# Interactions of Atmospheric Gases and Aerosols with the Monsoon Dynamics over the Sudano-Guinean region during AMMA

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#### Dear Editor,

We thank you and the reviewers for their positive comments and opinions. About their comments, we propose in the following some answers and corrections of the manuscript.

## 1 Report 1

#### 1.1 General Description of manuscript and General Comments

The authors use observations from the West Africa AMMA aircraft campaign in 2006 and an atmospheric chemistry model to diagnose the transport patterns and contributing sources to enhancements in carbon monoxide and PM 2.5 along a latitudinal transect from the Gulf of Guinea to the Sahel.

As presented currently the study appears anecdotal. It is not apparent that the features observed along a very limited longitudinal domain in West Africa apply to the rest of West Africa and to other years. Please clarify whether the findings in this study are generally applicable to the rest of West Africa and other years? If so, what do the outcomes from this study mean for past/present/future atmospheric composition or development of air quality and/or climate policy?

The present analysis is meant to be a case study for the year 2006, making use of the wealth of observations acquired during the AMMA programme. The interannual variability has not been investigated and is out of the scope of this paper. We revisited the AMMA 2006 observations to focus on the transport patterns and sources of carbon monoxide and PM2.5 over Southern West Africa, a region that has received less attention than the Sahel during AMMA.

We suspect that some of the features observed occur year after year over Southern West Africa monsoon but we cannot extrapolate from the results of our study. Here, we proposed a new approach focusing on two major pollutants (Carbon monoxide, CO, and fine atmospheric particulate matter, PM2.5) that can be transported far from the sources due to their long lifetime. They are certainly of paramount importance for air quality and climate policies development, but we feel that additional simulations are needed to address this, which is also beyond the scope of the paper.

Why not also compare the model to other parameters measured during the AMMA campaign to assist in interpreting transport patterns and contributing sources and diagnosing what causes differences between modeled and observed PM 2.5 and CO? These could include measure components of PM 2.5 (sulfate, ammonium, organic aerosol, nitrate), and CO precursor VOCs, for example.

We decided to focus on these two important atmospheric components (Carbon monoxide, CO, and fine atmospheric particulate matter, PM2.5) because the data were available. Aerosol Mass Spectrometer data were not available. For CO, we assume that we modeled the two main sources (i.e. Anthropogenic and biomass burning). Given the amount of VOCs, i.e. > 15 ppb according to Ancellet et al. (2011), VOCs oxidation must be very low (a few ppb).

#### **1.2** Specific Comments

We thank the reviewer for taking the time to go through the spelling and grammar of the manuscript. All proposed corrections have been taken into account in the revised version of the manuscript. Below, we provide answers to the more science oriented questions the referee has.

p. 2, Lines 17-18: The authors point to economic growth as a driver of emissions from industries, including gas flaring, but the reference they site does not mention economic growth as a driver.

The sentence has been changed: 'However, the economic growth over the region drives up anthropogenic emissions: the increase of industries including gas flaring (Asuoha and Osu, 2015), of local fuel-wood burning

for stoves and of traffic (Liousse et al., 2010; Hadji et al., 2012; Liousse et al., 2014) with more two-wheel vehicles using very poor fuel quality used (Ndoke and Jimoh, 2005; Assamoi and Liousse, 2010), which are suspected to quickly worsen the air quality.'

p. 2, Line 19: Do the authors mean 'air quality standards' or air quality guidelines? If from WHO these should be guidelines.

#### Corrected

p. 3, lines 9-12: Presumably DACCIWA will also contribute to understanding the change in atmospheric composition due to increases in emissions over a rapidly growing region?

The sentence has been changed: It will contribute to understanding the change in atmospheric composition due to increases in emissions over a rapidly growing region as well as the development of the next generation of accurate models to forecast weather and pollution in southern West Africa (Knippertz et al., 2015).

p. 3, line 31: Is a 1 month spin-up sufficient for carbon monoxide output from a model, when CO has a lifetime of 2 months?

As for all chemical species, and the principle of an area limited domain model, the lifetime of the species is not a constraint. The 'aged' concentrations are already modeled with the global climatological model, providing hourly boundary conditions. These concentrations are injected into our regional model depending on the wind direction and speed. The 'fresh' concentrations are explicitly hourly emitted in the domain.

The real constraint motivating the use of a spin-up time is the transport of the species into the domain: we want to ensure that for the first modeled hour, all possible species, due to a previous transport, are well present in the domain. There is no link to the lifetime, but depends on the transport and the domain size only.

p. 5, lines 22-23: Please point out the features that are similar to the Flaounas et al. (2010).

The paragraph related with Flaounas et al. (2010) has been modified such as: 'During this period, the precipitation location and rate will play a crucial role on the modeled surface  $PM_{2.5}$  concentrations. As a validation for this variable, the methodology of Flaounas et al. (2010) is used: precipitation rates are averaged between 8.5W and 8.5E. Day-to-day variability is smoothed by applying a moving average of  $\pm 2$  days. Figure 2 is directly comparable to the Flaounas et al. (2010) study using the same period and averaged region. In May and June, observed and modeled precipitations occur mainly over the ocean (below 5N). From late June on, the main precipitation areas move over the continent (above 5N) and reach the Sahel (at about 13N). Figure 2 shows that the modeled precipitation spatial patterns are in good agreement with the two satellite observations (TRMM and GPCP) presented in their study (see Figure 3 of Flaounas et al. (2010)). '

p. 6, line 14: Remove parentheses around the AERONET URL.

- p. 6, line 15: Space between number and units (400 nm instead of '400nm').
- p. 6, line 15: Provide units for '440-870'.

p. 10, line 17: 'analyzes' should be analysis.

#### All four points have been corrected.

p. 10, lines 26-27: Point out in Figure 5 the feature that indicates the arrival of the cold tongue.

This sentence has been revised: At the end of the period, when precipitation occurs inland and  $anth-PM_{2.5}$  is low, the meteorological situation changes suddenly over the ocean showing the cold tongue arrival located at the Equator, which is associated with increased wind speed between the Equator and the coast, as detailed by Meynadier et al. (2016).

p. 12, line 31: Fix units.

p. 14, line 11: 'perturbated' should be perturbed.

p. 15, line 5 (bottom of page): Dust is repeated.

#### All three points have been corrected.

p. 15, line 14: What does 'Figure ??' refer to? Is this Figure 11? Indicate on the figure the convective cell.

Thanks for picking this up. Yes indeed, we meant Figure 12 (previously 11). A red ellipse has been superimposed on Figure 12 to indicate the location of the convective cell.

p. 16, lines 29-30: There is no context for why the results in this work will be compared to DACCIWA. What new insights will be gained from this comparison that justify mentioning it here?

This sentence has been changed: Concerning air quality and climate policy development, we have shown that the export of anthropogenic pollutant from the Guinean coast toward the North could lead to cross boundary pollution plumes. This result will be confirmed by comparing to the 2016 DACCIWA campaign observations in order to propose strategy to reduce the atmospheric pollution in West Africa.

#### Figures:

Figure 3: What are the statistics in the first and third panel? Does this compare the modeled component to total AOD from the measurements? Whats the value in showing this? Why not just compare total modeled and observed AOD? The label for the modeled AOD components is confusing. The label is 'AOD Anthr.' and 'AOD Fires', but shouldn't is rather be biomass burning and all other components for clarity? The figure caption suggests this is what is shown.

There is no value to keep the statistics because the scores are very slightly improved by adding biomass burning emission. It has been removed. Concerning the legend, the labels ('AOD Anthr.' and 'AOD Fires') have been replaced (by 'with biomass burning' and 'without biomass burning'). The new figure is presented below (Figure 1 of this document).

Moreover, a detailed explanation of how the different contributions are obtained has been added in Section 2.2.

# 2 Report 2

#### 2.1 General Comments

This paper looks at the contribution of different emissions sources to CO and PM2.5 over W. Africa and the meteorological conditions that influence the transport of the pollutants within this region. It is a model study using a WRF coupled to CHIMERE and evaluated using measurements made during the AMMA project in 2006. It is the impact of the meteorological conditions on pollution concentrations particularly on the coastal region where the majority of people live that provides novel insight that is worthy of publication. I do have a few concerns that I would like to see addressed before publication.

#### 2.2 Specific Comments

It is not clear to me why in section 3.3, the model meridional simulation of CO and PM2.5 is evaluated against just two flights the on consecutive days (13-14 June), during which an MCS passes through the area (section 3.3.1). On the back of this the model is then used to quantify the modelled pollution source apportionment on a monthly basis. Why not evaluate the model over the whole month? There were flights on other days and with other aircraft. There are several papers published from the AMMA campaign that could have been used to help with this evaluation see Reeves et al (2010) (www.atmos-chem-phys.net/10/7575/2010/) that gives an overview of the chemical and aerosol characterisation and references therein. Satellite data could also be used for the evaluation. This would also help to evaluate the transport of biomass burning plumes into the region from south of the aircraft flight tracks.

The section 3.3 deals with modeled and observed CO and PM2.5 during two meridional flight trajectories on consecutive days (13-14 June) because it provides data from the coast to the Sahel, thus it fits exactly the scope of the study focusing on the Sudano-Guinean region. New insights are presented gained from the AMMA 2006 observations as we focus on the transport patterns and sources of CO and PM2.5 over the Sudano-Guinean region.

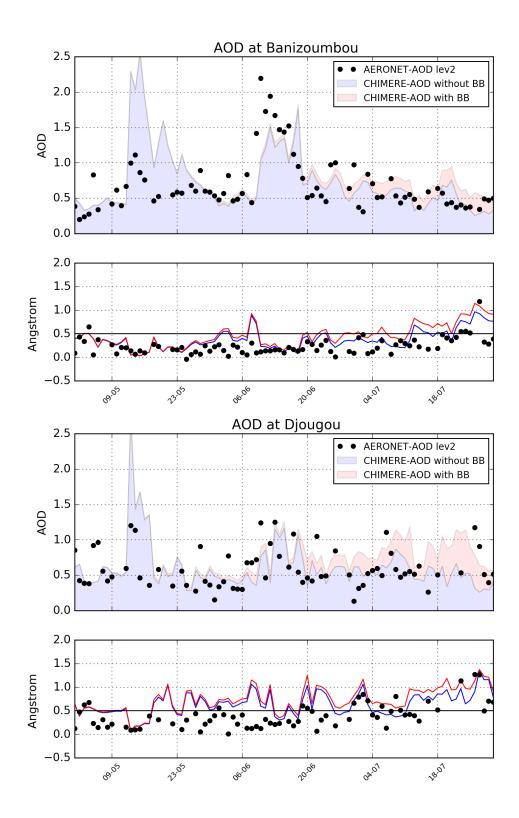


Figure 1: Observed daily averages of AERONET level 2 AOD and Angström exponent (black dots) at Djougou (Benin) and Banizoumbou compared to the modeled time series with a splitting to extract the relative contribution between without biomass burning emissions (including anthropogenic, biogenic, sea salt and mineral dust; all four in blue) and with biomass burning emissions (in red).

Moreover, it has been difficult to work with validated data from the AMMA database. Many datasets used in preliminary evaluations were partly unusable after contacting responsible people of the dataset. We have contacted FAAM team (Graeme NOTT) for data from the BAe 146 of the 20-21 July but it was not possible to produce PM concentration because: 'during this period no calibration of the PCASP was carried out by FAAM' and in the FORTRAN code, a mistake has been done using constant flow rate. Suzanne CRUMEYROLLE has provided the only aircraft data that we are confident but aerosol speciation was not available. The two flights used in our study are meridional transects that fit exactly with the purpose of the study.

How is the source apportionment (Section 3.3.1) determined? Are separate CO tracers used for the background, anthropogenic and biomass burning? How is this done for PM2.5, in particular considering how the aerosol scheme works? How is the formation of SOA considered? What about mixed aerosols? One of the main scientific questions addressed in this paper is the contribution of different sources to pollutant concentrations, so it is essential that a clear description of the methodology for determining this is included in the paper. Much of the analysis in the paper focuses on anth-PM2.5 so it must be clear how this is defined.

References describing how the aerosol scheme works in the CHIMERE model were missing in the manuscript. In the revised version of the manuscript, we now describe it as well as how the source apportionment is determined for CO and for PM2.5, which has been added in Section 2.2:

'Menut et al. (2016) have detailed and analyzed aerosol speciation and size distribution in the CHIMERE model during the summer 2013 over Europe and Africa using the AERONET network for AOD and EMEP network for PM concentrations. For the AOD calculation, the aerosol optical scheme in the CHIMERE model considers mixed aerosols following the 'core-shell' hypothesis detailed in Péré et al. (2009) and evaluated in Péré et al. (2010).

In order to quantify the PM2.5 source apportionment, we assume that it is possible to split aerosols in different families depending on the sources because their chemical compositions are different: Mineral, Biogenic, Salt and Anthropogenic. Given that anthropogenic and biomass burning aerosols have similar compositions, we have done two simulations with and without biomass burning emissions to split their contributions. The gas phase chemical scheme for SOA formation explained in Bessagnet et al. (2010) takes into account three anthropogenic and three biogenic hydrophilic species, three hydrophobic species with different saturations, and two surrogate compounds for the isoprene oxidation products.

The source apportionment has been determined for CO considering three main contributors (anthropogenic sources, biomass burning sources and long-range transport). Consequently, three simulations have been done: one without any emission source in the domain for the background concentration, one with the anthropogenic emission only, and a last one with the anthropogenic and biomass burning emissions.'

Section 4.22. I really do not understand the conclusions here. In Figure 7, surely a and b are the wrong way around, with the top plot having the 1 a.u. contour at around 7N near Cotonou and the bottom plot having it at 14N near Niamey? Perhaps I am getting muddled by this figure, but it seems extremely odd that the ratio of the coastal to Sahelian tracer is greater in the Sahelian region, especially since the tracer experiment uses arbitrary units and so does not consider the relative strengths of the tracer emissions in each region. The clarity of the discussion could be improved by attention to the English, but I think there is something scientifically wrong here.

We thank the reviewer for reporting this mistake. In this Figure (now Figure 8), a) and b) were the wrong way around. This has been corrected.

Real anthropogenic emissions represent the amount of what each city emits: their magnitude is thus representative of each location, the activity sectors and the population. After emissions, pollutants are transformed by transport, mixing, deposition and chemistry. In this case, and if we want a realistic estimate of a concentration far from the sources, it is necessary to have emissions really representative of the size of each urbanized area. The problem is different with the tracers: we don't want to have a realistic value, but just to know the percentage of what arrived at a remote location. Thus, we need to emit the same amount at every location to have the exact percentage at the studied remote location. This enables to quantify that in Niamey, Cotonou tracer concentration is about 9% (of the 1 a.u. isocontour presented in Figure 8-a), while in Cotonou, Niamey tracer concentration is about 0.03% (of the 1 a.u. isocontour presented in Figure 8-b).

In Section 4.2.2, a sentence has been added for clarity: 'The tracer experiment uses arbitrary units and considers the same quantity of tracers emitted in each town.'

The second paragraph of this section has been changed: 'Tracers emitted at the coast indicate that there is an important transport of coastal pollutants toward the North in the PBL. On the other hand, there is no significant transport of tracers emitted in the Sahel toward the coast. In Niamey, Cotonou tracer concentration is about 9% (of the 1 a.u. isocontour presented in Figure 8-a), while in Cotonou, Niamey tracer concentration is about 0.03% (of the 1 a.u. isocontour presented in Figure 8-b). In the HTAP anthropogenic inventories (presented in Figure 1), the anth-PM<sub>2.5</sub> (respectively anth-CO) is  $\approx 103$  (735) kg.km<sup>-2</sup>.day<sup>-1</sup> in Niamey and  $\approx 438$  (7707) kg.km<sup>-2</sup>.day<sup>-1</sup> in Cotonou. Therefore, an important part of the pollution over the Sahel has been emitted at the coast and it contributes to a maximum of anthropogenic pollution in June over the Sahel. In conclusion, the high concentration over the Sahel is due to the existence of a meridional atmospheric cell, which acts at accumulating pollutants emitted locally and remotely at the coast.'

P 2, 1 22-24: Several values are given for high concentrations of pollutants in these 3 lines. It would be helpful to give the time averages over which these measurements were made as the context of this paragraph is to compare them with the air quality standards which are for specified periods of exposure.

Time averages have been added: 'based on half-hour averages' for Baumbach et al. (1995); 'based on 1-min averages' for Dionisio et al. (2010); 'based on daily averages' for Boman et al. (2009)

P 4, l 24-25: Id like to see more details on how WRF is coupled to CHIMERE. Is the CHIMERE transport used or just the chemistry? Time steps for physical processes and chemistry?

In this study, we present offline simulations (CHIMERE is forced by WRF). For chemistry and aerosol simulations, the concentrations are calculated using the chemistry, transport, mixing and deposition equations implemented in the CHIMERE model. For clarity, two sentences have been added: 'The WRF and CHIMERE models are run offline on the same horizontal grids for the continental and regional domains' and 'The time step is set to 10 minutes for the physical processes and 5 minutes for the chemistry, which could change depending on the Courant-Friedrichs-Lewy condition.'

P 5, 1 24: It would be good to show plots of the TRMM and GPCP data to demonstrate the good agreement.

These two plots have been presented by Flaounas et al. (2010) as well as the comparison to the different WRF parametrization. We do not aim at focusing on the precipitation patterns in the present study. We have specifically pointed out comparable patterns of our Figure 2 and Figure 3 of Flaounas et al. (2010). The first paragraph of Section 3.1 has been modified such as: 'During this period, the precipitation location and rate will play a crucial role on the modeled surface  $PM_{2.5}$  concentrations. As a validation for this variable, the methodology of Flaounas et al. (2010) is used: precipitation rates are averaged between 8.5W and 8.5E. Dayto-day variability is smoothed by applying a moving average of  $\pm 2$  days. Figure 2 is directly comparable to the Flaounas et al. (2010) study using the same period and averaged region. In May and June, observed and modeled precipitations occur mainly over the ocean (below 5N). From late June on, the main precipitation areas move over the continent (above 5N) and reach the Sahel (at about 13N). Figure 2 shows that the modeled precipitation spatial patterns are in good agreement with the two satellite observations (TRMM and GPCP) presented in their study (see Figure 3 of Flaounas et al. (2010)). '

P 6, 1 3-8: Looking at Fig. 2, it seems to me that the precipitation is focused at 2N through much of June and that it is only until mid-late June that it shifts to more to 5N. This is not consistent with the text that says the pre-onset occurs in May.

The section 3.1 has been modified to be consistent with the figure and the three periods defined (see above).

P 9, 1 30: The high CO concentrations at the coast are not so continuous in late June and July.

'during the whole period' has been changed to 'from the beginning of May to late June'

P 10, l 5-8: Please explain more clearly how precipitation/convection impacts surface CO concentrations. How does it affect the vertical distribution?

In this section, we identify the changes which are analyzed in the following. The sentence: 'In July, the variability is mostly consistent with precipitation rates after the onset, which suggest that surface versus vertical distribution has changed by the convection associated with large scale precipitation.' has been replaced by 'In July, the variability is mostly consistent with precipitation rates after the onset, suggesting modifications of transport and deposition patterns by the convection associated with large scale precipitation.'

P 10, l 15-16: A cant make out any great difference between the pattern at 12N and 13N.

'at 12N' has been removed. This sentence was unclear because we were not comparing 12N and 13N. This paragraph has been modified (see above).

P 12, 1 27: Are diurnal patterns included in the emission inventories used?

'when the convection and NLLJ are weak' has been removed because diurnal patterns of anthropogenic pollution are mostly driven by diurnal variation of the emissions.

P 12, l 29-31: In Fig. 8 some of the pollution over the sea on the 10-11 June in the Hovmuller plot appears to progress northwards with time (i.e. bottom left towards top right) rather than be transported out to sea from the land. Fig. 9 suggests it may be coming from other cities further to the south.

This paragraph has been modified such as: 'There is a transition from low to high concentration of anthropogenic pollution from 8 to 12 June. Anthropogenic pollution is modeled over the sea from 10 to 11 June. It is interesting to note that precipitation occurs inland on 11 June (between 18 UTC and 00 UTC), then high modeled concentrations persist during the night of 11-12 June. This precipitation event reflects a change in the wind patterns, which induces a change in the transport of pollutants, leading to surface concentrations up to 8  $\mu g.m^{-3}$  in Cotonou.'

P 13, 1 33: How can you be sure that the plumes are 'overlaying' and not mixed?

You are right, it is mixed. We are analyzing the surface level. If the tracers associated to the different cities are at the same location, it means that the pollution from the different cities is mixed.

The English needs to be improved. I have listed some places where the understanding is not clear or incorrect because of the English, but there are many minor corrections that need to be made (e.g. the appropriate use of 'the' and 'a', use of singular and plural) that I have not listed.

## 2.3 Technical Comments

Ensure the initial letters of 'Guinean Gulf' are in uppercase, here and throughout the paper.

OK

P 1, l 5-6: It needs to be clear what the 38% relates to. 38% of PM.2.5?

This sentence has been modified from 'For  $PM_{2.5}$ , desert dust decreases from  $\approx 38$  % in May to  $\approx 5$  % in July;' to 'Desert dust decreases from  $\approx 38$  % in May to  $\approx 5$  % in July of  $PM_{2.5}$  concentration'

P 1, 1 9-10: It is not clear. Are the pollutants emitted near the coast concentrated in the Sahel?

In order to be clear, this sentence has been split in two sentences: 'Air masses dynamics concentrate pollutants emitted in the Sahel due to a meridional atmospheric cell. Moreover a part of the pollution emitted remotely at the coast is transported and accumulated over the Sahel.'

P 1, l 11: 'Refining the analysis' reword the English. Suggest 'Focusing the analysis'

OK

P 1, l 13: 'overlay' each other?

They are mixed and not overlaying. Corrected

P 1, l 13: 'high pollution level' is ambiguous. High concentrations? High attitude?

Indeed 'level is ambiguous. It should be either 'concentration' or 'altitude'. The manuscript has been entirely modified to remove this ambiguity.

P 2, l 2: 'washout the atmosphere' change to 'wash pollutants out of the atmosphere'

OK

P 2, l 4: 'air pollution'. Are natural components of the atmosphere pollutants? E.g. Sea salt aerosols? 'air pollution sources' has been replaced by 'aerosol and gas sources'

P 2, 17: 'in megacities' should be 'from megacities'.

OK

P 2, 1 31: 'since the last decade'. The English is not clear. Do you mean 'since' or 'during'. Note that the main AMMA campaign was in 2006, i.e. more than a decade ago.

'since the last decade' has been deleted.

P 3, l 13: 'This article is dedicated to the pollutants transport over the Guinean Gulf coastal region and focuses on two major pollutant concentrations:'. Reword the English, 'This article focuses on transport of pollutants over the Guinean Gulf coastal region, in particular on:'

OK

P 3, l 15: Replace 'have both an' with 'both have a'.P 3, l 17: Replace 'pollutants in the' with 'pollutants to the'.P 3, l 23: Replace 'refines spatially' with 'focuses on'.

All last three points have been corrected.

P 3, 1 27: It would be useful to provide a figure showing the 2 nested domains.

The latitudes and longitudes of the two domains are given in the beginning of Section 2. The results of the article concern only the West African region. Furthermore, we do not compare the coarse and fine resolutions. It is why we want to focus only on the regional domain (presented in Figure 1).

P 4, 1 6: Replace 'hourly interpolated' with 'interpolated hourly'.

OK

P 4, 1 8: 'better' than what?

The sentence has been changed and 'better' removed.

P 4, 127-28: Replace 'The anthropogenic emissions are estimated using the HTAP v2 (Hemispheric Transport of Air Pollution) annual totals for the year 2010 by the EDGAR Team,' with 'The anthropogenic emissions are estimated by the EDGAR Team using the HTAP v2 (Hemispheric Transport of Air Pollution) annual totals for the year 2010,'

#### OK

P 4, l 31-32: 'Taking into account vegetation fires emission fluxes is of primary importance to simulate West African pollution (Giglio et al., 2006).'- The English needs improving.

This sentence has been modified: 'Biomass burning emission from Central Africa is of primary importance to simulate West African pollution (Giglio et al., 2006)'

P 4, l 31 P5, l2: Be consistent with terms and their combinations: 'fire', 'vegetation', 'biomass burning'. How were the two parts split?

The manuscript has been revised to use only 'biomass burning'.

Since the incomplete combustion is both included in anthropogenic inventories (local urban burning) and forests biomass burning inventories, the simulation was designed to split these two parts. It is now explained in Section 2.2

P 5, l 4: 'a new global soil and surface datasets' singular or plural? P 5, l 4: 'a satellite-derived aeolian roughness length data' singular or plural?

This is singular because it is a unique dataset composed of two satellite retrievals.

The sentence has been modified such as: 'The mineral dust sources are obtained using the GARLAP (Global Aeolian Roughness Lengths from ASCAT and PARASOL) new global soil and surface dataset made from satellitederived aeolian roughness lengths with a 6 km spatial resolution, as detailed in Mailler et al. (2016).'

P 5, l 14-15: 'first analyze and quantify the temporal variability of the pollutants concentrations modeled in the urbanized areas along the Guinean Gulf coast during the whole AMMA-SOP1 period (Redelsperger et al.,

2006). First' There are two 'firsts' which is confusing. It states that the temporal variability of the pollutants will be analysed, but in the rest of the paragraph I can only see mention of AOD. What about any other pollutants?

This sentence has been modified: 'In this section, we analyze the temporal variability of precipitation, gas and aerosol during the whole AMMA-SOP1 period (Redelsperger et al., 2006).'

P 5, 1 29: Replace 'interactions constituted' by 'interactions, which are made up'.

OK

P 6, l 27: Replace 'lead' with 'leads'.

OK

P 7, l 5-6: English needs improving.

'Two flights made during a 'North-South land-atmosphere-ocean interaction' mission plans have been conducted over the Cotonou-Niamey meridional transect on 13 and 14 June 2006.' has been modified such as: 'We are studying two flights conducted along a meridional transect between Cotonou and Niamey on 13 and 14 June 2006 as part of a 'North-South land-atmosphere-ocean interaction' survey mission.'

P 7, 1 28: What is meant by 'a South-North gradient is expected moving closer to the Sahara'?

'a South-North gradient is expected moving closer to the Sahara' is unclear and it has been modified such as: 'For  $PM_{2.5}$ , a South-North gradient is expected with the highest concentrations close to the Sahara.'

P 7, 1 31 and P8, 1 4: What is meant by 'a gap of concentration'?

This has been replaced by: 'important increase of the concentration'

P 7, 1 7: To test if the model reproduces the MCS, why not compare modelled meteorological parameters with observed e.g. satellite precipitation.

A new figure has been added in Section 3.3.1 (presented in Figure 2 of this document), which aims at presenting the meteorological situation on 13-14 June as well as validating the model meteorology. The first paragraph of this section has been modified such as: 'We are studying two flights conducted along a meridional transect between Cotonou and Niamey on 13 and 14 June 2006 as part of a 'North-South land-atmosphere-ocean interaction' survey mission. During these two days, the WAM dynamics over the area were perturbed by the presence of a MCS. It developed over the Jos Plateau (Nigeria) around 16:00 UTC, reaching the Benin-Nigeria border at 20:00 UTC and moved southwestward across Benin overnight and into central Ghana, as already described in Flamant et al. (2009) and (Crumeyrolle et al., 2011). The model reproduces the location of this MCS but earlier than in the observations, i.e. reaching the Benin-Nigeria border at 10:00 UTC (Figure 4). The MCS interacts with the dust layer coming from the Sahara (especially from the Bodele depression), changing the dust load and vertical distribution over Benin and Niger. Associated with subsidence in the wake of the MCS, there is a lowering of the dust layer height (Flamant et al., 2009). '

P 7, l 8: Replace 'not realistic' with 'unrealistic'.

OK

P 7, 19: Why 'Nevertheless'?

This sentence has been modified: 'Nevertheless, the order of magnitude of the CO and  $PM_{2.5}$  concentrations are good in agreement with observations. '

P 8, l 31: Presumably by 'vegetation emissions' you mean biomass burning rather than biogenic. This needs to be clear and correct throughout the paper.

OK

P 8, l 33-34 and P 9, l 5 and 10: Units of ppb should be microg / m-3.

#### OK

P 10, l 10: Why 'more important'?

P 10, l 11: What 'increase'? The English in this sentence needs improving. Consider splitting it in two.

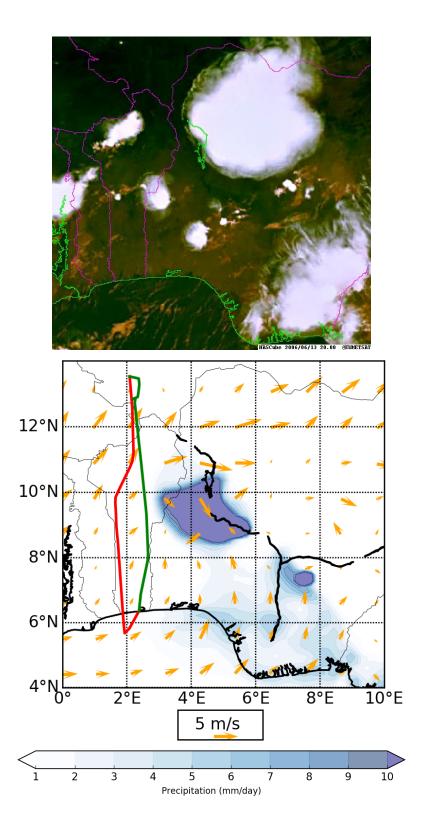


Figure 2: (top) EUMETSAT visible image of the Cotonou area of the 13 June 2006 at 20 UTC (from NAScube (http://nascube.univ-lille1.fr); (bottom) Map of Cotonou area for the 13 June 2006 at 12 UTC with wind vectors at 10 m (orange arrows), precipitation (blue shading). The two flight trajectories are displayed with the red line for the 13 June and with the green line for the 14 June.

For the two last points, the sentences: 'The same behavior is observed for the surface concentrations of  $PM_{2.5}$ . The week to week variability is more important. This increase is probably due to the longer CO lifetime compared with that of PM (being less chemically active and without settling), the CO concentrations are more homogeneously mixed in a large latitudinal area from the coast to more than 16 up to the North.', have been reworded: 'The same behavior is observed for the surface concentrations of  $PM_{2.5}$ . The week to week variability is greater than for anth-CO, which is probably due to the longer lifetime of CO compared with that of PM (being less chemically active and less prone to settling). CO is more homogeneously mixed than PM in a large latitudinal area spanning from the coast to latitudes higher than 16. '

P 10, l 13: A frequency has units of 1/time. Do you mean a periodicity close to 2 weeks?

#### OK

P 10, l 14: A wouldnt call these features plumes as they are a characteristic of a Hovmuller plot rather than a pollution plume.

'latitudinal plumes' has been replaced by: 'latitudinal patterns'

P 11, l 19: anti-clockwise?

Yes, it has been corrected.

P 12, l 25: Replace 'derives' with 'are transported'.

#### OK

P 14, l 11-12: Low and high anthropogenic pollution where? Contonou?

Yes, 'in Cotonou' has been added.

P 15, l 14: 'Figure ??', 11.

#### OK

Fig. 2. Caption 'regional domain' should be replaced by 'regional model domain'.

#### OK

Fig. 3. The legend only says 'anthr' but the captions says 'anthropogenic, biogenic and mineral dust'. Is the red line the sum of 'anthropogenic, biogenic and mineral dust' and 'biomass burning emissions'?

In red, this is anthropogenic plus biogenic plus mineral dust plus sea salt. The legend's labels ('AOD Anthr.' and 'AOD Fires') have been replaced (by 'with biomass burning' and 'without biomass burning').

Fig. 6 caption: what PM2.5 mass density is the light orange line meant to be? Shading should be lines.

We tried to remove shading but it leads to too much contours, which is difficult to read. In the end, we use shading for the meridional wind speed and contours for the anthropogenic PM2.5 concentration. The light orange line has been removed and we focuse only on anthropogenic PM2.5 concentration = 4.0  $\mu$ g.m<sup>-3</sup> (orange) and = 5.0  $\mu$ g.m<sup>-3</sup> (violet). The new figure is presented below (Figure 3 of this document).

Fig. 7 caption: Shading should be lines. It is not clear if one line is white or both are yellow.

As for the previous figure, we have removed the white line. The comment of this figure is now focused on only two isocontours. The new figure is presented below (Figure 4 of this document).

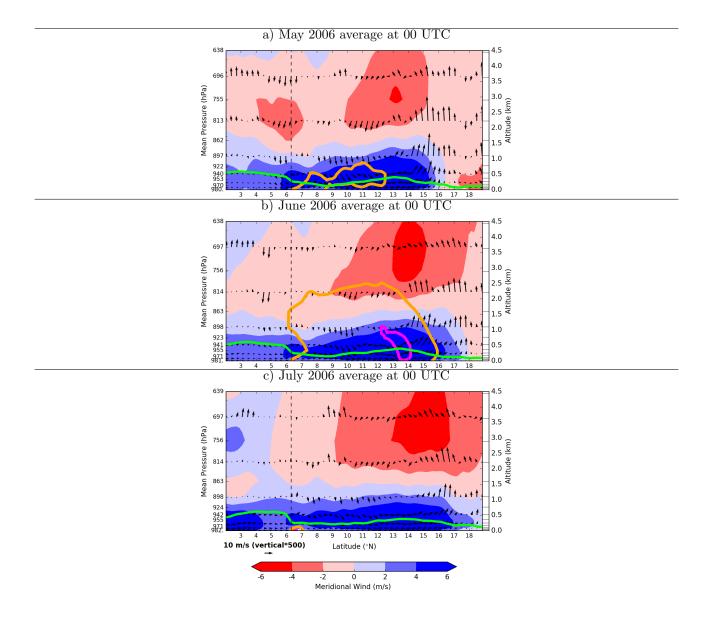


Figure 3: Vertical cross-section of the meridional wind (shading in  $m.s^{-1}$ ) mean over: a) May, b) June and c) July, at 00 UTC along a meridional transect from 2N to 19N and averaged from 2E to 3E including Cotonou (Benin) and Niamey (Niger). Orange and violet shading represent anthropogenic  $PM_{2.5}$  concentrations of = 4.0  $\mu g.m^{-3}$  and = 5.0  $\mu g.m^{-3}$ . Vectors represent the wind field in the plan of the transect (with an aspect ratio of 500 between the meridional and the vertical components). The green line is the PBL height (m). The grey vertical dash line is the latitude of the coast.

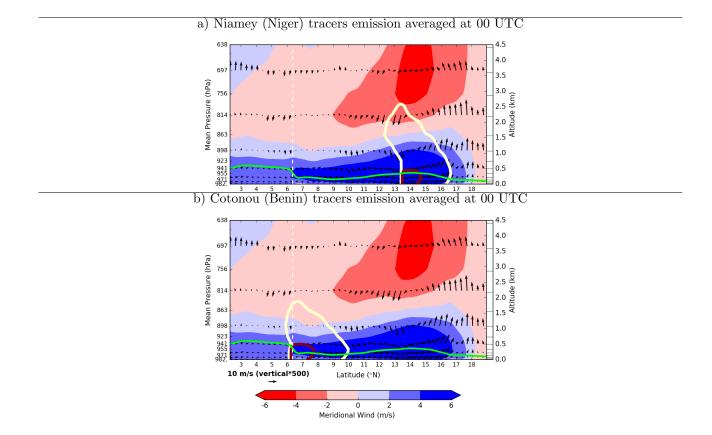


Figure 4: Vertical cross-section of the meridional wind (shading in  $m.s^{-1}$ ) along a meridional transect from 2N to 19N, averaged from 2E to 3E including Cotonou (Benin) and Niamey (Niger) and averaged over 20 to 30 June at 00 UTC. Isocontours represent gaseous tracers concentration continuously emitted (in arbitrary unit) from the 1 to 30 June at: a) Niamey (Niger) and b) Cotonou (Benin). Brown and yellow shading represent tracers concentration = 1 a.u. and = 10 % a.u. respectively. The green line is the PBL height (m). Vectors represent the wind field in the plan of the transect (with an aspect ratio of 500 between the meridional and the vertical components). The white vertical dash line is the latitude of the coast.

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# Interactions of Atmospheric Gases and Aerosols with the Monsoon Dynamics over the Sudano-Guinean region during AMMA

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Abstract. Carbon monoxide, CO, and fine atmospheric particulate matter,  $PM_{2.5}$ , are analyzed over the Guinean gulf Gulf coastal region using the WRF-CHIMERE modeling system and observations during the beginning of the monsoon 2006 (from May to July), corresponding to the Africa Multidisciplinary Monsoon Analysis (AMMA) campaign period. Along the Guinean gulf Gulf coast, the contribution of long-range pollution transport to CO or  $PM_{2.5}$  concentrations is impor-

- 5 tant. For PMThe contribution of desert dust  $PM_{2.5}$ , desert dust concentration decreases from  $\approx 38$  % in May to  $\approx 5$  % in July; biomass burning aerosol. The contribution of biomass burning  $PM_{2.5}$  concentration from Central Africa increases from  $\approx 10$ % in May to  $\approx 52$  % in July. The anthropogenic contribution is  $\approx 30$  % for CO and  $\approx 10$  % for  $PM_{2.5}$  during the whole period. When focusing only on anthropogenic pollution, frequent northward transport events from the coast to the Sahel are associated with periods of low wind and no precipitation. In June, anthropogenic  $PM_{2.5}$  and CO concentrations are higher than in May or
- 10 July over the Guinean coastal region. Over the Sahel, air masses Air mass dynamics concentrate pollutants emitted locally and remotely at the coast in the Sahel due to a meridional atmospheric cell. Moreover a part of the pollution emitted remotely at the coast is transported and accumulated over the Sahel.

Refining Focusing the analysis on the period 8 - 15 June, anthropogenic pollutants emitted along the coastline are exported toward the North especially at the beginning of the night (18 UTC to 00 UTC) with the establishment of the nocturnal low

15 level jet. Plumes originating from different cities overlay are mixed for some hours at the coast, leading to high pollution levelconcentration, because of specific disturbed meteorological conditions.

#### 1 Introduction

The interactions interaction between air pollution and climate in megacities is a challenging field of research (Baklanov et al., 2016). In the countries of the Guinean Gulf, the population has been growing rapidly during the last decades, accompanied by economic development. Parallel to industrialization, air pollution is increasing without any governmental control (Zhu, 2012).

During the dry season (*i.e.* November - April), when the Harmattan easterly wind is weak, high ozone concentrations and smog are observed over megacities such as Lagos or Cotonou when the Harmattan easterly wind is weak (Marais et al., 2014; Minga et al., 2009). During the wet season (*i.e.* May - October), the West African Monsoon (WAM) wind carries the pollutants northward, and local convective precipitations wash out pollutants out of the atmosphere. Two precipitation periods occur over

5 the Guinean Gulf coastal region in April-May and August-September. Between these two periods, the wind coming from the South is predominant (Janicot et al., 2008).

There are various <u>air pollution aerosol and gas</u> sources in the Guinean Gulf coastal region during the WAM. Sea salt aerosols are transported in the marine boundary layer, and mineral dust aerosols are transported in the Saharan Air Layer (SAL) above the monsoon air (Lafore et al., 2011). Biogenic components are emitted by tropical forests (Reeves et al., 2010), and the

- 10 urban air pollution in megacities (Liousse et al., 2014) from megacities leading to ozone production (Ancellet et al., 2011) (Liousse et al., 2014). In addition, pollutants resulting from incomplete combustion such as carbon monoxide and black carbon particles are coming from the Southern hemisphere due to biomass burning reach emissions, and are reaching the Guinean coast in June (Mari et al., 2007). Biomass (Mari et al., 2007) (Williams et al., 2010). In situ biomass burning plumes observations have shown-measured high ozone (≥ 60 ppb at 700 hPa) and carbon monoxide concentration (≥ 200 ppb at 700 hPa)
- 15 concentrations (Sauvage et al., 2004; Mari et al., 2011).

In Nigeria, Akeredolu (1989) have listed the different sources of particle loading: biomass burning (31.7 %), fugitive dust from roads (29.1 %), fuel wood burning (21.3 %), Harmattan dust (13.8 %), solid waste incineration (2.1 %), stationary sources (1.6 %), automobile exhaust lead (0.2 %) and gas flares (0.1 %). Since the 1990's, natural pollutions pollution from desert dust and vegetation fires remains biomass burning has remained important (Mari et al., 2011; Haywood et al., 2008). However,

- 20 anthropogenic pollution has increased: there is more the economic growth over the region drives up anthropogenic emissions: the increase of industries including gas flaring (Asuoha and Osu, 2015), of local fuel-wood burning for stoves and more of traffic (Liousse et al., 2010; Hadji et al., 2012; Liousse et al., 2014) with more two-wheel vehicles using very poor quality fuel (Ndoke and Jimoh, 2005) (Assamoi and Liousse, 2010), which are suspected to quickly worsen the air quality, partly due to the very poor fuel quality used (Ndoke and Jimoh, 2005; Ndoke and Jimoh, 2010); the economic growth over the region drives
- 25 up emissions by industries including gas flaring (Asuoha and Osu, 2015)air quality.

All studies of air quality monitoring studies have shown that the outdoor air quality standards WHO air quality guidelines (*i.e.* threshold concentrations) are largely exceeded. These thresholds are for CO: 35 ppm for 1 h and 9 ppm for 8 h exposure; and for PM<sub>2.5</sub>: 10  $\mu$ g.m<sup>-3</sup> annual mean, 25  $\mu$ g.m<sup>-3</sup> for 24-hour mean. For instance, in April 1993, Baumbach et al. (1995) have measured in Lagos (Nigeria) very high levels of carbon monoxide, CO, (up to 10 ppm, measured close to high traffic road)

30 and total particulate matter (up to 200  $\mu$ g.m<sup>-3</sup>) <u>based on half-hour averages in Lagos (Nigeria</u>). In Accra (Ghana), Dionisio et al. (2010) have measured PM<sub>2.5</sub> up to 200  $\mu$ g.m<sup>-3</sup> in a polluted street (based on 1-min averages) in streets polluted by wood stoves, heavy traffic and trash burning. In Ouagadougou (Burkina Faso) PM<sub>2.5</sub> observations (based on daily averages) reach 164  $\mu$ g.m<sup>-3</sup> (Boman et al., 2009) and CO concentration measured in-traffic frequently exceed all World Health Organization (WHO) guidelines (Lindén et al., 2008). The health impact of such air pollution is expected to be high and to increase <u>further</u> without any specific emission regulation (Lindén et al., 2012). It is therefore important to gain a better understanding of <u>the pollutants' pollutant</u> emissions and transport in West Africa. All these results have highlighted the high level of pollution in megacities<u>affecting</u>, <u>affecting also</u> remote places. However, there is no continuous air quality monitoring in West Africa, so existing studies are focused on local <u>sealescales</u>, short

5 time periodperiods, and few pollutants.

25

- Several observation campaigns have been dedicated to WAM since the last decade the WAM, notably the Africa Multidisciplinary Monsoon Analysis (AMMA), which was the first international program started to improve our knowledge of on all aspects of the WAM (Redelsperger et al., 2006). WAM modeling made progresses, however the Guinean Gulf coastal region is challenging to model because of the complex land-sea-atmosphere interactions.
- Along the coastline, there are several atmospheric cells acting at on different scales. The diurnal cycle of the land-sea breeze occurs at a local scale (a few kilometers). During the day, surface wind is linked with convection within the boundary layer, while at night there is the formation of the Nocturnal Low Level Jet (NLLJ) in response to the daily deep convection activity (Parker et al., 2005). At a regional scale (a few hundred kilometers), the monsoon wind from the South meets the Harmattan wind from the North, forming the Inter Tropical Discontinuity at the ground level (Flamant et al., 2007; Cuesta et al., 2009;
- 15 Karam et al., 2009; Pospichal et al., 2010), and leading to a complex vertical structure (Haywood et al., 2008; Lafore et al., 2011). Between these two scales, an additional meridional atmospheric cell is suspected in the low atmosphere enhancing convergence at the coast (Leduc-Leballeur et al., 2013), which. This cell results from a gradient of wind speed due to the meridional gradient of sea surface temperature (de Coëtlogon et al., 2014). The recent research program "Dynamics-Aerosol-Chemistry-Cloud Interactions in West Africa program" (DACCIWA) has been dedicated to the study of land-sea-atmosphere
- 20 interactions in West Africa. It will contribute to the understanding changes in the atmospheric composition due to increasing in emissions over a rapidly growing region, as well as to the development of the next generation of accurate models to forecast weather and pollution in southern West Africa (Knippertz et al., 2015).

This article is dedicated to the pollutants transport focuses on transport of pollutants over the Guinean Gulf coastal regionand focuses on two major pollutant concentrations, in particular on: Carbon monoxide and Particulate Matter with an aerodynamic diameter  $D_p < 2.5 \ \mu m$  (CO and PM<sub>2.5</sub> hereafter), which have both an both have a detrimental impact on health (Lelieveld

- et al., 2015). The scientific questions addressed in this work are:
  - What is the relative contribution of long-range transported and locally emitted pollutants in the to surface concentrations from the Guinean Gulf to the Sahel?
  - What is the impact of meridional atmospheric cells on the transport of pollutants emitted from coastal megacities?
- 30 The pollution patterns are analyzed during the 2006 AMMA period using several observational data sets in combination with numerical simulations of the meteorology as well as of the aerosol-gas chemistry and transport presented in section Section 2. Section 3 presents the main spatial and temporal patterns over the Sudano-Guinean region of the AMMA study case. Section 4 analyzes the anthropogenic pollution from the coast to the Sahel. Section 5 refines spatially the analysis on focuses

on the analysis of the coastal dynamics and pollution transport. Section 6 focuses on specific study cases. Conclusions and perspectives are given in section Section 7.

#### 2 Weather-Pollution modeling configuration

The modeling analysis was performed using the Weather Research and Forecasting (WRF) model for the meteorological fields, 5 which drives the CHIMERE model for the gaseous and particulate species concentrations. Two nested geographical domains are defined: a continental one to take into account remote sources and long-range transport from the Mediterranean sea to the tropic of Capricorn (27°S to 44°N; 38°W to 47°E); and a regional one, centered on the Guinean Gulf (1°N to 20°N; 23°W to 17°E). The WRF and CHIMERE models work are run offline on the same two horizontal grids. The time period simulated horizontal grids for the continental and regional domains. The simulated time period is April to end of July 2006, including a one month spin-up. 10

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#### 2.1 Meteorological fields with the WRF model

The meteorological variables are modeled with the regional non-hydrostatic WRF model (version 3.7.1) presented by Skamarock and Klemp (2008). The continental domain has a constant horizontal resolution of 60 km  $\times$  60 km, and the regional domain has a constant horizontal resolution of 20 km  $\times$  20 km for the regional one, both with 32 vertical levels from the surface to 50 hPa. We use a 2-way nesting with the WRF model.

The global meteorological fields are taken from the US Global Forecast System produced by the National Center for Environmental Prediction. It is read and hourly interpolated interpolated hourly by WRF using low frequency spectral nudging above the PBL in order to enable the PBL variability to be resolved by WRF (von Storch et al., 2000). We followed the recommendations of Flaounas et al. (2010, 2011) to configure the convection and planetary boundary layer schemes, which have

optimized a better model with a set-up for the entire optimized for the 2006 WAM, especially for the meridional gradient of 20 temperature and the low level circulation.

The Single Moment-6 class microphysics scheme (WSM6) is used allowing for mixed phase processes suitable for high resolution simulations (Hong and Lim, 2006). Li et al. (2015) have shown that WAM precipitation patterns are very sensitive to the radiation scheme, and the most realistic patterns were obtained with the Rapid Radiative Transfer Model for General

- Circulation Models (RRTMG) with the Monte-Carlo Independent Column Approximation (McICA) method of random cloud 25 overlap from Mlawer et al. (1997). The planetary boundary layer physics are computed using the Yonsei University scheme (Hong et al., 2006). The cumulus parametrization used is the ensemble Grell-Dévényi scheme, as Crétat and Pohl (2012) have shown that internal variability is much larger with the Kain-Fritsch scheme than for with the Grell-Dévényi scheme at the seasonal, intra-seasonal, and daily time scales, and from the regional to the local (grid point) spatial scales. The surface layer
- scheme is based on Monin-Obukhov with a Carslon-Boland viscous sub-layer. The surface physics are calculated using the 30 'Noah' Land Surface Model ) scheme with four soil temperatures and moisture layers Ek et al. (2003) (Ek et al., 2003).

#### 2.2 Chemistry-Transport with the CHIMERE model

CHIMERE is a regional chemistry-transport model (version 2017), fully described in Menut et al. (2013a); Mailler et al. (2016). The CHIMERE model has previously been used over the AMMA observation period but only dust aerosols were modeled (Schmechtig et al., 2011; Menut et al., 2009). In this study, all important gas and aerosol sources are included (anthropogenic,

5 biogenic, mineral dust, sea salt and biomass burning). The 32 vertical levels of the WRF model are projected on the 20 levels for CHIMERE from the surface to 200 hPa. We use a 1-way nesting with the CHIMERE model.

The anthropogenic emissions are estimated by the EDGAR Team using the HTAP v2 (Hemispheric Transport of Air Pollution) annual totals for the year 2010by the EDGAR Team, using inventories based on the MICS-Asia, EPA-US/Canada and TNO databases (available at *http://edgar.jrc.ec.europa.eu/htap\_v2*). Figure 1 presents the anthropogenic PM and CO emissions

10 over the regional domain and the Cotonou-Niamey meridional transect used for the analysis in the next sections, defined in longitude  $\lambda = 2^{\circ}$  East to 3° East, and in latitude  $\phi = 1^{\circ}$  North to 19° North.

Taking into account vegetation fires emission fluxes is Biomass burning emissions from Central Africa are of primary importance to simulate West African pollution (Giglio et al., 2006). This is achieved using the APIFLAME model (Turquety et al., 2013), which estimates aerosols and chemical species emissions produced by vegetation fires biomass burning. Since

15 the incomplete combustion is both included in anthropogenic inventories (local urban burning) and fires emissions inventories (biomass burning of forests) forests biomass burning inventories, the simulation was designed to split these two parts.
Biogenic emissions are calculated using the MECAN emissions acheme (Cuenther et al. 2006). The minerel dust courses

Biogenic emissions are calculated using the MEGAN emissions scheme (Guenther et al., 2006). The mineral dust sources are obtained using a the GARLAP (Global Aeolian Roughness Lengths from ASCAT and PARASOL) new global soil and surface datasets made from a dataset made from satellite-derived aeolian roughness length data lengths with a 6 km spatial resolution GARLAP (Global Aeolian Roughness Lengths from ASCAT and PARASOL) (Menut et al., 2013b), as detailed in

20 resolutionGARLAP (Global Aeolian Roughness Lengths from ASCAT and PARASOL) (Menut et al., 2013b), as detailed in Mailler et al. (2016).

The top and lateral boundary conditions are driven by LMDZ-INCA for aerosols and chemical species (Folberth et al., 2006). The time step is set to 10 minutes for the physical processes and 5 minutes for the chemistry, which could change depending on the Courant-Friedrichs-Lewy condition. It is also possible to release gaseous or particulate atmospheric tracers, which is a

25 powerful tool to analyze the pollution patterns.

Bessagnet et al. (2004) described the calculation of gaseous species in the MELCHIOR-2 (reduced) scheme and the aerosol scheme, which takes into account species such as sulphate, nitrate, ammonium, primary organic matter (POM) and elemental carbon (EC), secondary organic aerosols (SOA), sea salt, dust and water. All aerosols are represented using ten bins, from 40 nm to 40  $\mu$ m in diameter. Their life cycle is fully represented with emission, transport, chemistry and deposition (wet and dry).

30 The top and lateral boundary conditions are driven by LMDZ-INCA for acrosols and chemical species (Folberth et al., 2006). It is also possible to release gaseous or particulate atmospheric tracers, which is a powerful tool to analyze the pollution patterns Menut et al. (2016) have detailed and analyzed aerosol speciation and size distribution in the CHIMERE model during the summer 2013 over Europe and Africa using the AERONET network for AOD and EMEP network for PM concentrations. For the AOD calculation, the aerosol optical scheme in the CHIMERE model considers mixed aerosols following the 'core-shell' hypothesis detailed in Péré et al. (2009) and evaluated in Péré et al. (2010).

In order to quantify the  $PM_{2.5}$  source apportionment, we assume that it is possible to split aerosols in different families depending on the sources because their chemical compositions are different: Mineral, Biogenic, Salt and Anthropogenic.

- 5 Given that anthropogenic and biomass burning aerosols have similar compositions, we have done two simulations with and without biomass burning emissions to split their contributions. The gas phase chemical scheme for SOA formation explained in Bessagnet et al. (2010) takes into account three anthropogenic and three biogenic hydrophilic species, three hydrophobic species with different saturations, and two surrogate compounds for the isoprene oxidation products.
- The source apportionment has been determined for CO considering three main contributors (anthropogenic sources, biomass
   burning sources and long-range transport). Consequently, three simulations have been done: one without any emission source in the domain for the background concentration, one with the anthropogenic emission only, and a last one with the anthropogenic and biomass burning emissions.

#### 3 Temporal variability from May to July 2006

In this section, we first analyze and quantify analyze the temporal variability of the pollutants concentrations modeled in the

- 15 urbanized areas along the Guinean Gulf coast precipitation, gas and aerosols during the whole AMMA-SOP1 period (Redelsperger et al., 2006). First, the precipitation regimes are analyzed identified using Hovmoller diagrams. Second, AERONET surface stations data are used to quantify the three-months variability of the Aerosol Optical Depth (AOD). Finally, the relative contribution of several sources are quantified using the modelto gases and aerosols from several sources is quantified using both airborne observations and modeling. The two last points focus on three locations: Cotonou (Benin), Djougou (Benin)
- 20 and Niamey (Niger), which are representative of locations under several influences (the combined influence of mineral dust, anthropogenic pollutionand vegetation fires), biogenic and biomass burning components.

#### 3.1 Precipitations Patterns

During this period, the precipitation location and rate will play a crucial role on for the modeled surface  $PM_{2.5}$  concentrations. As a validation for this variable, the methodology of Flaounas et al. (2010) is used: precipitation rates are averaged between

25 8.5°W and 8.5°E. Day-to-day variability is smoothed by applying a moving average of ±2 days. Figure 2 is directly comparable to the Flaounas et al. (2010) study using the same period and averaged region. Results show In May and June, observed and modeled precipitations occur mainly over the ocean (below 5°N). From late June on, the main precipitation areas move over the continent (above 5°N) and reach the Sahel (at about 13°N). Figure 2 shows that the modeled precipitation spatial patterns are in good agreement with the two satellite observations (TRMM and GPCP) presented in their study (see Figure 3)

## 30 of Flaounas et al. (2010)).

The WAM is <u>due to driven by</u> the sea surface temperature decreases , <u>which forms a cold tongue</u>, <u>over the Gulf of Guinea</u> and over the Sahara, a low thermal pressure system appears called the Saharan Heat Low (Lafore et al., 2011). The temperature

gradient between the sea and the Sahara allows the monsoon system to progress inland reaching the Sahara in July (Hall and Peyrillé, 2006; Lavaysse et al., 2009). We can notice on Figure 2 that the The monsoon progression to the North is not linear . Two jumps are observed due to the complex sea-land-atmosphere interactions constituted of two steps: the monsoon 'pre-onset' occurs (Figure 2). Two jumps are modeled at the end of May when the main precipitation area associated to the Inter

5 Tropical Convergence Zone (ITCZ) located at 2°N moves to 5°N (Sultan and Janicot, 2000; Sultan and Janicot, 2003, 2003); and the monsoon 'onset' is another abrupt shift happening end of Junewhen the main precipitation area moves from 5° to 10°N (Janicot et al., 2008). The precipitation and at the beginning of June. Precipitation is associated with large scale squall-lines, creating Mesoscale Convective Systems (MCS) moving Westward (Hall and Peyrillé, 2006).

The meteorological simulation reproduces the two changes of the main precipitation area at the that have been previously

- 10 identified from climatological averages: the 'pre-onset' (*i.e.* end of May), when the main precipitation area associated to the Inter Tropical Convergence Zone (ITCZ) located at the equator moves close to the coast (Sultan and Janicot, 2000; Sultan and Janicot, 2003 and the 'onset' (*i.e.* the 'pre-onset') and *i.e.* at the beginning of July(*i.e.* the 'onset')), when the main precipitation area reaches the Sahel (Janicot et al., 2008). For these dates, simulated precipitations match very well AMMA observations which have shown that the 2006 monsoon onset date was the 10 Julyprecipitation matches very well with AMMA observations. In 2006,
- 15 the monsoon onset occurred on the 10 July with a 10-day delay compared to its climatological date, *i.e.* which is 24 June with a standard deviation of 8 days over the period 1968-2005 (Janicot et al., 2008)according to Janicot et al. (2008). Thus, three periods could be defined: before 'pre-onset' (in May), between 'pre-onset' and onset (in June), after onset (in July).

#### 3.2 Meridional aerosols content

In our studied region, surface <u>aerosol</u> concentrations in the cities are affected by several contributions. In addition to local emission, cities may be strongly impacted by biomass burning transported from the <u>Central Africa (Mari et al., 2007)Central</u> <u>Africa (Mari et al., 2007) (Williams et al., 2010)</u>, or by mineral dust transported from Sahara (Flamant et al., 2009).

The modeled daytime Aerosol Optical Depth (AOD) and Angström exponent are compared to observations from the AERONET network (Holben et al., 1998), available at *(aeronet.gsfc.nasa.gov)*. From the daily AERONET level-2 measurements AERONET-AOD at 440nm and (at 440 nm) and the Angström exponent 440-870(440 nm - 870 nm), AERONET-AOD is

25 calculated at 600 nm(at 550 nm) based on the Angström law. A spatial bilinear interpolation of the model outputs is performed at the station location.

Two AERONET stations are located close to the meridional transect studied (Figure 1): Banizoumbou (13.5°N, 2.1°E) in the suburb of Niamey in Niger, and Djougou in Benin (9.7°N, 1.6°E) North of the strongly urbanized areas around Cotonou. Comparisons are We compare a simulation made with and without biomass burning emission (with/without BB), presented in

#### 30 Figure 3.

There are two important events of coarse particles recorded at both sites, associated with a low Angström exponent (*i.e.* Angström exponent lower than 0.5 as in Ogunjobi et al. (2008)) and AOD greater than 1, between 13-14 May and between 10-13 June. The model captures the magnitude of these large scale dust events. During the studied period, the events of coarse particles are well reproduced (high or moderate AOD are generally associated with a low Angström exponent). There is an

increase of the Angström exponent, *i.e.* fine particles over the period, which is well reproduced by the model. Frequent fine aerosol events (high Angström exponent) have been monitored corresponding to low or moderate AOD, which are partially captured by the model.

The addition of the biomass burning <u>emission emissions</u> lead to an important plume of gas and aerosols reaching the Guinean <u>gulf</u> Gulf in June. Modeled AOD with biomass burning emissions are well in the range with biomass emission (bias is reduced)

- 5 gulf Gulf in June. Modeled AOD with biomass burning emissions are well in the range with biomass emission (bias is reduced) of the observations but the variability is not captured. The Angström exponent is associated in Maywith In May, the aerosol content is mostly composed of coarse particles (Angström exponent about 0.2), in June with of a fine/coarse mixture of particles (about 0.5), in July with a fine particles(of finer particles, especially at Djougou (Angström exponent about 0.8). The model is able to reproduce this increase of the Angström exponent, which suggests an aerosol origin transition, from a period dominated
- 10 by desert dust to a period of fine particles which could be <u>local</u> urban or/and biomass burning pollution from the South.

#### 3.3 Meridional aerosols and gases concentrations

In this section, firstly the modeled CO and  $PM_{2.5}$  concentrations are compared to <u>aircraft</u> observations collected during the AMMA campaign <u>-Secondly(3.3.1)</u>. Then, the different <u>contribution contributions</u> of the pollution sources <u>is\_are</u> analyzed from the modeled concentrations at the three studied sites (3.3.2).

#### 15 3.3.1 Airborne observations

Two flights made during a 'North-South land-atmosphere-ocean interaction' mission plans have been conducted over the Cotonou-Niamey meridional transect. We are studying two flights conducted along a meridional transect between Cotonou and Niamey on 13 and 14 June 2006. These two dayscorrespond to disturbed dynamics of the WAM due to 2006 as part of a 'North-South land-atmosphere-ocean interaction' survey mission. During these two days, the WAM dynamics over the

- 20 area were perturbed by the presence of a MCS. It developed in the vicinity of the Jos plateau in the North of Nigeriaover the Jos Plateau (Nigeria) around 16UTC, moving westward to the center of Ghana, which have already been described (*e.g.* (Flamant et al., 2009) and (Crumeyrolle et al., 2011)). Moreover, the :00 UTC, reaching the Benin-Nigeria border at 20:00 UTC and moved southwestward across Benin overnight and into central Ghana, as already described in Flamant et al. (2009) and (Crumeyrolle et al., 2011). The model reproduces the location of this MCS but earlier than in the observations, *i.e.* reaching
- 25 the Benin-Nigeria border at 10:00 UTC (Figure 4). The MCS interacts with the dust layer coming from the Sahara (especially from the Bodele depression), changing the dust load and vertical distribution over Benin and Niger. Associated with subsidence in the wake of the MCS, there is a lowering of the dust layer height (Flamant et al., 2007) (Flamant et al., 2009).

Modeled CO and  $PM_{2.5}$  concentrations are compared to aircraft measurements performed onboard the ATR-42 aircraft (with PCASP instrument for PM), which have been averaged at a 2-minute time step. The modeled values are interpolated

30 along the aircraft trajectories, in time between the two closest modeled hourly outputs, and vertically between the two closest model vertical levels and horizontally with a bilinear interpolation. For the two flights (13 July in the morning from Niamey to Cotonou, and 14 July in the afternoon from Cotonou to Niamey), Table 1 presents modeled and observed mean spatial value and range values and ranges of CO and PM<sub>2.5</sub> concentrations in the PBL (altitude lower than 1000 m) over three regions: Coastal

region including Cotonou (6.3°N - 9.0°N), Sudano-Guinean region including Djougou (9.0°N - 11.0°N), Sudano-Sahelian region including Niamey (11.0°N - 13.5°N).

For CO concentration, on 13 June, there is no clear gradient over the three regions but rather a constant concentration of about 170 ppb. On 14 June, a gradient is noticed we can noticed a gradient from the coast (200 ppb) to the Sahel (167 ppb).

- 5 For both days, the model predict an opposite predicts a gradient with the highest concentration over the Sahel. Over the coastal region, the observed CO concentration range is similar for the two flights (between 147 222 ppb), which is well in agreement with the modeled range (between 175 240 ppb). Over the Sudano-Guinean region, the observed range of variation is 161 182 ppb prior to the MCS (13 June), and it increases to 153 233 ppb after the MCS (14 June). The model is able to capture the larger range-variability on 14 June than on 13 June (205 247 ppb compared with to 218 245 ppb on 13 June). Over
- 10 the Sudano-Sahelian region, the observed range variability of CO concentration is also larger on 14 June (146 200 ppb) than on 13 June (149 - 174 ppb). This behavior is not reproduced in modeled concentrations. There is an over-estimation of the modeled CO concentration (positive bias of  $\approx 20$  ppb) for 13 and 14 June.

For the  $PM_{2.5}$  concentration concentrations, a South-North gradient is expected moving closer with the highest concentrations close to the Sahara. There is a clear gradient in the observed  $PM_{2.5}$  concentration mean on 13 June :-between the coastal region

- 15 (8  $\mu$ g.m<sup>-3</sup>for the coastal region, ) and the Sudano-Guinean region (50  $\mu$ g.m<sup>-3</sup>for the ), and almost the same concentration in the Sudano-Guinean region, and the Sudano-Sahelian region (56  $\mu$ g.m<sup>-3</sup>for the Sudano-Sahelian region). After the MCS, there is no clear gradient but rather the same concentration over the coastal and the Sudano-Guinean regions (39  $\mu$ g.m<sup>-3</sup>) and a gap of concentration over. However, there is an important increase of the concentration moving to the Sahel (up to 92  $\mu$ g.m<sup>-3</sup>). The ranges are variability is increased over the three regions: 37 - 42  $\mu$ g.m<sup>-3</sup> for the coastal region, 24 - 59  $\mu$ g.m<sup>-3</sup>
- for the Sudano-Guinean region, 50 139  $\mu$ g.m<sup>-3</sup> for the Sudano-Sahelian region. The modeled ranges match the observed one-ones for both days. The model reproduce reproduces a South-North gradient on 13 June, which is well in agreement with the observations. On 14 June, the model predict predicts the concentration gap between the coastal and the Sudano-Guinean regions (from 43 to 82  $\mu$ g.m<sup>-3</sup>), while it was observed between the Sudano-Guinean and the Sahelian regions.
- The 13 and 14 June 2006 correspond to disturbed These two days correspond to disturbed meteorological conditions, which may not be representative of the typical average concentrations. The model-observation comparison suggest suggests that the MCS is not well reproduced occurs later in the observation, which could in turn induce a not realistic unrealistic modeled pollution plume (for instance a biomass burning plume) over the Sudano-Sahelian region and the Sahel. Nevertheless, there is a over-estimation of the modeled CO concentration (positive bias of  $\approx$  20 ppb) for these two days the order of magnitude of the CO and PM<sub>2.5</sub> concentrations are good in agreement with observations.

#### 30 3.3.2 Monthly modeled pollution sources apportionment

In order to analyze the source apportionment, we consider that the CO mixing ratio is due to three major sources: background, anthropogenic and fires, and biomass burning, and that the  $PM_{2.5}$  mass concentration comes from five major types of pollution source: anthropogenic, firesbiomass burning, mineral dust, biogenic and sea salt (we assume that  $PM_{2.5}$  background concentration is negligible). For the whole period and for each month of the simulation, the The relative percentage of each source is

presented for CO in Table 2 and for  $PM_{2.5}$  Table 3 at the three studied locations: Cotonou, Djougou and Niamey, for the whole period and for each month of the simulation.

For the three sites, the average concentrations of surface CO increase during the whole period. The mean concentrations are very close for the three sites: 221 ppb in Cotonou, 227 ppb in Djougou, and 212 ppb in Niamey. There is a clear increase of the

5 CO from May (157 - 180 ppb) to July (267 - 280 ppb). This increase is due to the vegetation fire biomass burning sources from May (3 - 10 %) to July (40 - 49 %), while the anthropogenic and background concentrations are stable during the whole period and for the three all sites. It seems that the CO overestimation noticed in the previous section is linked with an overestimation of vegetation fire biomass burning emissions.

Considering PM<sub>2.5</sub> concentrations, there is on average over the whole period display a South-North gradient of concentra-

- 10 tions (30  $\mu$ g.m<sup>-3</sup> in Cotonou, 38  $\mu$ g.m<sup>-3</sup> in Djougou, 54  $\mu$ g.m<sup>-3</sup> in Niamey) on average over the whole period, consistent with the gradient of the dust contribution (15 % in Cotonou, 35 % in Djougou, 67 % in Niamey). From May to July and for the three all sites, the mineral dust contribution is in constant decrease. On the other hand, the vegetation fires biomass burning contribution increases (3 to 19  $\mu$ g.m<sup>-3</sup> in Cotonou, 1 to 17  $\mu$ g.m<sup>-3</sup> in Djougou, 0.6 to 11  $\mu$ g.m<sup>-3</sup> in Niamey), which eould seems to be overestimated as for CO concentration. PM<sub>2.5</sub> concentrations are dominated by natural sources. Nevertheless
- 15 anthropogenic Anthropogenic PM<sub>2.5</sub> concentrations range between from 3 to 5  $\mu$ g.m<sup>-3</sup>, which is about 10 % for the whole period and for the three sites.

In Cotonou, the average concentrations concentration of surface  $PM_{2.5}$  increases during the whole period, from 23 to 37  $\mu$ g.m<sup>-3</sup>. This mainly corresponds to the arrival of vegetation emissions biomass burning emission products, transported from Central Africa to the Guinean Gulf, with an increase from 11 to 52 % from May to July. On the contrary, the mineral dust contribution decreases during the period, from 38 to 5 %. The sea salt contribution increases from 3 to 6 ppb. During the three months, the anthropogenic and biogenic contributions remain stables stable at about 4 ppb- $\mu$ g.m<sup>-3</sup> and 6 ppb- $\mu$ g.m<sup>-3</sup>

respectively.

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In Djougou, the same behavior is observed but with some changes in the absolute values. The relative contribution of mineral dust decreases from 57 % to 14 %, while the fire-biomass burning contribution increases from 3 to 47 %. The anthropogenic contribution is slightly higher in June at about 5 ppb. $\mu$ g.m<sup>-3</sup>.

In Niamey, the dust contribution is important for the three months. It decreases by a factor 4, from 61 to 15  $\mu$ g.m<sup>-3</sup>, consistently consistent with observation of PM<sub>10</sub> in Banizoumbou in Niger (Marticorena et al., 2010), which is probably due to the reduction of local emission linked with the increase of vegetation cover. The relative contribution of anthropogenic pollution is slightly higher in June at about 5 ppb. $\mu$ g.m<sup>-3</sup>.

For CO or PM<sub>2.5</sub> concentrations, the anthropogenic contribution is always important in emissions contribute significantly to the total budget ( $\approx$  30 % for CO and  $\approx$  10 % for PM<sub>2.5</sub>). It is therefore important to better understand the daily variability of anthropogenic pollutants pollutant transport.

#### 4 Focus on anthropogenic pollutants from Cotonou to Niamey

This section focuses on anthropogenic pollution the horizontal variability and vertical structure of anthropogenic pollution. Only the contribution of anthropogenic sources is considered in  $PM_{2.5}$  and CO concentrations, from now on referred to as anth- $PM_{2.5}$  and anth-CO.

#### 5 4.1 Time-latitude variability at the surface

#### 4.1.1 CO and PM<sub>2.5</sub> concentrations

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The Cotonou-Niamey meridional transect includes the two specific cities extensively studied in the framework of the AMMA program: a coastal megacity (Cotonou in Benin) and a Sahelian city (Niamey in Niger). To highlight the latitudinal regional transport, modeled concentrations are presented with the same methodology as in the previous section with Hovmöller diagrams, corresponding to time-latitude average of variables (data are smoothed with a 5-day moving average, *i.e.*  $\pm 2$  days).

- Results are presented in Figure 5 for anth-CO and anth- $PM_{2.5}$ . For the two species, meteorological parameters are superimposed to on the concentrations. The precipitation Precipitation rate contours are defined for events with more than 10 mm/day over the Cotonou-Niamey transect. These are similar but not equivalent patterns to those presented in section 3.1 averaged over the entire West Africa.
- The Inter-Tropical Discontinuity (ITD) is the limit between the northward monsoon wind and the southward Harmattan wind (Flamant et al., 2007; Karam et al., 2009). The ITD could be defined as the isocontour of relative humidity (RH) equal to 20 %. We can notice that the location of the ITD marks associated with a sharp gradient in surface anthropogenic concentrations, with a decrease going further North from 100 to 20 ppb for CO, and from 5  $\mu$ g.m<sup>-3</sup> to 2  $\mu$ g.m<sup>-3</sup> for PM<sub>2.5</sub>.
- For anth-CO surface concentrations, high concentrations are continuously noticed during the whole period modeled from the beginning of May to late June at the coast where Cotonou is located, about 100 ppb around  $\phi = 6.3^{\circ}$ N. Over the Guinean gulfGulf, the concentration is low between 20 and 50 ppb. The second area with high anth-CO values corresponds to the Sudano-Sahelian region, where concentration vary between 60 and 100 ppb around  $\phi = 12^{\circ}$ N. Whereas high concentrations of anth-CO at the coast is are clearly related to local emissions, the high concentrations over the Sahel should could be either due to transport, or local emissions. Over the studied domain, anth-CO surface concentrations evolves evolve between 20 and 120 ppb.
- 25 There are in May and in June some high modeled surface concentrations in May and in June when rain occurs, such as around 11 June. In July, the variability is mostly consistent with precipitation rates after the onset, which suggest that surface versus vertical distribution has changed suggesting modifications of transport and deposition patterns by the convection associated with large scale precipitation. The precipitation Precipitation variability can thus explain only a part of the CO variability. It is necessary to also investigate the large-scale wind speed and direction.
- The same behavior is observed for the surface concentrations of  $PManth-PM_{2.5}$ . The week to week variability is more important. This increase greater than for anth-CO, which is probably due to the longer CO lifetime lifetime of CO compared with that of PM (being less chemically active and without settling), the CO concentrations are less prone to settling). CO is more homogeneously mixed than PM in a large latitudinal area spanning from the coast to more latitudes higher than 16° up

to the North. The temporal variability of surface PManth-PM<sub>2.5</sub> exhibits a frequency close to 2-weekperiodicity close to two weeks: during the whole modeled period, five higher concentration periods are observed from the coast to  $\phi \approx 16^{\circ}$ N. In addition in these latitudinal plumespatterns, local minima are modeled, for instance the 20 Juneat  $\phi \approx 13^{\circ}$ N. At  $\phi = 12^{\circ}$ N, there is an area of high concentration , which is present over the whole period. This may be related to vertical transport and will

5 be quantified in the next sections. The results showing similarities for both two pollutants in terms of time-latitude variability, the next sections will refine the analyzes only for PM analysis only for anth-PM<sub>2.5</sub>.

#### 4.1.2 Synoptic wind and pollution

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This section aims at analyzing the anth-PM<sub>2.5</sub> concentration temporal frequency variability through the surface wind speed and direction. The previous section has shown that low pollution levels are concentration is not always associated with precipitations. Results are presented in the same way using Hovmöller diagrams on Figure 6(Figure 6). For each figure, colored isocontours are superimposed corresponding to surface anth-PM<sub>2.5</sub> of 4 and 5  $\mu$ g.m<sup>-3</sup>.

For the surface wind speed, the lowest values are modeled along the coast during the whole period. The wind speed variability is weak from the coast to the Sahel. Periods of low wind speed are coincident with the highest values of surface PManth-PM<sub>2.5</sub>. At the end of the period, when precipitation occurs inland and anth-PM<sub>2.5</sub> is low, the meteorological situation

15 changes and the wind speed increases suddenly over the ocean showing the cold tongue arrival as previously described by Meynadier et al. (2016), when there are precipitations inland and low anthropogenic pollution levelslocated at the Equator, which is associated with increased wind speed between the Equator and the coast, as detailed by Meynadier et al. (2016).

Regarding the precipitations precipitation occurrences discussed in the previous section, the high surface anth-PM<sub>2.5</sub> concentrations modeled around  $\phi$ = 12°N are due to a combination of low wind speed and low precipitation rates. These meteorological conditions are representative of stagnation, which accumulate pollutants in the lowest layers of the troposphere.

For the surface wind direction, the main wind direction near the coast is the South-West quarter during the whole period. There is no obvious link between wind direction changes and  $PM_{2.5}$  highest values over the Cotonou-Niamey meridional transect.

The large scale variability of meteorological variables (precipitation and wind speed) controls the period of high anthro-

25 pogenic pollution from the coast to the Sahel. However, it does not explain if the high concentration over the Sahel are linked with local emissions or/and with pollutants transport from the coast.

#### 4.2 Monthly mean vertical structure

#### 4.2.1 Anthropogenic pollution

We now focus on the vertical structure of the lower troposphere from the surface to 4 km altitude in order to understand what is

30 responsible for hte causes of the high anth- $PM_{2.5}$  concentration over the Sahel. Monthly averages of anth- $PM_{2.5}$  concentration are analyzed together with wind eirculations circulation (monthly averages correspond to consistent meteorological periods, *c.f.* Section 3.1). In Figure 7, the modeled concentrations are spatially averaged over the Cotonou-Niamey meridional transect. Three isocontours (3, 4 and 5  $\mu$ g.m<sup>-3</sup>) are used to follow the anthropogenic pollution patterns. Results are presented at 00 UTC when the NLLJ is established (on the contrary, well established (during the day, the pollution is more mixed in the PBLby dry convection).

For the three months, the meridional wind is lower at the surface than in the boundary layer from the coast to ≈ 9°N,
highlighting the well established NLLJ. Above the northward monsoon flux, there is the SAL associated with southward winds. The highest southward wind speed in the core of the SAL between 11 and 16°N in latitude and 2 to 4.5 km in altitude is the African Easterly Jet (AEJ).

Regarding the wind, two atmospheric cells are noticed on Figure 7 modeled during the three months (Figure 7). There is a large cell going northward at the surface within the monsoon flow, and going backward toward the South with the SAL (or

10 AEJ), located at  $\approx 2$  km altitude, and between the coast and  $\approx 16^{\circ}$ N. There is also a small cell turning in the same direction (elockwiseanti-clockwise) at  $\approx 2$  km altitude, with the downdraft at 6°N and the updraft at 7°N. In May and June, the small cell is included in the large cell, while in July, they are disconnected. These two cells seem to interact with the anthropogenic pollution because the anth-PM<sub>2.5</sub> isocontours shape appear to be driven by the wind patterns.

Regarding anth-PM<sub>2.5</sub> concentration, these two atmospheric cells seem to interact with the anthropogenic pollution because 15 the anth-PM<sub>2.5</sub> isocontours shape appear to be driven by the wind patterns. The large atmospheric cell induces a recirculation of the modeled anthropogenic plume, ranging from  $\phi = 6$  to 18°N in latitude and 0.5 to 3 km in altitude (anth-PM<sub>2.5</sub> isocontour of 3 µg.m<sup>-3</sup>). We can notice that the recirculation center The center of this cell is located at  $\phi \approx 14$  °N during the three months studied. In May, an important part of the pollution from the coast is transported in altitude within the NLLJ above the PBL (displayed by the anth-PM<sub>2.5</sub> isocontour of 4 µg.m<sup>-3</sup>). In June, there is high concentration in altitude over the Sahel (displayed

20 by the anth-PM<sub>2.5</sub> isocontour of 5  $\mu$ g.m<sup>-3</sup>), which suggests that the atmospheric cell concentrates pollutants. Note that in July, the lowest latitude of the anthropogenic plume is moved northward, starting at  $\approx 10^{\circ}$ N, and less connected with the coasthigh anth-PM<sub>2.5</sub> concentration are only modeled along the coast and close to the surface.

Air masses transport anthropogenic pollutants from the coast to the Sahel. High surface concentrations of anth-PM<sub>2.5</sub> are modeled at the latitude of the coastal urbanized areas ( $\phi = 6.3^{\circ}$ N), leading to a plume to the North within the NLLJ and concentrating pollutants over the Sahel.

#### 4.2.2 Coastal versus Sahelian pollutants meridional transport

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A tracer experiment has been set-up to analyze if the main contributors to the Sahelian maximum are emitted locally or remotely at the coast. Gaseous tracers are released at the two major cities of the meridional transect (without any sink): Cotonou (Benin ) and Niamey (Cotonou in Benin and Niamey in Niger). The tracers tracer experiment uses arbitrary units and considers the same quantity of tracers emitted in each town. The tracers are constantly released from the 1 to 30 June. The emission altitude occurs in the PBL (0 - 500-200 m) from the 1 June to 30 June. Results are presented in Figure 8-averaged during the ten last days at 00 UTC considering as in the previous section. We consider either only coastal emissions or only Sahelian emissions in order to observe where air recirculation may concentrate the pollutants (Figure 8).

Tracers emitted at the coast indicate that there is a contribution an important transport of coastal pollutants over the Sahel. The ratio of Sahelian tracers concentration and coastal tracers concentration toward the North in the PBL. On the other hand, there is no significant transport of tracers emitted in the Sahel is between 10 % and toward the coast. In Niamey, Cotonou tracer concentration is about 9% (of the 1 %; *i.e.* with coastal emission 10 to 100 times higher, the anthropogenic coastal

- 5 pollution could be equivalent to the anthropogenic Sahelian pollution over the Sahel.a.u. isocontour presented in Figure 8-a), while in Cotonou, Niamey tracer concentration is about 0.03% (of the 1 a.u. isocontour presented in Figure 8-b). In the HTAP anthropogenic inventories (presented in Figure 1), the anth-PM<sub>2.5</sub> (respectively anth-CO) is at Niamey  $\approx$  103 (735) kg.km<sup>-2</sup>.day<sup>-1</sup> and at Cotonou in Niamey and  $\approx$  438 (7707) kg.km<sup>-2</sup>.day<sup>-1</sup> in Cotonou. Therefore, an important part of the pollution over the Sahel has been emitted at the coast , which and it contributes to a maximum of anthropogenic pollution
- 10 in June over the Sahel. In conclusion, the high concentration in altitude over the Sahel are due to atmospheric cells, which concentrate is due to the existence of a meridional atmospheric cell, which acts at accumulating pollutants emitted locally and remotely at the coast.

#### 5 Impact of coastal dynamics on anthropogenic pollution

In order to better characterize the coastal pollution, anthropogenic PManth-PM<sub>2.5</sub> for the period of 8 to 15 June 2006 are now 15 described at hourly temporal resolution. This week includes a large variability of low to high surface concentrations.

#### 5.1 Surface hourly pollution variability

The surface hourly anthropogenic PMhourly surface anth-PM<sub>2.5</sub> concentrations are shown on in Figure 9 over the Cotonou meridional transect. The highest temporal resolution shows the same variability as described in Figure 5 with the with Hovmöller diagrams. The beginning of the week associated with lower concentrations.

20 <u>, as already described in Figure 5.</u> For most days, the highest surface concentrations are modeled between 18 UTC and 00 UTC from the coast to 8°N. This coincides with the lowest boundary layer height, which concentrates urban emissions (i.e waste burning and traffic) in a thin layer (not shown). It seems that urban plumes start from the coastline, corresponding to the city, at the sunset (18 UTC) and derives to the North in the next few hours.

At the coast, anth-PM<sub>2.5</sub> concentration decreases at night (between 00 UTC and 06 UTC) and  $\frac{1}{2000}$ , increases again in the morning

25 (around 06 UTC), concentration increases again at the surface when the convection and NLLJ are weak, which follows traffic emissions. This feature follows traffic emissions, which are included in the anthropogenic emission inventories. It seems that urban plumes start from the coastline at sunset (18 UTC) and are transported to the North in the following hours.

There is a transition from low to high level concentration of anthropogenic pollution from the 8 to 12 June. Anthropogenic pollution is modeled over the sea from 10 to 11 June. It is interesting to note on the 11-12 June night that high concentrations

30 are modeled during the day when there is precipitation inland that precipitation occurs inland on 11 June (between 18 UTC and 00 UTC), then high modeled concentrations persist during the night of 11-12 June. This precipitation event reflects a change

in the wind patterns, which induces a <u>change in the</u> transport of pollutants<del>over the sea (with, leading to</del> surface concentrations up to  $\frac{3.8 \ \mu g.m^3}{1000}$  in Cotonou.

#### 5.2 Contribution of other cities at Cotonou

At the coast, the wind direction comes from the sector S-SW but there are diurnal variations , which could affect the air pollution along the coastline. In order to distinguish pollutants pollutant transport from the different coastal megacities, a tracer experiment has been set-upset up. The tracers are constantly released in the lowest model layers ranging from surface to 500 m above sea level. This altitude corresponds to emission below or within the NLLJ. Three point sources have been defined: Accra in Ghana (5.6°N,0.2°W), Lome in Togo (6.2°N,1.2°E) and Cotonou in Benin (6.4°N,2.4°E). The results are presented over the Cotonou meridional transect (Figure 10). We aim at evaluating the impact of the Cotonou local emissions versus emissions

10 from distant areas transported toward Cotonou. The emission (in arbitrary unit) has the same magnitude in each city without any daily variation.

As expected, the surface concentrations of Cotonou due to local emission only are the highest at the coast. The diurnal cycle of pollutants pollutant transport appears clearly with the highest concentrations exported at the beginning of the night (18 UTC), when the boundary layer height decreases quickly and when the establishment of the NLLJ occurs. Up to 9°N, high tracers concentrations are transported from Cotonou far from the point source. The Cotonou plume always transports tracers

15 tracers concentrations are transported from Cotonou far from the point source. The Cotonou plume always transports tracers up to 7°N; for some days, these plumes may reach the latitude of 9°N during the night between 18 UTC and 06 UTC.

At Cotonou, the modeled tracers concentration released from Lome or Accra are logically lower because the source points are not in the Cotonou meridional transectstudied. The same kind of northward transport is observed but pollutants transport from Accra and Lome reach Cotonou in the morning between 06 UTC and 12 UTC for the Lome plume, and in the afternoon between

- 20 12 UTC and 18 UTC for the Accra plume. This result is consistent with a transport speed between 10 and 20 km.hour<sup>-1</sup> (the Lome-Cotonou distance is about 150 km, and Accra-Cotonou about 300 km). Moreover, there is probably a diurnal variation of the wind direction, coming and synoptic wind direction from the sector S-SW during the day and SW-W at night because of the land/sea breeze, which carry more efficiently pollutants emitted during the night toward Cotonou.
- <u>S-W.</u> In arbitrary unit and with the same emission at the three cities, the concentrations modeled at latitude modeled
   <u>concentrations</u> between 7°N and 8°N are similar for the Lome and Cotonou plumes, and <u>in a lower extent lower</u> for the Accra plume. This means that the urbanized areas West from Cotonou (Lome or Acera) contribute as much as Cotonou to the anthropogenic pollution at those locations (when considering the same emission).

The comparison between the three plumes shows a specific behavior during 10-12 June as the Cotonou pollution is not exported to the North:

On 10 June, the Lome plume is clearly exported over the sea and very high concentrations are noticed over Cotonou. The same behavior is observed for the Accra plume with lower concentration because this location is further from Cotonou. Indeed, the Lome and Accra plumes reach Cotonou after being transported over the sea, which suggests that pollutants at the three cities have been transported Eastward, leading to plumes overlaying mixing at the Cotonou location.

- On 11 June, there are still high concentrations over Cotonou due to the Accra plume probably driven by the same meteorological conditions, but it does not affect the Lome plumedoes not have a specific behavior. This suggests a perturbation affecting especially Accra.
- The On 12 Junecorresponds to the most, there is an important transport of Cotonou emission pollution to the North. At the same time, this is the only day when Lome and Accra plumes do not reach Cotonou, as they are shifted to the North at 7°N when crossing the Cotonou meridional transect.

All these results suggest a fast change of the meteorological situation, leading to air pollution. In the next section, the specificity of the vertical wind structure during this period will be studied in detail.

#### 6 Disturbed atmospheric dynamics and pollution transport

10 In this section the analysis is refined to two periods of two days on 8-9 and 11-12 June 2006. 2006 in the Cotonou area. The first corresponds to non-perturbated non-perturbed monsoon flow situation leading to low anthropogenic pollution, the second corresponds to a disturbed perturbed situation leading to high anthropogenic pollution.

#### 6.1 Evolution of the vertical structure

In order to focus on the day/night transport from the coast to the North (described in section 5.1) and the changes in dynamical

- 15 regimes during this period (described in section 5.2), results are presented as vertical slices in Figure 11, averaged along the Cotonou meridional transect. The tracers concentrations, emitted separately at Lome and Cotonou, are presented as isocontours of threshold values: the emissions being arbitrary, the modeled concentrations are also arbitrary. But the same threshold is used for the two emissions locations, thus concentration magnitudes are comparable. The wind vectors (meridional/vertical components) are superimposed on the figures to highlight the vertical cells. The meridional wind is also presented as color
  20 shading for the NLL Lintancity.
- 20 shading for the NLLJ intensity.

5

The first period , (8 June at 23 UTC and 9 June at 11 UTC, ) corresponds to a classical monsoon case, often observed and described in the literature (Abdou et al., 2010; Lothon et al., 2008). At night, surface pollutants are concentrated in a shallow layer (less than 200m), corresponding to nocturnal surface layer and to the lowest part of the NLLJ (represented by the 'dark blue' shaded area in Figure 11). The Cotonou and Lome plumes are mixed. During the day, the convection induces more mixing

25 over land than over sea, where the boundary layer reaches 1500 mmixing in the boundary layer, which reaches 1500 m at 11 UTC over the continent. The Lome plume does not reach the coast, but it crosses the Cotonou meridional transect more further to the North  $> 6.5 (\approx 7^{\circ} N)$ .

On 8-9 June, an updraft-downdraft convective cell is clearly observed during the day and at night, with ascendent wind at  $7^{\circ}$ N and subsident wind at  $6.3^{\circ}$ N (the Cotonou site latitude). This circulation has already been observed for the whole studied

30 period in section 4.2.1. This is not a modeled land-sea breeze because it turns in the same direction day and night. Land-sea breezes have not been explicitly modeled because of the too coarse resolution (about 20 km).

The second period , (11 June at 23 UTC and 12 June at 11 UTC, ) corresponds to a disturbed case compared to what is usually observed in this region and for this month(for instance 8-9 June). Indeed, for the 11 June at 23 UTC, the NLLJ is not present near the coast and the wind is weak from the coast to 8°N. The modeled nocturnal PBL height is very low (less than 50m50 m). The Lome plume is not present over Cotonou. On 12 June at 11 UTC, air subsidence is modeled from 7°N to

5 10°N. The isocontours of concentrations due to emissions in Lome and Cotonou are at the same latitude, corresponding to an iso-latitudinal transport, along the coast. Compared to the 8-9 June, there is no coastal cell located over the emission region. But a A larger cell is modeled, with high meridional wind speed (up to 4 m.s<sup>-1</sup>). The subsidence, located at  $\phi > 7^\circ N$ , imports upper air masses from the free troposphere and blocks the northward transport of the coastal pollutants.

#### 6.2 Specificity of 11-12 June 2006

- In this last section analysis, we focus on the 11-12 June to understand which meteorological conditions have led to an important modeled anthropogenic  $PM_{2.5}$  event at Cotonou. Schwendike et al. (2010) have shown that a large MCS has occurred over Ghana due to convective instabilities at the border of Togo, Ghana and Burkina Faso. Some spots of convection ('Pop corn' convection) over a large region including Cotonou has been identified on 11 June. An isolated convective cell lasting a few hours coming from South-East and going moving North-West have has crossed the coastline over Cotonou at around 18
- 15 UTC (Figure ??Figure 12), which is well in agreement with the modeled location (Figure 13). When precipitation is inland between 19 UTC and 23 UTC(Figure 13 top), the wind speed is null over the coast because the monsoon flux is blocked (Figure 13 top). During these specific meteorological conditions, the high anth-PM<sub>2.5</sub> surface concentrations are thus due to an accumulation of pollution during a few hours (from 19 to 23 UTC).

The same tracer experiment as described in the previous sections (described in section 5.2) is used to test our hypothesis

20 about confirm the accumulation of pollutants and to distinguished plumes of the different cities. Gaseous tracers are released with the same emission at four cities: Accra (Ghana), Lome (Togo), Cotonou (Benin) and Lagos in Nigeria has been added (6.5°N,3.4°E).

We can notice (on Figure 13 -bottom) that the pollution emitted at the different cities West from Cotonou is mixed at  $\phi = 7^{\circ}$ N on 11 June at 19 UTC (Figure 13 -bottom). Six hours later, only the Cotonou plumes is responsible for the high anth-PM<sub>2.5</sub>,

25 which confirms pollutants accumulation because it has been blocked by the down-draft of the precipitation system during six hours.

This result demonstrates that during the monsoon period, specific meteorological conditions could lead to high pollution in the Guinean coast megacities, although most of the time pollution emitted along the coastline are quickly transported to the North.

#### 30 7 Conclusions

The West African pollution has been was studied using both models and observations during May, June and July 2006. This corresponds to the beginning of the WAM and West African monsoon and it includes the AMMA campaign observational

period. The focus is was on urbanized areas , located along the Guinean Gulf coast and known as large gas and aerosol emitters. In addition to these anthropogenic emissions, the coast is often under the influence of long-range transport of mineral dust and vegetation fires biomass burning emissions. The analyses are performed for CO and  $PM_{2.5}$  over a large domain to include all sources: Central Africa for biomass burning, Sahel and Sahara for mineral dust and a large part of the Guinea Gulf

5 for sea salt.

20

The first analysis was devoted to estimate the relative contribution of each source during the three months in Cotonou (Benin), Djougou (Benin) and Niamey (Niger). It was shown that the surface concentrations of  $PM_{2.5}$  constantly increase during the period. The mineral dust dust relative contribution remains low close to the coast, showing that in monthly average, pollution during this period is not dominated by mineral dust transport events. On the other hand, the vegetation fires biomass

10 <u>burning emissions</u> increase from May to July. The anthropogenic part is stable during the whole period for the three studied sites at  $\approx 50$  % for CO and  $\approx 15$  % for PM<sub>2.5</sub>.

The second part of the study was focused on analyzed the anthropogenic contribution of to CO and  $PM_{2.5}$  along a Cotonou-Niamey meridional transect. A transport of these pollutants These pollutants are transported from the coast to the north has been demonstrated up to as far as the Sahel (13°N). The Northward limit of the transport corresponds to the Inter Tropical

15 Discontinuity. It was also shown that there are alternating periods of high/low concentration from Cotonou to Niamey with a weekly frequency. To understand this variability, meteorological variables have been were investigated. The highest surface pollutants concentrations occurred when there is no precipitation and low wind speed.

In order to better understand the meridional transport and the occurrence of high pollutants pollutant concentrations over the Sahel ( $\approx 13^{\circ}$ N), monthly averages of vertical wind structure were analyzed. From May to June, a large atmospheric cell going from the coast to the Sahel remains present and it has been identified as responsible for the pollutant accumulation over the Sahel of pollutants emitted locally and remotely at the coast.

A focus has been done was put on coastal dynamics and pollution transport during a restricted period, from 8 to 15 June 2006, including which included high and low levels concentrations of anthropogenic pollution. To isolate the coastal dynamics impacts on several cities plumes from city plumes along the coastline, a tracer experiment was designed with emissions at Accra

25 (Ghana), Lome (Togo) and Cotonou (Benin). The tracer concentrations confirm that, in Cotonou, the modeled concentrations are both due to due to both local and remote emissions. A meridional transport of the anthropogenic pollution from the coast to the North has been highlighted at night linked with the Nocturnal Low Level Jet close to the coast.

Finally, two contrasted anthropogenic pollution situations were detailed analyzed in detail. The first situation (8-9 June) corresponds to low anthropogenic pollution during a 'typical' case of monsoon dynamics, while the second situation (11-12

30 June) corresponds to a disturbed meteorological situation due to a MCS. During the convective system. During 11-12 June, it was shown that air subsidence is modeled at latitude 7°N, which imports clean upper air masses from the free troposphere, limiting the northward transport of the coastal pollutants. The main results of this article will be compared to-

Concerning air quality and climate policy development, we showed that the export of anthropogenic pollutant from the Guinean coast toward the North could lead to cross boundary pollution plumes. This result will be confirmed by comparison

to the 2016 DACCIWA campaign observations in order to confirm and refine your conclusions propose strategy to reduce atmospheric pollution in West Africa.

Acknowledgements. The research leading to these results has received funding from the European Union 7th Framework Programme (FP7/2007-2013) under Grant Agreement no. 603502 (EU project DACCIWA: Dynamics-aerosol-chemistry-cloud interactions in West Africa). This work has been supported by the African Monsoon Multidisciplinary Analysis (AMMA) project. Based on a French initiative, AMMA was built by an international scientific group and is currently funded by a large number of agencies, especially from France, UK, USA and various African countries. The authors wish to thank the SAFIRE (Service des Avions Francais Instruments pour la Recherche

en Environnement) for preparing and delivering the research aircrafts (ATR-42). We thank the Philippe Goloub and Didier Tanre for theirs

effort in establishing and maintaining AERONET sites in Djougou (Benin) and in Banizoumbou (Niger).

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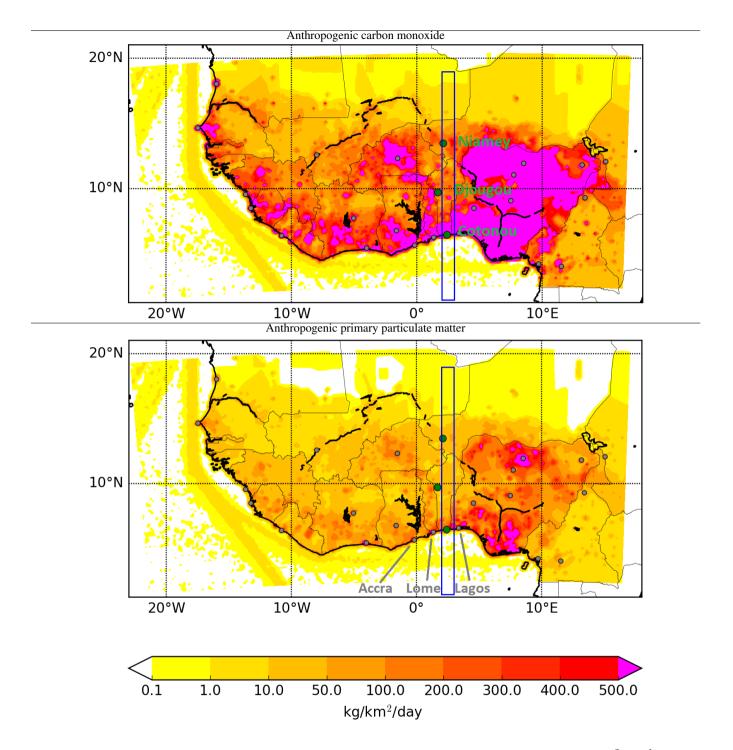
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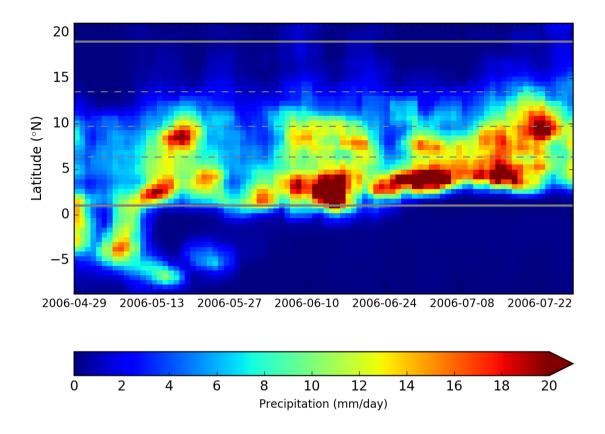
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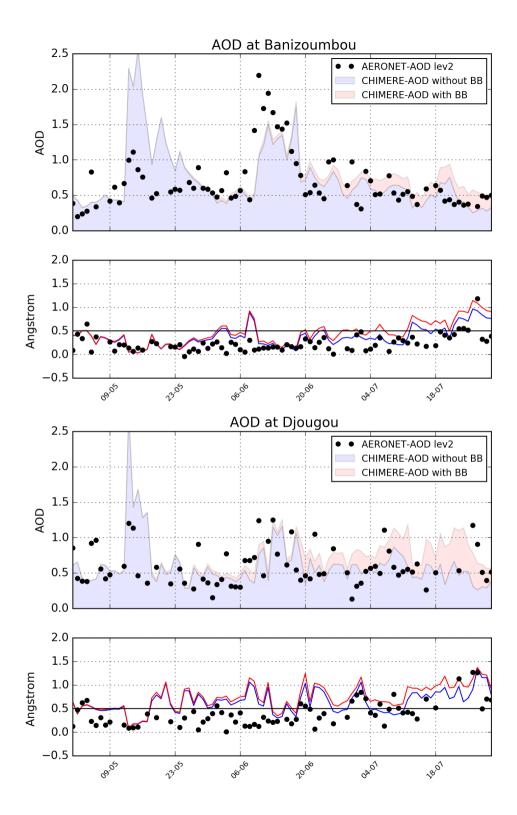
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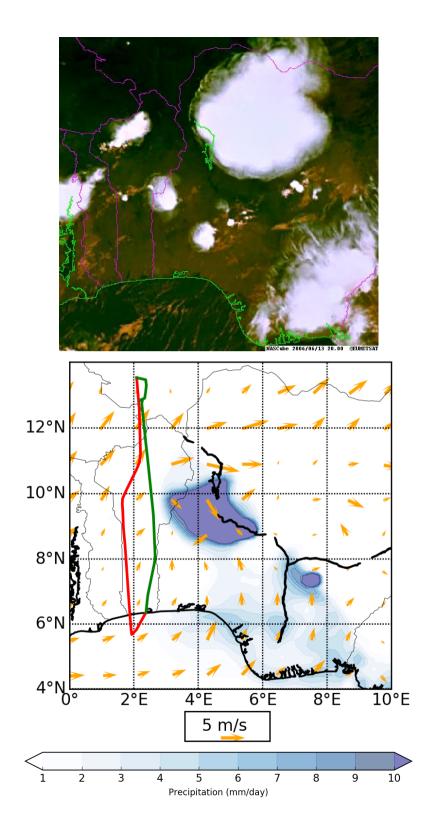
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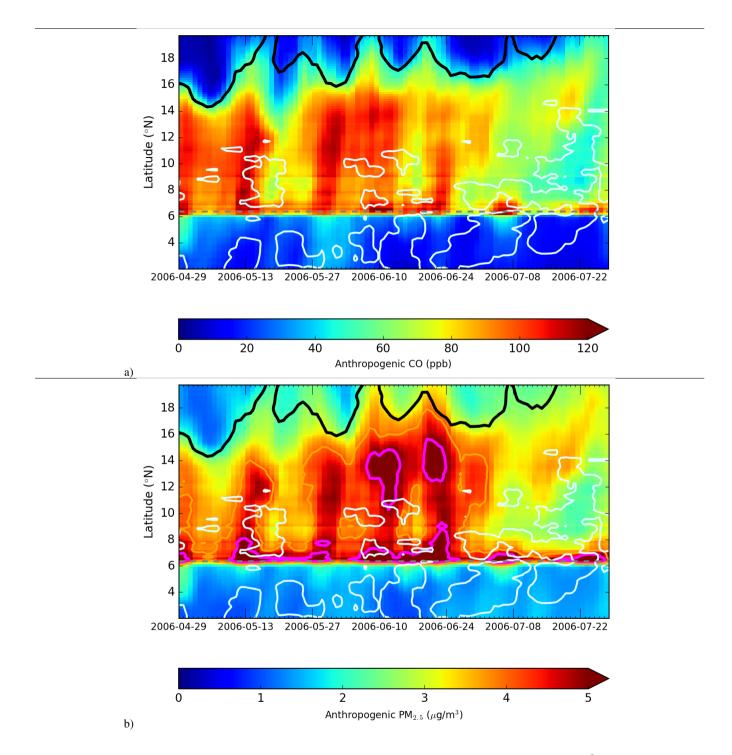
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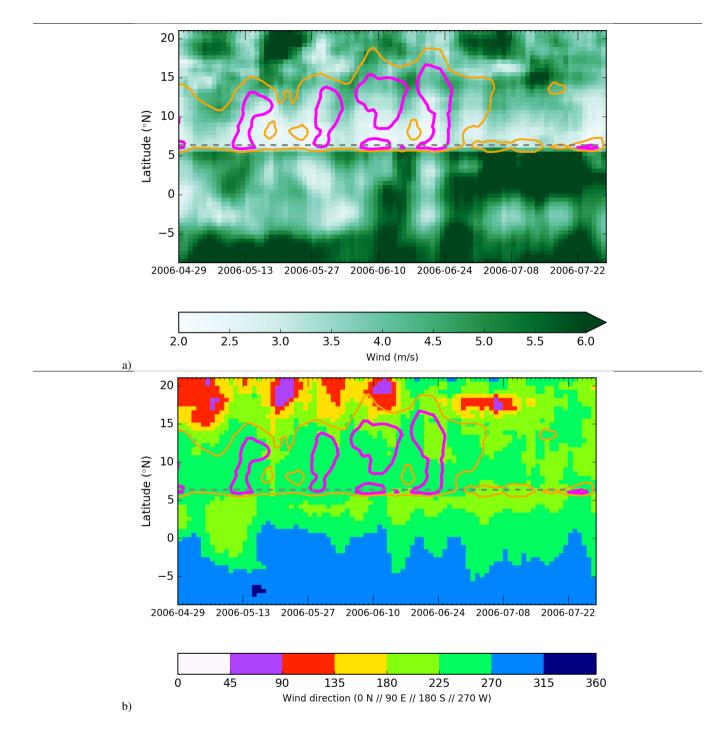
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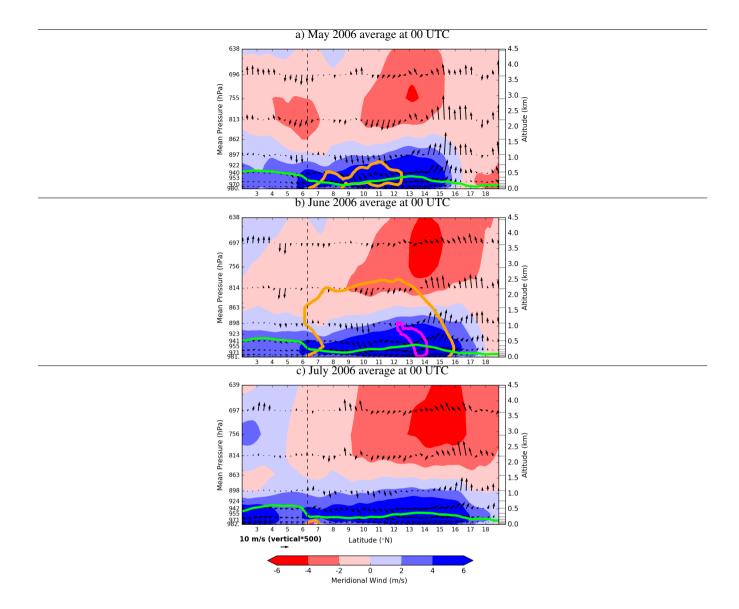
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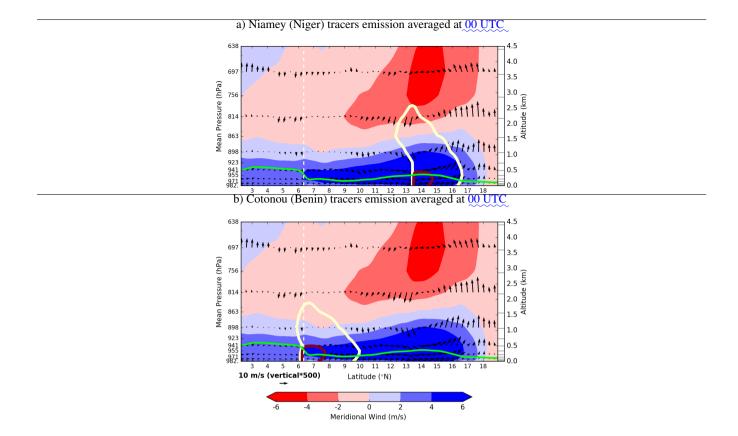
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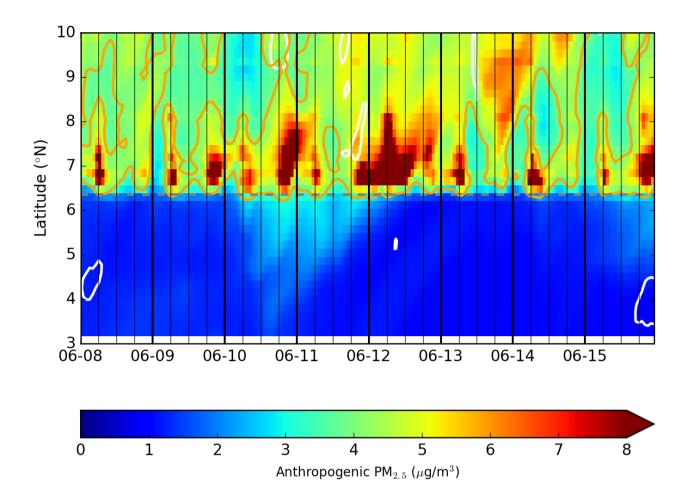
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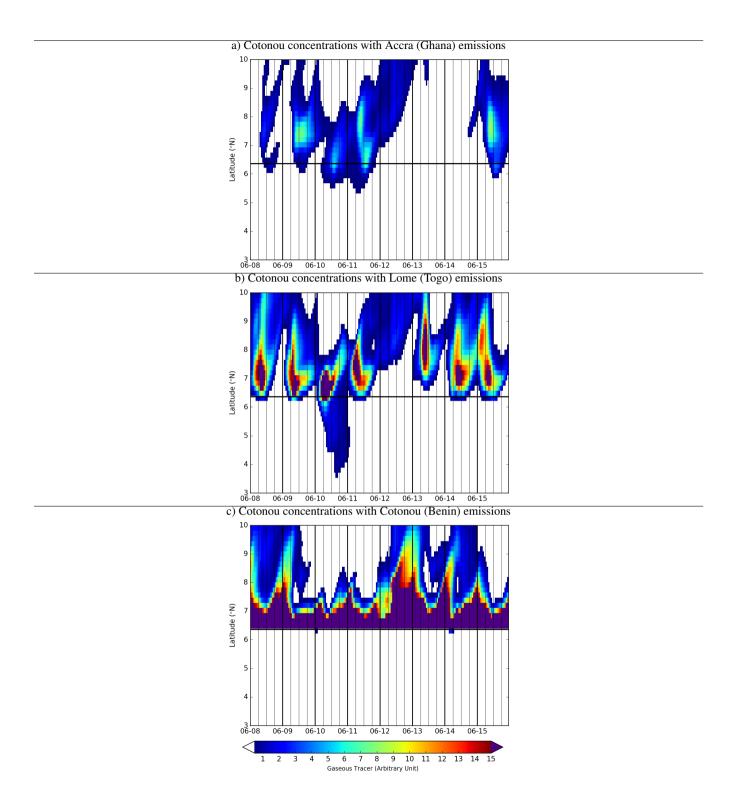
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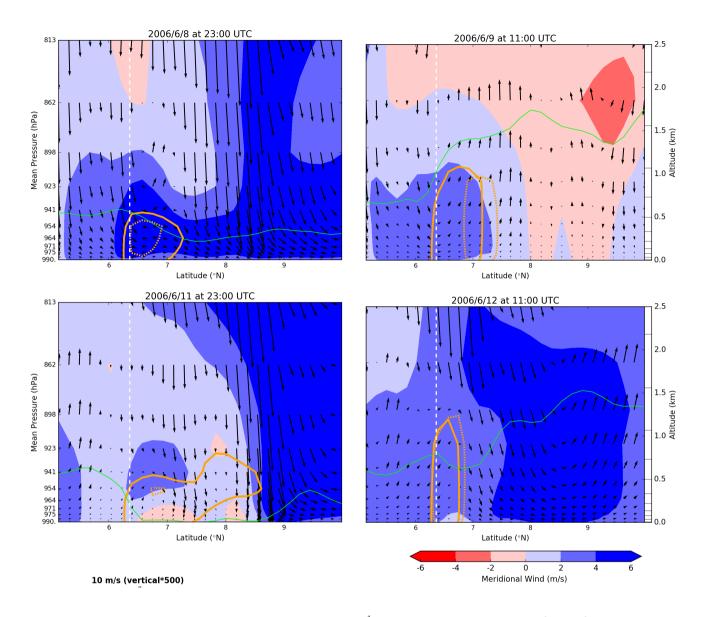
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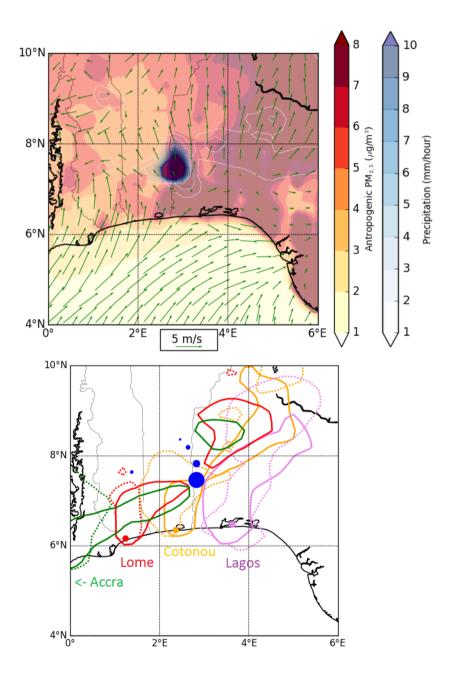
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Pollutants obs/mod	Coastal region	Sudano-Guinean region	Sudano-Sahelian region
i onutunto obb/mou	Coustai region	Suduno Guineun region	Suduno Sunchun region

Aircraft observations of 13 June 2006 from 10 UTC to 13 UTC											
	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max		
CO CHIMERE (ppb)	207.24	174.90	233.03	231.52	217.50	244.54	243.88	212.25	275.76		
CO Aircraft (ppb)	172.78	146.78	209.21	172.51	161.43	182.14	159.26	148.70	174.24		
$PM_{2.5}$ CHIMERE ( $\mu$ g.m <sup>-3</sup> )	23.95	19.26	26.01	28.96	25.46	33.87	55.15	35.18	79.93		
$PM_{2.5}$ Aircraft ( $\mu$ g.m <sup>-3</sup> )	15.67	7.57	33.02	49.72	22.33	77.88	55.81	34.22	72.20		

## Aircraft observations of 14 June 2006 from 13 UTC to 16 UTC

111													
	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max				
CO CHIMERE (ppb)	212.71	190.23	239.83	229.15	205.49	246.75	244.74	232.68	257.97				
CO Aircraft (ppb)	200.21	185.49	222.34	181.51	153.11	233.35	168.66	146.35	200.66				
$PM_{2.5}$ CHIMERE ( $\mu$ g.m <sup>-3</sup> )	42.79	28.23	64.33	82.25	64.43	92.93	84.01	79.65	91.46				
$PM_{2.5}$ Aircraft ( $\mu g.m^{-3}$ )	39.40	37.26	42.37	39.36	23.60	59.12	92.08	50.11	138.86				

**Table 1.** Range of 2-minute average modeled and observed concentrations of CO (ppb) and  $PM_{2.5}$  ( $\mu$ g.m<sup>-3</sup>) in the PBL (altitude lower than 1000 m) over three regions: Coastal region ( $6.3^{\circ}N - 9.0^{\circ}N$ ), Sudano-Guinean region ( $9.0^{\circ}N - 11.0^{\circ}N$ ), Sudano-Sahelian region ( $11.0^{\circ}N - 13.5^{\circ}N$ ).

СО	May-July		May		June		Ju	ly	
Cotonou (Benin)									
Average (ppb)	221.01		157.25		239.11		267.26		
Background (ppb and %)	73.30	33.17	73.58	46.79	72.82	30.45	73.48	27.50	
Anthropogenic (ppb and %)	64.75	29.30	67.83	43.14	64.65	27.04	61.77	23.11	
Fire Biomass burning (ppb and %)	82.96	37.54	15.84	10.07	101.64	42.51	132.00	49.39	
Djougou (Benin)									
Average (ppb)	226.78		180.28		240.65		259.85		
Background (ppb and %)	75.27	33.19	77.38	42.92	75.23	31.26	73.21	28.17	
Anthropogenic (ppb and %)	80.77	35.62	93.52	51.88	89.01	36.99	60.04	23.11	
Fire Biomass burning (ppb and %)	70.74	31.19	9.38	5.21	76.41	31.75	126.60	48.72	
		Niamey	(Niger)						
Average (ppb)	212.44		171.24		229.26		237.34		
Background (ppb and %)	78.95	37.16	82.72	48.30	78.69	34.32	75.42	31.78	
Anthropogenic (ppb and %)	82.54	38.85	83.92	49.01	98.24	42.85	65.96	27.79	
Fire Biomass burning (ppb and %)	50.95	23.98	4.60	2.69	52.33	22.82	95.97	40.43	

**Table 2.** *CO* (*ppb*) average and relative contributions (%) of each type of pollution source (background, anthropogenic, *firebiomass burning*) at Cotonou (Benin), Djougou (Benin) and Niamey (Niger). The time averaged periods correspond to each month of (May, June, July) and to the whole period (from May to July).

PM <sub>2.5</sub>	May	y-July May		ay	June		July		
Cotonou (Benin)									
Average ( $\mu$ g.m <sup>-3</sup> )	29.62		23.31		28.89		36.64		
Anthropogenic ( $\mu$ g.m <sup>-3</sup> and %)	3.45	11.64	3.44	14.75	3.44	11.90	3.47	9.46	
Fire Biomass burning ( $\mu$ g.m <sup>-3</sup> and %)	12.02	40.58	2.54	10.89	14.38	49.79	19.21	52.44	
Dust ( $\mu$ g.m <sup>-3</sup> and %)	4.35	14.68	8.89	38.15	2.34	8.10	1.74	4.76	
Biogenic ( $\mu$ g.m <sup>-3</sup> and %)	6.04	20.38	5.84	25.04	5.68	19.65	6.59	17.98	
Salt ( $\mu$ g.m <sup>-3</sup> and %)	3.77	12.72	2.60	11.17	3.05	10.56	5.62	15.35	
Djougou (Benin)									
Average ( $\mu$ g.m <sup>-3</sup> )	38.25		41.31		37.71		35.70		
Anthropogenic ( $\mu$ g.m <sup>-3</sup> and %)	4.10	10.72	4.38	10.60	4.59	12.16	3.35	9.38	
Fire Biomass burning ( $\mu$ g.m <sup>-3</sup> and %)	9.18	23.99	1.34	3.23	9.36	24.82	16.84	47.17	
Dust ( $\mu$ g.m <sup>-3</sup> and %)	13.55	35.43	23.73	57.44	12.05	31.95	4.83	13.52	
Biogenic ( $\mu$ g.m <sup>-3</sup> and %)	9.95	26.01	10.76	26.05	10.44	27.68	8.66	24.25	
Salt ( $\mu$ g.m <sup>-3</sup> and %)	1.47	3.85	1.10	2.67	1.28	3.39	2.03	5.68	
	Ν	Jiamey (N	liger)						
Average ( $\mu$ g.m <sup>-3</sup> )	53.76		71.59		52.56		37.09		
Anthropogenic ( $\mu$ g.m <sup>-3</sup> and %)	4.22	7.85	4.00	5.58	5.15	9.80	3.54	9.54	
Fire Biomass burning ( $\mu$ g.m <sup>-3</sup> and %)	5.73	10.66	0.56	0.78	5.47	10.41	11.15	30.06	
Dust ( $\mu$ g.m <sup>-3</sup> and %)	36.86	68.57	61.02	85.24	34.34	65.35	15.14	40.83	
Biogenic ( $\mu$ g.m <sup>-3</sup> and %)	6.31	11.75	5.58	7.79	7.08	13.48	6.31	17.01	
Salt ( $\mu$ g.m <sup>-3</sup> and %)	0.63	1.18	0.44	0.61	0.51	0.96	0.95	2.56	

**Table 3.**  $PM_{2.5}$  ( $\mu g.m^{-3}$ ) average and relative contributions (%) of type of pollution source (anthropogenic, <u>fireBiomass burning</u>, dust, biogenic, sea salt) at Cotonou (Benin), Djougou (Benin) and Niamey (Niger). The time averaged periods correspond to each month of (May, June July) and to the whole period (from May to July).