal region (about 0.3).

The comparison between MODIS-AOD and the modeled AOD by COSMO-ART shows an over-estimation over Nigeria (about 1 where MODIS-AOD is about 0.3). An AOD over-estimation is also modeled over the Guinean coastal region (about 0.2).

The comparison between MODIS-AOD and the AOD modeled by the GEOS-Chem model indicates that the two high AOD areas are well located but with an important under-estimation (about 0.8). Moderate AOD are modeled over the Guinean coastal region in agreement with observations.

The WP3 models show that modeled AOD is very variable, from a large under-estimation for GEOS-Chem to a good agreement for the CAMS-IFS model allowed by the MODIS-AOD assimilation (*Benedetti et al. 2009*). From the South to the North, three areas could be distinguished: the biomass burning area, the urban area, and the dust area. These three areas are present in CAMS-IFS, GEOS-Chem and ECHAM-HAM.

3.2 Temporal variability at AERONET stations

The AEROSol NETwork is a network of stations measuring aerosol properties with a sun-photometer. This network has been implemented by NASA and each station is operated by different research groups. The variables used in this section are the AOD and the Angstrom exponent. The AOD provides information on the aerosol variability for the entire atmospheric column. At a given wavelength, the AOD is the integration of the extinction coefficient due to aerosol absorption and scattering along the whole solar pathway (weighted to be independent of the time of the day). The Angstrom exponent gives information about the aerosol size, which could be considered as coarse particles (such as sea salt or dust) when it is below 0.5 (*Smirnov et al. 2002*). There are only two models with different AOD wavelengths available to calculate the Angstrom exponent (CHIMERE and ECHAM-HAM).

From the daily averages AERONET level-1.5 measurements, the AERONET-AOD is calculated or interpolated (depending on the station) at 550 nm. For each station, a spatial bilinear interpolation of the model outputs is performed, and daily modeled averages for daytime are calculated and compared with the daily AERONET averages.

Seven stations have been selected: three in the Sahel close to the mineral aerosol sources in the Sahara; three in SWA, two of which have been set up within the DACCIWA project; one in Central Africa close to the vegetation fire sources.

3.2.1 Sahelian stations

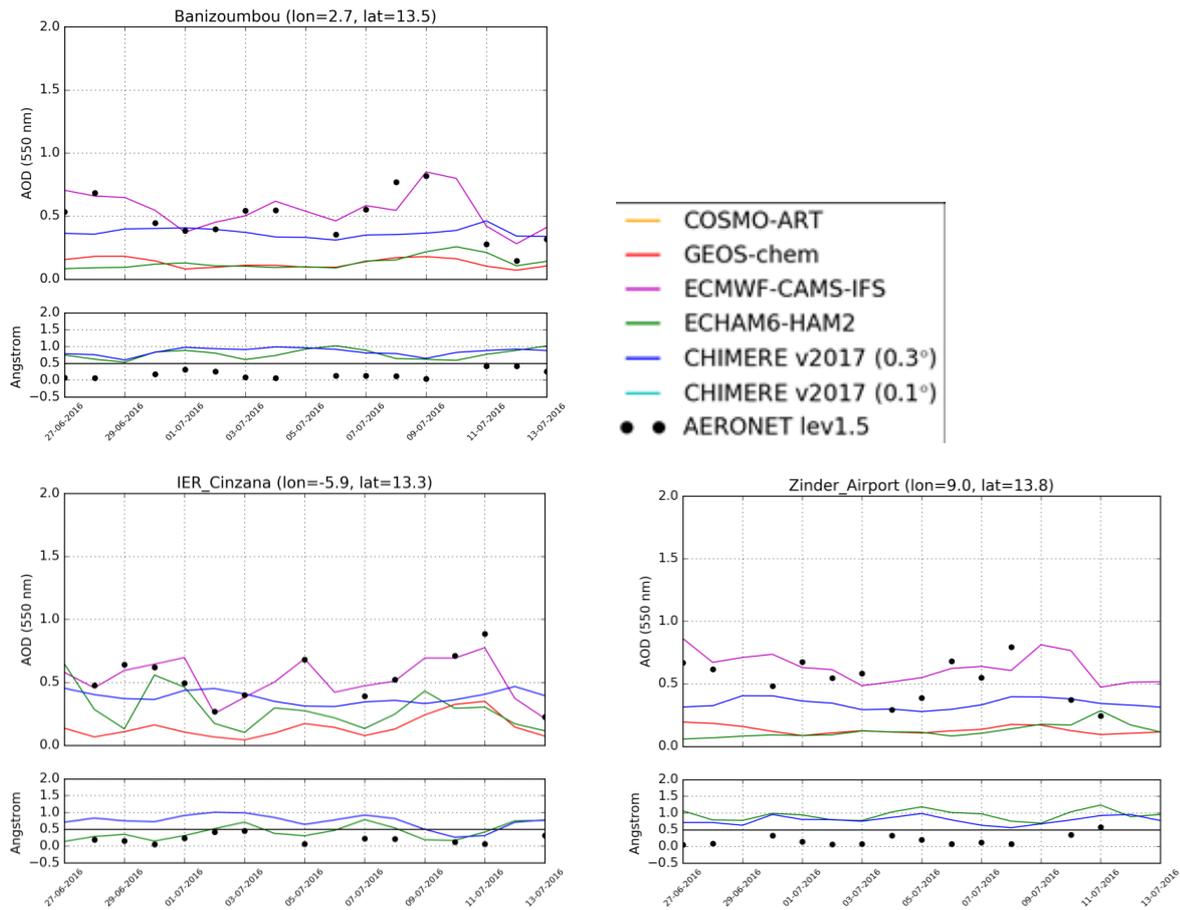


Figure 4 – Time series of average daily AOD and Angstrom exponent modeled by the WP3 models and observed at two Sahelian AERONET stations (Banizoumbou in Niger and Zinder in Niger) for the period 25th June to 13th July 2016.

The observed AOD in Figure 4 at the Sahelian stations, ranging from 0.3 to 0.8 and is comparable to the WP3 models for CAMS-IFS from 0.5 to 0.8, for ECHAM6-HAM2 from 0.1 to 0.3, CHIMERE (at 0.3°) from 0.3 to 0.4 and for GEOS-Chem from 0.1 to 0.2. The observed variability is higher than the modeled one.

The modeled Angstrom exponent is similar for the CHIMERE and the ECHAM-HAM models but there is an over-estimation for both models. This result suggests that there are too many fine particles and not enough total aerosol mass. Thus it seems that for the CHIMERE and ECHAM-HAM models, there are some dust emissions that are missing or under-estimated.

The highest value is seen on the 8th-9th July at both stations but only CAMS-IFS reproduces this AOD increase. This event, seen at the three Sahelian stations, is clearly related to a mineral dust event because the Angstrom exponent is about 0. It could be linked to dust emission in the Sahel from a Mesoscale Convective System (MCS) that has occurred at the border between Nigeria and Niger (Figure 5).

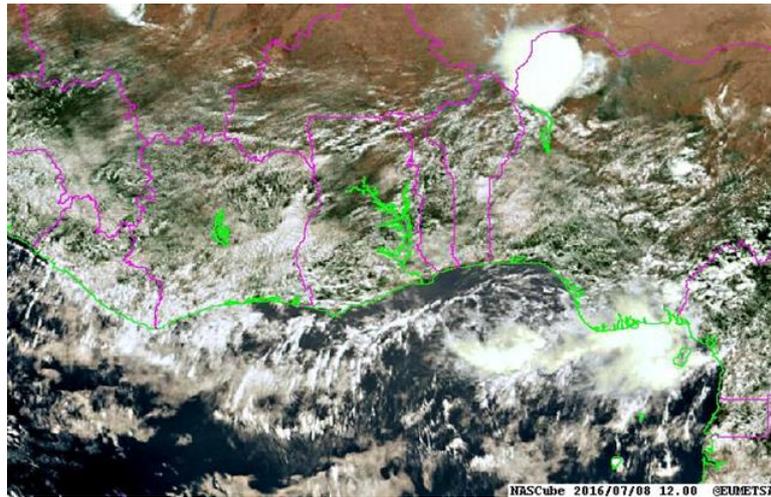


Figure 5 - EUMETSAT visible image on the 7th July 2016 at 12 UTC over the evaluation domain (1°S-15°N;12°W-12°E) downloaded from the NASCube website (Lille University).

In conclusion, the large scale dust transport from the Sahara is in good agreement with observations but the variability linked to the addition from the local dust emissions seems to be missing in the WP3 models, except for CAMS-IFS whose assimilation captures such event.

3.2.2 Central Africa station

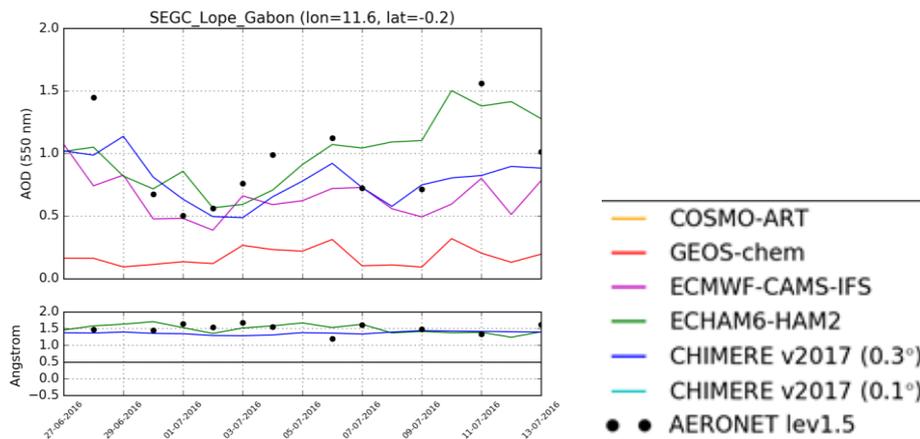


Figure 6 – Time series of average daily AOD and Angstrom exponent modeled by the WP3 models and observed at the central Africa AERONET station (Lope in Gabon) for the period 25th June to 13th July 2016.

In Gabon, close to the vegetation fire area, observed AOD ranges from 0.5 to 1.6 (Figure 6). The WP3 models reproduce this observed range well, for CAMS-IFS from 0.4 to 0.9, for ECHAM6-HAM2 from 0.8 to 1.6, CHIMERE (at 0.3°) from 0.5 to 1.2, but for GEOS-Chem the range is underestimated from 0.1 to 0.3.

The observed variability is not captured by the models, which suggests that the vegetation fires characteristics (emission quantity, injection height, location) or the meteorological variability is not well reproduced.

The modeled and observed Angstrom exponent are well in agreement at about 1.5, which confirms the presence of fine aerosols, probably associated with the vegetation fires.

3.2.3 Guinean coastal stations

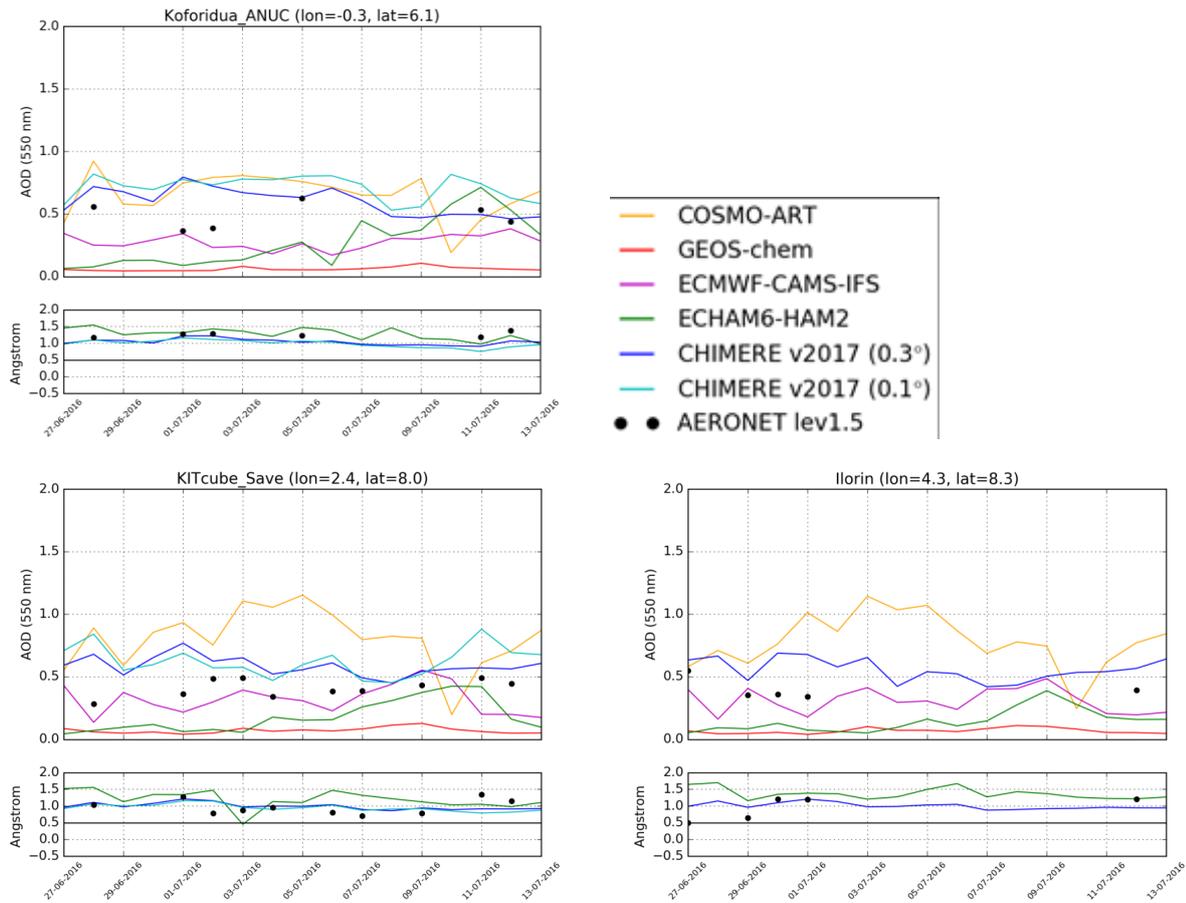


Figure 7 – Time series of average daily AOD and Angstrom exponent modeled by the WP3 models and observed at three Guinean AERONET stations (Koforidua in Ghana, Savè in Benin and Ilorin in Nigeria) for the period 25th June to 13th July 2016.

Over the Guinean coastal region, the observed AOD in Figure 7 range from 0.3 to 0.5. The WP3 models predict for CAMS-IFS from 0.2 to 0.6, for ECHAM-HAM from 0.1 to 0.5, for CHIMERE (at 0.3°) from 0.8 to 0.4, for CHIMERE (at 0.1°) from 0.9 to 0.4, for COSMO-ART from 0.4 to 1.2 and for GEOS-Chem from 0.1 to 0.2.

The modeled variability is higher than the observed one. There is a group of models, which predict AOD with a positive bias (CHIMERE and COSMO-ART) and the other group with a negative bias (ECHAM-HAM, GEOS-Chem, CAMS-IFS). CAMS-IFS is closest to the observations.

The modeled Angstrom exponent is similar for CHIMERE at both resolutions and ECHAM-HAM, which are comparable with the observations.

3.3 Case studies of the 1st and 11th July

Two case studies have been selected, which correspond to specific dates when the three DACCIWA aircrafts were flying in order to quantify the city emissions of Abidjan, Accra, Lomé and Cotonou (*c.f.* section 4.2).

3.3.1 1st July 2016

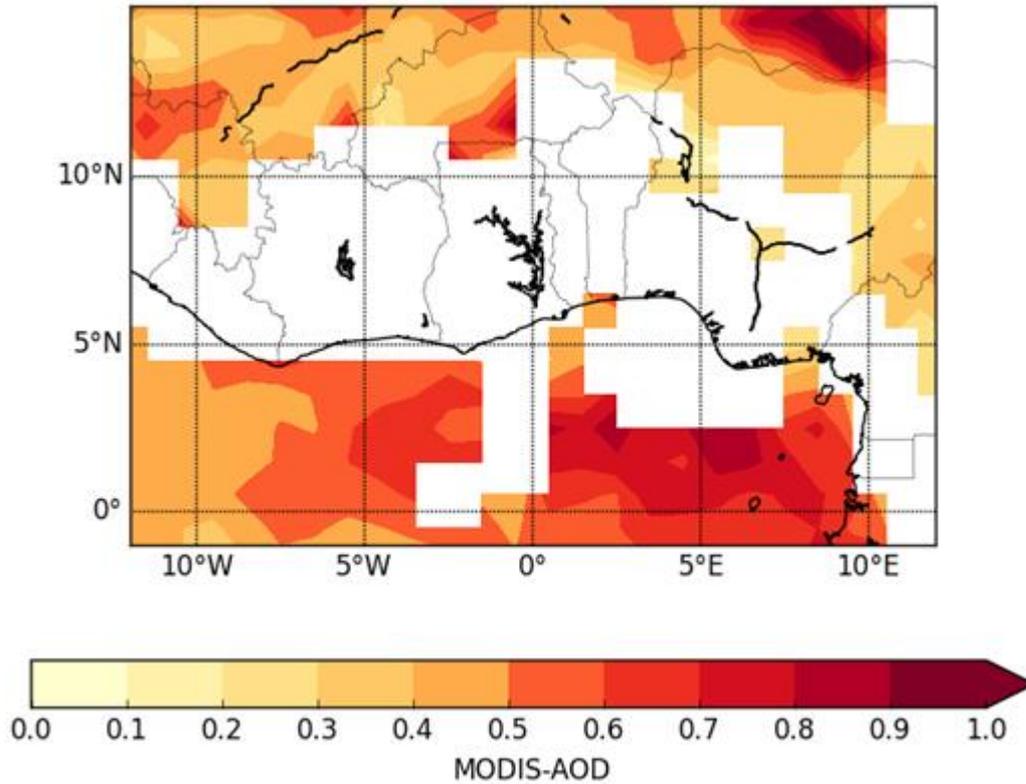


Figure 8 - MODIS-AOD average of the combined Dark-Target and Deep-Blue products acquired by Aqua and Terra (MYD08-D3 and MOD09-D3 products) for the 1st July 2016.

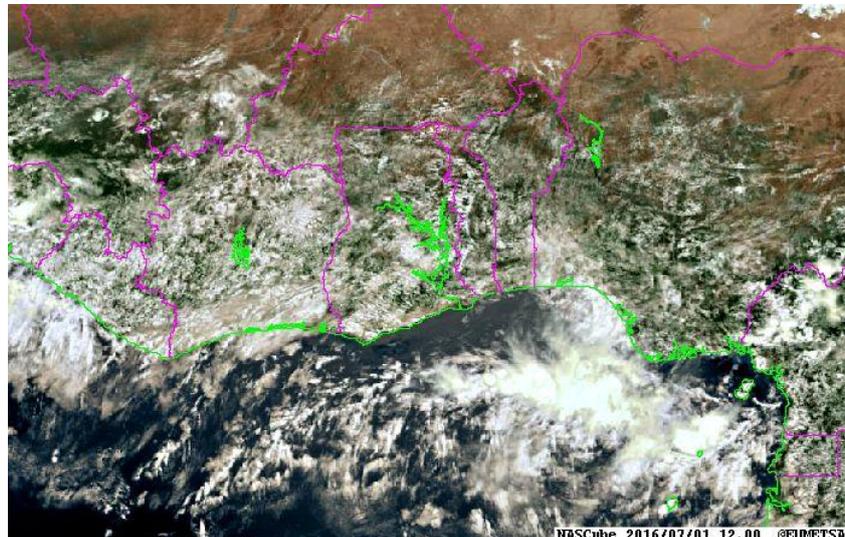


Figure 9 - EUMETSAT visible image on the 1st July 2016 at 12 UTC over the evaluation domain (1°S-15°N;12°W-12°E) downloaded from the NASCube website (Lille University).

On the 1st July, firstly we notice in Figure 8 from the MODIS-AOD observations that there are no values over the coast because of the significant cloud cover over our region of interest (confirmed by EUMETSAT observations in Figure 9). There are two high AOD areas, one over the ocean in the South of Nigeria and one over the South-East of Niger.

Figure 10 presents maps of modeled AOD together with AERONET measurements on the same color scale.

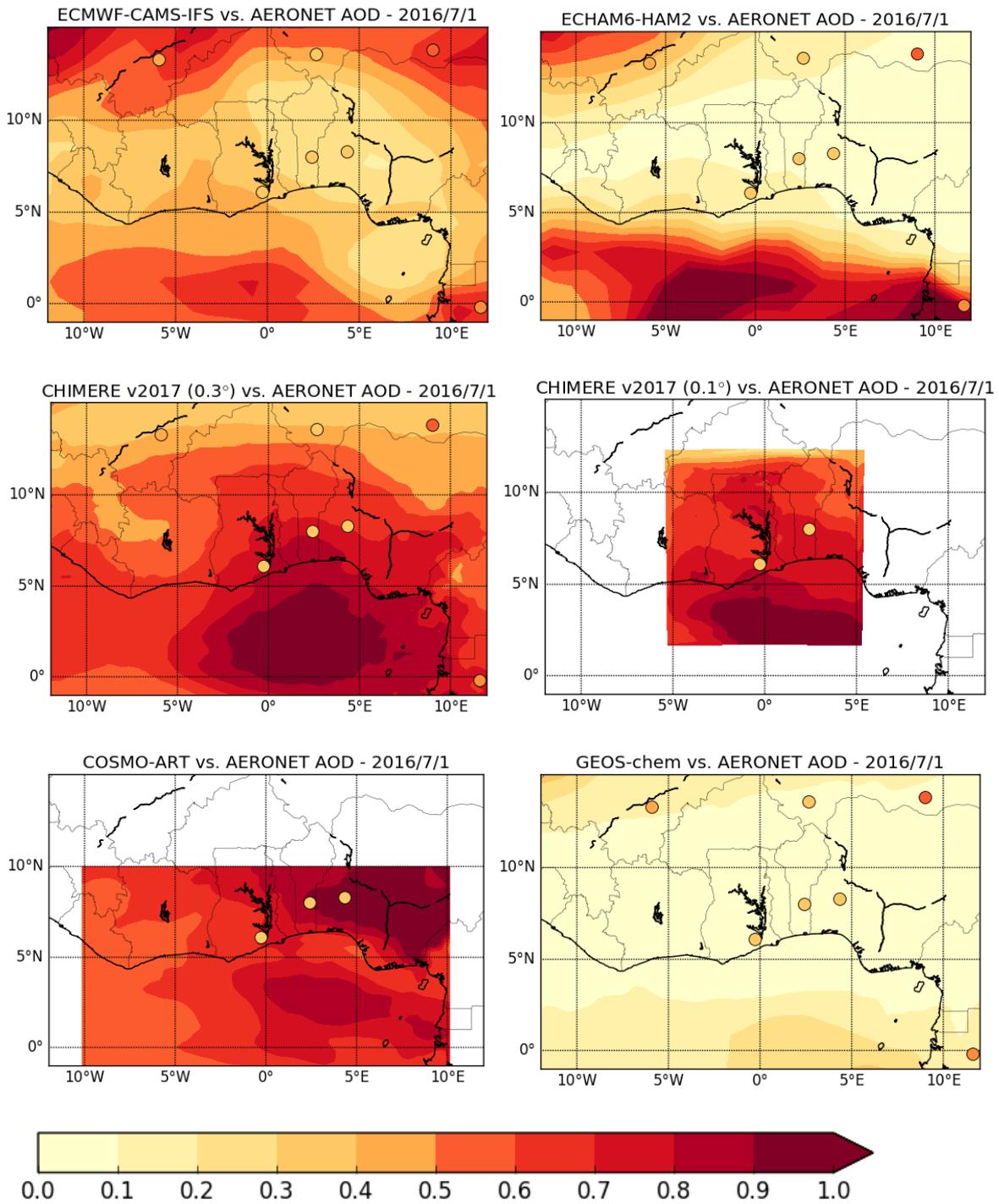


Figure 10 - AOD average of the six models (CAMS-IFS, ECHAM6-HAM2, CHIMERE 0.3° and 0.1°, COSMO-ART, GEOS-Chem) and daily AERONET level 1.5 measurements (dot with the same color scale) for the 1st July 2016.

AERONET-AOD at the seven stations range from 0.4 to 0.5. Over the Sahel and over the ocean, all models predict higher AOD. When looking at the AOD near the coast, observed AOD are about 0.4. The modeled AOD by CAMS-IFS model are between 0.3 and 0.4, for ECHAM-HAM between 0.1 to 0.3, for CHIMERE (at 0.3° and 0.1°) between 0.5 and 0.8, for COSMO-ART from 0.5 to 0.8 and for GEOS-Chem from 0.0 to 0.1.

3.3.2 11th July 2016

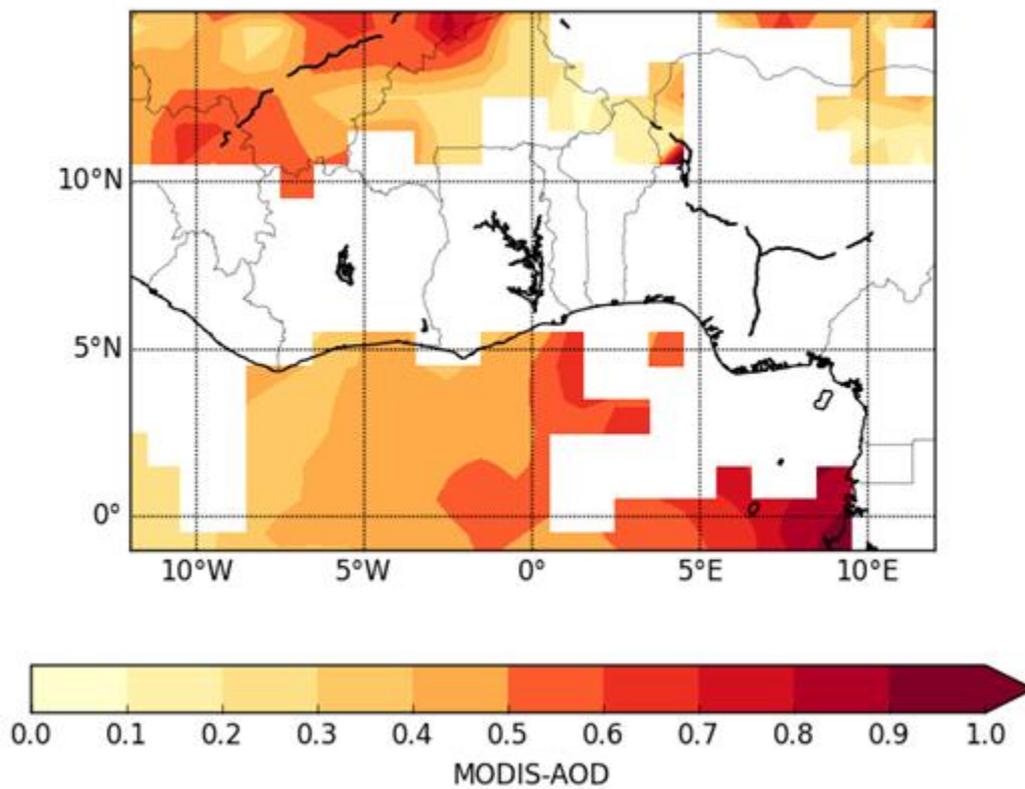


Figure 11 - As Figure 8 but for 11th July 2016.

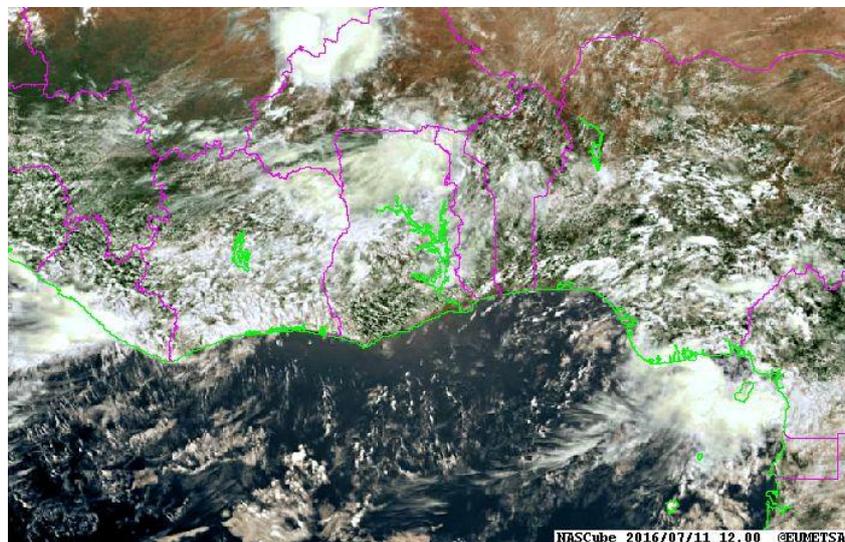


Figure 12 - As Figure 9 but for 11th July 2016.

On the 11th July as for the 1st July, we notice in Figure 11 from the MODIS-AOD observations that there are almost no values over the coast because of the important cloud (confirmed by EUMETSAT observations in Figure 12). There are the same two high AOD areas, which seem to be greater than on the 1st July over the ocean in the South of Nigeria, and lower than the 1st July over the South-East of Niger.

Figure 13 presents maps of modeled AOD together with AERONET measurements on the same color scale.

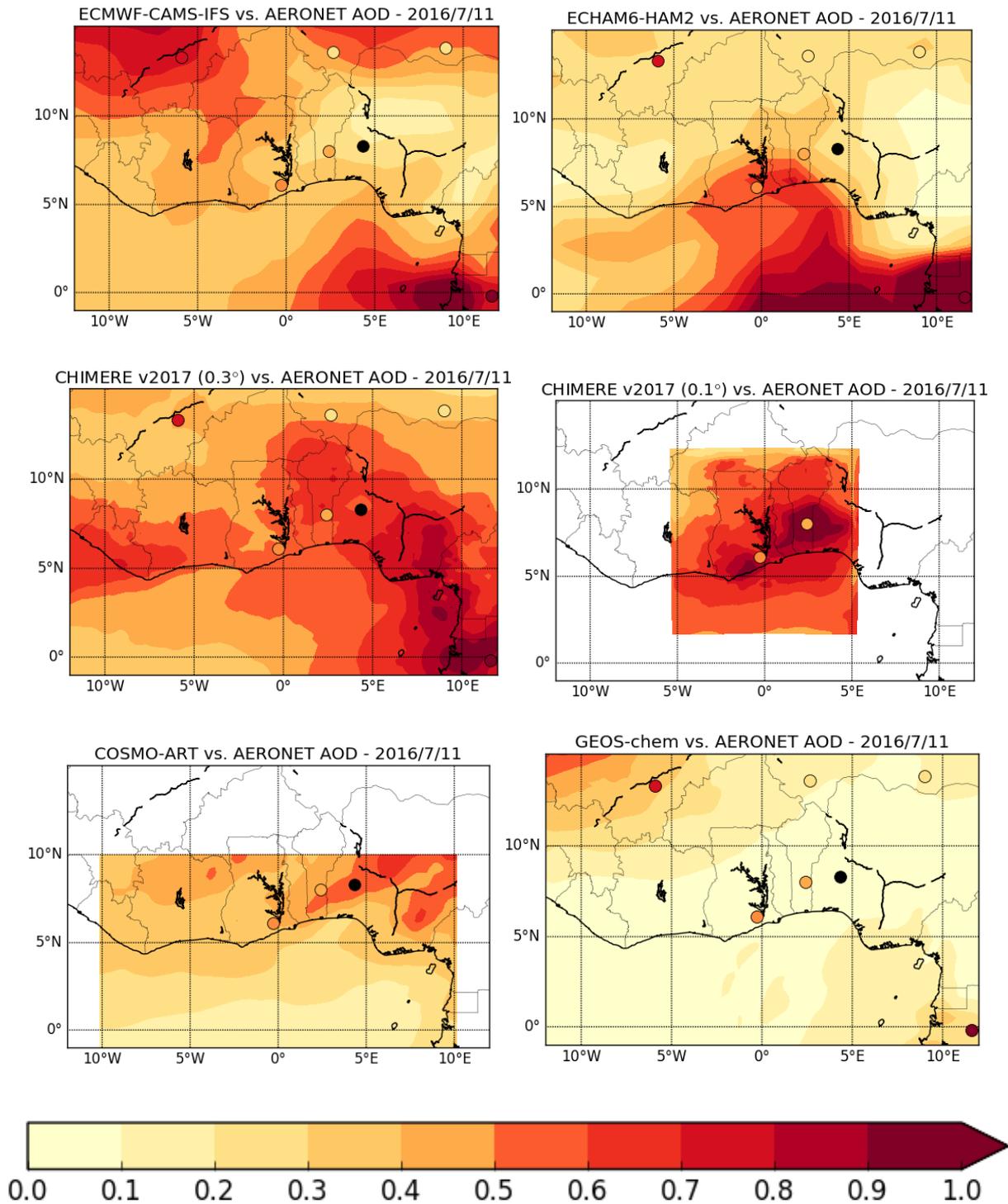


Figure 13 - AOD average of the six models (CAMS-IFS, ECHAM6-HAM2, CHIMERE 0.3° and 0.1°, COSMO-ART, GEOS-Chem) and daily AERONET level 1.5 measurements (dot with the same color scale) for the 11th July 2016. Black dot corresponds to no daily AERONET data available for this day.

AERONET-AOD at the seven stations range from 0.4 to 0.5. Over the Sahel and over the ocean, all models predict higher AOD. When looking at the AOD near the coast, observed AOD are about 0.4. The modeled AOD by CAMS-IFS model are between 0.3 and 0.4, for ECHAM6-HAM2 between 0.3 to 0.6, for CHIMERE (at 0.3° and 0.1°) between 0.5 and 0.9, for COSMO-ART from 0.1 to 0.5 and for GEOS-Chem from 0.0 to 0.1

In conclusion, this section has shown the importance of large scale aerosol transport toward SWA and also the difficulty in estimating the AOD close to the Sahara and the vegetation fires area. This induces an AOD range which is reasonable. However it includes a variability due to mineral dust or biomass burning which does not match the observations. Nevertheless, along the coast, the WP3 models AOD ranges are in good agreement with observations.

4 Gaseous species

This section is dedicated to three gaseous species concentrations: carbon monoxide CO, nitrogen dioxide NO₂ and ozone O₃ that have been monitored during the DACCIWA field campaign. The first part analyzes the observed and modeled hourly temporal variability at a ground-station in Savè (Bénin). The second part shows the comparison between airborne measurements and the modeled concentrations for some specific flights of the three aircraft. In the third part, the focus is on observed and modeled values in the lowest level of the troposphere, the Planetary Boundary Layer (PBL).

4.1 Temporal variability at the Savè ground station

From the CO and O₃ measurements at 60Hz performed by the University Paul Sabatier in Savè (Bénin), hourly averages have been calculated and compared with the model outputs. A spatial bilinear interpolation of the model outputs is performed to be compared with these observations.

4.1.1 Daily evolution

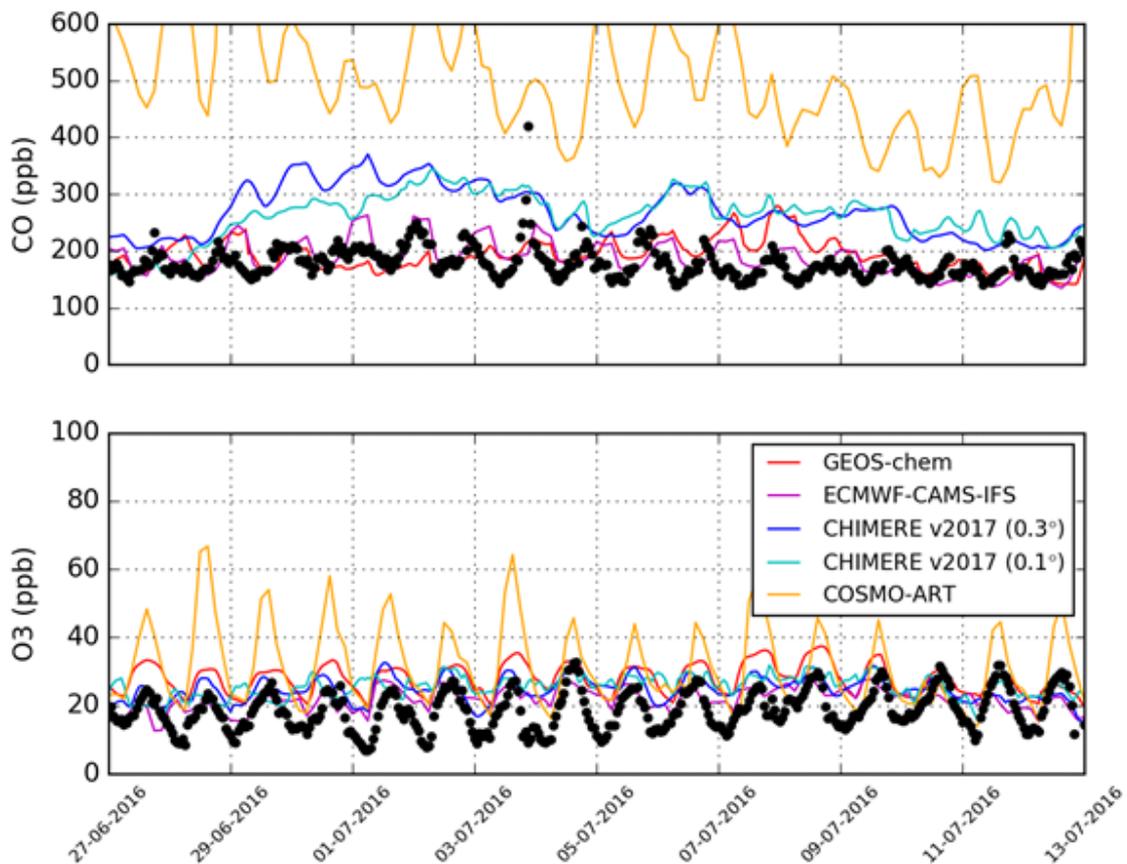


Figure 14 – Time series of average hourly carbon monoxide (CO) and ozone (O₃) concentrations (in ppb) modeled by the WP3 models (CAMS-IFS, CHIMERE 0.3° and 0.1°, COSMO-ART, GEOS-Chem) and observed (black dots) at the DACCIWA field campaign ground station in Savè (Benin) for the period 25th June to 13th July 2016.

Over the campaign period, the concentration of CO was stable at about 200 ppb and O₃ was stable at about 20 ppb (Figure 14). For both pollutants, there are no specific events.

For CO concentration, GEOS-Chem and CAMS-IFS are well in agreement with the observations but the other models simulate a much higher concentration. CHIMERE has a positive bias of about 100 ppb and of about 300 ppb for COSMO-ART. All WP3 models are able to replicate the CO maximum during the afternoon. GEOS-Chem and CAMS-IFS are in good agreement with the

range of the observations but the daily maximum is not well synchronized every day. The CHIMERE and COSMO-ART models predict CO concentrations with an over-estimation for almost the entire period, which suggests an unrealistic biomass burning or anthropogenic emissions, but the diurnal cycle seems to be consistent with the observations. None of the models are able to reproduce the high CO concentrations observed on the 4th July.

For O₃, there is an overall over-estimation by the WP3 models, ranging from a few ppb for GEOS-Chem and CAMS-IFS to more than 10 ppb for COSMO-ART. The period of O₃ increase seems to be related to local production by daytime photo-chemistry, rather than transport because it happens every day.

A clear diurnal cycle is observed every day for both pollutants, for CO from about 160 ppb at 7 UTC to about 200 ppb at 21 UTC, and for O₃, from about 10 ppb at 7 UTC to 30 ppb at 15 UTC. It is worth noticing that the daily maximum of CO occurs at the same time every day, which may be due to the reduction of the PBL height and less vertical mixing. It could also denote an plume coming from the urbanized coast due to the nocturnal wind structure (Parker *et al.* 2005).

There is an event of high CO concentration reaching 400 ppb on 4 July, which is not associated with an increase in O₃.

4.1.2 Diurnal cycle

In the previous section, we have noted a strong day to day cycles for CO and O₃ concentrations. In this section, we focus on modeled and observed CO and O₃ diurnal cycles, which has been computed for the observations (maximum, minimum and mean for each hour) and model outputs (mean for each hour) based on hourly concentrations. We analyze the ability of WP3 models to reproduce both the timing and the intensity adequately (Figure 15).

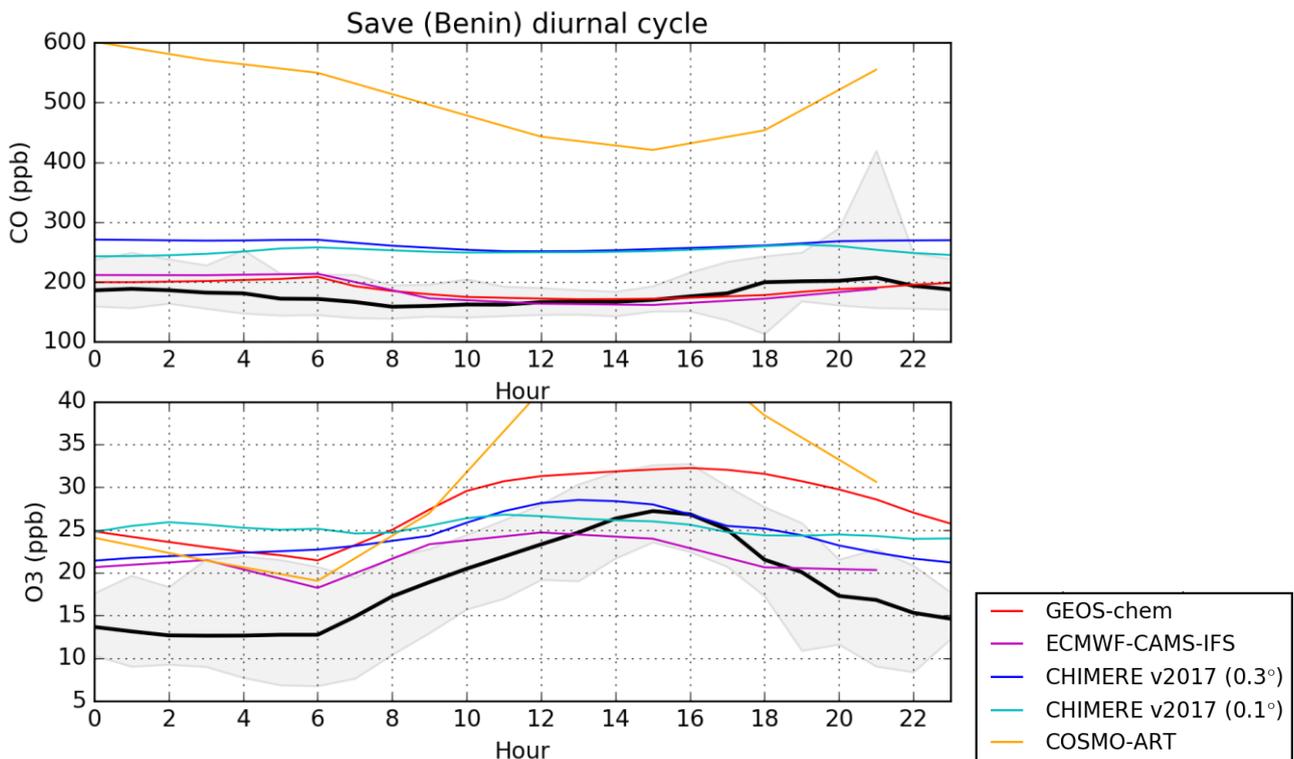


Figure 15 – Average diurnal cycle of carbon monoxide (CO) and ozone (O₃) concentrations (in ppb) modeled by the WP3 models (CAMS-IFS, CHIMERE 0.3° and 0.1°, COSMO-ART, GEOS-Chem) and observed maximum (upper grey limit), minimum (lower grey limit) and mean (black line) for each hour at the DACCIWA field campaign ground station in Savè (Benin) for the period 25th June to 13th July 2016.

The observed CO diurnal cycle has a clear minimum during the daytime at about 170 ppb and it increases at night up to 200 ppb. During the day, there is a constant increase from 8 UTC (about 160 ppb) to 17 UTC. From the beginning of the sunset, the CO concentration increases reaching 200 ppb from 18 UTC to 21 UTC, which suggests that PBL height reduction and/or less vertical mixing and/or enhanced urban pollution transport from SW to NE. After this, there is a decreasing phase until 8 UTC. The WP3 models are divided in two groups: the CHIMERE and the COSMO-ART models over-estimating; the GEOS-Chem and CAMS-IFS models with a very good agreement. All the models predict a decrease during daytime compared to the nighttime.

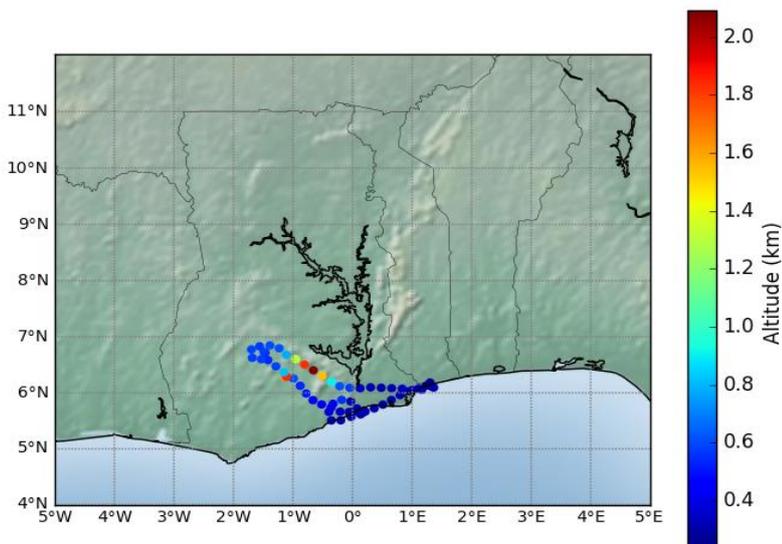
The observed O₃ diurnal cycle is in opposite phase to the CO one because it has a clear maximum during the day at 15 UTC (about 27 ppb) and a minimum from 1 UTC to 6 UTC (about 13 ppb). All the models predict a clear cycle with a maximum between 12 UTC and 16 UTC. There is an over-estimation similar for all the models (> 5 ppb). The increase of 14 ppb from 6 UTC to 15 UTC is well reproduced by the CHIMERE model, the CAMS-IFS model under-estimates the maximum, the GEOS-Chem and COSMO-ART models over-estimate the maximum.

4.2 Spatial variability during the 1st and 11th July case studies

The two case studies selected concern specific dates when the three aircraft were flying in order to sample city emission. In this section, we present CO, NO₂ and O₃ when available as well as a map of the flight trajectory. For each flight, the modeled values have been interpolated to be comparable with the aircraft measurements, in time between the two closest modeled hourly outputs, vertically between the two closest model vertical levels and horizontally with a bilinear interpolation.

4.2.1 1st July 2016

- From 10 to 13 UTC by the French ATR



In the daily summary, the crew have noted that: "Map out Accra plume, circumnavigate Accra, cross the power plant plume of Takoradi and extend west to capture biogenic emissions. This will be followed by further crossing of the power plant plume and a circumnavigation of Kumasi and following the Kumasi plume towards Togo. Sc cloud properties studied on Accra-Kumasi track and south-east of Kumasi."

2016/7/1 between 10 - 13UTC

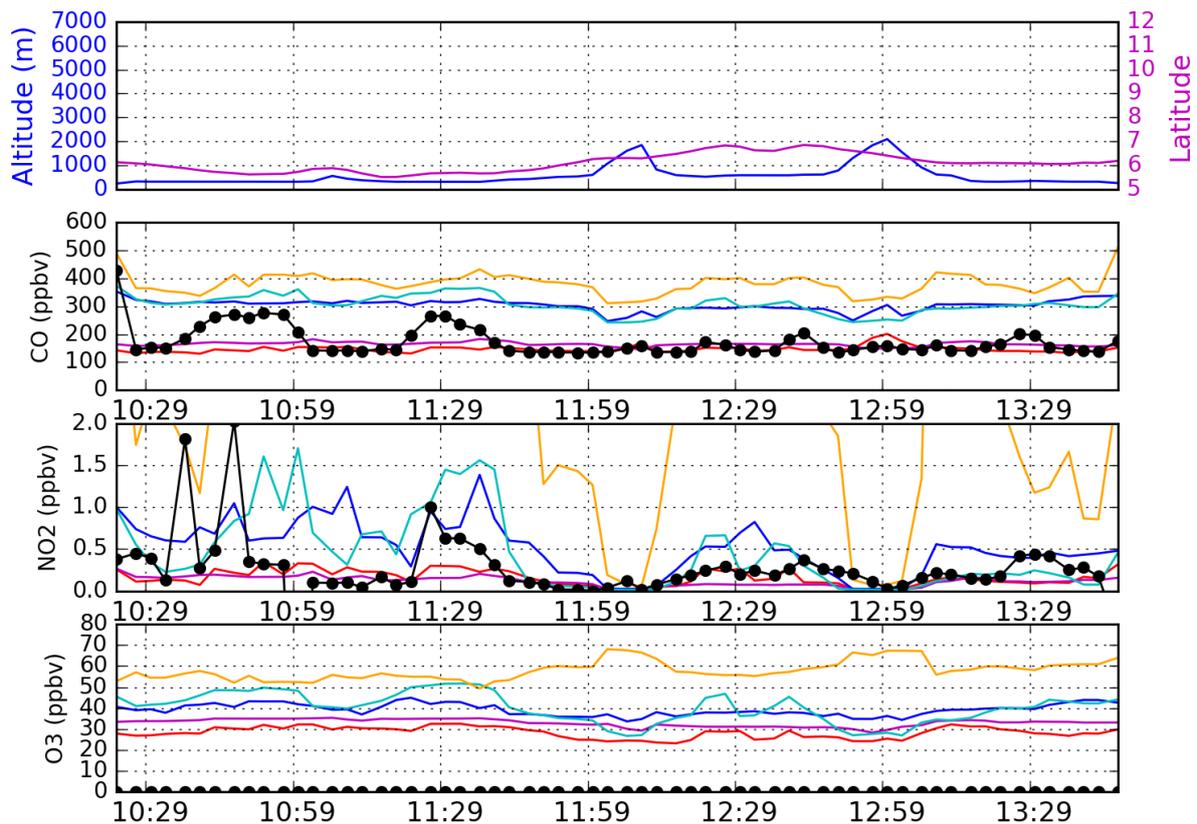


Figure 16 - Map of the flight trajectory on the 1st July 2016 of the French research ATR42 aircraft operated by the SAFIRE team, and the time series of the altitude (in m) with the latitude (in °N), CO concentration (in ppb), NO₂ concentration (in ppb), O₃ concentration (not available for this flight) observed and modeled with the WP3 models: CAMS-IFS (in violet), CHIMERE 0.3° (in dark blue) and 0.1° (in light blue), COSMO-ART (in yellow), GEOS-Chem (in red). Raw airborne observations at 1 Hz are averaged every 3 minutes (black dots).

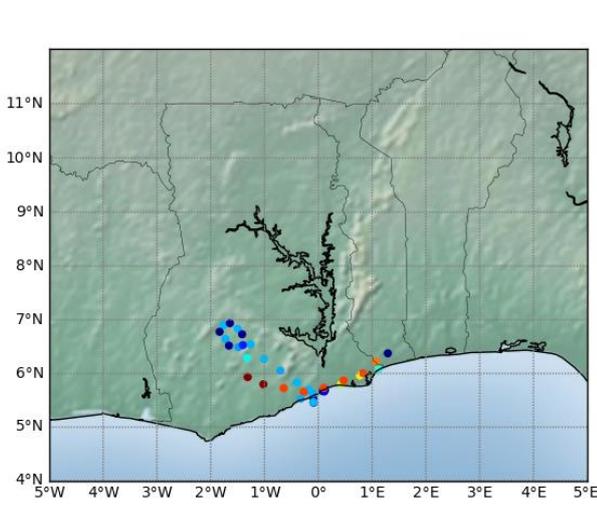
The ATR flight presented in Figure 16 was dedicated to the Accra city emissions. It was conducted mainly below 1000 m in the PBL. The first leg was along the coast until Accra, then flying around Accra and over the Takoradi power plant, then over the forest for biogenic emissions, and back to Lomé.

The observed CO concentration has a baseline at about 150 ppb with some peaks of the concentration up to 280 ppb. These peaks do not seem to be associated with a large scale feature like a biomass burning plume, but rather local plumes when crossing the power plant or the urban plumes. The modeled CO concentration could be divided in two groups, first CAMS-IFS and GEOS-Chem models predicting a correct baseline without any peaks, second CHIMERE and COSMO-ART models over-estimating CO background concentration. None of the models are able to reproduce the variability observed in CO concentration.

The observed NO₂ concentration has a huge variability from less than a ppb to 1.5 ppb because of its short life time. Two peaks are noticed close to Accra up to 1 ppb and two other peaks at the end of the flights, up to 0.3 ppb. The modeled NO₂ concentration presents the same division into two groups, CAMS-IFS and GEOS-Chem models predict the mean concentration well over the entire flight. CHIMERE and COSMO-ART over-estimate the NO₂ concentration around Accra. CHIMERE performs well to reproduce the intensity of the peaks.

The modeled O₃ concentration presents various levels from less than 30 ppb for GEOS-Chem to greater than 60 ppb for COSMO-ART.

- From 11 UTC to 14 UTC by the German Falcon



In the daily summary, the crew have noted that: "Flight included upwind and downwind measurements of Accra and Kumasi and measurement legs between Accra and Kumasi in dissolving stratus in areas with low and moderate pollution. Pollution was detected during the Accra and Kumasi emission sampling and in some measurement sections in dissolving stratus. Cloud measurements could be performed in two cloud layers at about 4000 and 9000 feet."

2016/7/1 between 11 - 14UTC

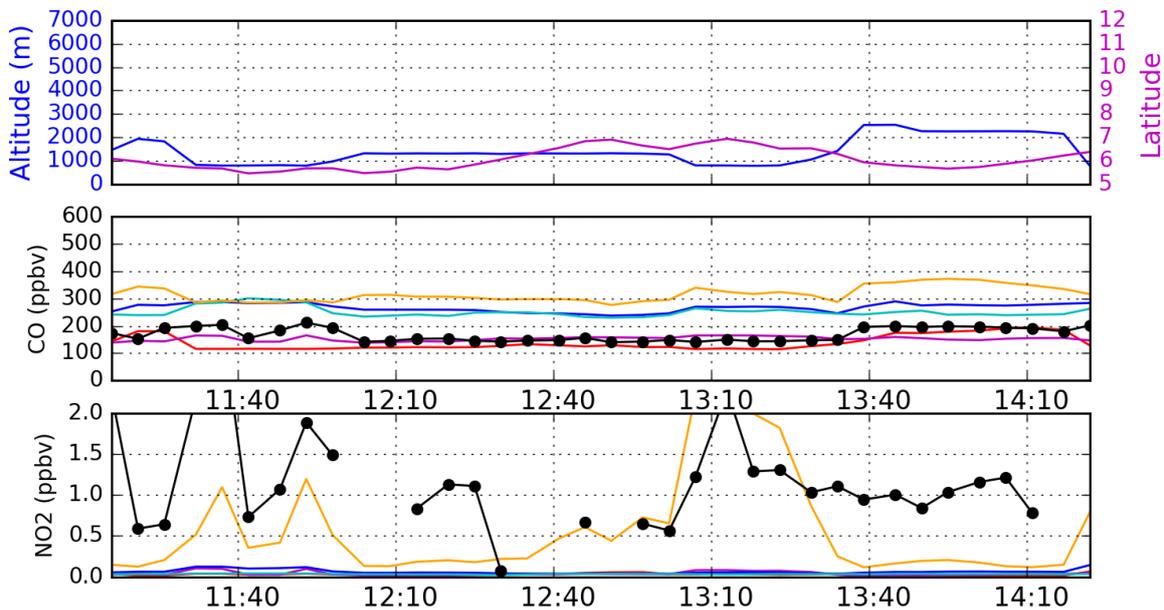


Figure 17 - Map of the flight trajectory on the 1st July 2016 of the German research Falcon aircraft operated by the DLR team, and the time series of the altitude (in m) with the latitude (in °N), CO concentration (in ppb), NO₂ concentration (in ppb) observed and modeled with the WP3 models: CAMS-IFS (in violet), CHIMERE 0.3° (in dark blue) and 0.1° (in light blue), COSMO-ART (in yellow), GEOS-Chem (in red). Raw airborne observations at 1 Hz are averaged every 3 minutes (black dots) with the standard deviation (as error bars).

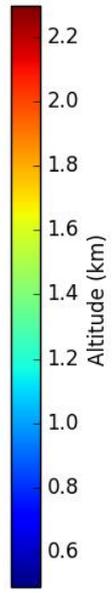
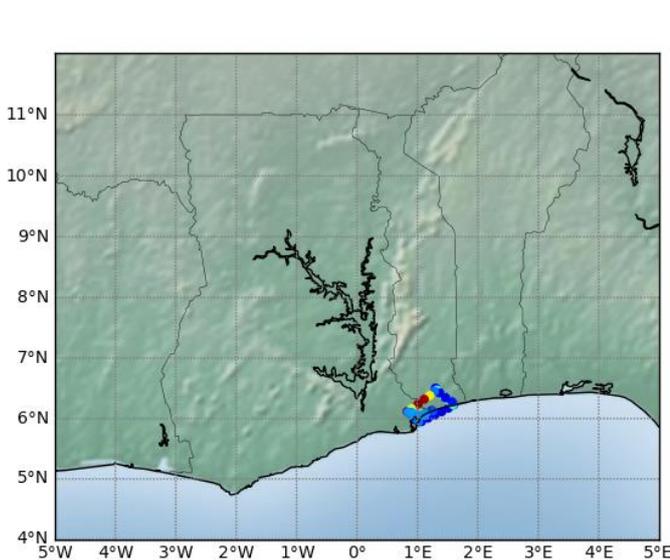
The flight of the Falcon on the 1st July followed a similar trajectory as the ATR one hour later and at higher altitude, which may be close to the limit between PBL and free troposphere. Figure 17 presents the trajectory and the observed and modeled concentrations.

For CO, we observed a baseline concentration at about 150 ppb with two peaks when flying around Accra reaching 200 ppb, whose increase is smaller than for the ATR flight probably because of the higher altitude, and another smaller increase over Kumasi. The modeled concentrations are also divided in two groups, which correspond to two different baseline concentrations at about 150 ppb and at about 250 ppb. It is worth noticing that the COSMO-ART and GEOS-Chem models, and to a lesser extent the CHIMERE model (with only one peak) present the peaks well synchronized with the observations when flying around Accra. It means that the urban plume is well located, thus the anthropogenic emission inventories of these models are realistic. As for the ATR flight measurements made earlier, they are no models able to reproduce the observed CO peak intensities, which could be attributed to anthropogenic emission missing and/or inaccurate plume direction and/or inaccurate PBL height.

For NO₂ concentration, we observed a large variability from > 1 ppb to < 1.5 ppb. The COSMO-ART model performs the best regarding this flight. Some peaks are very well reproduced in time and in magnitude. All other WP3 models do not predict the magnitude of the concentration, but rather a concentration almost stable at 0.1 ppb. This result suggests that either anthropogenic emissions or the height of the PBL are not well modeled except for COSMO-ART. Given that the CHIMERE model has the same anthropogenic emissions, it is most likely related to the height of the PBL, which is too low in all other models.

- **From 14 UTC to 17 UTC by the British Twin-Otter**

In the daily summary, the crew have noted that: "Take off from Lomé at around 2.15 after a successful power change over. Were held by ATC on route to our first waypoint. Did a run upwind of Lomé at 7000ft (above boundary layer). Then descended to 2000 ft on reciprocal run and headed along coast (south of the city) for the downwind runs. Background CO was ~150ppb and NOx ~1ppb. We then did 4 runs (10 minutes each) downwind of the city sampling the plume (at 2000, 2500, 3000 and back at 2000 ft). Some enhancement in CO, NOx, CH4 observed (80ppb, 1ppb, 70ppb respectively) at the 2000 and 2500ft runs. A distinct plume of NOx observed in the lowest run (up to 15ppb), with 2pppb SO2 also observed. We then headed back to the upwind legs (on a run north of the city), ascending to 7000ft to asses BL height (~5500ft). We then did two runs upwind at 3000 and 2500 ft (6 minutes each). We sampled 18 WAS bottles and 4 bags downwind and 8 bottles and 2 bags upwind."



of Lomé at 7000ft (above boundary layer). Then descended to 2000 ft on reciprocal run and headed along coast (south of the city) for the downwind runs. Background CO was ~150ppb and NOx ~1ppb. We then did 4 runs (10 minutes each) downwind of the city sampling the plume (at 2000, 2500, 3000 and back at 2000 ft). Some enhancement in CO, NOx, CH4 observed (80ppb, 1ppb, 70ppb respectively) at the 2000 and 2500ft runs. A distinct plume of NOx observed in the lowest run (up to 15ppb), with 2pppb SO2 also observed. We then headed back to

the upwind legs (on a run north of the city), ascending to 7000ft to asses BL height (~5500ft). We then did two runs upwind at 3000 and 2500 ft (6 minutes each). We sampled 18 WAS bottles and 4 bags downwind and 8 bottles and 2 bags upwind."

2016/7/1 between 14 - 17UTC

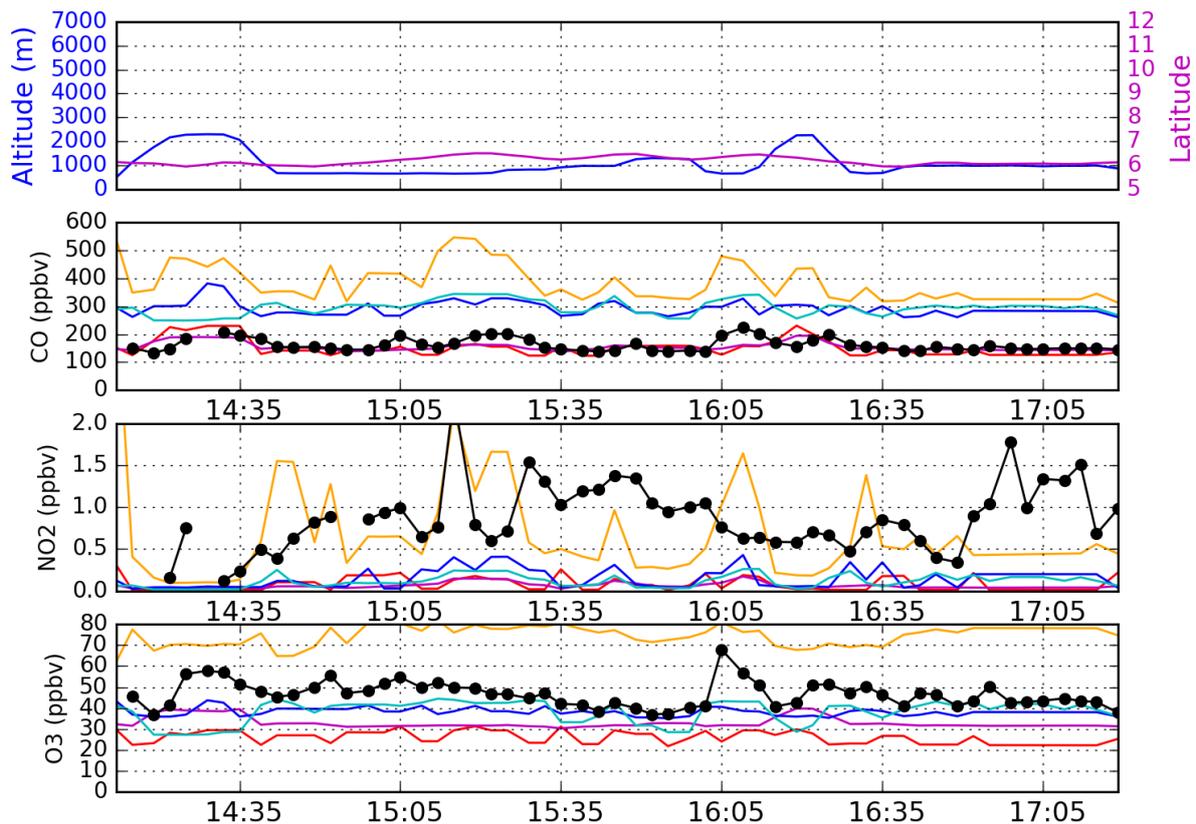


Figure 18 - Map of the flight trajectory on the 1st July 2016 of the British research Twin-Otter aircraft operated by the BAS team, and the time series of the altitude (in m) with the latitude (in °N), CO concentration (in ppb) , NO₂ concentration (in ppb), O₃ concentration (in ppb) observed and modeled with the WP3 models: CAMS-IFS (in violet), CHIMERE 0.3° (in dark blue) and 0.1° (in light blue), COSMO-ART (in yellow), GEOS-Chem (in red). Raw airborne observations at 1 Hz are averaged every 3 minutes (black dots).

- The flight of the Twin-Otter on the 1st July followed a different flight plan than the two others, flying around Lomé. **Figure 18** presents the trajectory and the concentrations. The flight was conducted mainly in the PBL at about 800 m, with two ascending/descending parts above 2000 m.
- For CO, the same two model groups are also relevant to reproduce the baseline concentration. In and above the PBL, there are some increases in the concentration from 150 ppb to 220 ppb. In the PBL, none of the models capture all the CO peaks, but some of the models do capture some of the peaks. Above the PBL (around 2000 m), all models predict an increase probably related to the biomass burning plume.
- For NO₂ concentration, observations range from < 1 ppb to about 1.5 ppb. Modeled variability is lower than observations, with the maximum modeled value by COSMO-ART at about 0.8 ppb. The ambient concentration around Lomé is about 1 ppb, which is under-estimated by all models.
- For O₃, we can see the baseline concentration is about 50 ppb, which is well reproduced by the CHIMERE model. When the plane is above 2000 m in the biomass burning plume, it reaches 70 ppb, which is not associated with high NO₂. Thus it does not seem to be locally created by photo-chemistry, but rather during the transport. None of the models are able to capture this feature.

- **Conclusions for 1 July**

During airborne measurements made on the 1st July, Accra, Kumasi and Lomé city plumes have been sampled, which gives information on how realistic the anthropogenic emission inventories are.

Firstly, for CO, the background concentrations in the PBL are observed at about 150 ppb. The models could be divided in two groups, with a baseline at about 150 ppb for the CAMS-IFS and GEOS-Chem models, and with a baseline at about 250 ppb for the CHIMERE and COSMO-ART models, which suggests a large scale CO bias, probably inaccurate vegetation fire emissions. Each model captures some peaks in the magnitude associated with the urban plume but there are no models which capture all of them, which suggests that an accurate anthropogenic emission inventory would be very valuable for SWA.

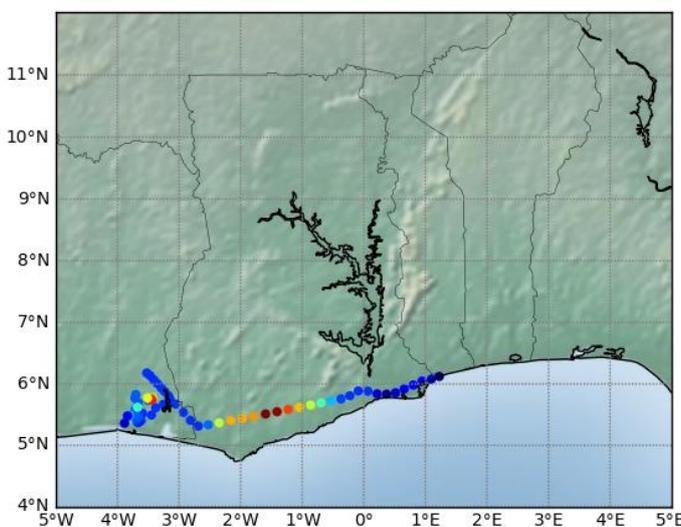
Secondly, for NO₂, observations range between > 1 ppb and < 1.5 ppb with a very high spatial variability. We have noticed that there are no models that are able to reproduce both the background concentration and the peaks associated with the urban plumes.

Thirdly, for O₃, the background concentration is about 50 ppb. The models range from 30 ppb to 70 ppb. The CHIMERE model reproduces well the background concentration. O₃ enhancements have been noticed in the biomass burning plume up to 70 ppb, but none of the models have captured this.

4.2.2 11th July 2016

- **From 7 UTC to 10 UTC by the French ATR42**

In the daily summary, the crew have noted that: *” Lomé – Accra - Abidjan: Way to Accra: Dense low level stratus immediately after TO, descent in more scattered sc to 1000 ft, climb necessary to 3000 ft, in sc layer, complicated to descend to 1000ft. Passing Accra at 3000 ft we decided to climb for sampling BB: 70 ppb O3, 8 µg tot org AMS, 150Mm-1 extinction, beyond- inside- BB layer legs.*



Solely accumulation mode in SMPS. 1000#, Leaving BB layer descending to msa, no way to come down below msa, but lots of µphys (stratus/sc – 4/8 but low) data at msa level all down from SW Ghana and crossing the Abidjan plume. Trial to descend to 1000ft and subsequent immediate climb. ATC sent A/C to track north – crossing of Abidjan plume forth and back, however at 3000ft. Have to have closer look at microphysics across the plum, since plume location has been crossed and localized / characterized. Finally sounding with max BB found more at

2300m (upper limit 2600m). Also again crossing cloud layer during sounding. Difficult for ATC: Monday (flight plan filed Sunday...), Sounding close to Abidjan...”

2016/7/11 between 7 - 10UTC

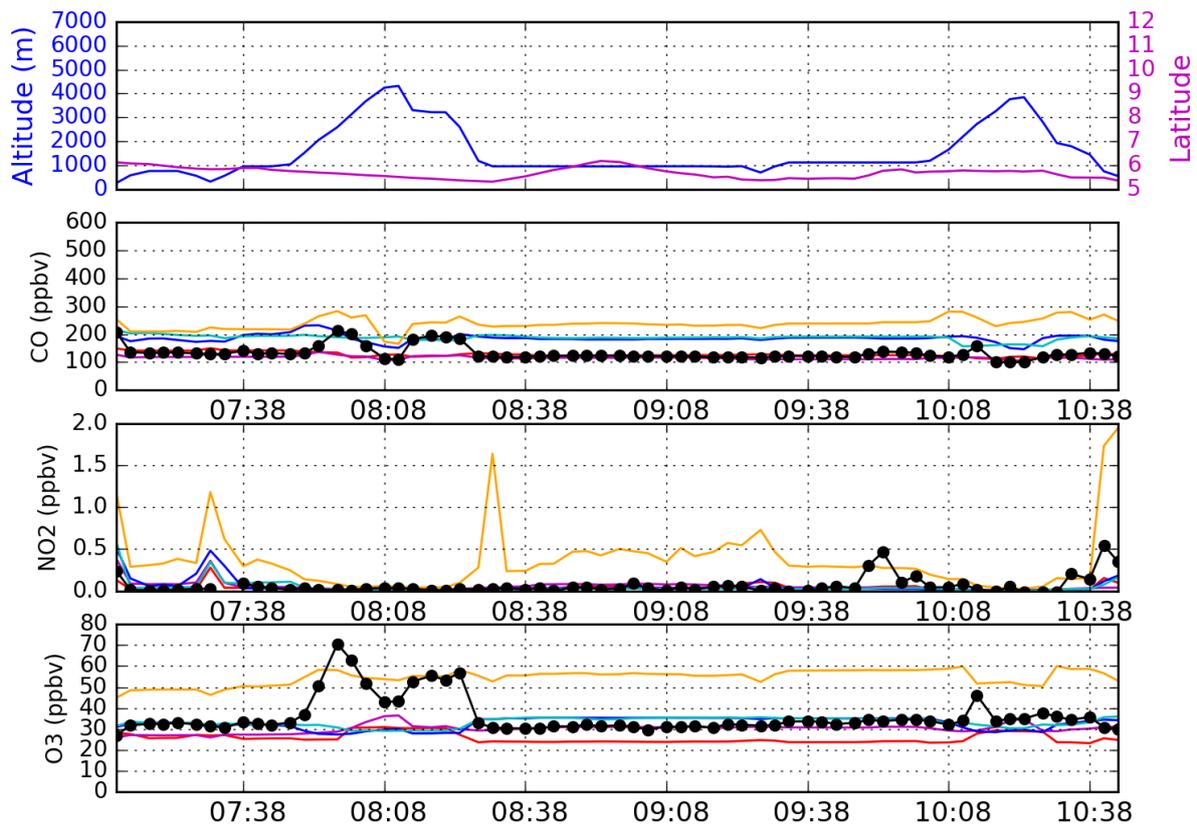


Figure 19 - Map of the flight trajectory on the 11th July 2016 of the French research ATR42 aircraft operated by the SAFIRE team, and the time series of the altitude (in m) with the latitude (in °N), CO concentration (in ppb) , NO₂ concentration (in ppb), O₃ concentration (not available for this flight) observed and modeled with the WP3 models: CAMS-IFS (in violet), CHIMERE 0.3° (in dark blue) and 0.1° (in light blue), COSMO-ART (in yellow), GEOS-Chem (in red). Raw airborne observations at 1 Hz are averaged every 3 minutes (black dots).

This flight of the ATR was conducted to sample both the Accra and Abidjan plumes (Figure 19). During the flight, a biomass burning plume was monitored two times above the PBL.

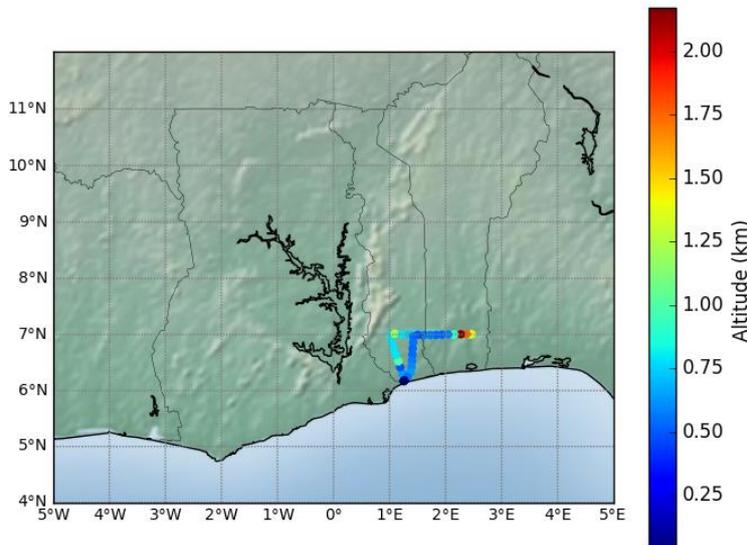
For CO, the background concentration is measured at about 120 ppb. This is reproduced by the GEOS-Chem and CAMS-IFS models but the COSMO-ART and CHIMERE models predict a background concentration at about 200 ppb, which is surprising because the CAMS-IFS and the COSMO-ART models have the same vegetation fires emission dataset (GFAS). It is interesting to note that COSMO-ART seems to capture the biomass burning plume because there are two peaks very well synchronized with the observations.

For NO₂ concentration, the flight did not cross an urban plume below the PBL. At 10:10, the border of the plume was probably measured with an increase up to 0.5 ppb. The models reproduce the low concentration but they do not capture the urban plume, except COSMO-ART, which over-estimates the NO₂ concentration, which may be due to the modeled PBL height.

For O₃ concentration, the baseline is about 30 ppb. The biomass burning plume is associated with high concentrations reaching 70 ppb at 2500 m when ascending and 60 ppb when descending half an hour later. The baseline is well modeled by all models, except COSMO-ART. There are no WP3 models capturing the O₃ increase in the biomass burning plume, which suggests that photo-chemistry producing O₃ has happened during the transport.

- **From 8 UTC to 10 UTC by the British Twin-Otter**

In the daily summary, the crew have noted that:” *Take off 8:22 Took off slightly earlier than planned as clouds were thin so we wanted as much time in them as possible. For this same reason we did cloud work first and aerosol work second. First leg to LS7 was performed initially below cloud then*



a profile above cloud. Cloud was broken so the rest of the leg was performed as an SLR in cloud at 2400 feet (MSA), this was close to our initial cloud top estimate. At around 8:35 on this leg we hit high NOx ~ 5ppb. This was assumed to be the Accra plume. PCASP concentrations remained unchanged. We remained in elevated amounts for ~ 3 mins (~6 nm). PCASP values on this leg were around 750-1000 cm-3 and CDP values were around 500 cm-3. At 8:37 the SMPS reboot started the instrument

working again and it measured a bimodal distribution with modes at 50 and 150 nm. At the beginning of run 2 (W->E) we began sawtoothing through cloud as it seemed more uniform. CDP number concentrations were around 400 cm-3 and cloud top was 3700 ft. Above cloud PCASP measured 900 cm-3 aerosol concentration. The cloud became more broken so we performed the rest of the run at 2400 ft. We think we hit the Accra plume again exiting at ~8:59, at which point typical max in-cloud CDP measurements dropped from 1000-1200 to 500-750 cm-3. At 9:09, we climbed to 3000 ft due to change in MSA at 2° longitude. There was a drop of CDP droplet conc to 400 cm-3 then a rise again to 700 cm-3 which may have been the Lomé plume. We then turned and climbed to 7000 ft to measure any cloud top inversion and sample above cloud aerosol. A biomass burning plume from central Africa had been forecast. We saw a general rise in PCASP concentration and some very narrow sharp peaks up to 7-8000 cm-3. We also saw 180 ppb CO. We then descended back it to the BL and PCASP below cloud measured 1500-1800 cm-3. We reached minimum alt of 1500 ft at 2 deg 6 sec E. Drizzle was spotted on the CIP during the reciprocal run. We had to abort this run slightly early to recover home. Filters were exposed from 09:53:55 - 10:16:55. They may have been exposed to a few seconds of cloud at the end. Landed 10:24”

2016/7/11 between 8 - 10UTC

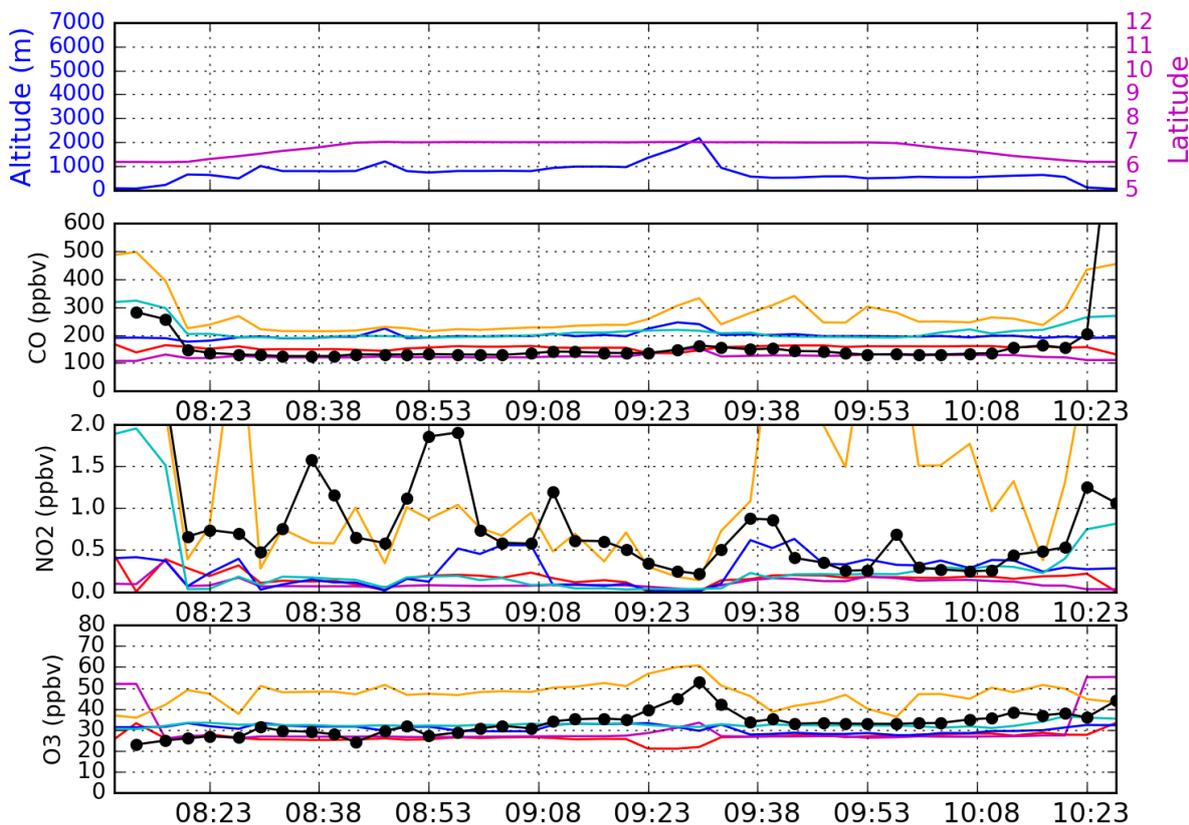


Figure 20 - Map of the flight trajectory on the 11th July 2016 of the British research Twin-Otter aircraft operated by the BAS team, and the time series of the altitude (in m) with the latitude (in °N), CO concentration (in ppb) , NO₂ concentration (in ppb), O₃ concentration (in ppb) observed and modeled with the WP3 models: CAMS-IFS (in violet), CHIMERE 0.3° (in dark blue) and 0.1° (in light blue), COSMO-ART (in yellow), GEOS-Chem (in red). Raw airborne observations at 1 Hz are averaged every 3 minutes (black dots).

This flight presented in Figure 20 was dedicated to stratus clouds. Flying between 1000 m and 2000 m, it has probably sampled the Lomé and the Accra plumes. The biomass burning plume has been crossed on the way back to Lomé airport.

For CO concentration, it is the same situation as for the ATR with the measured background of about 120 ppb. The concentration is very stable during the entire flight (except close to the airport). There are two groups of models, the GEOS-Chem and CAMS-IFS models are in good agreement, and the COSMO-ART and CHIMERE models over-estimate the background concentration with an 80 ppb bias. There is a small increase in the CO concentration when crossing the biomass burning plume from 120 ppb to 180 ppb.

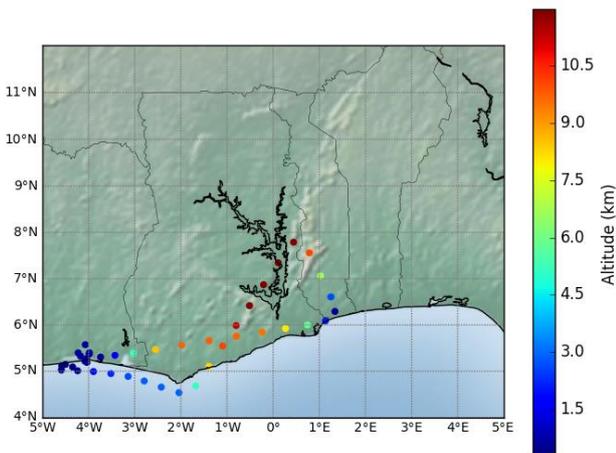
For NO₂ concentration, high levels are measured from the beginning in the Lomé plume then in the Accra plume from 0.5 ppb to 2 ppb. The COSMO-ART model is able to capture the Accra urban plume very well in term of magnitude and localization. All other models do not capture this plume.

For O₃ concentration, firstly near the airport it is very low (about 20 ppb) in high NO_x level. The concentration in the plume at 1000 m ranges from 20 ppb to 30 ppb. The WP3 models reproduce this level except COSMO-ART. When crossing the biomass burning plume, O₃ was measured up

to 50 ppb. The WP3 models did not reproduce this increase of 20 ppb. The CAMS-IFS and COSMOS-ART models simulate an increase.

- **From 10 UTC to 14 UTC by the German Falcon**

In the daily summary, the crew have noted that: *”Flight included upwind and downwind measurements by flying two circles around Abidjan at 2000 ft. After descending to 1000 ft above the ocean, one upwind leg and three downwind legs are performed to measure emission from an oil platform in the Espoir oil field. Between Abidjan and Accra a biomass burning layer was sampled between 7000 and 9000 ft (slow profile from 7000 to 9000ft, afterwards ~15min sampling at 9000ft). Climbing to FL370 a ~2km thick MCS outflow layer was sampled.”*



2016/7/11 between 10 - 14UTC

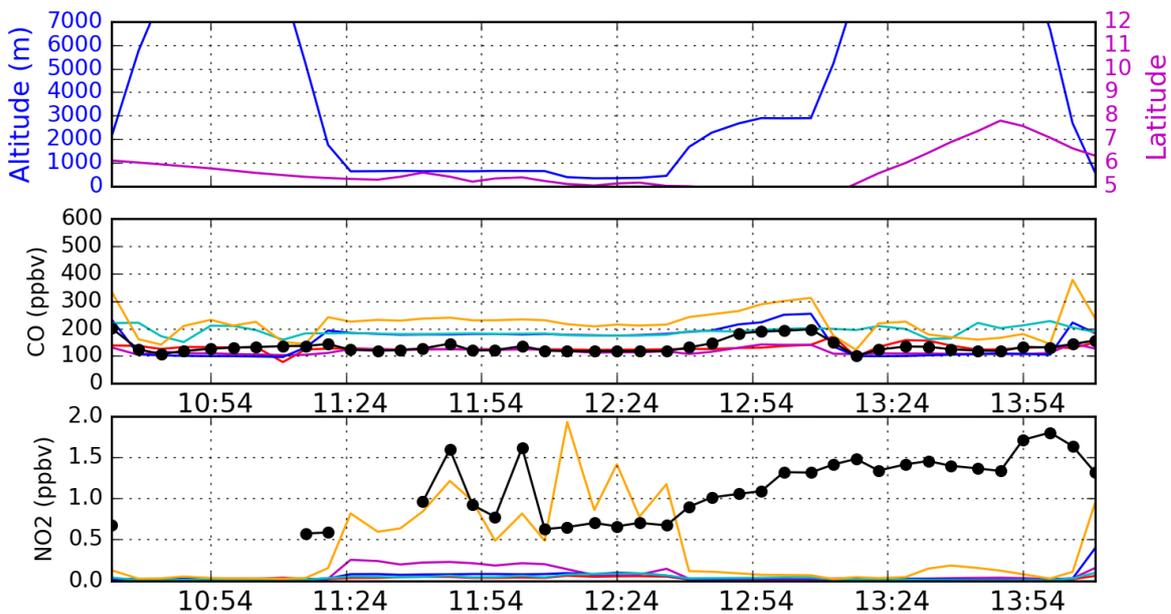


Figure 21 - Map of the flight trajectory on the 11th July 2016 of the German research Falcon aircraft operated by the DLR team, and the time series of the altitude (in m) with the latitude (in °N), CO concentration (in ppb), NO₂ concentration (in ppb) observed and modeled with the WP3 models: CAMS-IFS (in violet), CHIMERE 0.3° (in dark blue) and 0.1° (in light blue), COSMO-ART (in yellow), GEOS-Chem (in red). Raw airborne observations at 1 Hz are averaged every 3 minutes (black dots).

The objectives of this flight is to consider the Abidjan city emissions and also the pollution over the ocean from the ships as well as an oil platform (Figure 21). The second half of the flight sampled a biomass burning plume, then a mesoscale convective system (MCS) outflow.

For CO, the background concentration is also measured at about 120 ppb. The CO concentration is very stable from 1000 m to 8000 m altitude. There is only an increase associated with the

biomass burning plume from 120 ppb to 200 ppb. The separation into two model groups is still true, the GEOS-chem and CAMS-IFS models are in good agreement, and the COSMO-ART and CHIMERE models over-estimate the background concentration with a 80 ppb bias. Surprisingly, the higher resolution of CHIMERE predicts higher CO concentrations from 1000 m to 8000 m altitude, which could be linked to the different meteorology. Nevertheless, the higher resolution of CHIMERE captures well the CO concentration increase in the biomass burning plume.

For NO₂ concentration, when the Falcon arrived in the Abidjan area the NO₂ ranged between 0.5 ppb and 2 ppb. A clear decrease is associated with the biomass burning plume and the MCS outflow. All WP3 models predict an increase over Abidjan but there is only COSMO-ART able to capture the variability. It is particularly interesting to note that in the MCS outflow, NO₂ concentration are very stable at about 1.5 ppb from 3000 m to 8000 m, which suggests that fresh urban air has been lifted up quickly by the convection. All models predict very low concentrations because the meteorological fields do not capture this MCS.

- **Conclusions for 11 July**

The CO and O₃ background concentrations have been measured by the three aircrafts at about 120 ppb for CO and about 30 ppb for O₃. The WP3 models could be separated in two groups, the GEOS-Chem and CAMS-IFS models reproducing the CO background concentrations, and the COSMO-ART and CHIMERE models over-estimating the CO background concentration with an 80 ppb bias. For O₃, the WP3 models are able to reproduce the O₃ background concentration except COSMO-ART. Some enhancements up to 200 ppb for CO and up to 70 ppb for O₃ have been observed associated with the biomass burning plume by the three teams. But the WP3 models struggle to capture this important feature, especially the O₃ level in the plume, which suggests that the photo-chemistry happening during the transport from Central Africa to the Guinean Gulf is under-estimated in our state-of-the-art CTM panel.

The fresh urban plumes, monitored by the NO₂ concentration, are well captured by COSMO-ART in terms of range and variability except in the morning. All other models under-estimate the NO₂ concentration during the day, which could be linked to a low modeled PBL height and/or to inaccurate anthropogenic emissions. We have also seen that high NO₂ concentrations are observed in MCS outflow from 3000 m to 8000 m altitude, which have not been modeled, probably because the convective system was missing in the numerical simulations.

4.3 Gaseous species concentrations in the Planetary Boundary Layer

This section focuses on the PBL concentrations in order to analyze the local pollution, which is effectively breathed by the population. The raw airborne measurements are averaged in time every 3 minutes and compared with interpolated modeled concentrations (same as in the previous section). This represents more than 27 flight hours for CO and NO₂, and more than 16 hours for O₃.

4.3.1 Carbon monoxide in the PBL

Table 3 presents the modeled and observed statistics for CO concentration in the PBL. Some high CO concentrations could be attributed to the airport area, but the majority of the observations has been done flying around some of the major Guinean cities: Abidjan, Accra, Lomé and Cotonou. The observations range from 112 ppb to 1111 ppb with a mean value of 170 ppb. The models are divided in two groups: the GEOS-Chem and CAMS-IFS models, which have a low negative bias (-8 ppb and -30 ppb respectively); the CHIMERE and COSMO-ART models, which have a high positive bias (> 80 ppb). The observed and modeled minimum values follow the same separation. The maximum CO concentration is too low for all WP3 models with the exception of the COSMO-ART model which has the broadest concentration range.

Overall the scores of the five models show that the CO variability in the PBL is not in agreement with the observations, except COSMO-ART capturing a part of the variability (R = 0.2). The GEOS-Chem and CAMS-IFS models do not present the systematic bias, whereas the CHIMERE and COSMO-ART models have a positive bias.

Table 3 – CO concentration (ppb) comparison between airborne observations averaged every 3 minutes in the PBL (lower than 500 m altitude) and modeled values interpolated along the flight track by the WP3 models: CAMS-IFS (in violet), CHIMERE 0.3° (in dark blue) and 0.1° (in light blue), COSMO-ART (in yellow), GEOS-Chem (in red).

WP3 models	N	max obs	max mod	min obs	min mod	mean obs	mean mod	R	RMSE	Bias
GEOS-Chem	555	1111.09	280.2	112.44	71.63	169.68	161.86	-0.1	93.73	-7.82
CAMS-IFS	555	1111.09	191.69	112.44	70.67	169.68	141.56	0.01	89.43	-29.76
CHIMERE (0.3°)	555	1111.09	353.51	112.44	173.84	169.68	252.69	0.02	122.63	83.02
CHIMERE (0.1°)	555	1111.09	397.47	112.44	174.42	169.68	259.08	0.08	124.62	89.4
COSMO-ART	555	1111.09	882.65	112.44	168.1	169.68	334.82	0.19	226.65	165.14

4.3.2 Nitrogen dioxide in the PBL

Table 4 presents the modeled and observed statistics for NO₂ concentration comparison in the PBL. As in the previous section, some high concentrations could be attributed to the airport areas. Observed NO₂ concentrations from the different flights of the three aircraft range from 0 (because of the detection limit which is different for the instruments in the three aircraft) to 23.87 ppb, with a mean value of 0.59 ppb.

Except the COSMO-ART model, all WP3 models under-estimate the mean and maximum values. The results seem to be linked with the resolution, with the best scores for CHIMERE at 0.1°. The scores demonstrate that it is particularly difficult to capture the NO₂ variability because of its short

lifetime (some hours in the PBL). The COSMO-ART model is able to reproduce the observed range of two orders of magnitude.

Table 4 – NO₂ concentration (ppb) comparison between airborne observations averaged every 3 minutes in the PBL (lower than 500 m altitude) and modeled values interpolated along the flight track by the WP3 models: CAMS-IFS (in violet), CHIMERE 0.3° (in dark blue) and 0.1° (in light blue), COSMO-ART (in yellow), GEOS-Chem (in red).

<u>WP3 models</u>	N	max obs	max mod	min obs	min mod	mean obs	mean mod	R	RMSE	Bias
GEOS-Chem	555	23.87	1.42	0	0	0.59	0.19	0.07	1.64	-0.4
CAMS-IFS	555	23.87	0.64	0	0.01	0.59	0.17	0.01	1.75	-0.49
CHIMERE (0.3°)	555	23.87	1.39	0	0.03	0.59	0.34	0.01	1.63	-0.25
CHIMERE (0.1°)	555	23.87	2.44	0	0.02	0.59	0.3	0.08	1.64	-0.29
COSMO-ART	555	23.87	11.56	0	0.11	0.59	3.07	0.06	3.75	2.48

4.3.3 Ozone in the PBL

Table 5 presents the modeled and observed statistics for O₃ concentration in the PBL. The observations range from 11 ppb to 54 ppb with a mean value of 30 ppb. The COSMO-ART and GEOS-Chem models perform best to reproduce this observed range, however COSMO-ART has a significant positive bias (about 14 ppb). The CAMS-IFS model over-estimates O₃ in the PBL with the highest RMSE. The CHIMERE model is able to reproduce a part of the variability but the higher resolution does not improve the correlation coefficient (0.29 compared to 0.28). Overall the scores of the five models show that the modeled O₃ variability in the PBL does not fit with the observations, except CHIMERE capturing a part of the variability (R = 0.3).

Table 5 – O₃ concentration (ppb) comparison between airborne observations averaged every 3 minutes in the PBL (lower than 500 m altitude) and modeled values interpolated along the flight track by the WP3 models: CAMS-IFS (in violet), CHIMERE 0.3° (in dark blue) and 0.1° (in light blue), COSMO-ART (in yellow), GEOS-Chem (in red).

<u>WP3 models</u>	N	max obs	max mod	min obs	min mod	mean obs	mean mod	R	RMSE	Bias
GEOS-Chem	321	53.96	52.45	11.73	22.47	30.33	31.34	0.03	8.2	1.01
CAMS-IFS	321	53.96	74.75	11.73	25.48	30.33	40.56	0.07	19.95	9.87
CHIMERE (0.3°)	321	53.96	43.5	11.73	26.62	30.33	35.64	0.28	8.16	5.31
CHIMERE (0.1°)	321	53.96	46.36	11.73	29.81	30.33	37.06	0.29	9.04	6.73
COSMO-ART	321	53.96	58.22	11.73	19.9	30.33	44.08	-0.15	16.9	13.76

5 Conclusions

The five models GEOS-Chem, CAMS-IFS, COSMO-ART, CHIMERE with two resolutions and ECHAM-HAM (only aerosols available) were compared to observations in order to assess the aerosol and gaseous concentration spatial and temporal variability across SWA. A special focus was set to the different emission inventories to interpret the results.

In the first part, the modeled aerosol content by the WP3 models has depicted important discrepancies in adequately reproducing the observed spatial features. There are three main areas: in the North of the evaluation domain over the desert, where aerosols are mostly constituted of mineral aerosols; in the South over the Guinean gulf, where aerosols are mostly constituted from vegetation fires pollution, in the centre over the urbanized coastal area, where the pollution is mostly anthropogenic. The CAMS-IFS model performed the best because it includes MODIS-AOD assimilation.

At ground-stations in the Sahel, a high AOD temporal variability has been observed, which is captured by the CAMS-IFS and the CHIMERE models. In Gabon, high AOD over 0.5 are observed, which is consistently modeled by CAMS-IFS, CHIMERE and ECHAM-HAM. Over the coastal region, we have noted that the daily observed AOD is very stable from 0.3 to 0.6, which is underestimated by GEOS-Chem, ECHAM-HAM and CAMS-IFS, and which is slightly over-estimated by CHIMERE and COSMO-ART.

In the second part, gaseous concentrations of three major pollutants (CO, NO₂ and O₃) have been investigated at Savè (Bénin). There is a clear diurnal cycle every day for CO and O₃ concentrations. During daytime, there is a minimum for CO and a maximum for O₃. All WP3 models predict these cycles but the magnitude of the cycles is in good agreement with observations only for CO modeled by the GEOS-Chem and CAMS-IFS models.

Comparing modeled and observed concentrations in the PBL around some of the major Guinean cities: Abidjan (Ivory Coast), Accra (Ghana), Lomé (Togo) and Cotonou (Bénin), we have shown the lack of consistency between the models. The range of the modeled values matches with the observations but the variability of the three studied pollutants is not reproduced by the WP3 models. In the PBL, the evaluation has highlighted the important improvements that could be achieved using accurate anthropogenic emission inventories.

The evaluation has also depicted two specific dates corresponding to city emission objective flights for both aerosols and gases together with some meteorological elements. On the 1st and 11th July 2016, the aerosol content presents a spatial structure with low AOD close to the coast (about 0.3) and higher AOD over the Guinean Gulf and over Niger. We have seen CO range from 150 ppb to 220 ppb in the biomass burning plume, and up to 300 ppb locally within power plant plumes, which is well modeled by CAMS-IFS and GEOS-Chem. NO₂ concentrations present an important variability in urban plumes from 0.5 ppb to 2 ppb, which is well captured by the COSMO-ART and the CHIMERE models depending on the region. O₃ background concentrations are between 30 ppb and 40 ppb. Some enhancements reaching 70 ppb is noticed in the biomass burning plume. The WP3 models seem to under-estimate the photo-chemistry happening in the biomass burning plume.

We have seen that each model has specific strengths:

- The CAMS-IFS model has the best large-scale features of dust and biomass burning aerosol contents, thanks to the MODIS-AOD assimilation. The background concentration of CO and O₃ are also well in agreement with observations above the PBL.
- The GEOS-Chem model captures background concentrations of CO and O₃ are in good agreement with observations above the PBL and also in the PBL.
- The COSMO-ART model performed the best to reproduce the urban plumes location in the PBL.
- The CHIMERE model performed the best in the PBL to reproduce the NO₂ concentration variability. The CHIMERE model has been used with two different resolutions, which have led to some improvements to capture the NO₂ variability but not systematically. Consequently, the increase of resolution and computational time does not seem to be critical in order to improve the results.

We have pointed out three critical aspects explaining the spread of the model results: the meteorological fields in the PBL, the anthropogenic and vegetation fire emissions datasets. To begin with, accurate air mass dynamics in the PBL is needed, notably the diurnal evolution of the PBL height should be deeply investigated.

Four different anthropogenic emission inventories have been used in this evaluation: HTAP 2010, EDGAR v4.2 2008, ACCMIP RCP4,5 and MACCity. In the framework of the DACCIWA project in the WP2, a new anthropogenic emission inventory has been recently delivered. This model assessment has clearly shown how it could be valuable to get more accurate location and magnitude of the urban emissions.

For the vegetation fire datasets, there were four datasets used: GFED, GFAS, APIFLAME and ACCMIP RCP4,5. Figure 22 presents the vegetation fires detected by MODIS during the month of June 2016 that was presented on the 4th July during the daily meteorological and pollution forecast of the field campaign. It shows the huge area and number of fires occurring in this season in Central Africa. The results of this deliverable have demonstrated that the representation of this pollution source is of prime importance for the Guinean coastal region. Our results show clearly that the background level of CO depends on the vegetation fire inventory used.

The biomass burning plume is a ubiquitous feature of the pollution along the coastline. The three aircrafts have sampled this layer multiple times, where we have noticed CO and O₃ concentration enhancements. The WP3 models struggle to reproduce this layer. When it is reproduced, the O₃ level within the layer is not consistent with observations. It constitutes a very interesting research pathway because the photo-chemistry taking place in the biomass burning plumes during the transport over the ocean is not reproduced by any WP3 model.

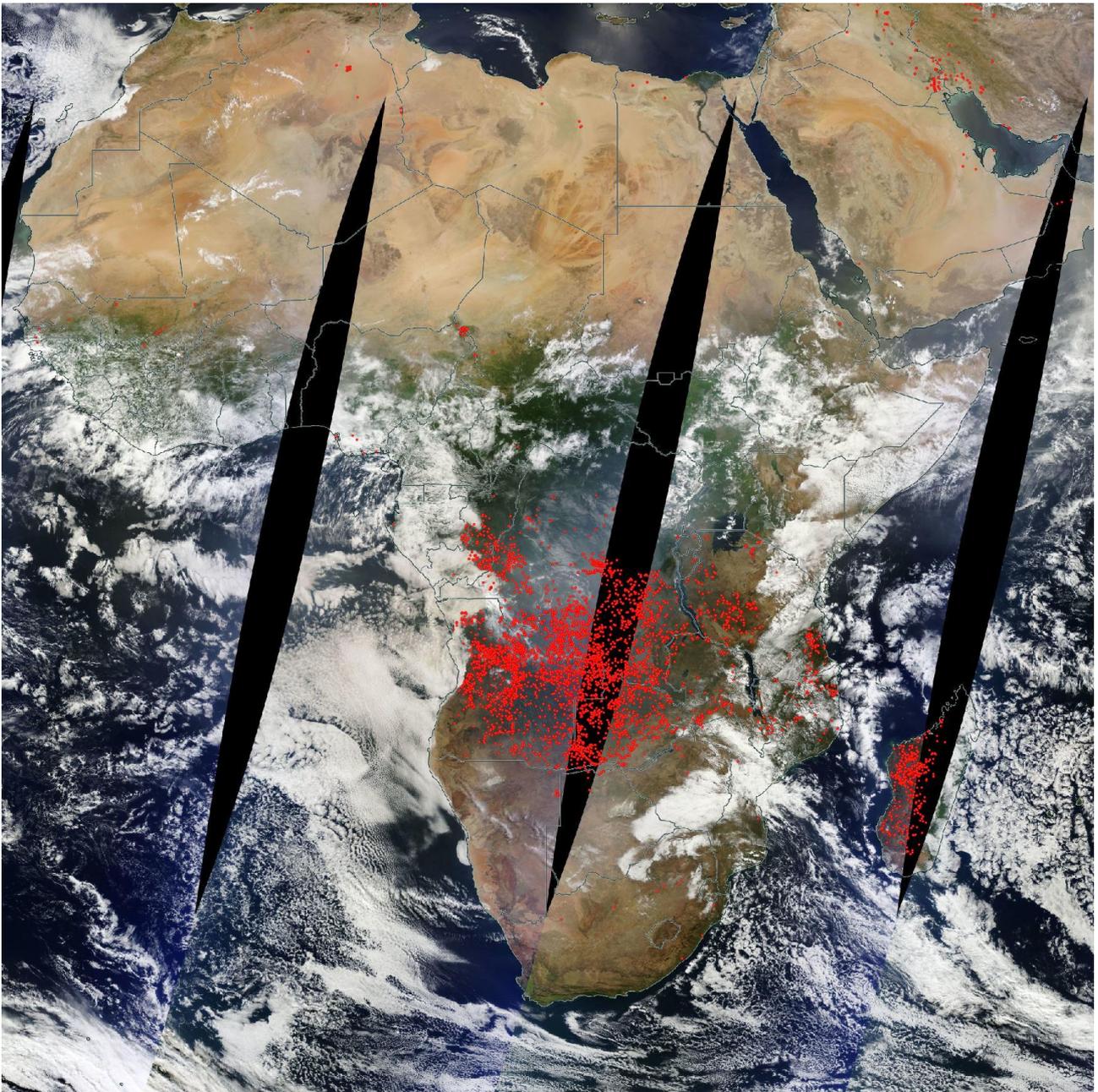


Figure 22 – MODIS visible image for the 3rd July 2016 presented during the aerosol/chemistry forecast on the 4th July 2016. Red dots represent the active fires detected from MODIS observations during the month of June.

6 Literature

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