

220 figure 3 and figure 4 respectively. The morphology of ambient particles are not
221 presented because of its scarcity and great variation. Most of the CH₄ and C₂H₄
222 combustion particles are considered as sphere while a small proportion of the
223 combustion particles have a wavy edge. Compared with CH₄ combustion particles,
224 C₂H₄ combustion particles seem to be deviate more from spherical shape. In addition,
225 the agglomerates of C₂H₄ primary particles are more branched and less spherical than
226 the agglomerates of CH₄ primary particles, which means that C₂H₄ particles have
227 larger contact area with the working fluid and thus could be more easily activated in
228 the CPC condenser. The different grayscale and fringe pattern of primary particles
229 from different sources imply that CH₄ and C₂H₄ may generate particles of different
230 chemical compositions.

231 *Experimental counting efficiencies*

232 Fig. 5 shows the experimental results at T_s=38 °C and T_c=29 °C when the
233 IUCPC was challenged with CH₄ and C₂H₄ particles. The counting efficiency
234 increased from 0 to 1 for both CH₄ and C₂H₄ particles as the particle size increased
235 from 13 nm to 19 nm, and C₂H₄ combustion particles exhibited a higher counting
236 efficiency at the same particle size. A plausible reason for the difference of the
237 counting efficiency between CH₄ combustion particles and C₂H₄ combustion particles
238 could be the differences in their hygroscopic properties. The hygroscopic properties of
239 test particles to the working fluid are normally influenced by two factors, namely the
240 composition of test particles and the morphology of the particles. Since particles are

241 not exposed to a high humidity environment before entering the condenser for the
242 sheath flow type CPC, they need to be activated within the period of the time in the
243 condenser, and thus the activation is more easily influenced by the hygroscopic
244 properties of the test particles compared to a full flow type CPC. In general, particles
245 featuring a higher hygroscopic property to the working fluid are more likely to be
246 activated in the condenser.

247 The difference in the composition of the CH₄ combustion particles and C₂H₄
248 combustion particles may influence the wettability of the two types of particles with
249 butanol, and thus influence the counting efficiency curve. C₂H₄ is considered to
250 generate more carbonaceous particles than CH₄ because C₂H₄ could form PAHs
251 (Poly-Aromatic Hydrocarbons) relatively easily, which are the soot precursor (Shukla
252 and Koshi, 2012).

253 The shape of test particles may be another reason contributing to the counting
254 efficiency difference. Compared with ideal spherical particles, non-spherical particles
255 experience higher drag in a differential mobility analyser (DMA), and this will lead to
256 a slight overestimation of their physical size based on their electrical mobility. On the
257 other hand, non-spherical particles may have more contacting line with the working
258 fluid, and thus have higher wettability to the n-butanol. Fig. 6 demonstrates that the
259 experimental counting efficiencies with the temperature difference of 9 °C for both
260 conditions $T_s=31\text{ °C}$, $T_c=22\text{ °C}$ and $T_s=36\text{ °C}$, $T_c=27\text{ °C}$. The former case showed
261 a higher overall counting efficiency, which may be due to the non-linear relationship

262 between the saturation pressure of butanol and the temperature. When the temperature
263 difference reduced from 9 °C to 6 °C, the D_0 (the particle size with which the
264 counting efficiency is just above 0) and D_{50} (cut-off diameter) increased from 12.2
265 nm, 13.6 nm to 14.8 nm, 17.9 nm, respectively. When the temperature difference was
266 further reduced, the D_{50} increased significantly and was much larger than 23 nm.
267 The results were not shown here because such large cut-off sizes are rarely found in
268 the real applications of CPCs.

269 Table 1 listed the cut-off size (D_{50}) and the lowest detectable size (D_0) of the IUCPC
270 operating at several working conditions when it is challenged with different particle
271 sources. When the temperature difference of the saturator and the condenser was set
272 to be 9°C, the cut-off size of IUCPC can be found to be more sensitive to the change
273 of the challenging particles as well as the average temperature of the saturator and
274 condenser than the lowest detectable size. The wettability of the ambient particles to
275 working fluid was thought to rank between that of CH₄ combustion particles and C₂H₄
276 combustion particles.

277 **Conclusion**

278 A sheath type CPC has been constructed to investigate the factors influencing the
279 counting efficiency when challenged with particles from a Bunsen burner and ambient
280 air. The experiments demonstrated that the counting efficiency is closely linked to the
281 combination of the saturator temperature and the condenser temperature, and to the

282 contact angle, which is an indicator of the wettability (or affinity) between particles
283 and the working fluid. When the temperature difference between the saturator and
284 condenser remains constant, the counting efficiency curve of the in-house CPC
285 challenged by ambient particles moved toward left as the average saturator and
286 condenser temperature decreased.

287

288 **ACKNOWLEDGEMENT**

289 This research is supported by National Natural Science Foundation of China
290 (91641119 and 41575119). This project is also supported by special fund of State Key
291 Joint Laboratory of Environment Simulation and Pollution Control (Grant No.
292 17K03ESPCP).

293 **REFERENCES**

- 294 Agarwal, J.K. and Sem, G.J. (1980). Continuous flow, single-particle-counting condensation nucleus
295 counter. *J. Aerosol Sci* 11: 343-357.
- 296 Aitken, J. (1881). Xii.—on dust, fogs, and clouds. *Transactions of the Royal Society of Edinburgh* 30:
297 337-368.
- 298 Amara, A.B., Dauphin, R., Babiker, H., Viollet, Y., Chang, J., Jeuland, N. and Amer, A. (2016).
299 Revisiting diesel fuel formulation from petroleum light and middle refinery streams based on
300 optimized engine behavior. *Fuel* 174: 63-75.
- 301 Biswas, S., Fine, P.M., Geller, M.D., Hering, S.V. and Sioutas, C. (2005). Performance evaluation of a
302 recently developed water-based condensation particle counter. *Aerosol Sci. Technol.* 39:
303 419-427.
- 304 Chen, L., Liang, Z., Zhang, X. and Shuai, S. (2017). Characterizing particulate matter emissions from
305 gdi and pfi vehicles under transient and cold start conditions. *Fuel* 189: 131-140.
- 306 Chen, L., Ma, Y., Guo, Y., Zhang, C., Liang, Z. and Zhang, X. (2016). Quantifying the effects of
307 operational parameters on the counting efficiency of a condensation particle counter using
308 response surface design of experiments (doe). *J. Aerosol Sci.*
- 309 Chen, L., Stone, R. and Richardson, D. (2012a). Effect of the valve timing and the coolant temperature
310 on particulate emissions from a gasoline direct-injection engine fuelled with gasoline and with
311 a gasoline–ethanol blend. *Proc. Inst. Mech. Eng. Part D-J. Automob. Eng.* 226: 1419-1430.

312 Chen, L., Stone, R. and Richardson, D. (2012b). A study of mixture preparation and pm emissions
 313 using a direct injection engine fuelled with stoichiometric gasoline/ethanol blends. *Fuel* 96:
 314 120-130.

315 Chen, L., Zhang, Z., Gong, W. and Liang, Z. (2015). Quantifying the effects of fuel compositions on
 316 gdi-derived particle emissions using the optimal mixture design of experiments. *Fuel* 154:
 317 252-260.

318 Collings, N., Rongchai, K. and Symonds, J.P.R. (2014). A condensation particle counter insensitive to
 319 volatile particles. *J. Aerosol Sci* 73: 27-38.

320 Coulier, P.-J. (1875). Note on a new property of the air. *Journal of Pharmacy and Chemistry* 22:
 321 165-173.

322 Espy, J.P. (1841). *The philosophy of storms*. CC Little and J. Brown.

323 Fletcher, N.H. (1958). Size effect in heterogeneous nucleation. *J. Chem. Phys.* 29: 572.

324 Giakoumis, E.G., Rakopoulos, C.D., Dimaratos, A.M. and Rakopoulos, D.C. (2012). Exhaust
 325 emissions of diesel engines operating under transient conditions with biodiesel fuel blends.
 326 *Prog. Energy Combust. Sci.* 38: 691-715.

327 Giechaskiel, B. and Bergmann, A. (2011). Validation of 14 used, re-calibrated and new tsi 3790
 328 condensation particle counters according to the un-ecé regulation 83. *J. Aerosol Sci* 42:
 329 195-203.

330 Giechaskiel, B., Wang, X., Gilliland, D. and Drossinos, Y. (2011). The effect of particle chemical
 331 composition on the activation probability in n-butanol condensation particle counters. *J.*
 332 *Aerosol Sci* 42: 20-37.

333 Hakala, J., Manninen, H.E., Petäjä, T. and Sipilä, M. (2013). Counting efficiency of a tsi environmental
 334 particle counter monitor model 3783. *Aerosol Sci. Technol.* 47: 482-487.

335 Hering, S.V., Stolzenburg, M.R., Quant, F.R., Oberreit, D.R. and Keady, P.B. (2005). A laminar-flow,
 336 water-based condensation particle counter (wpc). *Aerosol Sci. Technol.* 39: 659-672.

337 Hermann, M. and Wiedensohler, A. (2001). Counting efficiency of condensation particle counters at
 338 low-pressures with illustrative data from the upper troposphere. *J. Aerosol Sci* 32: 975-991.

339 Hoppel, W., Twomey, S. and Wojciechowski, T. (1979). A segmented thermal diffusion chamber for
 340 continuous measurements of cn. *J. Aerosol Sci* 10: 369-373.

341 Iida, K., Stolzenburg, M.R. and McMurry, P.H. (2009). Effect of working fluid on sub-2 nm particle
 342 detection with a laminar flow ultrafine condensation particle counter. *Aerosol Sci. Technol.* 43:
 343 81-96.

344 Iida, K., Stolzenburg, M.R., McMurry, P.H., Smith, J.N., Quant, F.R., Oberreit, D.R., Keady, P.B.,
 345 Eiguren-Fernandez, A., Lewis, G.S., Kreisberg, N.M. and Hering, S.V. (2008). An ultrafine,
 346 water-based condensation particle counter and its evaluation under field conditions. *Aerosol*
 347 *Sci. Technol.* 42: 862-871.

348 Jiang, J., Attoui, M., Heim, M., Brunelli, N.A., McMurry, P.H., Kasper, G., Flagan, R.C., Giapis, K.
 349 and Mouret, G. (2011). Transfer functions and penetrations of five differential mobility
 350 analyzers for sub-2 nm particle classification. *Aerosol Sci. Technol.* 45: 480-492.

351 Kousaka, Y., Niida, T., Okuyama, K. and Tanaka, H. (1982). Development of a mixing type
 352 condensation nucleus counter. *J. Aerosol Sci* 13: 231-240.

353 Kulmala, M., Mordas, G., Petäjä, T., Grönholm, T., Aalto, P.P., Vehkamäki, H., Hienola, A.I., Herrmann,

354 E., Sipilä, M., Riipinen, I., Manninen, H.E., Hämeri, K., Stratmann, F., Bilde, M., Winkler,
355 P.M., Birmili, W. and Wagner, P.E. (2007). The condensation particle counter battery (cpcb): A
356 new tool to investigate the activation properties of nanoparticles. *J. Aerosol Sci* 38: 289-304.

357 Lee, D.-W., Hopke, P.K., Rasmussen, D.H., Wang, H.-C. and Mavliev, R. (2003). Comparison of
358 experimental and theoretical heterogeneous nucleation on ultrafine carbon particles. *J. Phys.*
359 *Chem. B* 107: 13813-13822.

360 Lee, H. and Jeong, Y. (2012). The effect of dynamic operating conditions on nano-particle emissions
361 from a light-duty diesel engine applicable to prime and auxiliary machines on marine vessels.
362 *International Journal of Naval Architecture and Ocean Engineering* 4: 403-411.

363 Lee, S., Kwak, J., Lee, S. and Lee, J. (2015). On-road chasing and laboratory measurements of exhaust
364 particle emissions of diesel vehicles equipped with aftertreatment technologies (dpf, urea-scr).
365 *Int. J. Nav. Archit. Ocean Eng.* 16: 551-559.

366 Liu, W., Kaufman, S.L., Osmondson, B.L., Sem, G.J., Quant, F.R. and Oberreit, D.R. (2006).
367 Water-based condensation particle counters for environmental monitoring of ultrafine particles.
368 *J. Air Waste Manage. Assoc.* 56: 444-455.

369 Mamakos, A., Giechaskiel, B. and Drossinos, Y. (2013a). Experimental and theoretical investigations
370 of the effect of the calibration aerosol material on the counting efficiencies of tsi 3790
371 condensation particle counters. *Aerosol Sci. Technol.* 47: 11-21.

372 Mamakos, A., Martini, G. and Manfredi, U. (2013b). Assessment of the legislated particle number
373 measurement procedure for a euro 5 and a euro 6 compliant diesel passenger cars under
374 regulated and unregulated conditions. *J. Aerosol Sci* 55: 31-47.

375 Mertes, S., Schröder, F. and Wiedensohler, A. (1995). The particle detection efficiency curve of the
376 tsi-3010 cpc as a function of the temperature difference between saturator and condenser.
377 *Aerosol Sci. Technol.* 23: 257-261.

378 Millo, F., Vezza, D.S., Vlachos, T., De Filippo, A., Ciaravino, C., Russo, N. and Fino, D. (2012).
379 Particle number and size emissions from a small displacement automotive diesel engine:
380 Bioderived vs conventional fossil fuels. *Ind. Eng. Chem. Res.* 51: 7565-7572.

381 Otsuki, Y., Takeda, K., Haruta, K. and Mori, N. (2014). A solid particle number measurement system
382 including nanoparticles smaller than 23 nanometers. 1.

383 Petäjä, T., Mordas, G., Manninen, H., Aalto, P.P., Hämeri, K. and Kulmala, M. (2006). Detection
384 efficiency of a water-based tsi condensation particle counter 3785. *Aerosol Sci. Technol.* 40:
385 1090-1097.

386 Rongchai, K. (2014). The high temperature condensation particle counter (ht-cpc)—a new instrument
387 for the measurement of solid particulate matter, Ph. D. thesis, University of Cambridge,
388 Department of Engineering.

389 Sem, G.J. (2002). Design and performance characteristics of three continuous-flow condensation
390 particle counters: A summary. *AtmRe* 62: 267-294.

391 Shukla, B. and Koshi, M. (2012). A novel route for pah growth in haca based mechanisms. *Combust.*
392 *Flame* 159: 3589-3596.

393 Stolzenburg, M.R. and McMurry, P.H. (1991). An ultrafine aerosol condensation nucleus counter.
394 *Aerosol Sci. Technol.* 14: 48-65.

395 Tan, P.-q., Ruan, S.-s., Hu, Z.-y., Lou, D.-m. and Li, H. (2014). Particle number emissions from a

396 light-duty diesel engine with biodiesel fuels under transient-state operating conditions. *ApEn*
397 113: 22-31.
398 Wilson, J., Hyun, J. and Blackshear, E. (1983). The function and response of an improved stratospheric
399 condensation nucleus counter. *J. Geophys. Res.-Oceans* 88: 6781-6785.
400 Winkler, P.M., Vrtala, A. and Wagner, P.E. (2008). Condensation particle counting below 2 nm seed
401 particle diameter and the transition from heterogeneous to homogeneous nucleation. *AtmRe* 90:
402 125-131.
403

ACCEPTED MANUSCRIPT

404

Figure Captions

405 **Fig. 1.** The schematic diagram of the laminar flow cooling type CPC

406 **Fig. 2.** The schematic of the experimental set-up

407 **Fig. 3.** TEM image of CH₄ combustion particles

408 **Fig. 4.** TEM image of C₂H₄ combustion particles

409 **Fig. 5.** Experimental counting efficiencies at $T_s=38$ °C and $T_c=29$ °C. Symbols
410 represent the experimental data of CH₄ particles (circle) and C₂H₄ particles
411 (pentagram).

412 **Fig. 6.** Experimental (symbols) results of the in-house CPC challenged with ambient
413 air particles at different combinations of the saturator temperature and the condenser
414 temperature.

415 **Table. 1.** Cut-off diameter (D_{50}) and the lowest observable particles (D_0) for CH₄ ,
416 C₂H₄ combustion particles and ambient particles when the IUCPC operating at
417 different combinations of saturator and condenser temperatures

418 Table 1

Operating Conditions	Particle source	D ₅₀	D ₀
T _s =38°C T _c =29°C	CH ₄ combustion particles	16.1	13.5
T _s =38°C T _c =29°C	C ₂ H ₄ combustion particles	14.1	12.5
T _s =36°C T _c =27°C	Ambient particles	15.2	12.6
T _s =31°C T _c =22°C	Ambient particles	13.6	12.2
T _s =31°C T _c =25°C	Ambient particles	17.9	14.8

419

ACCEPTED MANUSCRIPT

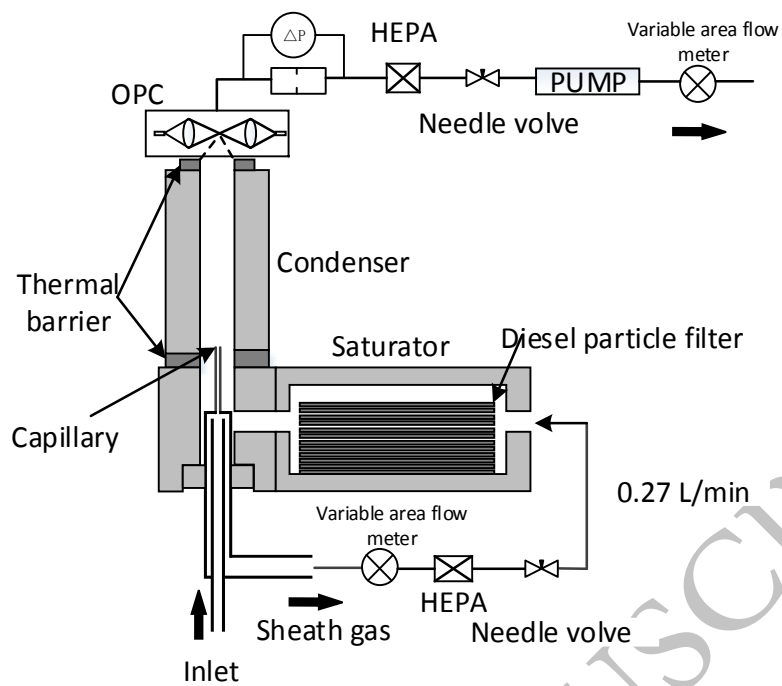


Fig. 1

420

421

422

ACCEPTED MANUSCRIPT

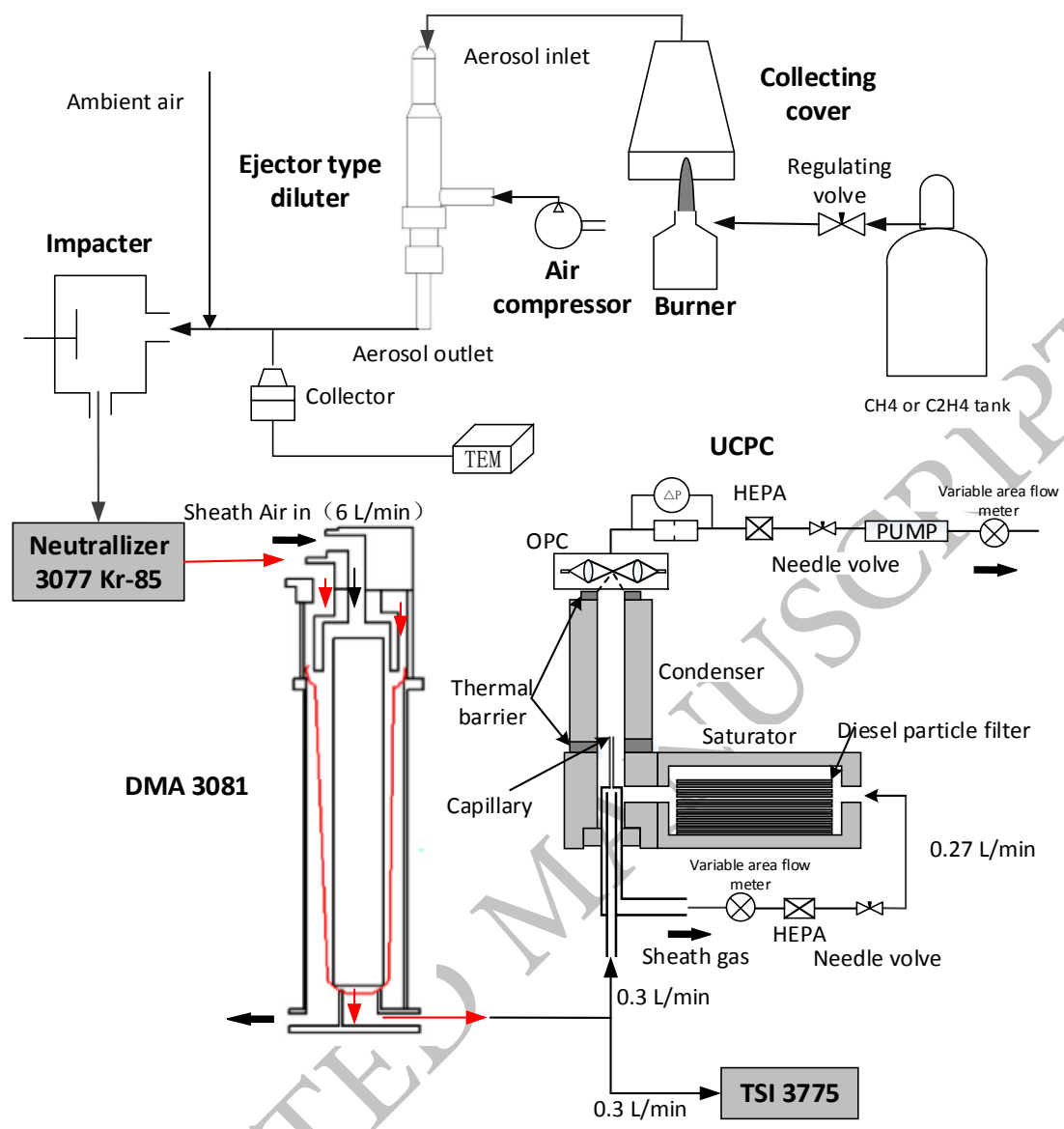
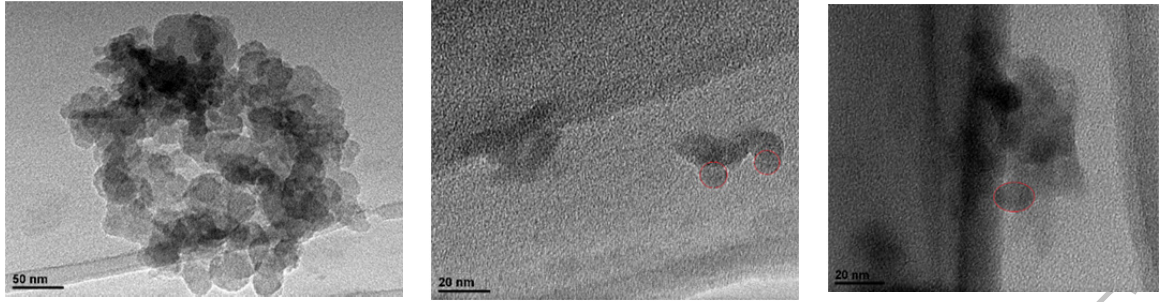


Fig. 2

423

424

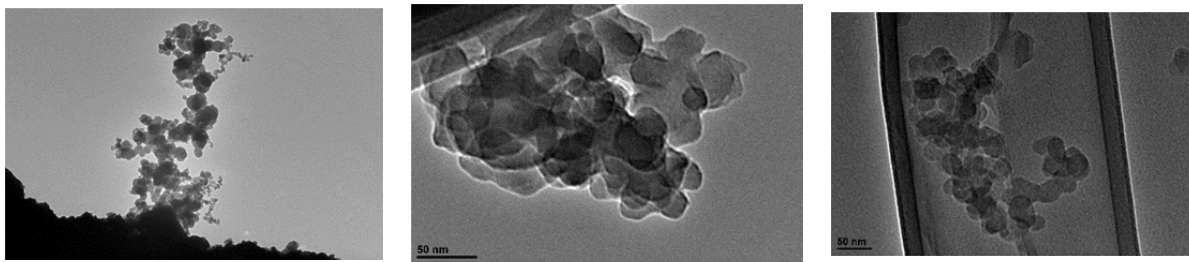
425



426

Fig. 3

ACCEPTED MANUSCRIPT

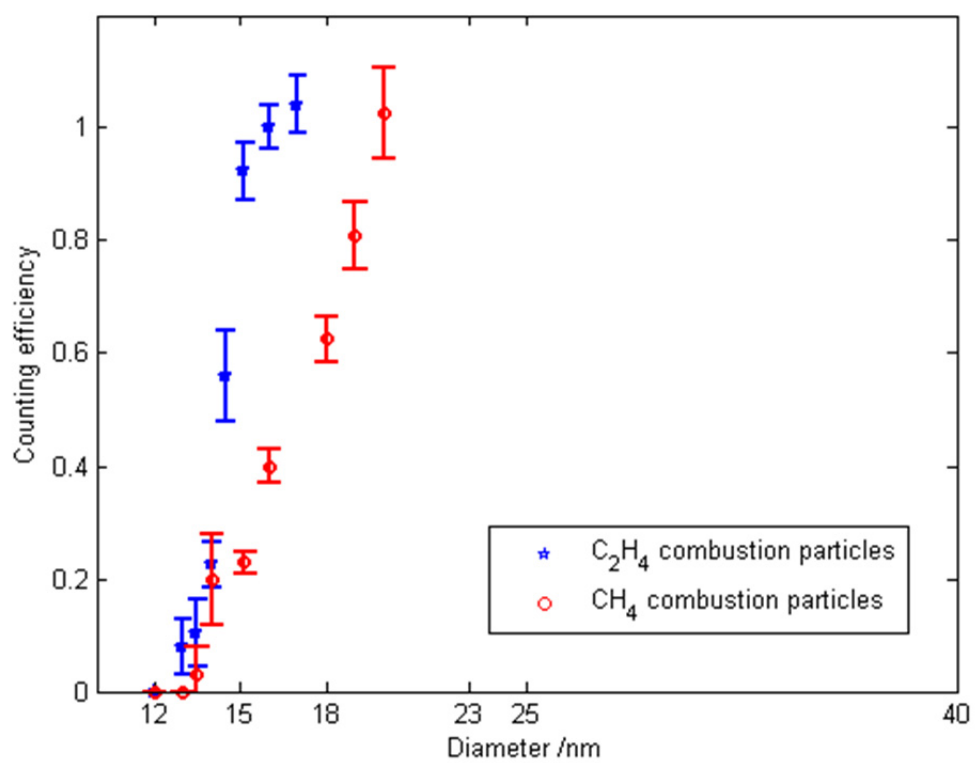


427

Fig. 4

428

ACCEPTED MANUSCRIPT



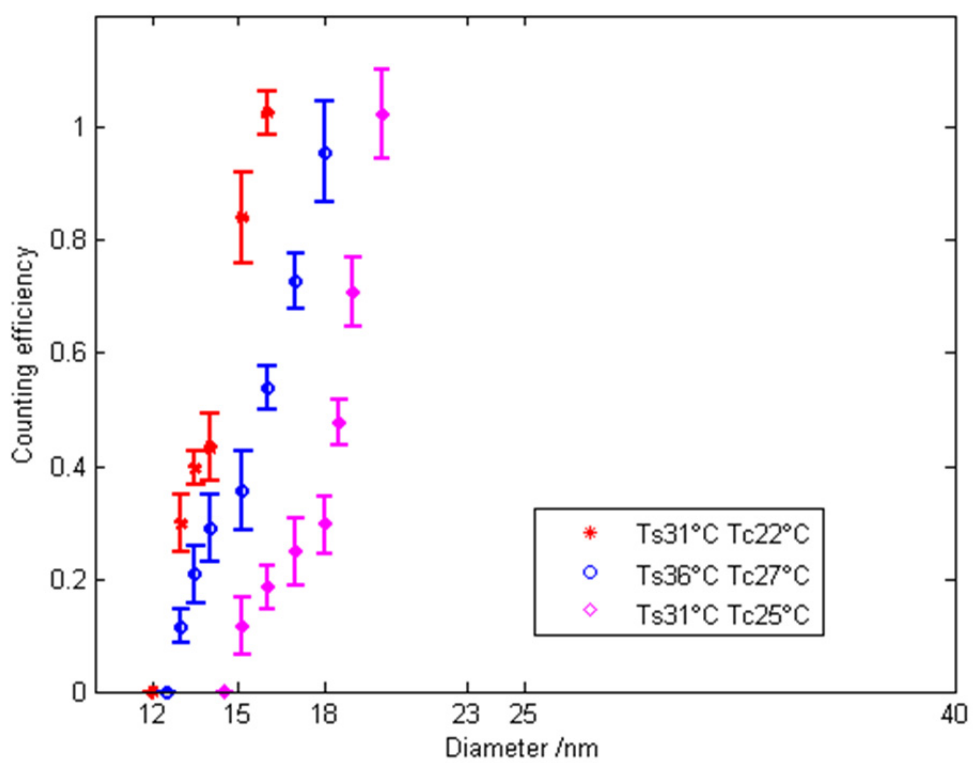
429

430

431

Fig. 5

ACCEPTED MANUSCRIPT



432

433

Fig. 6

ACCEPTED MANUSCRIPT