Dear reviewer,

The reviewer’s comments were highly insightful and enabled us to improve the quality of our manuscript. Our point by point responses to the each of the comments in the following pages. We hope that the revisions in the manuscript and our accompanying responses will be sufficient to make our manuscript suitable for publication in *Atmospheric Measurement Technique*.

Changes to the revised manuscript are shown in green.

We shall look forward to hearing from you at your earliest convenience.

Yours sincerely,

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OPEN DISCUSSION #3

Question 1

The manuscript is well structured, however the last two paragraphs of the introduction read like a summary. They forestall important results from the following sections. The introduction is not the right place for this information. Please change and shorten these last two paragraphs. My suggestion, instead of presenting results, you should write something like: "Traditional CPC geometries do not allow for a much smaller size and weight … they are not suited for batch production … we tried a new technique … based on …" This would fit perfect to the end of the introduction. But this is just a suggestion.

Answer 1

We thank for your kind advice.

We modified the '1st paragraph of Introduction' in the revised manuscript as following,

In this study, we developed a high-performance MEMS-based CPC that is portable, inexpensive, and power-efficient. Our system comprises a MEMS-based condensation chip and miniature optical particle counter (OPC). UFPs are grown to micrometer-sized droplets on the chip and the grown droplets are detected by the miniature OPC. New fabrication techniques including MEMS and 3D printing technologies are applied to CPCs; particle enlargement (i.e., the fundamental process of the CPC) can be realized on a chip-scale system, because the essential elements for growing droplets (i.e., channels, micropillar-type wick, heaters, temperature sensors) are integrated on a single glass slide. Accordingly, our system shows far more compact and cost-efficient than traditional CPCs even including their portable versions.

In addition to its compactness, our system also provides high degrees of accuracy and precision. A quantitative characterization using Ag particles proves that our system is capable of growing UFPs to micrometer-sized droplets, counting them one by one, and thereby measuring UFP number concentration with a high accuracy, which is comparable to commercial OPC. These results show that our system can potentially be used as a portable, low-cost, and high-precision UFP sensor for various fields (e.g., assessing UFP exposure, monitoring workplaces, tracing particle sources in high-precision industries with cleanrooms). Moreover, when combined with the recently developed miniature DMA, our system should also be able to perform onsite monitoring of the UFP size distribution with high resolution(Liu and Chen, 2016; Qi and Kulkarni, 2016).

Question 2

Title:

- Please write-out "MEMS", as this abbreviation is relatively unknown to the atmospheric community.

- Please remove "at a point of interest" because this statement I not very specific and a user of this new instrument will always only measure "at a point of interest". Moreover, I´m not a native speaker, but it seems for me that this wording might be understood as "sight-seeing point" as well.

Same for the abstract, remove "at a point of interest" as well; there it also causes a reference error, the "point of interest" is not "portable, …".

Answer 2

We thank for your advice. We wrote-out "MEMS" in the title, and removed the phrase, "at a point of interest", throughout the revised manuscript.

Question 3

Abstract:

- p. 1, l. 15: Please specify if the given size information (nm) are for particle "diameter" or "radius". Please do so in the whole manuscript.
- p. 1, l. 16: Please specify which "deviation" is meant, standard deviation?

Answer 3

Following your advices,

We modified the terms 'Abstract' in the revised manuscript as following,

Abstract. We present a micro-electro-mechanical systems (MEMS)-based condensation particle counter (CPC) for sensitive and precise monitoring of airborne ultrafine particles (UFPs) that is portable, inexpensive, and accurate. Our system consists of two main parts: a MEMS-based condensation chip that grows UFPs to micro-sized droplets and a miniature optical particle counter (OPC) that singly counts grown droplets with the light scattering method. A conventional conductive cooling–type CPC is miniaturized through MEMS technology and 3D printing technique, and the essential elements for growing droplets are integrated on a single glass slide. Our system is much more compact (75 mm × 130 mm × 50 mm), lightweight (205 g), and power-efficient (2.7 W) than commercial CPCs. In quantitative experiments, the results indicated that Our system can detect UFPs with a diameter of 13.4 nm by growing them to micro-sized (3.16 µm) droplets. Our system measured the UFP number concentration with high accuracy (mean difference within 4.1 %), and the number concentration range where our system can singly count particles was characterized as 7.99–6850 cm^{-3}. Thus, our system has a potential of being used for UFP monitoring in various environments (e.g., air filtration system, high-precision industries utilizing cleanrooms, indoor/outdoor atmospheres).

Question 4

p. 1, l. 22: "Monitoring of airborne ultrafine particles …yield(s !) enhancement in industry fields"

I can imagine what the authors meant with this sentence, but actually I do not understand it. Please rephrase.

Answer 4

We thank for your advice. We have rephrased the sentence and added a detailed explanation in the manuscript to help the reader understand.

We modified the '1st paragraph of Introduction' in the revised manuscript as following,

High-precision industries with cleanrooms also require UFP monitoring to trace their sources, minimize the contamination, and thereby increase the production yield. For instance, in case of the semiconductor industry, as the minimum feature size of the semiconductor devices approaches to 7 nm, particles with the diameter of a few nanometers are critical (Neisser and Wurm, 2015). Although the air-purification system equipped with ultra-low particulate (ULPA) filter eliminate the contaminants in the air entering the clean room, it cannot control the internally-generated UFPs during the manufacturing processes (e.g., chemical vapor deposition (CVD), metallization, wet etching) (Choi et al., 2015; Manodori and Benedetti, 2009). If they are deposited on electrodes of a chip, they cause the interruption of the current flow, making the whole chip unusable and thereby reducing the yield of the semiconductor device (Libman et al., 2015). In this regard, ISO 14644-12 has been recently developed to guide how to monitor UFPs in cleanrooms. In order to monitor the concentration field of UFPs in these environments where the spatial and temporal variations of UFP concentrations are enormous, a portable and low-cost sensors are required to establish simultaneous monitoring at multiple points or establish dense monitoring networks.

Question 5

p. 1, l. 22: Some reference in the introduction seem for me to be very old, many 199Xs. There might be some very fundamental among those, but the last 20 years definitely brought some progress. Please check if there are newer references.

Answer 5

Thank you for your advice. We added recent references to the first paragraph of the introduction.

We modified 'Introduction' part of the revised manuscript as following,

Monitoring of airborne ultrafine particles (UFPs), which are smaller than 100 nm, is needed in various fields for human health and yield enhancement in industrial fields(Donaldson et al., 1998; Donovan et al., 1985; Hristozov and Malsch, 2009; Li et al., 2016; Liu et al., 2015). UFPs are mainly generated from burning fossil fuels and are ubiquitous in urban air; they account for about 90% of the total particle number concentration(Kim et al., 2011; Kittelson, 1998; Shi et al., 1999). Because of dramatic developments in nanotechnology, engineered UFPs for commercial and research purposes have been produced at a large scale. These incidentally and intentionally generated UFPs are more harmful to human health than larger counterparts: UFPs have a higher chance to deposit in the lower respiratory system and are more toxic owing to their larger surface-to-volume ratios, which causes oxidative stress, pulmonary inflammation, and tumor development(Hesterberg et al., 2012; Hext, 1994; Li et al., 2003; Renwick et al., 2004). Thus, onsite monitoring is needed to assess and minimize UFP exposure. High-precision industries with cleanrooms also need UFP monitoring to increase the production yield. For instance, in the semiconductor industry, the UFP minimum linewidth of the chips is approaching 7 nm(Neisser and Wurm, 2015). Particles that are a few nanometers in size are critical because "killer particles" (i.e., the diameter is greater than half of the minimum linewidth) can render the whole chip unusable(Libman et al., 2015). Unfortunately, since UFPs in cleanrooms are generated during fabrication processes (e.g., chemical vapor deposition (CVD), metallization, wet etching), contamination can occur in any manufacturing stages(Choi et al., 2015; Manodori and Benedetti, 2009). In these circumstances, a portable and low-cost sensor is needed for onsite UFP monitoring to accurately evaluate adverse health effects and control the contamination level in cleanrooms to enhance the production yield.

We have added the references in the 1st paragraph of the introduction in the revised manuscript.

(1) Li, N., Georas, S., Alexis, N., Fritz, P., Xia, T., Williams, M. A., Horner, E., and Nel, A.: A work group report on ultrafine particles (American Academy of Allergy, Asthma & Immunology): Why ambient ultrafine and engineered nanoparticles should receive special attention for possible adverse health outcomes in human subjects, J Allergy Clin Immunol, 138, 386-396, 2016.

(2) Liu, L., Urch, B., Poon, R., Szyszkowicz, M., Speck, M., Gold, D. R., Wheeler, A. J., Scott, J. A., Brook, J. R., Thorne, P. S., and Silverman, F. S.: Effects of ambient coarse, fine, and ultrafine particles and their biological constituents on systemic biomarkers: a controlled human exposure study, Environ Health Perspect, 123, 534-540, 2015.

(3) Kim, K. H., Sekiguchi, K., Kudo, S., and Sakamoto, K.: Characteristics of Atmospheric Elemental Carbon (Char and Soot) in Ultrafine and Fine Particles in a Roadside Environment, Japan, Aerosol and Air Quality Research, 11, 1-12, 2011.

(4) Hesterberg, T. W., Long, C. M., Bunn, W. B., Lapin, C. A., McClellan, R. O., and Valberg, P. A.: Health effects research and regulation of diesel exhaust: an historical overview focused on lung cancer risk, Inhal Toxicol, 24 Suppl 1, 1-45, 2012.

Question 6

p. 1, l. 23: "UFPs are mainly generated from burning fossil fuels …". This statement is not generally true, the main particle formation process is gas to particle formation and fossil fuel burning might be dominant in cities only.

Answer 6

We thank for your advice and letting us improve the solidity of the manuscript.

We modified the 1st paragraph of 'Introduction' in the revised manuscript as following,

Monitoring of airborne ultrafine particles (UFPs), which are smaller than 100 nm, is needed in various fields for human health and yield enhancement in industrial fields (Donaldson et al., 1998; Donovan et al., 1985; Hristozov and Malsch, 2009; Li et al., 2016; Liu et al., 2015). While they have a variety of anthropogenic and natural source, in urban area, UFPs are largely generated from the vehicle exhaust (e.g., soot agglomerates, secondary particles from hazardous gaseous precursors) (Kim et al., 2011; Kittelson, 1998; Shi et al., 1999). Moreover, because of dramatic developments in nanotechnology, engineered UFPs for commercial and research purposes have been produced at a large scale. These incidentally and intentionally generated UFPs are more harmful to human health than larger counterparts: UFPs have a higher chance to deposit in the lower respiratory system and are more toxic owing to their larger surface-to-volume ratios, which causes oxidative stress, pulmonary inflammation, and tumor development (Hesterberg et al., 2012; Hext, 1994; Li et al., 2003; Renwick et al., 2004). Thus, onsite monitoring is needed to assess and minimize UFP exposure.

Question 7

p. 2, l. 6: Please insert "e.g." before mentioning the TSI 3007". There are other models as well, e.g. from KANOMAX.

Answer 7

We modified 'Description of the MEMS-based CPC' part of the revised manuscript as following,

However, commercially available CPCs are bulky and expensive; thus, they are impractical for onsite monitoring where the UFP concentration changes continuously. Although portable CPCs (e.g. model 3007, TSI Inc., USA) are currently on the market, they are still large in size (292 mm × 140 mm × 140 mm) and expensive (~10,000 USD) for ownership. Therefore, despite their advantages, CPCs are difficult to actively utilize for onsite monitoring applications.

We modified 'Conclusion' part of the revised manuscript as following,

In terms of compactness and cost-efficiency, our system is superior to conventional instruments. MEMS-based CPC has a far smaller physical volume than the reference CPCs and is less than 91.5 % of the portable CPC (e.g. model 3007, TSI Inc., USA) available in the market.

Question 8

p. 2, l. 35: I might have missed it, but you don´t specify in the whole manuscript which working fluid was used. Did you try different ones? This information is essential for this technical paper and should be provided to the reader.

Answer 8

We thank for letting us catch the mistakes in our manuscript. The kind of working fluid should be clearly stated in manuscript. We used Butanol as the working fluid of MEMS based CPC.

We modified '2 Description of the MEMS-based CPC' part of the revised manuscript as following,

Figure 1 shows the operating principle of MEMS-based CPC, which consists of a reservoir, saturator, condenser, and miniature OPC. To generate supersaturated vapor and hence grow UFPs to micro-sized droplets, our system utilizes a conductive cooling method. Butanol was used as working fluid. The saturator generates saturated vapor by heating the wetted wall with the working fluid.

Question 9

p. 2, l. 40: In the supplement is only one figure. It seems strange to me to have a supplement because of just one figure. I suggest to incorporate it into the main manuscript.

If I remember correctly, in the traditional CPCs the particles grow more or less to the same droplet size, but in this new type there is a strong increase in droplet size with initial particle diameter (Fig. S1). How far does this go? Are even 20 or 30 µm droplets generated? How does this affect the counting efficiency of your new CPC for larger particles (sedimentation)? Please comment in the manuscript on that, if this is an issue.

p. 5, l. 24: Again, how large do the droplet in your CPC get at maximum and is sedimentation really no problem?

Answer 9

We thank for your careful comments. Following your advice,

We moved the results of the droplet size distribution from the supplemental material to the revised manuscript, and have added the new section, '5.1 Droplet size distribution' in the result of the revised manuscript as following,

Figure 6 shows the size distribution of the droplets generated from the MEMS-based condensation chip. Monodisperse Ag particles in the size range from 20 to 140 nm were used as test aerosol and their number concentrations were fixed at around 2000 cm^{-3} by adjusting the valves of the dilution bridge. The sampling time for measuring each droplet distribution was 2 min, and the corresponding measurement uncertainty based on the Poisson statistics was 0.13 %. All the error bars at each data point represent the standard deviations. The commercial OPC (OPC-N2, Alphasense, UK) was used for measuring the droplet size distribution. It was reported that OPC-N2 was capable of not only measuring particles from 0.4 to 17.0 µm, but also having moderate counting performance compared to the reference OPC (PAS-1.108, Grimm Technologies) (Sousan et al., 2016). The measurement errors induced from the Mie resonance were not considered in this data. The average droplet diameter ($d_{d,avg}$) was 3.1 µm when particles with the size of 20 nm, slightly larger than the minimum detectable size (12.9 nm), were introduced. Since the lower detectable size of the optical detector in our system was 0.3 µm, introduced particles successfully grew into micrometer-sized droplets which were large enough to be counted by optical means. It was noted that the mean droplet size did not vary significantly above 40 nm. Also, most of the grown droplets were smaller than 10 µm, indicating that tens of micrometer-sized droplets, which could be attached to the inner walls of the condensation chip or optical detector via sedimentation, were barely generated.

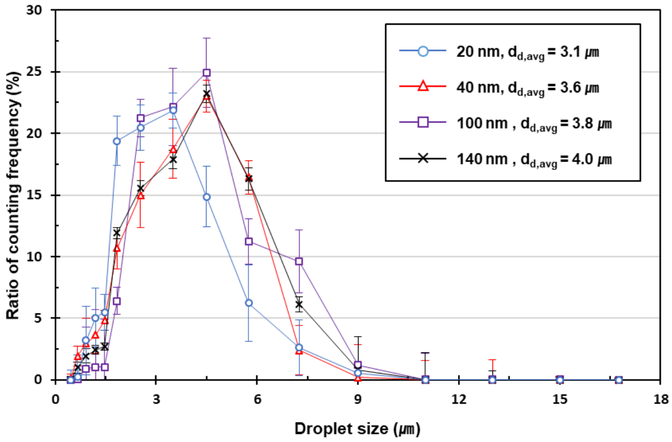

Figure 6: The size distribution of the droplets grown from the MEMS-based condensation chip when Ag particles of specific sizes were introduced.

We have added the reference which refers to the performance of OPC-N2 used in this study for measuring droplet size distribution in the revised manuscript.

Sousan, S., Koehler, K., Hallett, L., and Peters, T. M.: Evaluation of the Alphasense Optical Particle Counter (OPC-N2) and the Grimm Portable Aerosol Spectrometer (PAS-1.108), Aerosol Sci Technol, 50, 1352-1365, 2016

To evaluate the droplet loss in the condenser via sedimentation, we have performed the experiment, and added the result in '5.5 Performance comparison with the reference CPC' of the revised manuscript as followings,

Figure 10 shows the measurement results of our system when it was tilted like an inset image. Monodisperse Ag particles with 25 nm were introduced and their concentrations were step-wisely increased from 0 to 4000 cm^{-3}. Since the measurement was carried out for about 500 s at each angle, and the measurement uncertainty of each section was below 0.01 %. When our system was oriented perpendicular to the surface, the counting efficiency of our system was 2.04 %. When a 30° angle was applied, the counting efficiency was 7.07 %. At 60°, the measurement difference compared to the reference CPC exceeded 10 % (16.3 %). Thus, it was found that, at a tilt angle of 60° or less, MEMS-based CPC can monitor UFPs without the significant degradation of the accuracy.

The deviation of the counting efficiency induced from applying a tilt angle can be explained by the sedimentation of droplets in the condenser. At 0°, since the gravity direction was identical to the direction of the sample flow, the probability that grown droplets impacted on the condenser wall via sedimentation was negligible. However, with the increment of the tilt angle, the velocity vector of a droplet perpendicular to the channel increased, which lead to the decrement of the counting efficiency.

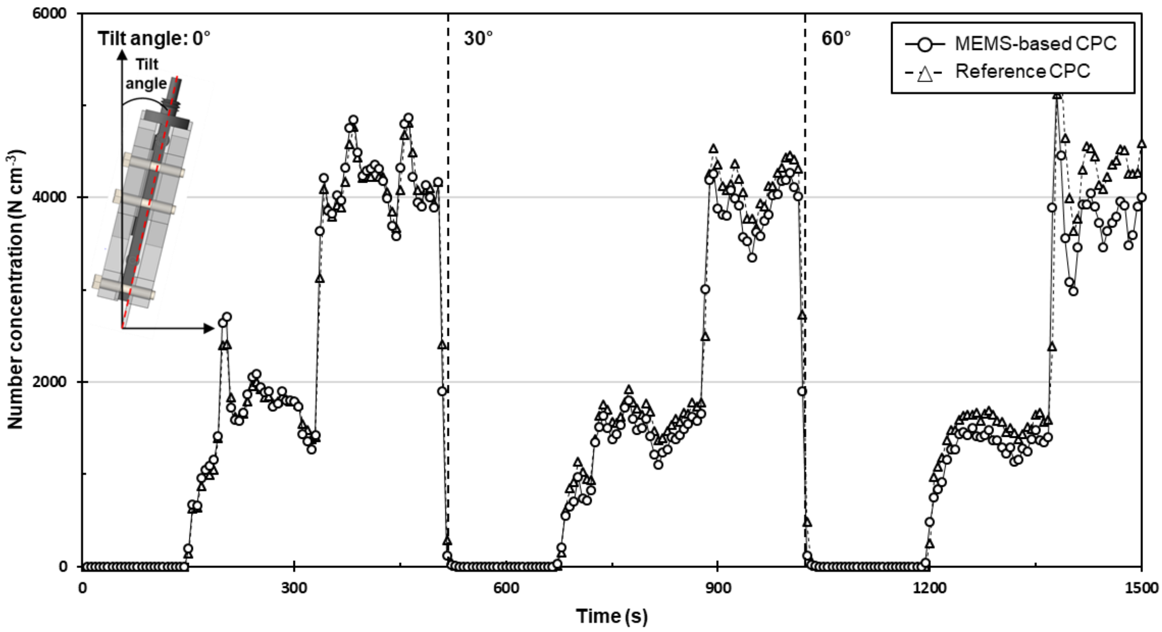

Figure 10: The time series of the number concentrations measured by MEMS-based CPC system when it was tilted.

Question 10

p. 3, l. 19: I did not fully understand what the micropillars in the condenser do. They are needed to prevent droplet formation, which could clog the channel, fine, but how exactly do they do this? Please explain more in detail.

p. 3, l. 22: Why is the pitch between the micropillars in the condenser not provided as number? All other dimensions are provided.

Answer 10

On rough surface, such as the surfaces with the micropillar array, wettability of liquid on the surface is increased. Wenzel proposed an equation for the actual contact angle (θ_e) as a function of static contact angle (θ_c), which can be expressed as

$$\cos\theta_a = r cos\theta_s \ ,$$

where r is the roughness factor, the ratio of the actual area to the projected area of the surface. This equation indicates that the micropillar array decrease the effective contact angle, meaning that it increases the wettability of working fluid and thereby suppresses the droplet formation on the wall.

We revised the content on the micropillar wick in '2 Description of the MEMS-based CPC' of the revised manuscript as followings,

In the condenser, while supersaturated vapor grows UFPs to droplets, some may condense on the wall and clog the channel. Thus, like the saturator, the condenser also had micropillar-type wicks. On the rough surface, the actual contact angle of a working fluid droplet is lower than the contact angle on smooth surface (Chen et al., 2013). Thus, micropillar array increases the wettability of working fluid, suppressing the droplet formation on the wall and draining the condensed working fluid to the reservoir. While the diameter and length were the same, the pitch of the micropillar-type wick (130 μm) was larger than that in the saturator (100 μm).

We added the geometric parameters of the MEMS-based condensation chip of the supplemental material as followings,

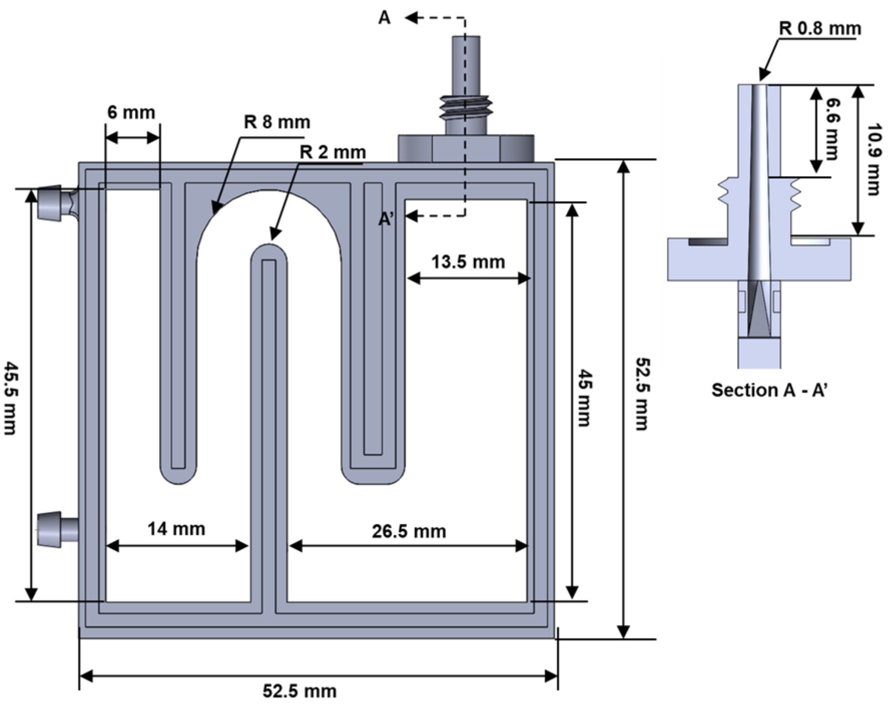

Figure S1: The geometry parameters of the MEMS-based condensation chip.

Question 11

p. 4, l. 20: How long were the sampling lines? Which flow splitter was used? How is the flow geometry there? Did you have the same volume flow to all instruments (probably not, see Fig. 4)? A flow splitter can introduce strong deviations in the particle number concentration for different instruments connected to the flow splitter, in particular when using different volume flows, because the particles are not necessarily distributed homogenously over the sampling line. Please add the information on how the flow split exactly looks like and how you guaranteed that all three instruments good the same particle number concentration. According to Fig 4c I would guess for very small particles this was not the case.

p. 3, l. 29: If the Reynolds number in the system is so low (below 32) don´t you get problems with secondary flows, i.e. convection? Could you check this for instance using the Richardson number? How are the diffusional losses for such a flow? Please provide some numbers.

Answer 11

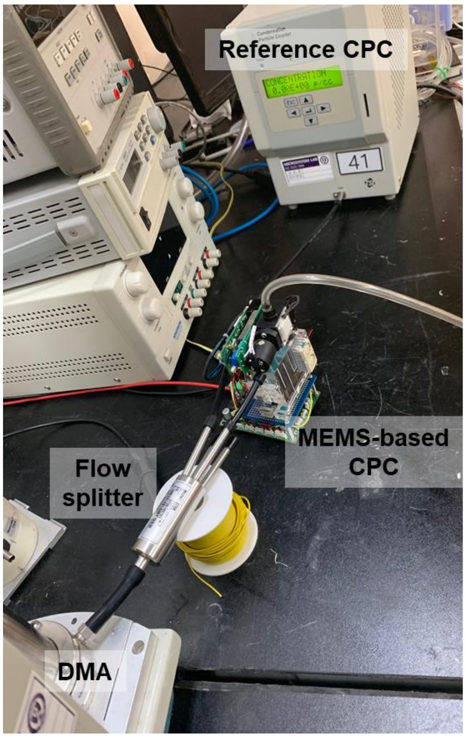

Figure R1: The optical image of the experimental setup for the performance test of our system

Figure R1 shows the optical image of the experimental setup. The CPC and aerosol electrometer were not used simultaneously; while aerosol electrometer was used for characterizing the counting efficiency and detectable concentration range, CPC was used for the performance comparison. Because of the large difference between the flow rates of the reference instrument and our system, the tube length of the reference instrument (either CPC or aerosol electrometer) was about 7 times longer than that of our system to guarantee the same transportation time. The following procedures were carried out to verify that particles with the same concentration were introduced into the two systems. First, to minimize the particle loss induced from the turbulence at the bifurcation, a flow splitter with a very small angle of cleavage (model 3708, TSI Inc., USA) was used. The tubes which leads to the both systems were electrostatic dissipative to minimize the electrostatic particle loss, and their lengths were carefully adjusted to match the transportation times. To verify that the particles which introduced into both systems have the same concentrations, it was confirmed that the counting efficiency was close to 100 % when particles with size of 100 nm were introduced (it was assumed that they were activated and grew into droplets with 100 % efficiency). Then, while reducing the size of the introduced particles to 40 nm by adjusting the voltages of a DMA, it was confirmed whether the counting efficiency remained constant. Through these procedures, it was verified that the concentrations of the particles delivered to the two systems were the same.

Instead of characterizing the loss using an analytical method, the loss of our system was characterized using the counting efficiency, since it is defined as the efficiency of the system at detecting the introduced particles, and thereby describes the overall transportation/activation efficiencies.

We added the procedures and assumptions for characterizing the loss and counting efficiency in 'Experimental setup' part of the revised manuscript as following,

Because of the large difference between the flow rates of the reference instrument and our system, the following procedures were carried out to verify that particles with the same concentration were introduced into the two systems. First, to minimize the particle loss induced from the turbulence at the bifurcation, a flow splitter with a very small angle of cleavage (model 3708, TSI Inc., USA) was used. The tubes which leads to the both systems were electrostatic dissipative to minimize the electrostatic particle loss, and their lengths were carefully adjusted to match the transportation times. To verify that the particles which introduced into both systems have the same concentrations, it was confirmed that the counting efficiency was close to 100 % when particles with size of 100 nm were introduced (it was assumed that they were activated and grew into droplets with 100 % efficiency). Then, while reducing the size of the introduced particles to 40 nm by adjusting the voltages of a DMA, it was confirmed whether the counting efficiency remained constant. Through these procedures, it was verified that the concentrations of the particles delivered to the two systems were the same. The loss of our system was characterized using the counting efficiency, since it is defined as the efficiency of the system at detecting the introduced particles, and thereby describes the overall transportation/activation efficiencies.

We modified the 'Figure 4' in the revised manuscript as followings,

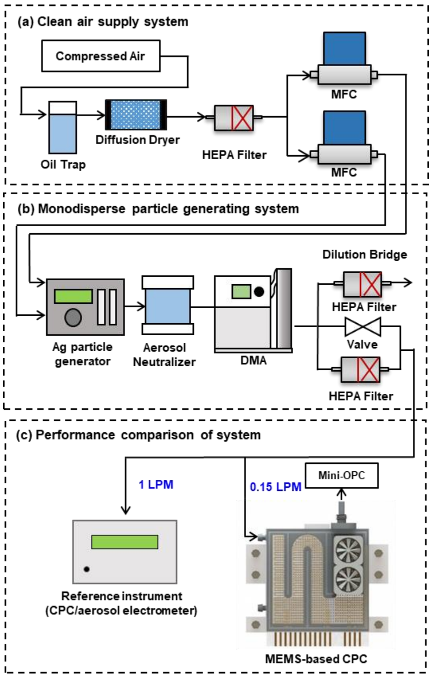

Figure 4: Schematic of the experimental setup for evaluating the performance of the MEMS-based CPC.

Question 12

p. 5, l. 7: The activation efficiency is described as "the condensation chip at growing droplets" which I do not understand or feel to say something wrong. The activation efficiency is the fraction of particles being activated to droplets in the condensation chip.

p. 5, l. 9: Please add "in particular for small particles below ca. 30 nm" after "… on particle size," because the mentioned dependencies are mainly valid for this range.

p. 4, l. 37: The "dry out-region" (maybe better "dry out region"?), how were they identified? The red areas in Fig. 5 "show" them, but I see no difference in the photo inside the read areas and outside.

Answer 12

We modified '2 Description of the MEMS-based CPC' of the revised manuscript as following,

where η_{trans}, η_{act}, and η_{OPC} are the efficiencies of a particle passing through our system, growing droplets at the condensation chip, and the OPC at counting droplets passing through its sensing volume, respectively. Because these three efficiencies are strongly dependent on the particle size, in particular for small particle below ca.30 nm the counting efficiency must be characterized as a function of the particle size.

We modified the mistype in '5.1 Working fluid transmission and evaporation' of the revised manuscript as following,

The dry-out region clearly did not form when the surface temperature was equal to the designed saturator temperature (40 °C) and even reached 70 °C. At 80 °C, the front of the working fluid started to recede, so a dry-out region formed.

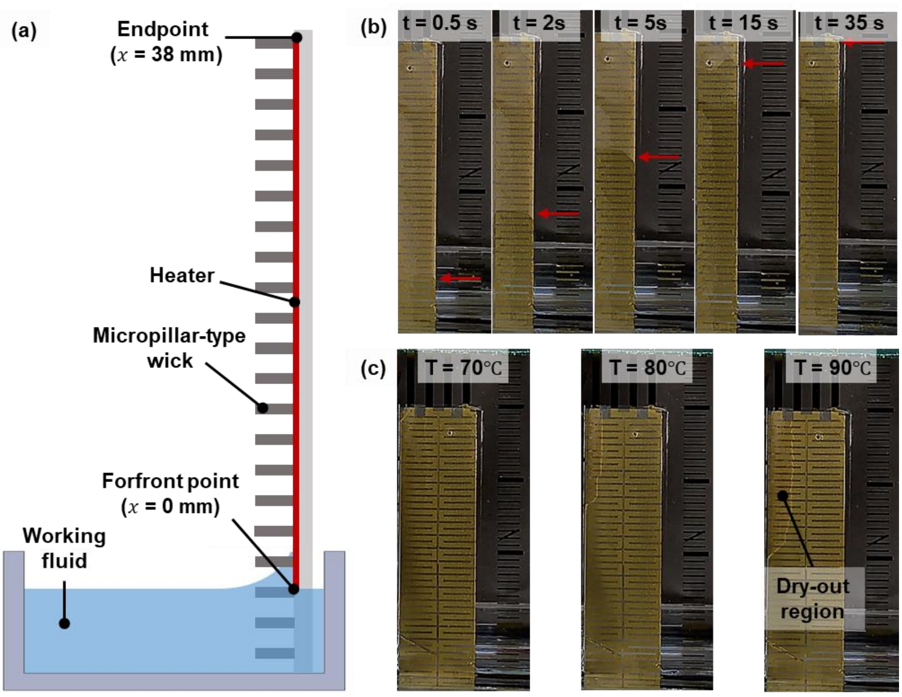

Figure 5: (a) Schematic of the capillary rise experimental setup; (b) selected video frames from the rise of the working fluid using micropillar-type wick; (c) the dry-out region formation as the surface temperature increased.

Question 13

p. 5, l. 16: Fig, 6, how often was the counting efficiency curve measured? The day to day slightly different set-up can influence the curve, hence it should be measured at least three times, ideally on different days. How were the temperature settings and how does the counting efficiency change with different temperature settings?

Answer 13

Although we conducted the counting efficiency experiment five times, the data points shown was a single measurement to make it look clear. We added the additional data points in the previous experiments.

Counting efficiencies for various temperature differences between the saturator and condenser at 20, 25 and 30 °C were characterized. The saturator temperature was not increased above 40 °C, which was the maximum operating temperature of the miniature OPC.

As your advice, we reinforced data in '5.3 Size-dependent particle counting efficiency' of the revised manuscript as following,

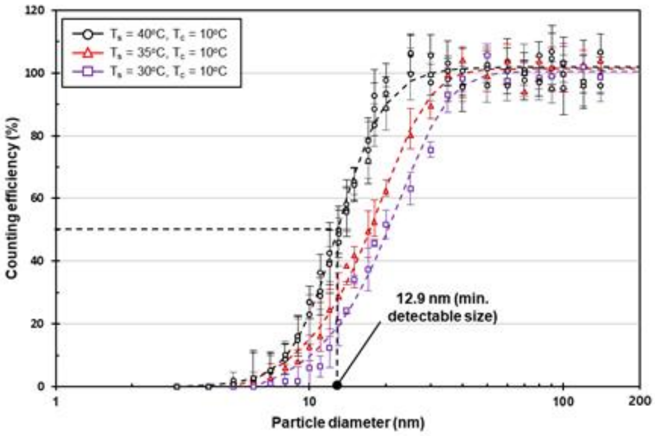

Figure 7: Particle counting efficiency of the MEMS-based CPC as a function of the particle size for various saturator temperature. The particle size at which the particle counting efficiency was fitted to 50% was 12.9 nm (T_S = 40 ° C), 17.3 (T_S = 35 ° C) and 20.4 (T_S = 30 ° C), respectively.

Figure 7 shows the size-dependent counting efficiency of the MEMS-based CPC. The size range of Ag particles was controlled concentration range to 1000-2000 cm^{-3}. The sampling times for each data point were 300 s, and the measurement uncertainty based on the Poisson statistics was 0.02 %. To evaluate the effect of the temperature difference, the counting efficiency was characterized when the condenser temperature (T_c) was 10 ° C and the saturator temperatures (T_s) were 30, 35 and 40 ° C. At 40 ° C (the design value of the saturator temperature), the same experiments were repeated three times to confirm the measurement reliability. When the saturator temperature was 40 ° C, it was found that our system detected 1% of UFPs with the size of 5 nm, and the detection efficiency increased sharply above 9 nm. This was primarily because the activation efficiency (η_{act}) increased when the particle size exceeded the Kelvin diameter. The transport efficiency (η_{trans}) also increased, because the diffusivity of a particle decreases with the increment of the particle size. The counting efficiency data were curve-fitted using

$$\eta_d = \alpha + \frac{(\beta-\alpha)}{1+(d_p/\gamma)^{\delta}} ,$$

where α, β, γ and δ are fitting constants of 101.96, 2.00, 12.99 and 4.70, respectively. The corresponding minimum detectable size is defined as the size at which particles are detected with 50 % efficiency and was found to be 12.9 nm. The detection efficiency was 90 % at 20.1 nm and reached 95% at 22.9 nm. It was close to 100 % and constant in the size range from 25 to 140 nm, indicating that the internal particle loss in this size range was negligible.

Question 14

p. 5, l. 20: "diffusivity of particles is inversely proportional to the size", is this true? How about the slip correction which brings a non-linear term into the particle diffusion problem?

Answer 14

We thank for letting us catch our mistake.

We corrected the wrong sentence '5.3 Size-dependent particle counting efficiency' in the revised manuscript as following,

The transport efficiency (η_{trans}) also increased, because the diffusivity of a particle decreases with the increment of the particle size.

Question 15

Fig. 1: I believe the miniature OPC scheme is incomplete. I cannot imagine that this OPC works in the forward scattering mode without using a beam blocker. Is this really true?

Fig. 5: Please add the unit "s" to the times provided above the photos.

Fig. 6.: Please provide uncertainty bars to the plot.

Answer 15

Figure R2 shows the (a) exploded view, (b) section A-A' and (c) section B-B' of the optical detector. It consists of the sensing chamber and optics (laser, cylindrical lens, elliptic mirror, optical detector, light trap). Introduced droplets are firstly arranged in a row in the acceleration nozzle (i.e., the outlet of the condensation chip) and enter the sensing chamber. The droplets then pass through the place where the condensed thin beam is irradiated. The mirror collects the scattered light from a droplet and redirect it to the optical detector.

When the laser beam passes through the cylindrical lens, the shape of the laser beam is not a point but a very thin surface. In addition, the acceleration nozzle at the chip outlet is only 0.8 mm in diameter and is located about 1.5 mm below the point where the beam passes. Therefore, as shown in Figure R2 (c), on the condition that the coincidence error does not occur (when two particles do not pass through the viewing volume at the same time), almost all the grown micro-droplets are counted in the optical detector.

In the perspectives of the structure and detection principle, the optical detector used in this study is similar to the high-precision OPC rather than dust sensors. Thus our system demonstrated particle counting performance, which was comparable to those of the reference CPC (model 3772, TSI Inc., USA).

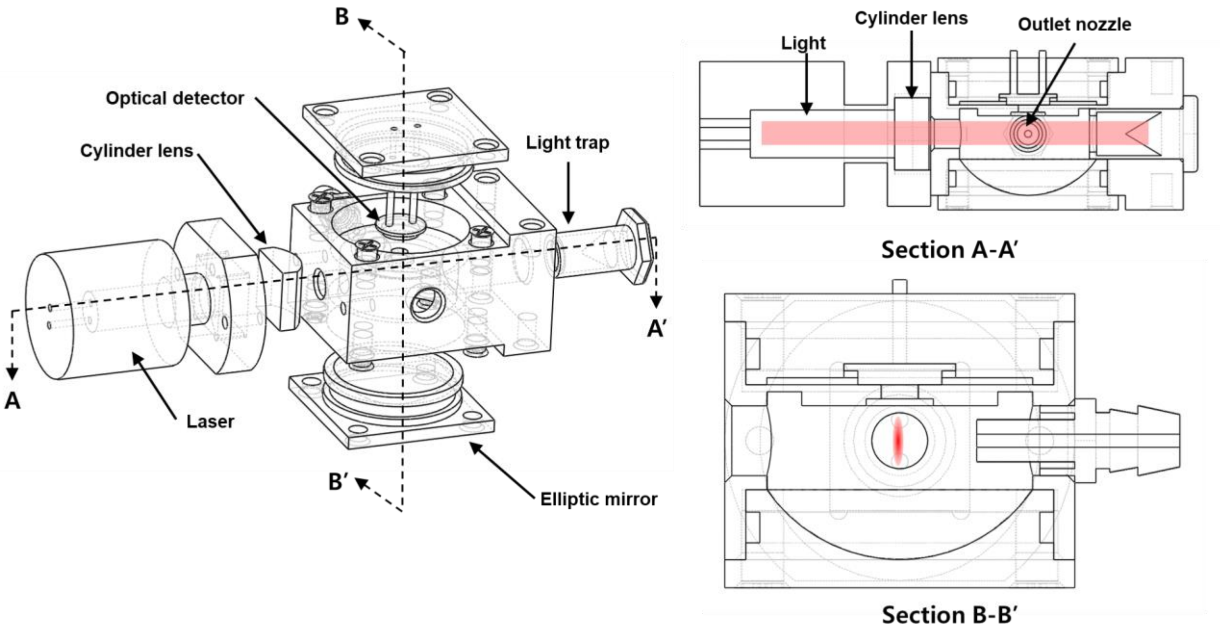

Figure R2: The (a) exploded view, (b) section A-A’ and (c) section B-B’ of the optical detector used in this study

<u>We modified the schematic diagram of the miniature OPC in ‘Figure 1’ of the revised manuscript as following,</u>

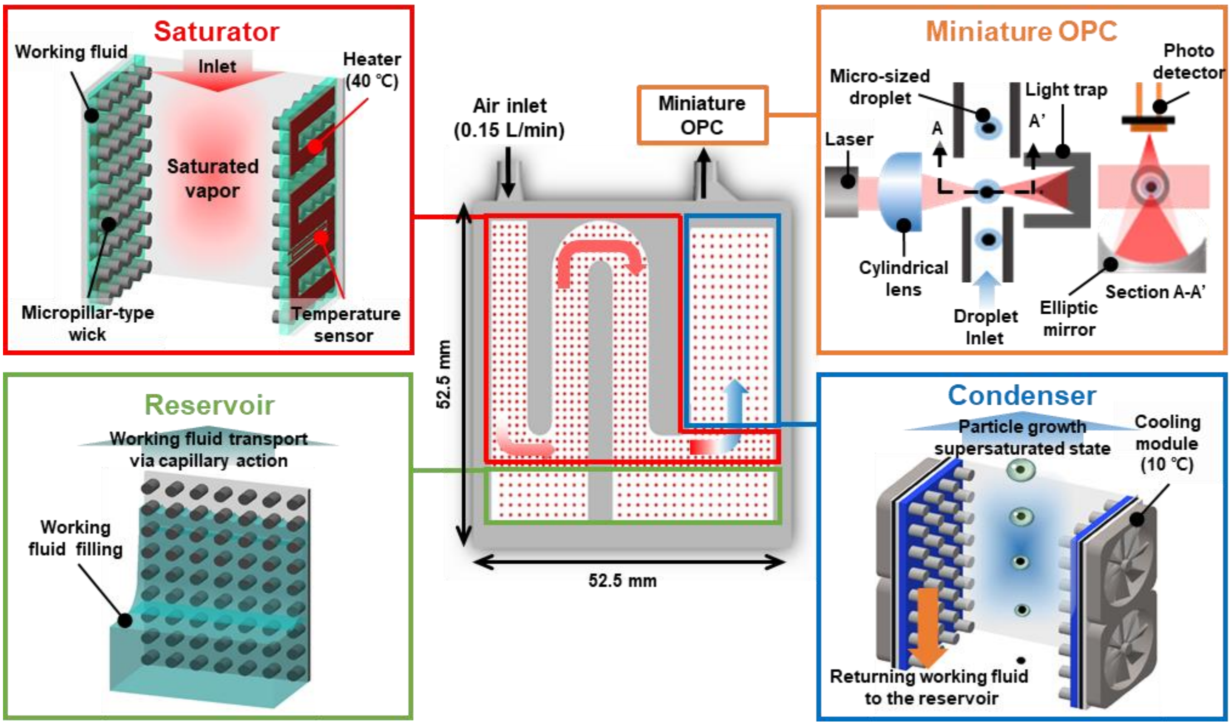

Figure 1: Schematic illustration of the MEMS-based CPC. MEMS-based CPCr parts: the reservoir, saturator, condenser, and miniature OPC. The reservoir supplies the working fluid to the saturator via capillary action by the micropillar-type wick. The saturator heats the working fluid to generate saturated vapor. The saturated air becomes supersaturated when cooled by the condenser. UFPs grow into micro-sized droplets in the condenser and are counted by the miniature OPC.

<u>We modified the video frames in ‘Figure 5’ of the revised manuscript as following,</u>

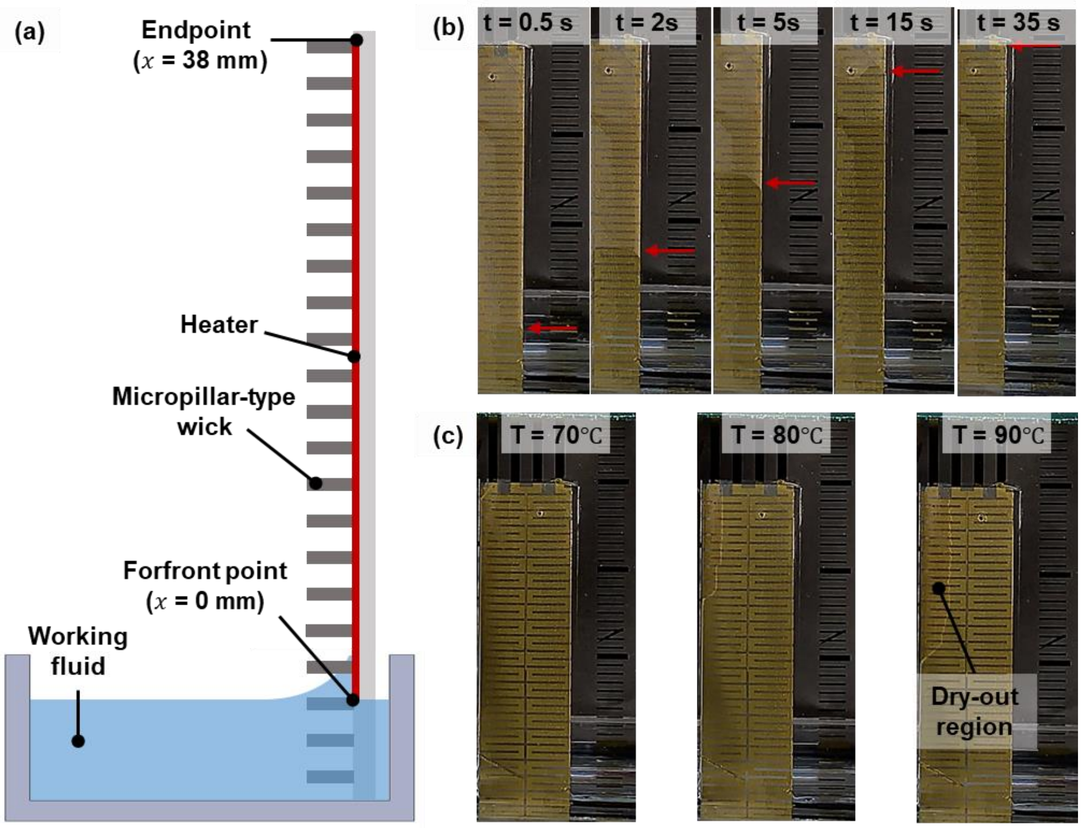

Figure 5: (a) Schematic of the capillary rise experimental setup; (b) selected video frames from the rise of the working fluid using micropillar-type wick; (c) the dry-out region formation as the surface temperature increased.

Please refer to Answer 12. Error bars were added to the size-dependent particle counting efficiency graph.

Question 16

p. 1 1. 12: Please remove “the” before “3D”.

p. 1, l. 17: Please replace “range of” with “range is”, otherwise a verb would be missing.

p. 1, l. 17: The correct CF unit for the particle number concentration should be “1/cm3 ”, without “N”. Please correct in the whole manuscript.

p. 1, l. 22: A space is missing in before the “(“…. This occurs several times in the manuscript, please correct.

p. 2, l. 7: Please remove “for ownership”, this addition is not needed here.

p. 2, l. 18: Please remove the comma after “chip”

p. 2, l. 33: Please insert “to” before “grow”.

p. 3, l. 2: Please exchange “proposed” with a different word, e.g. “new”. The MEMS CPC is existing, you do not “propose” it.

p. 3, l. 7: Please insert “micropillar” before “dimensions”.

p. 3, l. 18: Please insert “to” before “control”.

p. 3, l. 19: “… some” what? “may condense on the wall”, please specify that you mean the working fluid vapor.

p. 4, l. 14: Please exchange “They” with “The particles”.

p. 5, l. 4: Please delete “drawn”

p. 5, l. 38: “the lower concentration limit of the aerosol electrometer was relatively high” Please rephrase, there is no “lower concentration limit” for an electrometer, however, because of the electronic noise you cannot trust the measured concentrations below a few hundred particles per cubic centimeter.

p. 7: The format of some references are different compared to the others, e.g., Hajjam et al, 2010 or Kim et al., 2015. There are more, please check all.

Answer 16 - 1

We modified ‘Abstract’ part of the revised manuscript as following,

A conventional conductive cooling–type CPC is miniaturized through MEMS technology and ~~the~~ 3D printing technique, and the essential elements for growing droplets are integrated on a single glass slide.

Answer 16 – 2

We modified ‘Abstract’ part of the revised manuscript as following,

MEMS-based CPC measured the UFP number concentration with high accuracy (deviation within 4.1 %), and its detectable concentration range is 7.99–7200 cm^{-3}.

Answer 16 – 3

We will revise the relevant part of the manuscript according to the advice of the reviewer.

Answer 16 – 4

We modified in the 1st paragraph of the introduction in the revised manuscript as following,

Monitoring of airborne ultrafine particles (UFPs), which are smaller than 100 nm, is needed in various fields for human health and yield enhancement in industrial fields (Donaldson et al., 1998; Donovan et al., 1985; Hristozov and Malsch, 2009; Li et al., 2016; Liu et al., 2015).

Answer 16 - 5

We modified ‘Abstract’ part of the revised manuscript as following,

However, commercially available CPCs are bulky and expensive; thus, they are impractical for onsite monitoring where the UFP concentration changes continuously. Although portable CPCs (model 3007, TSI Inc., USA) are currently on the market, they are still large in size (292 mm × 140 mm × 140 mm) and expensive (~10,000 USD) ~~for ownership~~.

Answer 16 - 6

We modified in the 3st paragraph of the introduction in the revised manuscript as following,

UFPs are grown to micrometer-sized droplets on the chip~~,~~ and the grown droplets are detected by the miniature OPC.

Answer 16 - 7

We modified in the 3st paragraph of the introduction in the revised manuscript as following,

To generate supersaturated vapor and hence to grow UFPs to micro-sized droplets,

Answer 16 - 8

We will revise the relevant part of the manuscript according to the advice of the reviewer.

We modified 'Description of the MEMS-based CPC' part of the revised manuscript as following,

By using the MEMS technology, our system can generate supersaturated vapor and grow droplets on a chip-scale system for significant decreases in the size, weight, and power consumption.

Answer 16 - 9

We modified 'Description of the MEMS-based CPC' part of the revised manuscript as following,

The dimensions of the micropillar-type wick were experimentally determined to be capable of pumping the working fluid from the reservoir and spreading it over the entire surface of the saturator to ensure that the saturator wall is always in the wetted condition.

Answer 16 – 10

Corresponding parts in the manuscript have been modified with the advice of previous reviewers as follows.

We modified 'Description of the MEMS-based CPC' part of the revised manuscript as following,

Figure 2a shows the customized circuit for our system. The circuit, whose dimension is 90 mm x 65 mm, simultaneously reads the data from the miniature OPC, temperature sensor and flow sensor (model 00H220H024, Nidec Co., JP), and controls the power of the heaters, cooling modules and micro pump (model FS1012-1020-NG, IDT Co., USA) via a pulse-width-modulation (PWM) method. In order for our system to be a stand-alone device, the feedback loops based on the proportional-integral-differential (PID) algorithm is implemented in the micro control unit (MCU) of the circuit, and their gains can be easily controlled using serial communication.

Answer 16 – 11

We modified 'Description of the MEMS-based CPC' part of the revised manuscript as following,

In the condenser, while supersaturated vapor grows UFPs to droplets, working fluid vapor may condense on the wall and clog the channel.

Answer 16 – 12

We modified 'Experimental setup' part of the revised manuscript as following,

The particles were electrically charged by a soft X-ray charger (XRC-05, HCT Co., KR) and then classified to a specific diameter with two types of DMA: (1) nano DMA (model 3085, TSI Co. Ltd., USA) for particles in the size range from 3 to 10 nm, (2) long DMA (model 3081A, TSI Co. Ltd., USA) for particles in the size range from 5 to 140 nm.

Answer 16 – 13

<u>We modified 'Experimental setup' part of the revised manuscript as following,</u>

The counting efficiency (η_d) is defined as the efficiency of the system at detecting the ~~drawn~~ particles and describes the overall CPC performance. It is the product of three efficiencies:

Answer 16 – 14

<u>We modified 'Detectable concentration range' part of the revised manuscript as following,</u>

As shown in Figure 7b, relatively large fluctuations were observed at number concentrations of < 1000 cm^{-3} because of the electronic noise of the aerosol electrometer. However, at the number concentration range of 1000−5000 cm^{-3}, the overall difference between the concentrations of our system and the aerosol electrometer was only 4.1 %, which proves the high accuracy of our system.

Answer 16 – 15

We will revise the relevant part of the manuscript according to the advice of the reviewer.

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