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Atmospheric Chemistry and Physics Discussions

# Remote biomass burning dominates southern West African air pollution during the monsoon

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**Abstract.** Vast quantities of agricultural land in southern and central Africa are burnt between June and September each year, which releases large concentrations of aerosols into the atmosphere. The resulting smoke plumes are carried west over the Atlantic Ocean at altitudes between 2 and 4 km. As only limited observational data in West Africa have existed until now, whether this pollution has an impact at lower altitudes has remained unclear. The Dynamics-Aerosol-Chemistry-Cloud Interactions in West Africa (DACCIWA) aircraft campaign took place in southern West Africa during June and July 2016, with the aim of observing gas and aerosol properties in the region in order to assess anthropogenic and other influences on the atmosphere.

Results presented here show that a significant mass of aged accumulation mode aerosol was present in the southern West African boundary layer, over both the ocean and the continent. A median dry aerosol concentration of 6.2 µg $m^{-3}$ (standard temperature and pressure (STP)) was observed over the Atlantic Ocean upwind of the major cities, with an interquartile range from 5.3 to 8.0 µg $m^{-3}$. This concentration increased to a median of 11.1 µg $m^{-3}$ (8.6 to 15.7 µg $m^{-3}$) in the immediate outflow from cities. In the continental air mass away from the cities, the median aerosol loading was 7.5 µg $m^{-3}$, with an interquartile range of 4.2 µg $m^{-3}$. The accumulation mode aerosol population over land displayed similar chemical properties to the upstream population, which implies that upstream aerosol is a



significant source of aerosol pollution over the continent. The upstream aerosol is found to have most likely originated from central and southern African biomass burning. This demonstrates that biomass burning plumes are being advected northwards, after being entrained into the monsoon layer over the eastern tropical Atlantic Ocean. It is shown observationally for the first time that they contribute up to 80% to the regional aerosol loading in the boundary layer of southern West Africa during the monsoon season.

As a result, the large and growing emissions from the coastal cities are overlaid on an already substantial aerosol background. On a regional scale this renders cloud properties and precipitation less sensitive to future increases in anthropogenic emissions. Such high background loadings will lead to greater pollution exposure for the large and growing population in southern West Africa. These results emphasise the importance of including aerosol from across country borders in the development of air pollution policies and interventions in regions such as West Africa.

## 1 Introduction

West Africa is currently undergoing rapid urbanisation, population growth and industrial development. As a result of these large socioeconomic changes, anthropogenic pollution in the region tripled between 1950 and 2000 (Lamarque et al., 2010), and is expected to do so again from 2005 to 2030 (Liousse et al., 2014). Nevertheless, West African air quality is among the most poorly studied worldwide. As a result, these changes are being imposed on a largely unknown regional background (Zuidema et al., 2016; Knippertz et al., 2015).

Plumes of biomass burning pollution from further afield are known to impact the mid-troposphere above West Africa during the summer monsoon (Chatfield et al., 1998; Mari et al., 2008). These plumes are the result of vast quantities of agricultural land in southern and central Africa being burnt between June and September each year (Barbosa et al., 1999). The large-scale burning releases high concentrations of aerosols into the atmosphere, which are carried west over the Atlantic Ocean at altitudes between 2 and 4 km. This transport mechanism is reliant on the southern hemispheric African Easterly Jet; when the jet is active, vast intrusions of biomass burning pollution can be transported across the Atlantic Ocean, in some cases as far west as South America (Mari et al., 2008). A second pathway over land may also exist (Menut et al., 2018). Intrusions into southern West Africa have been well documented from in-situ and satellite data. To date, this phenomenon has been thought to be confined predominantly to layers between 2 and 4 km in altitude (Barbosa et al., 1999; Capes et al., 2008; Chatfield et al., 1998; Mari et al., 2008). Suggestions that the pollution may mix further down into the planetary boundary layer (e.g. Menut et al., 2018) have remained unconfirmed due to limited in-situ observations. Recent attempts to quantify the extent to which smoke over the Atlantic Ocean entrains into the boundary layer using model simulations have shown that different models can provide very different results (Das et al., 2018; Peers et al., 2016), which makes the collection and analysis of observational evidence on this matter particularly important.

Both near-field and remote sources of pollution are likely to have an effect on cloud properties, radiative forcing and human health in southern West Africa. During the onset of the West African Monsoon, aerosol becomes entrained



into newly-forming banks of monsoon clouds, so could have a resultant effect on rainfall patterns as well as the region's response to climate change. The emergence of megacities along the southern coast means that large numbers of people will be exposed to any atmospheric pollutants that exist in the region.

Airborne measurements made during the Dynamics-Aerosol-Chemistry-Cloud Interactions in West Africa (DACCIWA) campaign (Knippertz et al., 2015; Flamant et al., 2018) in June - July 2016 provided the opportunity to map aerosol properties southern West Africa extensively. Here, observations from the three aircraft employed during the campaign are used to examine the relative contributions of local and transported pollution towards the regional boundary layer (<1.9 km) aerosol loading in southern West Africa. The relative contributions of regional urban emissions and aged biomass burning aerosol from central and southern Africa towards this background are assessed. Biomass burning aerosol advected inland from remote sources is found to be the key driver of particulate pollution in the boundary layer across southern West Africa away from large urban centres.

## 2 Method

### 2.1 Airborne observations

The DACCIWA aircraft campaign took place during June and July 2016 and focused on the highly populated southern coastal region of West Africa. Science flying began on 29th June and concluded on 16th July 2016. Three aircraft took part in the campaign: the German Deutsches Zentrum für Luft- und Raumfahrt (DLR) Falcon 20, the French Service des Avions Français Instrumentés pour la Recherche en Environnement (SAFIRE) ATR-42 and the British Antarctic Survey (BAS) Twin Otter. All three aircraft were based at the military airport in Lomé, Togo (6.16°N, 1.25°E), though the ATR-42 flew to the Aéroport Félix Houphouët-Boigny in Abidjan, Côte D'Ivoire, twice, on the 6th and 11th July. In total, 50 scientific flights were carried out, which comprised 155 hours of scientific sorties. The DLR Falcon completed 12 scientific missions during the campaign, the ATR-42 completed 20 and the Twin Otter 18 (Flamant et al., 2018). The aircraft campaign took place after the monsoon onset and was characterised by a northward-shifted intertropical discontinuity, which resulted in unusually dry conditions for this time of year. This period has been described as Phase 2 of the 2016 West African Monsoon (Knippertz et al., 2017).

Several flights were conducted between Lomé and the air above a ground station in Savè, Benin (8.03 ºN, 2.48 ºE), around 250 km to the north-east, which were used to build up statistics on background aerosol concentrations and cloud-aerosol interactions. Other flight patterns included city emission flights, which targeted city plumes and flights over the sea. Flight paths for all three aircraft are shown in Fig. 1.

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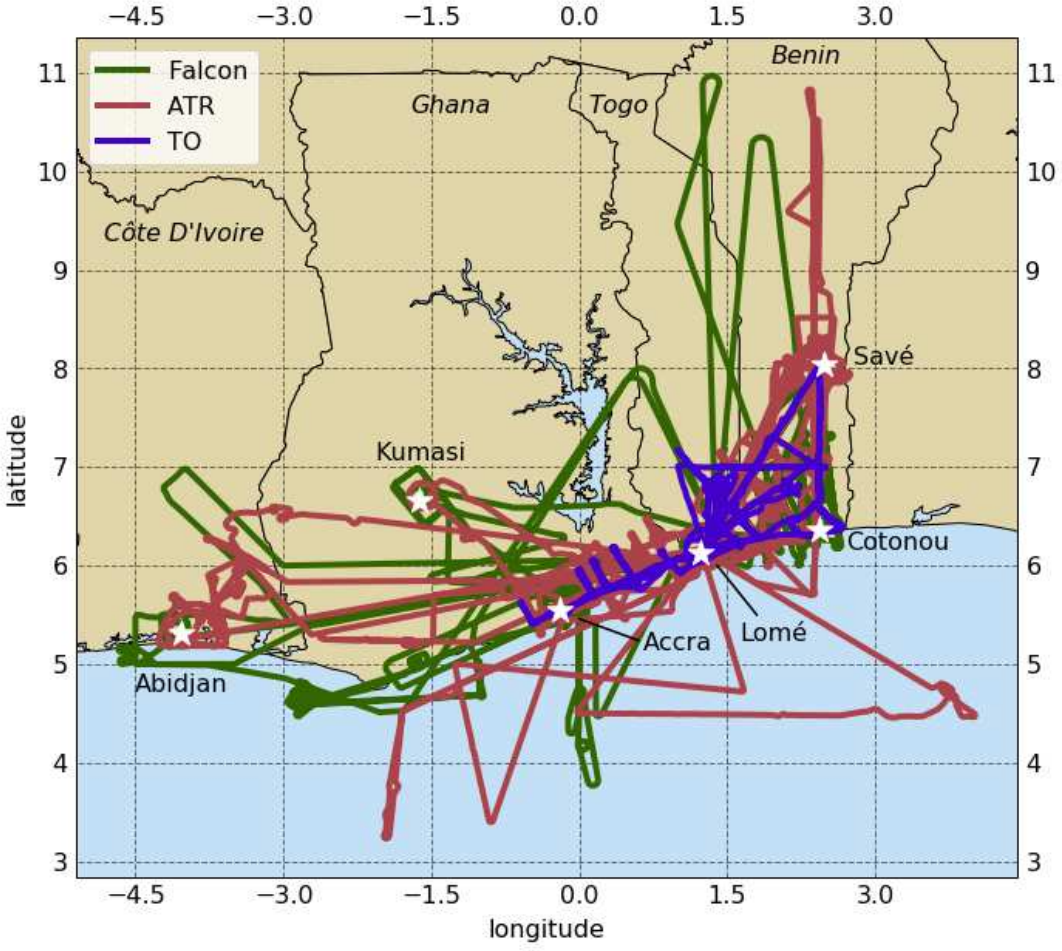


**Figure 1: Map showing the flight paths of the Falcon, ATR and Twin Otter aircraft during the DACCIWA campaign.**

Submicron aerosol chemical composition was measured using Aerodyne compact Time-of-Flight Aerosol Mass Spectrometers (AMS) (Drewnick et al., 2005; Canagaratna et al., 2007), mounted onboard all three aircraft. These produced mass spectra of the aerosol chemical composition with a time resolution of 20-45 seconds. A fragmentation table (Allan et al., 2004) was used to distinguish different compounds, yielding measurements of sulphate ($SO_4$), nitrate ($NO_3$), ammonium ($NH_4$) and organic compounds. An average collection efficiency of 0.5 was used throughout the campaign, which is standard for ambient measurements in similar environments (Middlebrook et al., 2012). The ionisation efficiencies of the instruments were calibrated several times througout the campaign using size-selected ammonium nitrate aerosol. A more detailed description of the ATR AMS data processing can be found in Brito et al. (2018).



Submicron aerosol size distributions were measured using a TSI Scanning Mobility Particle Sizer (SMPS) on board the ATR aircraft. This produced a size distribution of aerosol between 0.02 and 0.5 µm every 90 s. Condensation nuclei number concentrations were measured using Condensation Particle Counters (CPC) on board each of the three aircraft (a Brechtel Mixing CPC on the Twin Otter, modified TSI 3010 on the Falcon and TSI 3788 CPC on the ATR; lower size limits were 3 nm, 14 nm and 3 nm respectively).

In order to integrate these datasets successfully, the sensitivities of the instruments on all three platforms were compared. The transect between the coastal city of Lomé in Togo and the inland city of Savè in Benin was flown several times by each aircraft, which provided a basis for performing statistical comparisons. The median measurements of the AMS instruments on the ATR and the Twin Otter aircraft were within 20% of one another, although a larger interquartile range was observed in measurements from the ATR. The CPCs on board all three instruments showed a discrepancy in the median values of around 40%. Where applicable, measurements were corrected to standard temperature and pressure (STP).

The AMS data were compared for the take-offs and landings at Lomé airport. Despite calibration efforts, the AMS on the Falcon measured lower mass concentrations than the other two. This is believed to have been caused by a loss process at its inlet that affected the absolute, but not the relative measured mass concentrations of the different compounds. Therefore, only the proportional chemical distribution from the Falcon AMS is used here. It is indicated in the text where these data are included.

The West African Monsoon governs surface level wind patterns in southern West Africa during June and July. Southerlies associated with the monsoon bring surface air into West Africa that has been advected over the ocean for several thousand kilometers (Williams et al., 2007). The incoming air is then affected by large coastal cities before continuing inland. In order to study the influence of different sources, surface level aerosol was analysed in three regimes: ‘upwind marine’, ‘continental background’ and ‘urban outflow’. The first two include data collected above the ocean and over West Africa away from immediate urban sources, respectively. This distinction provides a direct comparison between upwind air entering the region from the south and that influenced by the coastal cities, Abidjan (Côte D’Ivoire), Accra (Ghana), Lomé (Togo) or Cotonou (Benin). The ‘urban outflow’ regime includes data from the centre of near-field urban plumes.

In all three cases, only aerosol below 1.9 km was considered. This comprises the boundary layer over land (Kalthoff et al., 2017) and the monsoon layer over the ocean. The monsoon layer here is defined to include air below 1.9 km, which is part of the southerly monsoon flow and feeds the continental boundary layer. When the air reaches land, a much deeper mixing results in the low boundary layer over ocean mixing with the air just above it to determine the concentration further inland. In the ‘continental background’ and ‘urban outflow’ regimes, data from below 100 m were removed to avoid bias, as the aircraft only flew at this altitude over land in the vicinity of the airport; the airport’s influence was found to be negligible above this altitude. ‘Urban outflow’ data were from the centre of the



near-field (<60 km) urban plumes emitted from the cities listed above. These data were confined to include only measurements where $NO_x$ levels were within the highest 5% measured during the campaign (3.2 ppbv).

### 2.2 Regional Modelling

The regional-scale model system COSMO-ART (Consortium for Small-scale Modelling – Aerosol Reactive Trace gases) was used to investigate the impacts of biomass burning on cloud microphysical properties. The model is based on the German Weather Service (DWD)'s operational weather forecast model COSMO (Baldauf et al., 2011), coupled with an aerosol model (ART) for online treatment of aerosol chemistry and dynamics (Vogel et al., 2009; Bangert et al., 2012; Athanasopoulou et al., 2014; Knote et al., 2011). This allowed feedbacks to be calculated between aerosols and radiation as well as between aerosols and clouds. For the model runs, EDGAR (Emission Database for Global Atmospheric Research) emission data were used for the anthropogenic emission of gases and aerosols. Natural emissions of biogenic volatile organic compounds, sea salt (Lundgren et al., 2013), mineral dust (Stanelle et al., 2010) and GFAS (Global Fire Assimilation System) emissions from vegetation fires (Kaiser et al., 2012; Walter et al., 2016) are calculated for each model time step. There was a spin-up time of 7 days (29 June to 5 July) and results were observed for 24 hours on 6 July.

Two simulations were performed for this study: one with biomass burning emissions included and the other without. The simulations were performed over a large domain covering West Africa and the south eastern Atlantic Ocean using a horizontal domain (D1) with a grid size of 5 km and 50 vertical layers. The output from D1 was used to supply boundary conditions for a smaller, nested domain (D2) covering southern West Africa (the DACCIWA region), with a resoluation of 2.5 km and 80 vertical levels (see fig. 2).

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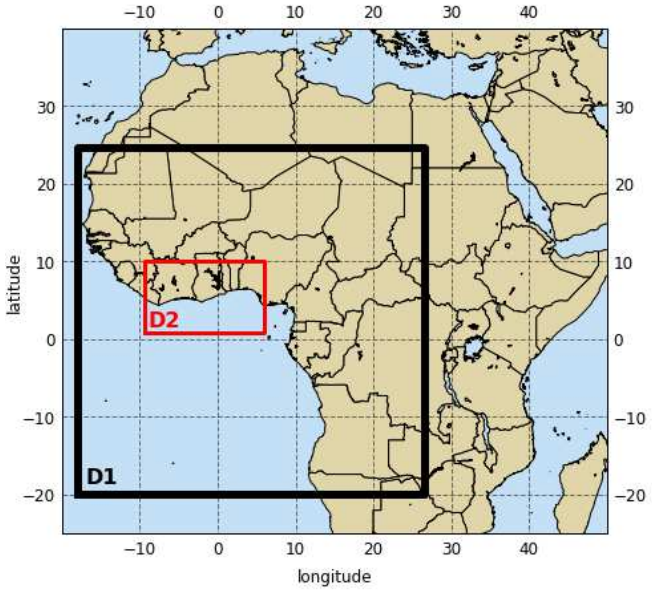


**Figure 2: The two nested domains used by the COSMO-ART model.**

## 3 Results and discussion

### 3.1 Observations

Figure 3 shows the aerosol particle number size distribution observed in each of the three regimes considered here. Significant variation can be seen in the number of smaller, Aitken mode particles. These particles are emitted from urban centres or formed from precursor gases and grow quickly in the atmosphere; large Aitken mode populations therefore indicate the presence of significant local sources. In the urban outflow regime, the Aitken mode concentration was often high, with a median number concentration of 3,400 $cm^{-3}$. In contrast, the Aitken mode was barely present in upwind marine air (median of 130 $cm^{-3}$). The number concentration of accumulation mode particles, with an average diameter near 200 nm, however, was remarkably consistent across the three regimes: 80% of the data lay within ± 30% of the campaign median in all cases. The median accumulation mode concentration was 600 $cm^{-3}$ STP in the upwind marine air and 850 $cm^{-3}$ STP in the continental background.

In the lower atmosphere during June and July, wind speeds in southern West Africa are generally low and wind comes from the south, becoming south-westerly as it approaches the coast. Therefore, within the boundary layer, cool, moist Atlantic air progresses towards the cities and is likely to carry their plumes inland (Knippertz et al., 2017). Nevertheless, the similarity between the accumulation mode concentration in the upwind marine and continental background regimes seen here suggests that the air mass already contained a large number of the accumulation mode particles prior to urban influence. Comparing the median accumulation mode number

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concentrations in the continental background regime (850 $cm^{-3}$ STP) and the upstream marine regime (600 $cm^{-3}$ STP) suggests that far from the source, city emissions and land-based biogenic sources contributed only an extra 40% on top of the incoming accumulation mode aerosol. This calculation assumes a constant influence across the region from incoming aerosol, and so likely represents a lower limit. Nevertheless, this implies that incoming pollution from the Atlantic has a considerable influence on the aerosol population over the land.

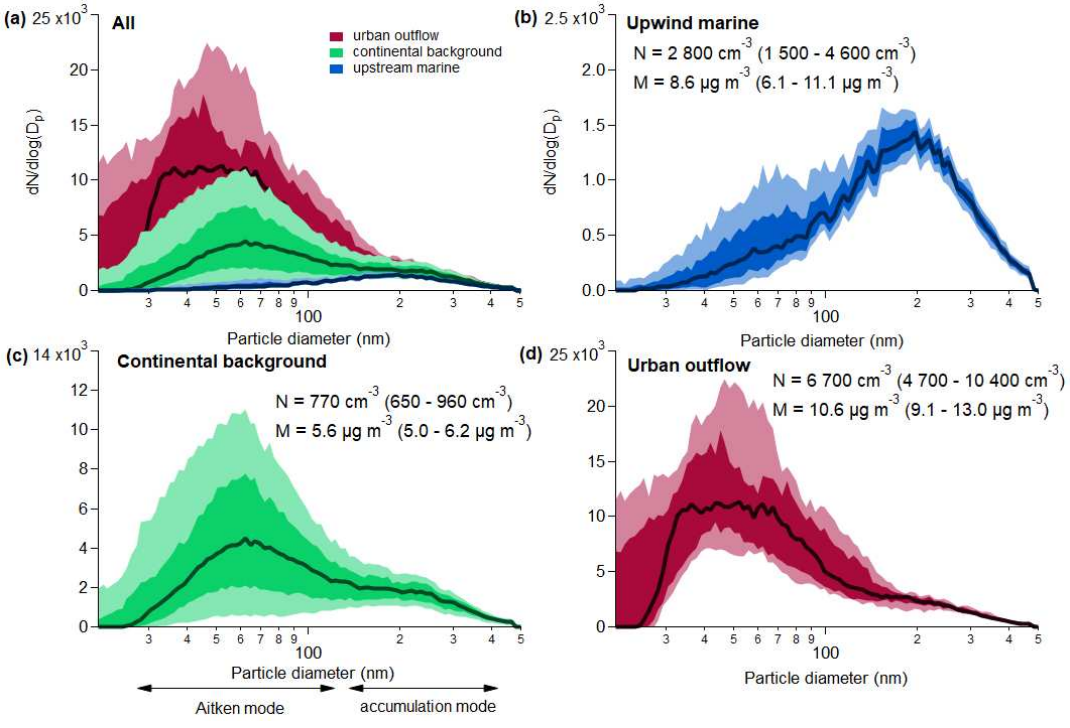


**Figure 3: Size distributions of aerosol in the urban outflow, continental background and upstream marine regimes, measured by the SMPS on board the ATR aircraft. For each regime, the median size distribution is shown by the dark line; the dark shading contains 50% of the data; and the light shading contains 80% of the data. The comparison of all three plots in panel (a) shows a stable accumulation mode that exists in all three regimes, centred at around 200 nm, while the smaller Aitken mode is much more variable. In panels (b-d), N shows the median total number concentration summed across the whole distribution, with the lower and upper quartiles shown in brackets; M shows the calculated aerosol mass, assuming an aerosol density of 1.3 g $cm^{-3}$, with the interquartile range again shown in brackets. The Aitken and accumulation modes are labelled in panel (c).**

The chemical composition of aerosols observed during DACCIWA supports the suggestion that much of the aerosol in the region originates upwind of the cities. Figure 4a shows aerosol mass and number concentrations in the three regimes, with mass classified by chemical species. The median total aerosol concentrations observed were 6.2, 11.1 and 7.5 µg $m^{-3}$ in the upwind marine, urban outflow and background continental regime, respectively, with interquartile ranges of 2.8, 7.1 and 4.2 µg $m^{-3}$. Although there was some day-to-day variability, which can be seen in the interquartile ranges shown in Fig. 3, there was no statistically significant variation in the median throughout the diurnal cycle (the variation in the median throughout the day was < 1 µg $m^{-3}$). Very little variation between the three

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regimes is seen in the proportional contribution of the different chemical species. The largest contribution to the measured $PM_1$ (particulate matter with a diameter smaller than 1 µm) was organic aerosol, which accounted for approximately 60% of the aerosol mass in all three regimes. Sulphate accounted for approximately 25% and nitrate was generally low, comprising 4-6%. Ammonium contributed around 11%. The largest aerosol mass loadings and number concentrations were observed in the urban outflow, although even here, accumulation mode aerosol present in incoming air could account for as much as 50% of the total mass. Figure 4b shows the proportional chemical distribution for the three regimes explored here alongside those for the elevated biomass burning aerosol layer commonly sampled at 2-4 km from the ATR and Falcon, and the free troposphere above 5 km from the Falcon.

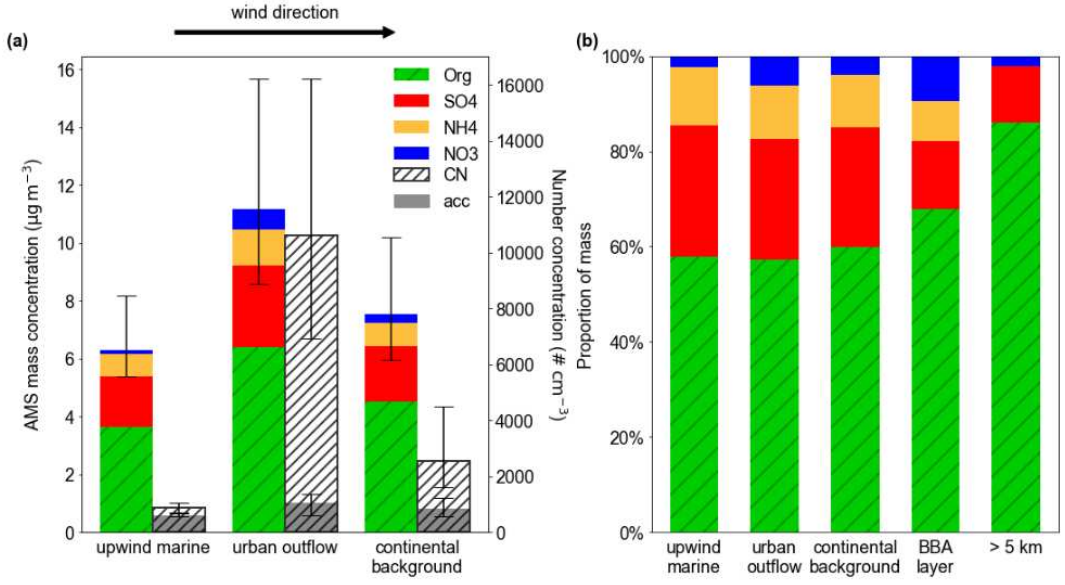


**Figure 4: (a) The chemical composition and condensation nucleus (CN) concentration in each of the three regimes. Coloured bars indicate aerosol mass concentration measured by the AMS. The CN bars show the total aerosol number concentration in each location (measured by the CPC), with shaded regions indicating the number of aerosol particles in the accumulation mode (derived from SMPS data shown in Fig. 3). Bars indicate the median, with error bars showing the interquartile range of observations (for total aerosol in the case of the AMS). A similar chemical distribution can be seen in each of the three regimes. (b) A comparison of the aerosol chemical distribution in all three regimes, alongside observations from the biomass burning layer at 3-4 km altitude (ATR and Falcon) and the free troposphere (> 5 km; Falcon) for comparison.**

The proportion of organic aerosol in the boundary layer was large compared with that seen in some other locations dominated by a mix of urban or biogenic emissions: Zhang et al. (2011) found that the global average organic fraction measured by the AMS is between 43% in remote locations and 52% downwind from urban centres. The contribution of nitrate here, in contrast, was lower than is typically seen in locations influenced by urban outflow. Zhang et al. (2011) found a global average contribution of 12% and 18% in downwind and urban locations, respectively, which is four times and triple what was observed here.

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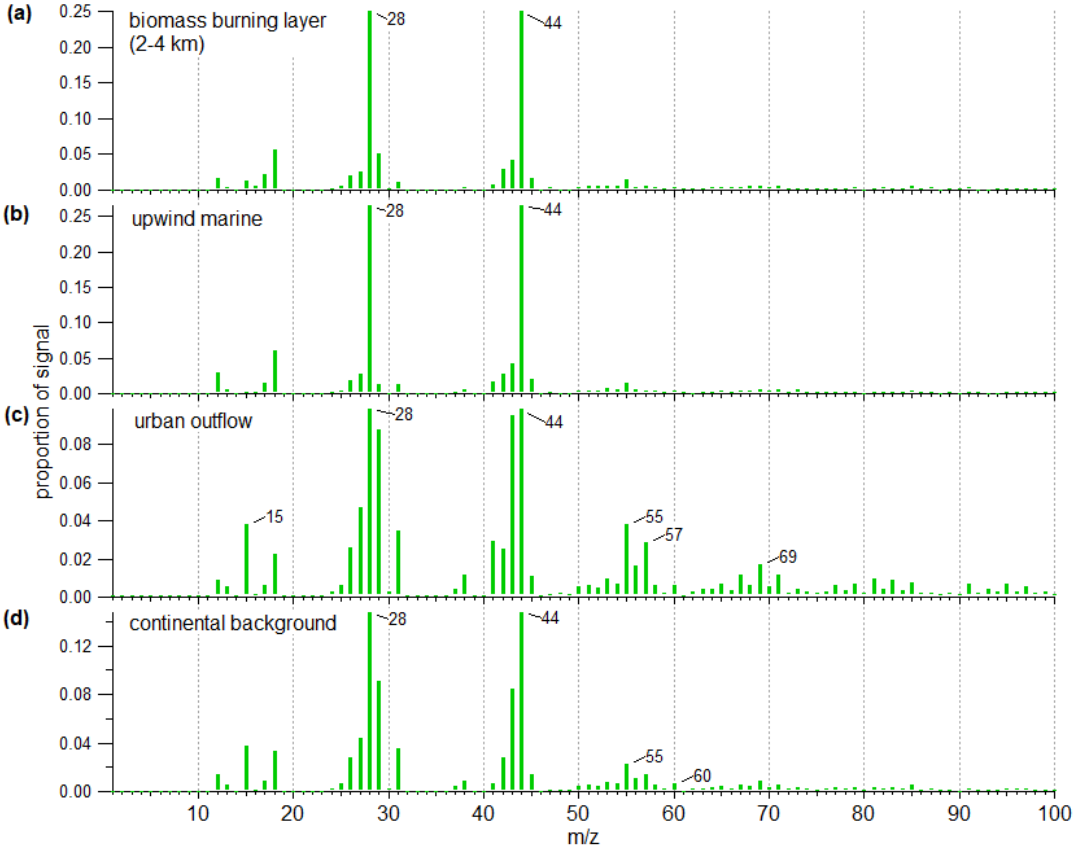


**Figure 5: Average organic aerosol mass spectra in (a) the 2-4 km biomass burning layer (from the Falcon AMS), and (b-d) each of the three regimes (ATR and Twin Otter). The influence of fresh urban emissions can be seen in the urban outflow regime and, to a lesser extent, in the continental background, demonstrated by the higher proportion of *m/z* 55 and 57, alongside larger hydrocarbon clusters. Fresh biomass burning is indicated by the peak at *m/z* 60. However, the dominant contribution in all cases is from *m/z* 28 and 44, both indicators of aged, oxidised organic aerosol.**

The organic mass spectra from the AMS on board the Twin Otter and the ATR for the three regimes are shown in Fig. 5, alongside the mass spectrum of the biomass burning layer at 2-4 km from the Falcon AMS, which is widely considered to have originated from central Africa (Flamant et al., 2018). All four spectra are dominated by aged aerosol, which is characterised by strong peaks at *m/z* 28 and 44 (Ng et al., 2010). Although the 2-4 km biomass burning layer showed a chemical distribution containing more organics and nitrate and less sulphate than aerosol observed lower in the atmosphere (Fig. 5b), the organic mass spectra shown here for this layer is very similar to that of the upstream marine aerosol, with *m/z* 28 and *m/z* 44 comprising 50% of the total organic mass in both cases. The urban outflow and, to a lesser extent, continental background mass spectra show features characteristic of urban pollution, including peaks at *m/z* 42, 55 and 91, which are associated with internal combustion engines (Ng et al., 2010), and a number of clustered hydrocarbon peaks, for example at *m/z* 65, 67 and 69 or *m/z* 79, 81 and 83. A peak can be seen in the urban outflow and continental background regimes at *m/z* 60, which is often associated with

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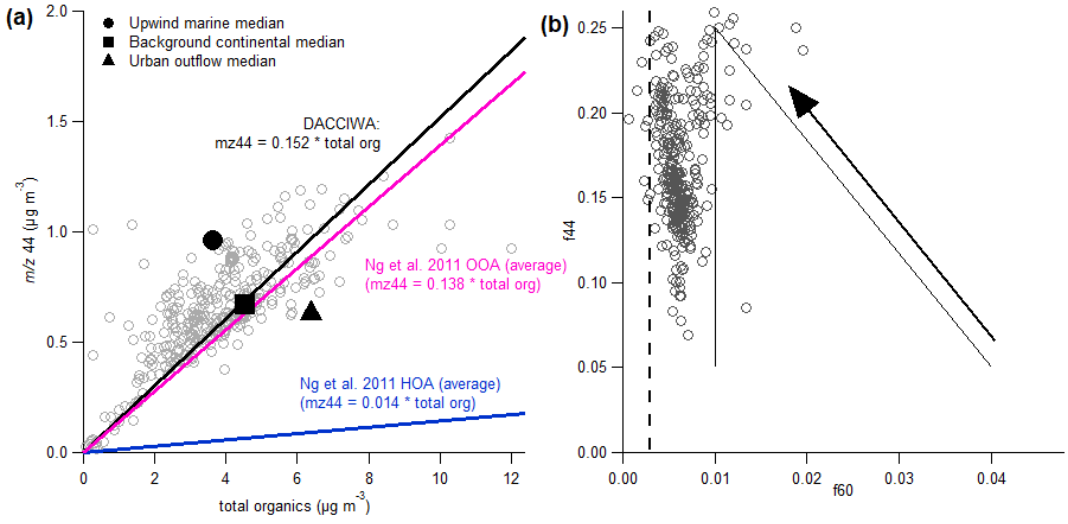
220 levoglucosan and other anhydrous sugars from biomass burning (Cubison et al., 2011) and likely arises from the widespread use of individual stoves for cooking, both in cities and in rural areas. This peak is associated only with fresh biomass burning; due to the oxidisation of anhydrous sugars in the atmosphere (Henningan et al., 2011), it would no longer be strongly visible in the spectrum after a few days of processing (Cubsion et al., 2011). These features together indicate that local urban and fresh biomass burning sources do contribute to the aerosol mass
225 loading in the boundary layer of southern West Africa. However, there appears to be a further, significant, and considerably more aged source, which is entering the region from the south and has resulted in all four mass spectra being dominated by the oxidised peaks *m/z* 28 and *m/z* 44.




230 **Figure 6: (a) Values of *m/z* 44, a strong indicator of aged organic aerosol, plotted against the total organic aerosol mass. The pink and blue lines show the average contribution of *m/z* 44 to the total for oxidised organic aerosol (OOA) and hydrocarbon-like organic aerosol (HOA). Grey circular markers show datapoints from the DACCIWA campaign. Median observations from each of the three regimes are shown as black shapes. (b) The fractional contribution of *m/z* 44 (f44) vs *m/z* 60 (f60). Fresh biomass burning aerosol is generally located within the triangle shown by the two black lines (Cubison et al., 2011) , with fresher aerosol lower in the triangle. While *m/z* 60 aerosol is present here, the low**
235 **contributions suggest that fresh biomass burning is not a dominant contributor towards the total organic aerosol mass.**

The dominance of aged aerosol in the overall population can be demonstrated further by comparing the magnitude of the *m/z* 44 peak with the total organic mass. Five-minute averaged datapoints from the Twin Otter aircraft are shown as markers in Fig. 6a. This dataset shown includes the continental background and urban outflow regimes; the median observation for each regime, including upwind marine, is also displayed. The *m/z* 44 contribution here
240 correlates well with the total organic mass, with *m/z* 44 contributing around 15% throughout the campaign.

The relationship between *m/z* 44 and the total organic aerosol mass can provide some insight into the source of ambient aerosol. The pink and blue lines in Fig. 6a show the averages across several campaigns from different parts of the globe for two different factors derived using positive matrix factorisation (PMF), as compiled by Ng et al.



(2011). PMF is a technique used to analyse the contributions of different aerosol sources to an AMS dataset and identify a mass spectrum associated with each source based on its variation in time. Two factors commonly identified by PMF include oxidised organic aerosol (OOA) and hydrocarbon-like organic aerosol (HOA). The highly oxidised OOA factors are generally associated with photochemically aged aerosol, with *m/z* 44 contributing a significant fraction of the total organic aerosol mass, as is shown by the higher gradient of the pink OOA line in Fig. 6a. The HOA fractions are often seen in pure fresh urban emissions. The contribution of *m/z* to the total organic aerosol is significantly lower in these cases, with the *m/z* 44 peak typically contributing less than 2% of the mass. This can be seen in the shallow gradient of the blue HOA line in Fig. 6a. In urban environments, mass spectra would be expected to have a large HOA component and thus, a large amount of scatter would be expected in the data, with the majority lying between the OOA and HOA lines. Here, the data are scattered predominantly around the OOA line, which suggests that the urban contribution is not the dominant factor in this dataset. The most significant proportion of the aerosol measured during the campaign is from aged, oxidised organic aerosol.

The presence of a peak at *m/z* 60 in the continental background air suggests that local biomass burning is present in the observed air mass. This is explored in more detail in Fig. 6b. It has been shown previously that fresh biomass burning contains a large fraction of *m/z* 60 (*f*60), a fragment of levoglucosan, which decreases as the plume ages. Furthermore, as the plume becomes more oxidised, the fraction of *m/z* 44 (*f*44) increases. Thus, fresh biomass burning emissions populate the bottom of the triangle shown in Fig. 6b, and move towards the top corner in the direction shown by the arrow as they age (Cubison et al., 2011). The dashed line to the left shows the expected baseline values for air not containing any fresh biomass burning. Here, *f*60 is consistently slightly higher than the baseline, suggesting the presence of some fresh biomass burning. However, *f*60 is not high enough at any time to suggest that fresh biomass burning is the dominant source of aerosol. The values of *f*44 are generally high, which again shows the prevalence of aged aerosol in the air mass.

### 3.2 Southern and central African biomass burning

The lack of variability in the accumulation mode concentration and composition is evidence that much of the aerosol observed during the DACCIWA campaign originated from a similar type of source. Furthermore, the similarity between these characteristics across the three regimes, including the upwind marine, identifies the dominant source to be outside the urban coastal region, upwind of all three locations. A closer inspection of the organic aerosol mass spectra during DACCIWA suggests the presence of a large mass of aged aerosol, with smaller contributions from fresh urban and fresh biomass burning sources. This evidence therefore shows that a large proportion of the aerosol mass in the continental West African boundary layer originates from the lower atmosphere over the eastern tropical Atlantic Ocean, and is present prior to influence from coastal cities.

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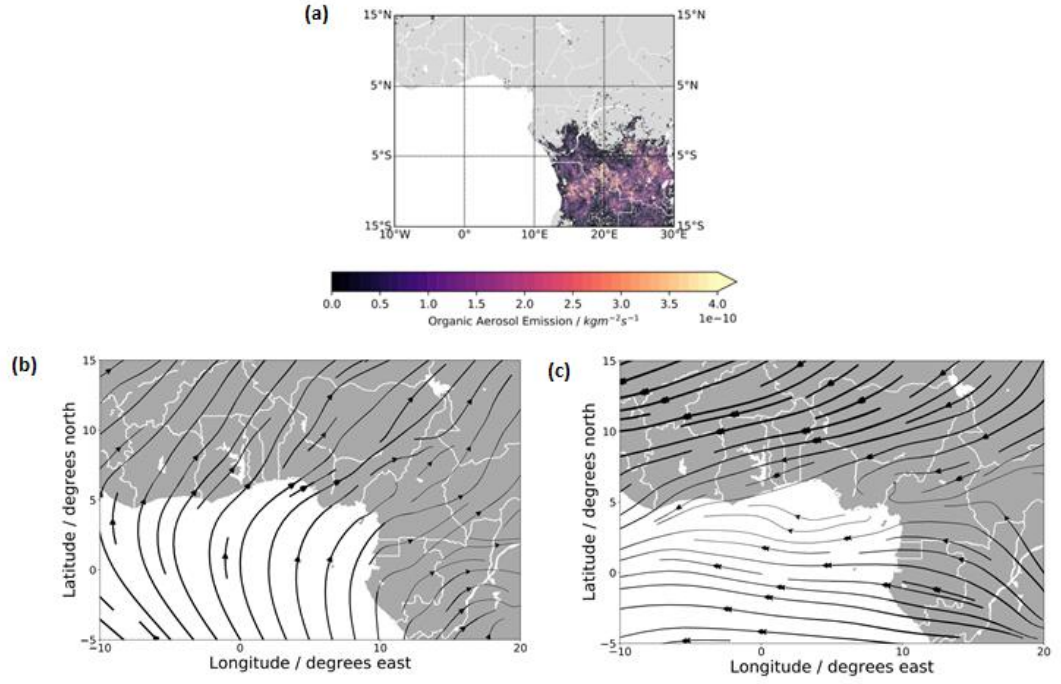


**Figure 7: (a) The location and intensity of organic aerosol emissions from biomass burning in central Africa during the DACCIWA campaign (June-July 2016) from the GFAS inventory (Kaiser et al., 2012) using biomass burning emissions data from EDGAR. (b, c) Wind stream functions at (a) the surface level, and (b) 750 hPa (approx. 2.5 km) from the NASA Global Modelling and Data Assimilation Office's GEOS-5 analysis. The line thickness indicates wind speed.**

One of the most significant fine-mode aerosol sources south of the coastal cities is the agricultural and savannah burning that takes place annually in central and southern Africa between June and September (Barbosa et al., 1999). Vast quantities of biomass burning aerosol are injected into the mid troposphere between 3 and 4.5 km as a result of these fires (Labonne et al., 2007) and carried west over the Atlantic Ocean by tropospheric winds at around 700 hPa (Das et al., 2017; Edwards et al., 2006). The location and intensity of these fires during the DACCIWA aircraft campaign is shown in Fig. 7, alongside modelled wind streams at ground level and at 750 hPa. At 750 hPa, the easterly currents that carry biomass burning pollution west out of central Africa can be clearly seen, while the surface level chart shows the southerly air stream that passes from the Atlantic Ocean into the southern West African boundary layer.

The observations of the aerosol composition in the continental boundary layer during the DACCIWA campaign described above show it to be characteristic of aged biomass burning aerosol. The proportion of organic aerosol was higher and nitrate was lower than would be expected in areas influenced primarily by urban outflow (Zhang et al., 2011). The mass spectra of organic aerosol below 1.9 km were dominated by peaks typical of aged, low-volatility aerosol, which closely resembled the mass spectrum of the 2-4 km biomass burning layer. Even in the urban outflow



regime, mass spectral features associated with near-field urban sources such as internal combustion engines were less prominent than would be expected from pure urban aerosol. This evidence supports the assertion that these central and southern African fires are the primary source of accumulation mode aerosol in southern West Africa during the summer monsoon season.

Recent observations carried out on Ascension Island to the south-west of the DACCIWA region (7.93 °S, 14.42 °W) as part of the US Department of Energy Atmospheric Radiation Measurement Layered Atlantic Smoke Interactions with Clouds (LASIC) campaign show that smoke from these fires can be detected at the planetary surface (Zuidema et al., 2018). This demonstrates that the central and southern African biomass burning aerosol plume is commonly entrained into the boundary layer of the remote tropical Atlantic Ocean to the south of the DACCIWA region. This confirms that there is a pathway for biomass burning aerosol to enter the boundary layer across large parts of the tropical eastern Atlantic. Once this biomass burning aerosol has been entrained into the boundary layer, the prevailing southerly trade winds at the surface will carry it northwards towards the coast of southern West Africa. There was little evidence of precipitation over the east Atlantic and dry deposition rates of accumulation mode aerosol over open ocean are low. Once entrained, any aged biomass burning aerosol from central and southern Africa would therefore be advected into the DACCIWA region with little further loss. It has been shown that biomass burning emissions in Africa are among the least variable in the world on annual timescales (Voulgarakis et al., 2015). This implies that this influence on the southern West African boundary layer is likely to be a consistent feature of the West African Monsoon.

In a recent multi-model evaluation, the extent of the plume's entrainment into the marine boundary layer over the Atlantic Ocean proved difficult to model consistently (Das et al., 2018), with many models showing the plume descending too rapidly. In contrast, Gordon et al. (2018) use the HadGEM climate model to show the plume remaining above the clouds between 2-4 km and not descending at all until approximately 10 °W. Results from the DACCIWA campaign verify the presence of regular biomass burning plume intrusions at altitudes of 2-4 km over the West African continent, with high aerosol loadings above 60 µg $m^{-3}$ being observed at this altitude in some cases (Flamant et al., 2018). This is consistent with research suggesting that the majority of the southern and central African biomass burning plume remains above the clouds over the Atlantic Ocean (Adebiyi et al., 2014; Das et al., 2018; Gordon et al., 2018). However, results presented here show that in addition, a significant proportion (up to 10%) of the aerosol mass from the biomass burning plume is being entrained into the boundary layer, where it is likely to have a significant impact on cloud properties and human health, particularly for the large population living along West Africa's southern coast.

### 3.3 Aerosol influence on cloud properties

Clouds' susceptibility to increased accumulation mode aerosol decreases when an aerosol background already exists. The relationship between aerosol concentration and cloud droplet number concentration is governed by a power law (Duong et al., 2011; McComiskey et al., 2008; Ramanathan et al., 2001; Terai et al., 2012), so increasing the aerosol number concentration has a proportionally greater impact on clouds that would otherwise have formed in

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clean air. Furthermore, the change in albedo from increasing the number of water droplets is greater in a cloud with an initially low concentration (Twomey, 1977). Below around 100 particles $cm^{-3}$, light scattering by low-level cloud is extremely sensitive to even small increases in aerosol concentration (Kaufman and Fraser, 1997; Kaufman et al., 2005; Ramanathan et al., 2001). This susceptibility decreases gradually; a similar change from a higher initial loading will have a substantially smaller impact. Lower susceptibility would be expected for a cloud forming in a region with an aerosol concentration of 600 particles $cm^{-3}$ or more.

During June and July, extensive low-level cloud forms along the West African southern coast (Knippertz et al., 2011; Schrage and Fink, 2012). It has been speculated that during the monsoon season, clouds above West Africa could be highly susceptible to increases in anthropogenic pollution (Knippertz et al., 2015). However, the presence of a significant quantity of biomass burning smoke in incoming wind from the Atlantic Ocean is likely to reduce this effect.

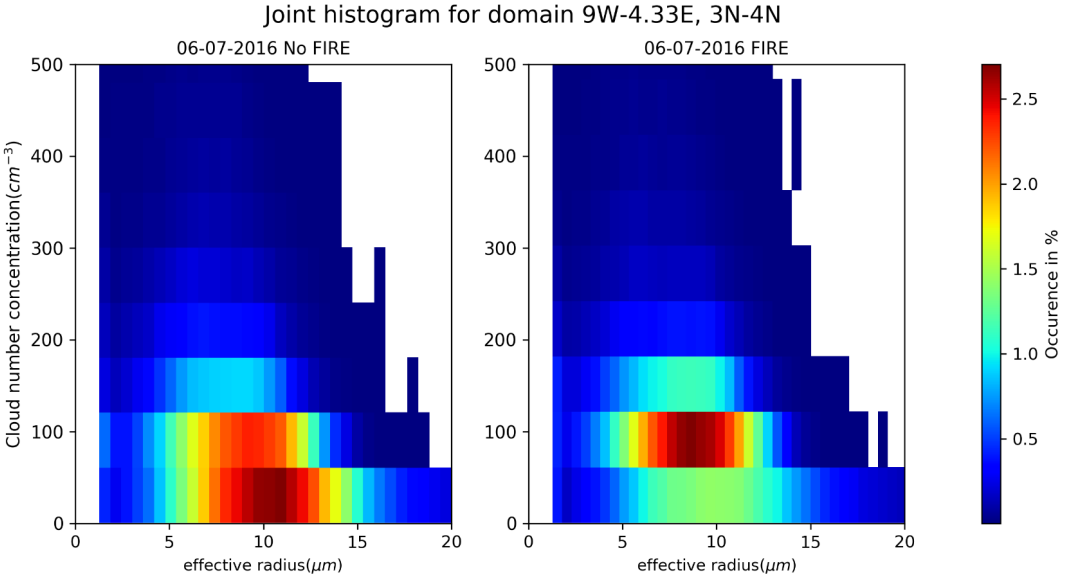


**Figure 8: Two dimensional histograms showing cloud properties in the NO FIRE (a) and FIRE (b) simulations conducted with COSMO-ART.**

Here, the effect of such an influx of biomass burning aerosol on cloud formation in the region was investigated using the COSMO-ART model. Two simulations were carried out for 6 July: one including biomass burning aerosol (FIRE) and one without (NO FIRE). These simulations are used here to illustrate the difference made to cloud properties by increasing the accumulation mode aerosol concentration. Figures 8a and b show two-dimensional histograms of cloud droplet number concentration and effective radius across the inner domain. In the NO FIRE case, the number concentrations are lower and the effective radii higher than in the FIRE case, with number concentrations increasing by up to 33%.



These results show that remote biomass burning aerosol creates a significant background loading that systematically perturbs the cloud field. Any increase in anthropogenic emissions will be superimposed onto this existing background, reducing its influence on cloud. Thus, while enhancements in cloud droplet number concentrations in near-field city plumes were observed during the DACCIWA campaign, the influence of these plumes became indistinguishable further afield as they dispersed into the background. On a regional scale, city plumes were of secondary importance. This conclusion is supported by observations of cloud droplet number concentrations carried out during the DACCIWA campaign (Taylor et al., 2018).

**4 Summary and conclusion**

Observations of aerosol measurements below 1.9 km were collated from the three aircraft that took part in the DACCIWA aircraft campaign during June and July 2016. A regional background of pollution was observed across southern West Africa, which contained around 6 µg $m^{-3}$ of dry aerosol in the accumulation mode and was dominated by aged organic matter. The lower atmosphere above the eastern tropical Atlantic Ocean, immediately upwind of the DACCIWA region, was similarly polluted. Mass concentrations of upwind pollutants here were typically around 80% of those over the land. Contributions from cities and local, small-scale biomass burning to the regional background was of secondary importance compared with this large aged aerosol mass. This aerosol background was attributed to large-scale biomass burning taking place in central and southern Africa. Emissions become entrained into the monsoon flow over the Atlantic Ocean and are advected northwards into the southern West African region.

The chemical composition of this aerosol background is consistent with aged biomass burning being advected over the continent in the boundary layer. Markers of oxidised aerosol dominated the organic mass spectra in all locations with a ratio to total organics that is typical for more aged aerosol. Urban aerosol and the signature of local biomass burning are present, but both play a minor role compared with the larger quantity of aged aerosol. Although there was some day-to-day variability in the total mass concentration, the aerosol background was observed across the entire region with very little variation in chemical composition, suggesting a large-scale, distant source. If this were related to locally-produced aerosol, greater variability would be expected across the region, with larger distinctions between urban outflow and rural measurements. Locally-produced aerosol would be unlikely to be observed over the ocean as far upwind of the coast as it has been observed here, and the composition of the upwind aerosol does not resemble recycled urban emissions. Biomass burning from central and southern Africa is the most likely source of a large-scale mass of homogeneous aerosol in this region. This conclusion is consistent with observations from other campaigns that show biomass burning smoke is present at this time of year in the boundary layer further south in Ascension Island (Zuidema et al., 2018).

Results presented here suggest that the biomass burning pollution accounts for up to 80% of the accumulation mode aerosol mass over the continent. Given this large moderating effect on the air pollution over West Africa at this time of year, the microphysics of the prevalent stratiform clouds in the West African Monsoon is likely already largely



perturbed even before near-field anthropogenic pollution is taken into consideration. Therefore, significant increases in anthropogenic pollutants could have a smaller perturbing effect than would have been the case if incoming air were less polluted (Taylor et al., 2018).

This study takes place in the context of a strong focus in the research community on the dynamics and effects of the African biomass burning plume. A number of campaigns, including LASIC, ORACLES, CLARIFY and AEROCLO (Zuidema, 2016) have recently been carried out over the Atlantic Ocean west of the African continent, with the aim of better understanding this problem and quantifying the direct and semi-direct aerosol effects of the plume, which can differ significantly depending on the altitude at which the plume spreads (Das et al., 2018). This study provides further motivation for understanding these processes, as it shows that the potential for biomass burning aerosol to become entrained into the southern West African boundary layer can have significant implications for large populations in West Africa, in addition to its effects on radiative forcing. During the DACCIWA campaign, it was only possible to gain a snapshot of the boundary layer aerosol, with a limited number of observations over the ocean to the south. It is therefore strongly recommended that more observations are carried out in southern West Africa and over the upwind Atlantic Ocean in the future, in order to document the transport of biomass burning aerosol into the continental boundary layer more comprehensively across larger timescales and areas. Ideally, observations would also be made along the secondary pathway suggested by the simulations of Menut et al., (2018). This will allow improved quantification of the impact on the population along the West African south coast.

The significant contribution of long-range emissions towards local pollution in southern West African coastal cities highlights the often unique challenges faced in policy creation in developing regions. The population in West Africa is currently almost 400 million and is expected to more than double in the next 30 years (UN, 2017), with a growing proportion living in cities along the southern coast. Thus, the boundary layer aerosol described in this paper will increase the $PM_1$ exposure of a large population by around 6 µg $m^{-3}$ from June to September, based on observational evidence presented here. Previous research has shown that during the monsoon season, submicron particles in southern West Africa absorb moisture and can easily grow to more than double their dry diameter (Deetz et al., 2018; Haslett et al., 2018). This would therefore enhance the aerosol mass loading from these particles, potentially close to the 10 µg $m^{-3}$ annual exposure recommended by the World Health Organisation (WHO, 2005).

During the dry season (November-January), high concentrations of desert dust from the Sahara and local biomass burning are advected into the region. Results presented here show that high levels of particulate are not confined to the local dry season but are present throughout much of the year as a result of long-range transport. This regional influx of aerosol presents a challenge for future management of air quality in countries across West Africa. Controlling air quality in these cities cannot be considered solely in terms of reducing local anthropogenic emissions. Rather, regional- and continental-scale sources of particulate, notably these large biomass burning sources, must be considered. This contrasts with air quality problems encountered in North America and Europe, where urban emissions contribute the majority of air pollution. Solely importing air quality strategies from these regions may therefore be unsuccessful in West Africa, given that transnational transport of particulate plays an



important role. Thought should be given to changes in land use practices in countries across the African continent to reduce the quantity of biomass burning if human exposure to particulate matter is to be limited.

***Author contributions.*** SLH, JWT, JB, AMB, SB, JS, CS, CD, TB, RD, AS, DS, CF, JD and IC were involved in the collection, quality assurance and analysis of observational data used in this manuscript. ME, EM, BV and AD developed the model code and carried out simulations. SLH prepared the manuscript with significant contributions from all authors.

***Data availability.*** The data from all three aircraft is publicly available on the SEDOO database (baobab.sedoo.fr/DACCIWA).

***Competing interests.*** The authors declare that they have no conflict of interest.

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