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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Towards a first classification of aerosol shrinkage events" *by* E. Alonso-Blanco et al.

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The authors would like to thank referee#3 for the useful comments which have helped to improve the original manuscript. We first address the general points raised by referee, followed by replies to the specific comments.

General comments:

This manuscript presents a classification scheme for atmospheric particle shrinkage events based on a 3.5-year field measurement dataset in an urban background site in Spain. As particle shrinkage events have been sporadically reported in only a handful of studies, this long-term dataset provides a unique opportunity to thoroughly study the types and possible reasons of particle shrinkage. As such, the study subject falls within

the scope of ACP and would be of interests to its readers. Although the manuscript is well written and the adopted scientific methods are fair, there are notable rooms for improvements; specifically, the inference made in the manuscript is rather descriptive and lacks quantitative information from “all” shrinkage events.

Reply - We understand the concerns of the referee#3, and as he/she suggests, quantitative information about “all” shrinkage events is now included in the work in three tables corresponding to each group of shrinkage events, collecting the most relevant information for each case identified. These tables can be found as supplemental information file in pdf format.

Specific comments:

1. In the Methodology, p.25237 lines 5-9, it is recommended to briefly provide what have verified during the intercomparison campaigns of the REDMAAS. In addition, have the authors synchronized all the collected data with varying time-average (i.e., 4.5, 7, 10 min, etc.) to a unified time resolution (e.g., 60 min)?

Reply to comment 1 - The authors agree with the referee’s suggestion. The text has been modified as follows:

“The equipment was checked and the maintenance activities were carried out frequently throughout the whole study period. Furthermore, the response of the equipment was verified during the intercomparison campaigns of the Spanish Network on Environmental DMAs (REDMAAS, in its Spanish acronym) that took place during the period 2010-2012 (Gómez-Moreno et al., 2010; Gómez-Moreno et al., 2015). The flow rates, the high-voltage source and the DMA calibration were checked and the CPC and SMPS were intercompared with other instruments to test the representativeness of their measurements over time.”

In relation to a unification of the time resolution of all collected data in this study, this issue was taken into account by the authors at the beginning of the paper. However, the

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standardization of the resolution of all data means losing the temporal resolution of the SMPS data, core of this study. Thus, the authors have considered more appropriate to maintain the original resolution of the different data that are discussed in this paper.

2. In the Methodology, p.25239 lines 1-9, please briefly provide the difference between Ia and Ib for benefits of the readers.

Reply to comment 2 - The authors agree with the suggestion of the referee. The text has been modified as follows:

"Group I, NPF+shrinkage events: shrinkages produced during the growth phase of the newly nucleated particles of the class I according to the methodology developed by Dal Maso et al. (2005). In turn, the NPF of class I has been categorized as of type Ia or Ib. The NPF of the class I corresponds to those events in which the growth and the formation rate can be determined with a good confidence level. If the particle formation is clear and strong, it is of type Ia, the rest of events belong to type Ib."

3. In the Methodology, p.25239 lines 15-19, please justify the use of "10% difference" as an indicator of atmospheric dilution. In the event of NPF, a substantial increase (much greater than 10%) of nucleation mode particles is common and expected, of which obviously may not be necessarily related to dilution. On the other hand, a 10 % difference of accumulation mode particles would be relatively difficult, of which is more likely related to dilution. I therefore suggest the "10% difference" criterion should be particle size-specific.

Reply to comment 3 - As the referee points out not only in the event of NPF, but also in the event of growth, the changes that suffer the particle concentration corresponding to the nucleation mode (N<30nm) responds to the dynamics of the process, e.g. the aerosol coagulation processes can substantially reduce the particle concentration in these particle sizes. Therefore, the authors have considered the presence of dilution in relation to the total concentration of particles (nucleation, Aitken and accumulation modes) when the ratio between the two event phases (NPF or aerosol growth process

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phase vs shrinkage phase) was higher than 10% difference. The authors understand that the criterion selected is not correctly explained, thus, the text has been modified as follows:

"Changes in aerosol concentrations due to dilution processes have also been studied. In this paper, the authors have considered have considered it has been a dilution phenomenon when the ratio between the two event phases (NPF or aerosol growth process phase vs shrinkage phase) was higher than 10%, because during the nucleation and growth phases may be reduced the particle concentration corresponding to the nucleation mode as a result of the dynamics of both process. This value coincides with the measurement uncertainty established in the ACTRIS SMPS standards (Wiedensohler et al., 2010)."

4. In the Results and Discussion, p.25242 lines 3-17, the authors attributed the seasonality of shrinkage events to increase production (photochemistry and biogenic VOC) and increased residence time for aerosol processes. The latter reason in specific is questionable and lack strong evidence. NPF events are commonly considered as "regional" phenomena that oftentimes span over a spatial scale of 10s- 100s km, and that the winds in the study area were actually stronger during spring and summer. See the following comments.

Reply to comment 4 - When the authors refer to the residence time of aerosols in the atmosphere in this work they mean the dynamics and atmospheric conditions in the study area during the occurrence of the shrinkages facilitate that the aerosol suffers changes and it makes more difficult its removal from the atmosphere.

5. In the Results and Discussion, p.25242 lines 18-24, the inference on the preferable meteorological conditions would be more reasonable to be based on "non-event" vs "event" days as the occurrence of particle shrinkage appears to be case-specific. In addition, it is important to describe and discuss the prevailing winds in order to relate the observed particle shrinkage to possible source regions (e.g., biogenic or anthro-

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pogenic).

6. In the Case study analysis, the authors attempted to generalize and attribute a specific type of particle shrinkage event to certain favorable conditions, prior to each case report. Although I find some inferences are reasonable, I am not fully convinced because of the lack of quantitative information from all the “event” days and that from the “non-event” days. It is strongly recommended to summarize those quantitative (statistical) information into a table to support the arguments.

Reply to comments 5 and 6 - The comments 5 and 6 are answered together because these are related to the same issues.

Firstly, the authors would like to emphasize that the aim of this work is to develop a first methodology of classification of aerosol shrinkage processes which permits to categorize these processes to the scientific community. In this case, the proposed classification, based on the processes that precede them, involves the description of these processes and consequently the discussion of their possible causes. However, the authors understand the concerns of the reviewer and following his/her suggestions, as it was indicated in the general comments, three tables corresponding to each group of shrinkage events with the most important information of each case identified are now included in the paper. These tables can be found as supplemental information file in pdf format.

Obviously, the authors have proposed, as the referee suggests, conducting a statistical study to determine what conditions for the occurrence of shrinkages vs non-occurrence are necessary, as well as, if possible, to characterize the formation pattern (aerosol sources, causes of its formation...) for each group of shrinkage identified. However, they consider that the number of identified events in this work is not enough to achieve statistically significant results. Therefore, and waiting to obtain a representative number of cases, a future article will examine the occurrence of these processes vs non-ocurrence.

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C9182/2015/acpd-15-C9182-2015-supplement.pdf

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