

459 this study, the flaming and the 'mixed' phases were collectively used under a single umbrella
460 term 'flaming phase', since the flame is still visible during the 'mixed' phase.

461
462 The distribution during the coking phase showed a near unimodal particle size distribution with a
463 GMD of 32.8 ± 5.1 nm and a mode of 31 nm (Fig. 8). The size distribution indicates that particle
464 number concentration gradually increases above 180 nm. Particle number concentration during
465 this phase is comparable to particle number concentrations during the flaming phase. A possible
466 explanation for this is that during the coking phase (at the top of the fuel bed) there will be some
467 coal still igniting and pyrolyzing at the bottom of the fuel bed. As the particles pass through the
468 burning red-hot combustion zone, they are burned resulting in the emission of particles with a
469 lower GMD. The results presented under this phase are similar to the one presented by Li *et al.*
470 (2017), however the study focused on particle size distribution from a coal fired power station. A
471 unimodal distribution was found but the average particle size was above 10 μm .

472
473 In Fig. 9, the coking phase was separated into two stages (i.e. early and late coking). Separation
474 of the combustion phase did not produce different number size distributions, with the distribution
475 similar to the entire phase average as indicated in Fig. 8. This is because the combustion
476 conditions at this stage are homogeneous, with no visible flame nor smoke. Heat produced is in
477 the form of radiant heat, and all volatiles have been driven out at the end of the flaming phase,
478 and only fixed carbon burns with oxygen adsorption and temperature limiting the rate of
479 combustion.

480
481 In Fig. 10, number size distribution and particle number concentration corresponding to the entire
482 burn sequence are presented. The average number size distribution for the whole combustion

483 sequence was found to be near bimodal, with a particle mode of 130 nm. After this mode, above
484 180 nm, particle number concentration was reduced gradually, and the GMD and mode were
485 determined to be 51.6 ± 2.0 nm and 50.6 nm, respectively.

486
487 Other studies also showed PSD from all combustion phases to be bimodal with particle
488 concentrations peaking between 30 and 150 nm (Hedberg *et al.*, 2002; Hays *et al.*, 2002; Zhang
489 *et al.*, 2011; Zhang *et al.*, 2012; Hosseini *et al.*, 2010). Bond *et al.* (2002) observed that when
490 burning coal briquettes particles are emitted in size range between 20 and 100 nm. Earlier lab-
491 based studies have found bimodal PSD from pulverized coal combustion with fine particle mode
492 peaking at around 100 nm (McElroy *et al.*, 1982).

493 Based on the results given in the figures above, where combustion phases were separated, and
494 size distribution have been reported at each stage offers a meaningful insight on this subject (Fig.
495 10). Since the combustion conditions are not constant throughout the whole burn-sequence, it is
496 essential to indicate number size distribution for each identical burn sequence. Looking at Fig. 10
497 only, one concludes that emissions of particles above 80 nm are attributed to the entire burn
498 sequence. While the contribution of larger particles is associated with the ignition and early
499 transitional combustion stage (i.e. ignition to early flaming) phase (Fig. 4-6), smaller particles are
500 associated with both flaming and coking phases (Fig. 7-9). The ignition phase produced particles
501 in the accumulation mode, which was influenced by smouldering conditions and the release of
502 volatile organic compounds under low-temperatures. As the combustion progressed, there was a
503 shift in particle mode from accumulation to Aitken, where particle diameter reduced during the
504 transitional stage. During late flaming to coking, the mode changed to nucleation, where particle

505 diameter further decreased under stable combustion conditions. This observation is similar to the
506 distribution reported in Zhang *et al.* (2012).

507

508 Summary and Conclusion

509 This paper presented results of number size distribution of the submicron particle fraction from
510 the combustion of D-grade coal in a high air ventilated brazier using the top-lit updraft method.

511 Particle size distributions were measured using a TSI NanoScan Scanning Mobility Particle Sizer.

512 The monitor measured PSD throughout the entire burn sequence (from ignition to coking). The

513 PSD curves were separated into three combustion phases: ignition, flaming, and coking. The

514 GMD of particle size distribution was estimated to be 51.6 nm for the averaged burn sequence.

515 Particle number concentrations were high during the flaming and coking phases compared to the

516 ignition phase. The GMD rapidly increased during the ignition phase and gradually decreased

517 during the flaming and the coking phases. Particle size distribution was bimodal across all

518 combustion phases for the D-grade coal used in our experiments but was unimodal during the

519 coking phase. The unimodal distribution suggest a homogenous combustion conditions. At this

520 phase there is constant radiant heat flow and identical particle emissions. During coking all

521 volatiles are driven out with only fix carbon burning under ideal combustion conditions and

522 sufficient air supply. Although the study does not attempt to investigate particle emission

523 composition, we can anticipate the particle emissions during coking phase is attributed to fly ash.

524 Despite limitation on particle size distribution being carried out on small scale coal combustion,

525 we have drawn lessons from other studies which focused on wood burning technologies. Study

526 conducted by several researchers reported a unimodal particle size distribution under ideal wood

527 burning condition (Hedberg *et al.*, 2002; Hays *et al.*, 2002; Zhang *et al.*, 2011; Zhang *et al.*, 2012;

528 Hosseini *et al.*, 2010); while McElroy *et al.* (1982) reported a bimodal PSD. Nevertheless, in all

529 studies a bimodal distribution is attributed to poor combustion conditions and agglomeration
530 process.

531
532 We have demonstrated in this study that improving combustion condition emits particles of
533 smaller diameter. This finding is worrying given the consequences as outlined in epidemiological
534 studies that smaller particles are easily inhaled and have a higher potential for uptake through
535 blood circulation. However, what remains to be investigated is the toxicity of these particles at
536 various combustion phases, to draw conclusions regarding the potential health effects of emitted
537 particles with varied number size distributions.

538
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546
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749 **Table 1.** Stove ventilation rate characterisation.

Brazier ventilation rate	Height (mm)	Dia. (mm)	Grate height (mm)	Area of holes below grate (cm ²)	Area of holes above grate (cm ²)	Total area of holes (cm ²)
High	370	290	185 (50%)	248 (61%)	159 (39%)	407

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751 **Table 2.** Fuel analysis specification.

Parameter (Air Dried Basis)	Standard Method	Slater Mine D-Grade Coal
Moisture content (%)	ISO 5925	1.8
Volatiles (%)	ISO 562	20.3
Ash (%)	ISO 1171	24.2
Fixed carbon (%)	By difference	52.0
Calorific value (MJ kg ⁻¹)	ISO 1928	23.4
Calorific value (Kcal kg ⁻¹)	ISO 1928	5590
Total sulphur (%)	ASTM D4239	0.63
Carbon (%)	ASTM D5373	62.6
Hydrogen (%)	ASTM D5373	2.76
Nitrogen (%)	ASTM D5373	1.0
Oxygen (%)	By difference	5.0
Total silica as SiO ₂ (%)	ASTM D4326	58.6
Aluminium as Al ₂ O ₃ (%)	ASTM D4326	27.6
Total iron as Fe ₂ O ₃ (%)	ASTM D4326	6.63
Titanium as TiO ₂ (%)	ASTM D4326	0.82
Phosphorous as P ₂ O ₅ (%)	ASTM D4326	0.55
Calcium as CaO (%)	ASTM D4326	2.30
Magnesium as MgO (%)	ASTM D4326	0.83
Sodium as Na ₂ O (%)	ASTM D4326	0.42
Potassium as K ₂ O (%)	ASTM D4326	0.79
Sulphur as SO ₃ (%)	ASTM D4326	1.10
Manganese as MnO ₂ (%)	ASTM D4326	0.12

752

753 **Table 3.** Particle number concentration per combustion phase relative to the entire burn sequence.

Phase	(#/cm³) N=3	Duration (minutes)	GMD (nm)	GSD (nm)
Entire Combustion Sequence	1,20E+07 (100%)	180	51.9	2.1
Ignition	2,71E+03 (0.02%)	20	109.8	18.4
Flaming	6,67E+06 (55%)	60	54.9	5.9
Coking	5,34E+06 (44%)	100	34.3	5,1

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755 **Table 4.** Contribution of combustion phase on PSD for the three modes (Nucleation, Aitken and
756 Accumulation).

Combustion phase	Nuclei (Dp<50 nm) (%)	Aitken (Dp 50- 100 nm) (%)	Accumulation (Dp>100 nm) (%)
Ignition	15,3	45,5	39,2
Flaming	46,8	30,8	22,4
Coking	83,9	13,4	2,6

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Figure Captions

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- Fig. 1.** Laboratory designed high ventilation stove.
- Fig. 2.** Schematic diagram of the SeTAR Centre testing rig.
- Fig. 3.** Particle size distributions corresponding to the background concentrations.
- Fig. 4.** Particle size distribution corresponding to the ignition phase.
- Fig. 5.** Particle size distribution during early and late ignition.
- Fig. 6.** Particle size distribution corresponding to the flaming phase.
- Fig. 7.** Particle size distribution during early and late flaming.
- Fig. 8.** Particle size distribution corresponding to the coking phase.
- Fig. 9.** Particle size distribution during early and late coking.
- Fig. 10.** Particle size distribution corresponding to the entire burn cycle.

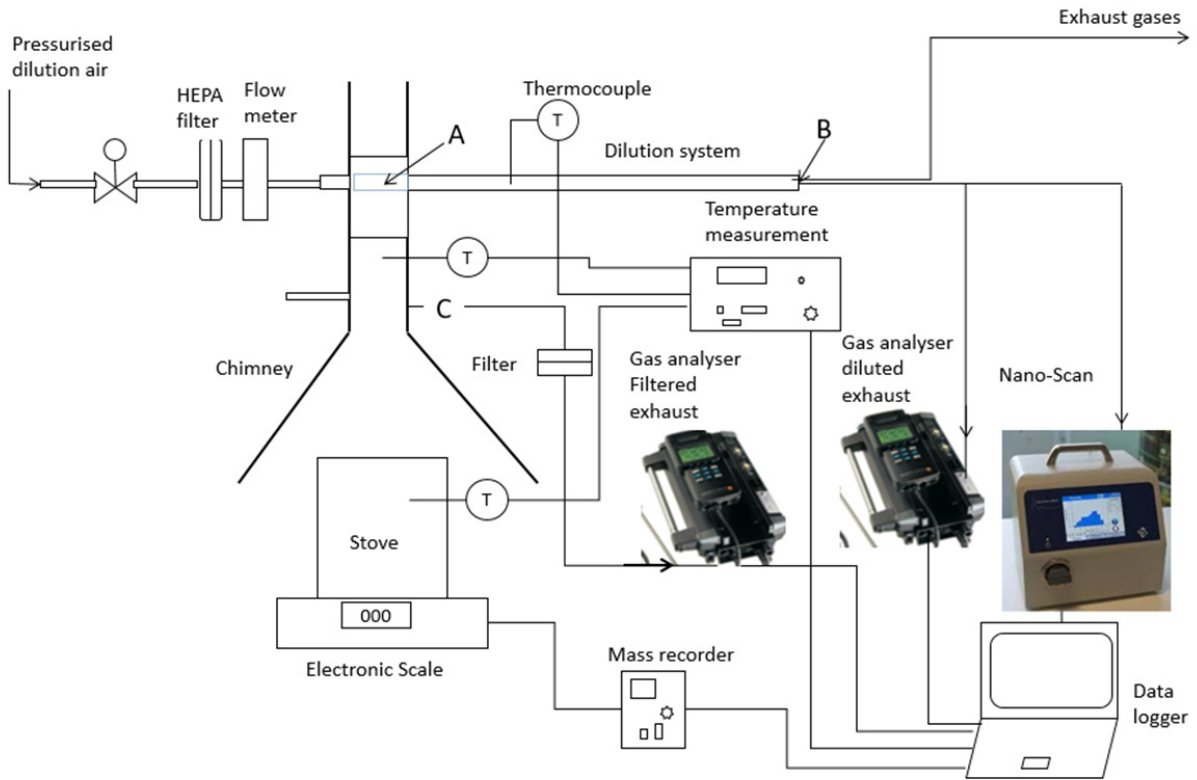
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Fig. 1.

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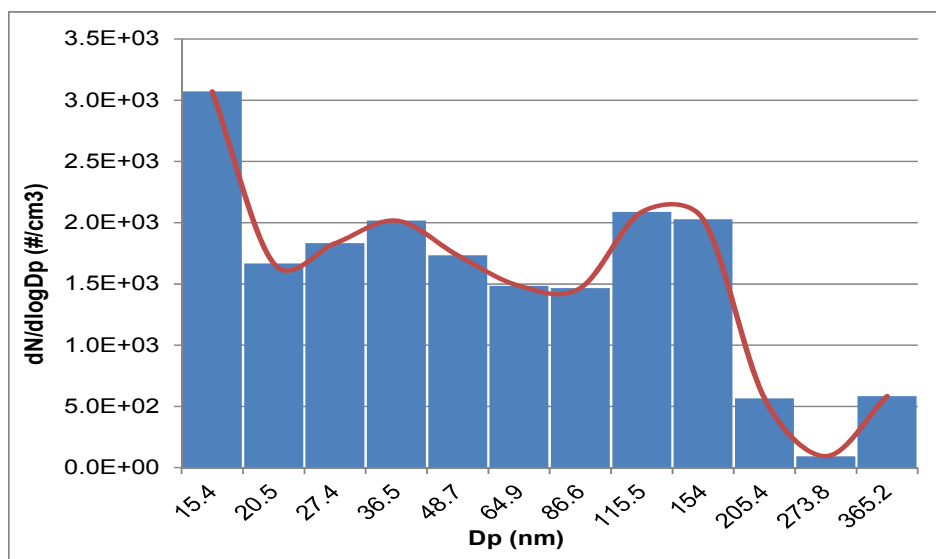
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Fig. 2.

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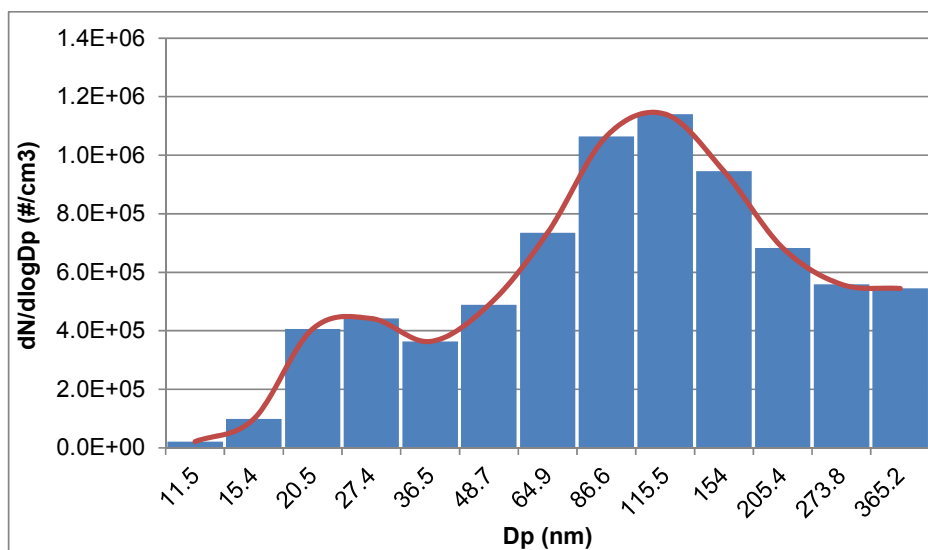
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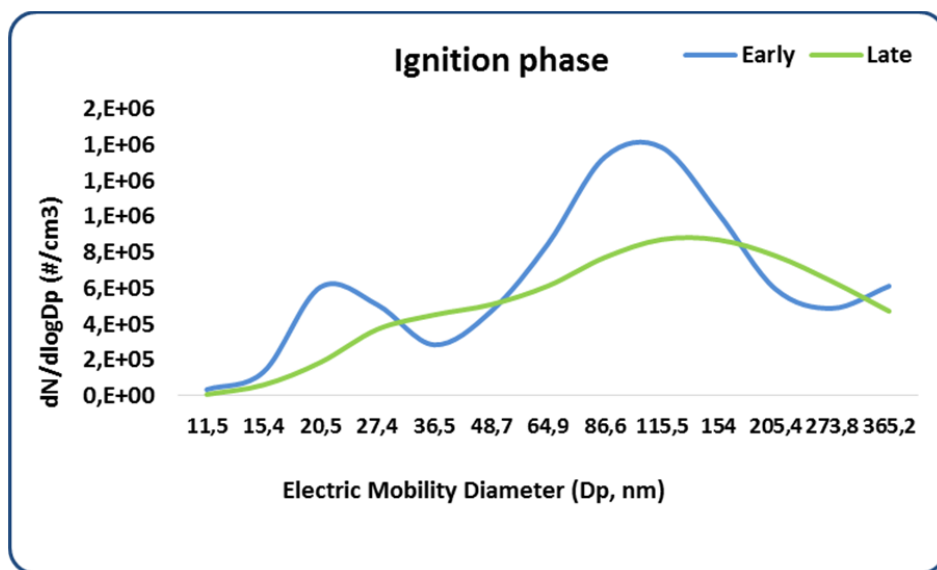
Fig. 3.



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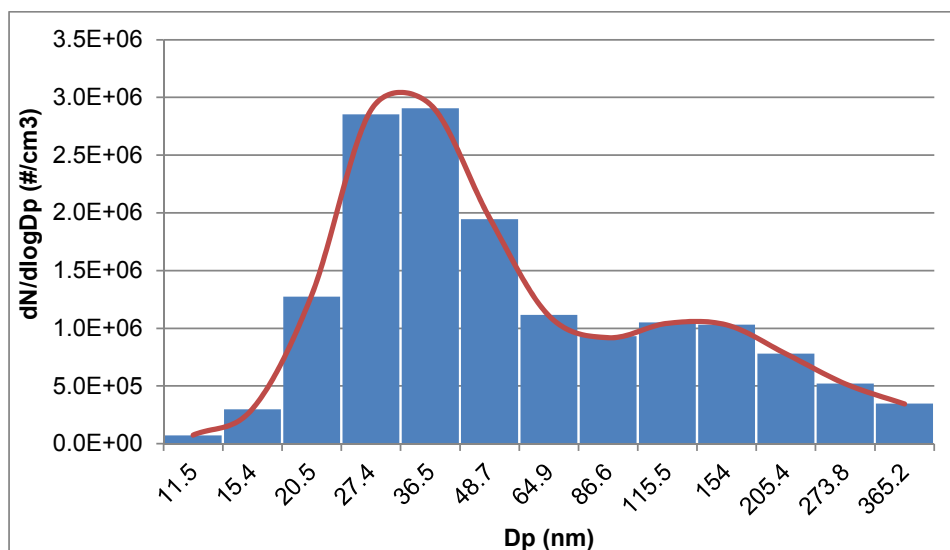
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Fig. 4.



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Fig. 5.



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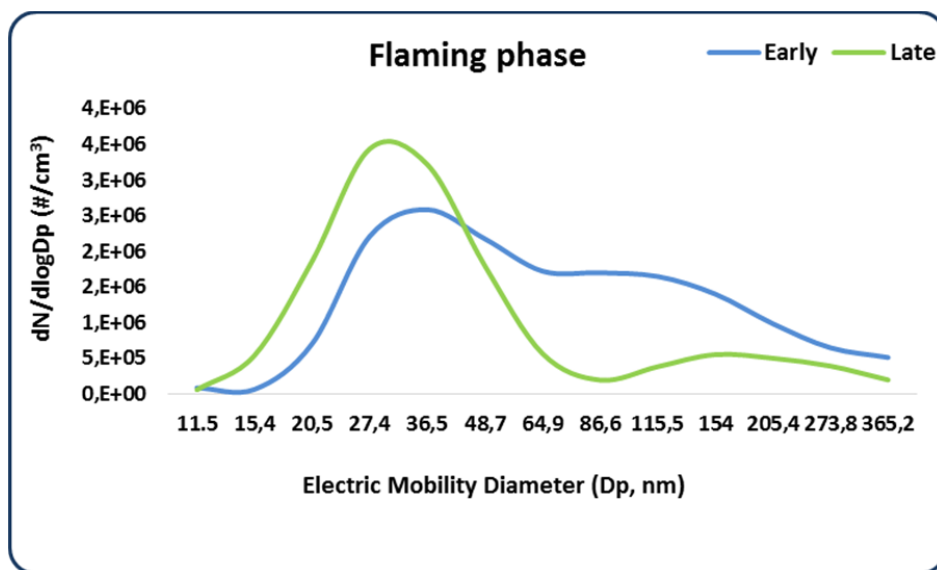
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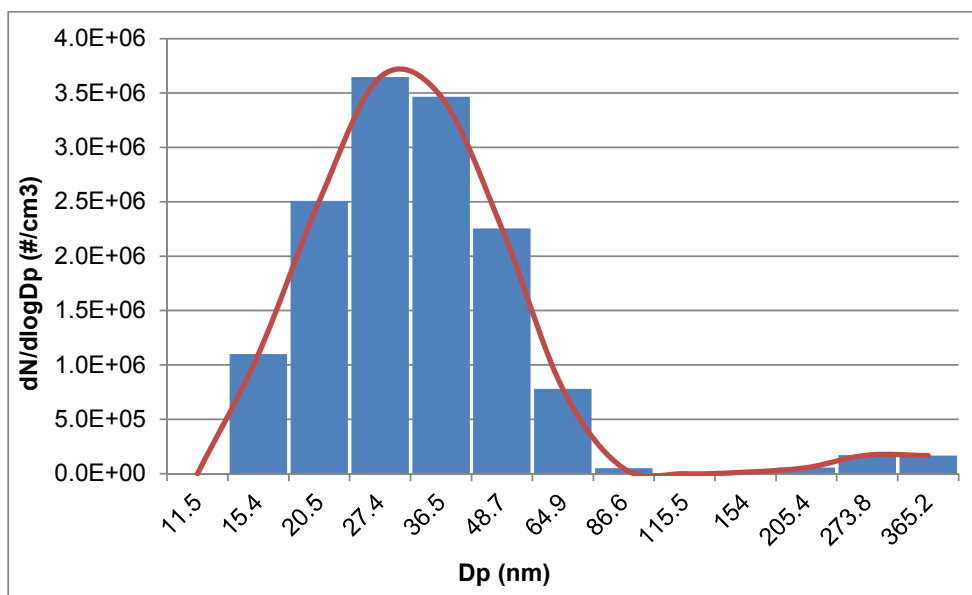
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Fig. 6.



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Fig. 7.



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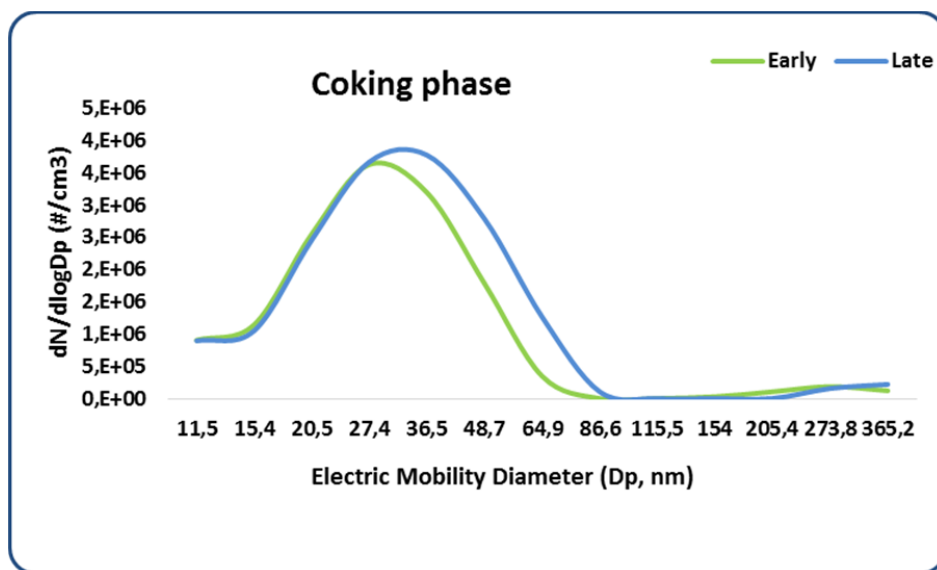
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Fig. 8.



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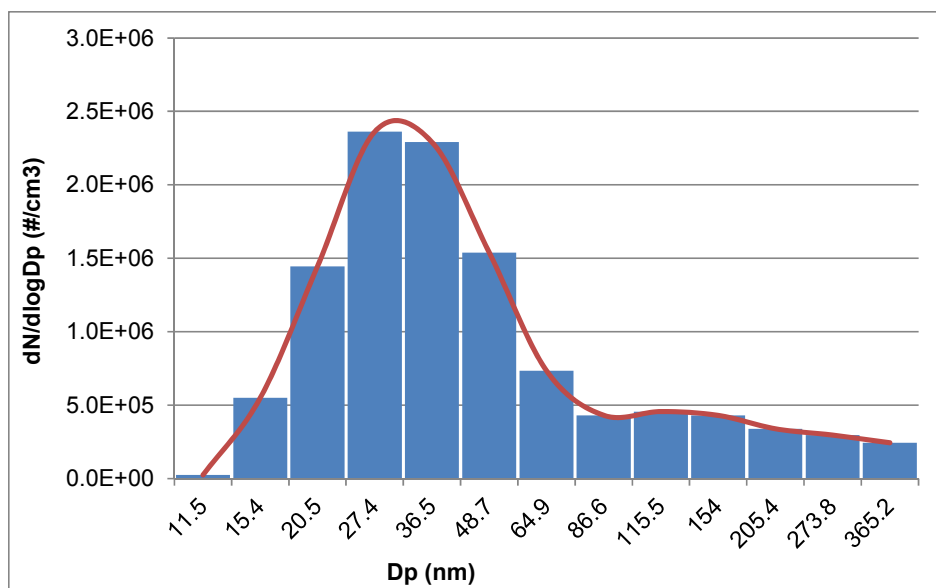
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Fig.9.



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Fig. 10.