Propriétés hygroscopiques de particules de suie exposées au radical OH : vers un possible nouveau mécanisme impliqué dans la formation des trainées de condensation

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Context

- Worldwide aircraft movements number in 2015 were 88 million flights, burned 106 million metric tons of fuel, corresponding to 7 billion passengers and up to 14 billion are expected by 2029 (in absence of pandemic) *(Airports Council International, 2016)*

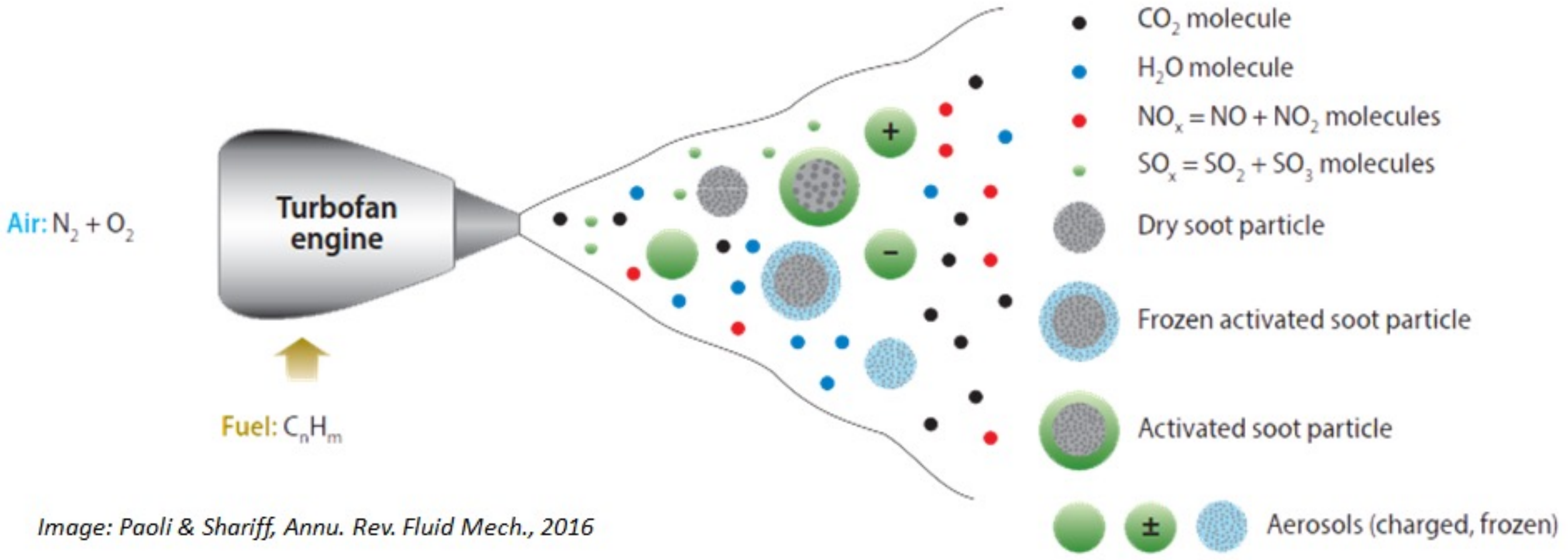


Image: Paoli & Shariff, Annu. Rev. Fluid Mech., 2016

condensation trails
= contrails

- Commercial aircraft engines emit hot gases and particulates such as **CO_x**, **H_2O**, **NO_x**, **SO_x**, **HC** and **soot particles** *(Paoli & Shariff, 2016; Hendricks et al., 2004)*

Context

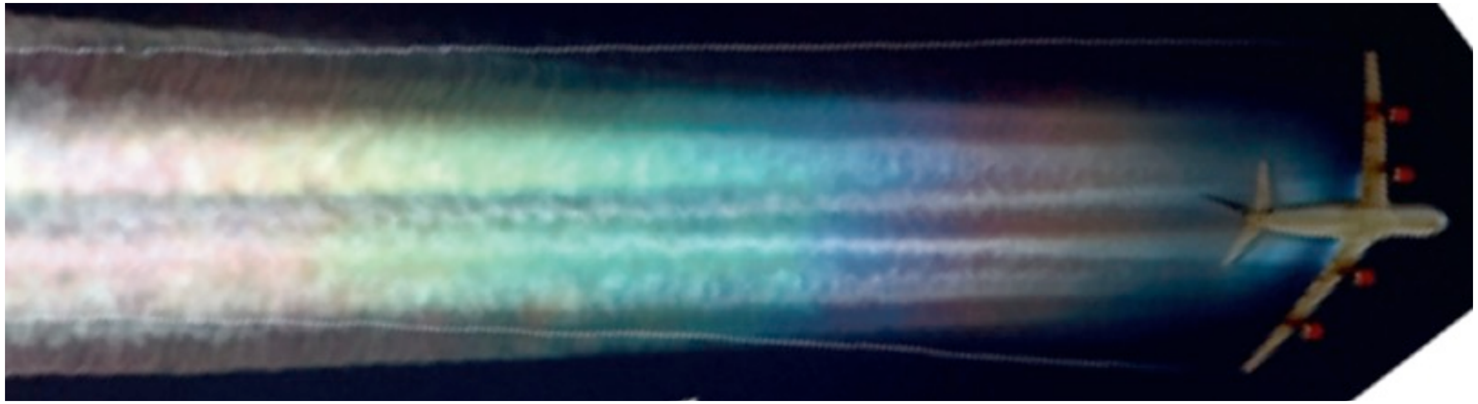
Contrails forms from the transformation of aeronautic soot particles into condensation nuclei for both liquid water (**CCN**) and ice (IN)

Whether certain conditions of (p, T, RH) are met, contrails can trigger the formation of cirrus-like clouds and affect the **radiative forcing of the atmosphere**.

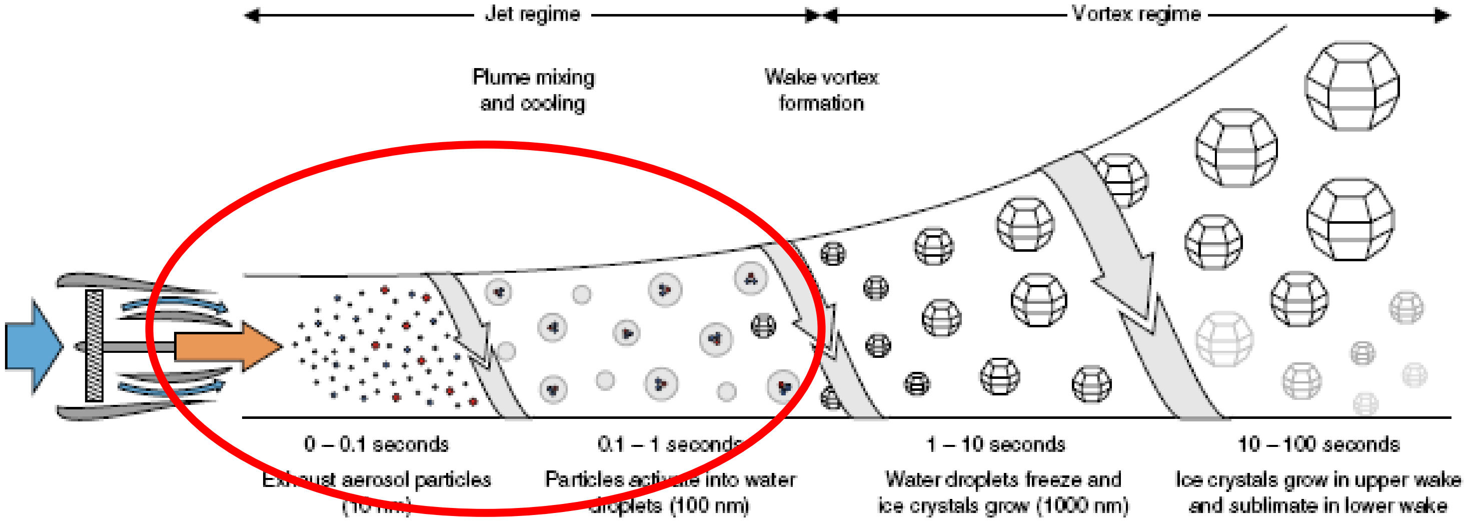
The contribution of Aircraft-induced forcing (AIC) accounts for more than 50 % of forcing fraction within aviation components, larger than CO2 (40%) *(B. Kärcher, Nature Comm. (2018)*

The contrails were estimated to cover about **0.1% of the Earth's surface** with larger regional values up to **15%**. *(R Sausen et al, 1999; Ponater et al, 2002)*

Contrails formation



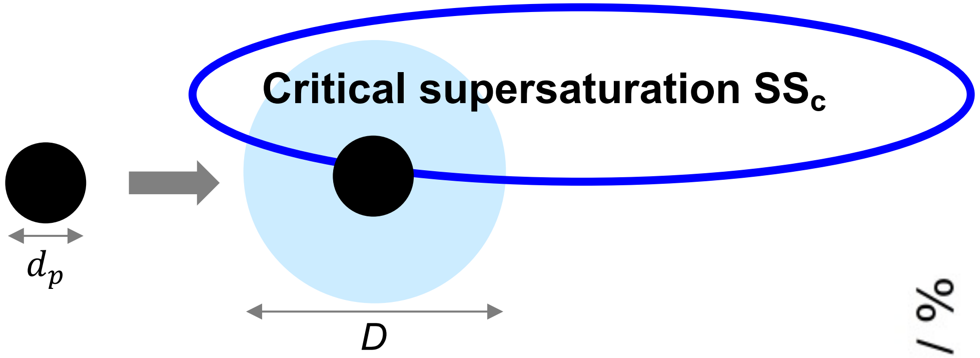
Petzold et al., J. Geo Research 102 (1997)



Kärcher, Nature Comm 9 (2018)

Formation of CCN according to the κ-Köhler theory

Cloud condensation nucleation



Critical supersaturation SS_c

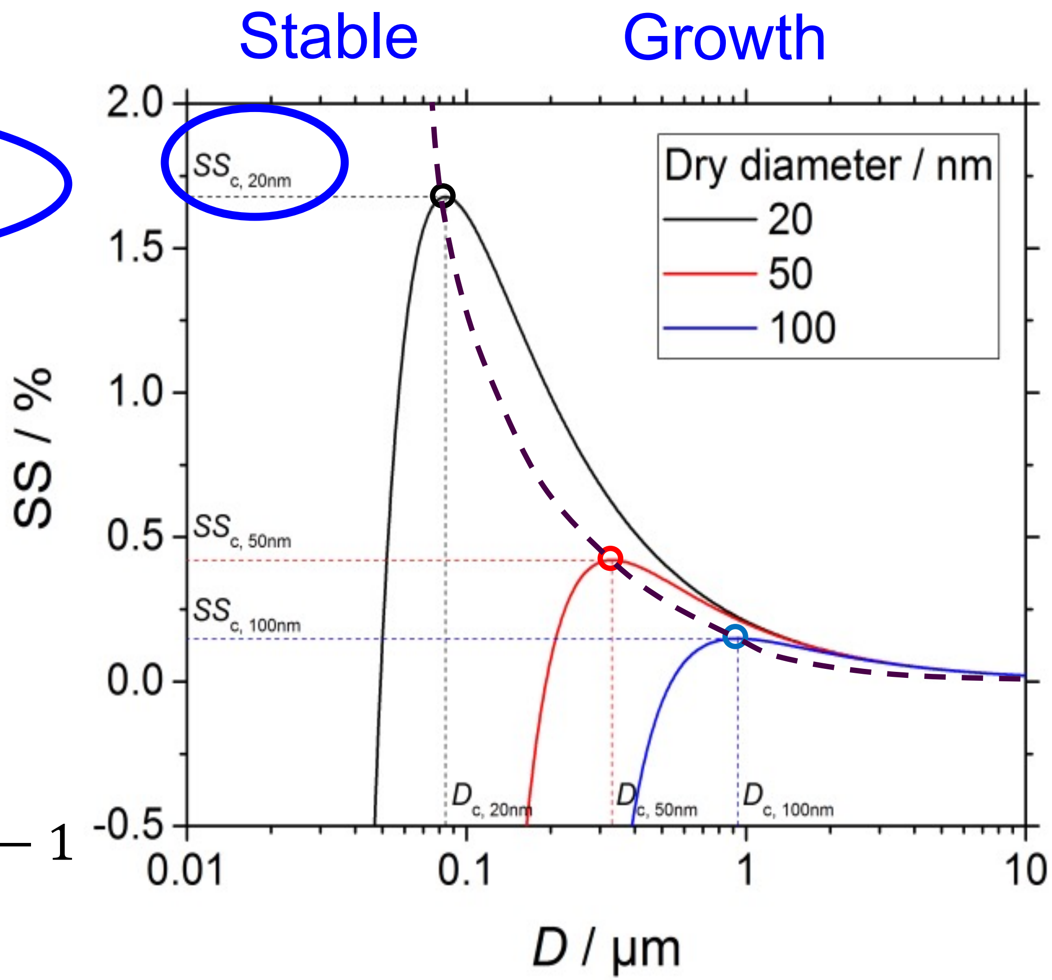
***SS*: supersaturation**

$$SS = \frac{p_v}{p_v^s} - 1$$

(Petters and Kreidenweis 2007)

$$SS(D) = \frac{D^3 - d_\mathrm{p}^3}{D^3 - d_\mathrm{p}^3(1-\kappa)} \exp\left(\frac{A(T,\sigma)}{D}\right) - 1$$

κ: hygroscopicity parameter



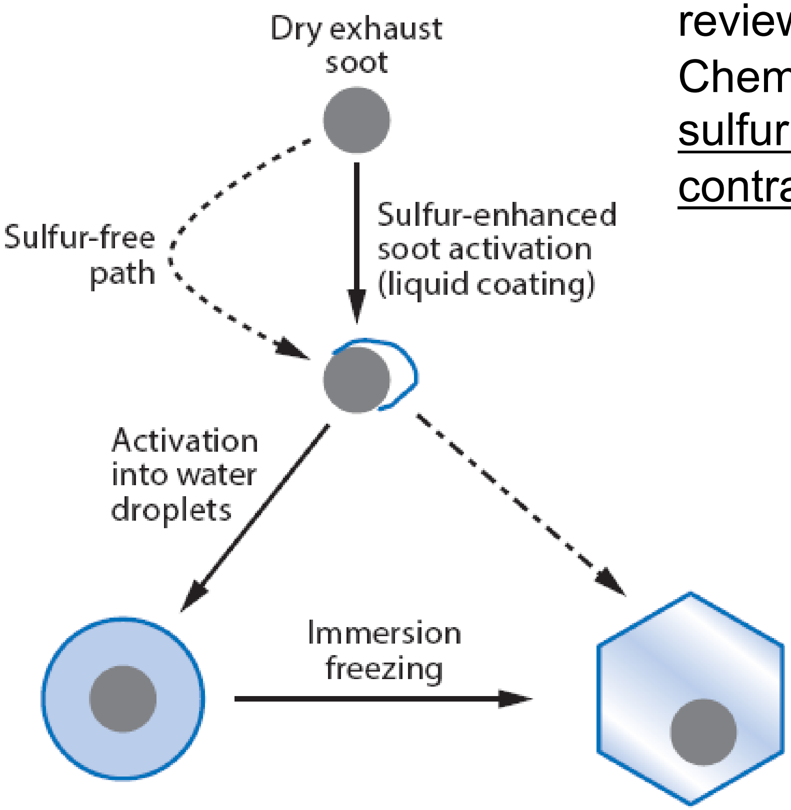
Ammonium sulfate $(HN_4)_2SO_4$ (**κ=0.61**)

Objective of our work

What happens in case of soot?

Many experiments have shown that freshly emitted soot particles, either from engines or laboratory flames, are hydrophobic and are therefore considered as poor CCN.

Sulfur is « introduced» to make soot hydrophilic



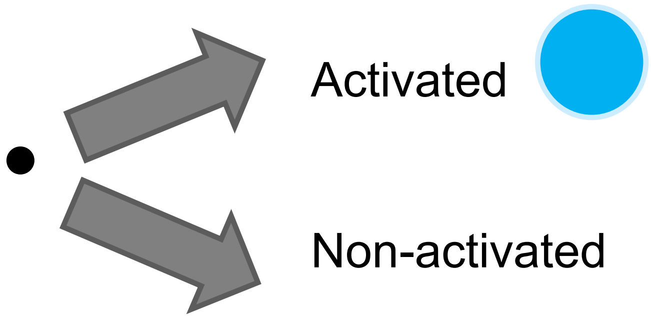
Paoli and Shariff, Ann.Rev. Fluid Mech 48 (2016)

There is no clear consensus in the literature about the role of sulfur in the formation of contrails. Thus, the recent review of the properties of contrails [Schumann et al., Atm. Chem. Phys. 17(2017)] reports that the changes in fuel sulfur content (FSC) were found to have minimal impact on contrail onset.

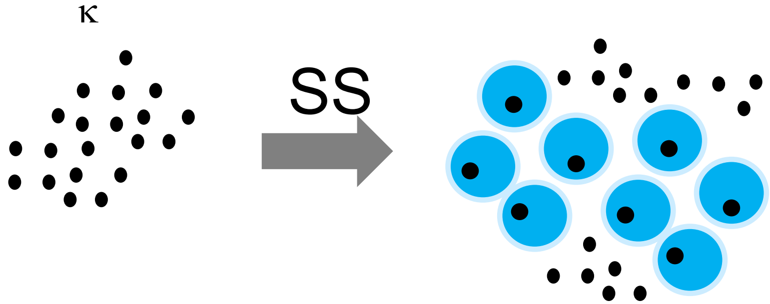
This is the starting point of our study which aims:

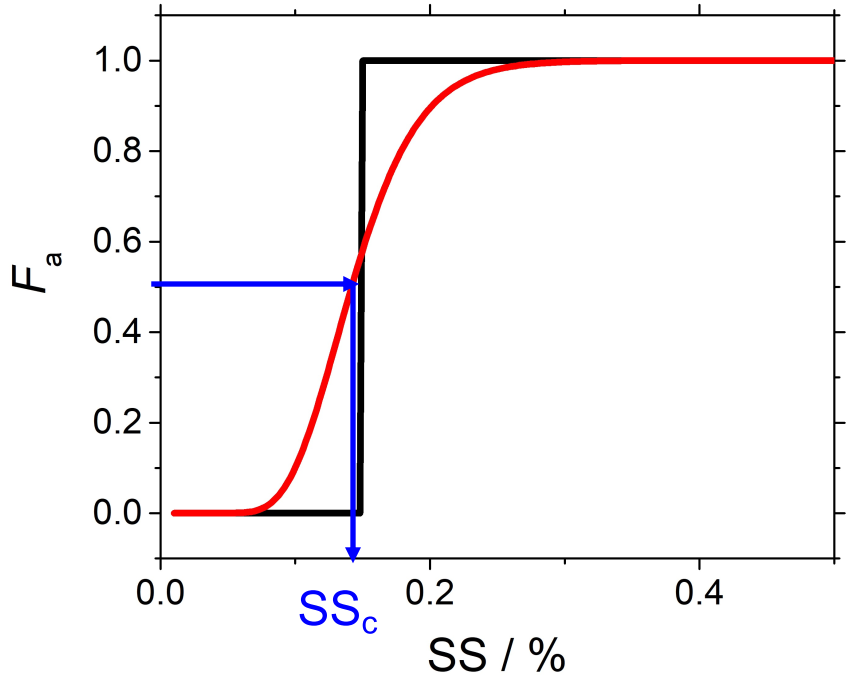
- To compare the hygroscopic parameters κ of soot formed by burning kerosene and a sulfur-free fuel (Diesel)
- To make the particles hydrophilic, we activate them by oxidizing them beforehand under OH radicals exposure, a known efficient activation agent.
- Then considering the oxidation occuring in the postflame of an aeronautic engine, we have examined wether this oxidation would be sufficient to activate the emitted soot particles. OH calculation were made using CFD simulation

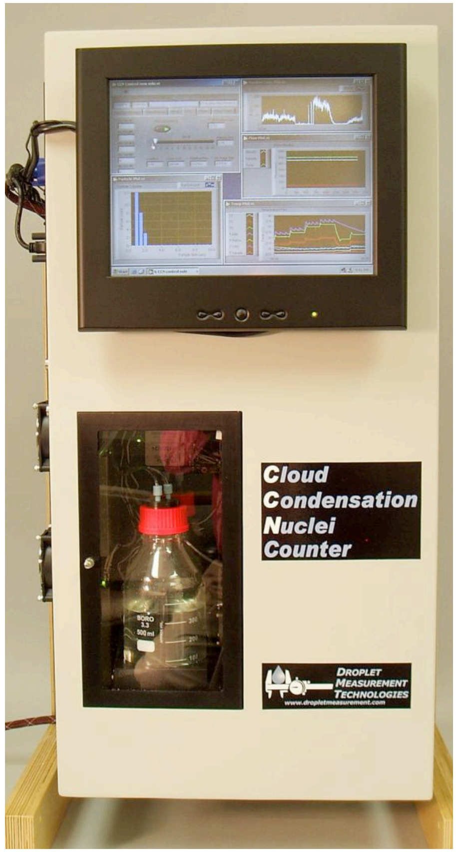
Measure the hygroscopic property in experiment



$$F_{\mathrm{a}}(SS) = \frac{N_{\mathrm{droplets}}(SS)}{N_{\mathrm{particles}}}$$

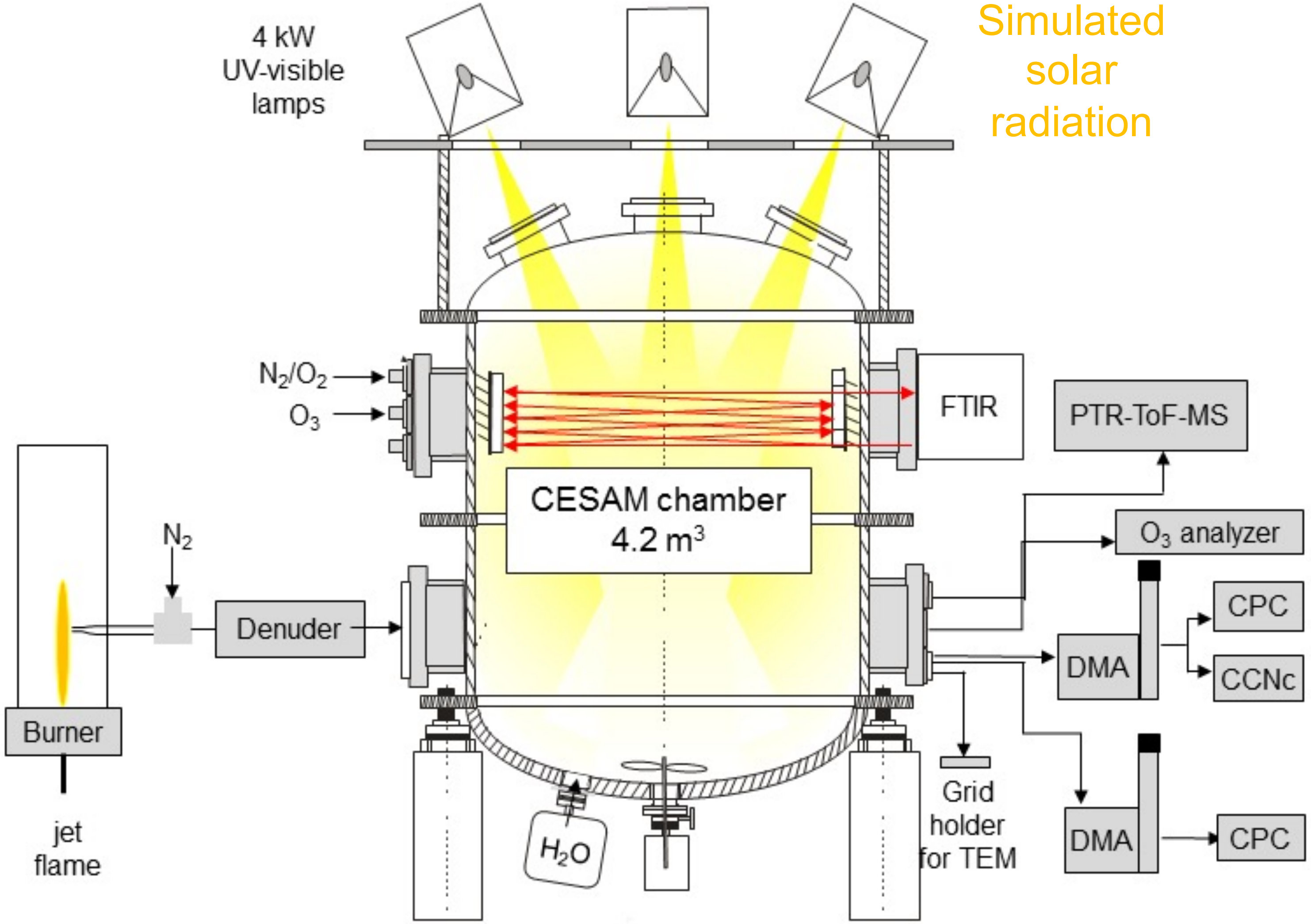


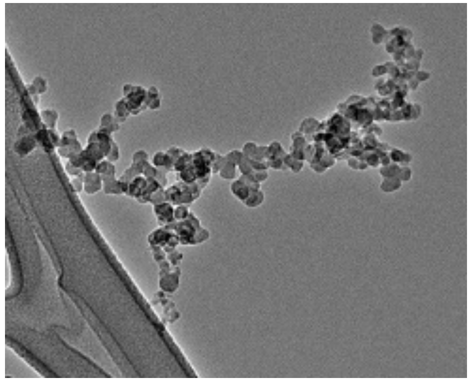




CCNc: Cloud Condensation Nuclei counter

Soot aging in the simulation chamber CESAM

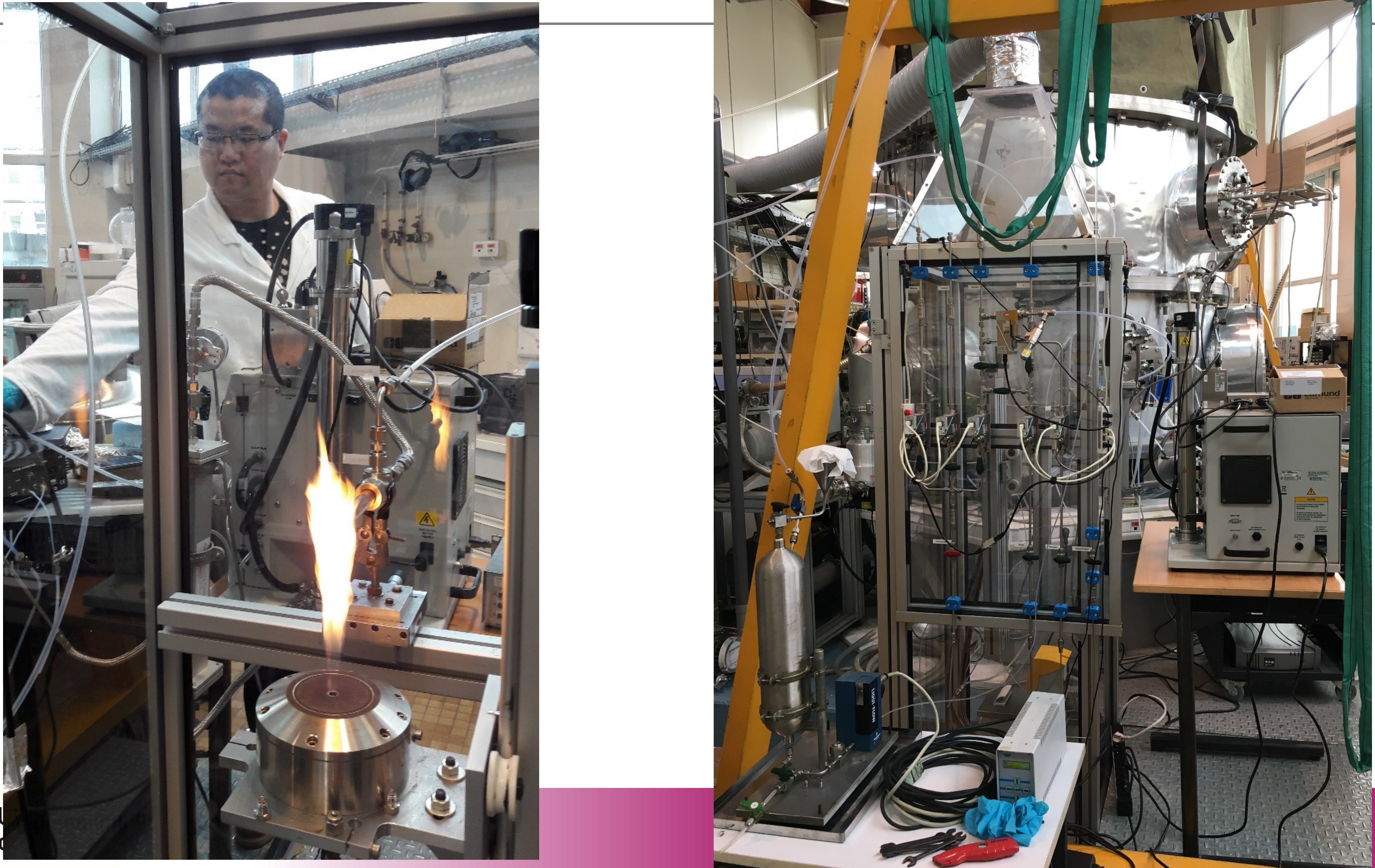




$$O_3 + hv \rightarrow O^{1D} + O_2$$
$$O^{1D} + H_2O \rightarrow 2\ OH$$

DMA: Differential Mobility Analyzer
CPC: Condensation Particle Counter
CCNc: Cloud Condensation Nuclei counter
SMPS: Scanning Mobility Particle Sizer
TEM: Transmission Electronic Microscopy

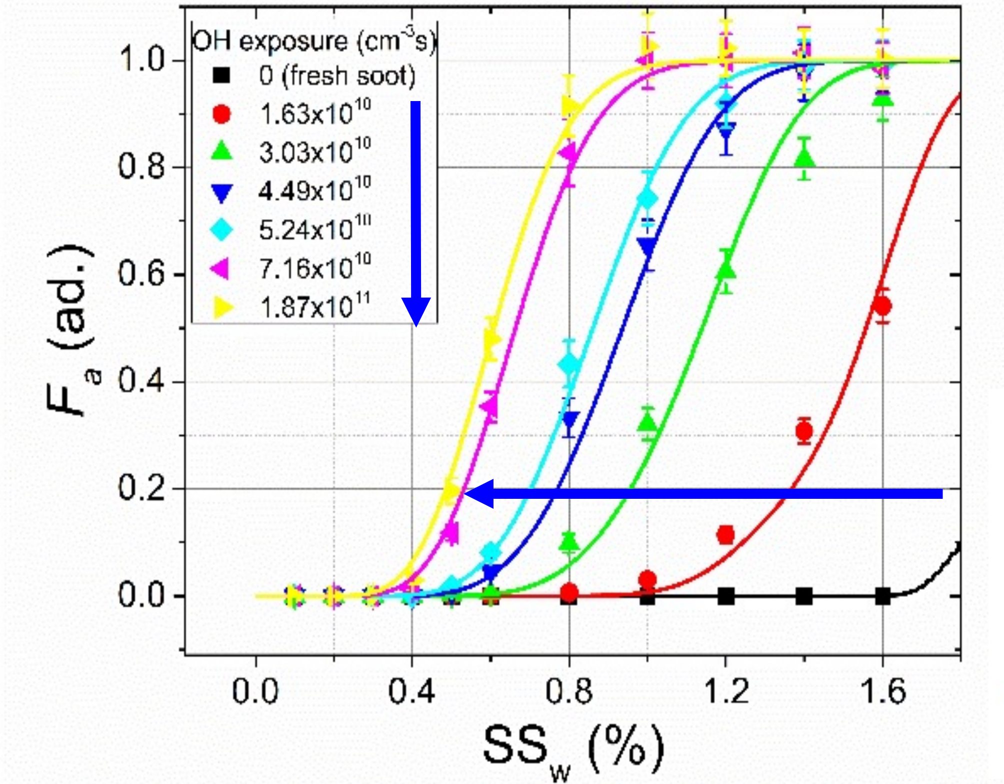
CESAM: Wang et al. Atm. Meas. Tech. 4 (2011)
Burner: Lemaire et al. Fuel 89 (2010)

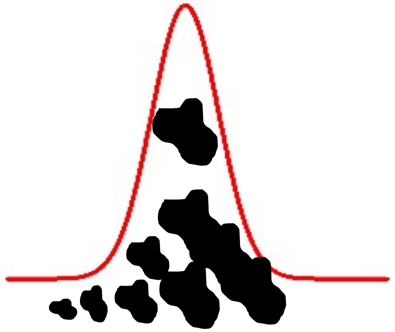


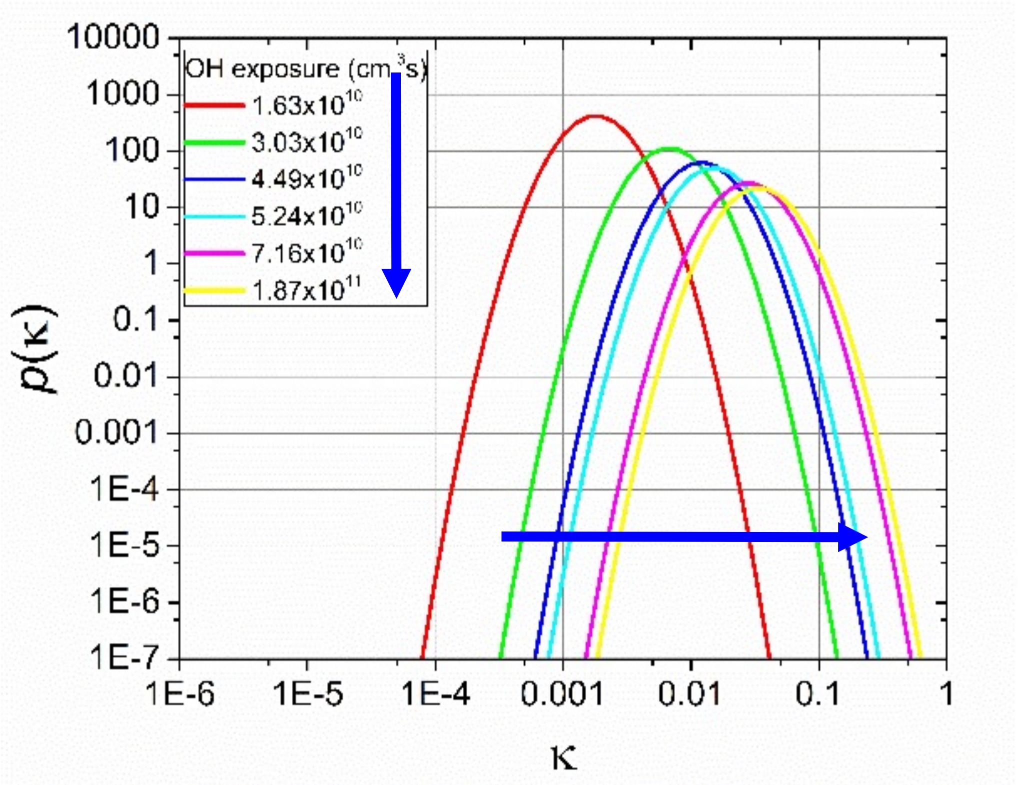
Modified κ-Köhler theory

Wu, Faccinetto, Grimonprez, Batut, Yon, Desgroux, Petitprez, Atmos. Chem. Phys. 20 4209 (2020)

OH exposure ($cm^{-3}.s$) = [OH] x time





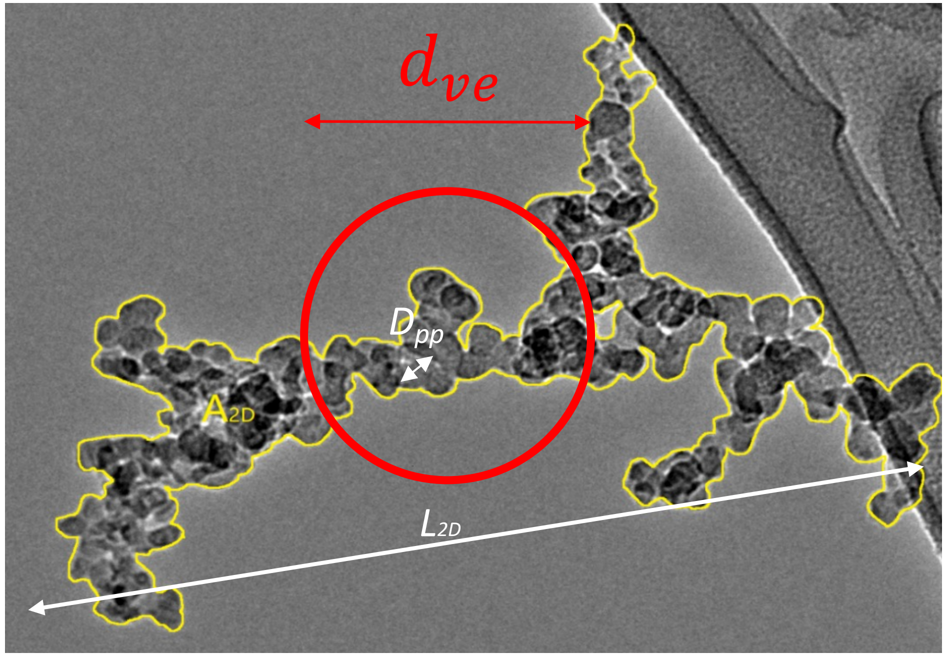


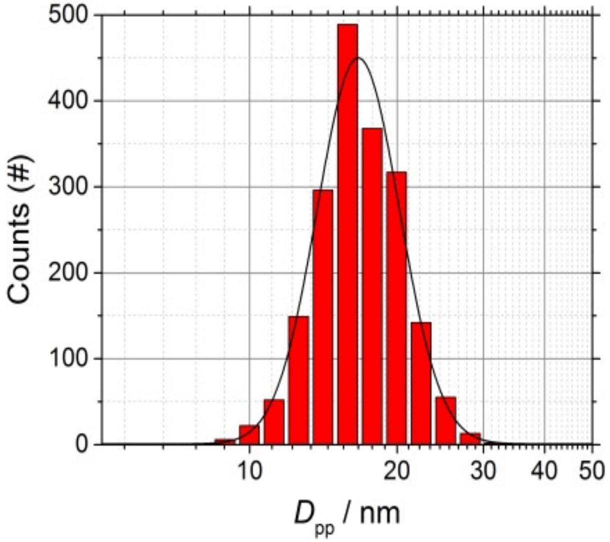
$$F_a(SS) = \sum_{\kappa=0}^{\infty}\left\{\frac{1}{2}-\frac{1}{2}\mathrm{erf}\left[\frac{\ln d_{ve}(\kappa, SS)-\ln \mu_{ve,geo}}{\sqrt{2}\ln \sigma_{ve,geo}}\right]\right\}p(\kappa)\Delta\kappa$$

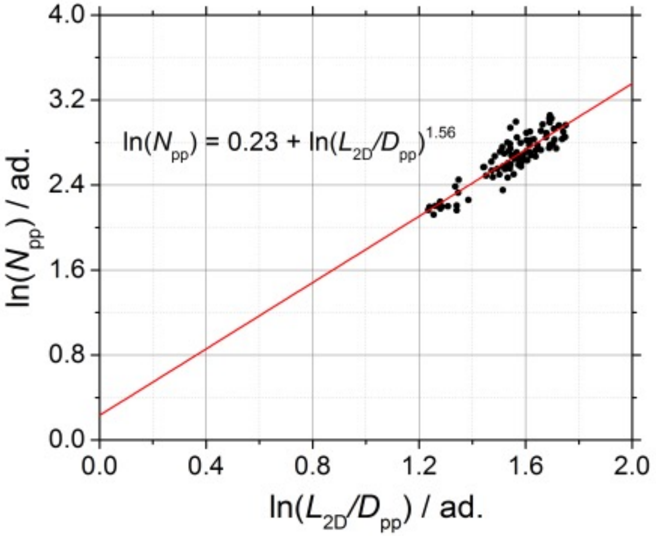
$$p(\kappa) = \frac{1}{\kappa \ln \sigma_{\kappa,geo}\sqrt{2\pi}}e^{-\frac{[\ln \kappa - \ln \mu_{\kappa,geo}]^2}{2\ln^2 \sigma_{\kappa,geo}}}$$

Mode and standard deviation of D_{ve}

Determination of the volume equivalent diameter D_{ve}

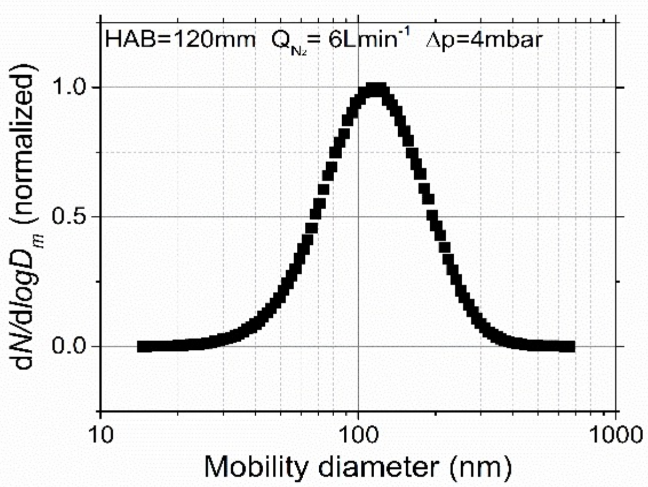


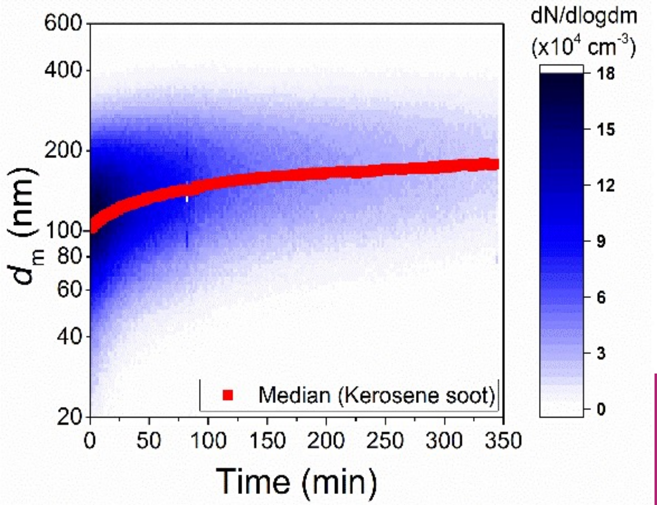




$$d_{\mathrm{ve}}(d_{\mathrm{pp}}, D_{\mathrm{f}}, d_{\mathrm{m}}) = d_{\mathrm{pp}}\left[\frac{d_{\mathrm{m}}}{d_{\mathrm{pp}}}\,\frac{C_{\mathrm{c}}(d_{\mathrm{pp}})}{C_{\mathrm{c}}(d_{\mathrm{m}})}\right]^{\frac{D_{\mathrm{f}}}{3\Gamma}}$$

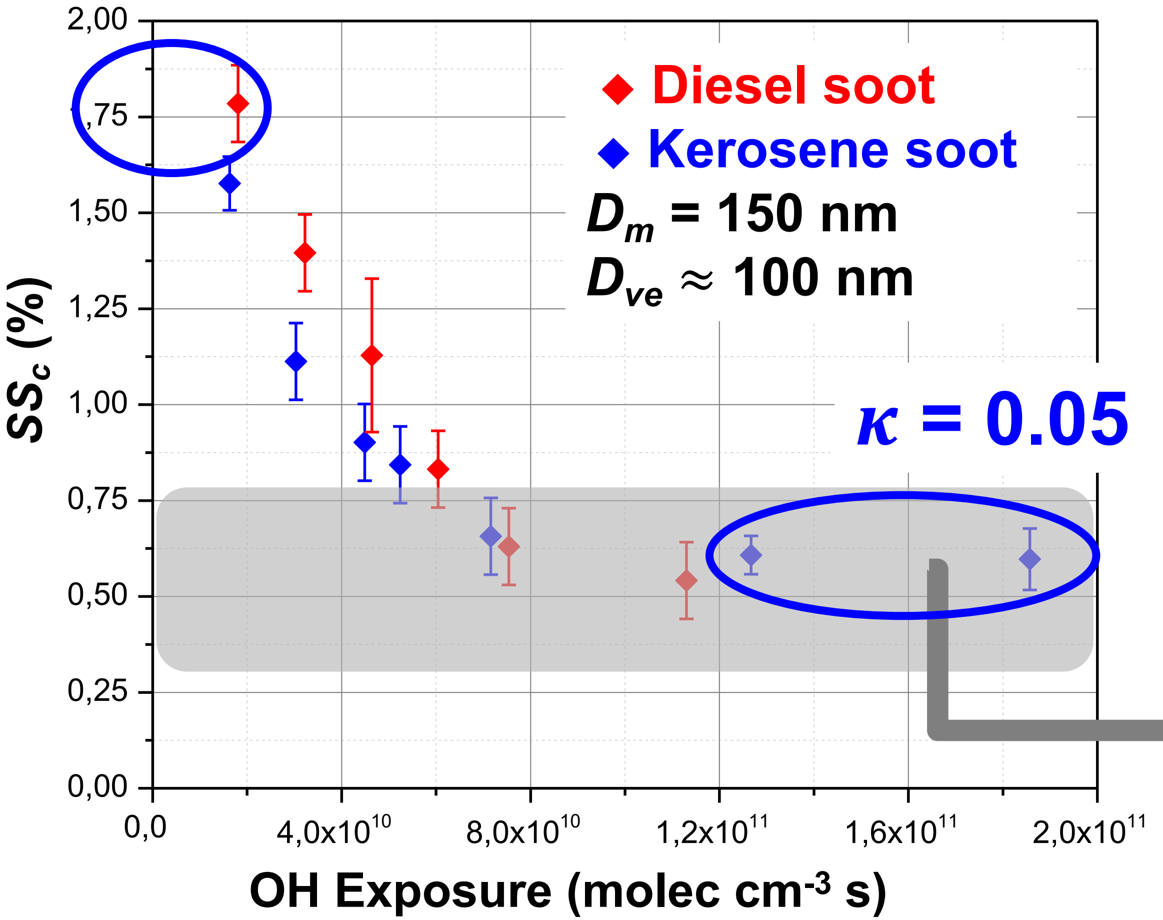
Yon et al. J. Aerosol Sci. 87 (2015)





κ values of kerosene soot and diesel soot exposed to OH radicals

κ <0.0001



« Fresh soot »

« Aged soot »

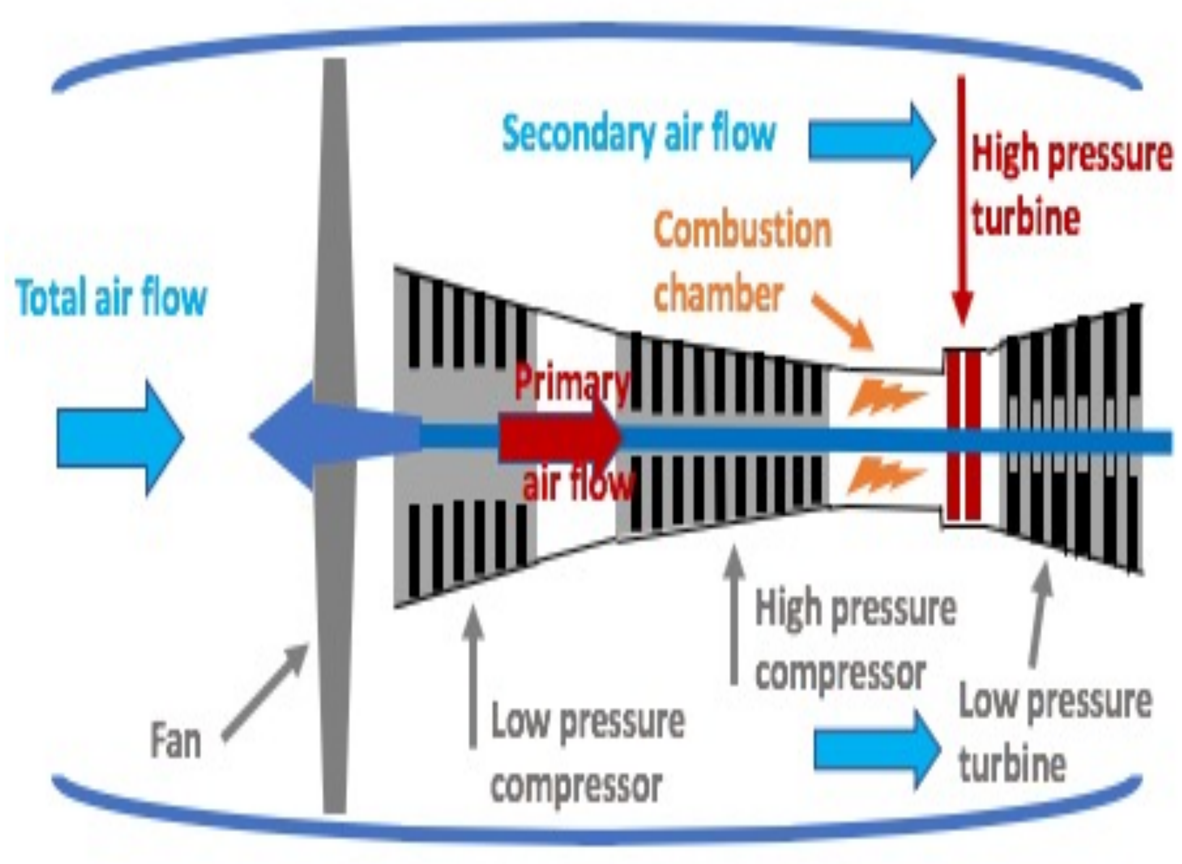
OH aging greatly promotes the hygroscopicity of soot particles. The possibility that soot exiting the turbine may have been sufficiently oxidized in the postflame zone of the combustor to become CCN has never been explored. This is another purpose of our work.

Cloud : *S* : 0,3 – 0,8 %

Pruppacher and Klett, “Microphysics of clouds and precipitations”, Springer 2010

Knowing that OH concentration in the troposphere is around 1.5 10^6 molecule/cm^3 and considering the range of OH exposures in the grey area, it means that between **9 and 28 hours is required to transform aeronautical soot in CCNs in the troposphere**

Mean OH mass fraction



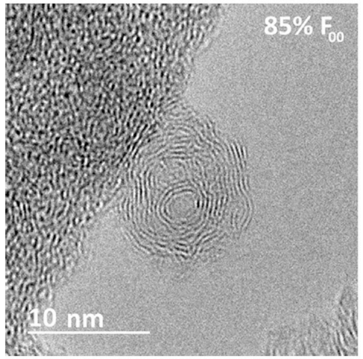
CFD calculations

- Large eddy simulation (LES) in the high pressure turbine is used to model the fluid particle and OH trajectories and their chemical evolution using the code AVBP of CERFACS
- time evolution of the mixture composition is calculated with the detailed chemical kinetics for dodecane using CANTERA
- Y(OH)= 10^{-4} which converts in [OH]=5×10^{15} molecule cm^{-3}
- **only 0.05 ms in the HP turbine is required to reach κ = 0.05 meaning turbine soot is sufficiently active to become CCN!**

Conclusion

- This work investigates the role of soot particles in the early steps of formation of contrails.
- to elucidate the role of sulfur compounds on the activation of soot, a comparative study is performed with soot sampled from a turbulent flame burning kerosene and sulfur-free diesel
- To make soot hydrophilic, soot are exposed to OH radicals in the simulation chamber CESAM before measuring their activated fraction.
- **OH exposure significantly enhances soot activation regardless of the sulfur presence in the fuel.**

HRTEM image of an aeronautical soot



Parent et al. Carbon 101 (2016)

- **CFD calculations in the HP turbine indicate that soot particles have enough time (0.05 ms) to be activated just from their OH oxidation within the turbine, making them potential CCN.**
- **This opens a new possible route to be considered for understanding the formation of first ice particles in contrails**

Thank you

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