Referee Comment for Manuscript AMT-2010-186 “Characterization of a new fast mixing type CPC and its application for atmospheric particle measurements” Wehner et al.

General Comments

The authors are to be congratulated on developing a new version of the mixing CPC with outstanding performance characteristics, particularly in terms of its fast response. The paper is well-written and easy to read. However, the analysis of the performance data leaves much to be desired.

The form of the equation used to model the counting efficiency has too many adjustable parameters which lead to an excellent fit of the data but otherwise create significant problems. The coefficients in Table 1 are insufficiently precise and do not allow for any interpolation or extrapolation to other saturator temperatures.

The analysis of the FCPC response as shown in Fig. 7 was done very poorly with the result that the mixing time was probably significantly overestimated. In light of this, the conclusions drawn from the analysis of Fig. 8 are quite questionable. The authors appear to be confused concerning the possible sources of limitation of the temporal resolution of the FCPC and how to analyze them. They also totally ignore the limitation imposed by the counting gate width.

These and other issues are elucidated below.

Specific Comments

page 5908, line 26 to p. 5909, ln. 4: Using the terminology as used by Wang et al. (2002), your primary reference, do not confuse “plumbing time” and “mixing time”. The former is a uniform delay easily compensated for in SMPS data analysis. The latter is the characteristic time τ in your Eq. (2) that leads to the exponential decay of the CPC response to a step change in the concentration. The mixing time is what sets the inherent limit on the temporal resolution of the CPC. Thus, “the relatively large time required to establish the supersaturation in the laminar flow condenser” (line 2) contributes mainly to the plumbing time and relatively little to the mixing time, especially for a CPC with sheath flow such as a TSI 3025 or 3776. On the other hand, “zones of recirculation” (line 4) in the saturator substantially contribute to the mixing time and the loss of temporal resolution. Again, for sheathed CPCs, this is not a factor as the aerosol does not go through the saturator.

Manufacturers of CPCs, such as TSI, often quote a “response time” as the delay in getting to 95% of the response to a step change in concentration. This amounts to the sum of the plumbing time plus three times the mixing time ($3*\tau$). Thus, in the TSI spec sheet for the 3776 the “response time” is quoted as <0.8 sec. However, in the same document there is a plot of the normalized response to a step change in concentration. From this it appears that the "response time" is roughly 0.6 s, the plumbing time 0.4 s and the mixing time 0.09 s. (The reason the "response time" calculated from the plumbing and mixing times does not quite agree with the "response time" read directly from the plot is at least partly due to the crudeness of reading values from the plot and also perhaps because this may not be a perfect exponential decay.)

For other laminar flow CPCs, Wang et al. (2002) report mixing times of 0.174 s and 1.35 s for the TSI 3025 and 3010, respectively, while Quant et al. (1992) report 0.1 s and 0.95 s for the same two instruments. Thus, your statement that "the temporal resolution of such CPCs is on the order of seconds" is too misleading and should be amended to perhaps cover only unsheathed CPCs.

p. 5910, ln. 8: The 0.06 s mixing time of the Wang et al. (2002) mixing CPC is <u>not</u> "at least 10 times faster" than the commercial laminar-flow TSI 3776 with a mixing time of about 0.09 s.

p. 5911, ln. 12 and following: If you found it necessary to cool the "condenser" block to get this device to work then you apparently failed to obtain adiabatic mixing as is the essential element of a true "mixing-type" CPC. With true adiabatic mixing or at least something very close, no subsequent cooling should be necessary to obtain growth to optically-detectable sizes. Thus, though your device works quite well, it is actually something of a hybrid of a mixing-type CPC (which does not have a cooled condenser block) and a laminar-flow CPC that does have a cooled condenser block. Either insulate your mixing T better or modify the nomenclature you use to describe your CPC.

p. 5911, ln. 16: With this small of an inner diameter for the condenser, the Reynolds number is on the order of 4600 so the flow is definitely turbulent. I would imagine this would enhance the losses near the end of the condenser and in the nozzle entering the optics at which point the droplets would likely be large enough to experience significant inertial effects. You need to comment on this and its possible relation to the maximum counting efficiency of your device which is noticeably less than 100% (Fig. 2).

p. 5911, ln. 24 and following: You need to elaborate more on your choice of the flow rates and particularly the flow fractions. In Wang et al. (2002) Fig. 3c indicates that for any combination of flow temperatures, there is a particular flow split that produces a minimum critical diameter. Note that this is not at exactly the same flow split as the maximum saturation ratio (Fig. 3b) as the critical diameter depends not only on the saturation ratio but also has an additional separate dependence on the temperature. For your chosen temperatures is your chosen flow split anywhere near that which gives the minimum critical diameter? If not, why didn't you choose the flow split that gave you the lowest possible D_{P50}? Some amount of discussion in the paper should be devoted indicating specifically what went into your selection of operating parameters.

p. 5912, ln. 24: Though the sampling lines were arranged in a symmetric pattern, according to the text the EM is drawing 2 L min^{-1} while the total flow drawn by the CPC is only 1 L min^{-1}. Thus, the diffusional losses in these lines would not have been equal. How did you account for this? You might consider including a schematic of the experimental setup including indications of the values of flows, especially if there are details of the setup that were not mentioned that somehow balanced the flows to the detectors.

p. 5913, ln. 8-9: This bit about the "error in diameter" needs more explanation. Are you referring to the possible degree of skewing of the DMA output distribution due to the slope of the input distribution? Or is this rather a simple statement of the width of the transfer function and therefore the DMA output distribution which does not imply any skewing of the mean of the distribution. Though this latter effect complicates the interpretation of the data, it is not really properly referred to as an "error". Disregarding significant slopes in the DMA input distribution, the standard deviation "sigma" of the transfer function does not relate to any uncertainty, or "error" in the mean, or centroid, of the transfer function and resulting DMA output distribution. It is simply a measure of the width of the distribution.

p. 5913, Eq. (1): Finding the proper function to fit data is often problematic. It is often an attractive option to add flexibility with additional adjustable parameters to accomplish this. Unfortunately, though one might find an excellent fit this way, the adjustable parameters often become highly cross-correlated with each other and the range of acceptable parameter combinations to get a good fit becomes quite large. It also means that a high degree of precision must be used in specifying the parameters in order to allow for accurate computations. And finally, the parameters lose any individual physical meanings. This has happened here. I have tried calculating D_{P50} from the parameters given in Table 1 and I do not get the correct results. This is no doubt due to insufficient precision of the values in the table. Also, the fitted parameters do not follow any clear trends with saturator temperature as physically meaningful parameters would. Therefore, it is impossible to use this information to interpolate or extrapolate to find the efficiency curve for different saturator temperatures.

As can be seen from Fig. 2, the efficiency curves all have basically the same shape. As indicated by Table 1 and Eq. (1), we really only need one parameter (T_{S}) plus the particle diameter to determine the counting efficiency. Thus, ideally we would find an equation for the counting efficiency with only one adjustable parameter. This could conceivably be D_{P50}, which is readily correlated with T_{S}. Besides D_{P50}, we know that we will need to somehow define something like the slope of the curve in the cutoff region or, equivalently, D_{P0}, the extrapolated lower endpoint of the efficiency curve. But we can see that D_{P0} will be highly correlated with D_{P50} such that we could readily find a fit (perhaps linear) to this relationship and substitute it back into our efficiency equation. Then we are back to a one parameter equation. (Though these two parameters will be highly correlated it is clear from their physical meanings that variations in one cannot be balanced by corresponding variations in the other such that there is little change in the actual efficiency curve. Thus, we should not end up with high uncertainties for the fitted values of these parameters.) Finally, the asymptotic limit of the counting efficiency for larger particles should be the same regardless of saturator temperature. Thus, the measurements for all values of T_{S} should be fitted at the same time with a common fitted parameter, a, for this asymptotic limit. Then you need to find the proper form of the equation that produces the correct shape of the interpolated bend between the sharply inclined cutoff region specified by D_{P50} and D_{P0} and the flat asymptotic limit specified by *a*. The grand fit of all the data will then have seven adjustable parameters – three D_{P50}'s, three D_{P0}'s and *a*. After that, find the fit that predicts D_{P0} from D_{P50} and substitute it back into the efficiency equation along with the universal value of *a*. The final result is an equation for the CPC counting efficiency that depends only on D_P and D_{P50}. Included with this could be an equation relating T_S and D_{P50}.

If you find it impossible to come up with a form of the counting efficiency equation as described in the previous paragraph, it may be possible to make adjustments to the values in Table 1 such that parameter *a* is a constant and parameters *b*, *c* and *d* show reasonable trends with changing saturator temperature such that they can be readily correlated with D_{P50}. Back substitution into Eq. (1) should then yield the desired result, though a high degree of precision in the equations for the fitted parameters would still be necessary. As mentioned earlier, the fitted parameters in Table 1 undoubtedly have high uncertainties such that they can be varied together in a specific way with little change in the resulting curve. The uncertainties in the average measured points are probably great enough to accommodate a significant amount of variation in the parameters without invalidating the fit. Of course, now the problem is to find a method of varying the parameters to achieve the desired goal.

p. 5913, following Eq. (1): There needs to be some discussion of the result of an asymptotic limit of 97+/-1% counting efficiency at large diameters. As this is fairly consistent for the different saturator temperatures it seems unlikely that it is a matter of Poisson uncertainty in the measurements. Rather, there is a consistent 3+/-1% bias from the expected 100% for larger particles. Where is this coming from?

p. 5914, lns. 10-13: Since you are claiming there are no ultrafine (nucleation) sources in the laboratory during the experiment, I am guessing that the experiment was run overnight and that the total aerosol concentration gradually dropped over the course of the evening. Whether or not this is true, the lower plot of Fig. 3 indicates that there was considerable variation in the concentration over the duration of the experiment. The scavenging of particles to the walls, other surfaces and to larger particles in the lab would have been significant during this three-hour period. The rate of scavenging of the smallest particles within the 3 to 7 nm range would have been faster than the larger particles, thus the fraction of these smaller particles would not have remained constant. Rather it would have decreased with time such that the ratio of the concentrations of the FCPC and TSI 3776 would have gradually increased as is indeed the case in the first 250 min of the upper plot of Fig. 3. The presence of this trend should be explained in the text.

I suspect the total concentration was experiencing a trend, either up or down, during the experiment as was the ratio of concentrations. This means that the total concentration and the ratio of concentrations would be correlated without proving a direct causal relationship because the shape of the size distribution was changing. This would result in a bias of the slope in the lower plot of Fig. 3. Indeed, the offset (30.8) is not likely to be of any physical significance. It would either imply that at zero concentration the FCPC reads 30.8 cm^{-3} or that the fit over the range from the measured data to the origin is non-linear.

Neither of these seems likely. Instead, the authors should calculate the uncertainty of their fit parameters to determine if the offset is significantly different from zero. If not, then the fit should be redone with the offset fixed at zero and only the slope is fitted. Presumably then, the slope will be significantly closer to the observed average ratio of 0.85, though it may still be skewed due to the bias from the correlation mentioned above. In the end, with skewed data for the lower plot of Fig. 3, it might be best to simply eliminate it.

This was something of an ill-conceived experiment as the shape of the size distribution was neither controlled nor measured. If the size distribution were measured, then perhaps it could have been used to verify the ratio of the total concentrations measured by the CPCs and its variation over the course of the experiment. Perhaps it would have been better to simply use aerosol from an atomizer with appropriate dilution. The concentration of the aerosolized solution should be such that it produces an aerosol of size range well above the cutoff regions of the CPCs.

p. 5915, lns. 13-14: I am unclear as to what point you are trying to make here. First of all, when you refer to uncertainty of the curve you appear to be referring to an uncertainty in diameter. Where is this uncertainty coming from? Is this from the earlier reference to Stolzenburg (1988) where you claimed an uncertainty range of 0.3-1.3 nm? Wherever that diameter range came from I don't see how it is relevant to a real temporal shift in the cutoff diameter which causes a change in the measured number concentration. A simple uncertainty in the cutoff diameter would not have this effect. Do not get confused between accuracy/bias, precision/uncertainty and real changes.

p. 5917, Eq. (2) and associated text: First of all, the equation written is unphysical; it goes to infinite measured concentration as time advances. Presumably, you intended to negate the argument of exp. This then would be the proper equation to describe the response of the CPC in the situation where the ambient concentration makes a step change at t_0 from N_0 to zero concentration. In this case, the preceding text should read something like this: "… the change of the measured concentration dN_i/dt is proportional to the difference ($\delta N = N_0 - N_i$) of the true concentration, N_0, and the measured concentration, N_i. This is just $-N_i$ in the case of a step change from $N_0 > 0$ to zero concentration at t_0. This results in a solution for $N_i(t)$ of the form: …". A more general form of the equation would be:

$$N_i(t) = N_1 + (N_0 - N_1) * \exp[-(t - t_0)/\tau] \qquad (2)$$

where now the step jump at t_0 is from N_0 to N_1. So your choices are to use the more general equation with appropriate preceding text similar to that above or to correct the sign in your existing equation and do a better job (similar to that above) of explaining the situation in the text preceding the equation.

p. 5917, Discussion of Fig. 7: In your analysis here you are assuming that the true ambient concentration made a step change here. Since you only have the response of the FCPC you don't actually know that. A step change would be the worst-case scenario and your analysis, if done properly (see below), would yield the maximum possible value of τ. The FCPC may actually perform better than that.

As noted above, the CPC response to a step change in the ambient concentration should be a sharp rise (in this case) of the measured concentration from the original ambient concentration followed by an exponential decay to the new ambient concentration. The 10 ms temporal resolution of the FCPC measurements is clearer too coarse to adequately define this behavior and the use of the linear interpolation leads to an overly large estimate of τ.

Four parameters are needed in the version of Eq. (2) above to properly model the FCPC response and thereby obtain a good estimate of the mixing time. The first two, N_0 and N_1, can readily be estimated from the measurement points immediately before and after the jump. The other two, t_0 and τ, must be obtained from points during the exponential decay. There must be at least two of these to determine these two parameters. The point at t = 660.277 s clearly falls during the decay. The previous point clearly does not and the following point is unclear. It is somewhat below the average value (N_1) of the following five points but this could conceivably still be within the natural variation at this new level. Thus, our choice out of necessity of designating it as still within the decay period is somewhat dubious. An eyeball fit of an exponential to the region of the jump then suggests that the initial rise of N_i, the theoretical measured concentration given a sufficiently high measurement frequency, began just before the first point on the decay curve at about t = 660.275-6 s while the e-folding point is at about t = 660.278-9 s. This gives an e-folding time of roughly τ = 3 ms. Since this was just an eyeball fit the result is a fairly crude estimate but it clearer indicates that the FCPC mixing time is much less than the 16 ms reported in the paper. Also note that if the second point used for the decay is actually at the new level then the fitted exponential must be even steeper and τ smaller yet.

But even the above analysis is not strictly correct as the measurement points do not represent instantaneous readings but rather 10-ms averages. This must be taken into account when fitting the exponential decay curve. For the first point on the decay curve, the average must extend back in time prior to t_0 so the average is greater than the theoretical instantaneous reading at the same point. For the next point, the average is over a section of the curve that is concave downward so the average is lower than the corresponding instantaneous reading. If we then fit our exponential to the instantaneous points which are now farther apart than the average points were, the curve will be even steeper and again τ will be smaller.

Ideally, this experiment (or a much simpler laboratory experiment using spark aerosol generation a la Wang et al. (2002) or a fast solenoid valve a la Quant et al. (1992)) should be repeated using a significantly shorter gate for the FCPC counting. For the 10-ms gate and the lower concentration, ~$2.5\text{x}10^4$ cm^{-3}, in Fig. 7 there must have been about 1250 counts per measurement point allowing less than 3% Poisson variation in the measurements. I think one could live with greater Poisson variation in the interest of getting more points on the decay curve to allow a more accurate determination of τ. If the electronics and computer are capable of working with a 1 ms gate then the Poisson variation would still be less than 9% and there should be plenty of points to define the decay curve and get an accurate determination of τ. If a 1 ms gate is not possible any decrease of the gate width from 10 ms would be advantageous.

p. 5917, Section 3.3.2, 1st paragraph: There are many problems with this discussion of the spectral behavior of the time series in Fig. 6. First of all, many of your readers, including myself, will not have dealt with this aspect of turbulence or spectral analysis since college. Thus, you must explain things more fully and provide references where appropriate. Specifically, the section beginning in line 18 with "From turbulence theory …" requires one or more references to back up your statements concerning the inertial subrange and the -5/3 exponent. Your statement beginning in line 21 with "… we can safely conclude …" needs a bit more explanation. I gather this is a result of the curves smoothing out beyond ~0.5 Hz and following a -5/3 slope. What is the origin of the noise below 0.5 Hz? The correspondence between frequency and eddy size needs noted explicitly. Why is the variation in the curve in Fig. 8 representing Poisson noise so great? Please give us some idea of how spectral analysis differentiates between coherent structures and Poisson noise.

p. 5919, In. 7: "The dropoff is … from mixing inside the FCPC." Given the reanalysis above of the response of the FCPC to a step jump in concentration, it would appear that the 10-ms counting gate is a greater limiting factor in the temporal resolution than the mixing time which appears to be notably less. Yet, in the "low-pass filtering" section of the curves in Fig. 8, the 100-Hz (10-ms) record and the 50-Hz (20-ms) record track together perfectly. This indicates that the gate time is not the cause of the filtering effect either. So what is causing this?

p. 5919, lns. 9-16: This section makes no sense. First of all, there are several independent factors which might limit the temporal resolution of the FCPC. There is no reason they should agree, i.e. all begin limiting the resolution at the same frequency. Thus, the way in which you try to relate the overall dimensions of the FCPC to the mixing time means nothing. Suppose you had an instrument with ideal plug flow from the inlet to the detector. The spatial and temporal structure of the sample would be transported intact from the inlet to the detector and the length of that transport would be irrelevant to the resolution of the instrument. The degree to which the FCPC falls short of this ideal is measured by the mixing time τ. This is all the internal information you need for this matter. The relevant physical dimensions you are looking for are those of the sample volume in the ambient as it is defined by the wind speed (2m s^{-1}), sample flow rate (0.3 L min^{-1}) and gate width (10 ms). The sample volume (0.05 cm^3) is just the sample flow rate times the gate width. It is assumed to be in the shape of a cylinder. The length (2 cm) of the cylinder along the direction of flow is given by the wind speed times the gate width. The diameter of the cylinder (0.18 cm) is readily calculated from the volume and the length. Fortunately, this sample volume diameter is very close to the ID of the ¼ inch tube that appears to be used as the inlet to the FCPC so the sampling was reasonably isokinetic. Otherwise, the diameter of the inlet might also be a dimension to be considered here. Clearly, the length of the sample volume will limit the spatial resolution of the sampling much more than the diameter. But the averaging effect of the length of the sample volume is identical to the effect of the finite sampling frequency (or gate width) as previously discussed. Thus, this analysis of critical dimensions provides no new information as to source of the apparent limit of the temporal resolution of the FCPC as indicated by the dropoff in the "low-pass filtering” section of Fig. 8.

Perhaps the dropoff is caused by something else and not by a limitation of the temporal resolution of the FCPC. The results from Fig. 7 would tend to support this. In any case, if the spectral analysis is to be included in this paper a more rigorous analysis should be presented. Among other things, the results from Fig. 7 and Fig. 8 should be compared more rigorously. I would imagine that the low-pass filtering effect of a first-order response system characterized by an e-folding time τ (or a finite gate width) has been analyzed thoroughly in the literature and there exists a simple equation with a corner frequency and everything that describes quantitatively the rolloff of the power spectral density as a function of frequency. This can be used to quantitatively compare the results of the two plots.

p. 5918, lns. 17-20: If you choose to keep the spectral analysis it may be useful to point out the essential tradeoff between temporal resolution limited by gate width or by Poisson noise. That is, as one shortens the gate width to increase the temporal resolution, the Poisson noise level increases such that it masks coherent turbulent structures at a lower frequency. There is then, for a given concentration, an optimal gate width to resolve the highest frequency of coherent structures.

p. 5919, ln. 12: As noted earlier, the 5 nm shift is real and its comparison to the uncertainty is largely irrelevant. You will see a change in the total concentration measured.

p. 5919, ln. 13: The change in the total number concentration is only negligible if there is only a negligible nucleation mode present. When there is a nucleation mode, it frequently makes up a large fraction of the total number so your statement would be false.

p. 5919, lns. 19-20: As noted earlier, the mixing time is probably much shorter than 16 ms. Also, commercially available laminar flow-type <u>sheathed</u> CPCs have mixing times significantly less than 50x16 ms = 0.8 s.

p. 5924, Fig. 1: According to this schematic, the optics exit flow and the saturator inlet flow are measured. Flow from the condensate trap is fixed at 0.2 L min^{-1}. Are all these flows at the same temperature? How much butanol vapor is in the optics and condensate trap exit flows? All this must be taken into account in calculating the aerosol inlet flow. Was this done?

p. 5928, Fig. 5: In the upper plot, the average of the ratio of FCPC to EM concentrations appears to be very close to 1.00 if not a bit higher for 30-nm particles. However, in Fig. 2 that efficiency is measured at about 0.96-0.97. Are these equal within the estimated standard deviations of their respective means? If not, do you have an explanation for that?

p. 5928, Fig. 5: The offset in the fit to the data in the lower plot, would most likely represent a zero error for the electrometer if it is significantly different from zero. Is it reasonable for the zero error to be this high? If the offset is not significantly different from zero then set it to zero and refit the data with a line through the origin.

Technical Corrections

page 5908, line 15: "… a sharp ramp due to a concentration change …"
p. 5908, ln. 19: "Condensation Particle Counters (CPCs) are …"
p. 5909, ln. 20: "… where the requirements … are even higher …"
p. 5909, lns. 22, 25: The word "aircraft" (without an 's') is normally used as both singular and plural.
p. 5909, ln. 22: Better wording for this would be "with a time resolution on the order of one second the spatial resolution resulting from the true air speed of the aircraft (~100 m s^{-1}) is 100 m."
p. 5909, ln. 25: "… aircraft observed only a few measurement points ~~only~~ in the region where new particles were formed."
p. 5911, ln. 10: "Swagelok" is misspelled.
p. 5912, ln. 1: "The typical duration of … flights …"
p. 5912, ln. 10: I assume you were only using one "furnace".
p. 5912, ln. 14: If this was a commercially available ^{85}Kr charger, give its manufacturer and model number. If this was something else, give a reference for it. Charger performance is becoming an increasingly important topic as the performance of SMPS systems are compared.
p. 5912, ln. 20: Define the acronym "EM".
p. 5912, ln. 20: The description of the test system would flow better if you finished describing the flow distribution after the dilution of the aerosol flow (i.e. mixing bottle, header, sampling lines, instruments) before giving details about the EM (i.e. flow rate and background offset) and CPC operating parameters.
p. 5912, ln. 23: "… with 11 ports downstream of the mixing bottle." Also, more information about the mixing bottle is needed to assure the reader that it is serving its intended purpose.
p. 5913, ln. 10: It would be clearer that you are talking about standard deviations of the means of the measured counting efficiencies, not diameters, if it was worded as "… standard deviations of the mean values at each diameter".
p. 5913, ln. 12: "… but in the cutoff region ~~slope~~ it can be higher …" "Cutoff region" or something similar would be clearer than "slope".
p. 5913, Eq. (1): Replace "x" with "D_P".
p. 5914, ln. 5: "… operated over a few hours …"
p. 5914, ln. 14: "… during the first 250 min" is a better way of saying this.
p. 5914, ln. 23: "…to check how stable the location of D_{P50} ~~the slope~~ is, (2) diameters above the cutoff region ~~slope~~, …" Again, "the slope" is ambiguous.
p. 5916, ln. 2: Substitute "environment" for "laboratory".
p. 5916, ln. 3: "Two-fold", not 'two-folded"
p. 5916, ln. 10: "… the time response of the FCPC is too low …"

p. 5916, ln. 15: "The FCPC was placed at ~~in~~ a height …"

p. 5917, ln. 9: For clarity as discussed above, use "mixing time", not "response time", twice in this line.

p. 5917, ln. 25: "The red and black curves represent …"

p. 5918, ln. 1: "… for the red curve a non-overlapping average …"

p. 5918, ln. 3: "… a 300-s long subrecord (t = 150-450 s) …"

p. 5918, ln. 17: "The flattening at the high-frequency end …"

p. 5918, ln. 22: "… still not completely noisy …"

p. 5919, ln. 4: "… shows a behavior similar to laminar …"

p. 5919, ln. 5: "… with increasing temperature differences D_{P50} is moving to smaller diameters."

p. 5919, ln. 6: "Here, temperature differences from 25 to 29K were chosen leading to D_{P50} between 8.7 and 6.5 nm." Give values of D_{P50} in same order as corresponding temperatures.

p. 5919, ln. 9: "… over a few hours …"

p. 5919, ln. 10: "… hours, except that ~~only~~ the D_{P50} ~~slope~~ of the counting efficiency curve is moving to larger particle sizes when ~~no~~ butanol is not filled continuously."

p. 5919, lns. 17, 19, 25: "… mixing ~~response~~ time …", three times

p. 5919, ln. 24: "… still not completely noisy …"

p. 5920, ln. 21: "… particle exchange with a twin CPC …"

p. 5923, Table 1: It would be useful if a copy of Eq. (1) were included here.

p. 5925, Fig. 2, caption: Though "quadrates" is technically a correct term to describe the symbols, it is unnecessarily obscure in this context. Please use "squares".

p. 5926, Fig. 3, caption: Replace "250 min from start" with "the first 250 min", twice.

p. 5931, Fig. 8, plot key: The dashed line representing $f^{5/3}$ is not a fit. Do not label it as such.