Response to reviewer comments for the paper "Effect of local and remote sources and new particle formation events on the activation properties of cloud condensation nuclei in the Brazilian megacity of São Paulo".

We thank the reviewer for valuable suggestions to improve our manuscript. We agree with the comments and made all the suggested modifications to our revised manuscript. Our responses to each of the comments (in black color) are provided below in blue color. We have highlighted the newly added text with a yellow color in the revised manuscript.

1. A major conclusion of the work is "local traffic emissions showed higher influence on activation parameters than remote sources". However, from Fig 8 to 11, I cannot see any data/information related to traffic source. From Fig 11, I will even conclude that remote sources (sea salt /biomass burning) had more significant influence on AR and Dact. These lead to a more serious problem of this work: the influencing sources of CCN were not clearly classified and many terms were used arbitrarily: diurnal, nocturnal, sea salt, biomass burning, NPF, non-NPF and traffic, the scopes of which overlap with each other. For example, in fig 11 and 10, why are diurnal/nocturnal and sea salt/biomburn put in the same figure? I think "diurnal" aerosols also received contributions from sea salt and biomass burning. Does "diurnal" here include both NPF and non-NPF? In Fig 11a, Fig 8b: "average" of what?

I strongly suggest the authors to state clearly which period (day or night) was dominated by which source. This could be possibly added in Table 1, since there are only 14 observation days. Then CCN properties should be calculated again according to the classification of influencing sources.

Response: In fact,we did not show information related to vehicular traffic in the figs. 8 to 11; hourly NOx average concentrations were added in figs. 9b and 1010, while NOx and CO mean in fig. 12b. The NOx and CO were measured by CETESB in a monitoring station at Marginal Tietê main road, which is distant about 4Km from the sampling site. The variability of NOx in this station is mainly associated with vehicular traffic, once fuel combustion of vehicles is the predominant source in this region. As can be seen in figs. 9b and 10c, NOx increase occurred during a first rush hour in the morning, which is clearly related with a decrease of particles diameter mode and AR (fig. 9b), and also with the increase of activation diameter (fig. 10c). Although the NOx decrease after midday is not associated with the decrease of traffic but with photochemical reactions and O_3 formation, briefly commented in Section 3.2, which contribute for secondary aerosol production. Therefore, NOx data corroborate the discussion presented in the next paragraph and included on page 15 in lines 13-19, which associated decrease and lower AR values during the day with vehicular traffic and secondary aerosol nucleation mainly.

The variability in NOx and CO concentrations measured at the Marginal Tietê is mainly associated with vehicular traffic, because fuel combustion is the predominant source of such pollutants in the MASP.As can be seen in Fig. 9b and Fig. 10c, an increase in NOx occurred during the morning rush and was clearly related to the decrease inAR and particle diameter mode (Fig. 9b), as well as to the increase in Dact (Fig. 10c). The NOx decrease after midday was not associated with the decrease in traffic volume but with photochemical reactions and secondary formation of pollutants, as previously discussed. We believe that the NOx and O3 variability corroborates our assumption, explaining the decrease in AR values during the diurnal period in the presence of high vehicular traffic volume and secondary aerosol formation.

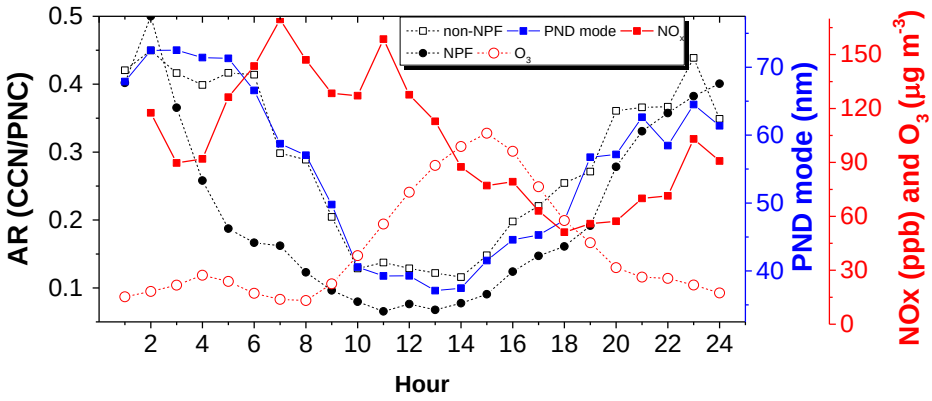


Figure 9.(b) - Mean hourly AR (at a supersaturation of 0.4%) on days with and without NPF events, plotted together with particle size mode, nitrogen oxides (NO_x) and O_3 in order to evaluate the effect of vehicle emissions on particle size and AR. The AR values for nocturnal and diurnal showed clear differences, whereas days with NPF events presented slightly lower values than did days without. Two processes contribute to particle size and AR decrease, namely primary aerosol from vehicular traffic (NO_x) and secondary NPF formed in the afternoon. PND – particle number distribution.

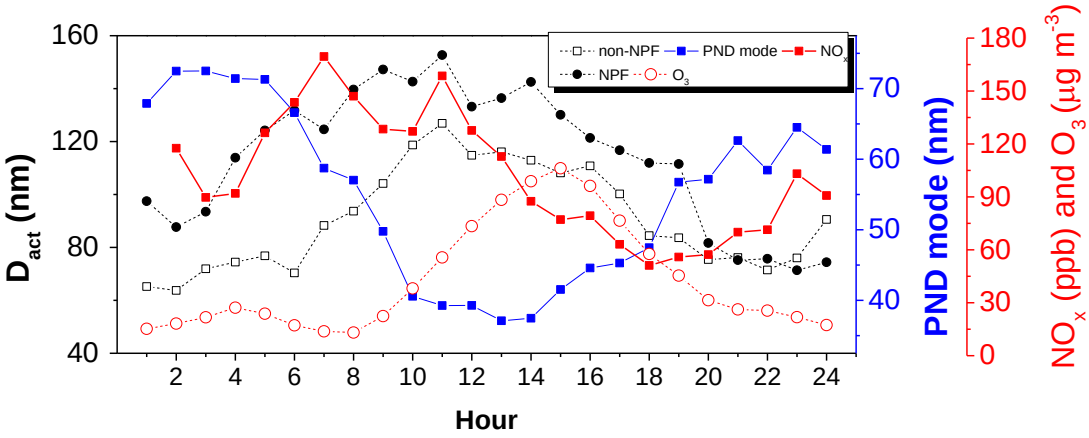


Figure 10. (c) - Time series of D_{act} on days with and without new particle formation (NPF) events, plotted together with particle size mode, NO_x and O_3. The D_{act} values were calculated for supersaturationof 0.4%. Biom burn – biomass burning; PND – particle number distribution.

We agree that sources of CCN was not clearly described, in order to improve the figs. 11 and 12, and show the influence of each remote source contribution, predominant in different periods, was marked in Table 1 the days under remote source and NPF events, based on sections 3.3 and 3.2 respectively. Local sources represented by vehicular emission, industrial and other sources were not showed on the table 1, because this source present daily emission, being a background condition for urban aerosol at SPMA, consequently local sources apportionment were not discriminated in this study.

Table 1. Diurnal data for meteorological parameters(temperature, relative humidity and rain), particle number concentration and cloud condensation nuclei during the period evaluated (19 August to 3 September, 2014). Peak diameter was calculated by size distribution mode. The last column, represent periods with NPF events and remote sources influence on MASP atmosphere.

Day		Date	Temperature (ºC)	RH (%)	Rain	PNC (cm^{-3})		Dp (nm)	Cloud Condensation Nuclei (mean ± SD)					Remote Source* and NPF	
			mean (range)	mean (range)	(mm)	mean ± SD	(range)		SS = 0.2%	SS = 0.4%	SS = 0.6%	SS = 0.8%	SS = 1.0%	Diurnal	Nocturnal
1	Tue	8/19/14	15 (11 - 20)	83 (64 - 96)	0.0	7654 ± 4582	1874 - 16607	50	1688 ± 569	1809 ± 624	2270 ± 830	2555 ± 965	2811 ± 1029		
2	Wed	8/20/14	16 (11 - 23)	80 (43 - 98)	0.0	13086 ± 8242	3335 - 35398	45	2240 ± 780	2385 ± 961	2969 ± 1191	3431 ± 1371	3770 ± 1611		
3	Thu	8/21/14	18 (13 - 27)	70 (32 - 95)	0.0	14133 ± 7647	3360 - 27144	54	2683 ± 1019	3082 ± 1451	3726 ± 1715	4201 ± 1831	4562 ± 1933		
4	Fri	8/22/14	19 (12 - 28)	62 (30 - 92)	0.0	15921 ± 7935	5529 - 35508	73	3368 ± 2664	2487 ± 1034	3208 ± 1512	3699 ± 1766	3960 ± 1890		
5	Sat	8/23/14	20 (13 - 28)	58 (28 - 87)	0.0	15923 ± 7644	5595 - 32061	72	3889 ± 752	4708 ± 1309	5633 ± 1588	6296 ± 1800	6860 ± 1894		
6	Tue	8/26/14	19 (16 - 23)	76 (48 - 96)	1.3	13091 ± 4965	5758 - 26105	92	2765 ± 411	5135 ± 1908	5961 ± 2242	6343 ± 2336	6678 ± 2522		
7	Wed	8/27/14	15 (10 - 20)	79 (55 - 93)	0.1	8627 ± 4965	1924 - 20416	50	1103 ± 549	1444 ± 954	1822 ± 1304	2229 ± 1405	2507 ± 1592		
8	Thu	8/28/14	13 (10 - 18)	84 (62 - 96)	0.0	11313 ± 6449	4920 - 25556	42	1432 ± 438	2103 ± 1181	2672 ± 1388	3117 ± 1536	3538 ± 1702		
9	Fri	8/29/14	13 (12 - 16)	85 (74 - 91)	0.0	9873 ± 3751	5170 - 17513	37	1609 ± 275	2201 ± 529	2747 ± 723	3146 ± 823	3444 ± 861		
10	Sat	8/30/14	16 (12 - 22)	80 (48 - 94)	0.0	12601 ± 8315	4486 - 30528	53	1945 ± 384	3144 ± 1142	3856 ± 1380	4304 ± 1632	4734 ± 1743		
11	Sun	8/31/14	18 (14 - 30)	83 (30 - 96)	2.2	7078 ± 4543	2781 - 18794	63	1573 ± 679	2144 ± 998	2525 ± 1159	2800 ± 1277	3036 ± 1366		
12	Mon	9/1/14	19 (15 - 26)	75 (48 - 95)	0.1	11014 ± 8580	2196 - 31674	38	782 ± 661	1315 ± 1011	1959 ± 1273	2336 ± 1263	2757 ± 1510		
13	Tue	9/2/14	19 (14 - 27)	79 (46 - 96)	3.6	14842 ± 10075	2294 - 32152	67	2138 ± 661	3861 ± 1564	4718 ± 1922	5281 ± 2073	5757 ± 2135		
14	Wed	9/3/14	18 (15 - 25)	80 (57 - 96)	2.2	7713 ± 7303	1347 - 26928	29	609 ± 328	727 ± 428	981 ± 580	1237 ± 701	1523 ± 834		
Mean	-	-	**17 (10 - 30)**	**79 (28 - 98)**	**0.9**	**11634 ± 3077**	**1347 - 35508**	**55**	**1987 ± 942**	**2610 ± 1264**	**3218 ± 1435**	**3641 ± 1512**	**3996 ± 1572**		

RH – relative humidity; PNC – particle number concentration; Dp – peak diameter; NPF – new particle formation; SS – supersaturation; SD – standard deviation.

* Emissions from local sources (vehicular traffic, industrial activity, etc.) occur daily, primarily during the diurnal period, and such sources were therefore excluded.

Days with NPF events	
Remote source apportionment	
Sea salt	Biomass burning

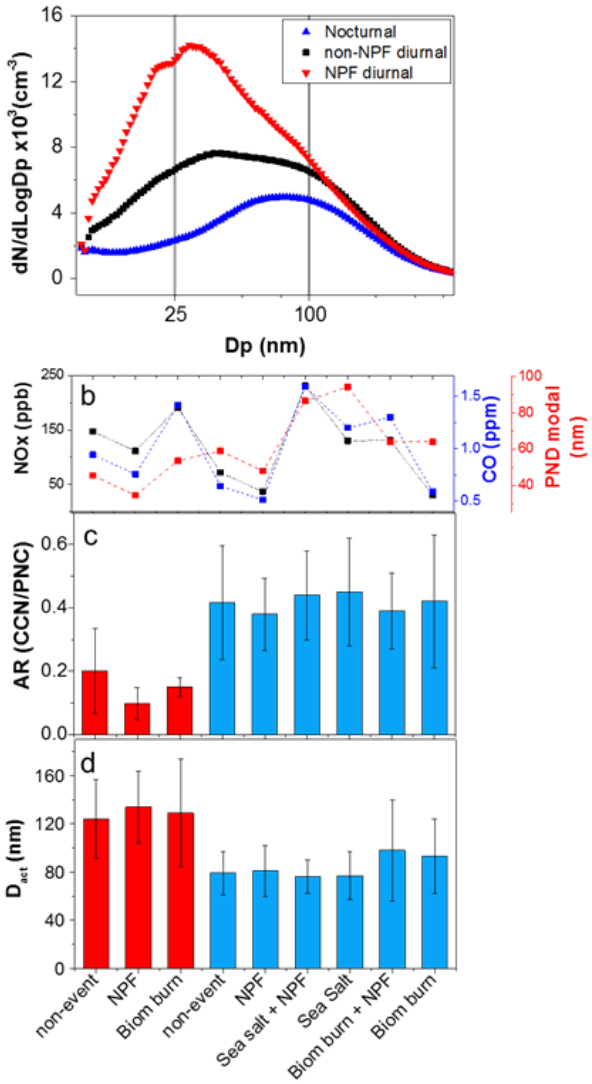
Based on days with and without the NPF as well as remote source events, CCN activation properties were recalculated, as shown in figs. 12c and d. The CCN activation properties related to event and non-event days were calculated for diurnal and nocturnal periods and represented in red and blue colors respectively. In addition, were separated means of nocturnal periods after NPF and non-NPF days, in order to evaluate the effects of particles formed during NPF after growth and also exclude this possible effect during biomass burning and sea salt events. Another, information is related to vehicular traffic variability, which was represented by NO_x and CO in Fig. 12b. New discussion related to Fig.12 was included on page 4 in lines 6-26 and also showed on the next two paragraphs.

On the basis of our data for diurnal and nocturnal periods with and without NPF and remote source events, we calculated the mean AR and Dact for specific periods (Fig. 12c and Fig. 12d), in order to identifythe influence thatdifferent events have onCCN activation. The CCN activation properties related to event and non-event days were calculated for the diurnal and nocturnal periods. The variability for diurnal and nocturnal periods was taken into account, and the events were calculated for each period. In addition, we calculated the mean AR and Dactvalues fornocturnal periods after NPF and non-NPF days,in order to evaluate the effects of NPF on CCN activation. Another important information is the variability in the volume of vehicular traffic, asestimated from the meanNOx and CO values(Fig. 12b), which was previouslyassociated with decreasedCCN activation. The overestimation of the volume ofvehicular traffic can result in the underestimation of effects of remote source events onCCN activation.

As can be seen in Fig.12c, diurnal periods with NPF showed lower AR values than did biomassburning and non-event days. However, AR values were similar among nights after NPF with those non-event and sea salt influence nights and also biomass burning after NPF events. The nocturnal increase in AR after NPF events was related to particle growth (Fig. 12c). Lower AR for nocturnal periods with sea-salt

influence after NPF can be attributed to smaller particle size, whereas sea-salt events in these nocturnal periods occurred after low-O3 NPF events (on days 7 and 8). As previously discussed, low-O3 NPFevents were characterised by the formation ofparticles that were smaller than those formed during high-O3NPFevents. The effect of another sea-salt event (day 4) onCCN activation might have been underestimated due tohigh traffic volume during thenocturnal period, as indicated by high concentrations of NOx and CO. The AR values were slightly lower during diurnal and nocturnal periods with biomass burning plumes only (i.e. without NPF events) than during those with non-events. In addition, during the nocturnal periods, the biomass-burning events promotedslightly higher Dact values than did NPF and sea salt, which is attributable to lower particle hygroscopicity. Although there was a lack of statistical significance, weobserved atendency forbiomass burningaffected air masses arriving at the MASP to decrease the activation properties.

Figure 12. (a) Average diurnal and nocturnal particle size.Average of CO, NOxand modal particle size(b), theactivated ratio (c) and diameter activation (d) for diurnal and nocturnal periods, as well during NPF, biomass burning and sea salt events.The diurnal period is showed in red and nocturnal in blue.Average for event-NPF were calculated for nights after NPF events.



2. The current source apportionment is not persuasive and could be a weakness of this work. Three sources were identified: biomass burning, seasalt, and vehicle emission. Industrial source was mentioned a few times in the manuscript, but was omitted in the data analysis. Then the “diurnal” is equivalent, by the authors, to local vehicular emission. It is not shown in the text what elements were analyzed with X-ray Fluorescence. Will these elements together with BC allow a more precise source apportionment?

Response: Some paragraphs were rephrased in section 3.3 in order to improve discussion, additionally title of this session was changed to **apportionment of biomass burning and sea salt remote sources,** to state more clearly the objectives of this section, related to the identification of biomass burning and sea-salt influence associated with local pollution sources for MASP aerosol.

In the next two paragraphs follow examples of reviewed paragraphs of section 3.3.

A combination of lidar, HYSPLIT trajectory and size-distributed chemical composition(Na, Ca, Ti, K, Cl P, Fe, Mn, Pb, Cu, Zn, S and BC) analyses were used in order to apportion the contribution of sea-salt and biomass burning withinthe air masses arriving at the MASP. Over the past 30 years, variousstudies conducted in the MASP have employed receptor modelling and aerosol chemical composition analysis in order to identify the main sources of atmospheric pollutants (Bouéres and Orsini, 1981; Andrade et al., 1994; Castanho and Artaxo, 2001; Sanchez-Ccoyllo and Andrade, 2002; Sanchez-Ccoyllo et al., 2008; Andrade et al., 2012). Those studies have determinedthat vehicle emissions account for 50–60% of fine particles, thus constitutingthe main source, followed by oil boilers (accounting for 20–40%), road dust (accounting for 10–30%), industrial emissions(accounting for 10–20%), biomass burning (accounting for 10–20%) and construction activities (accounting for ~10%). Some studies carried on in the MASP evaluated the elemental profiles of the principal urban pollution sources, characterising observed elements such as Mn, Pt, Ni, Cu, Pb, Cr and Zn as markers of gasoline and alcohol emissions (Silva et al.,2010). Diesel burning byheavy-duty vehicles is associated mainlywith BC and S. Suspended road dust, characterised inside and outside road tunnels, has been found to becomprise BC, Si, Al, Fe, Ca, Mg, K, Ti and S, which denotes a mixture of soil, pavement abrasion, tire wear, brakewear and vehicular emission (Hetem and Andrade 2016). In a general approach, other studies found BC to be related to biomass burning;Na and Cl to be related to sea-salt contribution; and Fe, Cu, Zn, Cr, Pb and Ni to be related to industrial emissions (Bzdek et al., 2012; Calvo et al., 2013; Taiwo et al., 2014).

Local pollution sources presents acontinuous contribution to atmospheric aerosol, functioning as background toMASP aerosol, emission of which are higher emission during the diurnal period and lowerduring nocturnal period, as demonstrated by the PNC variability (Fig. 2). The objective of this study was not todiscriminate amongthe main local sources but rather to identify periods during which remote sources were detectable, in order to evaluate how CCN activation properties are affected by such sources, in association with local sources, which cannot be excluded during urban aerosol measurements.

3. Some judgments made in the abstract are not well supported and probably biased. “weak effects of sea-salt and biomass burning aerosols could be observed on activation parameters as sea-salt showed a positive feedback” "vehicular traffic presented most negative effect on CCN activation." “NPF events showed a negative feedback to CCN activation.

First, it was not defined in the text what is negative feedback or positive feedback?

compared with what? second, from the Figures I would say sea salt has significant feedback on CCN parameters , but why is the effect claimed to be “weak”? third, it is generally accepted that NPF will eventually has positive contribution to CCN number. it is thus misleading to say NPF showed negative feedback to CCN activation.

Response: The abstract was rewritten in order to correct some not well supported or biassed information as follow.

Atmospheric aerosol is the primary source of cloud condensation nuclei (CCN). The microphysics and chemical composition of aerosols can affect cloud development and the precipitation process. Among studies conducted in Latin America, only a handful have reported the impact of urban aerosol on CCN activation parameters such as activation ratio (AR) and activation diameter (Dact). With over 20 million inhabitants, the Metropolitan Area of São Paulo (MASP) is the largest megacity in South America. To our knowledge, this is the first study to assess the impact that remote sources and new particle formation (NPF) events have on CCN activation properties in a South American megacity. The measurements were conducted in the MASP between August and September 2014. We measured the CCN within the 0.2–1.0% range of supersaturation, together with particle number concentration (PNC) and particle number distribution (PND), as well as trace-element concentrations and black carbon (BC). NPF events were identified on 35% of the sampling days. Combining TEC and BC data with an aerosol profile from Lidarand HYSPLITmodel analyses allowed us to identify the contribution of sea salt and biomass burning from remote regions on 28% and 21% of the sampling days, respectively. The AR and Dact parameters showed distinct patterns for diurnal and nocturnal periods. For example, CCN activation was lower during the diurnal periods than during the nocturnal periods, a pattern that was found to be associated mainly with local road traffic emissions. A decrease in CCN activation was observed on the NPF event days, mainly due to high concentrations of particles with smaller diameters. We also found that aerosols from sea salt and biomass burning had minor effects on Dact. For example, nights with biomass burning showed slightly higher Dactvalues than did non-event nights. Our results show that particulate matter from local pollution sources, mainly local road traffic emissions, has a greater effect on CCN activation parameters than those from remote sources.

4. Activation Ratio is discussed in this work and shown to have a negative "feedback". But what about CCN number increase/enhancement due to a NPF event? how is it when compared to non-NPF or local/remote episodes?

Response: In order to account CCN number enhancement during NPF, days were plotted CCN number mean for NPF and non-NPF days in Figure S2, which was included on Supplementary Information. Could be observed the enhancement of CCN number at nights after NPF events (Fig. S2), caused by th growth of high particle number formed during NPF, being this observation supported by previous studies by experimental measurements or modelling (Sihto et al., 2011). These new text was included in the discussions (page 14, lines 17-21) as follow.

In the present study,the AR increased substantially after each NPF event,although it was still lower than that observed on thenon-event days. In addition, weobserved increase in the number of CCN during thenocturnal periods after NPF events, as showed in Supplementary Information (Fig. S3), attributed tothe high rate of growth amongtheparticlesformed during NPF, an observation that is supported by previous studies (Sihto et al., 2011; Yue et al., 2011; Peng et al., 2014).

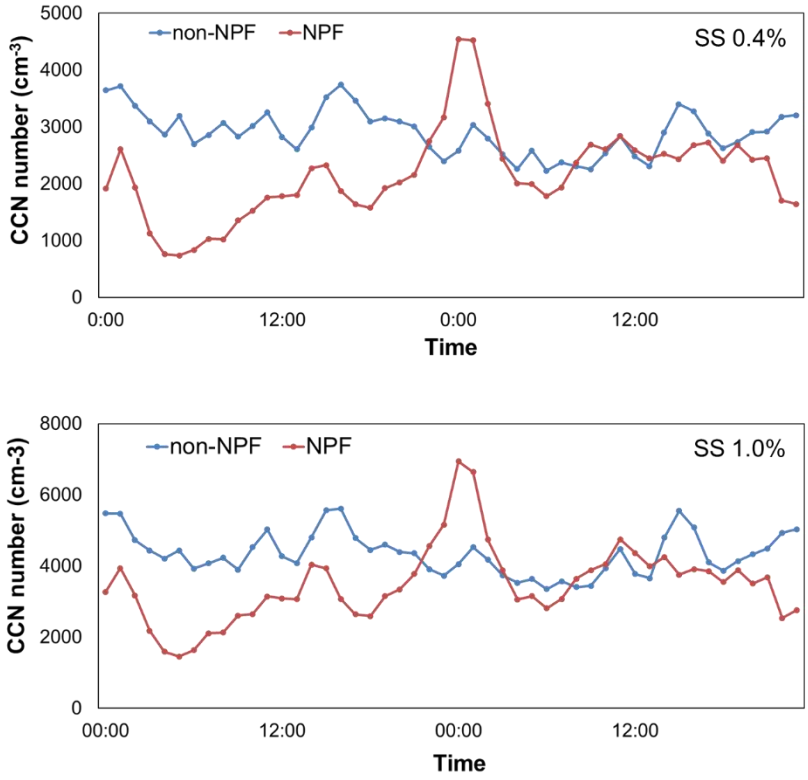


Figure S2. CCN number concentrations during NPF and non-NPF days. CCN averages were calculated for the48h period, being the first day select as NPF or non-NPF, while the second day was not considerate in this classification. During NPF days the CCN number is lower than non-NPF days in agreement with theprevious discussion about AR showed in Fig. 8. In this figure can be seen clearly the enhancement of CCN number for NPF days in comparison with non-NPF followed by similar values on the second day.

5. Other specific comments include:

1. There are many very short paragraphs in the text (like in Section 3.1 and 3.3). They should be reorganized.

2. Some terminology may not be used correctly: “Diurnal” or “daytime”? Page 2 line 26: hygroscope compounds Page 4 line 18: alcohol vapour Page 10 line 2: “frequency of hourly PNCs in three modes”, "frequency" or “percentage"?

3. Page 4 line 25-29: is the correction factor 1.3 or 1.15?

4. Some literature citations should be moved to Introduction section, like page 8 line 24-28.

5. Page 15 line 20: “It can be observed that nocturnal samples are most related with water soluble species as (NH4)2SO4, SOA, NO3- and marine air than the diurnal aerosol.”——— This is obviously not your observation.

6. There are many other typo- or grammar mistakes in the manuscript.

Response: Other specific comments:

1. The short paragraphs were reorganized improving the text structure.

All the specific comments as well as rewrite or new paragraphs are plotted in yellow crosshatch in the paper revised version.

2. Diurnal and Nocturnal periods are related to events occurred during daytime and night-time respectively. All other indicate corrections were made.

3. The correction factor employed was 1.3, additional test explaining this factor assurance has now been removed to avoid confusion. In addition, the paragraph was rephrased (page 5, lines 20-25) as follow.

Determining PNCs from the DMPS has been found to result in the undercounting of particles during ambient measurements, mainly due to lower DMA transfer probability or deviation in sampling and sheath flow rates (Almeida et al., 2014). Another deviation is related to the different diameter size range of particles measured by the DMPS (10–450 nm) and CCN (<10 µm), which can lead to overestimation of AR values, as calculated from the CCN/PNC ratio. A correction factor of 1.3 was applied to the entire data set in order to correct for undercounting during the measurement of PNCs and for overestimation of the AR. That factor was obtained by linear fitting of scatter plot data (CCN versus PNC) with AR values>1.

4. As suggested, some text and citations were moved for introduction section

5. As suggested, we have re-written this phrase (page 16, lines 28-31) as follow.

Our observations support the assumption that nocturnal samples typically comprise greater concentrations of water soluble species, such as (NH4)2SO4, SOA, NO3−, and of marine air than do diurnal samples. Our findings are also in keeping with those of other studies showing that aged aerosols present high hygroscopicity (Gunthe et al., 2011; Bougiatioti et al., 2011).

6. We have read the text of the manuscript to remove any grammaltical infelicities.