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Quantitative investigation on diesel exhaust: influence of dilution, residence time and hygrometry on soot particles

Quantitative investigation on diesel exhaust : influence of dilution, residence time and hygrometry on soot particles

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In most of the experimental studies, engine exhaust particle measurements are performed on diluted aerosols. Applying a high dilution ratio (>10:1) we avoid condensation and the analysis devices are preserved from dirtying. In addition, we know that for particle concentration levels lower than 10^4 or 10^5 part./cm^3, the effect of soot aggregates coalescence is not very significant during the measurement time. Nevertheless some studies, like toxicological experiments, require dilution ratios lower than 10:1. The aim of this experimental work is to quantify the influence of dilution ratio, residence time and hygrometry on the characteristics of soot particles accumulation mode. These measurements have been performed with a SMPS particle size analyzer on the exhaust of a recent 2L displacement diesel common rail engine. These experiments have been carried out as part of the TOPAASE* research program.

Toxicological exposition system and particle measurement results

The toxicological experimental setup has been presented by Dr. J.P. MORIN at the 4th ETH conference [1,2,3,4,5,6,7]. This original system has six rotary exposition chambers disposed in their incubator. Five of these chambers are used to quantify the dose effect with the application of different dilution ratios. The last one is used as a reference and the exposition atmosphere is free of particles and gaseous pollutants. These chambers are designed to receive several culture vials containing the biological samples. The process has four different functions: sampling of exhaust gases, preparation of the dilution gaseous mixture, primary dilution operation and exposition with the realization of the five secondary dilution ratios.
Particle measurements have been performed by SMPS in different parts of the circuit. We notice a significative shift (of 40 nm) in the particle size between the inlet and the outlet of the chambers. In addition, the particle size is also dependent on the dilution ratio. These results suggests that a coalescence phenomenon is probable, but we can not be sure about hygrometry having a significative role on aggregate growth. In fact, the hygrometry level undergoes to important changes during the dilution process, and the level of humidity in the exposition chambers is close to dew point conditions.
Three hypothesis can be listed and should be checked to explain soot aggregates growth.

- The effect of hygrometry on the measurement device (SMPS)
- The effect of hygrometry on soot aggregate growth rate
- The effect of coalescence ascribable to concentration of particles and residence time

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* Toxicologie, Pollution Atmosphérique, Aérothermochimie, Santé, Environnement (*TOxicology, Atmospheric Pollution, Aerothermochemistry, Health, Environement)*

Experimental setup

On the 7th slide we give a detailed representation of the experimental setup. The process has four different functions :

- The production of a regular flow of dry, clean and particle free air
- A system of humidifier which is able to give different levels of hygrometry
- The engine exhaust pipe with its sampling probe system
- The dilution tunnel fitted out with a SMPS measurement outlet, a volumetric flow meter and an hygrometry meter for the total humidity calculation.

Effects of hygrometry on SMPS

To check this hypothesis, a monodispersed polystyrene aerosol (PSL) has been generated and diluted with clean air to obtain five different hygrometry levels (9, 25, 39, 50.5, 60.5 RH%). The results (slides 8 and 9) show that the modal size of PSL is absolutely constant and that the distributions are superposed. This means that the SMPS analyzer is not sensitive to hygrometry under the dew point.

Effects of hygrometry on aggregates

Four physical parameters have been selected to check the second hypothesis: the geometrical mean diameter of distributions, the particle concentration, the aerodynamic soot volume fraction and the standard geometric deviation of the lognormal fitted distributions. These parameters have been measured versus the hygrometry level (RH%) for five exhaust gases concentrations. The results, given on the slides 10 to 12, show that there is no significative effect of the hygrometry level on soot aggregates. These results are in agreement with the works that Weingartner, Baltensperger and Burtscher published in 95 [8]. Indeed, they have shown that for combustion particles there is no significative growth effect except for high relative humidity condition, this means close to the dew point.

Effects of exhaust gases concentration

An important increase of the particle size versus the dilution ratio has been measured (slide 13). In fact, when the fraction of exhaust gases is about 50% the particles are more than 30% bigger than when it is only about 10%. Consequently, the product between the concentration and the dilution ratio decreases. However, a question remains. Is the soot aggregate size increase only ascribable to coalescence phenomenon? On the slide n°14 we can see an excellent proportionality of the aerodynamic volume fraction with the exhaust gases percentage. Even if we know that the compacity of aggregates can be dependent on the their mean size, it seems that there is no significative mass losses and, in other words, that most of particle disappearing can be explained by coalescence.

A confirmation of the soot aggregate growth phenomenon is given on the slide n°15 by an ELPI measurement.

Effects of residence time

It remained to quantify the effect of residence time on soot particles, to close this investigation. To do this, SMPS measurements were performed using different tubing lengths. Indeed, as higher is the concentration, higher is the coalescence, we have observed that higher is the residence time, higher are the particle sizes (slide 16). In addition we have noticed a significative decrease of the aerodynamic fraction with the tubing length increases (slide 17). This seems to indicate that we have particle losses by deposition.

Conclusion

To summarize, this study we can say that there is no effect of the sample humidity level on SMPS, that hygrometry is not responsible for soot aggregates growth but that coalescence is the only phenomenon able to explain the growth of the particles.

1 **: Le PRIEUR E., MORIN J.P., BION A., GOURIOU F., DIONNET F.** .« Toxicological Impact of Diesel Fuel supplementation with rapseed Methylester (RME) on the lung toxic potential of Diesel exhausts » *SAE Technical Paper 2000-01-2060, Paris **2000***.

2 **: Le PRIEUR E., MORIN J.P., VAZ E., BION A., DIONNET F.** « Toxicity of diesel engine exhaust: induction of pro-inflammatory response and apoptosis in an in vitro model of lung slices in bi-phasic organotypic culture » *SAE Technical Paper 2000-01-1928, Paris **2000***.

3 **: MORIN JP, Le PRIEUR E, DIONNET F, ROBIN L.** « The influence of a particle trap on the in vitro lung toxicity response to continuous exposure to diesel exhaust emission » *SAE Technical Paper 1999-01-2710, Vancouver **1999.***

4 **: MORIN JP, FOUQUET F, MONTEIL C, Le PRIEUR E, VAZ E, DIONNET F.** « Development of a new in vitro system for continuous in vitro exposure of lung tissue to complex atmospheres : Application to diesel exhaust toxicology ». *Cell Biol. Toxicology, vol. 15, pp. 143-152, **1999.***

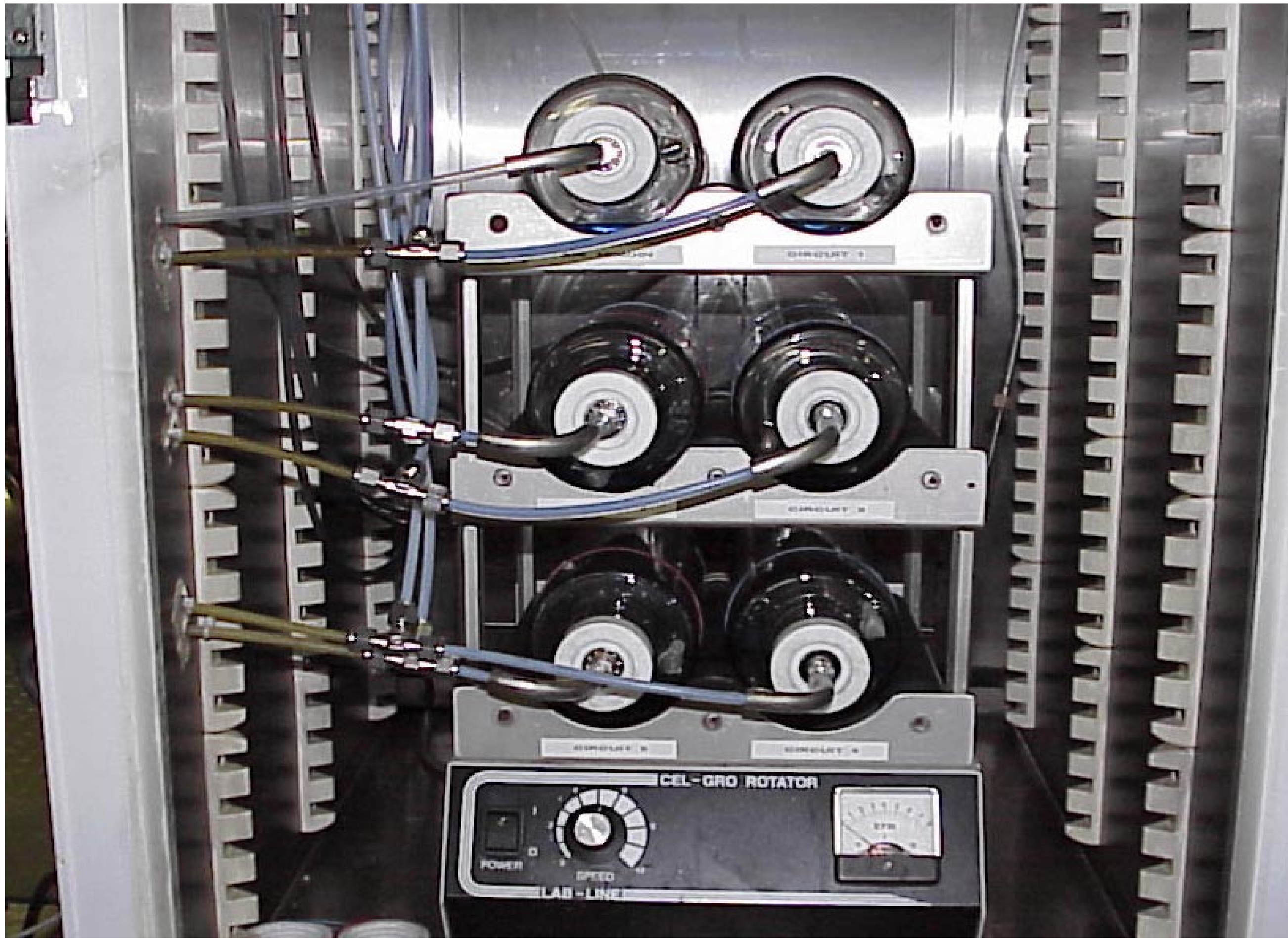
5 **: FOUQUET F., MONTEIL C., Le PRIEUR E., DIONNET F., MORIN J.P.** « In vitro modelization of inhalation toxicology : effects of diesel exhausts on organotypic culture of peripheral lung tissue ». *Toxicology letters , vol. 95-S1 Int. Congress of Toxicology, Paris **1998.***

6 **: MORIN JP., DIONNET F.** « Impact des polluants atmosphériques liés aux transports sur la santé : une nouvelle méthode d'étude toxicologique in vitro ». *Colloque PRIMEQUAL-PREDIT Pollution de l'air à l'échelle urbaine et locale, Paris 10 et 11 déc. **1997.***

7 **: Le PRIEUR E., MONTEIL C., FOUQUET F., DIONNET F., MORIN J.P.** « impact des échappements diesel sur le tissu pulmonaire : développement d'une nouvelle méthode d'étude toxicologique in vitro » *Société Française de Toxicologie, PARIS **1997.***

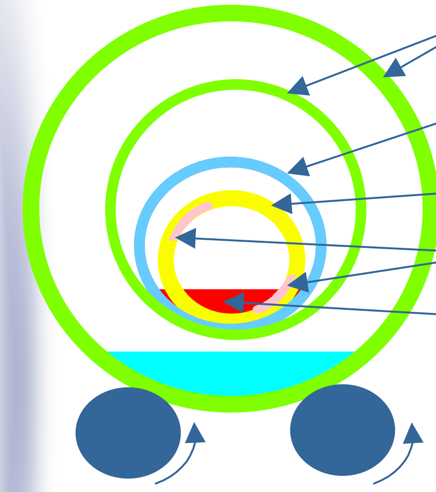
8 **: WEINGARTNER E., BALTENSPERGER U. and BURTSCHER H.** « Growth and Structural Change of Combustion Aerosols at High Relative Humidity » *ENVIRON. SCI. TECHNOL, vol. 29, n°12, pp. 2982-2986, **1995.***

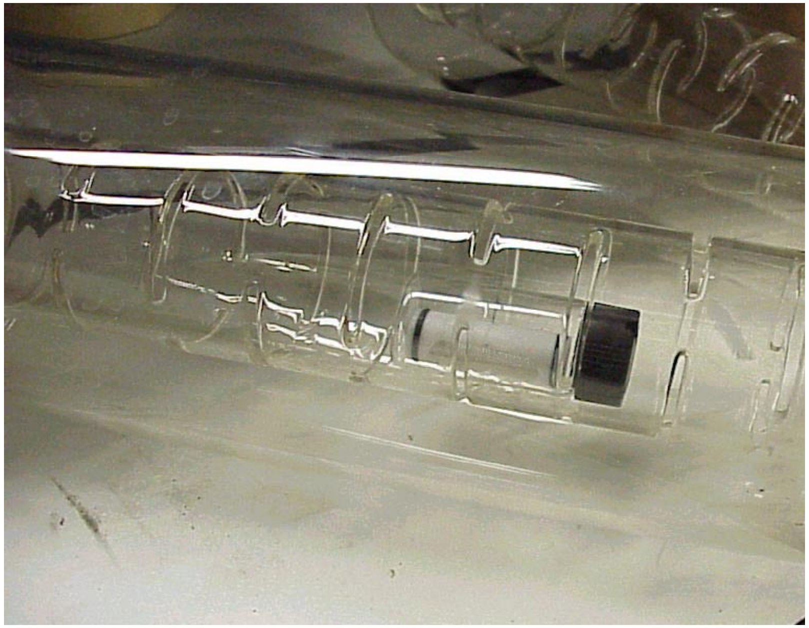
experimental setup for toxicological studies



exposition chamber design

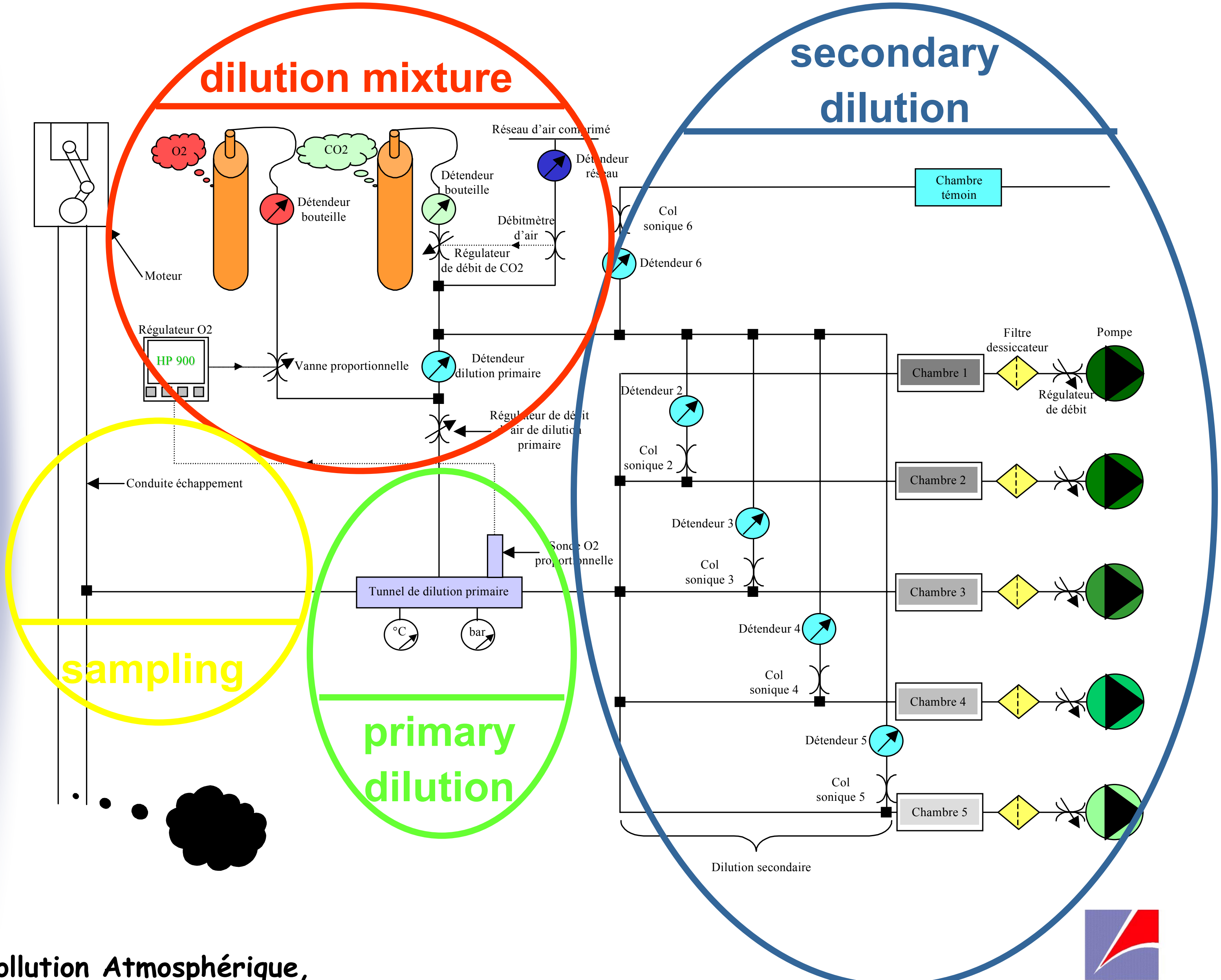
Original Design for Dynamic Organ Culture





Continuous exposure to a constant flow of controlled simple or complex atmospheres (gaseous and/or particulate matter) or solubilized compounds

principle of operation



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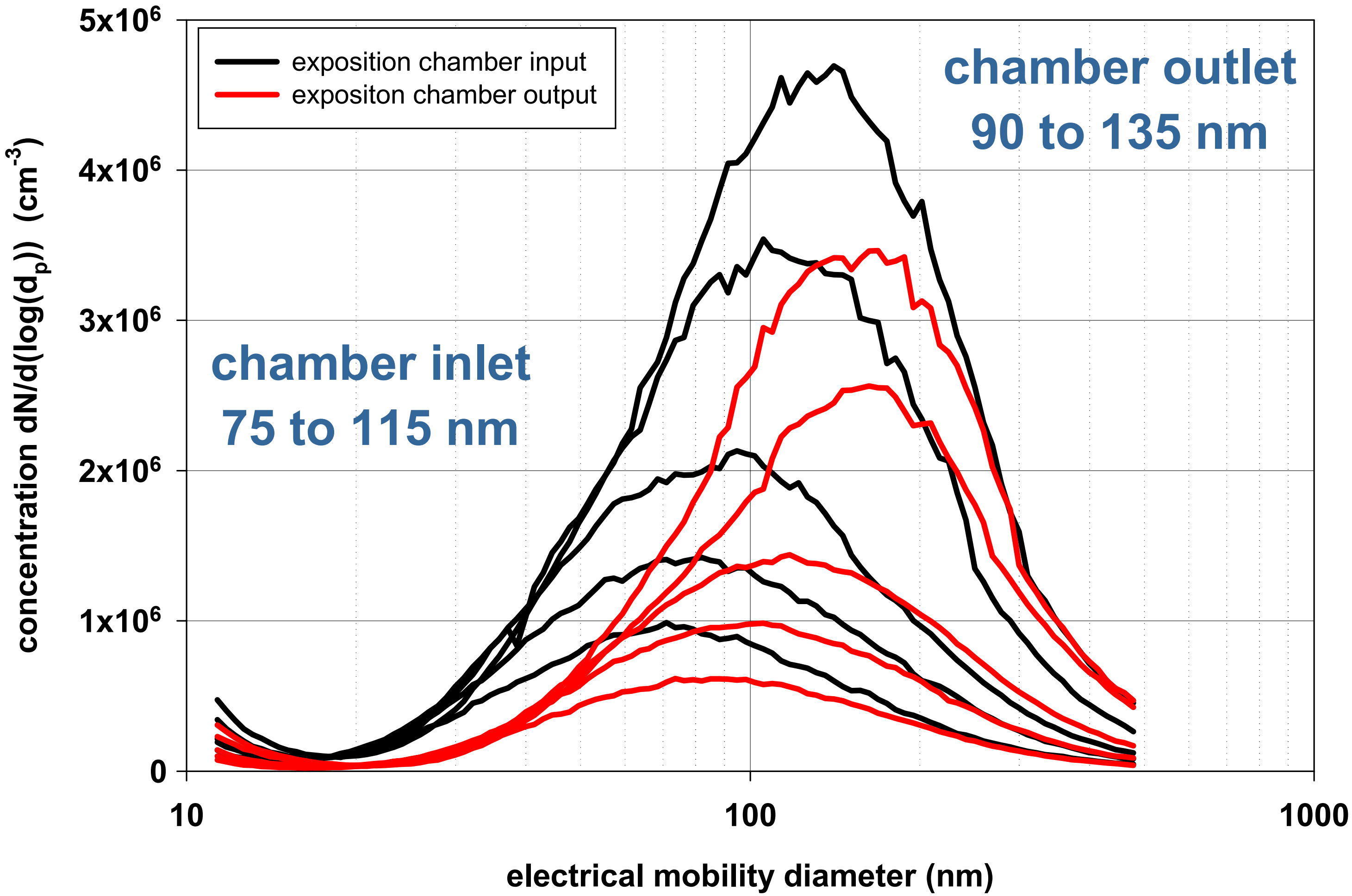
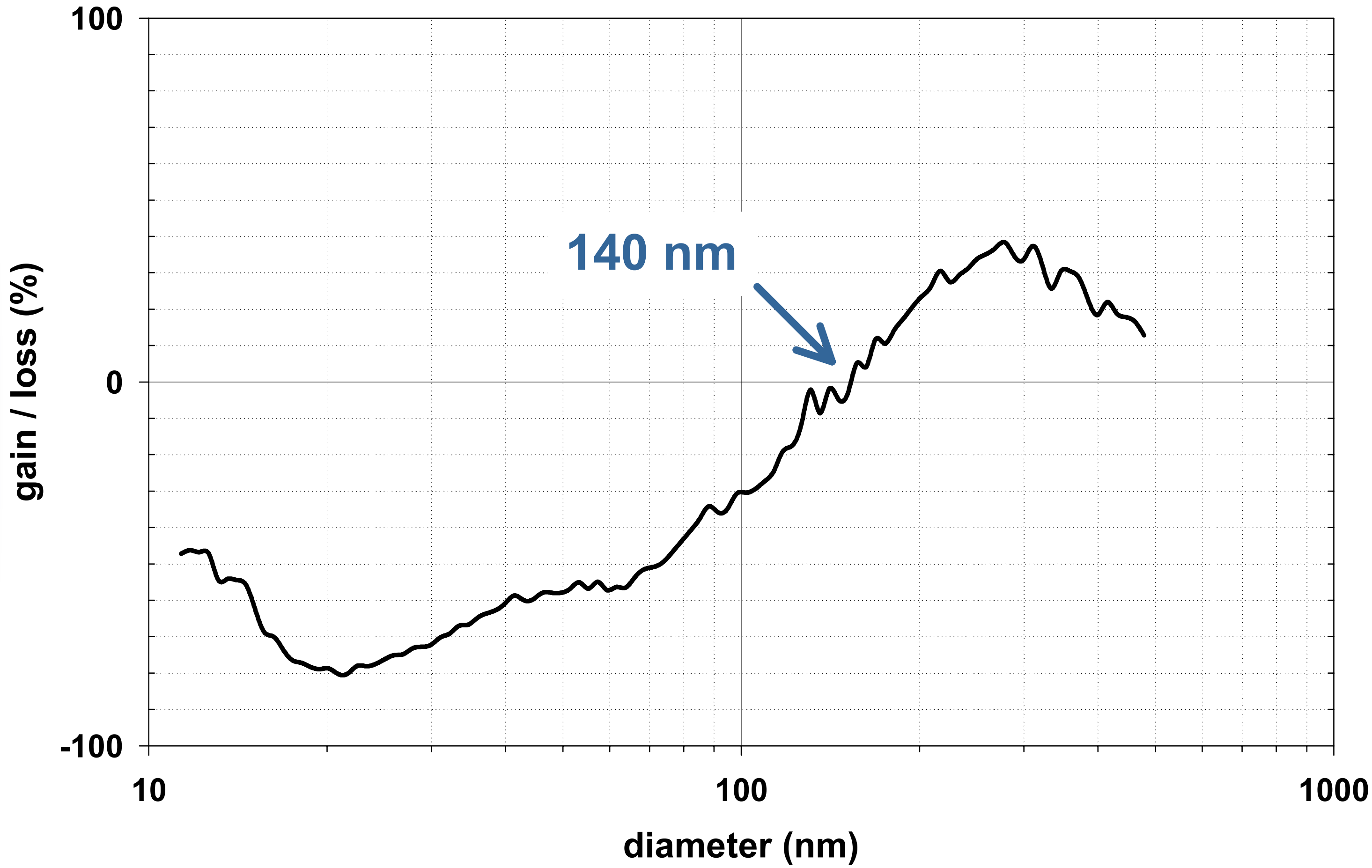
aggregates sizes / sampling point



outlet / inlet



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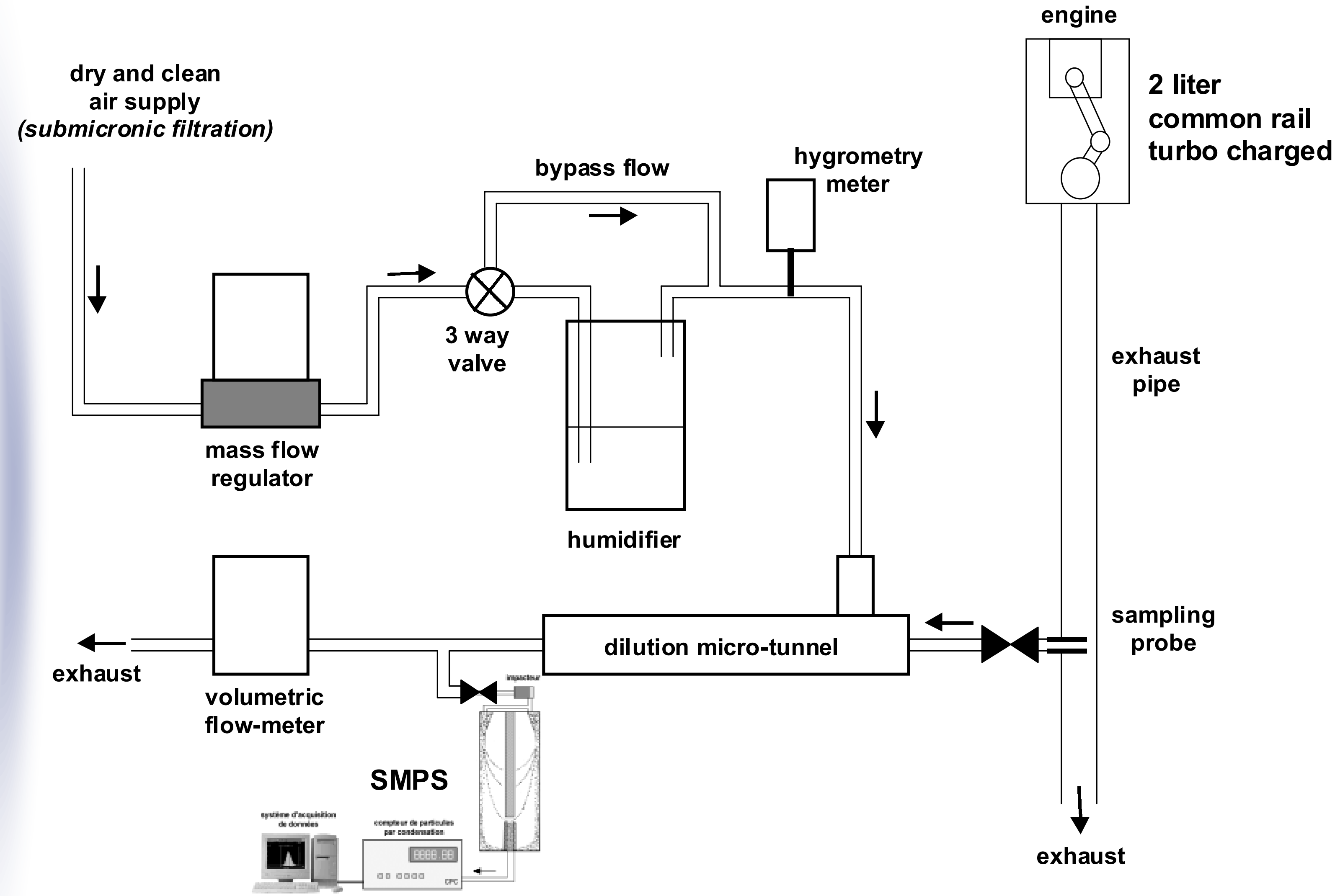
3 ways of explanation

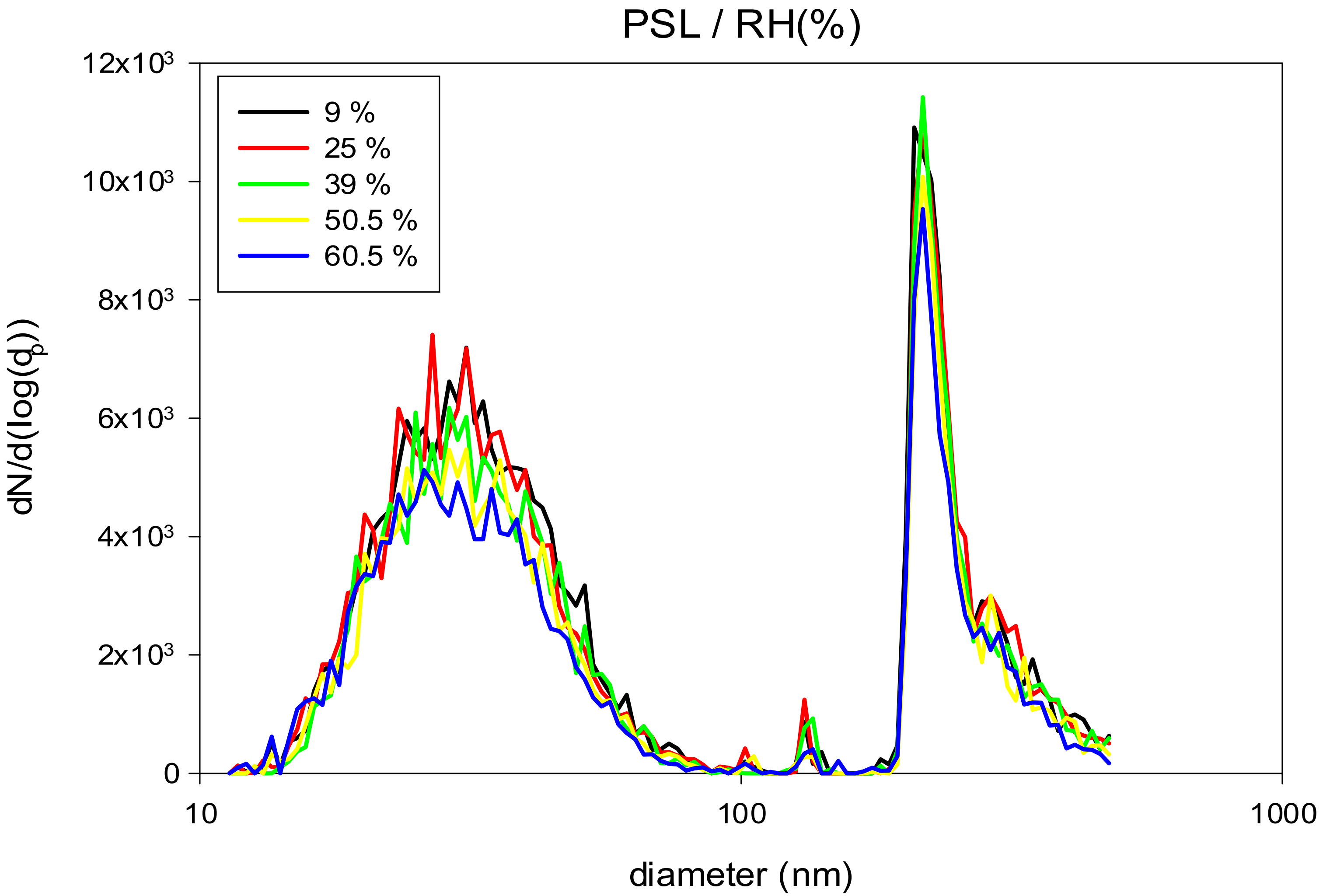
- **the effect of hygrometry on the measurement device (SMPS)**
- **the effect of hygrometry on soot aggregates**
- **coalescence of soot particles /**
  - concentration
  - residence time

experimental setup / engine



SMPS / hygrometry (1)

effect of hygrometry on the SMPS size analyser



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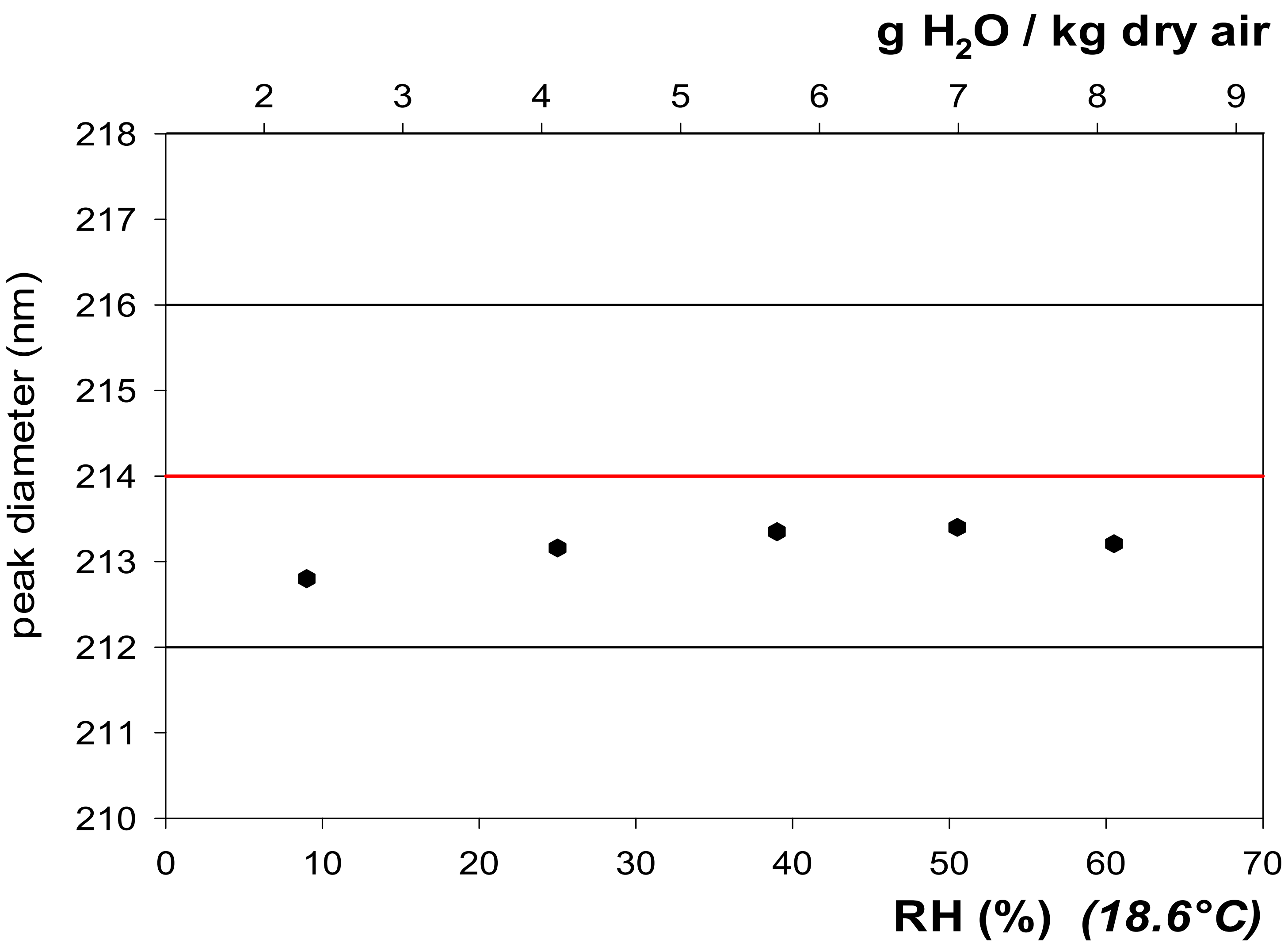
SMPS / hygrometry (2)

effect of hygrometry on the SMPS size analyser



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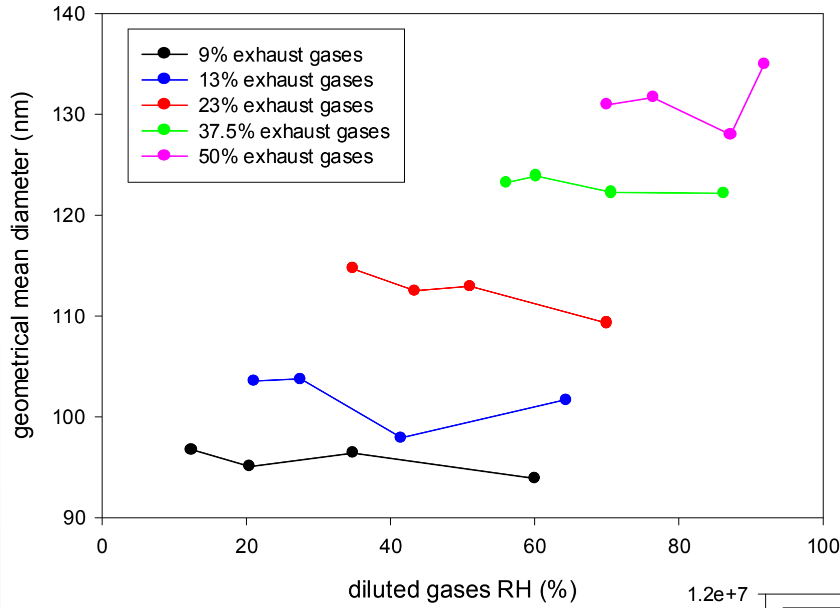
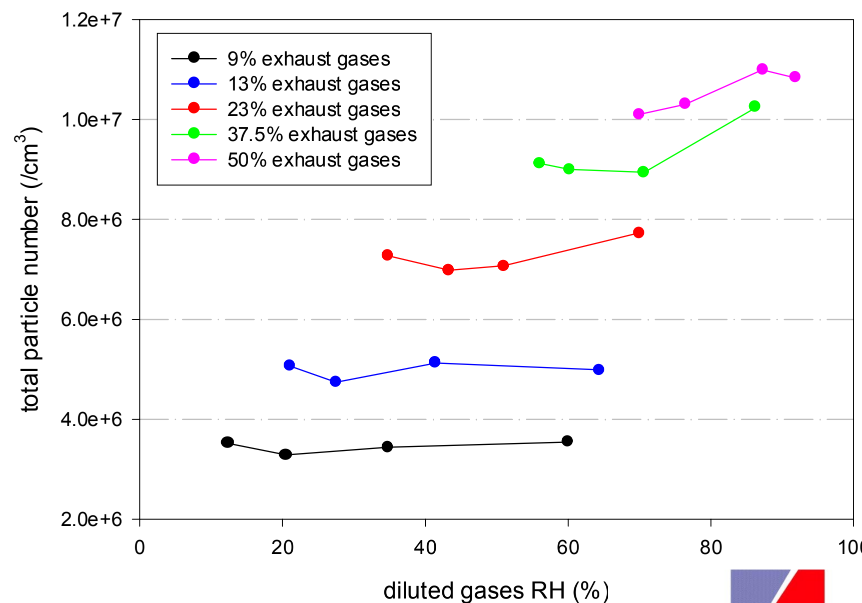
d & N / RH (%)

effect of hygrometry on soot aggregates





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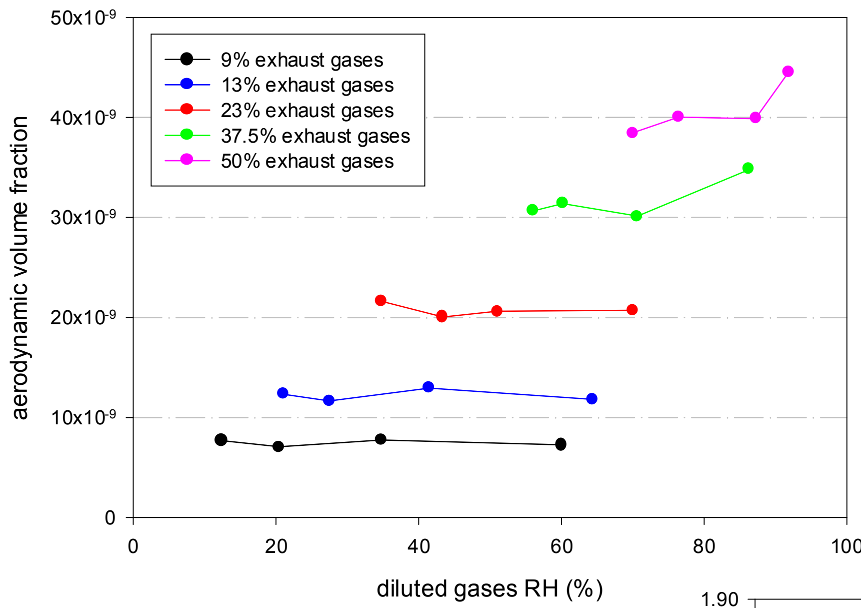
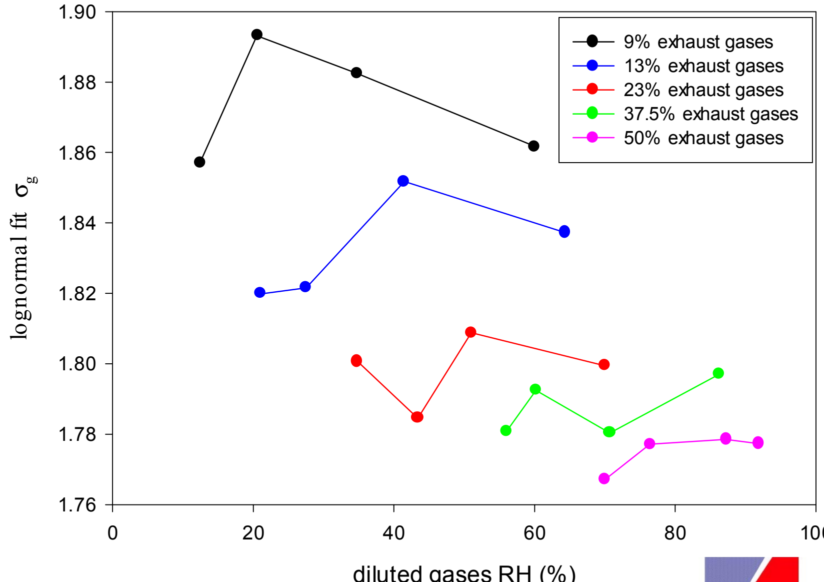
vol. frac. & σ_g / RH (%)

effect of hygrometry on soot aggregates





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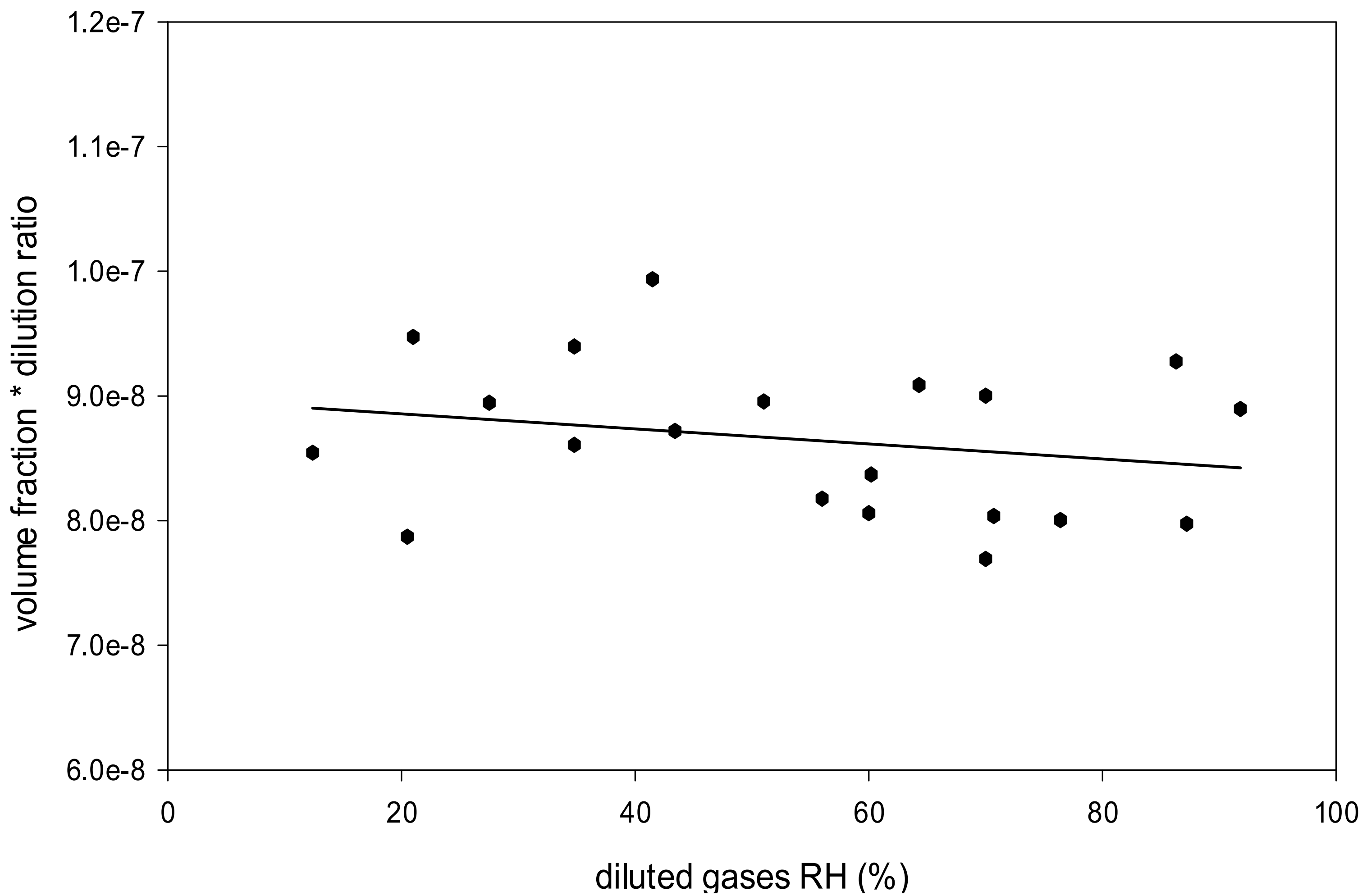
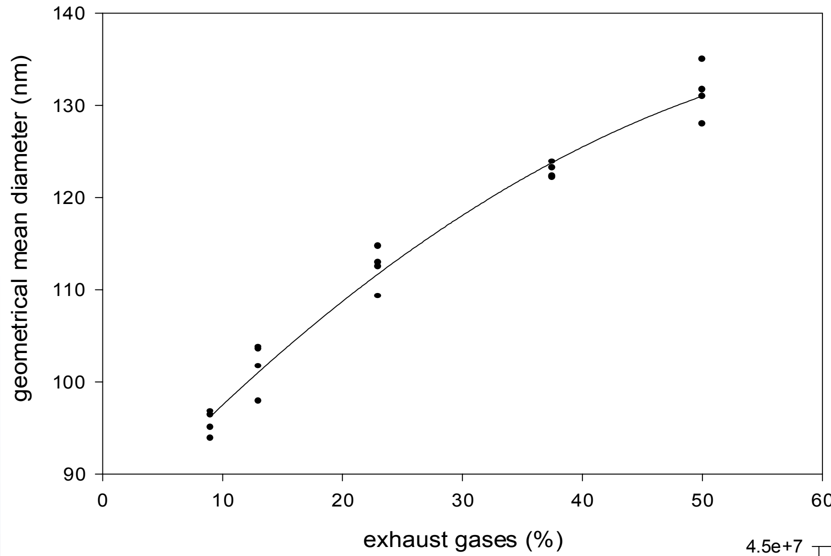
(volume fraction) x (dilution ratio) / RH (%)



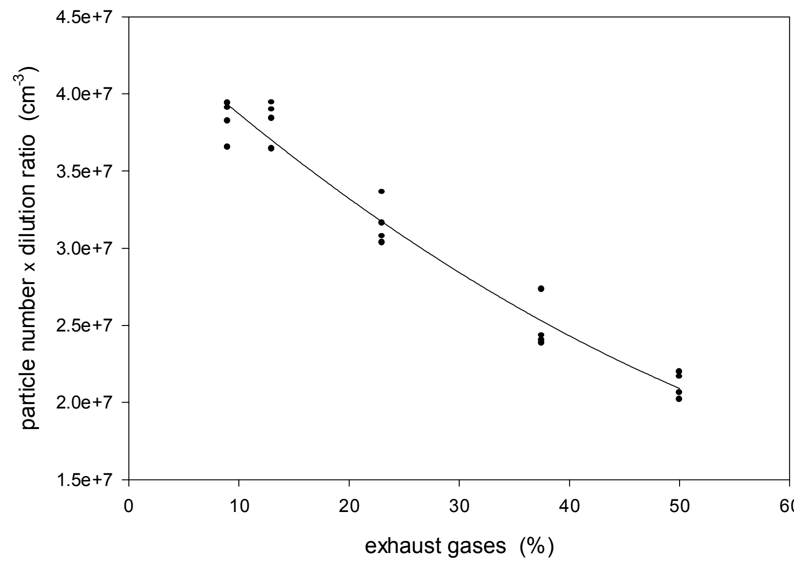
d & N / exhaust gases (%)

effect of exhaust gases concentration on aggregates



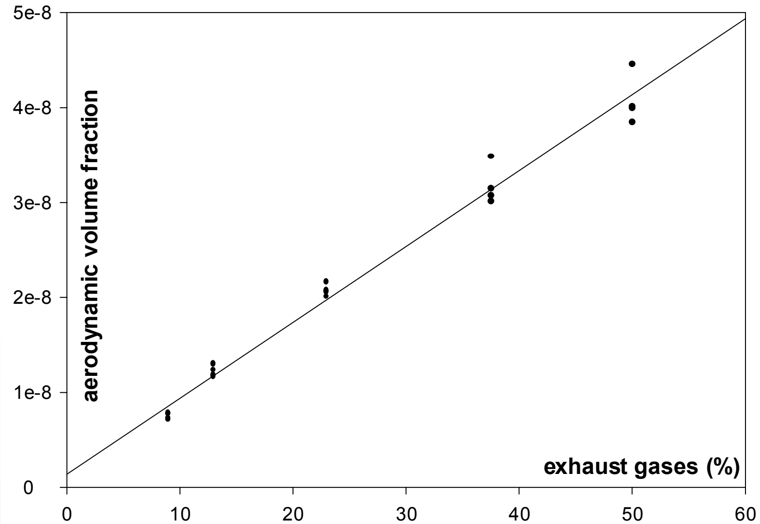
d ↗

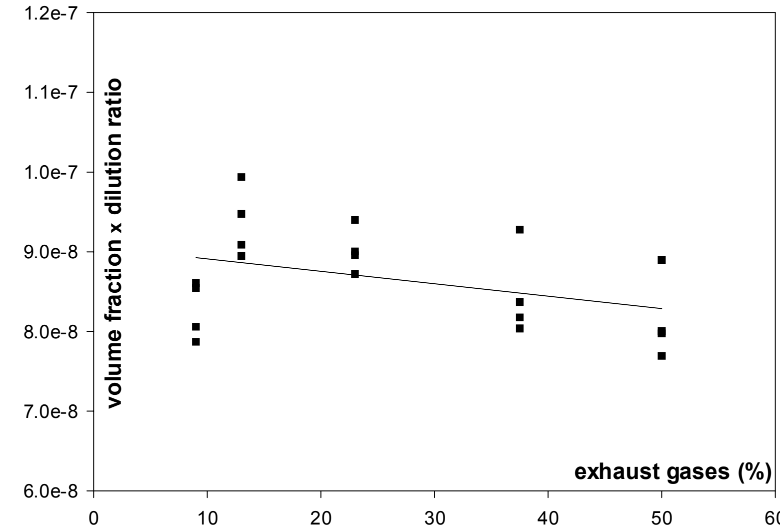
N x dil. ratio ↘



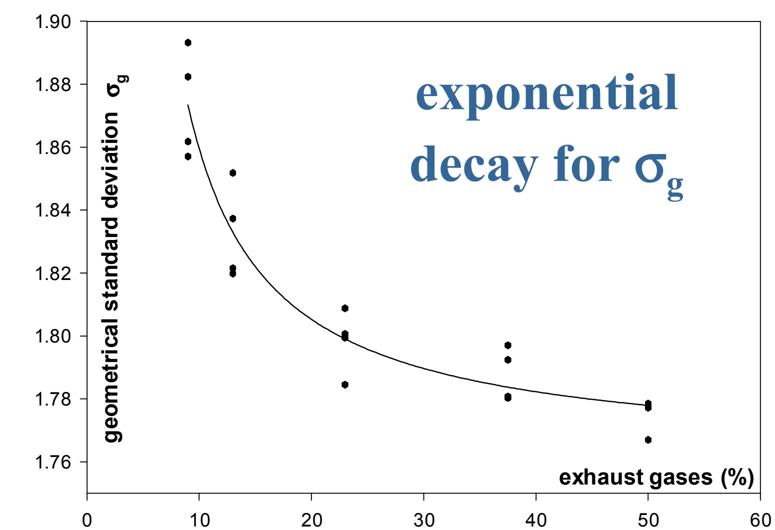
vol. frac. & σ_g / exhaust gases (%)

effect of exhaust gases concentration on aggregates



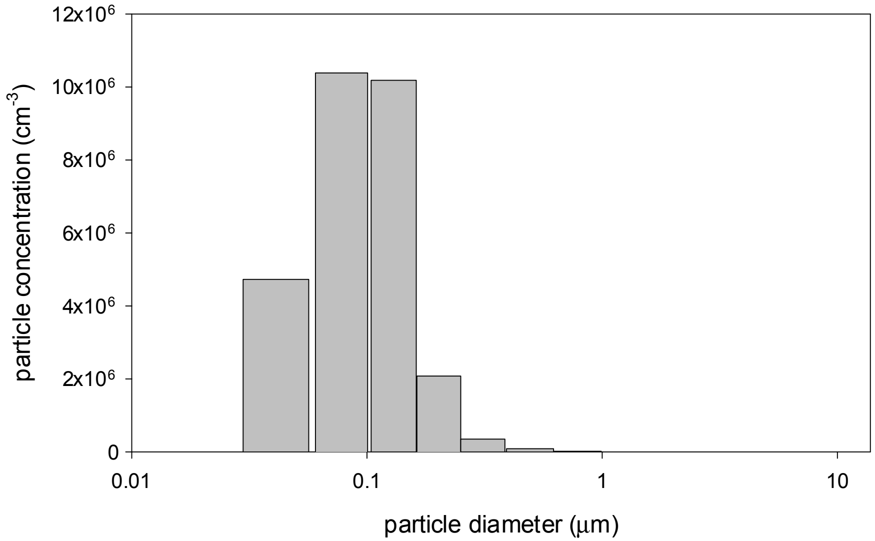


the aerodynamic volume fraction is proportional to the % of exhaust gases and vol.frac.x dil.ratio constant with EG(%)



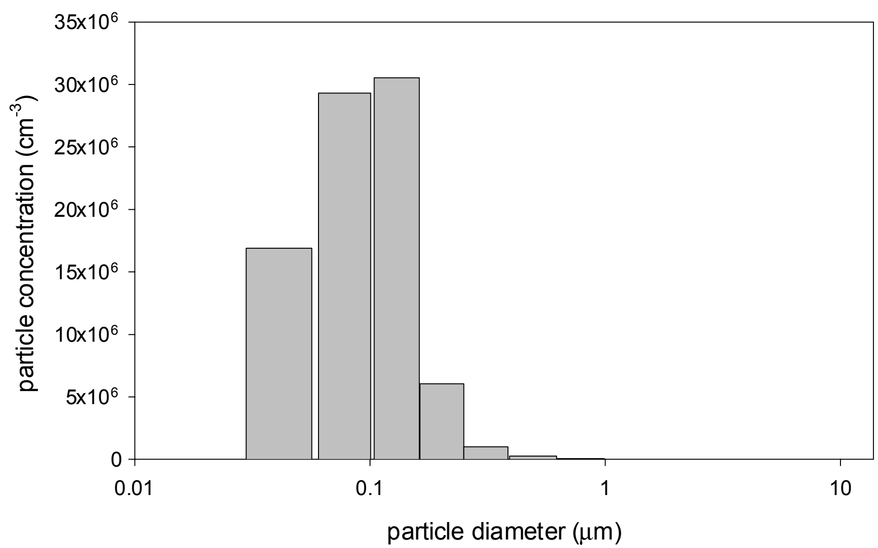
exponential decay for σ_g

particle size shift with ELPI



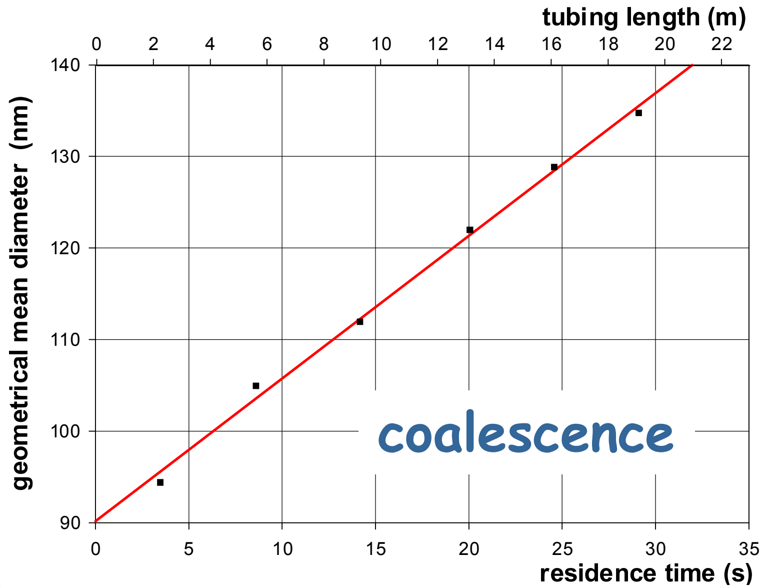
exhaust gases (10%)

exhaust gases (30%)

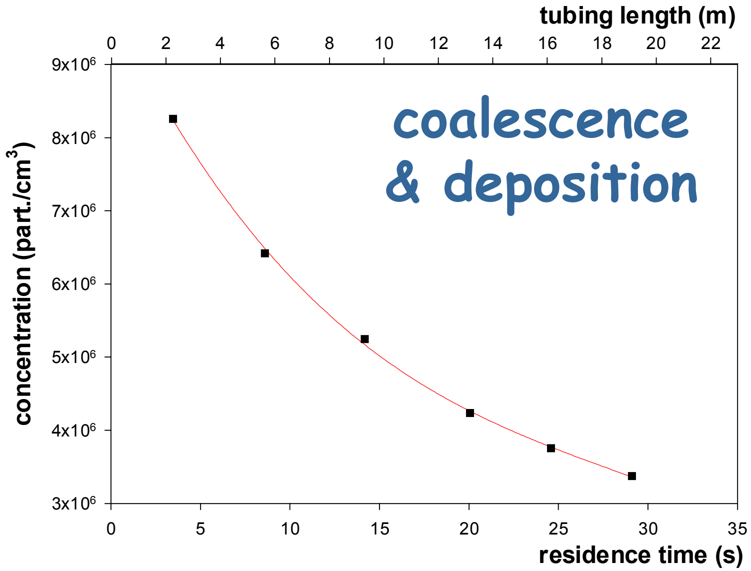


effect of exhaust gases concentration on aggregates

effects of residence time (1)



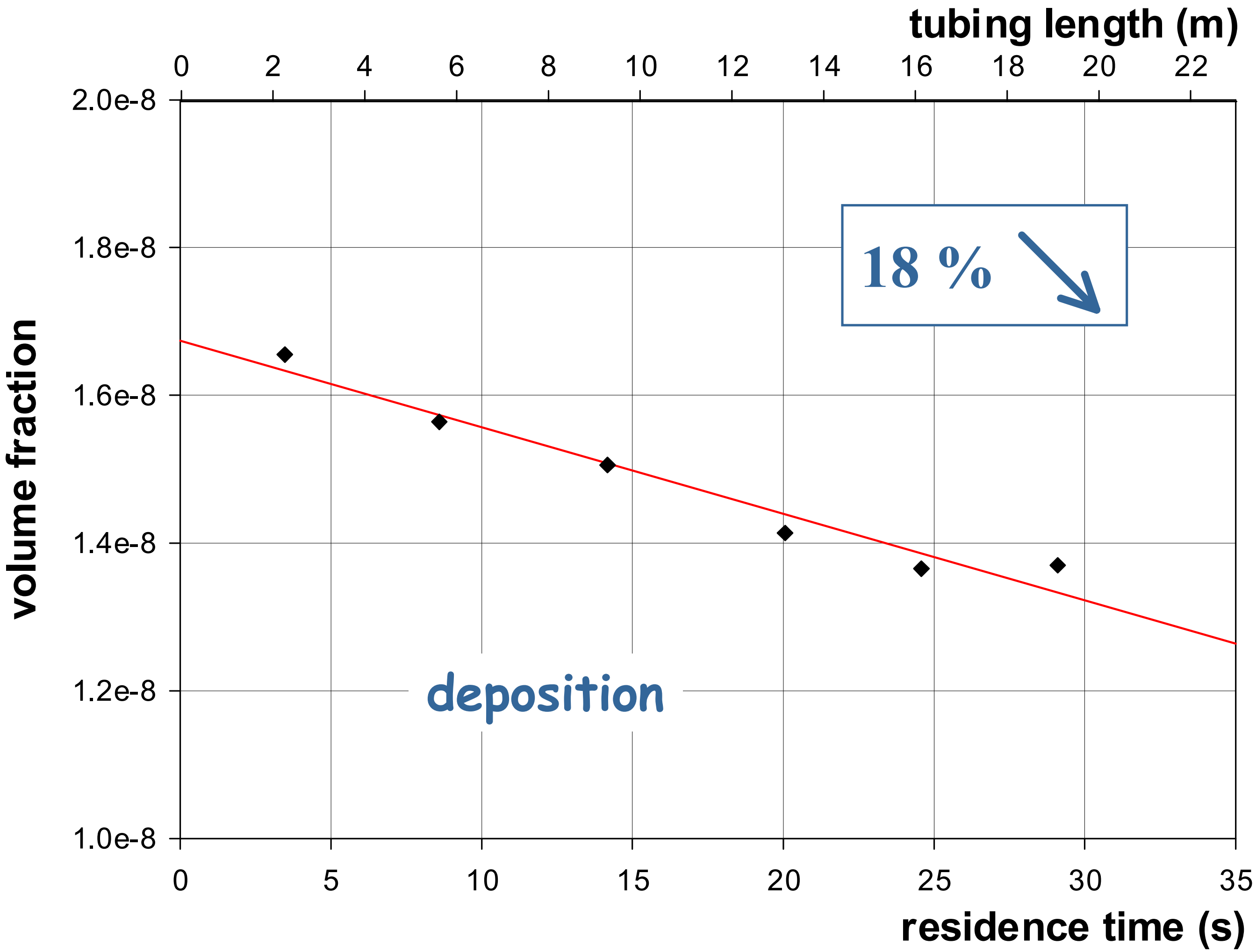
5 s / 95 nm
30 s / 135 nm
linear increase



decrease of 60%

effect of residence time on soot aggregates

effects of residence time (2)



conclusion

- no effect of hygrometry on SMPS
- no effect of hygrometry on aggregates
- particle growth detected by SMPS and ELPI
- noticeable effect of concentration and residence time on aggregates

- particle growth with concentration and residence time (coalescence)
- number decrease (coalescence) or (coalescence + deposition)
- volume fraction constant or decrease (deposition)