

# Response to reviewers: Intercomparison of airborne and surface-based measurements during the CLARIFY, ORACLES and LASIC field experiments

Paul Barrett et al.

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We thank both reviewers for taking the time to read through this paper and offer many constructive criticisms that have no doubt improved the manuscript. We recognise that the manuscript is long and the results were not presented as concisely as they could have been. We have attempted to rectify this through use of additional figures and removal of some tabulated materials to the Supplement. Whilst the text could have been shortened with the use of tabulated information about the instrumentation we felt that readability would have suffered and so kept the section broadly the same.

Use of ODR fitting was undertaken initially but we have taken onboard the suggestions to shorten the averaging period and have done so where possible and have now also included uncertainties on the ODR fit parameters. We now described the method in detail at the head of the results section.

We have concentrated on primary measured quantities and so moved some derived parameters such as dew point temperature, relative humidity and aerosol particle effective radius to the supplementary materials.

Some of the discussion has been moved in to the results section, including that around thermodynamics to make manuscript more readable. The discussion section is now more focussed on synthesis of results and outstanding issues, such as the impact of inlets, etc.

We present the responses to both Reviewer #1 and Reviewer #2 below. Comments are copied in grey italics for convenience. We do not include every change to the manuscript in here as that would be unwieldy, so we also upload a marked-up manuscript with differences highlighted. We have added references to relevant literature that has become available since submission.

## 1. Responses to Reviewer #1

### 1.1 Major

*1) The comparisons between the various instruments are based primarily on linear regression against mean values from long periods of flight. There are several problems with this approach:*

*a) The uncertainties quoted are for each instrument's inherent response time as installed in the aircraft. Yet averaging together many minutes of data will result in reduced uncertainties (if the same population is being randomly sampled). One would expect better agreement than the stated raw instrument uncertainties for such averaged data.*

*b) Regression should be applied using the highest time resolution data possible, rather than to just a few average values from these different "runs". A quick example: if there were only two "runs", using this manuscript's approach there would be only two values, and regression would be a perfect fit to the two data points. The agreement between instruments should be based on the highest resolution data reported, to which the stated uncertainties apply. If one were to fit to averaged values, uncertainties must be adjusted and accounted for in the regression. It would be very interesting to see the regression from the large dynamic range covered in the profile of the two aircraft; this would be a nice way to rigorously compare instruments in a challenging environment.*

44 1. Fits were performed using ODR originally, but this was not stated explicitly. Regressions have now  
45 been redone - and performed on 10s segments rather than flight leg averages. See below for  
46 details.

47 **1 a), b)**

48 The datasets tend not to be valid at the raw instrumental resolution due to the nature of sampling  
49 from the different platforms. In particular due to sampling through inlet systems and through  
50 pipework which can result in physical “smoothing” of the signals due to imperfect transport, and  
51 possible temporal offsets – which whilst we attempt to correct for this may still be present. Small  
52 timing errors may differ between instruments on the same platform and between platforms. In most  
53 instances e.g. optical absorption and extinction on FAAM the true fastest response possible has been  
54 demonstrated in the laboratory to be between 6 and 10 seconds. Therefore, we have first smoothed  
55 data to 10 s (i.e. 0.1 Hz) from the aircraft.

56 Data have been included for as wide a dynamic range as possible from the full flight intercomparison  
57 section. This includes the very dry and relatively clean troposphere at close to 6km and the polluted  
58 humid oceanic boundary layer. We do not include data specifically from the whole profile, as many  
59 instruments are not optimised for use during descents / pressure changes. We now show the data  
60 from the absorption measurements, and the problems can be seen in the artefacts in the NASA PSAP  
61 data, where there is a spike in data on red and blue channels, resulting in unrealistic looking single  
62 scattering albedo values. We feel that using the data from known good times in the free troposphere  
63 leg and the descent through the pollution layer in the free troposphere is a good compromise. We  
64 have also used observed CLARIFY PAS observations data to compute Angstrom exponents for all  
65 wavelength pairs for the airborne comparisons, rather than relying on the campaign mean from  
66 Taylor et al. (2020) as we had done originally.

67 Concentrations of pollutants, chemical and physical varied over the range that is presented – we do  
68 not include data that is below demonstrated (in the laboratory) limits of detection.

69 Data from LASIC must be treated differently, as the measurements are offset in space and time.  
70 Here we keep the observations as mean values and variability.

71 The errors in x and y and the ODR fits are taken as the standard error over the averaging period. We  
72 have now added commentary at the start of the results section that gives details of the method and  
73 the reasons for the choices made in the analysis. We are aiming to find the similarity or differences  
74 between the observations on two platforms, rather than construct a function that maps one set of  
75 observations on to the other. Of course, should downstream users want to obtain measurements  
76 with reduced uncertainties then they could average over any length of time of their choosing,  
77 considering natural spatial and temporal variability and we expect them to do this on a per-  
78 instrument basis as they require.

79 The fit parameters only changed by minimal amounts (a few percent), by changing from run  
80 averages to 10 s data - for example:

	Original ODR fit	New ODR fit
CO	$8 + 0.97x$	$9.5 + 0.95x$
O3	$-1 + 1.19x$	$-9.6 + 1.17x$
$\sigma_{SP}$ at 660 nm (PM10)	$-0.1 + 1.56x$	$-0.57 + 1.52x$
$\sigma_{SP}$ at 660 nm (PM1)	$-0.3 + 0.90x$	$-0.72 + 0.97x$

81

82 *c) The linear regressions appear to use one-sided least-squares fits. Because there are*

83 *uncertainties in both x and y parameters, a 2-sided regression, such as orthogonal*  
84 *distance regression, should be used to determine slopes and intercepts. Further, the*  
85 *regressions should account for the uncertainties in each parameter, whether averaged or*  
86 *not.*

87 Fits are performed using orthogonal distance regression, this was not stated in the original  
88 manuscript.

89 *2) Most of the data are presented in Table 3, which is so large as to be completely*  
90 *unwieldy and is extraordinarily difficult to read because it spans multiple pages.*  
91 *Generally it is much preferable to show data graphically. Instead of Table 3, I*  
92 *recommend a series of graphs of the key data that are discussed and analyzed (at their*  
93 *native resolution). For example, a plot of extinction coefficient for the two airborne*  
94 *platforms could be shown with all of the data covering the full dynamic range, with*  
95 *points perhaps colored by the run type (BL, FT, etc.). It may be most effective to use*  
96 *log-log plots to show the range of values clearly. The numerical values in Table 3 could*  
97 *go into an appendix or the supplemental materials, hopefully in a more compact format.*  
98

99 We agree that the table was too large.

100 New Fig. 5 now contains comparison plots of temperature and humidity, with the data from this  
101 portion of the table moved to the supplement. Aerosol number correlation plots are added to the  
102 PSD figure (new Figure 7). We have chosen log plots for humidity data and kept linear for others  
103 which we deem best to show the data.

104 Where possible we now show data points coloured by altitude. For some parameters we do not do  
105 this in order to preserve clarity.

106 Data for (new) Figs 5, 6, and 7 are shown as the 10 s values rather than run averages for airborne  
107 data.

108 Where data are now plotted the values from Table 3 are moved to the supplement. For the  
109 parameters that remain – they have been split in to sub-tables, and placed on landscape pages,  
110 reducing the number of pages of tables in the main manuscript. We have retained chemical  
111 composition measurements and derived properties as these are present only from the boundary  
112 layer at one point in time. The LASIC data (which compare badly) are included for completeness  
113 (item #5). Cloud physical properties are also tabulated as only one run was performed in cloud.

114 We are showing all the extinction data that it is possible to show – given the times we know that  
115 instruments were operating outside their valid operating parameters. For example, NASA extinction  
116 data requires scattering and absorption, but the PSAP which measures absorption does not perform  
117 well during the descent.

118 *3) There is extensive discussion of aerosol number concentration and effective radius.*  
119 *However, aerosol mass is extremely important as it is the parameter most often carried*  
120 *in models. Thus it would be very useful to compare integrated volume from the different*  
121 *size distribution instruments. I would suggest that Fig. 6 be converted to 6 panels, with*  
122 *a, b, and c showing, on a linear y-scale, the number concentration comparisons, and*  
123 *panel d, e, and f showing the volume concentrations on a linear panel. A log-log scale*  
124 *with almost 9 orders of magnitude on the y-axis can hide so much detail. For example,*  
125 *at ~2 nm in the current Fig. 6a, there is almost an of magnitude difference between the*  
126 *green line (FAAM PCASP1) and the others. Is this significant? When plotted on a linear*  
127 *scale we can see if this difference is a significant contributor to parameters we care*  
128 *about, such as integrated number or volume (mass).*

129 3) We have modified the particle size distributions (new Fig. 7) to show number and volume  
130 distributions. Linear y-scales are used for both. We chose to keep the elevated pollution plume and  
131 free troposphere data on the same figures (b) and (d) as the purpose is to show that the instrument  
132 can differentiate between the weak pollution plume and the cleaner surroundings – at least for  
133 particle number distributions. The particle volume distributions are shown to be poor – as there are  
134 so few particles at the larger diameters. We do not show cumulative distributions because there is  
135 no good way to integrate number or volume across multiple probes, without creating a composite fit  
136 - and that is beyond the scope of this study and left for individual research questions.

137 4) Figure 8. I had trouble understanding Fig. 8b. The y-label say it is the Angstrom  
138 exponent of absorption, but the caption says it is that for extinction. Is it derived using  
139 Eq. 2 applied to the absorption coefficient values shown in Fig. 8a? If so, why are the  
140 markers in 8b plotted at ~460 nm when the closest wavelength pairs are at 470 and 405  
141 nm? Please explain carefully how these values were derived. Also, it would make more  
142 sense graphically for these two plots to be side-by-side, to enhance the vertical scaling  
143 and make differences more evident.

144 Corrected extinction to absorption in caption.

145 Wavelength pairs for blue, green absorption from FAAM EXSCALABAR are 405 nm and 515 nm giving  
146 a mean of 460nm. NASA PSAP instrument has wavelengths 470 nm and 530 nm giving a mean of 500  
147 nm, as plotted.

148 We have replotted the figure with one panel above the other as suggested and narrowed the aspect  
149 ratio to permit printing in a single column rather than spanning both.

150 5) *Lines 950-956. The agreement between the AMS on the FAAM aircraft and the ACMS*  
151 *at the ARM site was quite poor, with factors of 3-4.5 difference. These data should be*  
152 *shown in Table 3, but are not. Poorly agreeing data can be just as important as data that*  
153 *agree well, so please show the values if they are part of a project data archive and not*  
154 *rejected for quality-controlled reasons independent of this comparison.*

155 This data is included in the revised version(see item 2.)

## 156 1.2 Minor

157 1) *Abstract. The data are described multiple times as agreeing "well". This should be*  
158 *changed to a more quantitative statement, such as "the data agreed within combined*  
159 *experimental uncertainty", if this is the case when the comparison is made at time*  
160 *resolutions for which the stated uncertainties are valid (see comment 1b above).*

161 Abstract – modified to remove “well” and added context.

162 2) *Line 186. Need period after "Beer's Law"*

163 Added period.

164 3) *Line 217. Two periods.*

165 Removed period.

166 4) *line 249. Change "dependant" to "dependent", here and elsewhere.*

167 Changed dependant to dependent globally.

168 5) *Line 255. I don't understand this sentence. Please clarify.*

169 Removed the sentence.

170 6) *Line 268. Do "rear" and "front" instruments refer to PSAPs or nephelometers?*

171 Added PSAP to line 271 for clarification.

172 7) *Line 283. Please state the flow rates to each optical instrument.*

173 Added line 285 - " The nephelometer drew at 30 L min<sup>-1</sup> and the PSAP 2 L min<sup>-1</sup>."

174 8) *Line 379. What are representative uncertainties for the absorption coefficient*  
175 *determined from the CAPS PMSSA instrument?*

176 Added line 377 - "The CAPS PM<sub>SSA</sub> measurement uncertainties for absorption coefficients are  
177 estimated in Onasch et al. (2015). For a typical SSA ~0.8 during LASIC, a conservative uncertainty  
178 estimate for the absorption coefficient is ~20%."

179 9) Line 397. Moore et al. (2021) provide a thorough analysis of refractive index  
180 sensitivities for the UHSAS. Moore, R. H., Wiggins, E. B., Ahern, A. T., Zimmerman, S.,  
181 Montgomery, L., Campuzano Jost, P., Robinson, C. E., Ziemba, L. D., Winstead, E. L.,  
182 Anderson, B. E., Brock, C. A., Brown, M. D., Chen, G., Crosbie, E. C., Guo, H., Jimenez,  
183 J. L., Jordan, C. E., Lyu, M., Nault, B. A., Rothfuss, N. E., Sanchez, K. J., Schueneman,  
184 M., Shingler, T. J., Shook, M.A., Thornhill, K. L., Wagner, N. L., and Wang, J.: Sizing  
185 response of the Ultra-HighSensitivity Aerosol Spectrometer (UHSAS) and Laser Aerosol  
186 Spectrometer (LAS) to changes in submicron aerosol composition and refractive index,  
187 Atmos. Meas. Tech., 14, 4517–4542, <https://doi.org/10.5194/amt-14-4517-2021>, 2021.

188 Added line 405 – "Moore et al. (2021) noticed similar behaviour in laboratory tests of a UHSAS for  
189 highly absorbing aerosols. Here we use the NASA P3 data for comparison with the outboard FAAM  
190 BAe-146 PCASPs." and reference to Moore et al. (2021).

191 10) Line 393. Although this is described in more detail in Wu et al. (2020), please  
192 provide a succinct explanation for why an empirical correction factor is needed for the  
193 SMPS, when it's quite a fundamental instrument.

194 Added line 393-399 – "Previously a comparison was made for CLARIFY data between estimated  
195 volume concentrations derived from AMS + SP2 total mass concentrations and PM1 volume  
196 concentrations from PCASP (assuming spherical particles). Estimated AMS+SP2 volumes were  
197 approximately 80 % of the PCASP derived values, which was considered reasonable within the  
198 uncertainty in the volume calculations (Wu et al., 2020) demonstrating consistency between inboard  
199 and outboard measurements. Discrepancies between SMPS (inboard) and PCASP (outboard)  
200 number concentrations remained however and so the SMPS concentrations were reduced by a  
201 collection efficiency factor of 1.8 to give better correspondence in the overlap region of the PSDs.  
202 The cause remains unknown."

203 11) Line 403. Perhaps just state "with updated electronics" rather than "with SPP200  
204 electronics". Or explain what SPP200 means.

205 Modified line 413 – "FAAM and NASA flew wing-mounted DMT PCASPs (Lui et al., 1992) with  
206 updated electronics (nominally SPP200, DMT 2021) which were exposed to the free airstream."

207 12) Line 417. Change "bin dimensions" to "bin boundary diameters".

208 Replaced bin dimensions with bin boundary diameters.

209 13) Line 418. The underwing PCASP is not only not adjusted for the "absorbing  
210 characteristics" of the BBA, but it's in general not adjusted for any varying refractive  
211 index, including water. This could make a significant sizing difference with in-cabin  
212 spectrometers.

213 This is discussed in results – new line 738-743 - Data for runBL were also available from the NASA  
214 UHSAS, first corrected for the characteristics of BBA as described in Howell et al., (2021), for  
215 diameters up to 0.5 µm (the stated upper size limit for the correction algorithm. Concentrations are  
216 larger than those reported by any of the PCASPs. By converting the FAAM PCASP2 bin boundaries to  
217 those for BBA equivalent refractive index it can be seen that the PSD more closely matches that from  
218 the UHSAS although concentrations are still lower. This demonstrates the importance of considering  
219 the material refractive index when combining measurements from multiple probes with differing  
220 techniques.

221 14) Line 641. What are linear regression "sensitivities"?

222 Replaced "sensitivities" with "slopes" globally.

223 15) Line 664. Data taken at or below detection limit are also of use, and should be  
224 plotted as suggested in comment 1b above.

225 Data from the FAAM AMS are not available for the altitudes above the boundary layer during this  
226 flight. The instrument was not able to detect material above the background, and so can not be  
227 included here. A fit to these low magnitudes would be biased by data which is known to be of poor  
228 quality.

229  
230 *16) Line 688. "Re" (effective radius) is not defined.*  
231 Equation for Re (effective radius) is defined on line 487.  
232 *17) Line 677 (and 955). Show the LACIS ACMS data in Table 3. Are they at least*  
233 *correlated?*  
234 Added LACIS ACMS data to table 4 showing that LACIS ACMS always overreads compared to FAAM  
235 AMS, but by varying ratios.

236 *18) Line 1080. Replace hyphen with a comma.*  
237 Replaced hyphen with comma.

238  
239 *References:*  
240 *Please ensure that all references comply with Copernicus' style guide. For example, for*  
241 *Baumgardner et al. the title is capitalized, as is Cotterell et al. (2021). This behavior is a*  
242 *result of reference manager software, which always messes up formatting and must be*  
243 *thoroughly checked manually.*

244 References checked and amended where required.

## 245 2. Response to Reviewer #2

246 We have included a key to acronyms as Table 8 and abbreviations have been checked.

### 247 2.1 Major

248 *In general, comparing measurements with different setups, actively dried or not, is not*  
249 *recommended. To ensure comparable conditions, one should care for RH below 40 %.*  
250 *Especially the RH is of crucial importance for filter-based absorption photometers. The*  
251 *observed gradient in the RH (Fig 4c) transposes into the airplane's piping and will bias t*  
252 *the absorption measurements due to the principle of differential measurement of the*  
253 *light attenuation behind the filter spots even if the cabin is heated to 30 °C (which also*  
254 *has implications for the volatile components of the aerosol particles). I.e., a sample at*  
255 *~80 % RH at ~12 °C outside equals inside at ~26 % at 30 °C. As shown in the profile,*  
256 *there was a change to ~1 % RH at ~20 °C outside, which equals 0.6 % at 30 °C inside.*  
257 *This relatively fast change of more than 25 % can significantly impact the filter-based*  
258 *absorption at NASA P3's PSAP or the TAP used on FAAM. However, the Nafion™ dryer at*  
259 *FAAM aircraft should dampen this effect significantly. The discussion must address this*  
260 *feature of the experimental setup.*

261  
262 **Relative Humidity is not controlled on all platforms:** we agree that this is a significant issue – but in  
263 many ways it is this aspect that has motivated this study. The platform operators here (and in  
264 general) are very distinct, some operate state-of-the-art unique instrumentation - e.g. FAAM and  
265 EXSCALABAR for optical extinction, versus commercial instrument on NASA and LASIC,  
266 nephelometers and PSAP for optical scattering and extinction. We want to understand the  
267 comparability of measurements made using these techniques – in part to understand the  
268 comparability of our measurements across the SE Atlantic basin between 2016 and 2018 and also  
269 because a number of historical datasets already exist using a range of these techniques.

270 We include the profile plot of optical absorption and comment on the suspect artefact in the PSAP  
271 sample from the elevated pollution layer. Added this to line 821:

272 *"The FAAM PAS data from the profile descent shows that absorbing aerosols are present in*  
273 *magnitudes greater than the lower threshold of the instrument in the boundary layer, runBL, and*  
274 *upper pollution layer, runELEV. Data follow similar trends from the NASA PSAP in the boundary layer.*  
275 *In the elevated pollution layer the NASA PAS data look suspect, for example signals from red and blue*  
276 *are nearly identical suggesting an unphysical Absorption Ångström exponent ( $A_{AP}$ ). This is likely*

277 *because the PSAP is not suitable for operating in regions where pressure or RH or other external*  
278 *factors are changing rapidly such as during descent, especially, as is the case, where the sample is*  
279 *not actively dried. These data should be treated with caution and are not used in subsequent*  
280 *correlations (Fig. 6 (h), (i)). Consequently, the data for  $\sigma_{EP}$ , from NASA (nephelometer + PSAP) should*  
281 *be treated with caution in the elevated pollution layer, when compared against the FAAM CRDS*  
282 *measurement which probes optical extinction directly. “*

283 We also have some discussion regarding RH already in section 5.4 which relates to the fact that the  
284 bias between LASIC and FAAM on the optical scattering measurements is in the opposite direction  
285 than might be expected from the un-dried LASIC sample. This continues into discussion around inlet  
286 sampling artefacts in section 5.5. We feel it is important to show these biases and consider the  
287 causes such that future campaigns may be better designed.

288 *Table 3 is way too large. One should consider presenting the content more*  
289 *comprehensibly, like with figures. E.g., the table content can be separated into the*  
290 *coefficients of the linear fitting and average values.*

291  
292 We agree and have removed much of the material to the supplement, partly by including new Fig. 5  
293 which compares temperatures and humidities and only keeping data which is not present  
294 graphically. We split the remainder in to multiple smaller and more targeted tables.

295  
296 *Figure 5 displays correlations of two variables consisting of uncertainty each. Hence a*  
297 *linear fit is not applicable, and an orthogonal fit accounting for both uncertainties should*  
298 *be applied. Moreover, it is unsuitable for fitting a linear behavior based on two*  
299 *observations. I would suspect that the statistical significance of those fits is small.*  
300 *Enhance the number of data points by decreasing the averaging window or address this*  
301 *in a deeper discussion.*

302 We were originally using Orthogonal Distance regression fits to account for uncertainty/variability in  
303 both x and y directions, and this is now made clear in the text at the start of Sect. 4 Results\*. We also  
304 take onboard the suggestion to reduce the averaging time (to 10s) where appropriate. This is done  
305 for the airborne comparisons. The fact that the data from the ground – airborne comparison are not  
306 collocated in space / time mean that this is not possible for this part of the comparison.

307  
308 \*Added line 615-634 - “When comparing measurements from two instruments, it is useful to  
309 explicitly consider statistical uncertainties, which differ between individual data points, and  
310 systematic uncertainties, which affect all data points from an instrument. Statistical uncertainties  
311 are large when instrument noise is large compared to the measured signal, and/or the measured  
312 property exhibits a high degree of variability within the sampling period. The effect of instrument  
313 noise can be minimised by choosing a longer averaging time and this is the approach we take for the  
314 comparisons between the BAe-146 and ARM site. The straight and level runs were designed to  
315 minimise the variability of measured properties during the comparisons, and we average the data to  
316 one point per run. Conversely, where a large statistical uncertainty is caused by real variation in the  
317 measured property within the measurement period, a shorter averaging time must be used. This is  
318 the approach we use when comparing the BAe-146 and P3 aircraft, and here we average the data to  
319 0.1 Hz to balance real variation with instrument noise.

320  
321

322 Once a set of points for comparison has been gathered, we compare the variables using orthogonal  
323 distance regression (ODR) with results summarised in Table 3 and shown in more detail in the  
324 Supplement (sect. S7). These straight-line fits utilise the uncertainty in both the x and y variables  
325 (taken to be the standard error, equal to the standard deviation divided by the square root of the  
326 number of data points), to produce a fit uncertainty that accounts for the measurement uncertainty  
327 of each data point used to produce the fit. Comparison between the different platforms can then

328 take place by comparing the slopes of the fits. Where they are different from unity both the  
329 statistical uncertainty of the fit and the systematic uncertainty in both instruments may contribute.  
330 When quoted in literature, this systematic uncertainty tends to be the calibration uncertainty,  
331 although other factors such as different inlets tend to make this uncertainty larger. Summary values  
332 of ODR fits for all parameters are to be found in Table 3. More completed tabulated results available  
333 in the Supplement (Table S2).”

334  
335 *Since a major point of the motivation is biomass burning aerosol, the discussion, and*  
336 *presentation of the aerosol particle light absorption coefficient is, in my opinion, not*  
337 *sufficiently addressed. Please also provide profiles of aerosol particle light scattering and*  
338 *absorption and a discussion of those.*

339 We now include a plot of the profile of aerosol optical absorption, Fig 4 (g) for completeness. We  
340 also now include the profile of aerosol optical scattering. We suspect the NASA PSAP data to have an  
341 artefact of sampling, and we discuss this in the text. It is likely related to the nature of changing  
342 pressure on the sample flow to the filter and is the reason that we do not include data from the  
343 profiles in the subsequent analysis of aerosol optical absorption. Ideally we would have loitered at a  
344 fixed altitude once we located the elevated pollution plume, but it is not possible to do such changes  
345 to the planned flight path when flying in formation as we were.

346 2.2 Minor

#### 347 **Abstract**

348 *Line 40: please add ° in the coordinates*

349 Added degree symbol

350 *Line 52: first appearance: Avoid using "well" when comparing devices. Please rephrase.*

351 Amended the text to remove ambiguous statements of “well”.

#### 352 **Instruments**

353 *Line 115: Although referenced, no details on the SMPS of the AMS rack are presented in*  
354 *section 2.4.2*

355 Added reference to SMPS in Sect. 2.6.

356 *Line 121: exemplarily for other referencing parts in the manuscript. For all references, a*  
357 *period should adjoin the subsection. Instead of 2.52, it should read 2.5.2.*

358 Changed the section numbering to have consistent format.

359 *Line 118: Provide details of the CPC by referring to section 2.6, i.e., their volume flow*  
360 *rate.*

361 Added CPC flow rates to Sect. 2.6, line 385-387, and reference to Sect 2.6 on line 119

362 *Line 120 and 124: What means good? Within which range?*

363 Added 10 to 30 % from the reference - “The inlet has been shown to efficiently transmit particles at  
364 dry diameters up to 4.0  $\mu\text{m}$  (McNaughton et al., 2007) with good agreement for submicron sized  
365 scattering aerosols between this and ground based tower observations to between 10 and 30%.”

366 *Line 126: Please provide the particle losses due to the tubing as a function of particle*  
367 *diameter used to correct those losses, e.g., in the supplementary material.*

368 These data were not corrected for modelled sampling losses due to pipework. Instead the sampling  
369 system was designed to minimise losses to a negligible level at the design stage of the instrument  
370 rack. The figures are not available here.

371 *Line 132: Please provide the period of the periodical change.*

372 Added details of PM10/PM1 switching regime.

373 *Line 160: split  $\text{ms}^{-1}$ ; otherwise, it is inverse milliseconds.*

374 Corrected the units.

375 *Line 186: Period after Beer's Law.*



376 Added a period.  
377 *Line 209 and repeatedly appearing along with the text: Please avoid judgmental*  
378 *adjectives such as "good."*  
379 Many instance of this have been amended with reference to errors, uncertainties, or else rephrased.  
380 *Line 217: Remove one period.*

381 Removed a period

382 *Line 259: (first appearance): Ensure the optical coefficients are properly subscripted.*  
383 Corrected optical coefficients.  
384 *Line 300 and 359: Use a uniform notation; Nafion™ or Nafion(TM)*  
385 Corrected to a standard notation.  
386 *Line 354 and 359: Explain where the dilution of the aerosol arises and the underlying*  
387 *reasons. Comment in which why this was accounted for. Leakage of the Nafion™*  
388 *membrane will bias the outside measurement with airplane cabin aerosol.*  
389 There is no accidental leakage, and aerosols can't pass across the Nafion(TM) membrane. Merely  
390 the instrument rack is designed such that the sample from outside is mixed with a clean filtered  
391 airstream, for reasons such as to provide a faster flow rate through instrumentation.

392 *Line 378: (first appearance): AAE (absorption angstrom exponent) is not  $\sigma_{ap}$ . Please*  
393 *change.*  
394 Checked and corrected instances of absorption Ångstrom exponent.

395 *Line 393: Comment or discuss where the factor of 1.8 originates from; Line 719:*  
396 *Comment on the underlying reasons for the empirical scaling factor used for the PSD.*  
397 Added commentary that details the processes of validating AMS volume concentrations with  
398 outboard PCASP, and then empirically scaling SMPS to better match the PCASP size distributions in  
399 the overlap region.  
400 *Line 400: According to the reference list, "Howell et al. (2020)" was published in 2021.*  
401 Corrected reference for Howell to (2021).  
402 *Line 428: Comment on the expected uncertainty omitting the refractive index correction*  
403 *of particles larger than 800 nm.*  
404 Added commentary to results section line 750: "A coarse aerosol mode was also present during  
405 runBL. At diameters larger than 0.5  $\mu\text{m}$ , where particle counts are much lower, Poisson counting  
406 uncertainties become significant: 40 % at 1.5  $\mu\text{m}$  and more than 200 % at 3.0  $\mu\text{m}$ . The bin  
407 boundaries of the PCASP and CDP have not been corrected for the material refractive index, which is  
408 not known. The 2DS is a shadow imaging probe and so not affected by the refractive index of the  
409 material. Detailed scientific analysis should account for the materials refractive index and not doing  
410 so here does limit the utility of the results in the probe cross-over regions. However, the magnitude  
411 of the differences between PCASPs is much larger than the combined uncertainties at supermicron  
412 diameters. The largest differences are apparent between the two probes on the FAAM BAe-146  
413 platform while FAAM PCASP2 and the NASA PCASP are in closer agreement. Only the FAAM CDP  
414 reported aerosol data in the particle diameter range 1-5  $\mu\text{m}$ , but, at larger diameters, data from 2DS  
415 probes on both aircraft cross over with CDP observations and show distributions with similar shapes.  
416 The cross over between CDP and PCASP is likely dominated by uncertainty in the larger sizes of the  
417 PCASP. This coarse mode will contribute to the total optical scattering from aerosol particles, as  
418 evidenced by the NASA runBL nephelometer data (Sect. 4.3.3) when switching between PM1 and  
419 PM10."

420 **Results:**

421 *Line 641: rephrase sensitivity to "the slope". Consistency: BAe-146 or BAe146. Choose.*  
422 Changed sensitivity to slope, and corrected to BAe-146.  
423 *Line 661: Discuss the differences in the measured CN between the two airplanes based*  
424 *on the cut-off of the CPCs.*

425 Line 709 - Added discussion on lower cut-off diameter of CPCs  
426 *Line 792: One could update Figure 9, including the separation between NIR and VIS, and*  
427 *add the corresponding integrated values.*

428 We feel that the diagram is suitable and note that the integrated values are presented in Table 8.

429 *Line 900: Please comment on the volatile nature of ammonium nitrate evaporating*  
430 *already at 20°C and its impact on the chemical composition measurements. See Schaap*  
431 *et al. (2004). Schaap, M., Spindler, G., Schulz, M., Acker, K., Maenhaut, W., Berner, A.,*  
432 *Wieprecht, W., Streit, N., Muller, K., Brüggemann, E., Chi, X., Putaud, J. P.,*  
433 *Hitzinger, R., Puxbaum, H., Baltensperger, U., and ten Brink, H.: Artefacts in the*  
434 *sampling of nitrate studied in the "INTERCOMP" campaigns of EUROTRAC-AEROSOL,*  
435 *Atmos. Environ., 38, 6487-6496, 10.1016/j.atmosenv.2004.08.026, 2004.*

436 **Added Line 980:** "Ammonium nitrate is semi-volatile at atmospheric conditions and to investigate  
437 this a model of evaporation of aerosols to the gas phase was developed after Dassios and Pandis  
438 (1999) was run for a range of atmospheric conditions and a sample temperature of 30° C and a  
439 sample residence time of 2 s. This showed that the worst case scenario losses of aerosol mass to the  
440 gas was 7 %, assuming unity accommodation coefficient, instantaneous heating upon sample  
441 collection and a single aerosol component. Pressure and relative humidity exerted much weaker  
442 controls (< 2 %). Sample residence times may well be longer on the aircraft, but the uncertainty is  
443 related to the differences between the sampling set-ups on the aircraft rather than absolute values  
444 which also reduces the impact of this on the comparisons"

445 *Line 1071: Provide a valuable reference for BBA density.*

446 Line 1124 - Added reference to Levin (2010) for BBA density.

## 447 **References**

448 *Add doi if available to each reference.*

449 DOI added where available.

## 450 2.3 General Comments

451

452 *Regarding tables: Table description on top of the tables.*

453

454 *The manuscript is very long. I recommend a revision in places that can be shortened.*  
455 *For instance, the instrument description part contains repetitive passages (e.g., gaseous*  
456 *components) and can be shortened, e.g., in the form of tables. A tabular overview of the*  
457 *instruments and corresponding parameters would be more understandable. After,*  
458 *differences between the airplanes and ARM-site regarding drying and instrument location*  
459 *(if necessary) can be explained.*

460 Descriptions moved to top of tables.

461 *Updating the colors of the fitting functions and adding the wavelength when optical*  
462 *coefficients are considered can improve figure 5.*

463 Some of the parameters in the very long Table 3 are now plotted, allowing us to move those  
464 segments of the results table to supplementary materials. We did consider rationalising some more  
465 of the text in section 2 relating to instrumentation descriptions. We considered using a table to  
466 outline the instrumentation, then referring to that table in the text. However, although long, we feel  
467 that the section is well structured which aids understanding and readability and that the many  
468 bespoke details of the individual set-ups mean that much of the text would have to remain anyway.  
469 We felt that a slight shortened but still long text, allied to a table that needed referencing would not  
470 in the end assist the reader.

471 *Figure 5, 6: Please provide the aerosol particles' volume and surface size distribution and*  
472 *their integrated and cumulative (along the diameter) sum values, e.g., in the*  
473 *supplementary material. Those would help comprehend the contribution of the different*

474 *aerosol populations to the optical properties since those are a function of the cross-*  
475 *section of the aerosol particles.*

476 **Fig 5, (new Fig 6)** this, and other figures have now been amended so that colours are used to  
477 distinguish that altitude of the measurements in most cases, or a particular instrument in others. We  
478 feel this has improved the figures. We have added the wavelength information where applicable.

479 **Fig 5, 6 (new Fig. 6, 7)**, we agree that some further information on the particle size distributions was  
480 required. In conjunction with this comments and comments in Review 1 we opted to show the  
481 particle number and particle volume distributions from the airborne comparisons these show a wide  
482 range of conditions. This is added to new Fig. 6. Volume (and mass) are parameters that models  
483 such as general circulation models tend to represent as prognostic variables. Showing these  
484 parameters gives an overview of how particles across the size range are sampled in comparison to  
485 one another.

486 The area distributions are included in the supplement. The optical properties are hugely important  
487 and a large focus of this study. There is significant complexity in the optical properties as a function  
488 of particle size, e.g. most biomass burning aerosol is sub-micron, and the composition of larger  
489 super-micron particles was not sampled. The optical properties depend strongly on composition and  
490 individual studies looking into these aspects of the science could be done, such as the study by  
491 Peers, et al. (2019).

492 We do not present cumulative distributions because we are relying on multiple probes to sample the  
493 full size range of aerosol particles. There is no obvious way to deal with the cross overs between  
494 individual probes and detailed study that produces a composite weighted fit is beyond the scope of  
495 the study. Likewise choosing an arbitrary size threshold at which to splice individual probes together  
496 would not be particularly instructive. Now we present both number and volume it is easier to see  
497 important features of the underlying aerosol size distributions.

498 *Comment on the different observed size ranges of the different AMS systems, i.e., the*  
499 *difference between ACSM and AMS when comparing the chemical composition. I am not*  
500 *an expert in that field, but could it be that this explains the observed difference?*

501 **AMS and ACSM differences:** There may be small size selection difference between the two  
502 instruments and sample inlets, of order 100 nm, but it is not envisaged that this is the driver of the  
503 differences. This is one set of comparisons that have been shown to be poor from this work, and  
504 unfortunately in this case we have not been able identify the underlying reasons.

505 Added line 1016 “The slight difference in quoted upper cut diameters of 600 nm (FAAM) and 700 nm  
506 (LASIC) do not explain these differences.”

507 *Line 1595: The specific instrument should be mentioned in the legend for each variable*  
508 *in all the figures. Change typo: its AAE (absorption angstrom exponent, not extinction*  
509 *angstrom exponent)*

510 Individual instruments are now on the legend and typo has been corrected.

511 *Figure 10a): Comment and discuss the discrepancy of one order of magnitude in the*  
512 *observed PSD of the 2DS and CDP.*

513 Overlap between 2DS and CDP poor at small end of 2DS – added comment on large sample volume  
514 uncertainties.

### 515 3. References:

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517 Moosmüller, H., Arnott, W. P., Hao, W. M., Collet Jr., J. L and Malm, W. C.: Biomass burning smoke

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520 Peers, F., Francis, P., Fox, C., Abel, S. J., Szpek, K., Cotterell, M. I., Davies, N. W., Langridge, J. M.,  
521 Meyer, K. G., Platnick, S. E., and Haywood, J. M.: Observation of absorbing aerosols above clouds  
522 over the south-east Atlantic Ocean from the geostationary satellite SEVIRI – Part 1: Method  
523 description and sensitivity, *Atmos. Chem. Phys.*, 19, 9595–9611, [https://doi.org/10.5194/acp-19-](https://doi.org/10.5194/acp-19-9595-2019)  
524 [9595-2019](https://doi.org/10.5194/acp-19-9595-2019), 2019.