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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of New Particle Formation (NPF) Events at Nearby Rural, Urban Background and Urban Roadside Sites" *by* Dimitrios Bousiotis et al.

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RESPONSE TO REVIEWERS

We thank the reviewers for their valuable comments, and respond point by point below.

ANONYMOUS REFEREE #2 Manuscript entitled 'Analysis of New Particle Formation (NPF) Events at Nearby Rural, Urban Background and Urban Roadside Sites' by Bousi-

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Discussion paper

otis et al. reports the occurrence of new particle formation events at three sites of different environments in the United Kingdom: Rural, urban background and near road sites. The authors study parameters of new particle formation such as frequency, growth rates, number concentration of sizes 16 – 20 nm, condensation sink, urban increment, nucleation strength factor and survival probability. The authors also report trajectory cluster analysis as well as the connection of NPF between the three different sites. In general, the manuscript, contains valuable data (three sites of different environments) and treasured statistics (7 years of data). In addition, the manuscript is well written and literature from around the world is acknowledged. However, the authors make big assumptions and conclusions without enough supporting data. The major concerns listed below need to be addressed before the manuscript is considered for publication in ACP.

Major Comments: 1. The authors report the observation of new particle formation events at three sites in the UK based on visual inspection of CPC (> 7 nm) and SMPS (> 16.6 nm). The general character of NPF events is missing. The lowest limit of the instrument is an issue and no big conclusions can be made before ensuring that the observed plume of particles is related to a new particle formation event. Authors should report how these events look like and whether they have a growing mode shape. Also, more characteristics of the growth should be reported such as possible shrinkage (see e.g. Salma et al. (2016)) and the size these particles reach. An example surface plot from each site should be added to the manuscript.

Let's take for example a regional event surface plot from Kerminen et al. (2018): figure 1, if we cannot observe the information below 16 nm, how can the authors prove that the increase in particle concentration is related to NPF, figure 2.

Figure 1 Regional Event example. Figure from Kerminen et al 2018.

Figure 2 Modified figure 1. The manuscript refers to many NPF studies from around the world, many of which report NPF starting from 6 nm (Salma et al., 2017), 3 nm

(Dal Maso et al., 2005), 1.7 nm (Kirkby et al., 2016) while their measurement starts from 16.6 nm. The authors should present evidence that these observed particles are related to new particle formation events, and not for example a traffic growing mode (Brines et al., 2015). RESPONSE: The dataset available, as mentioned in the text ranges from 16.6 to 604 nm. To overcome this limitation additional data was used to ensure the correct identification of the NPF events. To achieve this: âĂć CPC data was used to provide insight into whether there was an increase on the number of particles of smaller size. An increased number of particles in the size range 7 – 16 nm (provided by the CPC data) right before or at the same time when observed in the SMPS data was a necessary criterion for the occurrence of an event. âĂć High resolution pollution data was used alongside particle number concentration data in a side by side comparison. A sharp increase in the particle number concentration which was accompanied by a similar increase in the concentrations of pollutants was an indication that these particles were probably associated with pollutant emissions. This was mainly an issue in the roadside (MR) and to a smaller extent with the background sites. Increased particle number concentrations observed at times matching the morning or evening traffic rush hours were also ignored at MR as they always coincided with increased concentrations of pollutants. âĂć Meteorological data was used. This mainly applies to the urban background site (NK), being in close proximity to London city centre. The possibility of a plume of pollution originating from the London city centre was considered when the site was downwind of it. A power plant to the northeast of the rural background site (HW) was also considered as a possible source of particles, though the distance is larger. Finally, as mentioned in the text, Heathrow airport and its influence were also considered.

In addition to this, the criteria set by Dal Maso et. al. (2005) were fully considered and unless there was a clear new mode of particles at the lower size range of the nucleation mode with a clear growth for at least 3 hours, an NPF event was not assigned. An example of the appearance of the events for each site has been added in the manuscript. Additionally, a discussion of particle shrinkage at later stages, which was observed at

MR, is also added to the text. Due to the limitations of the dataset, events in which the newly formed particles failed to grow to greater than 16 nm could not be seen except in the CPC data. These were rare and due to lack of additional information about their development were ignored. This clarification has been added in the text. 2. Section 2.1: Which years are studied? RESPONSE: The years studied are 2009 – 2015. This information has been added in the text.

3. Section 2.1: Distance between the three sites should be mentioned. RESPONSE: The distance between MR and NK is 4.5 km. The distance between HW and London city centre is about 80 km. This information has been added to the text.

4. Section 2.2.1: Authors report a visual inspection of CPC and SMPS data. - How was this exactly done? Please elaborate. - Was there any kind of counter-calibration done between these instruments? RESPONSE: The method used was visual inspection of SMPS data supplemented by the use of CPC data to confirm the increase of the particle number concentration in the smaller size range (7 – 16 nm), as mentioned in (1). The text has been updated to clarify the method used. Both instruments are calibrated by the National Physical Laboratory according to the latest internationally recommended protocols.

5. Section 2.2.2: Calculation of the growth rates: - Size of growth rates should be mentioned. E.g. growth from 7 to 20 nm? To 50 nm? - How many points were taken in calculating the GR? - Line 290: Authors claim that GR in NK (4.4 nm/h) are higher than the regional events GR (3.9 nm/h), what is the error bar on these calculations? Accordingly, these growth rates might be similar. RESPONSE: As the lower size available was 16 nm, a calculation of the growth rate up to 50 nm was chosen (rather than up to 30 nm, which provided poor results in many cases due to the small range). The number of points taken depended on the development of the event and were considered from the start of the event until a) growth stopped, b) GMD reached 50 nm or c) the day ended. These points were added to the manuscript to clarify the method used. On the third point made, due to the large variation of the growth rates of the events,

the error bars are overlapped for the two groups of events. This has been included as a note in the text. 6. On line 178: the author mention nucleation mode, which is by definition number of particles between 3 and 25 nm, while the authors conduct a large study on a small fraction of this nucleation mode (16 – 25 nm). RESPONSE: We are not aware of a widely recognised definition of the nucleation mode, with the term taking in different size ranges in the literature. Regardless of that, in the text it is mentioned that "NPF events are considered when a distinctly new mode of particles which appears in the size distribution at nucleation mode size, prevails for some hours and shows signs of growth", which is accurate in relation to the criteria set for NPF event selection in this study. 7. Section 2.2.4: Reference to Kulmala et al. 2017, calculating P = CS'/GR. What GR was used here? See point 4. RESPONSE: The growth rate and condensation sink used are the ones calculated by the methods mentioned in the text. A clarification of this has been added in the text.

8. Section 3.1.1: Reference to Figure S1: cloudiness, and RH.... Is missing. Was cloudiness measured or calculated? RESPONSE: Cloud amount data, as for all other meteorological data were measurements provided by the Met Office, as mentioned in the text. A plot with average cloud amount for each site has also been added in the supplementary.

9. Section 3.1.2: How can the authors prove that NPF events are happening at the near road site and not transported to the location? RESPONSE: It cannot be stated with certainty whether the NPF took place at the site or particles were advected. What can be said though with confidence is that regardless of where the particle formation took place (either on the spot or in the close vicinity, as particles of that size range cannot travel to distances greater than some kilometers before either reaching detectable sizes or being diluted – especially in a polluted environment such as the London city centre), the new mode not only persists but it also grows for at least 3 hours. A clarification of this has been added in the text. If the events at the roadside site were due to advection, or a purely regional phenomenon, a much closer correlation of event days and growth

rates between MR and NK would be expected than was observed.

10. The authors make big conclusions regarding the SO2 driving mechanism of NPF which cannot be proved without adequate chemical speciation of the particles formed. These conclusions shall be minimized throughout the manuscript. Authors could try calculating sulfuric acid proxy from SO2 and CS (Petäjä et al., 2009). RESPONSE: In the text is stated that SO2 was found to be lower on event days compared to the average, which logically leads to the conclusion that either the greater concentrations of SO2 are associated with a more polluted environment with an increased condensation sink (which consequently has a negative effect in the occurrence of an event), or its concentration is adequate and it is not a factor affecting the occurrence of an event (positively or negatively). The calculation of the H2SO4 proxy was carried out and provided information that did not help in clarifying this point. It was found that the proxy was higher on event days at the background sites and gave an unclear result for the roadside. This result though provides no additional information as the increased values of the proxy are the result of the higher solar radiation and the lower condensation sink found during events. Changes were made in the text to "soften" these conclusions.

ANONYMOUS REFEREE #1 The MS mainly deals with the occurrence frequency, particle growth rate, condensation sink, nucleation strength factor, survival parameter and relationships among them at 3 different locations (rural, urban background and urban roadside sites) in the UK over several years. It contains valuable results and conclusions. Some parts of the MS should be elaborated better (some items are given below as examples), and they can definitely be handled and improved. There is, however, a conceptual weakness of the study related to the lower diameter limit of the SMPS system (of 16.6 nm) which can represent the largest source of inconclusive or ambiguous interpretations for the urban sites.

Major comment: 1. New particle formation and growth events are mainly identified, separated from emission sources and classified on the basis of particle number size distributions in the particle diameter range <20 nm (e.g. Kulmala et al., Nat. Protoc.,

7, 1651–1667, 2012). The diameter interval available for this in the evaluated work, namely 16.6–20 nm is quite narrow in particular, when you consider the logarithmic scale of the abscissa of size distributions. More importantly, the lower limit is requested to be even smaller (preferably below 10 nm or at 3 nm) for studies in urban atmospheric environments, where huge emission peaks can temporary dominate the smallest size ranges as well (Nieminen et al., Atmos. Chem. Phys., 18, 14737–14756, 2018). This property (16.6 nm lower limit) of the measuring system and its consequences for the data treatment, results and conclusions at the urban sites should definitely be discussed in detail, explained and resolved before any further opinion could be formed or decision can be made. RESPONSE: The limitations and consequences due to the available dataset, as well as the additions in the method to ensure the correct selection of NPF events are explained at length in the response to Referee #2 (earlier in this document). As a result of this, clarification of the method and the additional data used have been added to the text.

Some minor comments: 1. Lines 21, 69, etc.: it is advised not to start a sentence with abbreviation. RESPONSE: Text updated to address the comment.

2. Line 61: consider writing primary particles or emission sources instead of primary emissions. RESPONSE: Text updated to address the comment.

3. Lines 106–109: it is unusual to attribute particles with a diameter between 1.3 and 3 nm to road traffic emissions, and, therefore, this should be discussed and explained in more detail. RESPONSE: Text updated to accurately reflect the conclusions of the study mentioned.

4. Lines 149–151 or Table 1: supply more detailed data coverage, e.g. for each year or season of years. RESPONSE: Table has been added in the S.I. for detailed seasonal data coverage.

5. Lines 198–203: it is requested that the diameter of particles under consideration is specified as the growth rate changes with diameter. RESPONSE: Text updated to

include the size range of the particles considered in the calculation of the growth rate.

6. Lines 262, 263, Table 2: revisit your rounding off strategy. RESPONSE: Text and tables updated to follow a uniform rounding scheme.

7. Lines 462–463: remove; it is a repetition from lines 235–239. RESPONSE: Text updated to remove repeated information.

8. Fig. 2: it is unclear from the figure or related text which time interval was considered here. A number of NPF events of 90 at Harwell in summer (JJA, 92 days) should be clarified to avoid any misunderstanding. RESPONSE: The figure's description has updated to clarify the period plotted.

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