We thank Referee N°3 for his comments and suggestions that were very usefull for improving the manuscript. We have addressed the comments point by point below.
Also, 2% of the measurements previously classified as performed under non cloudy conditions in the first version of the manuscript were actually performed under undefined conditions regarding clouds. We now classify those 2% measurements as cloudy by default (i.e. filtered from our statistical analysis, for more safety). This leads to very minor deviations on some numbers of the manuscript.

Comment 1: P8152, l10: To analyze the vertical extent is also a purpose of this study. This should be mentioned in this line as well.

Reply 1: This is now clearly mentioned: "The main purpose of the present work was to characterize the spatial extent of the NPF process, both horizontal and vertical."

Comment 2: Please be more precise. Which processes?

Reply 2: These are mainly the processes which contribute to the production of gaseous precursors. This is now stated in the manuscript: "These observations suggest that nucleation events could be more influenced by local precursors originating from emission processes occurring above the sea, rather than linked to synoptic history".

Comment 3: P8152, l17: How was the analyses of the vertical extent performed? It should be mentioned that vertical soundings have been used.

Reply 3: It is now mentioned that vertical soundings were used, and the expression "high altitude" was removed :" Vertical soundings were performed, giving the opportunity to examine profiles of the N_{5-10} concentration and to analyse the vertical extent of NPF. Our observations demonstrate that the process is favoured above 1000 m, i.e. frequently in the free troposphere, and more especially between 2000 and 3000 m, where the NPF frequency is close to 50 %."

Comment 4: P8152, l18: "high altitude", this term is used at several places in the text. For someone doing research in the boundary layer evrything above 1000 m is "high", but for someone doing research in the upper troposphere/lower stratosphere this is rather "low". Therefore, I would suggest to write if possible above which altitude (e.g. above 100 m) or at "higher altitudes".

Reply 4: It is true that the use of the term "high altitude" might be ambiguous. It was thus replaced by the suggested expressions.

Comment 5: P8152, l25: Why "could"?

Reply 5: The use of "could" suggests that our conclusion regarding the particle GR is an hypothesis to explain our observations. Such caution is supported by the fact that our analysis of the particle GR is not based on numerical values that we can compare directly, but rather on the temporal evolution of average particle size distributions recorded at different altitudes. The use of "could" is also related to the fact that the number of SMPS size distributions included in the statistics is reduced for some altitude/time ranges (down to 29 above 3000 m at night).

Comment 6: P8153, l25: It should read “in” rather than “by”.

Reply 6: Correction was done.

Comment 7: P8157, l6: what is SD? As far as I remember the abbreviation has not been introduced yet.

Reply 7: P8157, l6: If we remember well we did not use the abbreviation in the manuscript that we submitted, but the expression “standard deviation”; maybe this was changed during the production process. The expression is explicitly written in the new version of the paper!

Comment 8: P8157, l14: I prefer trajectories six days backward in time. In my opinion three days are too short, but I know that it is common to use just three days.

Reply 8: Since the influence of air masses is mainly discussed regarding the occurrence of nucleation and the production of small particles (5 – 10 nm), we really believe that 3 days are enough, based on the life time of such small particles (also provided in the manuscript).

Comment 9: P8158, l21: “high” altitudes. Better to write above 1000 m.

Reply 9: Since Table A1 reports information for altitudes above 2000 m, “high altitudes” was changed into “above 2000 m”.

Comment 10: P8163, l28: “to be negative”…… Although there is nothing wrong with writing it this way, I would prefer that you write minus temperatures or temperature below zero.

Reply 10: Changed! “It is very clear that temperature is decreasing with altitude, especially above 3000 m where most of the temperatures are found to be below zero.”

Comment 11: P8165, l9-11: Something went wrong in this sentence. Please rephrase.

Reply 11: This sentence is now replaced by two shorter sentences: “During the time period 11:00 – 17:00 UTC, the size distributions are dominated by the nucleation mode. In fact, this mode includes 42% of the particles measured by the SMPS between 2000 and 3000 m, and 48% above 3000 m.”

Comment 12: P1865, l19-21: I have problems to follow how this suggest the speed of the particle growth. Could you be more precise and improve your explanations?

Reply 12: We have tried to improve our explanations. Here is the new version of the paragraph l17 – 21: “At night (17:00 – 05:00 UTC), the contributions of nucleation and Aitken modes to the total particle concentration are very similar between 2000 and 3000 m, being around 34% each, whereas above 3000 m the nucleation mode is dominant (46% against 36% for the Aitken mode). These observations suggest that between 2000 and 3000 m, nucleated particles are growing during the course of the day, leaving the nucleation mode, which thus includes a decreasing fraction of the total particle concentration, to reach the

Aitken mode. In contrast, it is likely that above 3000 m, particle growth is not as fast, since the nucleation mode displays particle concentrations which remain on average higher compared to the Aitken mode, even in the evening. Again, this observation suggests that particle growth could get slower with increasing altitudes."

Comment 13: P8167, l9: "first" obsolete?

Reply 13: Yes, removed!

Comment 14: P8167: Do I understand it correctly that you discuss size distributions that have been measured at the same day during different times? If yes, please write this more clearly. I think the reason for the decreasing coagulation sink with increasing altitude is due to the total number of particles you find in this altitude regions. Usually, as higher the total number of particles (with nucleation mode radii, like e.g. after a nucleation burst, is) as faster the coagulation.

Reply 14: No, here the GR was estimated from the shift of the nucleation mode seen on the average size distributions which are shown on Fig. 10, and which were calculated from the distributions recorded during the whole campaign, as explained at the beginning of section 3.2.1. It is true that "average" was not well used in our first sentence, which was changed to: "Particle GR were estimated from the shift of the nucleation mode diameter observed on the average SMPS size distributions between night time (17:00 – 05:00 UTC) and morning hours (05:00 – 11:00 UTC) for the altitude range 2000 – 3000 m (Table A1, Fig. 10)".

We also agree with the fact that decreased coagulation sinks are explained by lower particle concentrations. This is now clearly stated:" The results of this analysis are reported on Fig. 11, which indicates that particle life time increases with altitude. Such observation might be explained by decreasing total particle concentrations with increasing altitude, thus leading to lower coagulation sinks."

Comment 15: P8168, l7: Again I would suggest to write above 1000 m instead of high altitude.

Reply 15: "high altitude" was removed.

Comment 16: Figure 2: In the caption it could be added that the color coding of the trajectories corresponds to the sectors as given by the text colors.

Reply 16: The figure caption was changed accordingly to: "Illustration of the air mass back trajectories calculation along the flight path (black points) for flight 39 (2012/09/23). The colour coding of the trajectories corresponds to the sectors as given by the text colours."

Comment 17: Figure 15: Which mode is shown here? Or all four modes represented by the size distribution?

Reply 17: We do not have Figure 15 and we cannot find which figure is concerned by comment 17!

We thank Referee N°1 for his comments and suggestions, which we hope will help considerably improving the manuscript. We have addressed the comments point by point below. Also, 2% of the measurements previously classified as performed under non cloudy conditions in the first version of the manuscript were actually performed under undefined conditions regarding clouds. We now classify those 2% measurements as cloudy by default (i.e. filtered from our statistical analysis, for more safety). This leads to very minor deviations on some numbers of the manuscript.

Comment 1: Main conclusion of the study is a claim that lower FT over the studied region is a region where new aerosol particles are observed. It is correct, but on my opinion authors misinterpret the observations. There is also another plausible explanation. Altitude range where they observe majority on new aerosol particles is also shallow cumulus convection cloud layer. It has been shown in several publications elsewhere that new particle are formed in vicinity of these clouds. Specific feature is that these particles are usually observed in a narrow size range with mode between 15 and 30 nm. The same mode as shown on Fig. 10. I wonder how authors can claim that they observe new particle formation if the shape of the size distributions is clearly “closed” with no particles below 10 nm. If authors observed new particle formation they should see, while flying through such region, also many “open size distributions” co-located with high N5-10 aerosol concentrations. No such data are shown. Also horizontal extend of NPF events in Fig. 3 indicates that these features are of limited extend. Thus I would like to ask authors to carefully analyze data with respect to presence and altitude range of clouds in vicinity of measurements.

Reply 1: The “close” shape of the reconstructed size distributions as shown on Fig. 10 were originally “open” type size distributions that were fitted with a nucleation mode that extended below the SMPS lower size cut, as shown Fig. R1. The reconstructed size distribution does represent the real size distribution measured below 20 nm. This point is now mentioned in the manuscript. Fitting a size distribution that would include both the SMPS and the dual CPC measurements is tricky, as we have only one point for the 5-10 nm size range, and one for the 10-20 nm size range.

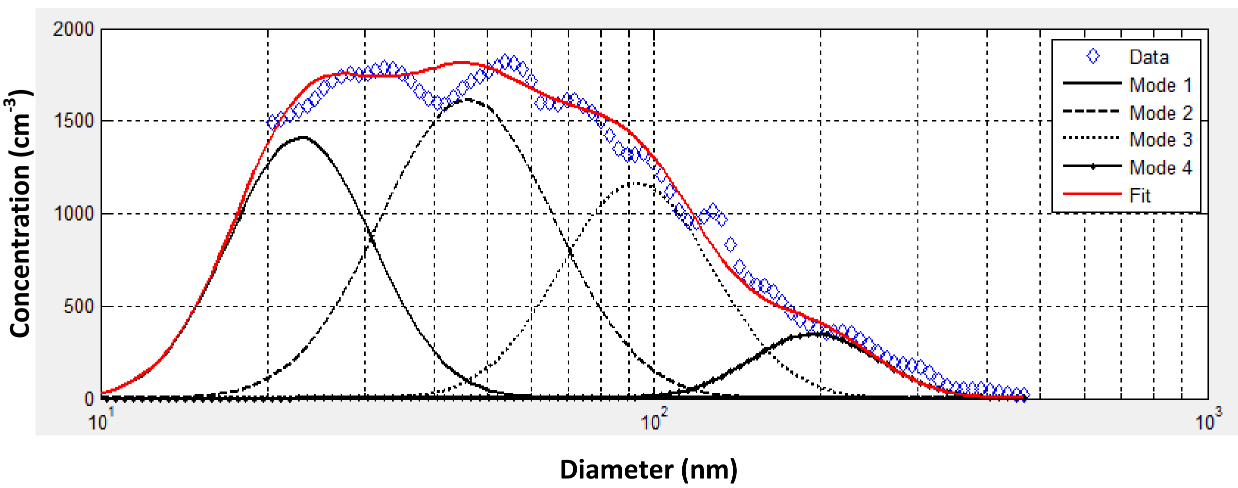

Fig. R1: Fitting procedure of an “open” SMPS size distribution.

However, Fig R2 shows that the time series of open SMPS size distributions do coincide with significant N_{5-10} concentrations.
Regarding the presence and influence of clouds on the NPF process, they are further discussed in reply 2. We indeed cannot exclude that cloud outflows might induce new particle formation on some cases, but further show that the horizontal extend of the NPF process are not only constrained by the presence of clouds.

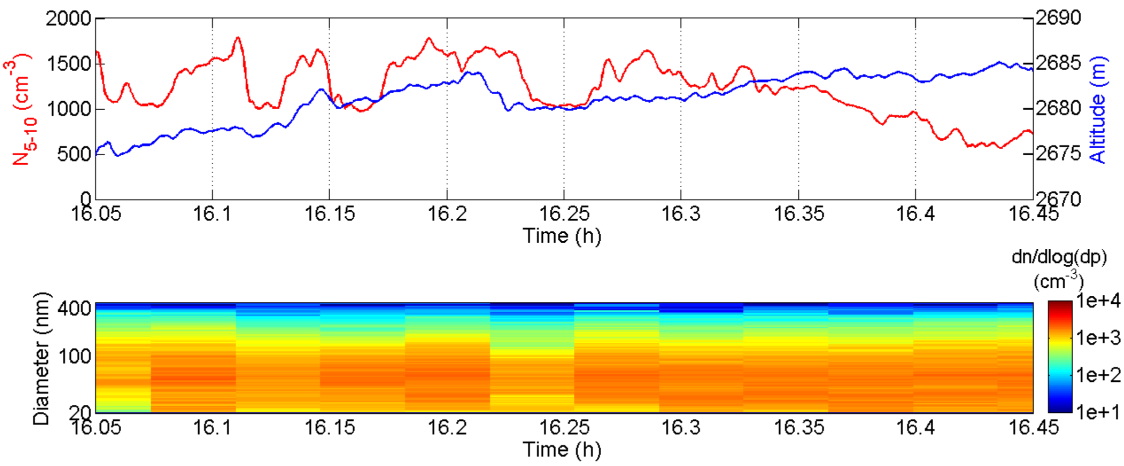

Fig. R2: N_{5-10} concentrations (upper panel) and SMPS size distributions (lower panel) observed at constant altitude during flight n°39.

Comment 2: Continuing from point 1), authors should use ATR core data, trace gas measurements and AMS data to show clearly that the air where they observed new aerosol particles is actually free tropospheric air and not the air recently transported by convection to the altitude where observations have been performed. Excluding the large scale advection the only mean of such transport is convection. If new particles are found in recently convectively lifted air which, then the results cannot be presented as nucleation in free troposphere, but it is additional observation on new particle formation associated with convective clouds and their outflow. Such air will have different chemical signature from FT air as well as different water vapor content. Without this rigorous analysis present conclusions are more of a speculation and not results of robust analysis.

Reply 2: The fact that the formation of the small particles detected at high altitude might have been influenced by inputs from the BL was suggested in section 3.1.4. However it was not clearly stated that such process might occur in the vicinity of clouds or in cloud outflows. In the revised version of the manuscript, the influence of BL intrusions and clouds on the NPF process is further discussed, using tracers such as specific humidity and black carbon (no gaz measurement available, and the AMS data will be specifically investigated in a forthcoming paper). Section 3.1.4 was thus changed to:

“The purpose of this section is to further investigate atmospheric parameters and/or processes which are associated to the higher probability of observation of small particles at higher altitudes.

Meteorological parameters, such as temperature and relative humidity (RH), as well as global radiation, were previously reported to influence the nucleation process. Global radiation, which is expected to be more intense at higher altitudes, and thus favor photochemical processes, including the oxidation of gaseous precursors involved in the nucleation process, could give a first explanation to the observed N_{5-10} vertical distribution. While low temperatures were also found to favor nucleation (Young et al., 2007), the role of RH seems to be more ambiguous. In fact, nucleation is likely to occur preferentially at low RH (Birmili et al., 2003), and both the nucleation rate and nucleated cluster concentration are reported to be anti-correlated with RH (Jeong et al., 2004; Sihto et al., 2006). However, nucleation events have been detected in the vicinity of clouds, where high RH are found (Clarke et al., 1998). Another aspect to consider is that among high altitude air masses, increased RH would also be associated to intrusions from the BL and hence more gaseous precursors and higher CS. The possibility for the small particles that were detected at high altitude to originate from NPF events associated with convective clouds and their outflow will be further investigated in the following.

Statistics concerning temperature and RH recorded during the studied flights are presented as a function of altitude range on Fig. 7. It is very clear that temperature is decreasing with altitude, especially above 3000 m where most of the temperatures are found to be below zero. The same trend is observed for RH, but with higher variability. N_{5-10} concentrations were also directly considered as a function of temperature, RH and humidity mixing ratio (g kg^{-1}), but the correlations between these meteorological parameters and the particle concentration was weak at all altitudes ($|R^2|<0.2$).

Figure 8 shows, for the different altitude ranges previously introduced, the median condensation sink (CS) calculated from SMPS size distributions recorded at constant altitudes, i.e. apart from vertical soundings. Up to 2000 m, the median CS does not significantly vary with altitude, being in the range 3.1 – 3.9×10^{-3} s^{-1}. A higher variability observed below 500 m could again be explained by more inhomogeneous conditions found at low altitudes. Above 2000 m, CS values are significantly decreased, with median values below 10^{-3} s^{-1}. These first observations suggest that higher nucleation frequencies found above 2000 m could be, at least partly, explained by lower CS. The fact that nucleation could be promoted at higher altitudes due to lower CS values was also reported by Boulon et al. (2011) at the puy de Dôme (PUY) station (1465 m a.s.l, France), where NPF is observed twice as frequently as at the BL station of Opme (660 m a.s.l.) located 12 km south east of the PUY.

A more complete analysis focussed on altitudes above 2000 m was then conducted to highlight the role of the CS in the nucleation process at higher altitudes. Figure 9 shows the correlation between N_{5-10} particle concentration and CS, separately for the two altitude ranges above 2000 m. The N_{5-10} shown are 130 second averaged values coinciding with SMPS measurements used for the CS calculation. Based on Fig. 8, we observe that ultrafine particle concentration and CS are positively correlated within each altutude range, especially between 2000 and 3000 m (R^2 = 0.48). The lack of measurements did not allow similar analysis at lower altitudes to compare with, but the fact that at higher altitudes, where the CS is usually low compared to BL stations, increased CS could favour the occurrence of nucleation has already been reported in the literature (Boulon et al., 2010; Rose et al., 2014). While lower CS values are typically reported on event days compared to non-event days at BL sites, increased CS are found on event days at high altitude stations (Manninen et al., 2010).

In the present study, we may hypothesis that some gaseous compounds are transported, together with the pre-existing particles, from lower altitudes, and that they may be further oxidized to more condensable species involved in the nucleation process. As previously mentioned, such processes might be favoured by convection associated with clouds and their outflow. In that case, the lifted air parcels where small particles are detected are expected to have different chemical signature from free troposphere air, as well as different water vapour content. Also, the fact that clusters might be formed at lower altitudes and then be transported together with larger particles above 2000 m cannot be excluded. In addition, it has been previously reported by several studies that the mixing of two air parcels showing contrasting levels of RH, temperature, condensation sink and precursors, could favor the occurrence of nucleation (Nilsson and Kulmala, 1998; Khosrawi and Konopka, 2003; Dall'Osto et al., 2013).

We further investigated the contribution of cloud processes and BL intrusions regarding the formation of new particles using tracers such as the black carbon (BC) concentration and the specific humidity, in addition to cloud cover. Unfortunately, there was no measurement available regarding the composition of the gas phase. Our analysis was focussed on the vertical soundings performed by the ATR-42 that allow a direct comparison of the vertical distribution of the parameters of interest (Fig. 5 and B1). *(New Fig. B1 is shown below)*

Among the 17 profiles previously associated to NPF in the FT (profiles n°2, 6, 13, 17, 18, 19, 26, 27, 29, 30, 31, 32, 33, 34, 35, 36 and 38), although cloudy conditions were filtered from our analysis, we retrieved that clouds were observed in the same altitude range as small particles in 4 cases (profiles n° 29, 30, 34 and 38). For 3 of them, collocated increases of the specific humidity (profiles n° 29 and 30) and/or BC concentration (profiles n° 30 and 34) were also found. These observations suggest that for those 4 particular cases, the formation of small particles was most probably induced in recently lifted air from convective clouds. For the remaining soundings, clouds were detected at lower altitudes: for soundings n° 2, 6, 13, 18, 19, 31, 33, 35 and 36, the vertical cloud profile was sparse, while it was denser for profiles n° 17, 26, 27 and 32. Missing data did not allow a complete analysis of soundings n° 18 and 36, which will thus not be further discussed. During sounding n°31, high N_{5-10} were found in the close vicinity of the cloud. The origin of small particles observed during profiles n° 6, 13, 27 and 32 could not be stated unambiguously, since they were observed at altitudes characterized by low BC concentrations but median specific humidity. In contrast, the vertical distributions associated to profiles n° 2, 17, 26, 33 and 35 clearly suggest the occurrence of NPF events in free tropospheric conditions, free of the influence of recent BL inputs. In addition, during sounding n° 19, small particles were detected around 1000 m and slightly below 4000 m, while increased specific humidity and BC concentrations were observed between 2000 and 2500 m.

We have shown so far that above the Mediterranean Sea, new particle formation was observed over large areas and could be favoured at higher altitudes, since particles in the size range 5-10 nm are mostly seen above 1000 m. However, the previous analysis did not always unambiguously answer the question regarding the conditions associated to the initial cluster (1-2 nm particles) formation, especially in terms of the degree of BL influence/intrusions. Nonetheless, these particles, whatever transported to or formed in the FT, in more or less polluted conditions, are expected to grow to larger diameters and might reach climate relevant sizes in the FT. The purpose of the next section is to investigate this growth process above 2000 m by analysing the shape of the SMPS size distributions."

Abstract and conclusion were also changed accordingly.

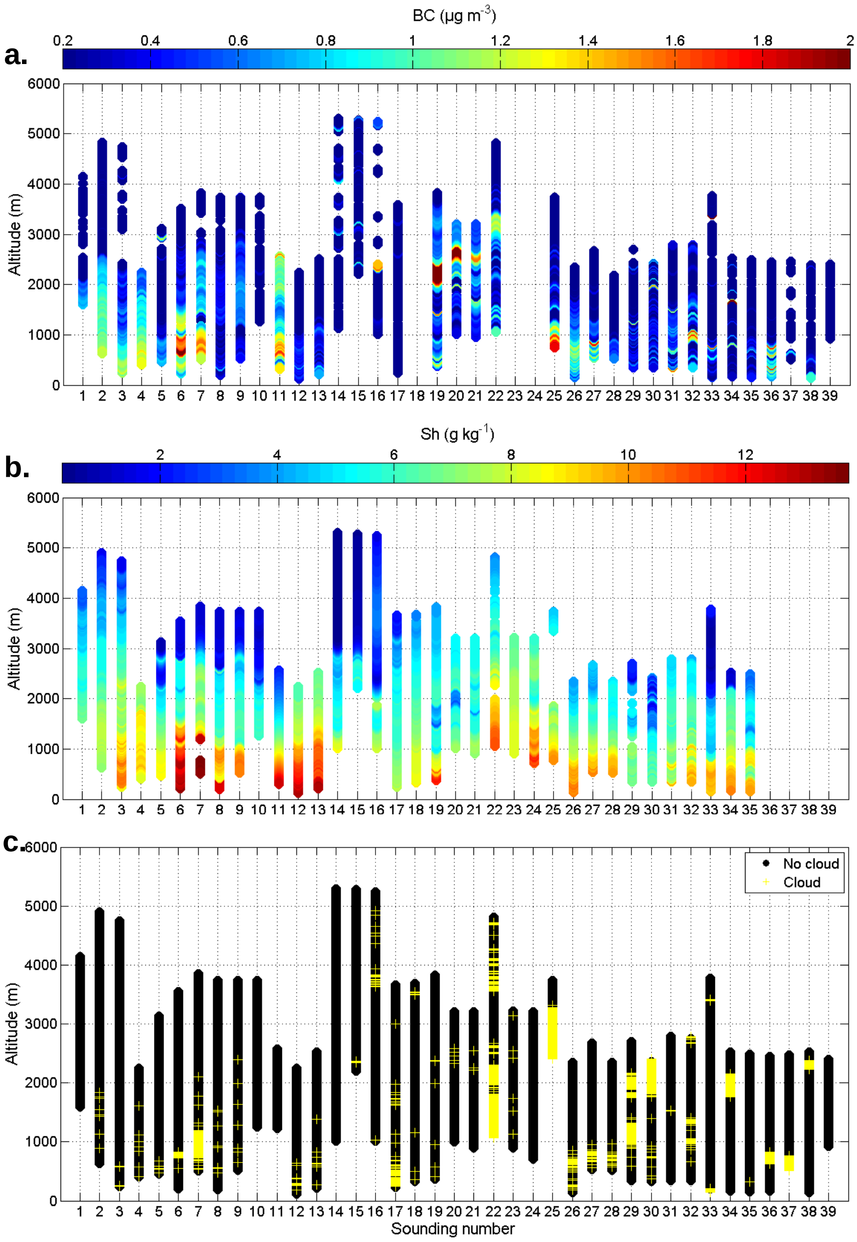

Fig. B1 Profiles of a. BC, b. specific humidity and c. cloud cover during the ATR-42 soundings performed above the sea.