Anonymous Referee #1

We thank reviewer #1 for constructive comments, we really appreciate their time spent reading our manuscript.

General comments: This paper demonstrates the unique aerosol and gas measurements via UASs and provides a promising data example for environmental research.

The authors presented a very useful payload for the atmospheric study and shared the exciting datasets from July 14 – July 18, 2018. However, analysis of the measurements is limited, and lack of a meaningful uncertainty estimation, which is critical for many data applications, such as the modeling evaluation.

RC1) Specific comments: P3, section 2.1: What is the typical flight operation? "The maximum endurance of these rotorcraft was about 15 minutes." (in line 61) Does that suggest the measurements with this platform only last for 15 mins? If so, it is too short for any study. Maybe the authors can explain how to operate this platform to provide meaningful vertical datasets?

AR1) For the LAPSE-RATE campaign we purchased a brand-new set of LiPo batteries, since it was the only way how to secure our operation. There was not enough time to make enough of charge-discharge cycles to rich the full batteries capacity. We put a lot of emphasis on safety during our operation, when battery voltage reached 23 V (6S 1600 mAh) during the rotorcraft ascend we started the descend to safely land. The progress in achieved maximum altitude is clearly visible from FIG 9 in the manuscript. If well cycled battery is used, the flight times are about 20 minutes, depended on environment (wind and temperature), our maximum achieved height ever with this rotorcraft was 1000 m AGL when flying in stabilized mode and conditions were perfect. Usually the combination of legal flight limits and the rotorcraft capabilities dictate the flight strategy. For example, we can choose, when measuring aerosols, do we want to characterize whole vertical column within the legal limits (3000 feet) but with poor statistics or rather have better statistics and give up on full height of the column. We made our choice and were able to observe high altitude NPF event.

Following was added, line 64: "This time was sufficient to achieve 893 m AGL (the height close to maximum COA allowed 914 m AGL), when flying in stabilized flight mode and ideal low wind conditions."

RC2) P4, section 2.2, line 89, is this "basic meteorological sensor" the Arduino Bosh BME280 sensor?

AR2) Yes, we meant Arduino Bosh BME280 to be basic meteorological sensor. It will be clarified in revised version of the manuscript.

RC3) P4, section 2.2, There is no uncertainty or accuracy information about the CPCs and OPC. The BME280 sensor accuracy mentioned in this section (in line 100) are all based on manufacturer statements, and very different from the comparison difference discussed in line 106-107. Does the field environment affect those manufacturer's accuracy? Were other sensors compared with high precision "siblings" in the laboratory, like CO2 sensors? If so, that information should be included here too. Does the KSU Matrice 600 Pro contain any other met sensors? POPS was not shielded from direct sunlight during the flights. Does that cause any overheating? What is the optical chamber temperature during the flights?

AR3) Following text was added to line 109: "The CPC calibration was done the same way as described in Hämeri et al. (2002}, the uncertainty of d50 values was determined to be +/- 0.8 nm. The total count was compared to table standalone CPC (model 3772, TSI Corp.) with the accuracy of about 20 %, when the

ambient air was sampled. There was no inter-comparison done for OPC-N2 counters against any more precise OPC."

In Barbieri et al. 2019 it was discussed that placement of the sensor (shielded/not-shielded, aspirated/notaspirated) plays dominant role rather than its own accuracy and uncertainty, that was also valid in our case, sensors shielded but not forcefully aspirated. When BME280 sensors are calibrated in environmental chamber – well controlled conditions, against national standard at FMI, their response is very satisfactory, please see FIG 1 attached.



Figure 1. T and RH calibration for 9 pcs of BME280 sensors against FMI national standard.

The KSU Matrice 600 Pro did contain Meteorological sensor (iMetXQ2, International Met Systems, Grand Rapids, MI, USA) borrowed from Oklahoma State University. However, the sensor data were found corrupted, and we were not able to recover the files for this manuscript.

Following text was added to line 170: "The airborne POPS was in direct sunshine only for a limited time due to the flights being short and was being cooled by an airflow during the flights. As a consequence, the highest optical chamber temperature recorded was 33C. The ground POPS was exposed to longer durations of direct sunshine and less airflow. As a result, the highest optical chamber temperature recorded was 53C. The manufacturer does not give a specific recommendation for the optical chamber temperature, but after reviewing the data raised no concerns about the overheating. As such, we believe that the temperatures are within acceptable ranges, especially for the flight unit."

RC4) P6, section 3.1.1, the ground meteorological comparison is meaningless because two sites are 15 km apart. Maybe change the Fig 4 (Surface vs. MURC) to diurnal variation plot (time vs. T, RH, P).

AR4) In general, during LAPSE-RATE campaign there was a lack of reference measurements. We will change the Fig. 4 to diurnal plot as suggested by reviewer, but we would like to keep the MURC data too even though they were measured at different height and 15 km distance. Figure 5 was removed from the manuscript.

RC5) P7, section 3.1.2, if I understand the section 2.2 correctly, a duplicate CPC, OPC, and POPS were operated on the ground. Do you compare them with the flying version on the ground?

AR5) Yes, we performed a short comparison (about 5 minutes) before each flight among the particle counters to check their performance, mostly visually from the laptop screen, if the number concentrations roughly correspond to each other. The rotorcraft with particle module was not in the same location as surface module, neither the particle counters were using the same inlet. The rotorcraft was standing on the camping table approximately one third of the height of surface module which was placed on the car roof. Since the provided comparison is rather semi quantitative.

The comparison of rotorcraft particle module to surface module for CPC could be seen in attached FIG. 2, we must point out that each CPC was calibrated to different D50 cut-off, the most pronounced disagreement could be seen on Jul 16th when a weak NPF took place also at the surface (the red circles).



Figure. 2 Inter-comparison of CPCs mounted on rotorcraft particle module (CPC1 and CPC2) and surface module (CPC Ground).

The comparison of OPCs in particle and surface module is shown in attached FIG. 3. In some cases, the OPC on particle module shows higher concentrations than the OPC on surface module. This might due to rotorcraft proximity to dusty surface during comparison. Similarly, when we compare normalized concentration per bin, the OPC on particle module slightly overcounts in all bins, see FIG. 4.



Figure 3. Daily comparison of total number concentration of OPCs mounted on rotorcraft particle module (OPC-N2_RC) and surface module (OPC-N2_Surface).



Figure 4. Daily comparison of normalized concentration per bin of the OPCs mounted on rotorcraft particle module (OPC-N2_RC) and surface module (OPC-N2_Surface).

There was no intentional comparison made for the pair of POPS counters, however we made a comparison of total particle number concentration using the air unit data just before the flight, when the KSU rotorcraft was ready for take-off, e.g. height was zero or close to zero, see Fig 5. The particle concertation data are slightly biased toward higher counts of air unit, on average about 10%.



Figure 5. Comparison of POPS air and surface unit particle number concentration.

We will add following to manuscript, line 253: "Also, a short inter-comparison (~5 minutes) was performed before each flight among the surface and airborne particle counters to check their performance, mostly visually from the laptop screen. Based on data postprocessing, the CPCs of particle and surface modules compared well within the manuscript stated uncertainty of 10%, except for July 16th when NPF at the surface level took place. This is due to different calibrated cut-off diameter of each CPC. The OPCs compared within factor of 2, however it has to be considered that very low particle concentrations were measured, about 2 cm⁻³ in the OPCs size range. There were no inter-comparison measurements made for POPS instruments. For more details please see the manuscript discussion."

RC6) Figure 8, "POPS and OPC-N2 overlap well over eight size bins" may overstate the comparison. The figure is in log scale, and POPS seemed to be a factor of 2 of the OPC-N2.

AR6) True, it is on average about factor of 2, the expression will be softened in revised version of manuscript.

The following was added to line 250:" The particle number size distributions measured by POPS and OPC-N2 overlap within factor of 2 over eight size bins..."

RC7) P8, section 3.2.1, The BME280 sensor has a +2 C difference comparing to the MURC. Will this difference constant for the hysteresis temperature profile? Will the observed T and RH data scientifically useful?

AR7) The temperature difference of 2 C of BME280 when compered to MURC, was the same as factory calibrated Vaisala AQT400 sensor mounted on the same rotorcraft. Many other sensors participated in the comparison showed deviation of same magnitude and direction, for details see Barbieri et al. (2019). The observed difference is very difficult to extrapolate to vertical profile, and most probably it will not be constant in whole column. In general, any comparisons to standards (P, T, RH, winds, particle counts) are of high value. The best way to do it is a comparison with in-situ measurements at different heights on the high towers or against tethered balloon system. During LAPSE-RATE the best we had was MURC with measurements at one height, 18 m AGL.

RC8) P9, section 3.2.2, it is great to see the new particle formation detection capability developed here. It will be beneficial to correct the over-counting behavior of CPC2 before calculating the delta(CPC). Line 266, if the descending rotorcraft would push aerosol particles downwards. Would the ascending flight push aerosol particles upwards? What is the ascending and descending rate of this flight?

AR8) The overcounting of CPC2 is given mostly by fluctuations in flow which is not recorded. Both CPCs are intentionally calibrated to different cut-off diameter 7 and 14 nm and if no particles smaller then 14 nm are measured, e.g. monodisperse aerosol is used in lab, the uncertainty in count will be still 10 % only due to flow instability. All CPC data are already corrected since their flow rate deviates from nominal 0.7 lpm. The TSI 3007 is a good instrument, but it was not meant for UAV measurements.

The overall movement of the airmass will be down in any case, 11 kg UAV needs to push 11 kg of air down to keep hovering.

Line 268: "Our ascent rates were approximately 5-8 and 3-5 m.s⁻¹ and descent rates were about 2-5 and 2-3 m.s⁻¹ for flights with particle module (FMI-PRKL1) and gas module (FMI-PRKL2), respectively.

RC9) Line 273, the site is about 700 m AGL. Will the 1000 AGL Flexpart dispersion model represent the 700 m AGL condition?

AR9) Here, our focus is on the air mass origin of the elevated layer of nucleation-mode particles discussed in the previous paragraph. We chose 1000 m AGL to make sure that the Flexpart simulation represents air mass movements for the elevated layer and not the surface layer below 700 m AGL.

In case of such layered structure of the atmosphere as seen on July 18th it is uncertain how accurately the underlying meteorological data can represent the vertical structure of the wind profile. Therefore, running the simulation close to the lower edge of the layer (700 m AGL) would increase the risk of simulating the surface layer instead of the elevated layer.

The following was added, line 309: "We used the Flexpart dispersion model to investigate the air mass history of the elevated layer of nucleation-mode particles observed on July 18th."

RC10) P10, section 3.2.3, It sounds that the platform motion has an impact on the POPS data. Please quantify the impactor. Does the author characterize the inlet loss for all the aerosol instruments – CPC, POPS, and OPC? It is critical to know the inlet loss for OPC during the descending and ascending because the concentration is very low – 5 cm⁻³.

AR10) Impact on POPS data had only fast movement during horizontal transects with inlet placed horizontally and facing the direction of the movement. There were very few such flights, but we have noticed the bias. This will be clarified in revised manuscript.

The following was added, line 312:" The occasional scatter in POPS data was caused by fast motion of rotorcraft when the ambient air was pushed against POPS' inlet facing the flight direction during horizontal transects and created pressure drop that internal pump did not handle very well, resulting in bias in POPS measured volumetric flow. However, there were only few such flights."

Line 165: 'During LAPSE-RATE the airborne POPS used a horizontally oriented naked inlet about 9 cm (3.5 inch) long with inner diameter of 1.7 mm (0.069 inches).

Line 172:" A duplicate POPS instrument was operated as a ground reference and was located approximately 1.8 m above the ground level and used a vertically oriented tube inlet of approximately 45 cm (18 inches) long and with an inner diameter of 3.175 mm (0.125 inches). The penetration through the inlet was estimated to be ~92% for 3 um particles and better for smaller ones.

Line 106: "Each of the CPCs used 30 cm inlet made of conductive tubing, led upwards to the center of the rotorcraft where both lines were merged to additional 10 cm piece of conductive inlet tubing, also facing upwards. Penetration for such inlet is ~90 to 99% for particle between 7-100 nm and ~99 % for 100 nm - 1 um. The CPC calibration was done the same way as described in Hameri et al. (2002), the uncertainty of d50 values was determined to be +/-0.8 nm. The total count was compared to desktop full-size more precise CPC (model 3772, TSI Corp.) with the accuracy of about 20%, when the ambient air was sampled. There was no inter-comparison done for OPC-N2 counters against any more precise OPC. Both OPCs, surface and particle module, were used with no additional inlet, those OPCs are not meant to be used with any kind of inlet due to use of fan for aerosol intake. On the rotorcraft the OPC was mounted from the bottom and middle of the carbon plate of the module, thus shielded from airmass movement and propeller eddies."

Anonymous Referee #2

RC1) The manuscript "Measurement Report: Properties of aerosol and gases in the vertical profile during the LAPSE-RATE campaign" by Brus et al. provides a description of UAS platform measurements of particles, trace gases and meteorological parameters in the San Luis Valley of Colorado in July 2018. The measurements were made as part of a multi-institution campaign associated with the ISARRA meeting that was held in Colorado that summer. The authors provide substantial details regarding the instruments, platforms and operation, and as such, the manuscript serves to provide a solid example of the kind of research that can be accomplished, and likely most effectively, using sUAS platforms. The manuscript is well organized and fairly well written, but the readability would benefit from careful copyediting for usage and punctuation. Overall, given the focus of the paper on the description of measurements and measurement systems rather than scientific analysis, I would think the manuscript would be a better fit in AMT than ACP.

AR1) We thank reviewer #2 for constructive comments, we really appreciate their time spent reading our manuscript. We will use Copernicus free copyediting service if our manuscript proceeds to publication. The submission to a new "ACP Measurement Report" was suggested by Editor as appropriate fit.

General comments:

RC2) The authors should be consistent with the description of the team (FNMI-KSU) or teams (FNMI and KSU), both are used in the manuscript, e.g. L42 "A flight team", L44 "by the FMI and KSU teams".

AR2) This will be corrected in revised version of manuscript, "FMI and KSU teams" will be used.

RC3) In terms of lower atmosphere/boundary layer studies, height AGL is more relevant or important to know than absolute altitude (MSL). Similarly, for diurnal processes such as boundary layer development, using local time is clearer than UTC, which requires the reader to calculate the local time to interpret the data.

AR3) The use of MSL altitude and UTC time was based on LAPSE-RATE organizers and participants decision. The standardization of all data sets produced during LAPSE-RATE and their interpretation was a must. The driving force for such decision were the needs of known in advance end users, the modeling and forecasting community.

We are going to use both in the manuscript text, the UTC and local time in parenthesis. In all figures the altitude MSL was changed to height AGL.

RC4) The discussion of the vertical profile observations is mostly descriptive rather than analytical, with the exception of the final paragraph of 3.2.2 (NPF), which would still benefit from substantially stronger arguments to derive the conclusions from the observations. Figures 4 and 5 are somewhat tangential to the focus of the manuscript. The inclusion of the descent data in Figure 9 clutters the picture when it has been argued in the text that the ascent data are believed to be more reliable. A single panel with both ascent and descent for comparison could be included instead to illustrate the description in the text.

AR4) Yes, we are aware of limited analysis provided in the manuscript, since it was submitted as "Measurement report".

The following test was added, line 253: "Also, a short inter-comparison ~5 minutes was performed before each flight among the surface and airborne particle counters to check their performance, mostly visually from the laptop screen. Based on data post-processing, the CPCs of particle and surface modules compared well within the manuscript stated uncertainty of 10%, except for July 16th when NPF at the surface level took place. This is due to different calibrated cut-off diameter of each CPC. The OPCs compared within factor of 2, however it has to be considered that very low particle concentrations were measured, about 2 cm⁻³ in the OPCs size range. There were no inter-comparison measurements made for POPS instruments. For more details, please see the manuscript discussion."

Figure 4 will be changed to time series, figure 5 will be omitted. Figure 9 will be split to single panels of separate ascent and descent profiles.

Specific comments:

RC5) L14: The opening sentence of the introduction is a bit absolutist. Perhaps "Most air pollution, including both gases and particulates, is released near the surface from various sources." The specific mention of wet deposition seems out of place since it is discussed nowhere in the manuscript, it could be omitted.

AR5) Manuscript text changed accordingly.

RC6) L18: The sentence "The atmospheric boundary layer can be investigated. . ." lists many measurement methods and relevant references, but then just stops without any following statement about why this list was presented. UAS are merely included as one item instead of the paragraph being used to set up the paper by describing why one might use sUAS instead of, or to complement, other methods.

AR6) UAS method will be elaborated in revised version of the manuscript.

The following text was added, line 22:" Unmanned Aerial Systems (UASs) have recently gained popularity as measurement platforms in atmospheric research. Generally, the UASs possess number of advantages over conventional platforms, since they are lightweight, low-cost, and highly maneuverable with exceptional spatial and temporal coverage. However, the UASs as measurement platforms still lack specifically designed instrumentation, since the off-the-shelf instruments were typically used. Such approach often requires extensive validation in order to ensure accurate and reliable observations in an atmospheric column,"

RC7) L46: suggest replacing "leveraging" with "utilizing" or "using"

AR7) Changed to "using".

RC8) L72: "employed a parachute system" was the parachute actually used regularly for landing, or was it there in case of emergency (power failure) and not actually needed during the campaign?

AR8) Parachute system was not used regularly for landing, only in case of emergency to save costly payload. It will be clarified in revised version of the manuscript.

The following text was added, line 78:"..and automatic launch, it was not used regularly for landing, only in case of emergency to save costly payload."

RC9) L79: If 15.5 kg GTW / 5.5 kg PW would have been too much for operation at the altitude of the SLV, what were the actual KSU Matrice operational weights for LAPSE-RATE?

AR9) The following text was added, line 85:"The actual weight of DJI Matrice 600 Pro was 9.5 or 10 kg depending on battery set used. The payload maximum total weight was about 1.1 kg: DJI Zenmuse X3 (221 g), GoPro Hero 7 (117 g), naked POPS (700 g), and Meteorological sensor (30 g, iMetXQ2, International Met Systems, Grand Rapids, MI, USA) borrowed from Oklahoma State University. Not all payload parts were used together during all flights."

RC10) *L85: "These modules were easily detached. . .swapping between sensor modules but since you were using two aircraft, did you actually swap the payloads? It didn't seem so from the description of operations.*

AR10) True, the modules were not exchanged during the campaign.

The following text was added, line 94:" These modules could be easily detached from the rotorcraft frame, allowing for swapping between sensor modules to meet the requirements of a given flight. However, this was not necessary since pair of rotorcraft was available during the LAPSE-RATE".

RC11) L90: "The CPCs were calibrated" perhaps "were set", "were adjusted"

AR11) Change to "were adjusted".

RC12) L92: The voltage applied to the TEC (or Peltier cooler) sets [or regulates], the temperature difference between the saturation and condensation regions.

AR12) Changed accordingly.

RC13) L94: given the cut-off (50%?) diameters of 7 and 14 nm how did the temperature instability limit the detection of sub-20 nm particles, how much did the cut-points vary? Was this incorporated in uncertainty, or was the delta-T measured and compensated?

AR13) The calibration was done the same way as described in Hameri et al. (2002), the uncertainty of D50 values was determined to be +/- 0.8 nm. The minimum and maximum set points for TEC, 1000 and 2000 mV, are the limiting factor to observe full nucleation mode range up to 20 nm. Using lower or higher setpoints would lead to instrument instability, this claim is based on personal communication with TSI technician. Using lower setpoint than 1000 mV could lead in range extension up to 20 nm, but compromising the device stability. We used values in safe range thus assuming the device is operating in steady state stable mode.

The following text was added, line 109:" The CPC calibration was done the same way as described in Hameri et al. (2002), the uncertainty of d50 values was determined to be +/-0.8 nm. The total count was compared to desktop full-size more precise CPC (model 3772, TSI Corp.) with the accuracy of about 20%, when the ambient air was sampled."

RC14) L96: "The platform was enclosed on all sides except the bottom with polylactic acid (PLA) foam to shield"; "Bosh" \rightarrow "Bosch"; "pressure (P), temperature (T) and relative humidity (RH).

AR14) Corrected accordingly.

RC15) L100: "6 ms and ±1 hPa for P, 1 s and ±0.5 °C for T, and 1 s and ±3% for RH."

AR15) Corrected accordingly.

RC16) L120: "oxygen"? Assumed to be ~20% of P?

AR16) Cannot comment on this, it is Vaisala proprietary compensation algorithm, confidential.

RC17) L128: suggest stating here that the AQT400 was not useful in the SLV environment.

AR17) This part just describes all sensors, usefulness of AQT400 is clearly discussed in part 4 Concluding remarks line 390.

RC18) L146: How long was the horizontal inlet?

AR18) Corrected accordingly to "...naked inlet about 9 cm (3.5 inch) long...".

RC19) L147: What was the nature/purpose of the POPS "custom electronics"?

AR19) The term simply refers to POPS being a research instrument and the electronics being custom-made by the manufacturer rather than an off-the-shelf solution.

RC20) L149: What is the expected effect of the POPS not being shielded from the sun over the duration of a profile? Did you evaluate the effect by running the instrument on the ground for the same period to see if a measurable effect occurred?

AR20) We expect the effect on the flight POPS to be small due to it being exposed only for short durations and being constantly cooled by an airflow. POPS electronics are rated for up to +85C. The effects on the ground POPS are harder to quantify. However, given that the ground POPS measurements stayed relatively flat despite the increase in temperature over the duration of the measurement we expect that the effects due to lack of shielding were small. An inter-comparison between airborne and surface POPS unit is given in answers to reviewer1.

The following text was added, line 170:" The airborne POPS was in direct sunshine only for a limited time due to the flights being short and was being cooled by an airflow during the flights. As a consequence, the highest optical chamber temperature recorded was 33 C."

Then, line 174:"The ground POPS was exposed to longer duration of direct sunshine and less airflow. As a result, the highest optical chamber temperature recorded was 53 C. The manufacturer does not give a specific recommendation for the optical chamber temperature, but after reviewing the data raised no concerns about the overheating. As such, we believe that the temperatures are within acceptable ranges, especially for the airborne unit."

RC21) L163: Typically, MSL would be denoted "altitude" and AGL "height".

AR21) Corrected accordingly: "...achieved height 893 m AGL (i.e. altitude was 3201 m MSL).".

RC22) L178: "A detailed description of the daily meteorological conditions in the SLV during the LAPSE-RATE campaign"

AR22) Corrected accordingly.

RC23) L181: The data presented in figures 4-7 seem to indicate that the surface measurements were not continuous, but only occurred during portions of each day.

AR23) Correct, "continuous" surface measurements were done with the battery power surface module only during UAV operation. No other surface measurements of aerosols and meteorological parameters were present in place of operation.

The following was added, line 211:" ...aerosol concentration (CPC and OPC-N2), but only during the UAV operation."

RC24) L219: "according to the US Environmental Protection Agency"; EPA or USEPA are not needed since there is no further reference in the paper.

AR24) There is a lack of any reference to PM measurements in the SLV, our goal was to provide at least some data. However, the sentence will be omitted in revised version.

RC25) L230: "based on which the goal" perhaps "the goal of which was to reach the highest altitude possible in a very short time."

AR25) Corrected accordingly.

RC26) L256: From figure 10 it appears that the bias between the two CPCs was fairly systematic, raising a question about either the determination of cut-off diameter or relative sampling efficiency. This potentially affects quantitative analysis of the results, but does not necessarily impact the qualitative conclusions drawn.

AA26) In Fig. 10c in the boundary layer the difference between the CPCs with 7nm and 14nm cut-offs is less than 10%, which is within the expected accuracy of TSI 3007. Considering that TSI 3007 has higher uncertainty and lower accuracy than a full-size desktop CPC when used in lab, less than 10% of concentration (in left hand side plot), could be considered as an excellent agreement. Compared to delta_CPC > 3km this constant bias is less than 5%.

The following text was added, line 109:" The CPC calibration was done the same way as described in Hameri et al. (2002), the uncertainty of d50 values was determined to be +/-0.8 nm. The total count was compared to desktop full-size more precise CPC (model 3772, TSI Corp.) with the accuracy of about 20%, when the ambient air was sampled."

RC27) *L273*: The discussion of the Flexpart back trajectories and interpretation of the resulting footprint (shown in Figure 11) is rather superficial and speculative.

AR27) Here, our focus is on the air mass origin of the elevated layer of nucleation-mode particles discussed in the previous paragraph. Please see also reply to reviewer1.

Flexpart footprint shows that power plant emissions cannot be ruled out as a source of the elevated layer of nucleation mode particles, which is relevant information with respect to earlier observations discussed in the previous section.

We have modified line 309 as: "We used the Flexpart dispersion model to investigate the air mass history of the elevated layer of nucleation-mode particles observed on July 18th."

RC28) *L293*: The description of the effect of aircraft motion on the POPS sampling is somewhat vague. How did the motion cause the effect? Was it the same for the horizontal and vertical inlet configurations (L145-147)?

AR28) Impact on POPS data had only fast movement during horizontal transects with inlet placed horizontally and facing the direction of the movement. There were very few such flights, but we have noticed the bias. This will be clarified in revised manuscript.

The following was added , line 330: "The occasional scatter in POPS data was caused by fast motion of rotorcraft when the ambient air was pushed against POPS' inlet facing the flight direction during horizontal transects and created pressure drop that internal pump did not handle very well, resulting in bias in POPS measured volumetric flow. However, there were only few such flights."

RC29) L298: "was 5 cm-3, similarly averaged PM1" should either use a ";" or ", while"

AR29) Corrected accordingly.

RC30) L304: "evaporation" "transpiration" if just from plants or "evapotranspiration" if from the environment (surface + plants).

AR30) Corrected accordingly.

RC31) L310: "from a BME280 sensor that showed a bias of about. . ."

AR31) Corrected accordingly.

RC32) Figure 10: Suggest labeling the panels in order (A, B, C) and change the references in the text to match

AR32) Corrected accordingly.

References:

Hämeri, K., Koponen, I.K., Aalto, P.P. and Kulmala, M: The particle detection efficiency of the TSI-3007 condensation particle counter, J. Aer. Sci., 33, 10, 2002, https://doi.org/10.1016/S0021-8502(02)00090-3

Measurement report: Properties of aerosol and gases in the vertical profile during the LAPSE-RATE campaign

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Abstract. Unmanned Aerial Systems (UASs) are increasingly being used as observation platforms for atmospheric applications. The Lower Atmospheric Process Studies at Elevation - a Remotely-piloted Aircraft Team Experiment (LAPSE-RATE) in Alamosa, CO, USA during July 14th - 20th, 2018 investigated and validated different UASs, measurement sensors and setup configurations. Flight teams from the Finnish Meteorological Institute (FMI) and the Kansas State University (KSU) partici-

- 5 pated in LAPSE-RATE to measure and investigate properties of aerosol particles and gases in the lower atmosphere. During the experiment, the performance of different UAS configurations were investigated and confirmed to operate reliably resulting in a scientifically sound observational dataset. As an example, concentration of aerosols - including two new particle formation events, CO₂ and water vapor, and meteorological parameters in the atmospheric vertical profile were measured during the short experiment. Such observations characterizing atmospheric phenomena of this specific environment would have not been
- 10 possible in any other way, and thus, demonstrate power of UASs as new, promising tools in atmospheric and environmental research.

1 Introduction

Most air pollution, including gases and particulates, is atmospheric aerosol particles and are released at near the Earth's surface the ground level from various sources. The dispersion, transport and removal from the atmosphere of these particles and

15 gases depends very much on wet deposition and atmospheric dynamical properties such as turbulent mixing (Tunved et al., 2013). The mixing height of the atmospheric boundary layer and wind speed and direction are influenced by a number of factors, including the amount of solar and surface stored energy and terrain inhomogeneity (Carbone et al., 2010). The atmospheric boundary layer can be investigated through various means, including balloon-borne soundings, tethersondes, dropsondes and hot-air balloons (e.g. Laakso et al., 2007; Greenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2009; Nygård et al., 2017), towers (e.g. Heintzenberg et al., 2017), towers (e.g. Heintze

- 20 al., 2011; Andreae et al., 2015), remote-sensors, including ceilometers, sodars, Unmanned Aerial Systems (UASs), Doppler lidars and radar techniques (e.g. O'Connor et al., 2010; Schween et al., 2014; Hirsikko et al., 2014; Vakkari et al., 2015) and conventional research aircrafts (Hermann et al., 2003; Twohy et al., 2002; Benson et al., 2008). Unmanned Aerial Systems (UASs) have recently gained popularity as measurement platforms in atmospheric research. Generally, UASs possess numerous advantages over conventional platforms, since they are lightweight, low-cost, and highly maneuverable with exceptional spatial
- 25 and temporal coverage. However, use of UASs as measurement platforms still lacks specifically-designed instrumentation, since the off-the-shelf instruments were typically used. Such an approach often requires extensive validation in order to ensure accurate and reliable observations in an atmospheric column, (e.g. Ramanathan et al., 2007; Jonassen et al., 2015; Kral et al., 2018; Barbieri et al., 2019; de Boer et al., 2020a; Girdwood et al., 2020).

The Lower Atmospheric Process Studies at Elevation - a Remotely-piloted Aircraft Team Experiment (LAPSE-RATE; de

- 30 Boer et al., 2020a) campaign broadly investigated the influence of continental, inhomogeneous terrain on lower atmospheric properties, land-air interactions, and the vertical distribution of aerosol particle and gas properties. LAPSE-RATE took place in the greater San Luis Valley (SLV) of Colorado during July 14th 20th, 2018. The experiment was organized in conjunction with the 6th Conference of the International Society for Atmospheric Research using Remotely-piloted Aircraft (ISARRA) and included over 100 participants from a number of universities, governmental and industrial teams. These participants supported
- 35 the coordinated deployment of 35 different UAS, completing over 1300 flights and 250 flight hours in less than a week. Distributed research flight plans were made and carried out to observe several atmospheric phenomena, including evolution of the atmospheric boundary layer during morning hours, the diurnal cycle of valley flows, convective initiation, and also concentration of gases and properties of aerosol particles.

LAPSE-RATE flights were conducted under both Federal Aviation Administration (FAA) Certificates of Authorization
40 (COAs) and FAA Part 107, with the COAs supporting flights up to altitudes of 914 m (3000 feet) above the ground level. In addition to the aerial assets, a variety of ground-based observational assets were deployed including the University of Colorado's Mobile UAS Research Collaboratory (MURC), the University of Oklahoma Collaborative Lower Atmospheric Mobile Profiling System (CLAMPS; Klein et al., 2015), two University of Colorado Doppler lidar systems, numerous radiosondes, and mobile surface instrumentation associated with vehicles and small towers deployed by a variety of institutes. For additional
45 information on LAPSE-RATE, please see de Boer et al. (2020a, b, c) and Bell et al. (2020).

- A flight team including two people from the Finnish Meteorological Institute (FMI) and one person from Kansas State University (KSU) participated in LAPSE-RATE. This publication summarizes the measurement setups and observations made by the FMI and KSU teams during the campaign. Section 2 provides an overview of the deployed rotorcraft and measurement modules, as well as an introduction of the validation techniques applied to the deployed sensors. A summary of the collected
- 50 meteorological, aerosol and gas observations demonstrates the benefits of <u>using leveraging</u> UAS for making measurements of these quantities (section 3). Finally, a concluding section outlines overall successes of the aerosol and gas payloads deployed by FMI and KSU during LAPSE-RATE. As a summary, the deployment of UAS-borne sensors provided new perspectives which could have not been collected with other profiling methods (e.g. ground-based remote-sensing instrumentation).

2 Materials and Methods

55 During LAPSE-RATE, the FMI and-KSU teams werewas stationed at a sampling site along the County Road 53, approximately 15 km north from Leach Airport (37°54'32.94"N, 106°2'6.83"W, see Fig. 1). The location was generally very quiet, surrounded by farmland on all sides, and only sporadically disturbed by passing local farm trucks. The elevation of the operations site was 2291 m (7516 feet) above the mean sea level (MSL). The FMI team conducted flights under a FAA COA with a maximum allowed altitude of 914 m (3000 feet) above the ground level (AGL). The KSU team conducted flights under FAA Part 107 with a maximum altitude of 121 meters (400 feet) AGL.

2.1 Technical description of rotorcraft

The FMI team deployed two rotorcraft (FMI-PRKL1 and FMI-PRKL2) during LAPSE-RATE. Both of these aircraft shared the same configuration and were custom-built around the Tarot X6 hexacopter platform. The maximum endurance of these rotorcraft was about 15 minutes, using brand new 22.2 V, 16000 mAh rechargeable lithium polymer (LiPo) batteries. This

65 flight time was sufficient to achieve 893 m AGL (the height close to maximum COA allowed 914 m AGL), when flying in stabilized flight mode and ideal low wind conditions. A maximum gross takeoff weight of the rotorcraft was close to 11 kg.

Flight missions were carried out in a manual (stabilized) or loiter (GPS position fix) mode, and autonomous flight missions were not carried out during the campaign. The flights were conducted using a 3DR Pixhawk PX4 flight controller running the Ardupilot software. The rotorcraft airframe was 960 mm (rotor-to-rotor) in diameter. Both rotorcraft used the same propulsion

- 70 system consisting of 340 kV brushless motors, 40A Electronic Speed Controllers and 18 inch (5.5 inch pitch) carbon fiber propellers. Such a setup supported lifting approximately 2 kg of active payload (i.e. scientific instrumentation). The rotorcraft also utilized a first-person viewer (FPV) video link, allowing real-time video broadcast from the aircraft to be viewed on a ground station monitor. The video link provided visual confirmation of the UAV performance together with telemetry information overlaid via an On Screen Display (OSD). UAV flight performance was recorded with a high resolution Digital Video
- 75 Recorder (DVR) and stored for post-flight analysis. Finally, to avoid any high energy impacts and to protect the expensive scientific instrumentation on board in case of a multi-level safety system failure, we employed a parachute system using a 60" parachute rated to 7 m.s⁻¹ descent speed for a 12 kg UAV. The parachute system had an independent power system and was configured for both manual and automatic launch, it was not used regularly for landing, only in case of emergency to protect the costly payload.
- The KSU rotorcraft was a DJI Matrice 600 Pro without any modifications, aside from payload attachments. The rotorcraft was controlled with the Matrice 600 Pro remote controller. Both DJI TB47S (6×4500 mAh, 22.2 V) and DJI TB48S (6×5700 mAh, 22.8 V) rechargeable LiPo batteries were used, alternating between battery types between flights. The maximum gross take-off weight recommended by the manufacturer was 15.5 kg, which would have resulted in a maximum payload capacity of roughly 5.5 kg. However, such a take-off weight would likely have been challenging given the high elevation of San Luis
- 85 Valley. <u>The actual weight of DJI Matrice 600 Pro was 9.5 or 10 kg depending on battery set used. The payload maximum total</u> weight was about 1.1 kg: DJI Zenmuse X3 (221 g), GoPro Hero 7 (117 g), naked POPS (700 g), and Meteorological sensor (30

g, iMetXQ2, International Met Systems, Grand Rapids, MI, USA) borrowed from Oklahoma State University. Not all payload parts were used together during all flights. During all flights the rotorcraft was additionally equipped with a DJI Zenmuse X3 eamera, and an additional GoPro HERO 2018 on some flights to provide alternative visual perspectives.

90 2.2 Description of payload modules

Both FMI rotorcraft carried custom-built payload modules consisting of a carbon fiber frame made out of tubes (O.D.=12 mm). A carbon fiber plate (30×20 cm and 2 mm thick), on which scientific instruments were attached, was centrally-mounted on the module. The $33 \times 30 \times 20$ cm (H×W×D) frame was attached between the rotorcraft's retractable landing gear on the rails supporting the battery plate (Fig. 2 and 3). These modules couldwere be easily detached from the rotorcraft frame, allowing for swapping between sensor modules to meet the requirements of a given flight. However, this was not necessary since a pair of 95 rotorcraft was available during the LAPSE-RATE. The first FMI rotorcraft (FMI-PRKL1) was equipped with a FMI-developed particle measurement module. This module consisted of two condensational particle counters (CPC, model 3007, TSI Corp., total count in range from 0.01 to $>1.0 \,\mu\text{m}$), a factory-calibrated optical particle counter (OPC, model N2, Alphasense, 16 bins size resolved range from 0.38 to 17 µm) and a basic meteorological sensor Bosch BME280 - (P, T and RH). The CPCs were 100 set calibrated to different cut-off diameters (7 and 14 nm, respectively, c.f. Altstädter et al., 2015, 2018). Such a configuration enables observation of freshly nucleated particles in a diameter range between 7 to 14 nm. The voltage applied to the Thermal Electric Device (TED) of a CPC regulates corresponds directly to the temperature difference between the saturation and condensation er regions. This temperature difference determines how fast particles grow to CPC detectable sizes (i.e. the cut-off diameter). Therefore, limitations to instrumental stability and sensitivity to changing saturator-condenser temperature differences, limited the ability to observe the full nucleation mode range up to 20 nm. TED values of 2000 and 1000 mV were 105 used for CPC1 and CPC2, respectively. Each of the CPCs used a 30 cm inlet made of conductive tubing, led from the sides upwards to the center of the rotorcraft, where both lines were merged to additional 10 cm piece of conductive inlet tubing, also facing upwards. Penetration for such an inlet was estimated to be between 90 and 99% for particles between 7-100 nm and 99% for particles between 100 nm and 1 µm. The CPC calibration was done the same way as described in Hämeri et al. (2002), the uncertainty of d_{50} values was determined to be ± 0.8 nm. The total count was compared to a desktop, full-size, more precise CPC 110 (model 3772, TSI Corp.) with an accuracy of about 20%, when the ambient air was sampled. There was no inter-comparison done for OPC-N2 counters against any more precise OPC. Both OPCs, in the surface and particle module, were used with no additional inlet as those OPCs were not meant to be used with any kind of inlet due to use of a fan for aerosol intake. On the rotorcraft the OPC-N2 was mounted from the bottom and middle of the carbon plate of the module, thus shielded from air 115 mass movement and propeller eddies. In addition to the CPCs and OPC, FMI-PRKL1 included an Arduino Bosch BME280

- sensor, located below the modular platform, to measure pressure (P), temperature (T) and relative humidity (RH). The platform was covered from all sides except the bottom, with polylactic <u>acide</u> (PLA) foam to protect sensors from solar radiation and keep the particle module thermally stable. The BME280 sensor was shielded from solar radiation, but not forcefully aspirated. The BME280 sensor has a manufacturer-stated response time and accuracy of 6 ms, ±1 hPa for <u>P pressure</u>, 1s, ±0.5 °C for <u>T</u>
- 120 temperature, and 1s, ±3% RH for RH relative humidity.

The second FMI rotorcraft (FMI-PRKL2) was equipped with a second FMI-developed module, carrying instrumentation to measure gases and basic meteorological parameters (P, T and RH). The gas module consisted of a flow-through CO₂ concentration sensor (Carbocap model GMP343, Vaisala Inc.), a CO₂ and water vapor analyzer (model Li-840A, Li-Cor Envir.) and a sensor to measure concentrations of CO, NO₂, SO₂, and O₃ (model AQT400, Vaisala Inc.). An Arduino BME280 sensor

- 125 measured pressure, temperature and humidity. The BME280 sensor was mounted identically to the respective sensor on the FMI-PRKL1. Both BME280 sensors showed a difference of about +2 hPa in pressure, +2 °C in temperature, and -12% in RH, when compared to measurements from the MURC platform during LAPSE-RATE inter-comparison flights (for details please see Barbieri et al. (2019). Both CO₂ sensors were forcefully-aspirated using micro-blowers configured as air pumps (Murata, model MZB1001T02) and leveraging custom 3D-printed housings connected to the sensor exhaust. A Gelman filter
- 130 in the sample airstream in front of both CO₂ sensors prevented contamination of the optical path. Micro-blowers were supplied with 12V DC resulting in a constant flow rate of 0.6 L.min⁻¹ to continually flush measured air through the sample cells. The GMP343 and LI-840A are non-dispersive infrared (NDIR) gas analyzers based upon a single path, dual wavelength infrared detection system. The GMP343 has a manufacturer stated accuracy of ±3 ppm ±1% of the measured reading and the LI-840A has an accuracy better than 1.5% of the measured reading for both CO₂ and water vapor.
- Both CO₂ sensors were laboratory-calibrated before and after the LAPSE-RATE campaign and showed no drift in calibration. The sensors were calibrated against standard carbon dioxide gases (traceable to WMO CO₂ scale X2007 at the FMI) at several concentrations (zero gas and 436 ppm before the campaign, and 370, 405.4 and 440.2 ppm after the campaign). Also, both sensors were tested in the lab against a calibrated high precision gas concentration analyzer (model G2401, Picarro, Inc.) at ambient CO₂ concentration. The GMP343 data were biased on average -3.4 (±1.3) and LI-840A 0.3 (±1.1) ppm. Data from
- 140 the GMP343 sensor were post-processed to account for pressure, temperature, RH (obtained from BME280 sensor) and oxygen by using the proprietary compensation algorithm of the probe obtained from Vaisala. For the Licor LI-840A, the built-in option for internal pressure, temperature and water vapor compensation of CO₂ concentration was used. Licor LI-840A water vapor was calibrated in dew point measurements against DewMaster Chilled Mirror Hygrometer (Edgetech Instruments Inc.).
- The AQT400 (Vaisala Inc.) gas sensor is based on proprietary advanced algorithms and electrochemical technology. The sampled gas causes a reduction or oxidation reaction in an electrochemical cell, thereby creating a weak electric current that is directly dependent on the volume of the measured gas. The gas content is subsequently calculated using an advanced compensation algorithm. The detection limits of the Vaisala AQT400 are 5 ppb for NO₂, O₃ and SO₂ and 10 ppb for CO, and the in-field accuracy is ±25, 60, 200 and 50 ppb for NO₂, O₃, CO and SO₂ respectively.
- A third FMI-developed sensor module was operated on the ground, consisting of a condensational particle counter (CPC, model 3007, TSI Corp.), an optical particle counter (OPC, model N2, Alphasesne) and a TriSonica Mini Weather Station (Applied Technologies, Inc.). The manufacturer stated accuracies for pressure, temperature and RH are ±3 hPa, ±0.5 °C, and ±3% RH, respectively. The surface sensor module was covered from all sides with PLA foam to protect sensors from solar radiation and keep the particle module thermally stable. The module was placed on the roof top of a car at about 2 m from the ground, the TriSonica Mini WS was mounted on a 45 cm long carbon fiber tube on top of the surface sensor module, i.e. about
- 155 2.75 m from the ground and about 75 cm from the car roof.

All FMI-PRKL1 particle module and meteorological sensors were logged at a rate of 1 Hz, except the OPC-N2 that was logged at rate of 0.5 Hz. The sensors included in the FMI-PRKL2 gas module were logged at different rates, with the meteorological variables, CO₂ and water vapor analyzer (LI-840A) logged at 1 Hz, the flow-through CO₂ concentration sensor (Carbocap, GMP343) logged at 0.5 Hz, and the gas sensor (AQT400, Vaisala Inc.) logged once per minute. The surface sensor

- 160 module data were logged at 1 Hz, except the OPC-N2 that was logged at rate of 0.5 Hz. Data from both rotorcraft and the surface sensor module were logged as ASCII comma separated files to embedded Raspberry Pi 3+ minicomputers using simple Python scripts. This information was aligned with position and orientation information from the autopilot in post-processing, using cross-correlation techniques to align the signals in time.
- The KSU Matrice 600 Pro was equipped with an optical particle counter (POPS, Handix Scientific LLC) with 16 size bins covering a range from 0.132 to 3.648 µm and measuring at 1 Hz. During LAPSE-RATE the <u>airborne</u> POPS was used both with a horizontally oriented naked inlet <u>measuring approximately 9 cm (3.5 inch) long</u> with an inner diameter of 1.7 mm (0.069 inches). and a vertically oriented tube inlet of approximately 45 cm (18 inches) long and with an inner diameter of 3.175 mm (0.125 inches). POPS included custom electronics, and its data were logged to a microSD card. POPS was attached to the top surface of the rotorcraft body with Velcro tape. It was not shielded from direct sunlight during the flights, but was kept in shade
- 170 while on the ground. The airborne POPS was in direct sunshine only for a limited time due to the short flights duration and was cooled during flight by airflow relative to the platform's motion. As a consequence, the highest optical chamber temperature recorded was 33 °C. A duplicate POPS instrument was operated as a ground reference and was located approximately 1.8 m above the ground level and used a vertically oriented tube inlet of approximately 45 cm (18 inches) long and with an inner diameter of 3.175 mm (0.125 inches). The penetration through the inlet was estimated to be 92% for 3 µm particles and better
- 175 for smaller ones. The ground POPS was exposed to a longer duration of direct sunshine and less airflow. As a result, the highest optical chamber temperature recorded was 53 °C. The manufacturer does not give a specific recommendation for the optical chamber temperature, but after reviewing the data no concerns were raised about the potential of instrument overheating. As such, we believe that the temperatures are within acceptable ranges, especially for the airborne unit.

2.3 Auxiliary data

- 180 Air mass history was investigated using the Lagrangian particle dispersion model Flexpart, version 9.02 (Seibert and Frank, 2004; Stohl et al., 2005). European Centre for Medium-Range Weather Forecasts (ECMWF) operational forecasts were used as meteorological input to the Flexpart back trajectories. The spatial resolution of ECMWF forecast is 9 km horizontally and variable in the vertical (137 model levels) and we obtained the model fields at 0.1 degree latitude-longitude resolution for an area of 10°N 75°N and 175°W 70°W. The temporal resolution of this ECMWF product is 1 hour. We obtained potential
- 185 emission sensitivity fields from backwards simulations with Flexpart to estimate air mass history (Seibert and Frank, 2004). The simulations were run 96 hours backwards and output was saved at 30 min time resolution. We used also mixing height of the atmospheric boundary layer (mixing layer height = MLH) from the ECMWF operational forecast calculated as the mean of four nearest model grid points to the measurement location.

3 Results and Discussion

- 190 Altogether, the FMI team completed 38 vertical profile flights. Of these, 14 flights were completed with the particle module and 24 flights with the gas module. The maximum achieved <u>height was</u> 893 m AGL (i.e.<u>altitudeheight</u> 3201 m MSL). Vertical profiles were performed by alternating flights between the particle and gas modules, with approximately 30 minutes between flights. The surface module logged continuously during the whole flight operation period. The first six flights conducted on July 15th were test flights designed to evaluate and fix several issues that arose due to transport of equipment between Finland
- 195 and Colorado. The KSU team completed in total 33 flights with their payload, including 40 individual vertical profiles. It should be noted that some of these profiles were redundant, made within a few minutes of each other, and repeated in the exact same location as another profile. The redundant flights were completed to test consistency of the instrument. All profiles were measured up to 121 meters AGL. During the first three days (July 15th-18th), the FMI <u>and</u>- KSU teams flight missions focused on vertical profiling of aerosol particle and gas properties. During the last sampling day (July 19th), the FMI and- KSU teams
- 200 joined the other LAPSE-RATE participants in order to conduct flights evaluating cold-air drainage from local valleys during the morning hours.

3.1 Ground observations

3.1.1 Meteorological variables

Weather forecasting and modeling support for the LAPSE-RATE campaign was provided by the National Weather Service

- 205 forecast office in Pueblo, CO and the National Center for Atmospheric Research (NCAR). In general the San Luis Valley is dry during summer months, though convections occur frequently over the surrounding mountains. A convection results in an afternoon thunderstorm which, in favorable wind and moisture conditions, can occasionally precipitate over the valley. The detailed description of <u>dailyday-to-day</u> meteorological <u>conditionssituation</u> in <u>the</u> SLV during LAPSE-RATE campaign was given in de Boer et al. (2020a).
- 210 The surface sensor module measured continuously meteorological variables (temperature, relative humidity, pressure, wind speed and direction) and aerosol concentration (CPC and OPC-N2), but only during the UAV operation. We compared observed meteorological variables to those obtained with MURC. MURC was equipped with a 15-meter high expandable mast mounted with several meteorological sensors including a Gill MetPak Pro Base Station that provided barometric pressure, temperature, and humidity; a Gill 3D sonic anemometer for 3D wind measurements and an R.M. Young Wind Monitor anemometer that
- 215 provided a redundant horizontal wind measurement (see de Boer et al., 2020c). During the majority of the campaign, MURC was positioned at the Leach Airport, about 15 km south from our location (Fig. 1). The MURC tower was the only reference measurement point in the vicinity of our location. Figures 4 A) and B) present diurnal temperature and relative humidity measured with TriSonica mini weather station mounted on FMI surface module together with MURC data for the same time span. Comparison of observed temperatures suggests that just before midday, when ambient air temperature exceeds 20 °C
- 220 and is likely homogeneously distributed over a large area, differences between values were within accuracy of sensors. During morning hours localized gradients in ambient conditions were more pronounced, resulting in significant differences (up to 4

°C) between observations at the two locations. Observations for pressure follow similar diurnal trends at the two locations (Fig. 4C). However, pressure measured by the TriSonica mini is on average 3 hPa higher than the values measured by MURC. Constant bias corresponds to about 13 meters higher elevation of MURC sensors compared to TriSonica. Wind speed and

5 direction observations with the TriSonica mini and MURC (Fig. 5) showed qualitatively good agreement in both quantities throughout the campaign days. Differences in these quantities are subject to local wind conditions due to surrounding roughness elements. A true comparison of wind sensor performances must be made side-by-side as was done during the LAPSE-RATE campaign for wind sensors deployed on UAVs (Barbieri et al., 2019). Unfortunately, our surface module was not part of the inter-comparison.

230 3.1.2 Aerosol particle concentration

The combined FMI and-KSU teams werewas the only ones to perform aerosol measurements during LAPSE-RATE. The aerosol number concentrations measured with the CPC of the surface sensor module were relatively stable during the whole campaign (July 15th -19th), and a similar diurnal cycle was observed every day (Fig. 5). The average total aerosol number concentration measured was around 1551 cm⁻³, with minimum and maximum concentrations around 1211 and 2249 cm⁻³, respectively, over the whole campaign. A slight increase in aerosol concentration occurred during the midday hours, between 235 approximately 16:00 UTC and 19:00 UTC (from 10:00 to 13:00 local time; Fig. 5). Increased anthropogenic activity in the San Luis Valley during morning hours is believed to result in an increase in production of primary particles from traffic and also creation of a pool of precursors for gas to particle conversion which activate due to daytime solar radiation. On average the total aerosol number concentration measured by POPS at the surface was about one third of that measured by CPC, around 532 cm⁻³, with minimum and maximum concentrations around 338 and 787 cm⁻³, respectively, over three days of POPS measurements 240 (Fig. 5). Occasional spikes in aerosol number concentration, up to 40 000 cm⁻³ for CPC and up to 16 000 cm⁻³ for POPS and lasting up to 3 minutes, were removed from datasets. Such high concentration peaks were caused by farm vehicles passing the sampling location. The number concentration of particles measured by OPC-N2 (particles diameter between 0.38 to 17 um) was generally very low, with an average concentration of about 1.2 cm⁻³, a minimum concentration of 0.7 cm⁻³ and a maximum concentration of 5.3 cm⁻³ (Fig. 6). Coarse mode particle number concentration made up only a marginal fraction 245 of the total aerosol number concentration. Corresponding average PM10 values were observed to be around 4.4 μ g.m⁻³, with

a minimum 0.8 μg.m⁻³ and a maximum of 127 μg.m⁻³. Such total particle number concentrations and particulate matter mass concentrations are typical for rural areas, which the San Luis Valley undoubtedly is. Continuous air quality measurements are sparse throughout Colorado, and according to The Environmental Protection Agency (EPA) website an average PM2.5
 concentration of 18±5 µg.m⁻³, PM10 concentration 58±23 µg.m⁻³ published as averaged maximum 24-hour concentration.

The particle number size distributions measured by POPS and OPC-N2 overlap within factor of 2 well over eight size bins between 0.46 to 3.5 µm, and together both particle counters cover accumulation and coarse modes (Fig. 7). The diurnal changes in number size distribution were minimal and no condensation neither evaporation processes were observed.

Also, a short inter-comparison (\sim 5 minutes) was performed before each flight among the surface and airborne particle counters to check their performance, with comparisons being completed through visual comparisons of real-time data on the laptop screen. Based on data post-processing, the particle and surface module CPCs compared well within the manuscript stated uncertainty of 10%, except for July 16th when NPF at the surface level took place. This difference is a result of the different calibrated cut-off diameters implemented in each CPC. The OPCs compared within factor of 2, however it has to be considered that very low particle concentrations were measured, about 2 cm⁻³ in the OPCs size range. There were no inter-comparison measurements made for POPS instruments. For more details, please see the manuscript discussion.

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3.2 Vertical profile observations

3.2.1 Reliability of observations

The measured profiles of temperature (Fig. 8) and relative humidity (Fig. 9) were divided between ascending and descending datasets by recorded altitude parameters. The data profiles showed quite a strong hysteresis in observed temperature and relative humidity, resulting in significant offsets between the ascending and descending profiles. Therefore, the observations are presented separately as solid curves for ascending legs and dashed curves for descending portions of the profiles. The hysteresis is a result of the flight strategy, based on which the goal of which was to reach as high altitudes as possible in a very short time. The BME280 Arduino sensor response was not fast enough to equilibrate to the quick changes in ambient conditions. Our ascent rates were approximately 5-8 and 3-5 m.s⁻¹ and descent rates were about 2-5 and 2-3 m.s⁻¹ for flights

- 270 with particle module (FMI-PRKL1) and gas module (FMI-PRKL2), respectively. Based on our experience, the strong hysteresis disappears when the ascent and descent rates are close to 1 m.s⁻¹ or slower. The correction of hysteresis error was not made, since it is the coupled effect of the wet and dry RH values and the elapsed time from the RH condition change during the fast vertical movement of the rotorcraft. The ascent data are probably closer to "true" profile and are preferred over the descend data.
- 275 Measured profiles of aerosol particles did not show any lag in instrument response during ascending and descending legs, and therefore both profiles were bin-averaged using 1 and 10 m vertical bins. Conversely, measured profiles of CO_2 revealed a slight lag in sensor response time, though both ascending and descending profiles were within the boundaries of measurement scatter. Therefore, despite the apparent time lag, we proceeded similarly as in the case of aerosols, both ascending and descending profiles were averaged using 1 m and 10 m vertical bins.
- 280 Temperature and relative humidity vertical profiles showed expected behavior; a decrease in air temperature with altitude during the first three days, July 16th-18th. A temperature inversion was observed during the first two flights completed in the early morning of July 18th. The temperature on the ground gradually increased due to increasing solar energy, resulting in the gradual development of a convective boundary layer and a general warming of the environment near the surface. At the same time, relative humidity was observed to generally increase with decreasing temperature as would be expected in well-mixed
- 285 conditions, though occasional single layers were detected with the opposite behavior (Figs. 8 and 9). During the last flight day of the campaign, when the FMI and- KSU teams joined the common LAPSE-RATE mission to monitor cold air drainage, a temperature inversion was observed. During the first flight day (on July 16th) the relative humidity was not recorded since the calibration constants were not read correctly.

3.2.2 New particle formation in the atmospheric vertical profile

290 Vertical profiles of particle number concentrations suggest that new particle formation (NPF) events were observed as an increase in particle number concentration in the nucleation mode (<20 nm) of the number size distribution. With the employed instrumentation suite, NPF can be monitored as a difference in particle number concentrations between readings from CPC1 (cut-off D_{50} = 7 nm) and CPC2 (cut-off D_{50} = 14 nm). It should be noted that in some cases the CPC2 slightly over-counted particles compared to CPC1, resulting in negative Δ CPC values. However, the difference in observed counts remains within 295 the uncertainty of the instrument (<10%) at all times.

For the entire dataset, we have attempted to separate collected measurements into three different NPF classification regimes. These can be classified as: 1) No NPF event (July 17th, see panel <u>BA</u> in Fig. 10); 2) A weak NPF event (July 16th, panel <u>A</u> **B** in Fig. 10); and 3) A strong NPF event (July 18th, Fig. 10 panel C). During July 16th, NPF event developed throughout the day, with no NPF in the morning, a slight increase in particle number concentrations, a peak in NPF during midday followed by slight decrease in particle concentration towards the afternoon. NPF was observed at the surface throughout the observation

period.

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During July 18th, NPF event was observed from early morning (8:03 local time) and continued until midday (10:59 local time), see panel C in Fig. 10. During this event, NPF took place only at high altitudes (about 3000 m MSL or 700 m AGL) and it was not observable at the surface at all. In panels A-C of Fig. 10 the data only from ascending flights are shown for clarity since the descending rotorcraft would push aerosol particles downwards. This bias caused NPF events to be visible at much

lower altitudes (about 2600 m MSL or 300 m AGL) compared to observations made during ascending flight. Previously, similar elevated layers of nucleation-mode particles have been observed in a number of different environments including for instance clean marine (e.g. Wehner et al., 2015), boreal forest (e.g. Leino et al., 2019) and other continental locations with varying amount of anthropogenic influence (e.g. Wehner et al., 2010; Platis et al., 2016; Altstädter et al., 2018; Junkermann et al., 2018; Qi et al., 2019). Junkermann et al. (2018) linked their observations with elevated emissions from fossil fuel combustion.

However, other sources likely contribute to particle formation in the cleaner environments.

We used the Flexpart dispersion model to investigate the air mass history of the elevated layer of nucleation-mode particles observed on July 18th To investigate the origin of air masses we ran the Flexpart dispersion model. The 96 h backwards footprint for July 18th at 20:00 UTC arriving to SLV at 1000 m AGL is presented in Fig. 11. On this day, the bulk of the air mass is

315 transported from West and was over the Pacific Ocean 72-96 h before arrival at San Luis Valley. A minor fraction of the air mass has continental origins between the measurement location and west coast of North America. A branch of this continental contribution covers the location of San Juan coal fired power plant approx. 200 km SW of the San Luis Valley (Fig. 11). Therefore, it is possible that the elevated layer of nucleation mode particles would originate in the power plant emissions. Decoupling of the layer above 1 km from the surface can be observed as substantial drop in water vapor concentration at about 200 500 m AGL (see Fig. 10 panel C).

320 500 m AGL (see Fig. 10 panel C).

The presented observations on NPF support the following conclusions: 1) Precursor vapors can be distributed within mixing height of the atmospheric boundary layer resulting in NPF in deep vertical extend (July 16th, Fig. 10 panel AB). 2) In addition

to vapor precursors, sink due to background aerosol for particles from NPF must be small enough. Levels of background aerosol concentrations are discussed below and support this conclusion. 3) Influence of long-range transport can be observed

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in the vertical profile. Free tropospheric NPF is known to be a significant source of secondary aerosol over both marine and continental regions (e.g. Hermann et al., 2003; Benson et al., 2008; Merikanto et al., 2009). In such a situation, the surface area concentrations are usually observed to be very low (e.g. Twohy et al., 2002).

3.2.3 Accumulation and coarse mode aerosol particles

Vertical profiles measured with POPS up to 140 m AGL are presented as diurnal 1 m averages in Fig. 12. The profiles did
not show any variation with increasing altitude, the averaged particle concentration of 672 cm⁻³ with minimum and maximum concentrations 500 and 1727 cm⁻³, respectively. Flat profile concentrations correspond to those measured with POPS at surface. Similarly, averaged diurnal number size distribution did not go through any observable dynamical changes resulting from growth or evaporation of particles. The occasional scatter in POPS data was caused by fast motion of the rotorcraft, resulting in acceleration of the ambient air against the POPS' inlet in line with the flight direction during horizontal transects. Such acceleration created a pressure drop that the internal pump system did not handle very well, resulting in bias in POPS measured

volumetric flow. However, there were only few such flights.

Vertical profiles of particles measured by OPC-N2 did not show any significant variation with increasing altitude, and thus indicating a well-mixed boundary layer throughout the measurement column (Fig. 13). The averaged particle concentration was 5 cm⁻³, <u>while</u> similarly averaged PM1, PM2.5 and PM10 were 3.1, 4 and 9.7 µg m⁻³, respectively. The main contribution to mass was made by aerosol particle load close to the surface. The result implies a lack of elevated plumes, despite the fact that there were extensive forest fires in south Colorado before and during the LAPSE-RATE campaign.

3.2.4 Gas concentrations in the atmospheric vertical profile

- Concentrations of CO_2 in the vertical profiles depend on both local sources and sinks as well as on atmospheric conditions including altitude, atmospheric mixing state and transport. The CO_2 and H_2O vapor profiles showed higher concentrations only near the surface due to sources from soil and vegetation canopy <u>evapotranspiration respiration and evaporation</u>. Averaged semi-diurnal (morning to early afternoon) vertical profiles of CO_2 and water vapor during July $16^{th} - 19^{th}$ for both probes (Vaisala GMP343 and Licor LI-840A) are presented in Fig. 14. Even though both probes were calibrated in the laboratory at sea level with satisfactory results, they showed different absolute CO_2 concentrations in the field (SLV). The difference in CO_2 concentration increased with increasing altitude, and the GMP343 was found to measure CO_2 gradients independent of
- the time of day. This might be the result of imprecise pressure compensation for the GMP343 as provided by Vaisala (see the Section 2 for details). Since we compensated GMP343 with temperature, RH and pressure obtained from BME280 sensor that showed <u>a bias</u> of about +2 °C in temperature, -12% in RH and +2 hPa in pressure during the LAPSE-RATE inter-comparison measurements against MURC (Barbieri et al., 2019), we also accounted for that bias in compensation resulting in an increase of CO₂ concentration by only 2 ppm. Very little variation in CO₂ concentrations was observed as a function of altitude on

355 July 16th and July 17th (Fig. 14), indicating a well-mixed boundary layer. The same holds for water vapor concentration.

However, on July 17th elevated surface concentrations were observed. During July 18th and 19th, profiling started earlier in the morning (about 13 p.m. and 12 a.m. UTC, 7 and 6 a.m. local time, respectively) and we were able to observe gradients in CO₂ concentration between the rotorcraft and the ground level. These gradients extend to approximately 100 m from surface on July 18th and 250 m on July 19th. Similarly for water vapor concentration, on July 18th and 19th the gradient extends the whole profile.

Figure 15 demonstrates a notable change in CO_2 concentration with increasing altitude on July 19th, where the first five flights from 11:44 to 13:41 UTC show a near-surface CO_2 gradient. This gradient is detected to include a decrease of approximately 30 ppm (450 to 420 ppm for the LI-840A, and 430 to 400 ppm for the GMP343) in first 350 m above surface. During the subsequent five flights (14:11 to 16:42 UTC, 8 and 10 a.m. local time), the CO_2 concentration profiles began to be more uniform all the way to the surface, with hints of a slight negative gradient observed with the LI-840A. A negative gradient

- was not observed with GMP343 sensor, probably due to its lower sensitivity. Such a negative gradient could be explained by ground vegetation photosynthetic uptake and the vertical transport of CO_2 to higher altitudes, as reported e.g. by Li et al. (2014). However, no ground flux measurements were present during the campaign to support this hypothesis. Water vapor concentration showed an opposite trend, with the profile being flat in the early morning (about 7 g.kg⁻¹) and the concentration
- at the surface gradually increasing up to 8.5 g.kg⁻¹, and at 350 m above the surface it was 6.75 g.kg⁻¹ for the last flight at 16:42 UTC.

Unfortunately, we were not able to acquire vertical profiles or ground level concentrations of NO₂, O₃, CO and SO₂ with the Vaisala AQT400 sensor. All data collected during LAPSE-RATE were far below the manufacturer declared detection limits. The AQT400 sensor is rather suitable for detection or identification of sources with higher concentrations, though this is not supportive of detailed measurements in clean environments with very low background concentrations of both gases and particulate matter. Furthermore, the AQT400 standard calibration is valid for the range 800 - 1200 hPa, and given the elevation of the San Luis Valley (about 2300 m MSL) the ambient pressure during the campaign was around 780 hPa at the surface, impacting the accuracy of the calibration significantly.

4 Concluding remarks

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380 Flight teams from the Finnish Meteorological Institute and the Kansas State University participated in LAPSE-RATE campaign in Sun Luis Valley, CO, USA during July 14th - 19th, 2018. The teams were operating from the same location and created a sound dataset on the properties of aerosol particles and gases in the atmospheric vertical profile. During the campaign the FMI modular system with off-the-shelf instrumentation was tested.

From deployment of UAS-borne off-the-self sensors in a high-altitude environment we learned that particle counters used during the LAPSE-RATE campaign worked flawlessly together. Even though they were deployed on different platforms the obtained number size distributions overlapped well over several size bins. The particle counters together covered aerosol particles ranging between 7 nm to 18 µm in diameter, however with a gap in the size range between 15-130 nm. Such a setup in principle allows very affordable observations of new particle formation events (nucleation mode), particle growth by condensation and coagulation (accumulation mode), and dust episodes (coarse mode) in vertical profile. We also learned that

- the Vaisala GMP343 CO₂ concentrations and vertical profiles do not match Licor Li-840A after post-processing the data due 390 to not very precise pressure correction provided by manufacturer. The precision and response times of Li-840A remain better. Water vapor content profiles obtained by Li-840A compared well against those calculated from radiosonde profiles. The air quality sensor Vaisala AQT400 is more suitable for environments with elevated levels of gas pollutants, in clean background location like SLV the measured values remained lower than the manufacturer stated detection limits.
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During the short measurement campaign features of aerosol particles, CO₂ and water vapor specific to the Sun Luis Valley were acquired. As an example, a new particle formation event at altitudes at and above 700 m above the surface, decoupled from the ground, was observed. This is an observation that could not easily be obtained by known remote sensing methods, but is easily collected using airborne in-situ methods.

Data availability. Data sets collected by Finnish Meteorological Institute and Kansas State University during LAPSE-RATE were published 400 together under the LAPSE-RATE community at Zenodo open data repository https://zenodo.org/communities/lapse-rate/.For data collected using the platforms outlined in this paper, the Finnish Meteorological Institute FMI-PRKL1, FMI-PRKL2 and FMI surface module data sets can be all found here: https://doi.org/10.5281/zenodo.3993996, Brus et al. (2020a). The Kansas State University KSU M600 and KSU surface data sets can be found both here: https://doi.org/10.5281/zenodo.3736772, Brus et al. (2020b).

Author contributions. D.B., A.H., G.B., O.K. and V.V. writing, reviewing and editing of manuscript, D.B., J.G., O.K. conducted the mea-405 surements. D.B. made dataset analysis and validation. V.V. made Flexpart analysis.

Competing interests. The authors declare no conflict of interest.

Acknowledgements. Authors would like to acknowledge following foundation for providing financial support of the project: KONE foundation, ACTRIS-2 - the European Union's Horizon 2020 research and innovation programme under grant agreement (No 654109), ACTRIS PPP - the European Commission under the Horizon 2020 - Research and Innovation Framework Programme, H2020-INFRADEV-2016-410 2017 (Grant Agreement number: 739530), Academy of Finland Center of Excellence programme (grant no. 307331) and the US National Science Foundation CAREER (1665456). In addition, Limited general support for LAPSE-RATE was provided by the US National Science Foundation (AGS 1807199) and the US Department of Energy (DE-SC0018985) in the form of travel support for early career participants. Support for the planning and execution of the campaign was provided by the NOAA Physical Sciences Division and NOAA UAS Program Office. Finally, the support of UAS Colorado and local government agencies (Alamosa County, Saguache County) was critical in securing site 415 permissions and other local logistics. D.B. and J.G. would like to especially acknowledge Dave L. Coach for acting as a Pilot-In-Command

for FMI team. Handix Scientific, LLC is acknowledged for providing their POPS instruments for the campaign at no cost.

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Figure 1. Map of wider San Luis Valley, the insert shows location of FMI - KSU team spot, and distance from Leach Airport. The airfield is located approximately 3.2 km ENE of the commercial district of Center, Colorado and 32 km NNW of Alamosa, Colorado. Background map courtesy of ©Google Maps.



Figure 2. Particle module attached to Tarot X6 hexacopter at the take-off/landing spot - blue tarp.



Figure 3. FMI-KSU team at dedicated spot during LAPSE-RATE campaign, gas module attached to Tarot X6 hexacopter during airborne operation. Ground module with TriSonica mini can be seen on the roof of SUV.



Figure 4. Diurnal values measured with TriSonica mini mounted to FMI surface module compared to observations by MURC tower positioned at Leach Airport A) temperature, B) RH and C) pressure. The local time is minus 6 hours from UTC



Figure 5. Total aerosol number concentration (minute averages) measured with CPC TSI 3007 (from July 15th to 19th) and POPS (from July 16th to 18th) at the surface.



Figure 6. Total aerosol number concentration (minute averages) measured with OPC-N2 at the surface from July 15th to 19th.



Figure 7. Aerosol number size distribution (diurnal averages) measured with OPC-N2 and POPS (plotted separately) at surface from July 15th to 19th. The grey area represents overlapping sizes of bout both particle counters.



Figure 8. Vertical profiles of temperature measured by BME280 Arduino sensor from July 16^{th} – July 19^{th} , solid lines (<u>left column A</u>)–ascend data and dashed lines (<u>right column B</u>)–descend data



Figure 9. Vertical profiles of relative humidity measured by BME280 Arduino sensor from July 16^{th} – July 19^{th} , solid lines <u>(left column A)</u>–ascend data and dashed lines (right column B)–descend data



Figure 10. Vertical profiles of total aerosol measured with two CPC TSI 3007 in particle module during July 16^{th} - 18^{th} , CPC1 with cut-off diameter $D_{50} = 7$ nm (solid lines) and CPC2 with $D_{50}=14$ nm (dotted lines). Observation of new particle formation (NPF) event: A) A weak NPF event measured throughout the measurement heights on July 16^{th} , the event developed throughout the day with no NPF in the morning, followed by a slight increase in particle freshly formed particle number concentration during midday and decreasing during the afternoon. B) no NPF event measured on July 17^{th} . C) A strong NPF event was observed on July 18^{th} from 700 m AGL (3000 m MSL) upwards through early morning (8:03 local time) and midday (10:59 local time). Water concentration vertical profiles measured with Licor LI-840A and calculated from balloon sounding's parameters demonstrates the decoupling of free troposphere nucleation from the atmospheric boundary layer.



Figure 11. Emission footprint calculated by Flexpart dispersion model for July 18th 96h backwards, arriving at 20:00 UTC at 1000 m AGL.



Figure 12. Daily averaged vertical profiles of total particle count and number size distributions measured with POPS, from July 16th-18th.



Figure 13. Averaged vertical profile OPC-N2 coarse particle total count - black line from July 16th-18th. Averaged vertical profiles of PM1, PM2.5 and PM10 values measured with OPC-N2-black line, from July 16th-18th.



Figure 14. CO_2 and water vapor vertical profiles measured with Licor LI-840A and Vaisala GMP343 on July 16th-19th. Data plotted as 1 meter (dotted lines) and 10 meter averages (solid lines). Please note different <u>heightaltitude</u> on daily figures.



Figure 15. CO_2 and water vapor vertical profiles measured with Licor LI-840A and Vaisala GMP343 on July 19th. Early morning data measured with LI-840A show visible gradient with higher CO_2 concentrations at the ground level, the profile flattens with increased Sun activity. The profiles measured with Vaisala GMP343 show weaker gradient because of its lower sensitivity. Water vapor profiles were firstly flat with gradient developed with time due to increased solar radiation.