

Boundary layer nucleation as a source of new CCN

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Boundary layer nucleation as a source of new CCN in savannah environment

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Abstract

The South African savannah region is complex environment of air pollution and natural emissions influenced by a strong seasonal cycle in biomass burning and strong precipitation. However, the scarcity of long-term observations means that our knowledge of controlling aerosol processes in this environment is very poor. Here we use a new dataset of 18 months of aerosol observations to understand the factors that control aerosol properties, and in particular cloud condensation nuclei. We find that biomass burning produces a strong source of primary CCN-sized particles during the dry winter season. However, measured CCN-sized particle concentrations remain high during the wet summer season despite the lack of burning and high wet removal rates. We show that during the wet season, a substantial fraction of CCN-sized particles originate from boundary layer new particle formation, whereas primary sources dominate during the dry winter season. The large contribution of boundary layer nucleation to CCN concentrations during the wet season is found to be due to high particle formation and growth rates and low pre-existing particle concentration in the beginning of particle formation. Based on the estimated seasonal cycle of condensable sulphuric acid and organic vapours, higher growth rates during the wet season are attributed to vapours of biogenic origin. Global model results for this region have the same seasonal cycle in nuclei growth rates but the opposite cycle in particle formation rates, and both rates are much lower than observed. In contrast, the same model tends to capture the seasonal cycle in particle concentrations at many other global sites where nucleation is an important process. These results point to deficiencies in our understanding of biogenic emissions and the factors controlling nucleation and growth in such dynamic environments.

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1 Introduction

Clouds, especially aerosol-cloud interactions, constitute perhaps the largest source of uncertainty in predicting the behaviour of the Earth's climate system (IPCC, 2007; Jones et al., 2009; Khain, 2009). The influence of aerosols on the reflectivity, lifetime and precipitation patterns of clouds depends principally on the number concentration of particles able to act as cloud condensation nuclei (CCN). Atmospheric CCN originate either from primary particle emissions or from new-particle formation. In both cases, CCN number concentrations are affected by various aerosol transformation processes taking place in the atmosphere (Andreae and Rosenfeld, 2008).

In southern Africa, the characteristics of CCN have been studied in two field campaigns, SAFARI 2000 and ARREX (Ross et al., 2003). One of the main findings of these studies was that CCN concentrations during the wet season were comparable to or even higher than those during the dry season. This is surprising, since particle emissions from biomass burning are at their highest and aerosol wet removal is at its lowest during the dry season.

The most plausible explanation for the above findings is the existence of a significant ultrafine (<100 nm diameter) CCN source during the wet season. One such source, not considered by Ross et al. (2003), is atmospheric new-particle formation (Laakso et al., 2008; Vakkari et al., 2011), which has been observed globally in different environments (e.g. Kulmala et al., 2004). Earlier observations have demonstrated that aerosol particles formed in the atmosphere may produce new CCN in both clean and heavily polluted environments (e.g. Lihavainen et al., 2003; Laaksonen et al., 2003; Kuang et al., 2009; Wiedensohler et al., 2009). The potential importance of atmospheric new particle formation for regional and global CCN budgets has been demonstrated also using global models (Spracklen et al., 2008; Makkonen et al., 2009; Merikanto et al., 2009; Pierce and Adams, 2007, 2009; Wang and Penner, 2009; Yu, 2009; Kazil et al., 2010), even though uncertainties related to these studies are still large.

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In this study, we investigate CCN production associated with new particle formation over southern Africa, with aim to deepen the understanding on previous findings of Ross et al. (2003). Our main hypothesis is that CCN have different dominant sources during different seasons: atmospheric new particle formation with condensational growth mainly by biogenic vapours during the wet season, and biomass burning or other primary sources during the dry season.

We base our analysis on detailed aerosol measurements, supplemented by trace gas and meteorological observations, conducted over an 18 month period in 2006–2008 at a background surface site in a savannah biome. The analysis is completed by modelling simulations with a global aerosol microphysics model.

2 Methods

2.1 Measurements

Measurements were made in the Botsalano game reserve in North-West Province, South Africa (25°32′28″ S, 25°45′16″ E, 1424 m a.s.l.). The reserve is located about 50 km north of the nearest city, Mafikeng, with approximately 260 000 inhabitants. One of the largest regional pollution sources in North-West Province, Rustenburg mining region, is located approximately 150 km east of Botsalano. Vegetation of the measurement location is typical for mixed bushveld (Laakso et al., 2008).

The comprehensive measurement description is given by Laakso et al. (2008) and Vakkari et al. (2011) and here we list only briefly the measurements used in this study.

The sub-micron aerosol number size distribution was measured with a Differential Mobility Particle Sizer (Hoppel, 1978; Aalto et al. 2001) in the size range from 10 to 840 nm. The sample was drawn through a Digital PM2.5 inlet (Digital Elektronik AG, Switzerland). Prior to sizing, the particles were dried with a Nafion-drier (Perma Pure LLC, USA) and then brought to a known charging state with a Ni-63 beta-active neutralizer. The particles were classified with a Vienna-type (length 0.28 m) Differential

Mobility Analyzer (Winklmayr et al., 1991) and counted with a TSI Condensation Particle Counter (CPC) model 3010. The time resolution of the system is 7.5 min.

Gases (SO_2 , NO_x , CO and O_3) were monitored with one-minute time resolutions using a set of gas analyzers sharing a PTFE-sampling line. Sulphur dioxide was measured with a Thermo-Electron 43S, NO_x with a Teledyne 200AU, CO using a Horiba APMA-360 and ozone with an Environnement s.a. 41A gas analyzer. The gas data were corrected based on on-site multipoint calibrations.

Local meteorological parameters (temperature, potential temperature gradient, relative humidity, wind speed and direction, photosynthetically available radiation (PAR) and amount of precipitation) were logged with a one-minute time resolution. All the instruments were checked and maintained weekly, and a full service was made approximately every three months.

The measurement period utilized in this study started on 23 July 2006 and lasted until 4 February 2008.

2.2 Data analysis methods used on measurement data

The data analyses were carried out using MATLAB program, which was developed to correct the data with calibrations and automatically filter out questionable data. Such values were recorded quite often after frequent electricity breaks. Furthermore, all the gathered data were checked visually to make sure that the questionable data points were excluded.

The formation rate of 10 nm particles (J_{10}) and particle growth rates (GR) for the size interval 10–30 nm were calculated based on the method described in detail in Dal Maso et al. (2005). Values of the condensation sink (CS; Fuchs, 1963), representing the inverse lifetime of non-volatile condensable vapours, were calculated from measured particle number size distributions as in Dal Maso et al. (2005). As the relative humidity at our site is very low (on average, 37%) during the particle formation and growth period, and the hygroscopic growth factors of the particles not known, CS is calculated for dry particle sizes. As the hygroscopic growth of aerosol particles is significant in

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relative humidities above 50 % only (Zhou, 2001) our assumption of dry radius should be safe.

Furthermore, to estimate the contribution of sulphuric acid on particle growth we calculated the so-called “sulphuric acid proxy”, $[H_2SO_4]_{pro}$, based on $[SO_2]$, solar radiation intensity and CS (Petäjä et al., 2009, proxy method 1; Vakkari et al., 2011). Since ambient OH radical concentrations are expected to be proportional to the radiation intensity (Rohrer and Berresheim, 2006), $[H_2SO_4]_{pro}$ is expected to vary as the real gaseous sulphuric acid concentration. However, as this proxy is developed for boreal forest, we consider the absolute values only indicative.

Without direct CCN measurements, we estimated CCN concentrations from particle number size distributions obtained from DMPS measurements by assuming that all particles larger than a certain threshold size act as CCN. Four dry particle threshold sizes (60, 80, 100 and 120 nm) were selected in order to cover different water vapour saturation ratios achieved inside clouds and to take into account the effect of aerosol chemical composition on the CCN activity (Dusek et al., 2006; Hudson, 2007; Quinn et al., 2008). Furthermore, the range of 60–120 nm represents reasonably well the minimum dry diameters of particles observed to act as cloud droplets (Henning et al., 2002; Komppula et al., 2005; Mertes et al., 2005).

Cloud condensation nuclei resulting from new particle formation were determined using the procedure depicted in Fig. 1. After identifying the days with particle formation, we identified the periods with particle formation and growth by visual inspection. To characterize the aerosol formation event we calculated variables such as the number concentrations of particles larger than a certain size before and after the aerosol formation event, minimum, mean and maximum sizes that growing particles reach during the event, as well as trace gas concentrations, sulphuric acid proxy and average meteorology during the formation and growth period.

In total, based on our combined measurements (Laakso et al., 2008) we calculated 56 different variables (like average gas concentrations and meteorology) for the formation and growth periods. During most of the days, determination of the end of nucleation

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mode growth was clear and took place at latest before the atmospheric mixing of next morning started. Due to the selection criteria requiring clear nucleation mode growth, the events utilized in this study are likely to be regional rather than related to individual locations with high emissions. The air mass history was determined using back-trajectories. The hourly 96-h back-trajectories were calculated with the HYSPLIT 4.8 model (Draxler and Hess, 1998, 2004).

2.3 Modelling

To interpret our observations we utilized a global aerosol model GLOMAP capable of simulating CCN formation from primary and secondary sources (Spracklen et al., 2005, 2006). The GLOMAP aerosol microphysics model is an extension of the TOMCAT 3-D global chemical transport model (Chipperfield, 2006). GLOMAP is an offline model where the large-scale transport and meteorology is specified from 6-h European Centre for Medium-Range Weather Forecasts (ECMWF) analyses. We ran the model for the same period as the observations (23 June 2006–4 February 2008), with an initial 4 month spin-up. Model microphysical processes include nucleation, coagulation, condensation of gas-phase species, in-cloud and below-cloud aerosol scavenging and deposition, dry deposition and cloud processing (Spracklen et al., 2005).

The model considers two aerosol size distributions described with 20 size bins spanning from 3 nm to 10 μm in dry diameter. One of the distributions is hydrophobic containing freshly emitted organic carbon (OC) and elemental carbon (EC). The other distribution is hydrophilic and contains sulphate, sea salt, and aged OC and EC. The size-resolved CCN concentrations are obtained as the sum of particles exceeding the threshold sizes in the two distributions. The model has a horizontal resolution of $\sim 2.8^\circ \times \sim 2.8^\circ$ and 31 vertical levels between the surface and 10 hPa. The model is linearly interpolated to the location of Botsalano measurement station.

Carbonaceous aerosol emissions from large scale biomass burning are obtained from the Global Fire Emission Database version 3 (GFEDv3) based on satellite data averaged over the years 1997–2002 (van der Werf et al., 2006). According to the

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database, the burning emissions are strongly seasonal and peak during the dry winter period in Southern hemisphere Africa and are negligible during the wet season (November-April). The annual variability of total carbon emissions in the whole Southern hemisphere Africa is approximately $\pm 15\%$, but larger differences may exist locally.

Anthropogenic carbonaceous and sulphuric emissions are based on the AEROCOM emission inventories for the year 2000 (Dentener et al., 2006). These emissions are kept constant throughout the year. The applied AEROCOM emissions and assumed particle sizes are explained in more detail in Spracklen et al. (2006). We assume that 2.5% of sulphur is emitted as primary sulphate and the remaining sulphur is emitted as SO_2 .

Condensable species include sulphuric acid and condensable secondary organics vapours. In the model we assume that the condensable secondary organic vapours derive entirely from biogenic monoterpenes. Modelled monoterpenes emissions are taken from the GEIA database (Benkovitz et al., 1996). Monoterpenes are oxidized by OH, O_3 and NO_3 to form a first-stage oxidation product (with 15% yield) that is assumed to condense with zero vapour pressure onto existing aerosol. The secondary organics scheme is explained in Spracklen et al. (2006).

New particle formation is modelled using two nucleation schemes. For the free troposphere we use a binary homogeneous $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ nucleation scheme by Vehkamäki et al. (2002). The model predicts binary homogeneous $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ nucleation to take place mostly in the upper troposphere. For the boundary layer we use the empirical kinetic nucleation scheme (Kuang et al., 2008). In the model the latter scheme produces the in-situ nucleation events at the site, while the former scheme contributes to background particle concentrations through downward transport of free tropospheric particles. In the boundary layer the formation rate of 1-nm particles is given by

$$J_1 = k \times [\text{H}_2\text{SO}_4]^2, \quad (1)$$

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where k is the kinetic prefactor. Here we use $k = 2 \times 10^{12} \text{ cm}^{-6} \text{ s}^{-1}$, that in GLOMAP gives a best agreement, on average, with modelled and observed particle number concentrations in various locations around the world (Spracklen et al., 2010).

Model simulations were carried out with primary particle emissions only (PR), PR and binary homogeneous nucleation (taking place mainly in upper troposphere) (PR + UTN), and finally also including boundary layer nucleation (PR + UTN + BLN). The impact of different sources was estimated by comparing CCN concentrations between different simulations. In addition, the simulated size distributions were analysed with the visual method described in Sect. 2.2 to allow direct comparisons with observations.

3 Results and discussion

3.1 General conditions during measurements

The meteorological characteristics for the site were discussed in detail in Laakso et al. (2008) so we only shortly summarize the results here. Typically, the summer temperatures vary between 15°C and 30°C and the winter temperatures between 5°C and 20°C . The wet summer season with significantly enhanced biological activity and high VOC emissions (Günther et al., 1995; Otter et al., 2003) is from October until April with some occasional rains outside this period. The dry winter season is characterized by frequent wild fires and increased domestic heating by small scale combustion and is typically from late April until early September. September is a spring month which typically represents special characteristics as it is still part of the dry season with high primary emissions, but also with significant biogenic emissions from the vegetation.

During the re-analysis of the Botsalano data, we found that in few cases the precipitation sensor was showing unrealistic high values (Odedina, 2009) with rain intensities above 60 mm h^{-1} . Fortunately, such data are infrequent and the results of our previous article (Laakso et al., 2008) are not significantly changed.

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The seasonal meteorology and transport pattern of the region is best illustrated in Garstang et al. (1996). The dominant meteorological conditions are anticyclonic recirculation which dominates during the winter, easterly disturbances frequent in summer and westerly disturbances observed throughout the year. Especially during the anticyclonic recirculation, pollution re-circulates in the atmosphere for up to 20 day long periods.

To check the spatial representativeness of our data, we calculated hourly air mass trajectories for the whole measurement period. Based on visual inspection, we decided to divide the trajectories into three groups based on their origin, typical flow patterns described above, and information from SAFARI 2000 emission inventory. The area division, shown in Fig. 2, is the following: (1) "Polluted": air mass cross over the Rustenburg and/or Johannesburg and/or Vereeniging, (2) "Limpopo": air mass arrives via a trajectory following the northern border of South Africa, but do not cross the industrialized Highveld, and (3) "Other": the trajectories arrive from less populated areas in Karoo and Botswana directions. The reason for this kind of source area classification was the location of the measurement station near the western brink of the polluted Rustenburg-Pretoria-Johannesburg-Vereeniging region. In our location air masses coming from this direction were always polluted, with high SO₂ and accumulation mode particle concentrations. The Botswana-Karoo direction, by contrast, is little influenced by anthropogenic pollution, especially industrial sources. The Limpopo direction is the most common arrival pattern came contains some emissions from domestic biomass burning sources but little industrial pollution.

Based on this analysis, we found no clear annual pattern for the origin of air masses for the days used in our analysis, which indicates that the potential variation is rather related to the season than differences in flow patterns.

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3.2 Aerosol formation and growth rates

During the observation period, new particle formation was observed on a total of 254 (or 69 %) of the days (Vakkari et al., 2011). For this study, we chose 187 new particle formation days with a clear nucleation mode growth up to sizes above 20 nm diameter.

The particle data coverage for the each month varied between 20 % and 100 %, typical monthly data coverage being approximately 80 %. Assuming similar new particle frequency for the days with missing data, days with nucleation with clear growth (and thus utilized in our analysis) was found to take place in approximately 50 % of the time.

Figure 3a shows observed 10-nm particle formation rates (Vakkari et al., 2011).

The 10-nm particle formation rates have a clear minimum during the dry winter season and higher values during the spring and autumn, showing slightly reduced rates during the wet mid-summer. This behaviour is similar to that observed in other environments (Kulmala et al., 2001; Kerminen et al., 2004; Pierce and Adams, 2007; Kuang et al., 2009; Vakkari et al., 2011) and related to the availability of the condensable vapours participating in the particle formation at ~ 1 nm size and subsequent growth to 10 nm.

Overall, the observed particle formation rates were higher than those typically observed in other remote environments and comparable to or lower than those observed in polluted environments (Kulmala et al., 2004; Kulmala and Kerminen, 2008).

Figure 3b represents the modelled 10-nm particle formation rates. The modelled rate is only approximately 10 % of the observed rates indicating a clear underestimation of nucleating and/or condensing vapours. As the modelled SO_2 -concentrations leading to formation of sulphuric acid are similar to observed ones, we attribute the missing fraction to vapours of organic origin. When the simulation data was studied in detail, we also found that unlike in observations, the pre-existing particle concentrations did not show a dip in accumulation mode during the onset of new particle formation. This is due to the underestimated aerosol removal mechanisms in residual layer/free troposphere especially during wet season, leading to an overestimation of the condensation to the pre-existing particles. Characteristic for the area, major fraction of the precipitation

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comes from multi-cell thunderstorms with strong updrafts (Tyson and Preston-Whyte, 2000) leading to challenges in estimating the particle wet removal mechanism in global models. Also, in the model we have used a fixed k in Eq. (1), although it has been observed to vary by 3 to 4 orders of magnitude between individual sites (e.g. Kuang et al., 2008). It might be possible that k also varies inter-annually, possibly due to the influence of organics (Mertes et al., 2010; Paasonen et al., 2010).

Figure 3c shows the observed 10–30 nm particle growth rates. Similarly to formation rates, the growth have a clear maxima in spring and autumn, a minimum during the dry winter season and a smaller minimum during the wet mid-summer. This cycle is similar to those observed in various other environments (Dal Maso et al., 2005; Qian et al., 2007; Manninen et al., 2010; Pryor et al., 2010; Cheung et al., 2011; Yli-Juuti et al. 2011).

Figure 3d represents the modelled particle growth rates. Again, these values are approximately 10–20 % of the observed ones indicating significant underestimation of condensable organic vapours and overestimation of the pre-existing particle surface. The annual cycle, except for a small mid-summer dip is similar to the observed ones. The smaller growth rate also leads to a larger fraction of depleted particles during the particle growth to 10 nm (Kerminen et al., 2004; Pierce and Adams, 2007; Kuang et al., 2009).

The annual cycle of particle growth given by the model follows clearly the observed data and indicates the role biogenic volatile organics have on particle growth. In contrast, the seasonal cycle of modelled J_{10} is different from the one observed. This may, combined with seasonal cycle of particle growth, indicate the contribution of organics also on initial nucleation.

Figure 4a shows the seasonal variation of contribution of sulphuric acid on observed particle growth rates (Vakkari et al., 2011). Instead of the absolute value of the proxy, which is less accurate, the main visible feature in the figure is the seasonal cycle: during the wet season the proxy is able to explain a smaller fraction of the particle growth than during the dry season indicating the contribution of organic vapours in the

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particle growth. The same trend is also visible in modelled data, Fig. 4b, supporting the role of organic vapours on seasonal cycle of particle growth rates.

3.3 CCN-sized particle formation via new particle formation

Figure 5a and b shows the monthly measured and simulated concentration of CCN-sized particles and the relative increases of particle concentrations due to new particle formation and growth (Fig. 5c and d). Both observations and model results show a similar absolute number of different CCN-sized particles, particularly from October to March. The main difference is the dry winter period, when model indicates lower concentrations than observed. A likely explanation for this difference is the underestimated wild fire emissions in the model.

The increase in CCN-sized particle concentration was further analysed by applying the method described in Fig. 1 to both observed and modelled particle data.

Figure 5c and d show the enhancement factor of CCN-sized particles due to new particle formation. The factor was obtained by dividing the number of CCN-sized particles after a particle formation event by that prior to the event. Both observations and model show that new particle formation was not able to affect the CCN-concentrations during the dry seasons. In contrast, the observations show a significant increase of these particles during the wet season. This suggests that new particle formation is a significant source of CCN-sized particles during the wet season. In model results, this increase is not directly seen because of underestimated particle growth rates.

In observation data, the CCN production by nucleation during the wet seasons is likely a result of multiple factors. Besides the higher growth rates, one of them was the fact that the value of CS during the morning inversion breakup just before the new particle formation event was almost two times higher during the dry season ($4.5 \times 10^{-3} \text{ s}^{-1}$) than during the wet season ($2.6 \times 10^{-3} \text{ s}^{-1}$). The reason for the greater CS during the dry season is likely due to anticyclonic re-circulation limiting the venting of pollution, limited in-cloud and below-cloud scavenging and greater biomass burning emissions.

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The modelling results were also analysed by simulating particle concentrations without boundary layer new particle formation, to estimate the importance of BL nucleation. The resulting CCN-sized particle concentrations were clearly lower during the wet season (Fig. 5b). This suggests that the enhancement factor obtained from the visual analysis of the measurement data is likely to underestimate the total effect of new particle formation on CCN concentrations. In addition to boundary layer nucleation, the model runs containing only primary emission suggests that particles originating from upper tropospheric nucleation make a significant contribution to simulated CCN concentrations of all sizes throughout the year (Fig. 5b).

Figure 6 shows the minimum, mean and maximum size of particles in the nucleation mode at the end of growth in observations (Fig. 6a) and simulations (Fig. 6b) obtained as explained in Fig. 1. Clearly, the particles reached larger sizes during the wet season (see also Fig. 3a, b). Since the value of CS during the period of particle growth did not vary significantly between the different seasons (Vakkari et al., 2011), the larger size of nucleated particles during the wet season is mainly due to higher condensable vapour concentrations at that time of the year.

Another look to the simulation data is presented in Fig. 7 which illustrates the predicted CCN concentration at 0.3% supersaturation. The seasonal impact of boundary layer nucleation on composition-resolved CCN concentrations is similar to size-resolved CCN concentrations, supporting the size-resolved CCN counting method used in analysis of observations. Also in case of composition-resolved CCN, modelled boundary layer nucleation made a significant contribution during the wet season.

4 Conclusions

This study investigated the effect of new particle formation on CCN concentrations in a savannah environment in southern Africa. We found that during the wet season, a substantial fraction of CCN-sized particles originates from boundary layer new particle formation, whereas primary particulate sources dominate during the dry winter season.

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The large contribution of boundary layer nucleation to wet season CCN concentrations is mainly due to high particle formation and growth rates combined with the smaller pre-existing aerosol surface area during the onset of nucleation due to mixing of cleaner air from above. Based on the estimated seasonal cycle of sulphuric acid and model simulations, higher growth rates during the wet season are attributed to condensable vapours of biogenic origin.

In our previous analysis of global particle concentrations using the same global model (Spracklen et al., 2010), we showed that the seasonal correlation of observations and the model from 36 sites was improved by adding the same nucleation mechanism as used here. The fact that the model predicts the incorrect seasonal cycle in particle formation rate in the South African savannah environment suggests that other factors, most likely biogenic emissions, are controlling the nucleation process in this environment. The model nucleation rate (at 2 nm) is assumed to depend on sulphuric acid alone and particles grow from 3 nm by condensation of sulphuric acid and secondary organic material. However, clearly the formation rate of 10 nm particles is more strongly influenced by organics than this simple approach would predict. Possibly the nucleation rate also depends on organics (Metzger et al., 2010) or the formation rate at 10 nm could also be affected more strongly by the organic-driven growth rate treated highly simplified in global models (Donahue et al., 2011; Riipinen et al., 2011; Yu, 2011). We emphasize that in the model we assumed the organics to condense on the pre-existing particles kinetically, leading to larger growth rates of nucleated particles compared to a case when organics are partitioned according to particle volume (Riipinen et al., 2007).

In addition to the importance of organics on CCN-formation, results also reveal a strong impact of boundary layer structure on boundary layer nucleation and particle growth. However, as we do not have only surface observations, further investigations of this phenomenon require either in-situ airborne measurements or size-resolved remote sensing techniques. From a modelling perspective, this highlights the importance of sub-grid scale processes affecting the larger scale regional particle formation.

Despite observed differences between the measurement data and modelling results, both methods support the importance of boundary layer nucleation and organic compounds on regional CCN formation in savannah environment.

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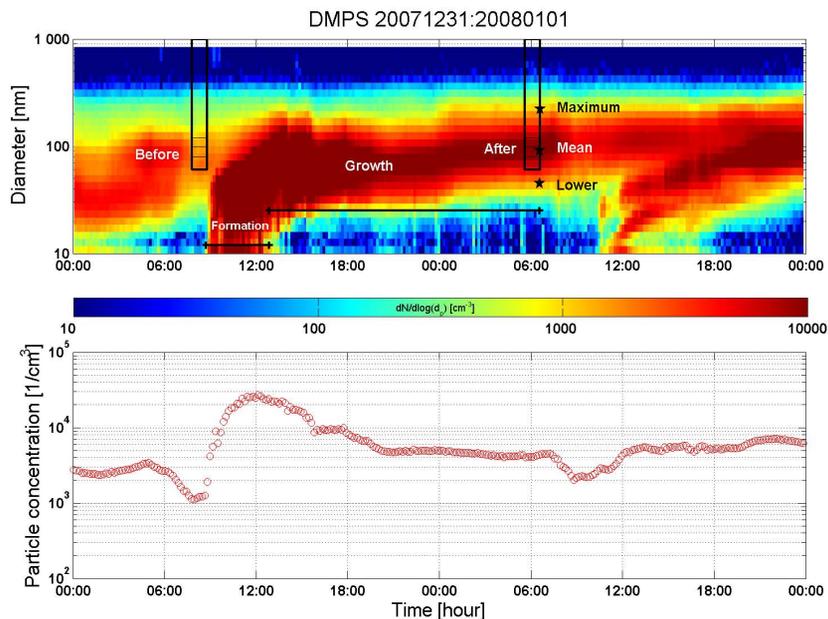


Fig. 1. Evolution of the particle size distribution during a particle formation event showing the different periods where values for analysis are calculated. “Before” is the one hour period before the new 10-nm particles appear, typically just after atmospheric mixing which results in the observed decrease in total particle concentration and breakup of the temperature inversion. “After” is the one hour period at the end of particle growth, but before the start of the next nucleation event. Small horizontal lines in “before” and “after” represent the lower size limits for CCN; 60 nm, 80 nm, 100 nm and 120 nm. “Formation” is the period when 10-nm particles appear whereas “growth” is the period from end of “formation” until “after”. The values used are median values for each period. The formation period is not the same as nucleation period as the particles nucleate at sizes of $\sim 1\text{--}2$ nm. This means the particles appear at 10 nm only approximately 0.5–4 h after they are nucleated.

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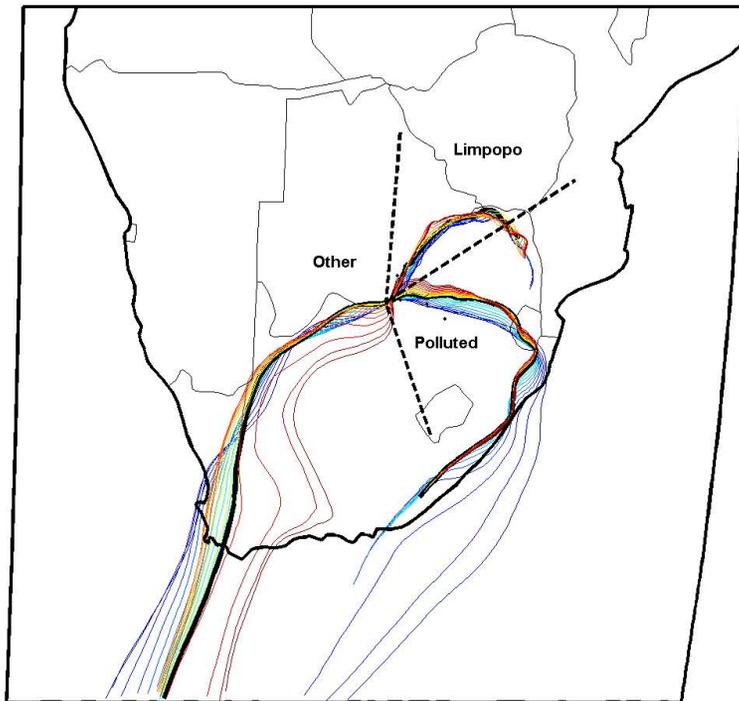


Fig. 2. HYSPLIT back trajectories arriving at the measurement site for three different days demonstrating the area division used in our analysis. The colours of the trajectories indicate the time of the day of arriving at Botsalano.

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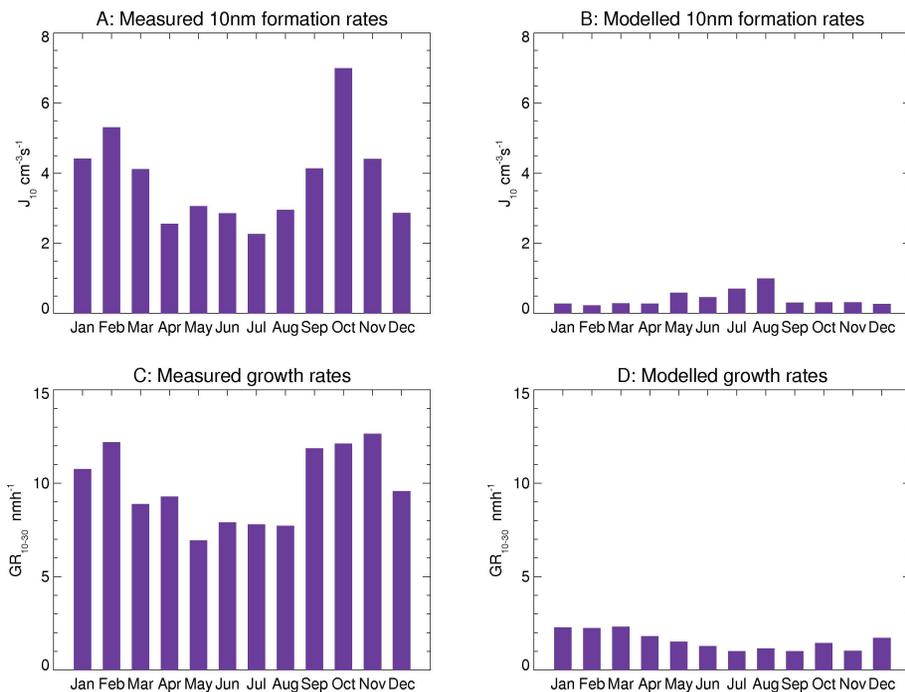
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**Fig. 3.** Observed (left) and modelled (right) particle growth and formation rates.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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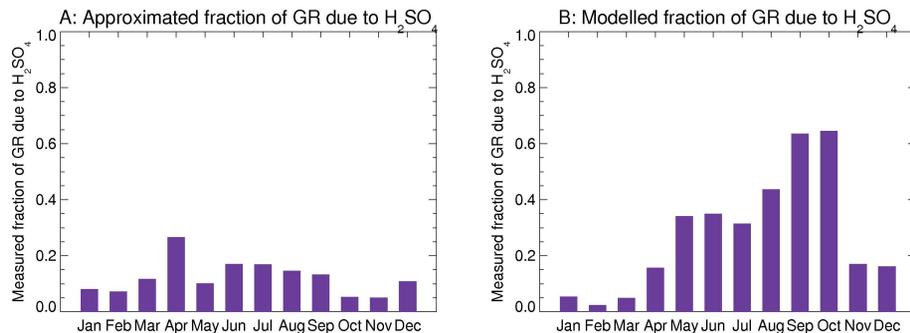


Fig. 4. Approximated fraction of particle growth due to sulphuric acid for observed (left) and simulated (right) results.

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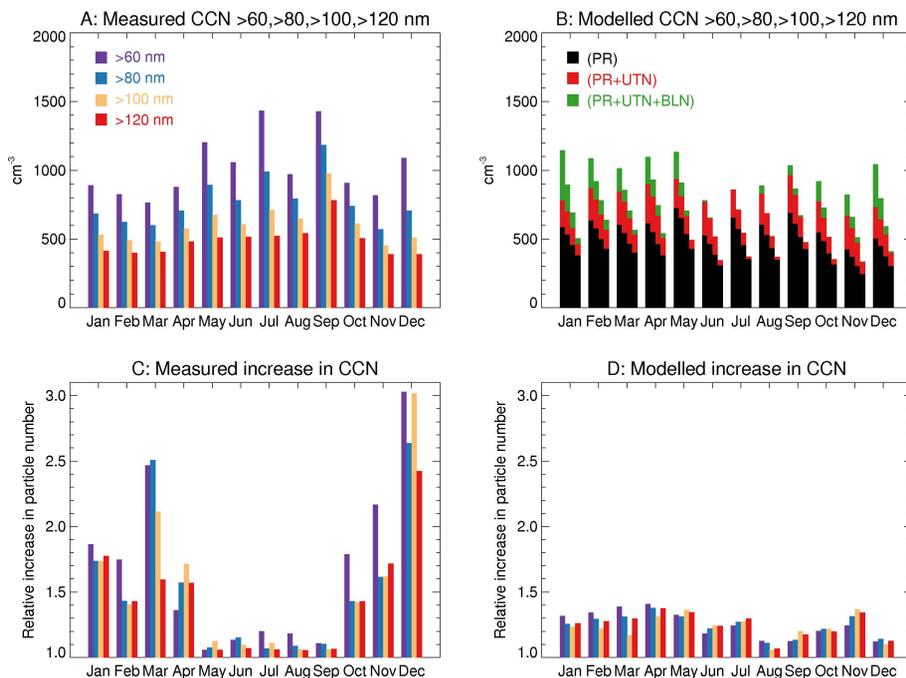


Fig. 5. (a) Measured monthly median concentration of particles above 60 nm, 80 nm, 100 nm and 120 nm; (b) Modelled monthly median concentration of particles above 60 nm, 80 nm, 100 nm and 120 nm; (c) Observed relative increase of CCN-sized particles; and (d) Simulated relative increase of CCN-sized particles.

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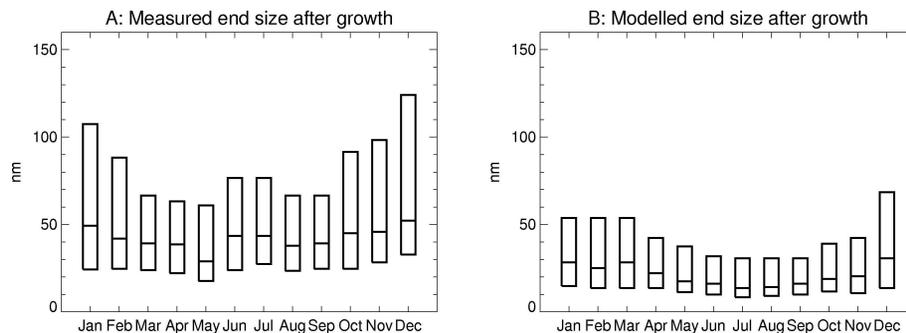


Fig. 6. Smallest, mean and max size of growing nucleation mode at the end of the event **(a)** in observations; and **(b)** simulations.

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Boundary layer nucleation as a source of new CCN

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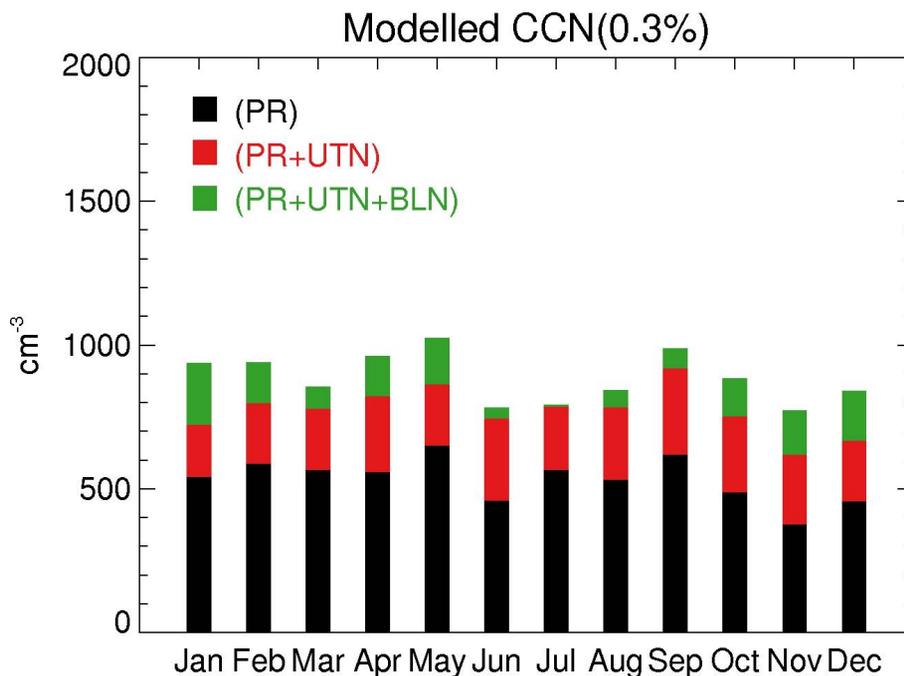
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Fig. 7. Modelled size and composition resolved CCN concentrations at 0.3% supersaturation when accounting for the composition of particles by using a hygroscopicity parameter, κ (Petters and Kreidenweis, 2007).