

**Multiple daytime
nucleation events**

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Multiple daytime nucleation events in semi-clean savannah and industrial environments in South Africa: implications of the driving factors

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Abstract

Two or three consecutive daytime nucleation events followed by subsequent growth were observed in a polluted industrial and moderately-polluted rural environment in South Africa on 108 and 31 days, respectively, based on two years of measurement at each site. In order to describe and understand these observations we analysed particle size distribution data together with air mass back-trajectories, forecasts of the atmospheric mixing layer evolution as well as trace gas and meteorological data. After rejecting days having notably changing air mass origin and wind directions, we were able to investigate selected nucleation events affected by temporal changes in air from specific origins. As a result of our analysis, we propose that the first nucleation and growth event of the day was driven by mixing of a residual layer rich with SO₂, oxidized to sulphuric acid, into the surface coupled boundary layer. The second event of the day started usually before midday and was sometimes associated with SO₂ emissions from the industrial origin. However, our analysis indicates that also vapours other than sulphuric acid, most likely oxidation products of biogenic organic vapours, seem to be required to initiate and sustain especially the second event. We demonstrate that analysis of multiple nucleation events during a single day offers a new way to investigate the factors driving atmospheric nucleation.

1 Introduction

Atmospheric aerosol particles have drawn considerable attention due to their health and climatic impacts (ACIA, 2005; IPCC, 2007). Formation of new aerosol particles via nucleation and their subsequent growth to larger sizes have been observed in a vast variety of environments, ranging from clean arctic air to heavily-polluted megacities (Kulmala et al., 2004; Hirsikko et al., 2011). Atmospheric aerosol formation contributes to cloud condensation nuclei concentrations in the global atmosphere (e.g. Merikanto

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et al., 2009; Pierce and Adams, 2009) and therefore influence the indirect radiative effects of aerosols (Kazil et al., 2010; Makkonen et al., 2012).

Although scientific understanding on atmospheric aerosol formation has increased substantially during the last few years (Kerminen et al., 2010), significant uncertainties related to the factors driving or controlling the spatial and temporal variability of this process remain. The vast majority of nucleation events have been observed to take place during daytime (Kulmala and Kerminen, 2008), suggesting the central role of photochemical reactions and possible assistance by turbulent mixing in the atmosphere (Janssen et al., 2012). Systematic investigation of this topic has, however, been hampered by (i) the possibility of having more than one active nucleation mechanism in the atmosphere, (ii) the apparent participation of several different vapours in the nucleation process (Paasonen et al., 2010; Zhang, 2010), and (iii) the coupling of both nucleation and growth with meteorological conditions and the presence of pre-existing larger particles (Boulon et al., 2011; Kuang et al., 2010; Wu et al., 2011).

In this paper, we propose a new approach to investigate the factors driving or controlling atmospheric nucleation, i.e. the analysis of cases with multiple nucleation events during sunlight hours on the same day. While the presence of multi-event days have been reported before (Sunı et al., 2008; Svenningsson et al., 2008; Manninen et al., 2010; Hirsikko et al., 2012), this phenomenon has not been systematically investigated. We base our analysis on continuous measurements made in a rural savannah site (Laakso et al., 2008; Vakkari et al., 2011), and on a polluted site surrounded by formal and informal settlements, and mining and metallurgical industries in South Africa (Hirsikko et al., 2012). From these analyses we suggest possible mechanistic explanation for the observations. We consider our approach to be applicable to other type of environments as well.

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2 Measurements and methods

Aerosol particle size distributions together with supplementary meteorological parameters and trace gases were monitored at Botsalano (latitude: 25.54° S longitude: 25.75° E, 1400 m a.s.l.) and at Marikana in South Africa (latitude: 25°41'54.51" S, longitude: 27°28'50.05" E, 1170 m a.s.l.) during July 2006–February 2008 and February 2008–May 2010, respectively (Laakso et al., 2008; Vakkari et al., 2011; Hirsikko et al., 2012; Venter et al., 2012). Botsalano is a background site in a semi-clean savannah environment influenced by industrial and urban emissions. The measurement site at Marikana is in the middle of the relatively densely populated and heavily-industrialized Bushveld Igneous Complex, from where the majority of the world's platinum group metals and chromium are produced.

The measurement instruments, data processing and the data quality assurance have been discussed in Hirsikko et al. (2012), Venter et al. (2012), Vakkari et al. (2011) and Laakso et al. (2008), therefore we only give a brief introduction here. Aerosol particle size distributions were measured with a Differential Mobility Particle Sizer (Winklmayer et al., 1991; Mertes, 1995; Jokinen and Mäkelä, 1997) and charged particle size distributions with an Air Ion Spectrometer (Mirme et al., 2007) in the diameter ranges 12–840 nm and 0.8–42 nm, respectively. Various instruments were deployed for monitoring meteorological parameters (i.e. wind speed and direction, global radiation) and trace gases (i.e. sulphur dioxide).

Particle formation event classification was based on the classical method to identify growing modes of freshly-nucleated particles (Dal Maso et al., 2005). Thus, non-growing occurrence of nucleation mode sized (smaller than 25 nm in diameter) particles were not included in the analysis. The air mass history for the nucleation event periods were calculated by using the model Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT, version 4.8) of the Air Resources Laboratory at the National Oceanic and Atmospheric Administration (Draxler and Hess, 2004; Air Resources Laboratory, 2011). The estimate of the boundary layer height (mixing layer depth) was obtained

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from the MARS database of the European Center for Medium-Range Weather Forecasts (ECMWF) (Beljaar et al., 2001; www.ecmwf.int/research/ifsdocs/CY37r2/index.html). The ECMWF runs their Ensemble Prediction System model twice a day, i.e. at midday and midnight (UTC). The temporal resolution of the data was three hours. The surroundings of the station in $1^\circ \times 1^\circ$ square were represented by 0.2° grids. In addition to temporal evolution of global radiation intensity, cloudiness over the measurement area was analysed using geostationary satellite images from MSG/SEVIRI, obtained at 30 min time resolution from Cloud-Aerosol-Water-Radiation Interactions (ICARE) on-line database (<http://www.icare.univ-lille1.fr>).

The condensation sink (CS), which inhibits nucleation and growth of freshly-nucleated particles, was calculated according the formula presented by Dal Maso et al. (2005) and references therein. Based on the measured sulphur dioxide (SO_2) concentrations, global radiation and values of CS we calculated a proxy for temporal evolution of sulphuric acid according to the procedure derived by Petäjä et al. (2009). However, the absolute proxy concentrations have to be considered as indicative only, as the method has not been tested against measured sulphuric acid data from environments comparable to Botsalano or Marikana.

3 Results

3.1 Multiple nucleation and growth events at Marikana

From 559 days on which new particle formation took place (Hirsikko et al., 2012), 108 days with two or three nucleation and growth events were selected. During most of these days, we were able to follow the growth of nucleated particles up to several tens of nanometers, which suggest that this phenomenon has an atmospheric importance, e.g. in terms of cloud condensation nuclei production.

From the 108 multiple event days, we rejected 16 days from the analysis because of too large gaps in the ancillary data. Considering the accuracy of the modelled

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trajectories (Stohl, 1998; Riddle et al., 2006) we selected 24 days for which the origin and path of air masses were similar for the two successive nucleation events and no major changes (60° or more) in local wind direction occurred between the events. The aforementioned decisions ensured a possibility to investigate temporal changes in various parameters specific for the air masses of certain origin and path. Although the selection criteria seem harsh, the remaining days represent the most optimal situations from which mechanistic information regarding multiple events could be obtained.

On the subset of 24 days, concentrations of SO_2 and sulphuric acid and the value of CS varied significantly between two successive nucleation and growth events (Fig. 1). The first event on each day was observed after the sunrise and it was always associated with a growing mixing layer, increasing concentrations of SO_2 and H_2SO_4 (proxy) and often also with increasing values of CS (as high as 0.03 1/s). The high peak values of CS and SO_2 concentration in the morning are likely to originate from downward mixing of a night-time residual layer rich in industrial emissions (Hirsikko et al., 2012; Venter et al., 2012). The second nucleation and growth event of the day was sometimes associated with a decreasing value of CS and/or renewed higher estimated sulphuric acid concentration. However, much of these cases occurred with decreasing or even smaller estimated H_2SO_4 concentrations compared with the first event of the day (Figs. 1 and 2).

The third nucleation and growth event of the day, if observed, took place late in the afternoon and was always associated with air masses different from those arriving at the site during the morning. These episodes occurred during daylight and freshly-formed particles did not always form a well-defined growing mode (Dal Maso et al., 2005).

Clouds reduce photochemical reaction rates. Therefore, the effect of clouds was removed by further selecting the days when clouds were not observed around the end of the first nucleation event of the day. We found that presence of clouds between the successive events may have been one reason to stop nucleation on 21 of above discussed days. During the 3 cloud-free days, the first nucleation and growth event of the

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day took place simultaneously with increasing SO_2 and sulphuric acid concentrations when surface coupled boundary layer was already mixed up to several hundreds of meters (Fig. 2). The first event stopped when H_2SO_4 concentration was still increasing on two of the three days. As an example, on 28 March 2008, sulphuric acid concentration was decreasing when the second new nucleation and growth event of the day started (compare with Fig. 1), which suggests that some other vapours were required to enhance and sustain the nucleation and growth. Similar observations were made on 24 September 2008. However, on 23 November 2008, the second nucleation and growth event of the day was observed when a new increase in sulphuric acid concentration occurred, even though the peak H_2SO_4 concentration remained lower compared with the first event.

The above discussion leads to a question whether the first nucleation event began in the boundary layer coupled or decoupled with the surface (e.g. Stratmann et al., 2003; Laakso et al., 2007; Siebert et al., 2007). Despite the observations of the increasing and decreasing concentrations of SO_2 and subsequently produced H_2SO_4 , scatter plots of medians of these parameters do not allow us to unambiguously conclude that sulphuric acid was more important during either one of the two events of the day (Fig. 2). However, due to sources and temporal behaviour of sulphuric acid concentration in the area, as well as the nature of biogenic vapours we suggest the latter to be an important driving factor for the second particle formation event of the day. A decreasing value of CS between nucleation events results from the increasing mixing volume of the boundary layer.

3.2 Multiple nucleation and growth events at Botsalano

In Botsalano, 31 multiple particle formation and growth event days were observed. However, on 21 of these days either one of the two events did not show all the growth features typical for proper nucleation event (Dal Maso et al., 2005). Therefore, we analysed eight days, which fulfilled requirements of non-changing origin and path of air masses as discussed in the Sect. 3.1. For these analysed days, the air masses arrived

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mainly from the south and south-west, which has previously been associated with moderate formation and growth rates, as well as limited influences from anthropogenic sources (Vakkari et al., 2011). On these days, during the first event of the day particle growth from nano-sizes began immediately after the sunrise when also an increase in the H_2SO_4 concentration was evident (Fig. 3). However, at this site the growing mode was not observable after 5–12 nm on many of the analysed days. During the second nucleation event of the day, the particle growth was also observed from the small ion sizes when boundary layer was already growing, and the growth could be followed over 20 nm.

The observations indicate that clouds had probably affected observed particle formation on three days. On some days, a slightly decreasing H_2SO_4 concentration was seen when the first particle formation event stopped. However, no clear connection between ground based data and stop of the first nucleation event of the day was always observed. The second nucleation and growth event of the day was typically associated with an increasing H_2SO_4 concentration and sometimes also with a decreasing value of CS. This was especially the case on days when the growth of particles during the first event of the day was suppressed at small sizes ($\sim 5\text{--}7$ nm). In contrast, on 28 July 2007 (Fig. 3), we could not indicate any clear reason for the start of the event, suggesting major contribution from other vapours.

4 Summary and conclusions

We analysed 139 days having two or three consecutive nucleation and growth events during one day. The observations were made in a residential and industrial environment (Marikana) and in a rural savannah site (Botsalano) in South Africa during four years (Vakkari et al., 2011; Hirsikko et al., 2012). Our analysis indicates that the first nucleation event of the day typically took place in the morning after the sunrise when the boundary layer was growing and mixed with a residual layer(s) having high concentrations of SO_2 (quickly oxidizing to H_2SO_4), and sometimes increased number of

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back-ground aerosol particles. After the dilution and consumption of suitable vapours, formation of new particles stopped. The fast growth of nucleated particles was observed to continue at Marikana but often to be suppressed at Botsalano. The second nucleation and growth event of the day took place before midday. The observations indicated that while sulphuric acid was the most probable candidate for initiating the first event, other vapours were probably needed for promoting the start of the second event. Other studies (e.g. Paasonen et al., 2010; Riipinen et al., 2011) have shown that low-volatile organic vapours formed in the atmosphere from volatile precursors can have substantial effects on both nucleation and growth of nucleated aerosols. In South Africa, such vapours are likely to be present due to active emissions from the local biosphere (Laakso et al., 2008).

It is possible that some of the analysed multiple events were actually not separate phenomena, but rather a single event interfered by clouds or some other mechanism discussed above. Even though, we suggest the presented conclusions to be valid. As a future it is suggested that boundary layer dynamics and structure measurements, as well as organic species concentrations should receive more attention, since such information could offer additional mechanistic insight.

We suggest that the presented analysis method would be applicable in various kinds of environments and would provide valuable information on atmospheric particle formation. The deduction that the first nucleation event is driven by sulphuric acid vapour from an atmospheric residual layer is relevant to other environments surrounded by industries emitting SO₂. However, it is likely that in the clean background environments, vapours other than sulphuric acid are responsible for the consecutive nucleation and particle growth events.

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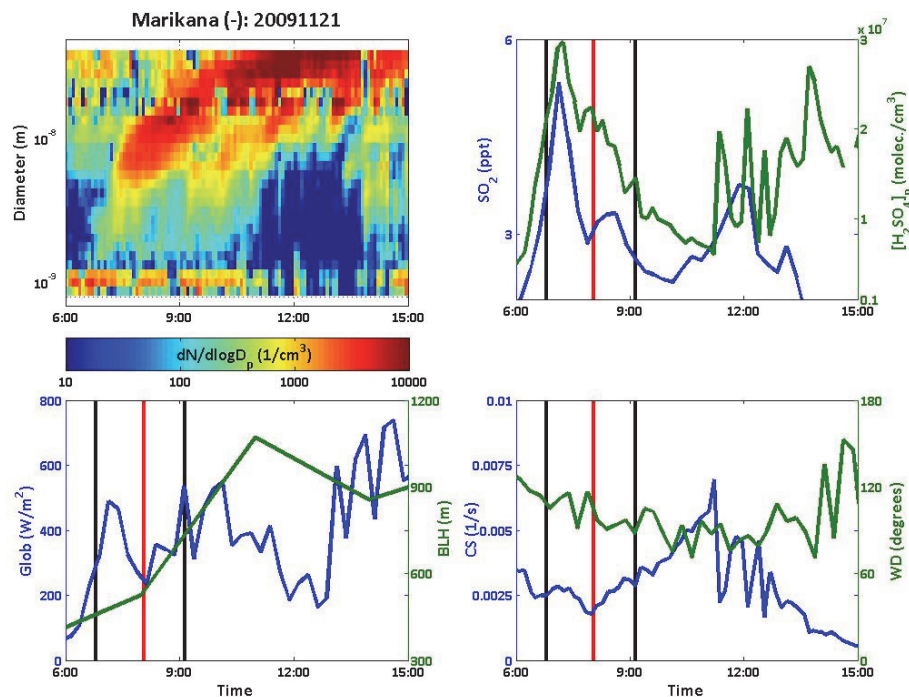


Fig. 1. Example of two consecutive negative ion nucleation events (top left panel) at Marikana on 21 November 2009. Concentration of SO_2 and H_2SO_4 -proxy (top right panel), values of CS and wind direction (bottom right panel), and boundary layer height and global radiation (bottom left panel) are also presented. The first black and red lines indicate the start and end of the first nucleation event, while the second vertical black line indicates start of the second event.

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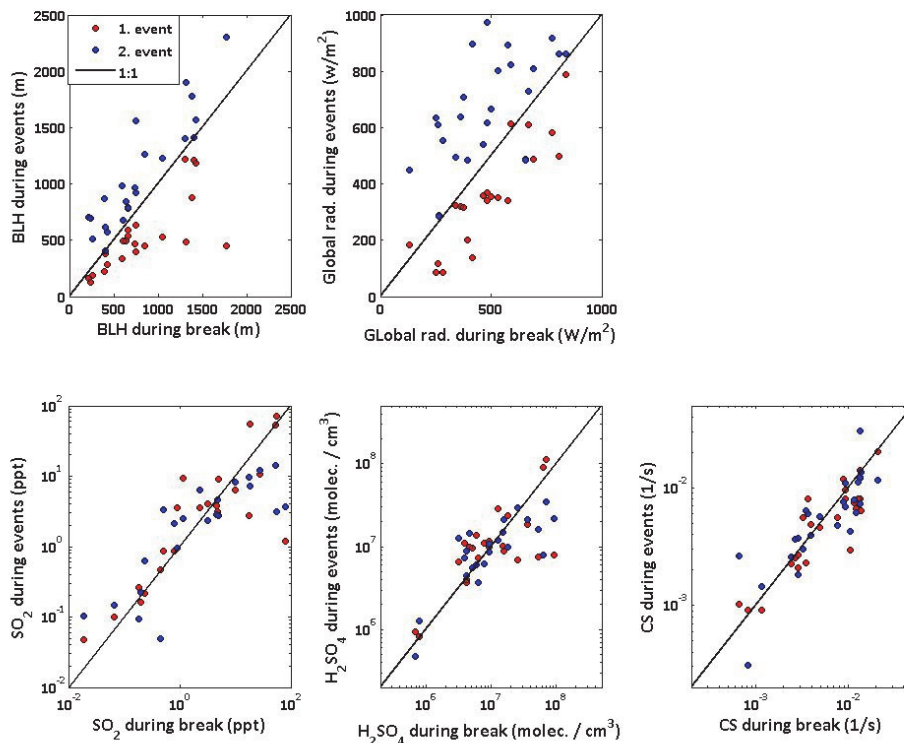


Fig. 2. Median values for mixing layer depth (BLH), global radiation, SO_2 and H_2SO_4 proxy concentrations and CS during the first (red markers) and the second (blue markers) particle formation event versus the corresponding parameters during the break between the events. Days having same airmass origin during successive growing modes from Botsalano and Marikana were included here.

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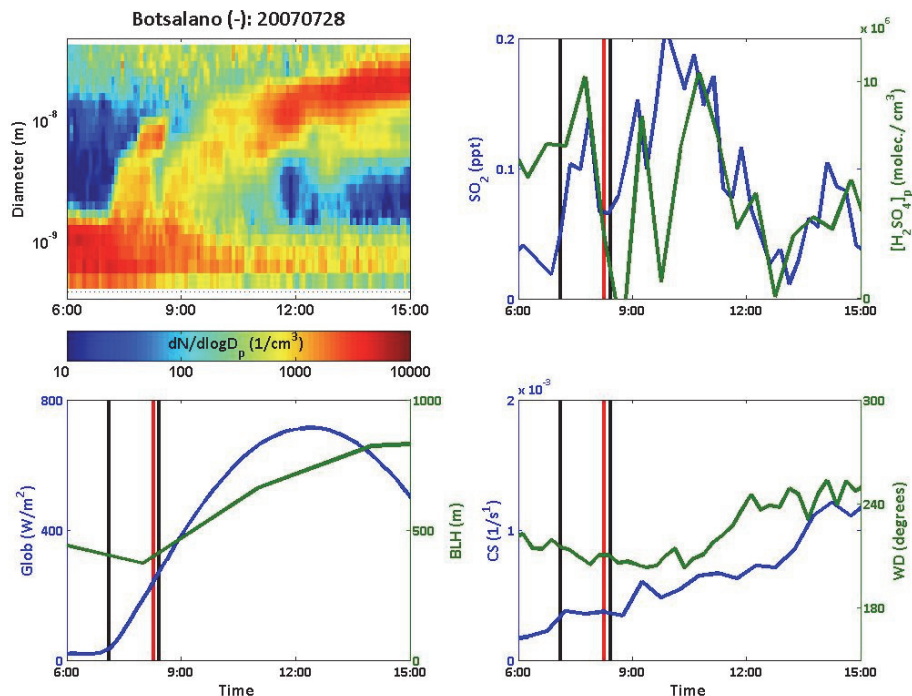


Fig. 3. Example of two consecutive negative ion growth events (top left panel) at Botsalano on 28 July 2007. Concentration of SO_2 and H_2SO_4 -proxy (top right panel), values of CS and wind direction (bottom right panel), and boundary layer height and global radiation (bottom left panel) are also presented. The first black and red lines indicate the start and end of the first nucleation event, while the second vertical black line indicates start of the second event.