

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.

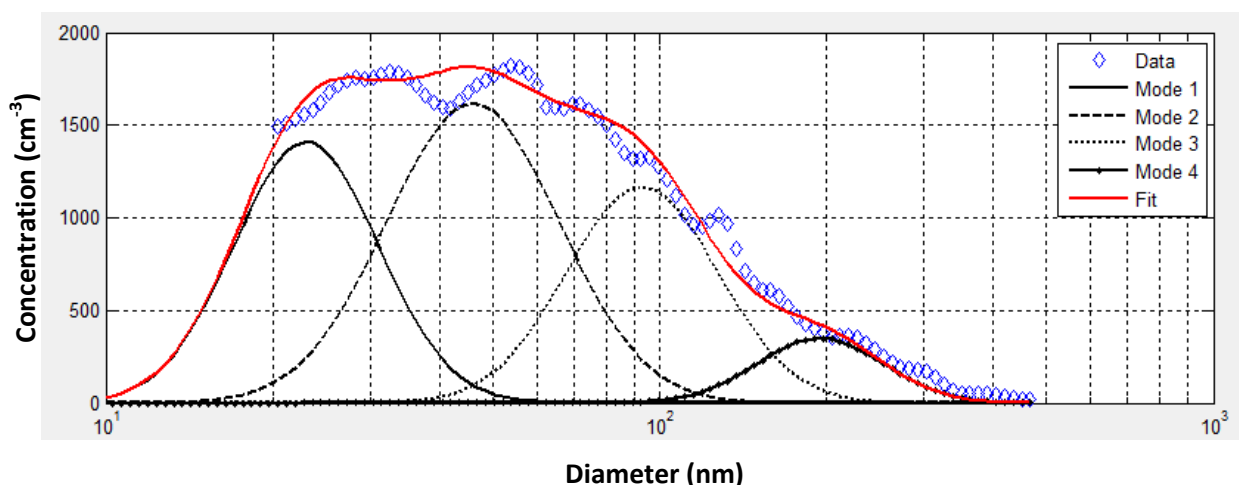


Fig. R1: Fitting procedure of an “open” SMPS size distribution.

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.

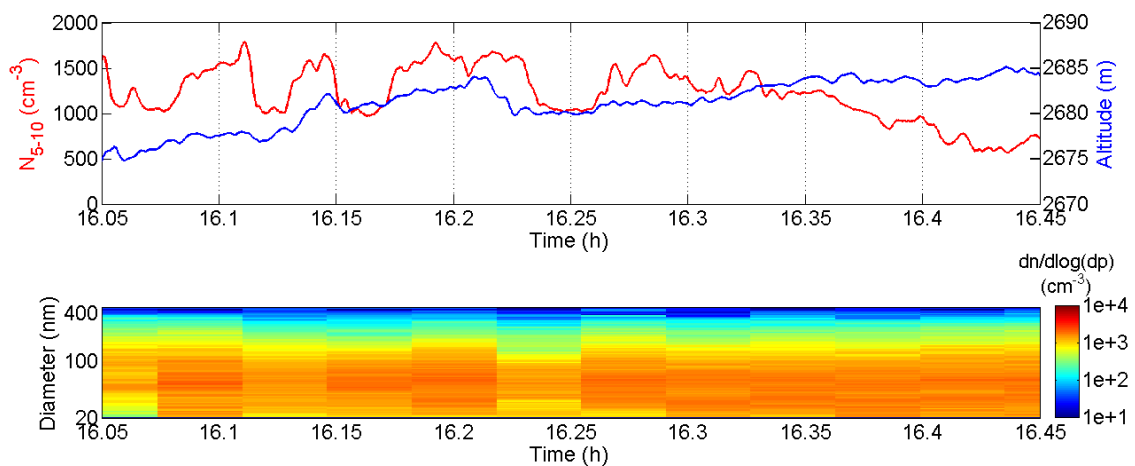


Fig. R2:  $N_{5-10}$  concentrations (upper panel) and SMPS size distributions (lower panel) observed at constant altitude during flight n°39.

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 ( $\text{g kg}^{-1}$ ), 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  $3.1 - 3.9 \times 10^{-3} \text{ s}^{-1}$ . 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  $10^{-3} \text{ s}^{-1}$ . 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 altitude 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 hypothesize 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.

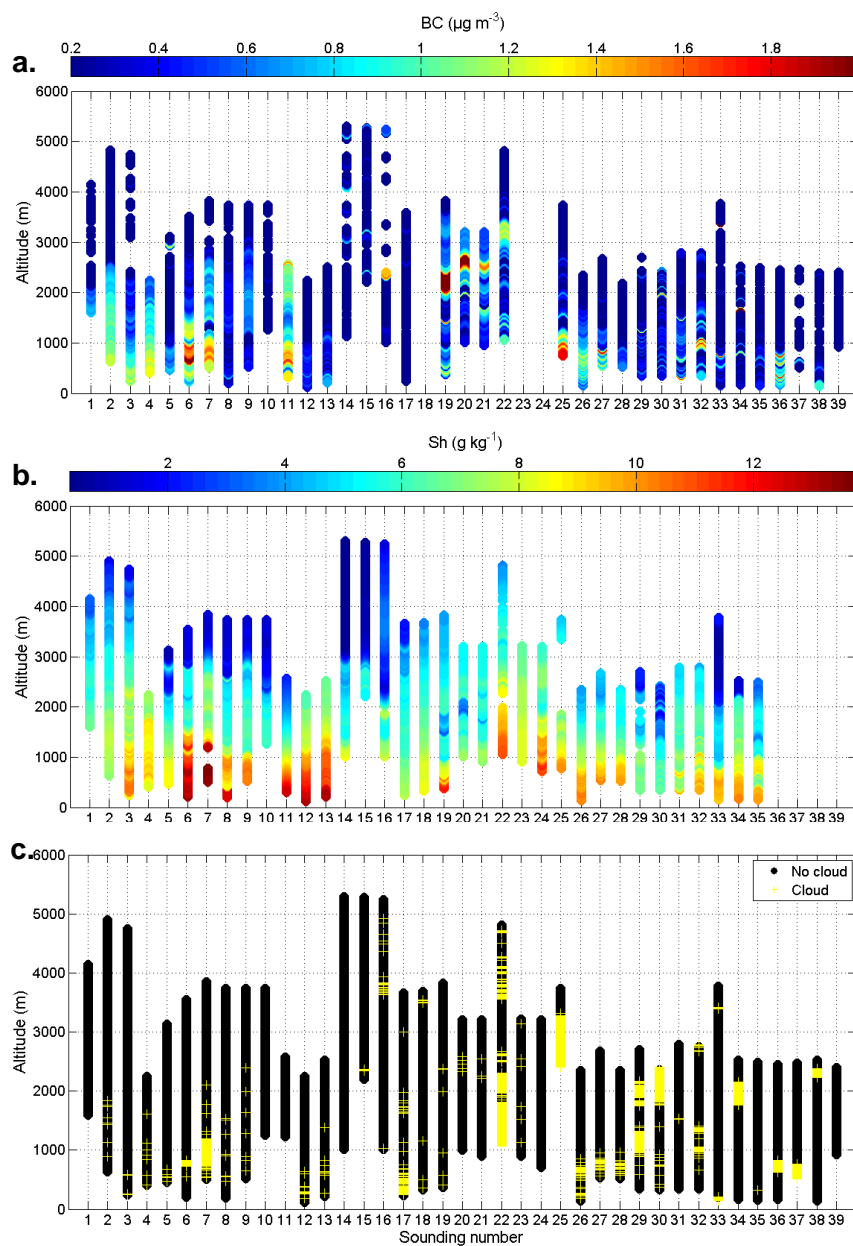


Fig. B1 Profiles of a. BC, b. specific humidity and c. cloud cover during the ATR-42 soundings performed above the sea.