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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Spatial Extent of New Particle Events over the Mediterranean basin from multiple ground-based and airborne measurements" *by* Kevin Berland et al.

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We thank Referee N°2 for his comments and suggestions that were very useful for improving the manuscript.

Comment 1: I wonder why the authors chose 16 nm for calculating the particle formation rate (and minimum size for calculating GR). In both Ersa and Finokalia, size distribution measurements are available down to about 10 nm. Values of J10 are much better comparable to other studies than J16.

Reply 1: It is true that providing J10 instead of J16 would have ease the comparison with other studies. However, as can be seen in Fig. 7, sub-16 nm concentrations were

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most of the time very noisy in Cap Es Pinar, most probably because of a sampling line instrumental issue, and thus did not systematically allow for J10 calculation. This is now clearly stated in Section 3.2, and the fact that the comparison might be done carefully with J10 is now also explicitly mentioned, both in Section 3.2 and 4.1.2: "While formation rates (J) are usually calculated for 10 nm particles (J10), sampling line issues causing high variability of the sub-16 nm concentrations in Cap Es Pinar (see Fig. 7) only allowed for calculations involving larger diameter particle concentrations (J16). In order to ease the comparison between Ersa and Cap Es Pinar, a similar size range was applied for J calculation from the Ersa dataset. For comparison with the literature, one has to keep in mind that J16 are lower than J10, due to coagulation effects during the growth of the particles from 10 nm to 16 nm." "Besides different environmental conditions which might explain these differences, one has to keep in might that J16 values are expected to be lower than J10 because of the coagulation processes which cause particle loss during their growth."

Comment 2: While equation 1 is mathematically correct, the last correction term in it is based on a very narrow size range. This can make J very sensitive to this correction term. Have the authors investigated this sensitivity? An additional problem related to this is that also GR undetermined based on this very narrow size range. The authors state that the median GR in Finokalia is slightly larger than GR reported in an earlier study for a wider size range (16-20 nm vs 7-20 nm, lines 202-205). However, the difference is not slight at all, but a factor of 4! This larger difference makes me suspicious about reliability of GR determined here using the very narrow size range. This problem concerns also the GR calculated for Ersa: Figure 4 shows a few very high (= unrealistic) monthly-mean GR values.

Reply2: The choice of 20 nm as an upper limit for GR calculation was driven by the fact that in many cases, particle growth beyond 20 nm was not linear. We however investigated the variability of the GR using different size ranges (16-20 nm and 15-25 nm) for the three case studies discussed in the second part of the paper. Based on

this sensitivity study, it seems that the variability of the calculation within a given size range is higher than between the two size ranges. However, we cannot ensure that comparing GR16-20 with GR7-20 would lead to similar conclusions, so comparison with the literature is now performed with emphasis on the uncertainty on the GR calculation, due to both high size range and small size interval that was chosen for the calculations. " The values obtained at Finokalia are in the upper range of the values reported by Manninen et al. (2010) at European sites for 7 – 20 nm diameter particles (1.8 – 20 nm h-1, mean value 4.4 nm h-1). Especially, the values calculated in this work are on average higher compared to those obtained at other European coastal sites such as Cabauw (2.1 - 19 nm h-1, mean value 6.7 nm h-1) and Mace Head (2.7 – 10 nm h-1, mean year value 5.4 nm h-1) (Manninen et al., 2010). Higher growth rates are expected in environments with high solar radiation and emissions, such as the Mediterranean basin. However, the median value reported here is also higher than the one reported for Finokalia from the years 2008-2009 in the size range 7 – 20 nm (5 nm h-1) (Manninen et al., 2010). This result may be explained by the higher size range used here for the GR calculation (16-20nm instead of 7-20 nm), which leads to higher values because GR usually increases with particle size, but also higher uncertainty because of the narrow size range. " Also, the fact that GR are indeed high is expected for high radiation and emission areas.

Comment 3: I wonder why the authors did not report how frequently NPF takes place during the same days between the different station pairs. This kind of information is quite essential when investigating the spatial extend of atmospheric NPF.

Reply 3: The information regarding long-term measurement in Ersa and Finokalia is already provided in the text (l252-254). Concerning the intensive campaign, the information is available in Table S1. We have however included one additional sentence in Section 4.2.1:" As reported in Table S1, during this 41-days period, NPF was observed to occur at one station (at least) on 23 days. Among these 23 event days, 8 events were observed on the same day on two stations at least. This frequency of simultane-

ous NPF events occurrence is very similar to the one observed at Korean coastal sites (5 out of 21 observation days, Kim et al. 2016). NPF was detected at all sites on August 9th, and three events were reported on the same day for each of the station pairs Ersa – Finokalia and Ersa – Mallorca, and one event for the pair Finokalia - Mallorca."

Comment 4: The concept "nucleation area" should be explained better than done here in the main text. By the way, 9 km or 40 km does not represent area, but rather a diameter or some other length measure of an area.

Reply 4: The method we used to estimate the location where nucleation is triggered upstream the station is now explained in the main text (Section 4.2.2) rather than in the supplementary. It is true that most of the information we provide is distance instead of area, so the text was changed accordingly when necessary. Eg: "On July 5th, previous calculations lead to distances of at least 9 km (Ersa) and 40 km (Cap Es Pinar) upstream the stations, which thus cannot allow further conclusions on the simultaneity of a large NPF covering the spatial area of both stations."

Comment 5: The authors state that particle size distributions showed similar trends in Ersa and Cap Es Pinar during the intensive campaign (line 264). By simply looking at Figure 7, I cannot agree with this statement. First, the time axis of this ïňĄgure is so squeezed that it is almost impossible to detect diurnal evolution of size distributions during individual days. Second, the occurrence of NPF event starting from the lowest sizes (10-20 nm) do not seem to co-inside very well between these two stations.

Reply 5: As mentioned in the title of section 4.2.1, the aim of Fig. 7 is only to provide a global overview of the time evolution of the particle size distribution at the three stations during the intensive campaign. We clearly believe that at this "campaign scale", Fig. 7 highlights 3 sub-periods during which all three stations display higher nucleation frequencies. However, we agree with the fact the comparison between the sites cannot only rely on this global approach, that is why Section 4.2.2 is dedicated to a more detailed analysis to describe the similarities/differences between the events observed

on the same days at the three stations.

Comment 6: In addition to the couple of studies mentioned in the introduction, the authors should summarize/discuss a few other earlier studies in which the spatial extend of regional NPF has been studied using multiple stations. This could be done either in introduction, or later in the paper when discussing the results in more detail. Examples of such studies include: Vana et al 2004, JGR 109, D17201; Komppula et al 2006, Atmos Chem Phys 6, 2811-24; Hussein et al. 2009, Atmos. Chem Phys 9, 4699-4716; Jung et al, 2013, Atmos Chem Phys 13, 51-68; Jun et al 2014, Atmos Pollution Res 5, 447454; Kim et al 2016, Atmos Res 168, 80-91; Salma et al 2016, Atmos Chem Phys 16, 8715-28.

Reply 6: We thank the reviewer for this useful list of references. We used the references for works related to comparisons of NPF events detected at multiple background sites, but the ones involving urban areas, which are very specific and would not help understanding our results.

Comment 7: The main stated result of this paper is that the spatial extend of NPF is several hundreds of km over Mediterranean. I am not fully convinced that the results really show this because 1) the estimated nucleation areas are rather small (10-40 km in length), 2) it remains unclear how frequency NPF is observed in at least 2 of the stations during the same day, and 3) the available air craft data do not really support this statement either.

Reply 7: 1) One of the methodologies used in this paper to assess the spatial extend of NPF in the Mediteranean area, (i.e. investigating similarities in NPF time occurrence between several stations) is very similar to the one used by several authors that draw the same conclusion for other environments. We additionally calculated the minimum areas in which nucleation occurred. The fact that our calculation gives a minimum area, and not the totality of the nucleation spatial extend is now better explained in the text. 2) This information was present in the manuscript, but it is now better highlighted in

the conclusion: “NPF formation was observed to occur simultaneously at least at two of the three stations on 8 days over the 41 days of observation, which confirms the frequent occurrence of regional scale NPF events in the Mediterranean area. “ 3) Aircraft data do show that NPF occurs over a large spatial area, but give additional information on geographical gradients and hence indicate that the regional NPF event may have different sources (continental, marine, high altitude). This is now better specified in the conclusion: “Airborne measurements confirmed the regional spatial extend of NPF events, and further showed regional NPF events can have different sources. The selected events depicted contrasting situations where particles were initially probably formed above the continent for one of them, both in the boundary layer and in the free troposphere, and probably formed above the sea for the other.”

Minor comments: they were all addressed

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