Observations of new particle formation in enhanced UV irradiance zones near cumulus clouds

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 mentionned.

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).

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 treshold concentration (1000 cm^{-3}) should be argued in more detail by the authors.

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

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

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%?

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 mentionned 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)…

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.

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

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".

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?

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

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.

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)".

P12429, L26: Moreover

P12430, L3: Measurements

P12431, L10: Re formulate the phrase “near cloud edge of clouds”

References

Ehn, M., Thornton, J. A., Kleist, E., Sipilä, M., Junninen, H., Pullinen, I., Springer, M., Rubach, F., Tillmann, R., Lee, B., Lopez-Hilfiker, F., Andres, S., Acir, I.-H., Rissanen, M., Jokinen, T., Schobesberger, S., Kangasluoma, J., Kontkanen, J., Nieminen, T., Kurtén, T., Nielsen, L. B., Jørgensen, S., Kjaergaard, H. G., Canagaratna, M., Maso, M. D., Berndt, T., Petäjä, T., Wahner, A., Kerminen, V.-M., Kulmala, M., Worsnop, D. R., Wildt, J. and Mentel, T. F.: A large source of low-volatility secondary organic aerosol, Nature, 506(7489), 476–479, doi:10.1038/nature13032, 2014.

Khosrawi, F. and Konopka, P.: Enhanced particle formation and growth due to mixing processes in the tropopause region, Atmos. Environ., 37(7), 903–910, 2003.

Manninen, H. E., Nieminen, T., Asmi, E., Gagné, S., Häkkinen, S., Lehtipalo, K., Aalto, P., Vana, M., Mirme, A., Mirme, S., Hõrrak, U., Plass-Dülmer, C., Stange, G., Kiss, G., Hoffer, A., Törő, N., Moerman, M., Henzing, B., de Leeuw, G., Brinkenberg, M., Kouvarakis, G. N., Bougiatioti, A., Mihalopoulos, N., O’Dowd, C., Ceburnis, D., Arneth, A., Svenningsson, B., Swietlicki, E., Tarozzi, L., Decesari, S., Facchini, M. C., Birmili, W., Sonntag, A., Wiedensohler, A., Boulon, J., Sellegri, K., Laj, P., Gysel, M., Bukowiecki, N., Weingartner, E., Wehrle, G., Laaksonen, A., Hamed, A., Joutsensaari, J., Petäjä, T., Kerminen, V.-M. and Kulmala, M.: EUCAARI ion spectrometer measurements at 12 European sites – analysis of new particle formation events, Atmos Chem Phys, 10(16), 7907–7927, doi:10.5194/acp-10-7907-2010, 2010.

Rose, C., Sellegri, K., Freney, E., Dupuy, R., Colomb, A., Pichon, J.-M., Ribeiro, M., Bourianne, T., Burnet, F. and Schwarzenboeck, A.: Airborne measurements of new particle formation in the free troposphere above the Mediterranean Sea during the HYMEX campaign, Atmos Chem Phys Discuss, 15(6), 8151–8189, doi:10.5194/acpd-15-8151-2015, 2015.

Siebert, H., Beals, M., Bethke, J., Bierwirth, E., Conrath, T., Dieckmann, K., Ditas, F., Ehrlich, A., Farrell, D., Hartmann, S., Izaguirre, M. A., Katzwinkel, J., Nuijens, L., Roberts, G., Schäfer, M., Shaw, R. A., Schmeissner, T., Serikov, I., Stevens, B., Stratmann, F., Wehner, B., Wendisch, M., Werner, F. and Wex, H.: The fine-scale structure of the trade wind cumuli over Barbados – an introduction to the CARRIBA project, Atmos Chem Phys, 13(19), 10061–10077, doi:10.5194/acp-13-10061-2013, 2013.

Weber, R. J., Clarke, A. D., Litchy, M., Li, J., Kok, G., Schillawski, R. D. and McMurry, P. H.: Spurious aerosol measurements when sampling from aircraft in the vicinity of clouds, J. Geophys. Res., 103(D21), 28337, doi:10.1029/98JD02086, 1998.

Young, L.-H., Benson, D. R., Montanaro, W. M., Lee, S.-H., Pan, L. L., Rogers, D. C., Jensen, J., Stith, J. L., Davis, C. A., Campos, T. L., Bowman, K. P., Cooper, W. A. and Lait, L. R.: Enhanced new particle formation observed in the northern midlatitude tropopause region, J. Geophys. Res. Atmospheres, 112(D10), D10218, doi:10.1029/2006JD008109, 2007.