Review on

„Aircraft-derived particle fluxes distinguish entrainment zone and decoupled layer nucleation in marine boundary layers“

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General comment:

This paper describes aircraft-based observations of aerosol particles, primarily in the size range between 3 and 10 nm, which are assumed to be newly nucleated. Turbulent particle fluxes are estimated using additional parameters and wavelet analysis. The technique is discussed as having advantages over other methods, and the detailed description therefore takes up a large portion of the manuscript. Although the determination of particle fluxes remains a major challenge and is therefore of great significance, in my view the classification and interpretation of the results can and should be improved. A key issue here is the imbalance between the details of the flow determination and the interpretation of the results. To take it to an extreme, the conclusions could already be drawn from the sign of the particle fluxes, since the absolute value is not interpreted in the slightest and thus loses its significance. In contrast, in some places the interpretation of observations of particle formation involves a high degree of speculation. Unfortunately, the work is also not comprehensively contextualized with the help of additional literature on particle formation in marine boundary layers; this becomes particularly evident in the last two chapters (see specific comments on this).

For this reason, this manuscript requires a thorough revision (“major revisions”) regarding the interpretation and contextualization of the observations and analyses.

As this review proceeds, I will offer both general and specific comments. However, I hope that these comments will be received constructively and will contribute to a successful revision of the manuscript, as I believe the measurements are definitely worth analyzing and publishing.

Abstract :

It is somewhat misleading to attempt to deduce the location of new particle formation using the flux of particles in the size range between 3 and 10 nm. Firstly, these particles have already aged and grown, and secondly, the flux only indicates the direction and quantity per time in which particles are transported. Perhaps this should be formulated more clearly.

It has become common practice to use the following phrasing (or something similar): When ultrafine particles in the range of 3 to 10 nm are measured, we refer to them as “freshly nucleated particles," but never as "new particle formation," since this process definitely cannot be observed with these measurements. As I continue reading the manuscript, I notice that you use a similar term later on; perhaps this should simply be clarified a little earlier.

I always find it a bit difficult when people talk about new particle formation on the one hand and CCN on the other; these are completely different modes, and it remains questionable whether locating NPF will lead to a better understanding of the number concentration of CCN. Why not argue that we want to understand the budget of aerosol particles in general? It is not at all clear whether the small particles will ever grow into CCN. I think a couple more sentences could be added here.

Incidentally, vertical profiles of aerosols have already been observed above the same island at almost the same time, which also indicate freshly formed aerosol particles. https://doi.org/10.1175/BAMS-D-19-0191.1 and references in there (see also my comment about the "discussion" and "conclusion" at the end of my coments).

line 106: please specify what you mean with "excellent sensitivity", without numbers or more details such phrases should be avoided

line 116/117: What is the background to this criterion? Why 10% ?

Section 2.3.2
At the beginning of the chapter, you describe the problem of temporal synchronization. But what I find missing is a statement about how you specifically correct the problem in your aircraft measurements. It all remains a little vague. In the second paragraph of this section, you then describe statistical collection errors, which has nothing to do with the title. I also think that the comparison with the mast measurements distracts from your specific problem—I'm actually interested in how you deal with your challenges.

For example, in line 156ff, you first describe the vertical distribution of turbulence intensity and then the sampling time required to determine a turbulent flux with a statistical accuracy of 10%. Each part on its own is fine, but the connection between them is not clear to me. In the next sentence, you refer to the spatial resolution of the measurements as a function of the sampling frequency of a sensor, which has nothing to do with the previous statement—a red line of the story is missing here.

line 161: The statement that 30-minute averages based on mast measurements "easily" fulfill stationarity conditions is somewhat harshly worded. I also don't quite understand why you are venturing onto such thin ice in this context.

Line 167: I don't quite understand the sentence; do the two CPCs now draw their sample air from the same inlet—if so, what is the problem? What do you mean by "same inlet" and "from different positions"—please clarify (maybe a sketch would help here)

Why is the time lag for the two CPCs not constant or did I misunderstand this part? Are the flows in the inlet system not regulated? Or is there even a pressure dependency? This should be clarified in this context, because in general I would assume a constant time lag. Under what conditions do you determine a time lag of 0 or even one second? Shouldn't this have a significant influence on particle flow determination?

I also think that if you discuss figures in such detail, they should appear in the paper itself and not in the supplement. I generally have a problem with discussing figures from the supplement—I would avoid that entirely. Either a figure contributes to the narrative of your manuscript, in which case it belongs in the manuscript, or you mention it without further discussion.

Line 187 ff: In line 189 you mention that the time lag is negligible but in the last sentence you state that for each case individual time lags are determined: i) what do you mean with "case" and ii) why do you have to determine a time lag when it is negligible?

I think the discussion you are having here is necessary, but in some places, it needs to be presented and clarified a little better.

Regarding the spectra in Fig. 1: All spectra are shown up to the Nyquist frequency of 0.5 Hz; while for the vertical wind, the spectrum—especially the one calculated with FFT—certainly flattens out for frequencies above 0.3 Hz and transitions into noise, I am surprised that no flattening can actually be observed for the particle measurements. This somewhat contradicts your statement regarding the CPC behavior – right?

Why can't the integral time scale in Sec 2.5 be determined to estimate the sampling error? Shouldn't that be easy to do using the autocorrelation function?

Line 278: Isn't the response of a CPC also a question of number concentration and therefore of counting statistics? What is the argument for the 3s resolution?

Is there an explanation for the flattening of the wind spectrum? I would have expected slightly better performance from the system.

Perhaps it's also a question of style, but you should really avoid repetition in an academic text; here you start mentioning the advantages of wavelets over FFT again – I would avoid that.

Line 285: what is the "true value" of a flux? I thought the eddy covariance is what direct follows the theory – right?

Line 287/288: I cannot quite follow your argument regarding the high-frequency part of the spectrum. Of course, there are limits to the resolution and representation of the high-frequency components in the spectrum, but these are mostly technical limitations and not a fundamental problem with FFT. There are airborne atmospheric turbulence measurements with high-resolution hot-wire anemometers down to the cm range, and the spectra look absolutely clean until they drop off steeply due to dissipation.

The figure caption of Fig 2 could be expanded a little bit to better understand what is displayed with the different lines.

I think at the end of Sec 2.8, it would be good to mention approximately how large the loss is in determining the flow when you can no longer resolve the integral length scales yourself. This does not anticipate any results from the next chapter, but would simply illustrate how important this correction is.

Starting the results section with a table whose parameters are only described very superficially is not convincing. You begin the chapter by stating that two case studies will be described in more detail, and then start with a table showing six flights—somehow, this doesn't quite fit the picture. Here, too, the table caption is very brief; perhaps I overlooked it, but what does the last column mean?

About Fig 3:
My first impression of this figure is, why do I need all this detailed information? Somehow, there is no convincing motivation for this figure. For example, why is panel d with the chemical components necessary if the topic is particle fluxes? Panel b describes the flight path and the sampling strategy—okay, that's important, but why don't you show a map with the flight path marked on it? No one is going to visualize the flight path based on the coordinates—so I'm not learning anything from panel b at first glance. If you exclude all cloud passages from the analysis, why are they not already marked in panel a)? It is also somewhat confusing that one first looks at the sections with "significant number concentrations" in panel a (shown in blue) and then looks at panel c and realizes that these sections were in clouds and are therefore excluded from further analysis. I suggest simplifying this figure significantly and focusing on the things that are evaluated and analyzed—that would make the figure much more convincing. In its current form, it looks like a "quick look plot" in which readers can search for interesting passages instead of the author highlighting the interesting passages and events. The same holds true for Fig. 6.

About Fig 4:
You are mixing a time series with the three excerpts from vertical profiles here; based on the times for the profiles, it is possible to get a rough idea of how everything is connected, but a little help would be useful. I would also consider whether it would make sense to separate the vertical profiles and time series into two figures. The color scale in the upper figure is somehow unconvincing with regard to the tick labels and very close to the legend of the vertical axis—at first, I thought the color scale belonged to the axis label "Airplane height," but that's just a technical note. In the caption the "Stot" should be "S_{tot}" – right?

Line 373: Regarding the comment on the high particle concentration at 12:18Z: I can't imagine that burst droplets produce particles in the 10 nm size range – but maybe I'm wrong. Could it be possible that you flew into your own exhaust plume? Interestingly, the size distribution directly at the edge of the cloud returns to the more typical distributions before the cloud-free part begins.

line 378 I have difficulty with the statement that a strong temperature gradient is an indicator of an entrainment layer. If there is no process that promotes vertical mixing, there is no entrainment even with a strong gradient. A strong gradient initially prevents vertical mixing, and only either shear flow or downward buoyancy due to cloud top cooling for example can cause mixing. But I agree with you, of course, that a strong temperature gradient combined with increased variance in vertical velocity is an indication of entrainment—it's just worded a little awkwardly at the beginning.

Could you please explain in more detail what exactly you mean by your statement regarding the moisture gradient (lines 386 to 388)? I don't really understand this explanation.

Regarding Figure 5: In principle, I find plots of this kind very illustrative, but unfortunately the flight section with the color-coded particle concentration is so small that it is impossible to relate it to the cloud situation. In the explanations accompanying this figure, the presentation of the observations, explanations, and then interpretations (also in comparison to other studies) are completely mixed up, which does not exactly improve readability. I suggest clearly separating explanations and interpretation.

Another important aspect that becomes particularly clear in Fig. 5: Technically speaking, the manuscript deals with the most accurate determination possible of ultrafine particle fluxes. The fluxes are determined for the two SPE periods selected in Figs. 3 & 4, but the amount of the fluxes is not used at all in the further discussion. To put it somewhat provocatively, wouldn't the sign of the particle flux be sufficient for the following discussion? Is the large difference in the calculated particle flux for the two events discussed at any point?

You are still in the chapter entitled "Results" and are discussing very general possible scenarios in which particle formation can occur. Are there any indications of "cold air outbreaks" over the Azores? Probably not, so in my opinion this discussion belongs in the introduction or discussion section, if anywhere, but not in the results section.

line 426: The discussion about a possible contribution of nucleation mode particles to CCN is also highly speculative and goes far beyond the observed results—so it does not belong here.

In the "Discussion" section, you first state that determining particle fluxes is important for a better understanding of particle formation. However, in the sections from lines 490 to 499 and 500 to 511, you describe in much greater detail the conditions prevailing in the two different regions where you measured the NPF. Although the particle flux itself is mentioned, the sign alone would have sufficed for the conclusion you draw from it. As a reader, I wonder why you undertook this significant technical effort to determine the flux, including a very thorough error analysis—which I greatly appreciate—if the magnitude of the flux plays no role whatsoever in your discussion.

The interpretation of the event's horizontal extent of a few kilometers in terms of "convective rolls," on the other hand, is highly speculative and rather superfluous in this context—why are you venturing onto such thin ice with this?

In the following "Conclusions" chapter, it sounds a bit as if your measurements have turned conventional wisdom regarding nucleation in marine environments on its head—that goes a bit too far: there are very detailed and well-founded observations of small, newly formed particles even over the ocean.

Take, for example, the work of Collin O'Dowd, who regularly observed nucleation at low tide along the Irish coast (Mace Head, https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2000JD000206).

Likewise, the pioneering work of Dave Covert (https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/92JD02074) and many others.

And then there was even a study that dealt with particle fluxes over the same area and almost at the same time as your measurements (https://acp.copernicus.org/articles/22/10007/2022/), which you completely ignored.

How could you present a more convincing explanation of your measurements?
Of course, the mass balance equation for aerosol particles cannot be solved based solely on the particle flux, but one could at least roughly estimate the contribution of small particles to the total concentration; after all, your original motivation for this topic was to highlight the importance of small particles for CCN development. If you want to keep up this motivation, not address this point and conclude the paper with it?