

We thank the editor and reviewers for the handling and commenting of our revised manuscript. Please find below our point-by-point responses to each of the reviewers' comments, including the modifications we have made to the manuscript. Editor comments (EC) and reviewer comments (RC) are in black text and the answers to reviewers (AR) are given in **blue text**, and excerpts of the revised manuscript are given in *blue italic text*.

### **Answer to editor's comments**

EC1: L16: you here use the term “payload”, but then subsequently describe MoMuCAMS as the container for instruments, which would seem to be the real payload.

AR1: That's correct. We changed the word payload to platform, which is a more generic term describing the enclosure with all the included systems.

EC2: L20: should “a board computer” —> “a single-board computer”?

AR2: Done

EC3: L22: m3 superscript

AR3: Corrected.

EC4: L23-25: something about how the flexibility allows for different instrument configurations to be used depending on the science focus of the observations?

AR4: The following sentence has been added at line 25:

*Different instrumental combinations are therefore possible to address the specific scientific focus of the observations.*

EC5: L28: “hitherto not yet characterized” —> “previously unreported”?

AR5: The sentence has been changed as suggested.

*Here we present a characterization of the specifically developed inlet system and previously unreported instruments, most notably a miniaturized scanning electrical mobility spectrometer and a near-infrared carbon monoxide monitor.*

EC6: L31: “the feasibility of” could be omitted

AR6: It has been removed.

EC7: L39: “allows to observe” —> “allows observations of”

AR7: Corrected.

EC8: L43: maybe “key challenges in atmospheric science” since there is a realm realm of aerosol science that is unrelated to the atmosphere

AR8: The sentence has been rephrased as suggested.

*One of the key challenges in atmospheric science is understanding the large heterogeneity of aerosol particles in space and time.*

EC9: L56: “Poles” —> “Polar regions and alpine valleys are two environments where”

AR9: Done.

EC10: L106: “flies” —> “carries”?

AR10. Indeed, this sounds better. It has been corrected.

EC11: L131: “is” —> “was” or “has been” since you are referring to the deployments so far, but other platforms could be used

AR11: Done.

EC12: L160: “STAP” —> “Single-channel Tricolor Absorption Photometer (STAP; model 9406...”

AR12: Done.

EC13: S3.4: choice of refractive index (not stated/discussed) can make a difference in the analysis you have conducted. Calibration was performed with PSLs which have a RI ~ 1.61, but tropospheric particles are likely to be lower (more in the 1.50 – 1.55 range (real component) for org-sulf particles, have an imaginary component (e.g. for biomass burning/soot), and be quite different for mineral/salt (as well as possibly not spherical)), which affects sizing and consequently the measured number given the impact on the low cut size and the steepness of the size distribution in that size range.

AR13: We agree that the aerosol refractive index and shape can have a large effect on the sizing accuracy of the POPS (and most other OPCs). However, it is out of the scope of this work to characterize this effect, especially considering that it has already been investigated by previous studies (see for example Mei et al., 2020). As correctly pointed out by the editor, the representative refractive index for tropospheric particles is in the 1.50 – 1.55 range which is close enough to that of PSL to only have a minor effect on the sizing accuracy of the POPS. This was also shown in the Mei et al., 2020 study mentioned above. Nevertheless, we acknowledge that in environments with aerosols having markedly different optical properties – for example, in arid regions with a high concentration of mineral dust – the data could be significantly affected and should be treated accordingly. We added a comment for the reader at line 282:

*Note that the sizing characterization was performed with PSLs with a refractive index of 1.59. The refractive index of tropospheric aerosol particles typically is in the 1.50 – 1.55 range which is close enough to that of PSL to only have a minor effect on the sizing accuracy of the POPS. However, if measurements were to be conducted in environments with aerosols having markedly different optical properties – for example, in arid regions with a high concentration of mineral dust – the data could be significantly affected and should be treated accordingly.*

EC14: L280: describe spacing of size bins, e.g. log spaced?

AR14: The bins are indeed log spaced. The information has been simply added in the sentence at line 281-282.

*We follow therefore their recommendations by setting the POPS size resolution to 16 log spaced bins to minimize sizing errors.*

EC15: L287: “particles with diameters between 142 and 186 (bins 1 to 3) are wrongly detected” —> “the measurement of particles with diameters less than 186 nm (bins 1 to 3) are affected by measurement artifacts that result in inflated apparent particle counts that scaled with particle concentration.”

AR15: The sentence has been corrected as suggested.

EC16: L296: “in parallel of” —> “in parallel with”

AR16: Done.

EC17: L301: Might clarify/restate that the SEMS – POPS comparison shown in Fig 6b (y-axis indicates  $D_p = 180\text{-}1500$  nm) is using a slightly different lower POPS diameter  $> 186$  nm (bins 4-16—correct?) And as you have noted, the comparison is also mobility diameter vs optical diameter.

AR17: An indication about the slightly different size ranges was already present at line 305. We completed it by restating the fact that the difference in types of measured diameters and the exact size range of the POPS.

*Note that the size range of each instrument differed slightly because of respective bin limits and different types of measured diameters (i.e. mobility versus optical diameter / POPS size range for bins 4 to 14 = 186 to 1480 nm).*

EC18: L433: “are typically applied with MoMuCAMS” —> “have been utilized for sampling with MoMuCAMS”

AR18: Done.

EC19: L459: “p0 and pb is” —> “p0 and pb are”

AR19: Done.

EC20: L548: ”surface layer (SL)”

AR20: (SL) has been added.

EC21: L566: “biased to surface” —> “biased the surface”?

AR21: Indeed. It has been corrected.

### **Answers to reviewer #1 comments**

RC1: Line 56: „Poles“ should be polar

AR1: The sentence has been corrected as follows:

*Polar regions and alpine valleys are two environments where a stable boundary layer is commonly observed.*

RC2: Lin 147: please add the resulting maximum height at the typical 45° inclination angle with the 800m tether

AR2: The height at the given angles has been added. The revised sentence reads as follows:

*Generally, the zenith angle tends to stabilize between 45 to 50° at around 8 to 10 ms<sup>-1</sup>, which corresponds to a maximum altitude between 515 and 565 m a.g.l. for an 800 m long tether.*

RC3: Line 193: “height” probably has to be “eight”

AR3: Indeed, it has been corrected.

RC4: Line 194: the largest particle diameter shown in the plot is actually 3000 nm

AR4: That’s true. It was a mistake in the text. It has been corrected to 3000 nm.

RC5: Line 222-223: The plateau efficiency (A) of both CPCs is not reached at 7 nm in plot S5! Please correct

AR5: We agree that the plateau is actually located more between 8 and 9 nm, although, following equation 1, values at 7 nm already indicate counting efficiencies around 100%. The text has been revised to:

*The detection efficiency for both aMCPCs reaches a plateau between roughly 8 and 9 nm, which is in agreement with the manufacturer's specifications.*