Supplement of Atmos. Chem. Phys., 17, 9567–9583, 2017
https://doi.org/10.5194/acp-17-9567-2017-supplement

Atmospheric Chemistry and Physics
Open Access

Supplement of

Spatial extent of new particle formation events over the Mediterranean Basin from multiple ground-based and airborne measurements

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1. Long term measurements from Ersa and Finokalia: meteorological parameters

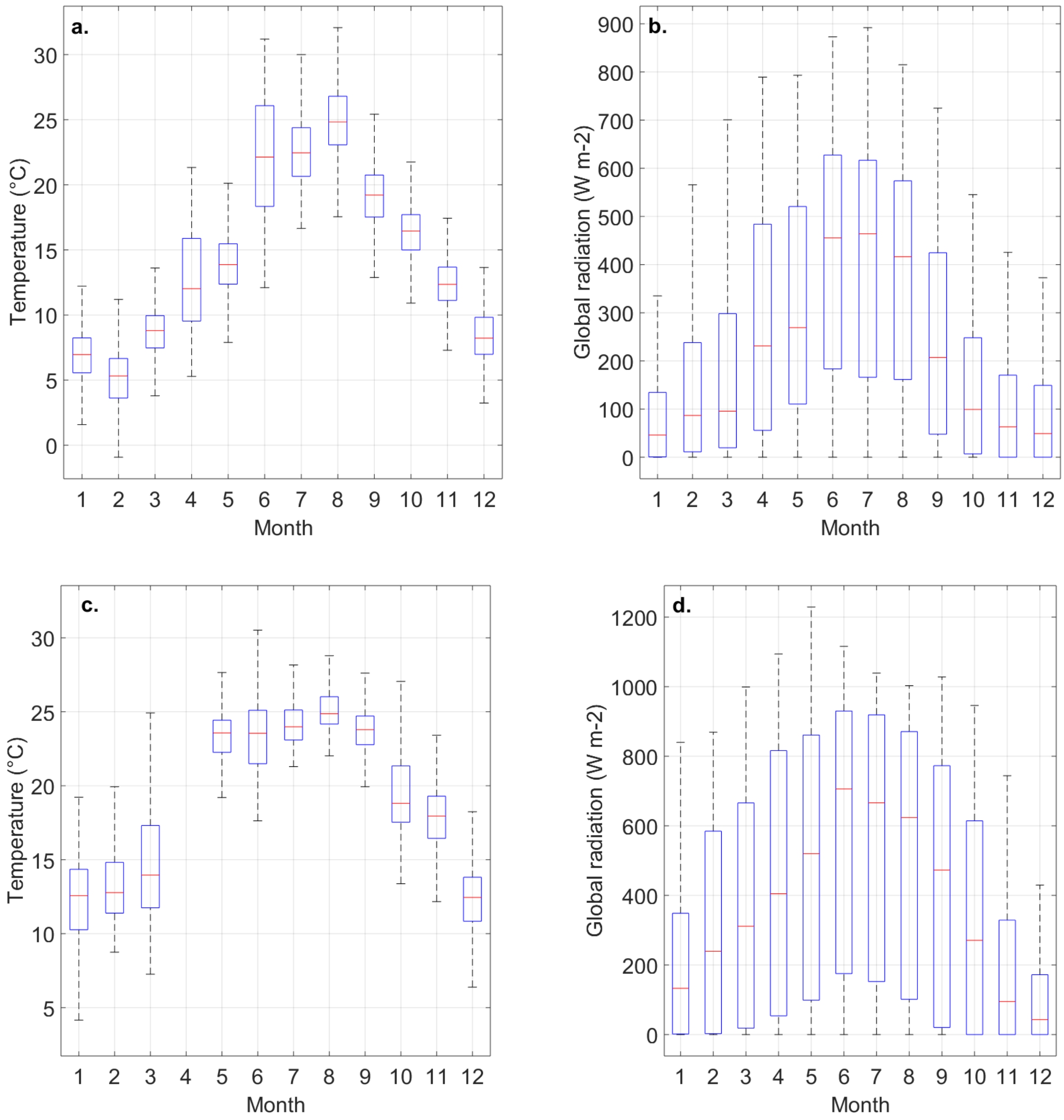

Fig. S1 Seasonal variations of temperature and global radiation in Ersa (a. and b., respectively) and Cap Es Pinar (c. and d., respectively) for 2013. Measurements from the time period 8:00 – 18:00 (UTC) are included in the statistics. Missing temperature values in Cap Es Pinar (April) are due to instrumental issues. Red line represents the median of the data, bottom and top edges of the box symbolize the 25th and 75th percentile, respectively. The dashed lines represent the remaining of the data still within the relevant statistical limit.

2. Intensive campaign during summer 2013

1.1 Global overview

Table S1: Detection of NPF at the ground based stations and onboard the ATR 42.

		July														
		3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Finokalia	SMPS										■	■				
Ersa	SMPS		■	■	■			■								
Mallorca	SMPS		■	■												
	ATR		⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄

		July													
		18	19	20	21	22	23	24	25	26	27	28	29	30	31
Finokalia	SMPS				■		■	■			■	■		■	
Ersa	SMPS												■	■	■
Mallorca	SMPS											■	■		
	ATR		⧄	⧄	⧄	⧄	⧄				●			●	

		August											
		1	2	3	4	5	6	7	8	9	10	11	12
Finokalia	SMPS		■	■	■		■			■	■		
Ersa	SMPS	■	■							■	■	■	■
Mallorca	SMPS									■			
	ATR	●	⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄	⧄

● NPF event observed on board of the ATR-42

■ NPF events

⧄ Absence of airborne measurements

1.2 Ground-based measurements: Case studies

1.2.1 Event of July 5th

Figure S2 shows the diurnal variation of particle concentrations recorded on July 5th at Ersa and Finokalia in the size range 11 – 15 nm (N_{11-15}), and at Ersa, Finokalia and Cap Es Pinar in the size ranges 15 - 20 nm (N_{15-20}) and 20 -25 nm (N_{20-25}). At Ersa, the occurrence of NPF was detected from the smallest SMPS diameters (N_{11-15}), with an increase of all concentrations from 08:00 UTC. In contrast, in Cap Es Pinar, the concentration of the smallest size range available at the station (N_{15-20}) does not display significant variations, while N_{20-25} shows an increase from 13:30 UTC to 17:30 UTC. These first observations suggest that NPF may not be initiated in the direct vicinity of the site at Cap Es Pinar, but rather in a neighboring area. Besides the temporal offset bewteen events at the two different stations, particle concentrations reach higher values at Ersa compared to Cap Es Pinar, with maximum N_{15-20} of 344 cm^{-3} and 53 cm^{-3}, respectively. Similar observations are made for larger particles, with N_{20-25} maximum value three times higher in Ersa compared to Cap Es Pinar (487 against 161 cm^{-3}). Thus it is possible that NPF have been trigerred earlier in the vicinity of Ersa and in the form of a stronger event compared to Cap Es Pinar.

Concerning Finokalia, July 5th was classified as undefined day regarding the occurrence of NPF, and this classification is supported by Fig. S2. As expected, particle concentration in the smallest size ranges does not display significant increase, but rather a series of peaks that do not follow the critiria of a NPF event.

As previously mentioned, newly formed particles were not detected from the smallest diameters in Cap Es Pinar, suggesting that NPF could have been triggered away from the station. In order to give more insight into the spatial extent of the nucleation area and further investigate the connection between the observations from Ersa and Cap Es Pinar, we estimated the location where 20 nm particles observed at the stations had initially been formed.

Figure S3 shows the 24 hours back trajectory of the air mass sampled at t_{max}, ie when the maximum of the 20 nm particles concentration was obtained at each observation site. In order to assess the variability of air mass origins, back trajectories of air masses reaching the station 1 and 2 hours, prior and after t_{max}, are also shown. Based on Fig. S3., air mass origins remained almost constant at both sites during the time window in the scope of this study. Air masses arriving at both stations are of northerly origin. Hence it is not the same air parcel that is traveling from one station to the other.

The place where nucleation is likely to start upstream the station, estimated from GR_{15-25}, is indicated by a green star on the air mass back trajectory calculated at t_{max}. Concerning Cap Es Pinar (Fig S3, right panel), this point is at a relatively short distance from the station, around 40 km, which does not allow us to assess a large spatial extend to the nucleation process itself. However, this analysis does not exclude that nucleation occurred at the same time at larger distances from the site, but over a short period of time, and that these particles are advected to the site later on at larger diameters (resulting in an apparent growth observed on the size distribution). Note that similar calculations could have been performed considering 30 nm particles since they are also clearly formed through NPF (Fig S2).

In the case of Ersa (Fig S3, left panel), we defined a nucleation zone between the point where nucleation was initially triggered upstream the station and the station itself, assuming that nucleation occurred at the measurement site since particles in the smallest SMPS size ranges were detected (Fig S2). The corresponding area is relatively small, only 9 km long, mainly because the GR_{15-25} calculated for this event is particularly high (16.37 nm.h^{-1}). However, it is worth noticing that this area is an underestimation of the actual nucleating zone for two main reasons: first because the use of GR_{15-25}, leads to an underestimation of the distance between the place where nucleation starts upstream the station and the station itself, and also because it is probable that nucleation continue to occur in the air mass downstream Ersa.

As previously mentioned, GR_{15-25} is ~ 16.4 nm.h^{-1} in Ersa, leading to an average J_{20} of 2.4×10^{-1} cm^{-3}.s^{-1}. Significantly lower values are reported for Cap Es Pinar, with a particle growth rate ~ 7.8 nm h^{-1} and 6 times lower compared to that reported for Ersa (0.41×10^{-1} cm^{-3} s^{-1}).

1.2.2 Event of July 29th

The event of July 29th was on a first approach weaker compared to that of July 5th. In Cap Es Pinar, N_{15-20} and N_{20-25} started to increase around 10:15 UTC, reaching a first maximum around 11:15 UTC and a second around 13:30 UTC (Fig S4). In Ersa, particle concentrations increases are seen later, around 11:15 – 11:30 UTC, to reach a first maximum at 11:45 UTC and a second one at 13:00 UTC for N_{11-15} and N_{15-20}; the peak of N_{20-25} is observed later, around 14:10 UTC. These observations suggest that the NPF process would be initiated slightly earlier in the Mallorca region, but occurs faster in the vicinity of Ersa since maximum concentrations are obtained almost simultaneously. This hypothesis is supported by the higher GR_{15-25} reported for Ersa compared to Cap Es Pinar (8.9 nm h^{-1} against 4.8 nm h^{-1}). Unlike the case of July 5th, N_{15-20} concentrations are comparable at both stations (maxima around 120-160 cm^{-3}), as a result of similar J_{20} values, being on average 7.88×10^{-2} cm^{-3}s^{-1} in Ersa and 7.75×10^{-2} cm^{-3}s^{-1} in Cap Es Pinar. In contrast, N_{20-25} is on average higher in Cap Es Pinar compared to Ersa (302 cm^{-3} against 201 cm^{-3}).

In Finokalia, the evolution of the concentrations displays similar features as those observed on July 5th during nucleation hours, with successive peaks attributable to NPF with less clarity. Additionally, concentration increases are seen for all sizes between 11 and 25 nm from 20:00 UTC, suggesting the occurrence of a night time NPF event, shown in more details on Fig.S5. Nocturnal concentration increases are also seen in Ersa (Fig S4). However, they are detected simultaneously for all size ranges between 20 and 80 nm (Fig S5), thus suggesting the implication of other sources than NPF. As a result, the connection between the observations at Finokalia and those at other stations located in the western part of the Mediterranean basin is not clear on July 29th.

Concerning Ersa and Cap Es Pinar, we have shown so far that on July 29th NPF seems rather simultaneous at the two stations, with similar structure and temporal evolution. This could indicate that in this case the spatial extension of the event covers both stations (although the air mass backtrajectories are quite different). We also estimated the closest distance where nucleation started upstream each site (Fig S5). Since particle concentrations were observed to increase from the lowest SMPS sizes at the stations, nucleation areas were outlined between the measurement sites and the locations previously defined.

Again, this was done under the assumption that nucleation occurred at the stations themselves, which could not be directly checked in the absence of additional cluster measurements. The nucleation zones display larger dimensions compared to those estimated on July 5th (61km for Cap Es Pinar and 66 km for Ersa) but still remain far too limited to overlap both stations. However, as already mentioned, the methodology used in this work is only able to predict a lower limit of the actual nucleation area, and thus does not allow ascertaining that events are disconnected.

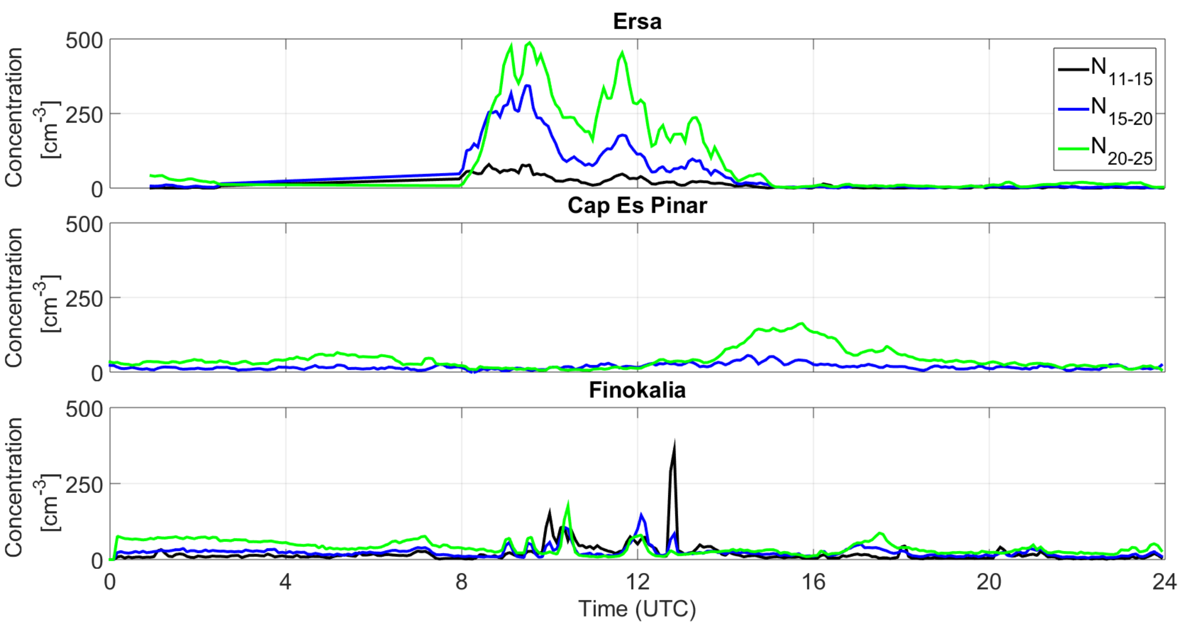

Fig. S2 Temporal evolution of particle concentrations in the size range 11 – 15 nm ($N_{11\text{-}15}$), 15 – 20 nm ($N_{15\text{-}20}$) and 20 – 25 nm ($N_{20\text{-}25}$) for July 5th event.

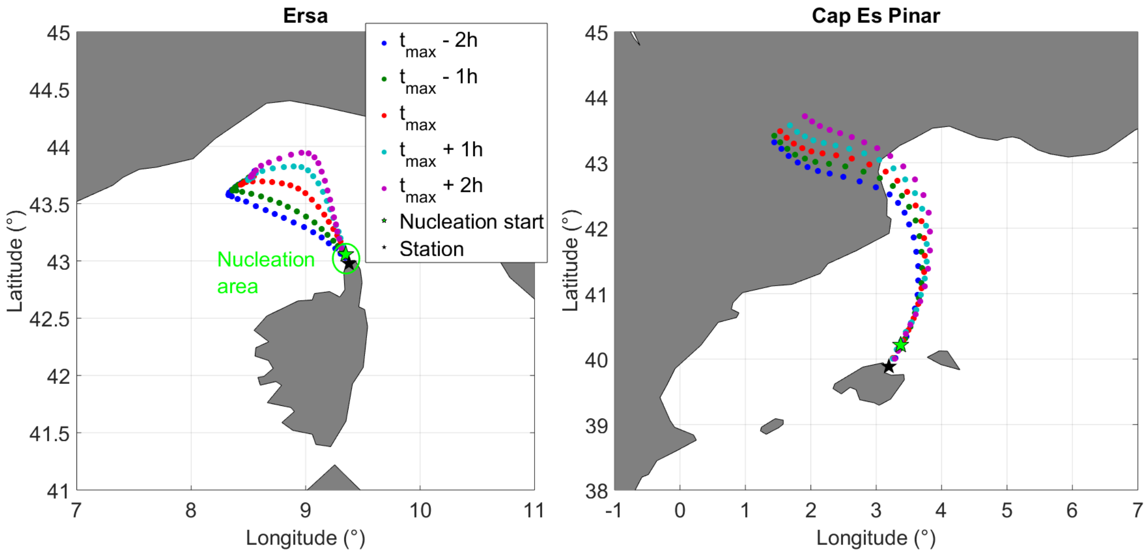

Fig. S3 Backtrajectories of air masses sampled in Ersa and Cap Es Pinar on July 5th at t_{max}, when 20 nm particles concentration is maximum, and during the two hours that precede and follow this maximum. The location where nucleation initially occurs upstream the station is marked with a green star.

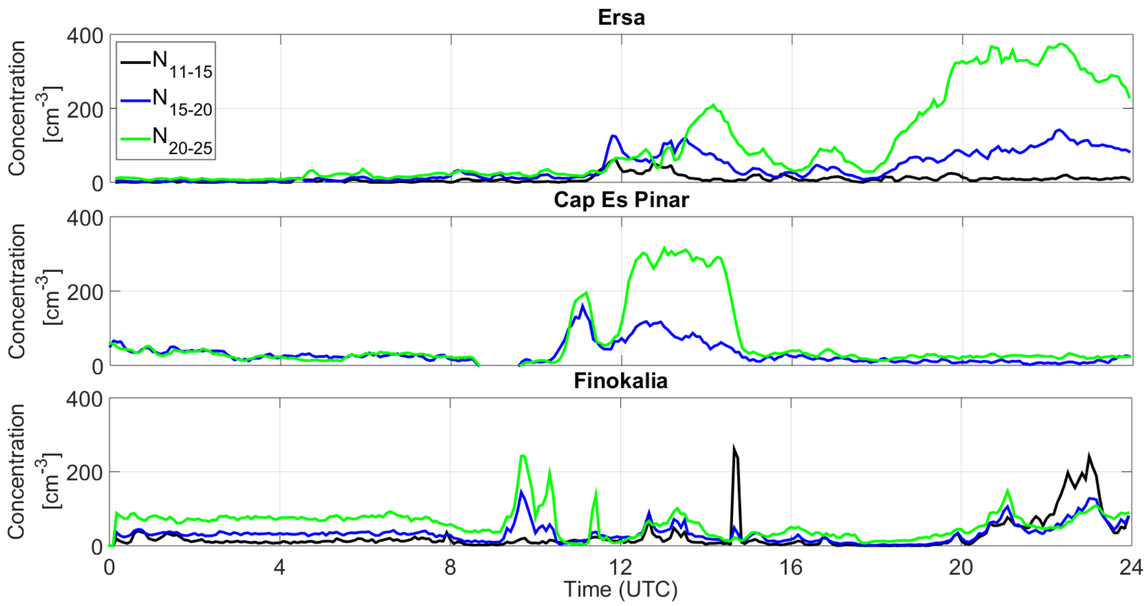

Fig. S4 Temporal evolution of particle concentrations in the size range 11 – 15 nm ($N_{11\text{-}15}$), 15 – 20 nm ($N_{15\text{-}20}$) and 20 – 25 nm ($N_{20\text{-}25}$) for July 29th event.

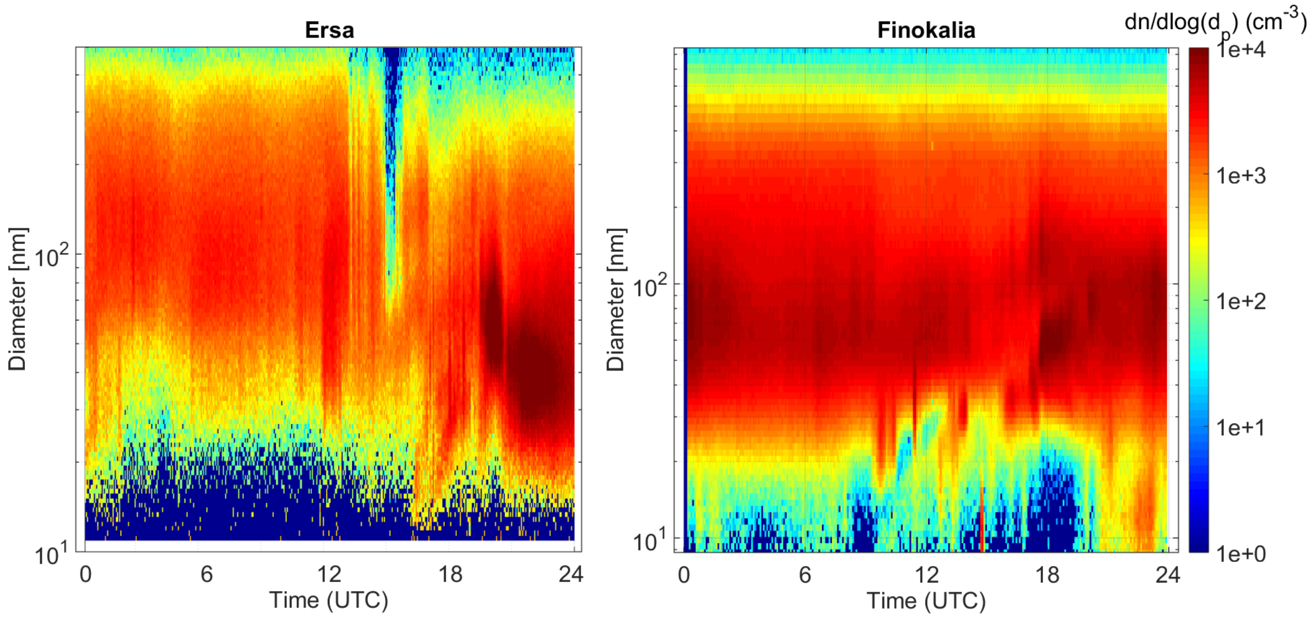

Fig. S5 Particle size distribution at both stations, Ersa and Finokalia , July 29th.

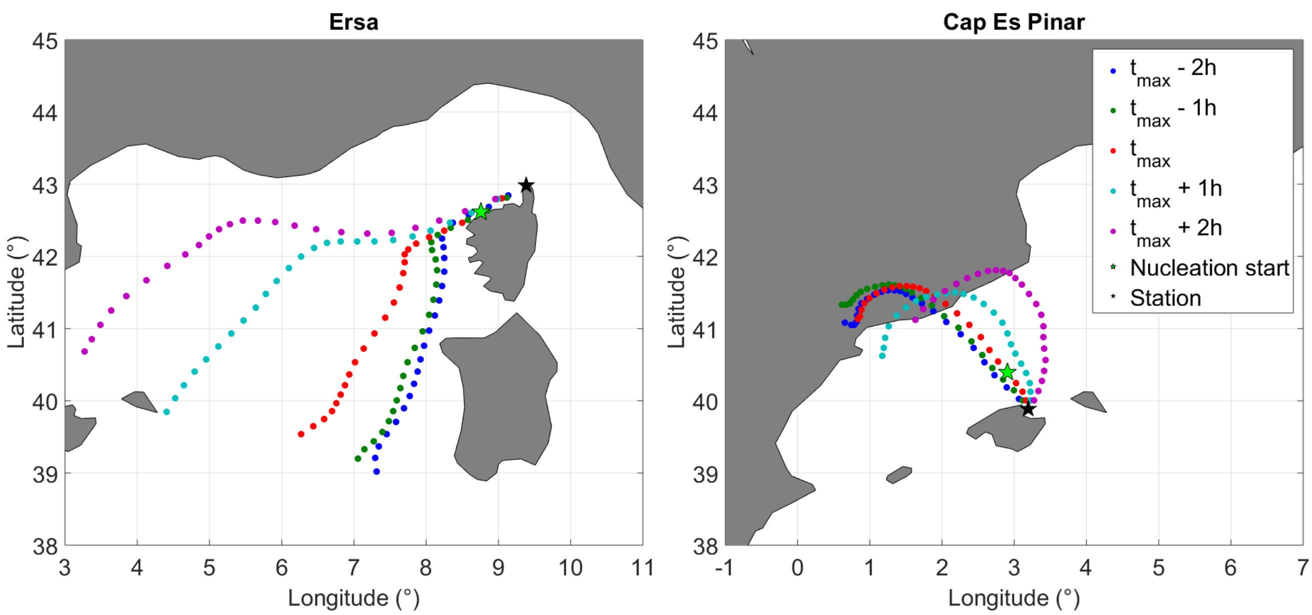

Fig. S6 Back trajectories of air masses sampled in Ersa and Cap Es Pinar on July 29th at t_{max}, when 20 nm particles concentration is maximum, and during the two hours that precede and follow this maximum. The location where nucleation initially occurs upstream the station is marked with a green star.