First of all we would like to thank the reviewers for their comments and suggestions. We considered them as explained below and think that the paper has been improved. See the point-to-point response below:

Reply to Reviewer 2

The paper by Wehner et al. discusses helicopter-borne measurements of aerosol particles conducted in the framework of the CARRIBA campaign performed in 2010 and 2011. The main goal of this study was to investigate the influence of clouds on the occurrence of NPF. 91 NPF events were detected using a CPC and a FCPC. Most of them were observed in the vicinity of clouds, suggesting that such cloud regions could favour the NPF processes. Increased UV radiation could especially enhance the oxidation of gazeous compounds which are involved in the formation of new particles. Finally, the authors provide an estimation of the particle growth rate, with values similar and even higher than the values reported for coastal areas.
I recommend the publication of this paper as it provides new measurements of aerosol size distributions and observations of NPF over open ocean, processes which remains poorly documented. However, I have several comments and recommendations which should be addressed in a revised version. Especially, I think that in order to really highlight the fact that NPF could be favoured in the vicinity of clouds, more comparisons with clear sky should be provided. Also, artefacts linked to sampling and other parameters previously shown to influence the formation of new particles should be mentioned.

Specific comments
P12430, L24-25 : The authors quickly claim that they observe NPF : in my opinion, they should also evoke the possibility for a fraction of the small particles they observe to be an artefact, resulting from the fragmentation of cloud droplets impacting the aerosol inlet (Weber et al., 1998).

This is a good point which needs to be considered. We found another paper which has recently been published by Craig et al. (2013) dealing with the topic of droplet shattering. In the abstract they summarize the different effects: 1. Aerodynamic breakup of droplets occurs when the Weber number of a droplet exceeds a critical value and depends on the droplet size and the magnitude of the relative motion of the droplet and the local air mass. Impaction breakup of a droplet occurs when the droplet's impaction breakup parameter , K, which is a combination of Weber and Ohnesorge numbers, exceeds a critical value.
In our case, the true air speed of 20 m s^{-1} is too low to reach the critical value for aerodynamic breakup of droplets. Impaction breakup depends on the droplet size and the true air speed. In our case with the true air speed of 20 m s^{-1} we reach the critical values for droplet diameters of approximately 26 μm. Most of the clouds observed during CARRIBA contained droplets smaller than this critical value (Schmeißner et al., 2015); however, this effect needs to be considered within clouds. Our NPF cases occur mostly out of clouds, some of them have no cloud contact at all, and thus they cannot be the result of an artefact caused by droplet shattering.
Another aspect excluding this type of artefact is that the different CPCs use different inlets: the FCPC has a very short line and the inlet is located on the upper side of ACTOS while the TSI CPC is connected with the same inlet as SMPS and OPC and is located below ACTOS. Thus, we would not expect artefacts to occur at two inlets at the same time.

We add in the manuscript:
"Theoretically, such particles can also be artefacts, created by droplet fragmentation at the inlet (Weber et al., 1998; Craig et al., 2013). In our case with a true airspeed of 20 m s^{-1} aerodynamic breakup will not occur, and impaction breakup would occur only for cloud droplets larger than approximately 26 μm in diameter. Most of the cloud droplets observed

during CARRIBA were smaller (Schmeissner et al., 2015), thus breakup would not occur. Another aspect excluding such artefacts is that most of the events were observed out of clouds. Furthermore, two different inlets were used for the different CPCs, but all events were observed at both CPCs at the same time.”

Unfortunately, the instruments used in the present study only allow the criteria which is used for the detection of NPF to be based on particle concentration, and not on particle size (I will discuss SMPS measurements in a next comment). Thus, I believe that the choice of the threshold concentration (1000 cm^{-3}) should be argued in more detail by the authors.

The threshold value of 1000 cm^{-3} has indeed been chosen arbitrarily. The total number concentrations in the marine boundary layer on these days has been very stable between 100 and 300 cm^{-3}, thus 1000 cm^{-3} was by more than a factor of three above the normal variations.

We added: ”This threshold was arbitrarily chosen but exceeds the background value by more than a factor of three.”

P12431, L5-6: Regarding the sentence “Interestingly, almost all of the NPF events were observed in the vicinity of clouds”. Can you provide the ratio of measurements made far from clouds compared to those close to cloud? Is this a statistically relevant number of cases to support your statement?

Unfortunately we have continuous measurements of irradiance for the campaign in 2011 only. However during this campaign 38 out of 44 cases were directly connected to a cloud and an enhancement in radiation was measured. These 38 NPF events in the vicinity of clouds can be divided into two categories, depending on the relative position of ACTOS to the respective cloud and the solar azimuth angle of the sun. Cases with ACTOS between cloud and sun, such as ACTOS approaching and leaving the illuminated side of an individual cumulus, are characterized by increased reflectivity in the UV spectrum (Vant-Hull, 2007). Cases where ACTOS is in a position above a cloud are included in this category and this first category cover 32 of the observed 38 cases. In the remaining 6 NPF events the cloud is positioned between ACTOS and the sun suggesting that ACTOS and SMART—HELIOS were probing in a cloud shadow. Thus, these cases are associated with increased UV radiation which cannot be explained with the flight direction relative to the cloud-sun-axis alone. Here, maybe 3D effects occur, i.e. the enhanced irradiance is caused by reflections at another part of the cloud.
For obvious cloud-free cases, one opportunity is that the cloud has been dissolved very recently which happens frequently and on very short time scales. Thus, the fact ‘far from a cloud’ could also mean that there was a cloud before. For 11 out of the 91 cases no direct connection to a cloud has been observed or the cloud was more than 300 m away and no cloud below was detected using the camera. But the downward looking camera was not always available and we cannot prove all these cases. For several cases the meteorological parameters show a cloud-like behavior and one would assume that a cloud may have just disappeared. That means, for most cases we cannot exclude an influence of clouds, therefore, we decided to focus our study on the cases clearly connected with clouds. But of course, there might be other parameters such as turbulent mixing which is discussed below and in the conclusions of the manuscript.
In summary, we cannot answer this question, because we cannot exclude cloud influence for cases looking like ‘far from a cloud’ on the first view. We can confirm only a connection between cloud, irradiance and NPF, but we cannot exclude the opposite.

We modified the sentence on page 12436:”Together with the fact that 38 out of 44 cases of NPF are directly related to clouds the increase of $F_{\lambda}^{\uparrow}$ seems to be an important factor for particle production in cloudy regions.”

to
"During this campaign 38 out of 44 cases were directly connected to a cloud and an enhanced irradiance has been measured. These 38 NPF events in the vicinity of clouds can be additionally divided into two categories, depending on the relative position of ACTOS to the respective cloud and the solar azimuth angle of the sun. Cases with ACTOS between cloud and sun, such as ACTOS approaching and leaving the illuminated side of an individual cumulus, are characterized by increased reflectivity in the UV spectrum (Vant-Hull, 2007). Cases where ACTOS is in a position above a cloud are included in this category, which comprises 32 of the observed 38 NPF events associated with clouds. In the remaining 6 NPF events the cloud is positioned between ACTOS and the sun suggesting that ACTOS and SMART—HELIOS were probing in a cloud shadow. Thus, these cases are associated with increased UV radiation which cannot be explained with the flight direction relative to the cloud-sun-axis alone. Here, maybe 3D effects occur, i.e. the enhanced irradiance is caused by reflections at another part of the cloud.
Together with the fact that 38 out of 44 cloud cases are directly connected to an increase in $F_{\lambda}^{\uparrow}$ the UV irradiance seems to be an important factor for new particle production in cloudy regions."

Also, when considering Fig. 2 from Siebert et al. (2013), one could think that the altitude ranges investigated in clear sky conditions were lower compared to in-cloud measurements, which can also influence the occurrence of NPF. Can you clearly show that at similar altitudes, the presence of clouds favour the NPF process compared to clear sky?

As stated above for 11 out of 91 cases we did not find a clear connection to a cloud or the distance to the next cloud was at least 300 m. This does not exclude any influence of clouds. For the majority of cases clouds were present and also enhanced irradiance compared to cloud-free regions has been observed. We had two measurement flights without any cloud contact of ACTOS, but clouds were present in the area. During each of these flights one NPF event without visible cloud contact has been observed. During other flights with cloud contact between 1 and 13 NPF events have been observed. We have no measurements taken without any cloud in the area. Thus we cannot compare these different conditions directly. However, we did not observe any new particle formation in the sub-cloud layer, i.e. the cloud-free area below the clouds. And the cloud layer is characterized by the occurrence of clouds during our campaigns. Thus we cannot compare to cloud-free conditions in these altitudes.

P12431, L12-13: Could you please develop your explanation supporting the fact that artificial particle production can be excluded?

We think this is dealing with the same topic as the fragmentation of droplets at the aerosol inlet which is answered above and manuscript modifications are given there. One general argument is that we use two different inlets at different positions on ACTOS, this excludes artificial particle production at the inlet.

P12431-12432, sections 4.1-4.2: Again, in order to better evaluate the role of the cloud and the influence of its proximity, can you compare in-cloud/clear-sky measurements performed at similar altitudes? Can you identify a threshold distance cloud-helicopter from which the influence of the cloud becomes significant? Can you calculate a "cloud free" and a "cloud" NPF frequency as additional information to the global frequency of 83%?

The total number of observed events was 91, while for 11 of them no direct connection to a cloud from our data could be obtained. But partly, no downward looking camera was

available to identify a potential cloud below, thus we cannot exclude the cloud influence for these cases. For some of the 2h-measurement flights only one NPF event of a few seconds was found. Thus the overall frequency of these events is low.
The identification of such a threshold distance is difficult because for some cases we cannot exclude that there was a cloud below or there has been a cloud before which has been dissolved recently. As stated above, we conclude that enhanced irradiance due to cloud effects plays an important role but some events do not clearly fulfill these criteria. However, we do not think that enhanced irradiance alone is sufficient for new particle production. The connection with turbulent mixing at cloud edges causing droplet evaporation and local supersaturation creates perfect conditions for new particle formation and growth.

Concluding we can state that we never found NPF in the sub-cloud layer which was always cloud-free. The Cloud layer was never cloud-free, thus we cannot compare different conditions here.

We added in the conclusions: "No new particle formation events have been observed in the always cloud-free sub-cloud layer, while cumulus clouds were present during all flights in the cloud layer."

Also, I think that vertical profiles performed at the beginning of the flights should be used in order to highlight potential preferential altitudes where increased concentrations of small particles are found, as recently shown in Rose et al. (2015). Despite the fact that the present study is focused on the influence of the cloud, this could help to introduce other parameters that were previously reported to affect the formation of small particles and which are not mentioned in the current version of the manuscript: cleaner conditions found at higher altitudes (e.g. Manninen et al., 2010), decreased temperature (Young et al., 2007), mixing of two air parcels with different characteristics (Khosrawi and Konopka, 2003)...

This is a really good point. Yes, we do have vertical profiles available from every flight, thus also from the 24 measurement flights where NPF were observed. In general the structure is very similar between the different days. A deeper look into the specific heights of the individual events shows that the NPF occur between heights of 600 m and 2200 m above ground, but always within the cloud layer, i.e. above the sub-cloud layer and below the trade inversion (see also Siebert et al., 2013). The interesting point is that for most cases all events of the flight occur in a very narrow height range, i.e. within 200 – 300 m. In some cases these layers are connected with a gradient in the particle number concentration, i.e. cleaner air might be involved. However, the structure of each individual profile varies for each case and it is beyond the scope of this study to add 24 profiles as well. Example profiles are given also in Siebert et al. (2013). On both days discussed NPF was also observed: on April 20 between 900 and 1000 m height and on April 22 around 1500 m height. In both cases, these were measurement altitudes where variations in the total particle number concentration were observed.
But the vertical structure between both days is very different, on April 22, the sub-cloud layer is significantly cleaner than the air aloft while on April the mean concentration is higher in the sub-cloud layer and similar to that in the cloud layer, except the interfacial layer in between shows a significant minimum. Thus, mixing with cleaner air may play a role for the NPF. This mixing is probably enhanced do to the increased turbulence at cloud edges where mixing of air parcels with different characteristics is continuously going on.
This is consistent with the results from Koshrawi and Konopka (2003), who showed that for the tropopause region nucleation and condensation may increase significantly due to turbulent mixing.
As a final point it needs to be mentioned that enhanced UV irradiance produces a sufficient amount of precursor gases required for nucleation and growth (e.g. Weber et al., 2001).

We added in section 4.3, page 12433:" NPF events have been observed at heights between 600 m and 2200 m, always within the cloud layer. Interestingly, NPF during individual measurement flights occurred within a narrow height range, i.e. within 200 - 300 m. These height ranges were often connected to a change in the number concentration in the vertical profile. Thus mixing with cleaner air may play a role for the NPF event."
And in the conclusions:" From vertical profiles we found variations in particle number concentrations in the height where NPF occurred for the majority of cases, thus mixing with cleaner air may also be one aspect forcing nucleation and growth. It is likely that the connection of the different issues is required: enhanced UV radiation to produce the precursor gases, cloud edges as a region with increased turbulence, and mixing with cleaner air reducing the condensational sink."

P12432, L1-2: I think that it is confusing to affirm that "there are no anthropogenic sources". If NPF is promoted in the vicinity of clouds, as suggested by the authors, we can believe that a significant fraction of the gaseous compounds involved in the particle formation are transported from lower altitudes through convection processes associated with clouds (and then further oxidized to more condensable species). In such a case, part of these compounds might have an anthropogenic signature.

This is correct. What we mean was that there are no sources for particulate emissions in the regions which exclude primarily emitted particles as a source for the particle bursts. However, the precursor gases leading to new particle formation may have an anthropogenic origin.

We changed the sentence in the manuscript to: "There are no anthropogenic particle sources…"

P12432, L11-13: It is true that SMPS size distributions show different signatures, suggesting the occurrence of NPF. However, how can you explain the "closed" shape of the size distribution when NPF is believed to occur?

The size distribution is one example. The SMPS is scanning over a diameter range of 6 - 230 nm within 120 seconds. Thus, during a typical NPF event of a few seconds, the SMPS is only able to sample within a very narrow size band. Due to a smoothing effect caused by a combination of continuous scanning (of different sizes) and a non-zero response time of the SMPS-internal CPC, the detected size distribution appears to be closed.
If this narrow size band is affected by NPF, increased concentrations can be also found in the NSD, but if the respective diameter range is not affected the NSD looks like every other before or after the event. However, there was not a single NSD found during an event with a maximum larger than 50 nm, only a few with maximum concentrations below 20 nm (like the one presented in Figure 3). But again, the SMPS measures at one diameter during the event, this is not related to the real maximum in the burst. Of course, there were also NSDs centered on smaller diameters, but this does not mean that the particles were smaller. The only thing we can conclude it that the particles were above our detection limit of 7 nm.

We added:
"However, the presented NSD is only representative of one NPF event and different events might exhibit a different distribution. Furthermore the maximum of the NSD does not show the real maximum of the NPF event, because the SMPS was measuring in a certain diameter range, defined by the measurement program."

P12432, L13-14: Considering the fact that the detection limit of the FCPC is 7 nm, I do not understand the following sentence: "Around 20 s later between 52 940 and 52 950 sod the event was observed in N_{FCPC} corresponding to a diameter between 10 and 20 nm".

This is closely connected to the point above. The SMPS scan starts at the lowest diameter of 6 nm and scans slowly upwards in voltage and diameter while the diameters are lognormally equally distributed. Thus, my estimate was that 20 s later, the selected diameter is in the range between 10 and 20 nm, 120 s later it reaches the upper end of 230 nm.

We added: "…. corresponding to a selected diameter in the SMPS between 10 and 20 nm. 120 s later, the SMPS reaches the upper end and measures at 230 nm."

P12436, L1-2: I might have missed the information but if I am right, the wavelength corresponding to $F_\lambda^\uparrow$ shown in Fig. 6-8 is not stated (only in the caption of Fig. 9), which does not ease the understanding of the obvious connection between NPF and increased UV radiation in the vicinity of cloud suggested by the authors.
Also, can you explain such an increase of UV radiation?

Sorry for the missing wavelength, we added this information to the figure caption of Figure 6, as well as in the text, where all presented parameters are introduced.
Cloud surfaces increase the reflectivity in the whole spectrum, particularly in comparison with a darker background, e.g. the ocean. This has been measured by e.g. Eck et al. (1987). Above the ocean they observed a reflectivity of less than 0.1, above clouds the reflectivity varied between 0.52 and 0.76 depending on the cloud type.

We added on Page12434:"The reason is the higher reflectivity of the cloud surface compared to the darker ocean (e.g., Eck et al., 1998)."

P12437, L9: You should also clearly mention the initial size you assume for the clusters.

Within our estimate we really started from nothing, i.e. < 1 nm. With regard to the growth time it does not play a role if we start from 0.4 or 0.8 nm, this has a minor effect.

We modified the sentence to:" Assuming the particles must reach d = 7 nm (starting at a cluster size below 1 nm)…"

P12437, L10-15: The growth rates which are presented here are huge, even higher than what is reported for coastal areas. The authors believe that such growth rates might be explained by ELVOCS, as suggested by Ehn et al. (2014). However, Ehn et al. (2014) mainly discuss how such compounds can be involved in the formation and growth of aerosol particles over forested regions, which are known to house a large pool of biogenic organic compounds. Thus, I would like the authors to balance their last results regarding particle growth and/or give more explanation regarding their relevance.

Similar growth rates have been reported for Mace Head (Kulmala et al., 2004), thus they have been observed also on longer timescales. Another aspect that must be considered is that the growth rates are observed on small temporal and spatial scales, where turbulent mixing may play a major role the growth can be significantly increased. In our case the NPF occurs at cloud edges, i.e. the evaporation of cloud droplets may play a role for the production of precursor gases. These gases are probably transported over long distances from continental regions to the cloud layer in the Caribbean marine boundary layer. However, we can only speculate which gases are the most relevant ones for particle formation and growth. Typical concentrations of sulfuric acid are too low to contribute much to the growth. Thus, probably organics might be involved and it has been demonstrated that ELVOCs may contribute very efficiently to particle growth. Anyhow the saturation vapor pressure of condensing vapors should be very small for effective molecular flux into growing newly formed particles, and ELVOCs fulfill these criteria. However, forest areas are a well-known *source for ELVOCs but the questions are if there are other sources and how far ELVOCs might be transported without any significant change or modification. One would not expect them to keep their structure while passing the aqueous phase. On the other hand ELVOC precursors could live inside clouds and convection could transport ELVOC precursors to cloud edges as well as SO2 as SA precursor. The solar radiation outside the cloud and precursors coming from evaporating cloud are able to make photochemical reactor efficient enough to produce ELVOCs.*

We added: "..because the saturation vapor pressure of condensing vapors should be very small for effective molecular flux into growing newly formed particles, and ELVOCs fulfill these criteria. ELVOC precursors could live inside clouds and convection could transport ELVOC precursors to cloud edges as well as SO_2 *as sulfuric acid precursor. The solar radiation outside the cloud and precursors coming from evaporating cloud are able to make photochemical reactor efficient enough to produce ELVOCs."*

Other comments

P12425, L2-5: Regarding the sentence "On the other hand, clouds strongly influence the incoming solar radiation, thus they may also influence the formation and distribution of aerosol particles, e.g. new particle formation (NPF)". I would suggest to rephrase this sentence to remove the fragment "e.g. new particle formation (NPF)".

We do not think that this part should be removed. Furthermore, we added the word 'by' and modified the fragment to: "e.g. by new particle formation (NPF)".

P12429, L26: <u>M</u>oreover

Done.

P12430, L3: <u>M</u>easurements

Done.

P12431, L10: Reformulate the phrase "near cloud edge of clouds"

Done. Now "near cloud edges"

<u>References:</u>

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