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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Multiple daytime nucleation events in semi-clean savannah and industrial environments in South Africa: implications of the driving factors" *by* A. Hirsikko et al.

Anonymous Referee #2

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Summary

The first author has done an excellent job in writing a clear and concise manuscript and in developing the argument for the presented hypotheses. This is a difficult assignment given the very significant limitations of the observational data set, which provides a only a very weak support for these hypotheses (although the hypotheses may be correct). The weaknesses of the data arise from the absence of altitude-resolved SO2 measurements, gas phase H2SO4 measurements, and measurements of nucleating

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compounds other than H2SO4, from the presence of clouds in the majority of the analyzed cases, from the very small fraction of events that support the hypotheses, and from features in the data which are in conflict with the hypotheses. These points are discussed in more detail below. To the authors' credit, they thoroughly disclose and discuss the data.

Overall, the manuscript develops plausible hypotheses from observations and from circumstantial evidence, but the data offer only a very weak support for their validity, and it would be up to future studies to place these hypotheses on solid footing. This approach to publishing results can be seen in a negative and in a positive light: On the one hand, the authors can claim precedence if their hypotheses are proved correct by future, more thorough research, without having to do the research themselves, while they take little risk if their hypotheses are not proved correct, or are even disproved. This approach has become more acceptable in an environment in which the number of publications and citations are an indicator of quality. On the other hand, one could say that the authors have done their best to extract the most from the available observations, and that the conclusions are only as good as the data. Even if one favors the latter, more positive assessment, it must be clear that the conclusions of this manuscript are very weak because the data are very weak, and because of this, I do not recommend this manuscript for publication.

Weakness of data

Marikana station

The authors analyze and discuss the role of clouds for the observations:

"We found that presence of clouds between the successive events may have been one reason to stop nucleation on 21 of above discussed days."

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The number of total days which met the (evidently stringent) criteria for analyzability was 24 at the Marikana station, which left three days without clouds. These are discussed as follows:

"The first event stopped when H2SO4 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 H2SO4 concentration remained lower compared with the first event."

In the conclusions, the authors write

"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."

I agree with the authors that one possible explanation for the second nucleation event could be a nucleating compound other than H2SO4. However, for the Marikana station, there are only 2 (!) days where the presence of clouds or increased H2SO4 during the second nucleation event are not equally valid hypotheses. These two days represent an extremely small sample of positive outcomes to support the following conclusion of the manuscript:

"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."

The fact that this conclusion is weak is highlighted by the use of the words "probable"

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and "probably".

Botsalano station

At the Botsalano station, multiple particle formation events occurred on 31 days, but only on eight of these days the particles showed growth behavior that is typical for a nucleation event. The authors write:

"The observations indicate that clouds had probably affected observed particle formation on three days."

later,

"The second nucleation and growth event of the day was typically associated with an increasing H2SO4 concentration and sometimes also with a decreasing value of CS."

From the eight days with proper nucleation events, clouds may have affected nucleation on three days, and increasing H2SO4 was typically observed during the second nucleation event. Hence the hypothesis and conclusion that "... other vapours were probably needed for promoting the start of the second event" is not supported by the data with the exception of very few days.

Boundary layer height

Figure 3 shows that the boundary layer height is decreasing during the start of the first nucleation event. This is at odds with the conclusion

"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."

Although the hypothesis that mixing of air rich in SO2 into the boundary layer initiates

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nucleation in the morning may be correct, the data do not seem to provide unambiguous support for this.

SO2 levels

Observed SO2 mixing ratios between 0.1 and 5 ppt are shown in Figure 1 and 3. In Figure 2, it is shown that the SO2 mixing ratio is < 10 ppt for the majority of the first nucleation events during a day. SO2 levels < 10 ppt are very low, more common in the clean marine boundary layer, but inconsistent with the polluted industrial and moderately-polluted rural environments described in this paper. Also, SO2 < 10 ppt is usually too low to produce significant nucleation in the boundary layer.

Hence the SO2 units are probably ppb rather than ppt. However, if the units are indeed ppt, then one of the hypotheses of the paper, "... 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" cannot be upheld. Rich in SO2 means hundreds of ppt or more. In addition, SO2 in the ppt range would be inconsistent with the polluted industrial and moderately-polluted rural environments described in the paper: again, polluted means hundreds of ppt of SO2 or more.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 26029, 2012.