UNIVERSITY OF EASTERN FINLAND

Effects of photochemical aging on the chemical and optical properties of exhaust emissions from a small-scale jet engine burner

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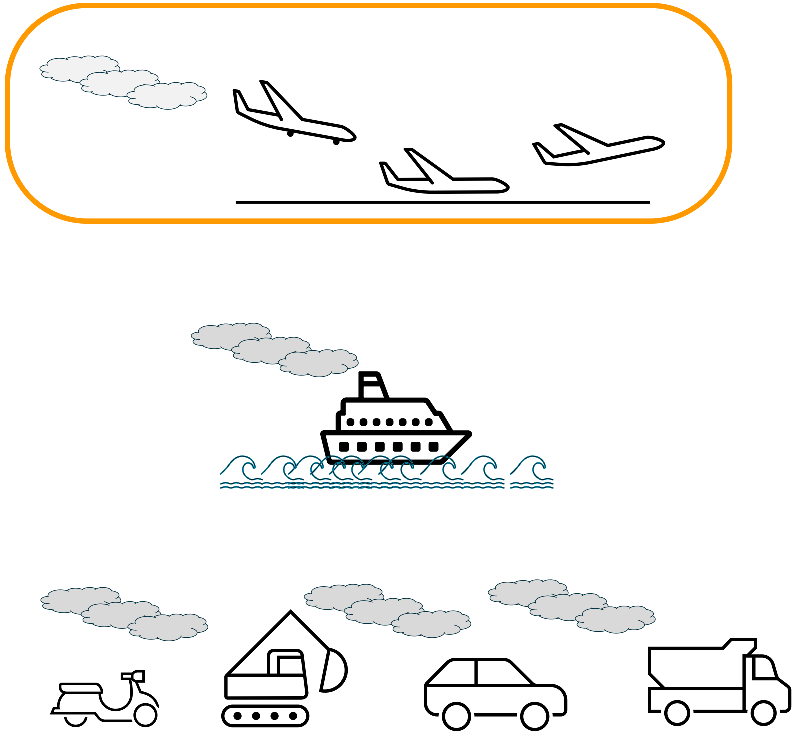
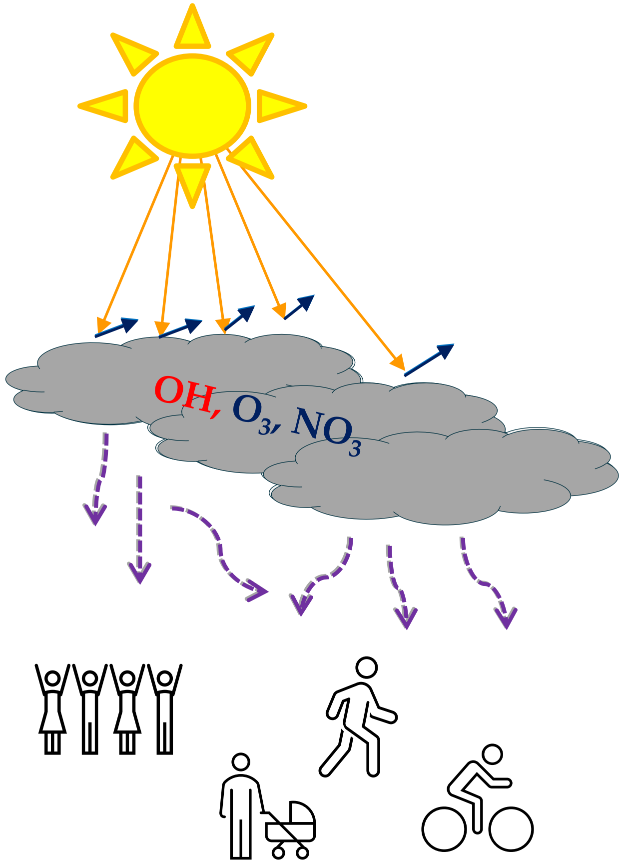
Background





→ Exposure resulting in adverse health outcomes

→ Impact on radiative forcing

ULTRHAS

ULtrafine particles from TRansportation – Health Assessment of Sources

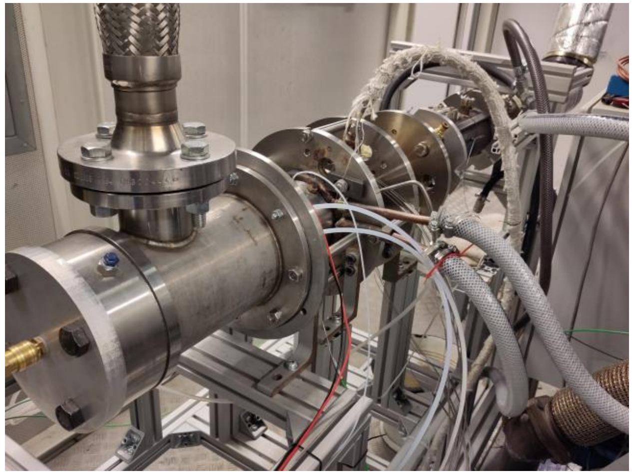
Objectives

- Determine secondary aerosol formation potential for aircraft emissions
- Investigate how photochemical aging influences the aircraft exhaust aerosols:
  - Chemical and physical properties relevant for health impact assessment
  - Aerosol optical properties relevant for climate impact assessment

ULTRHAS

ULtrafine particles from TRansportation – Health Assessment of Sources

Experimental design: combustor rig



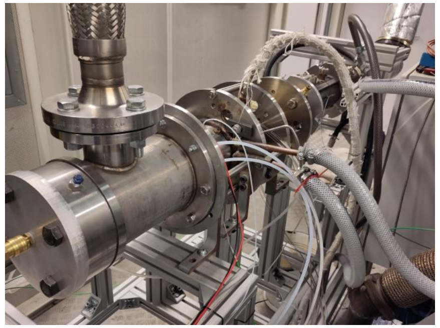
- Combustor rig, incl. combustion chamber of a turbine engine

  ! no compressor, no turbine, no lubrication oil

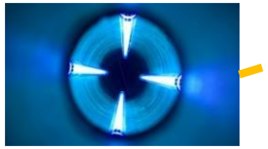
- Fuel: kerosene-based jet fuel (JP-8)

- One operation mode (idle, 7% nominal load) representing the average of a land- and takeoff cycle

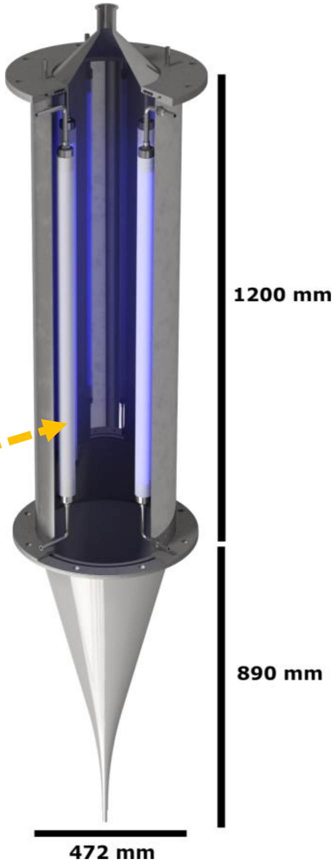
Experimental design: photochemical aging in the Photochemical Emission Aging Flow Reactor (PEAR)[1]



Dilution by 1:50 or 1:200



- 4 x 254 nm UV lamps
- Flow rate 120 lpm



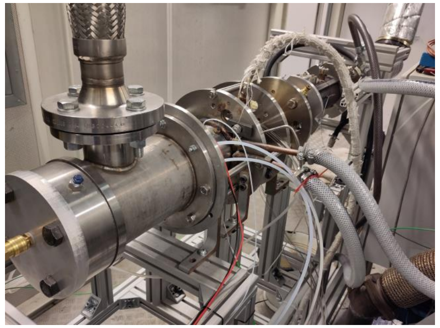
$$O_3 + hv \rightarrow O_2 + O(^1D)$$
$$H_2O + O(^1D) \rightarrow 2OH$$

Hydroxyl radical (OH) exposures equivalent to

1) ~2 days in atmosphere (DR 1:50)
2) 1-7 eqv.d (DR 1:200)

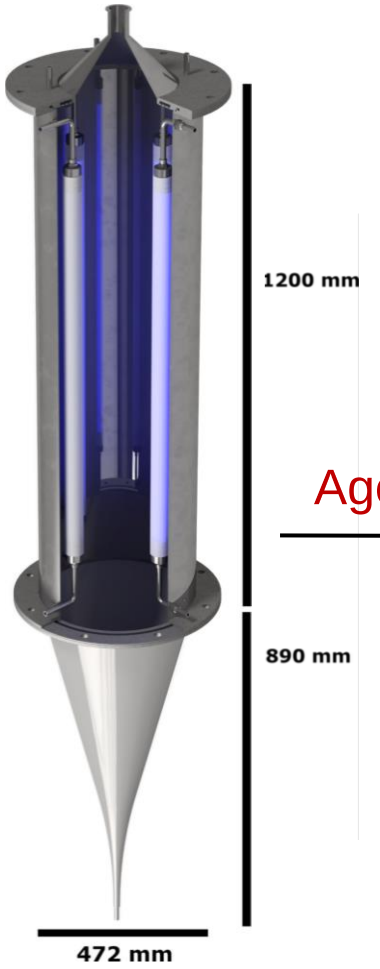
[1] *Ihalainen et al., 2019. A novel high-volume Photochemical Emission Aging flow tube Reactor (PEAR), Aerosol Science and Technology. DOI: 10.1080/02786826.2018.1559918*

Experimental design



Dilution by 1:50 or 1:200

Online characterization of the gas phase (NO_x, SO_2, CO_2, CO (*FTIR*), gaseous hydrocarbons (*FID*))



Aged exhaust

Fresh exhaust

Chemical-physical characterization

Online particle phase measurements (*instrument*)

- Particle number and size (*SMPS, FMPS*)
- Particle mass and composition (*HR-AMS, aethalometer, TEOM*)
- Particle density (*AAC-SMPS)*

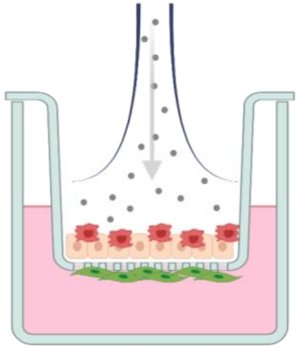
Online gas phase measurements

- *HR-ToF-PTR* : organic gas phase
- *FTIR*: CO2, CO, SO2, NOx

Offline sampling of both PM and VOCs

→ Chemical composition by total carbon analysis
→ Light absorption by UV-vis spectroscopy
→ Morphology (*TEM & SEM*)

Toxicological assessment (DR 1:50)



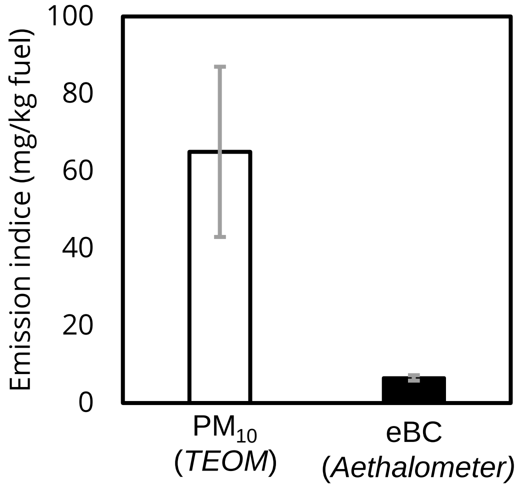
Air-liquid interphase

Cell exposures at different dilutions and doses

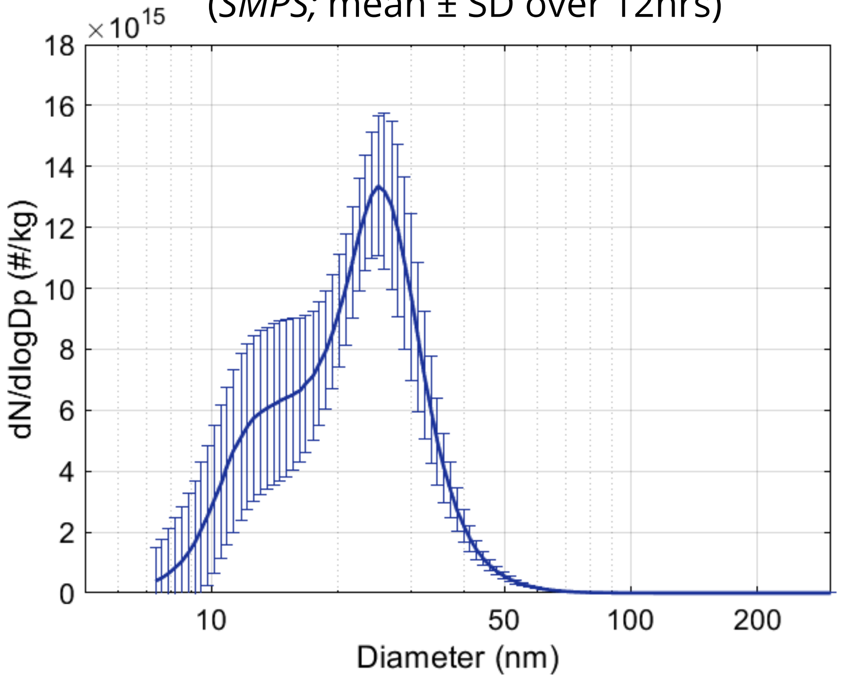
Online measurement of

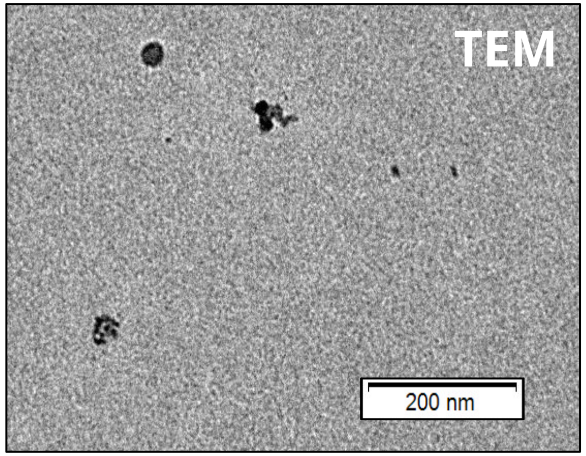
- reactive oxygen species
- oxidative potential

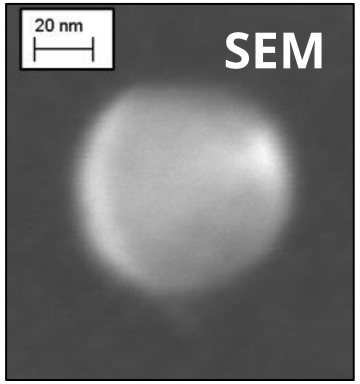
Fresh exhaust: bimodal distribution of mainly spherical, organic particles



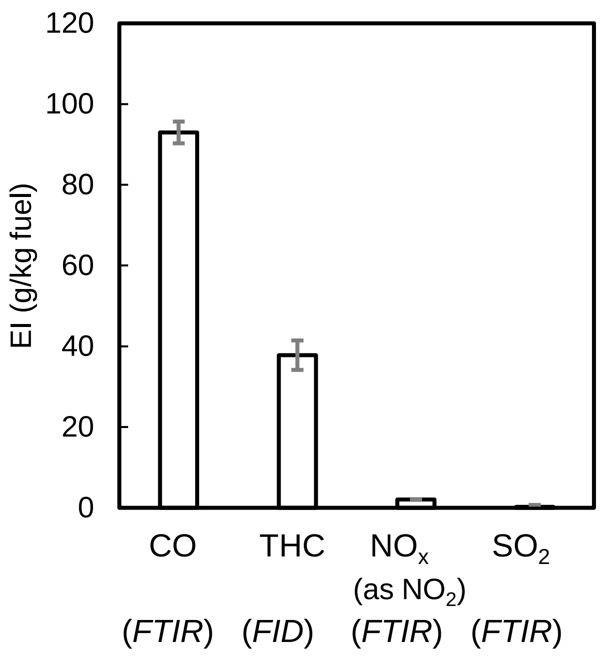
Particle number distribution
(*SMPS;* mean ± SD over 12hrs)



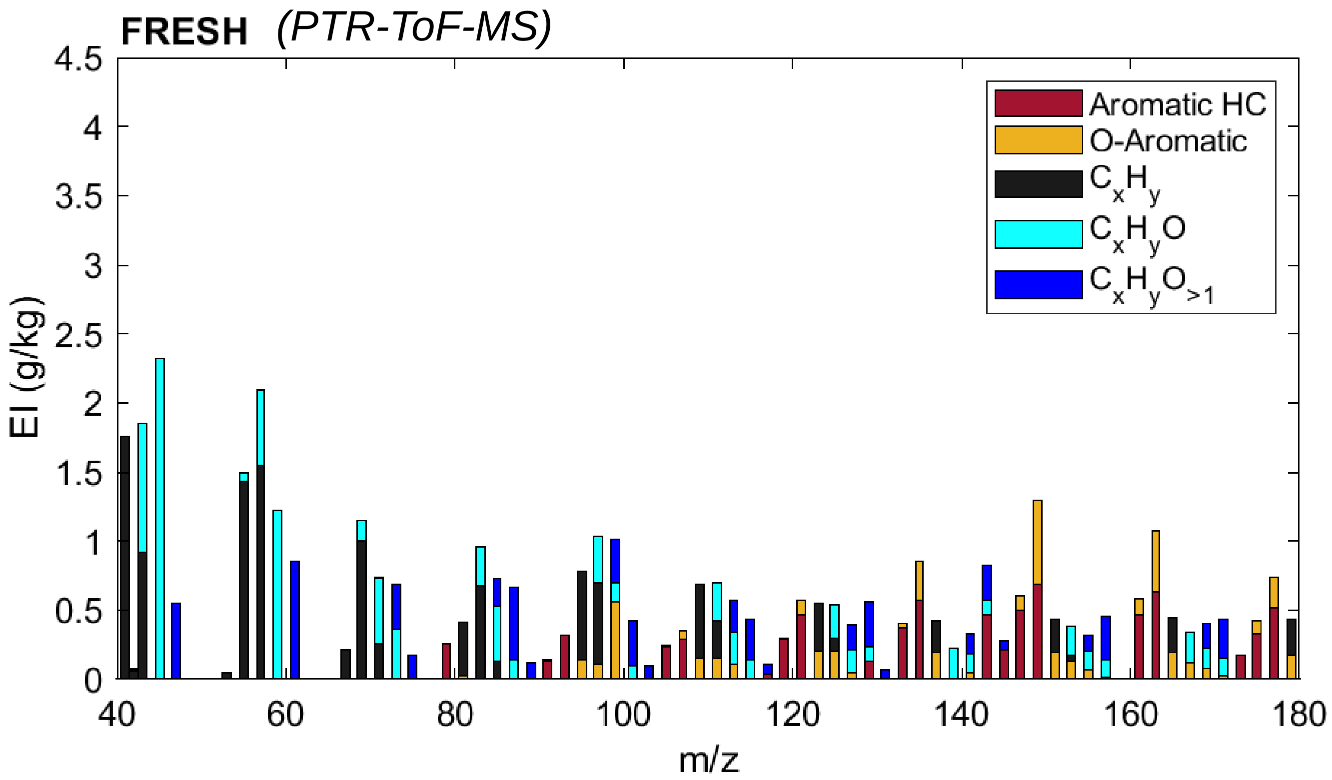




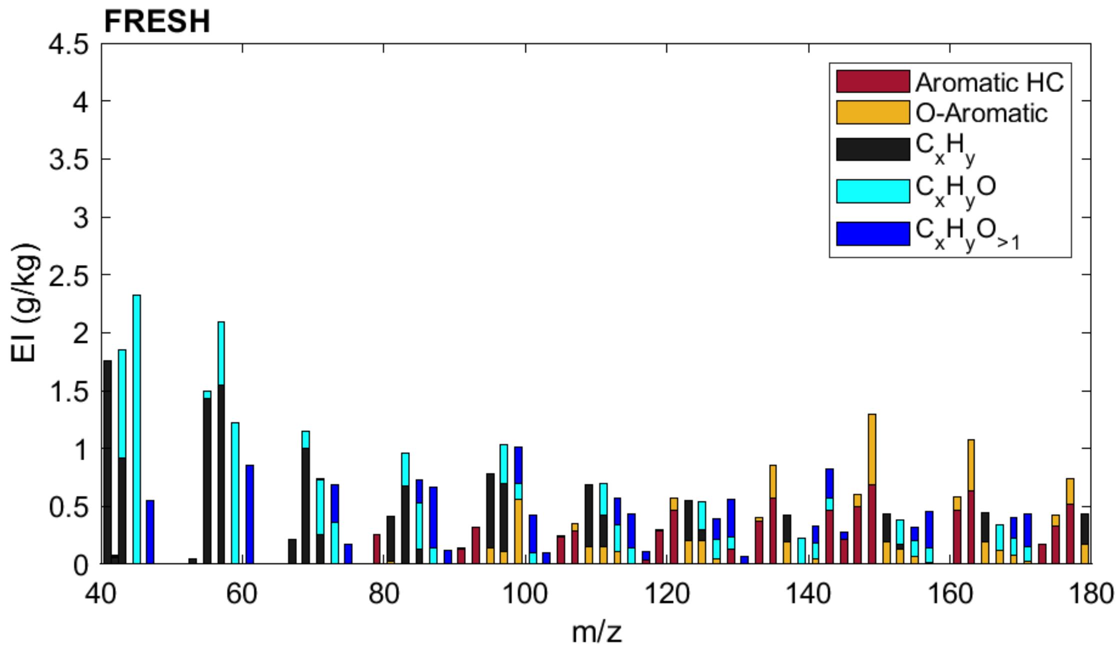
Fresh exhaust gases: CO2, CO, and organic gaseous species

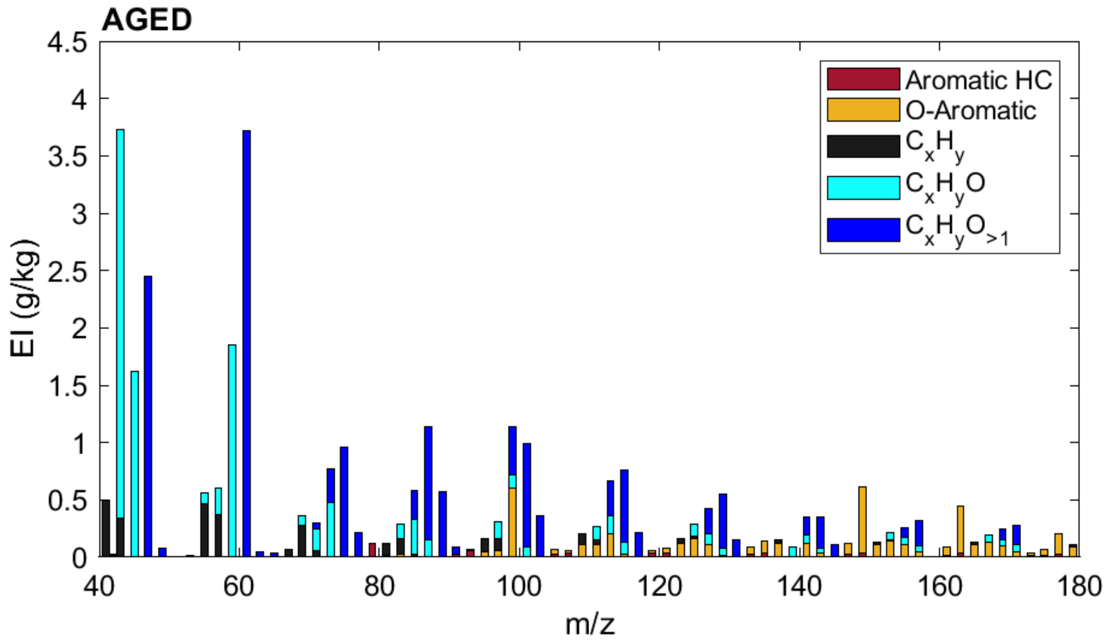


(mean ± SD over 12hrs)



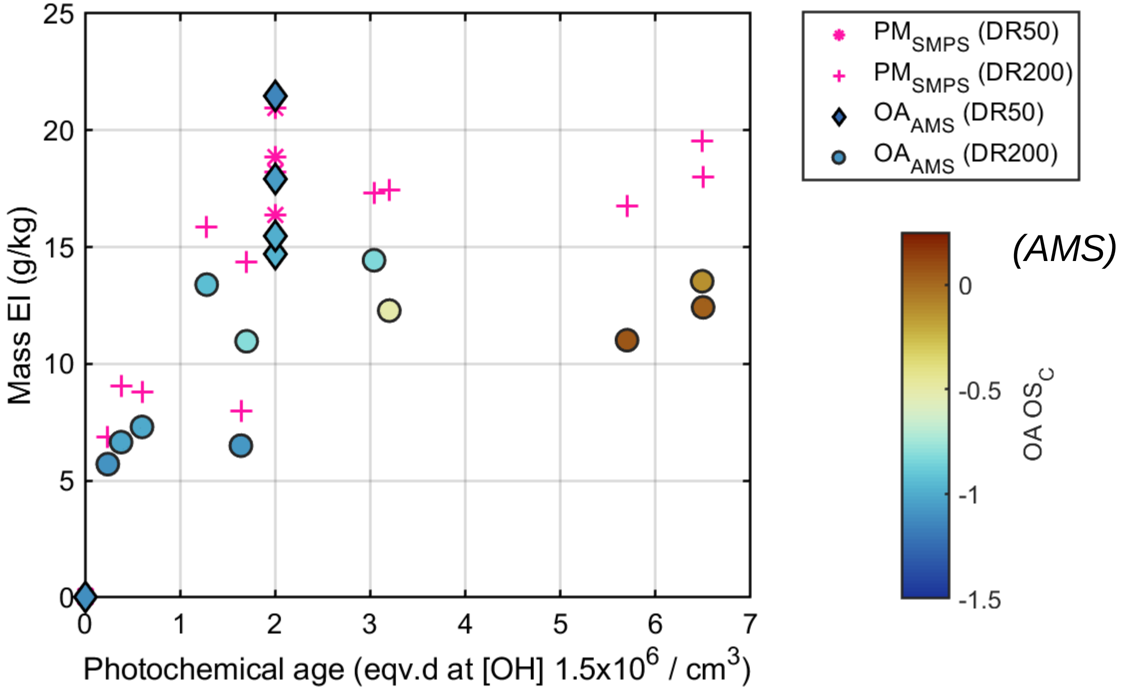
Organic gaseous pollutants altered by photochemical processing

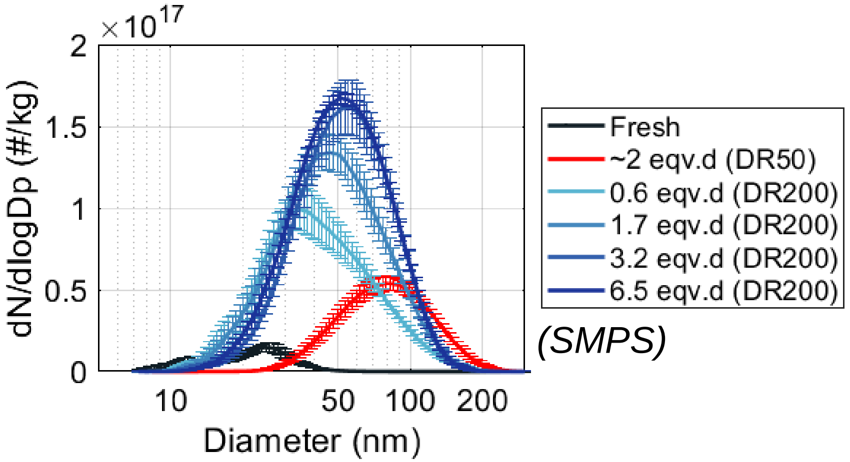




(*PTR-ToF-MS;* average over 12h (fresh) or 16h (aged))

Mass enhancement by a factor of ~300, organic aerosol becomes increasingly oxidized





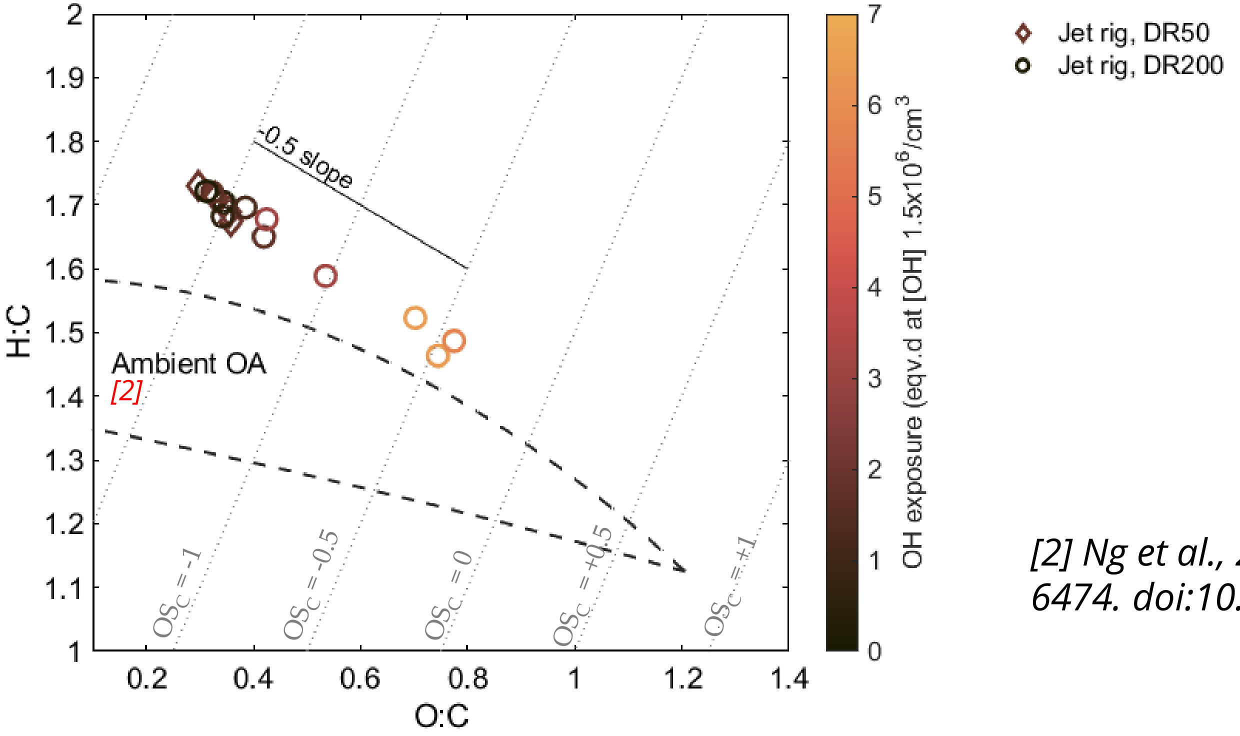
Dilution by 1:50:
GMD ~ 80 nm
N ~ 2e16 #/kg

Dilution by 1:200:
GMD ~ 50 nm
N ~ 6e16 #/kg

Average carbon oxidation state

$OS_C = 2 \times O:C - H:C$

Organic aerosol composition development typical for NO_x –limited conditions



(AMS)

[2] Ng et al., 2011. Atmos. Chem. Phys., 11, 6465–6474. doi:10.5194/acp-11-6465-2011

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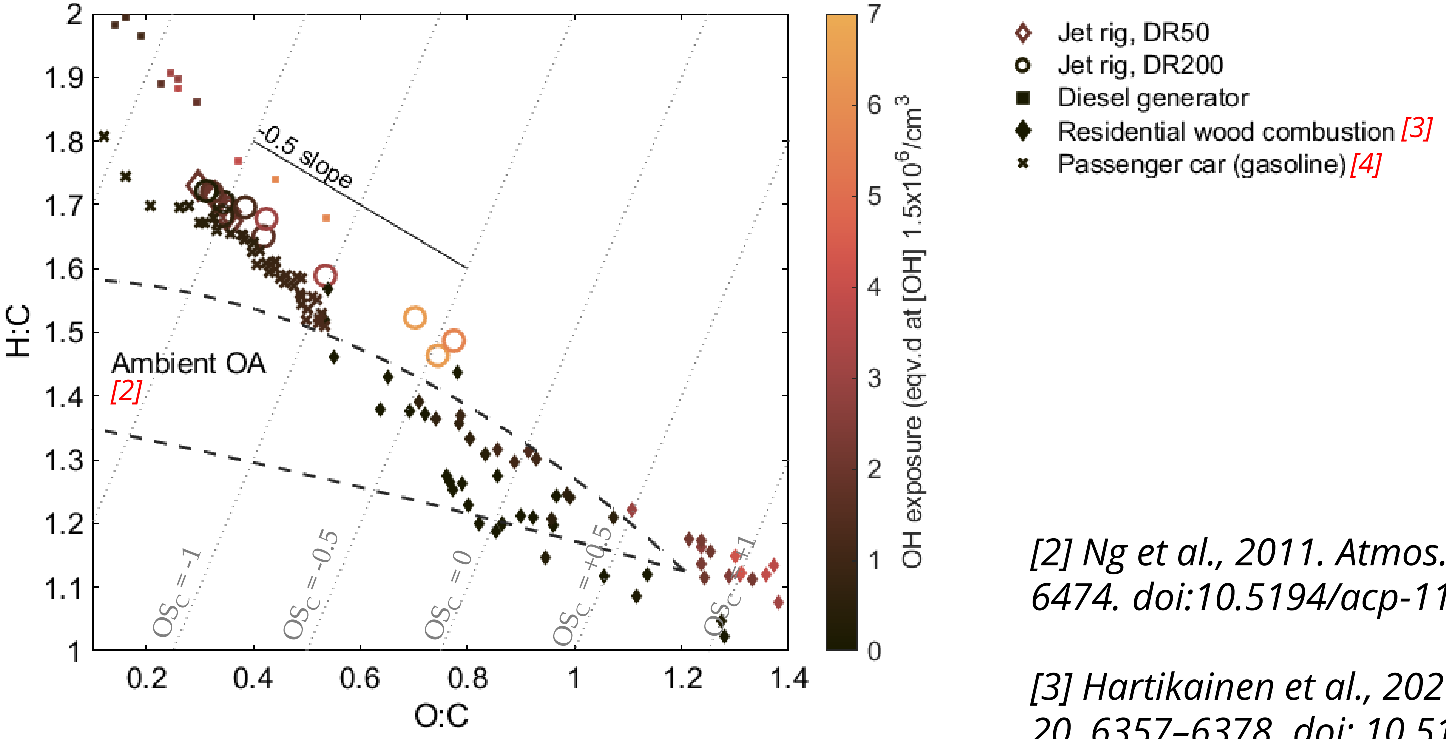


(AMS)

[2] Ng et al., 2011. Atmos. Chem. Phys., 11, 6465–6474. doi:10.5194/acp-11-6465-2011

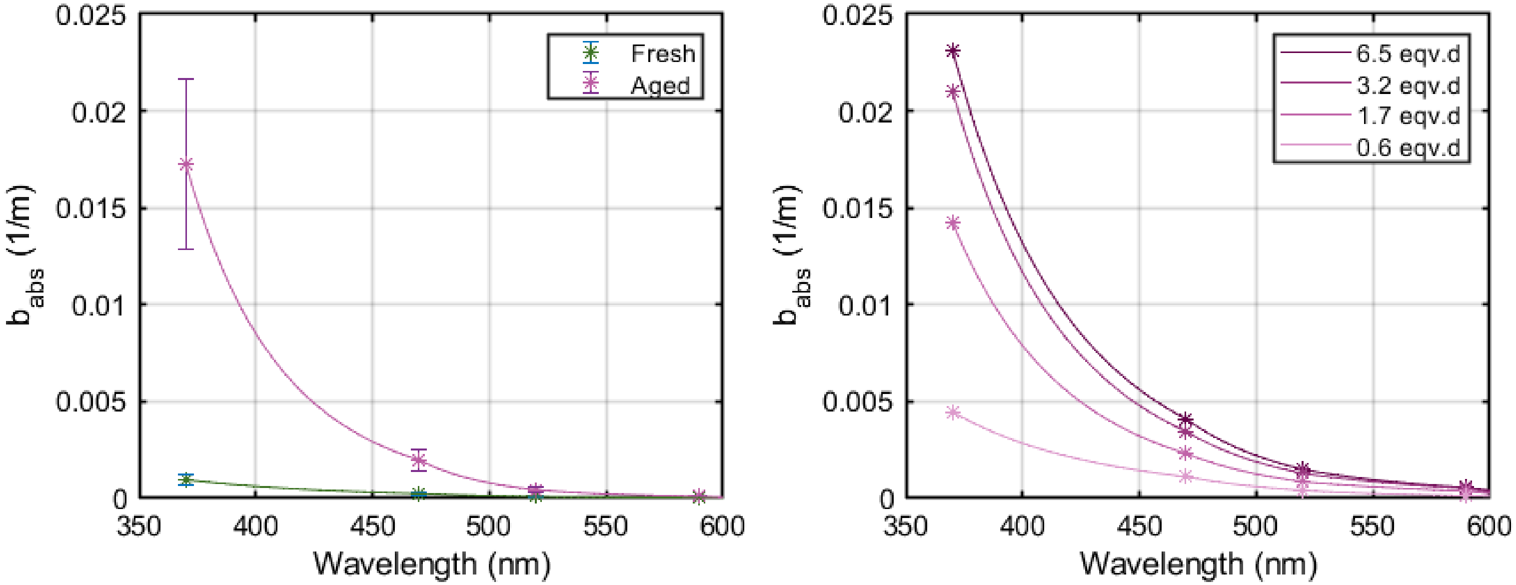
[3] Hartikainen et al., 2020. Atmos. Chem. Phys., 20, 6357–6378. doi: 10.5194/acp-20-6357-2020

[4] Hartikainen et al., 2023. J. Aerosol Sci., 171, 106159. doi: 10.1016/j.jaerosci.2023.106159

Aging increased light absorption by the exhaust

Absorption coefficients in raw exhaust conditions, measured by the *aethalometer*

- for ’brown carbon’ only (=attenuation fit to 660-950 nm subtracted)

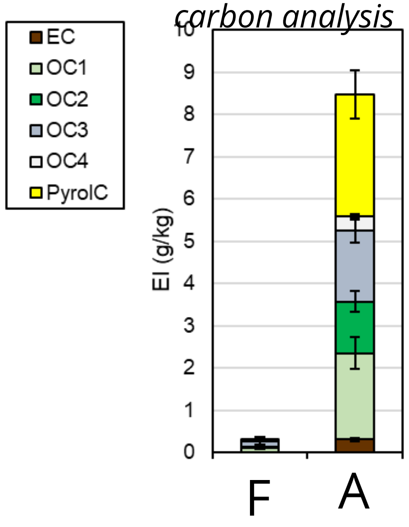


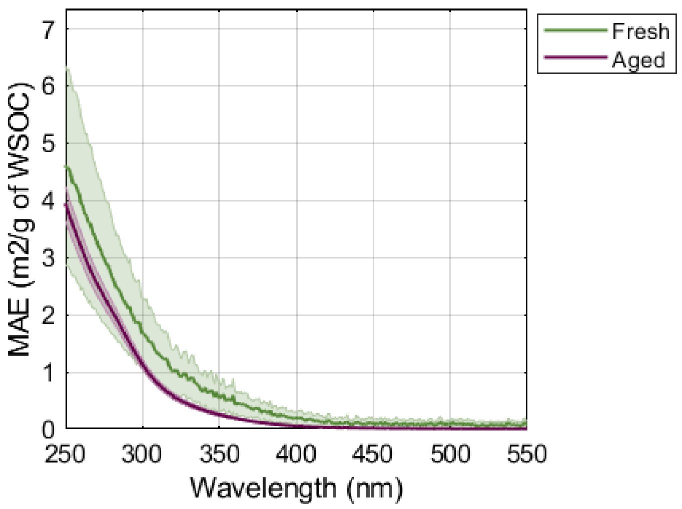
mean ± SD of means of 4 h experiments (n=4)

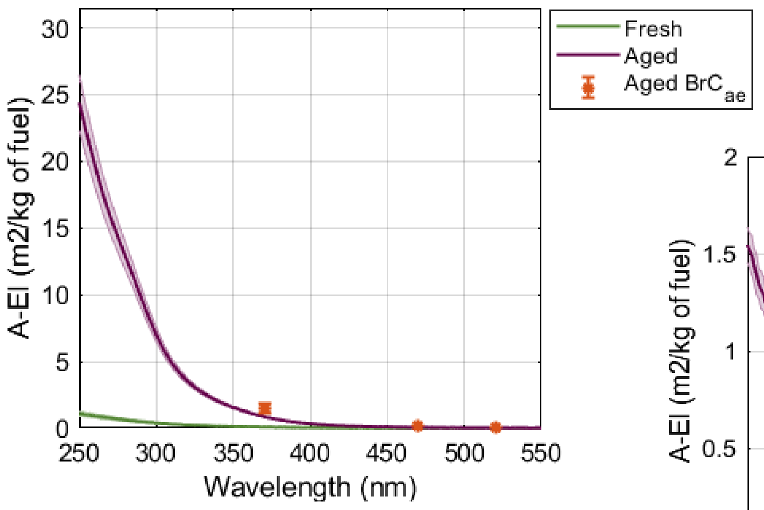
Absorption by water soluble organic carbon (WSOC)

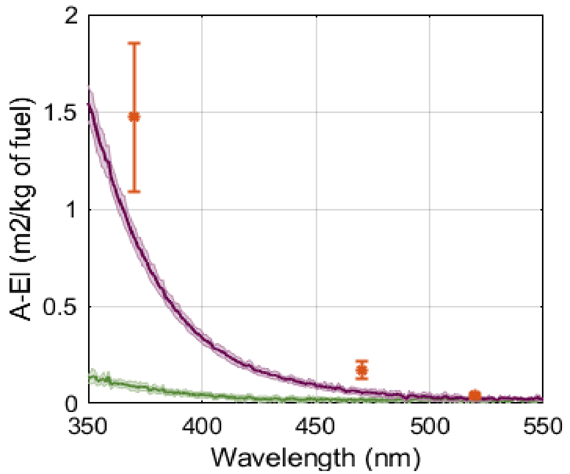
- Mass absorption efficiencies (MAE) slightly higher for fresh WSOC
- Total absorption by combustion of 1 kg of fuel (A-EI) increases in line with the OC enhancement

Thermal-optical carbon analysis



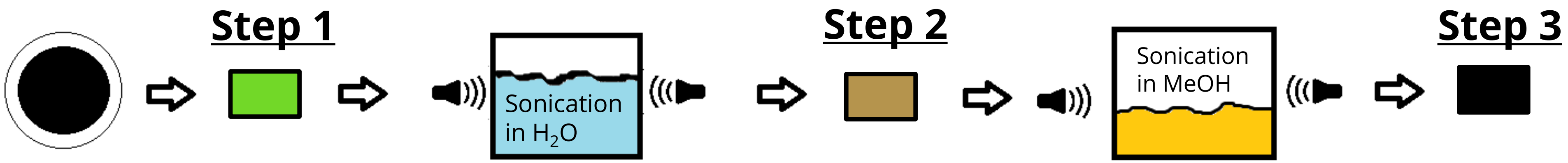


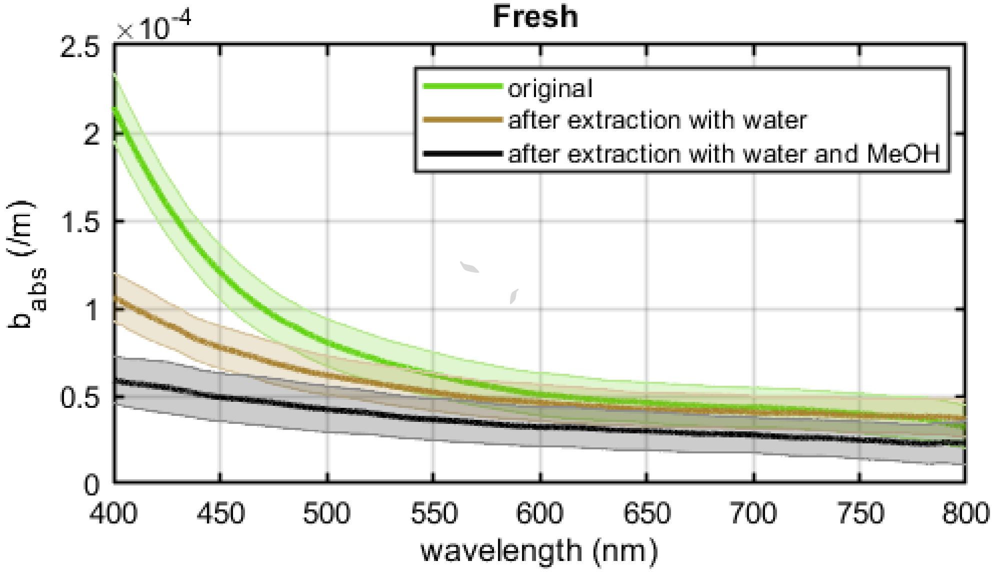


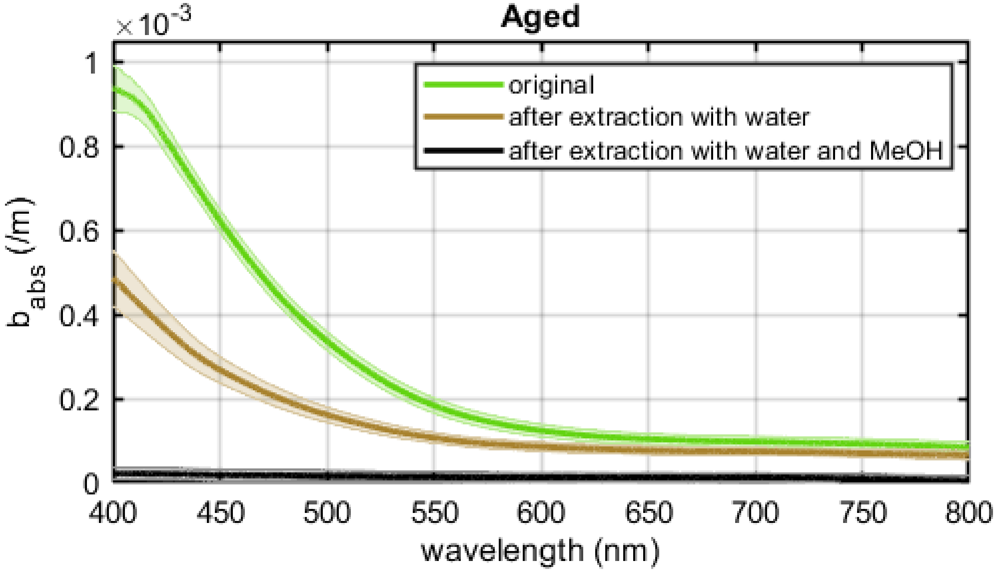


(*UV-VIS Spectrometer*, mean ± SD, n=4 for fresh, 3 for aged exhausts)

Absorbance also by water insoluble organic carbon

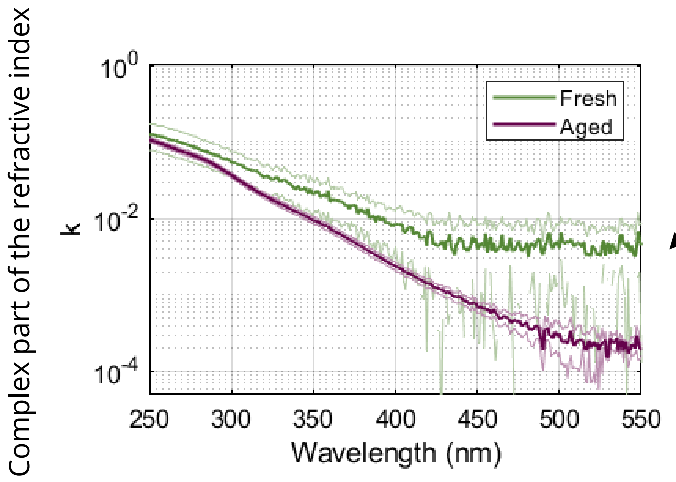






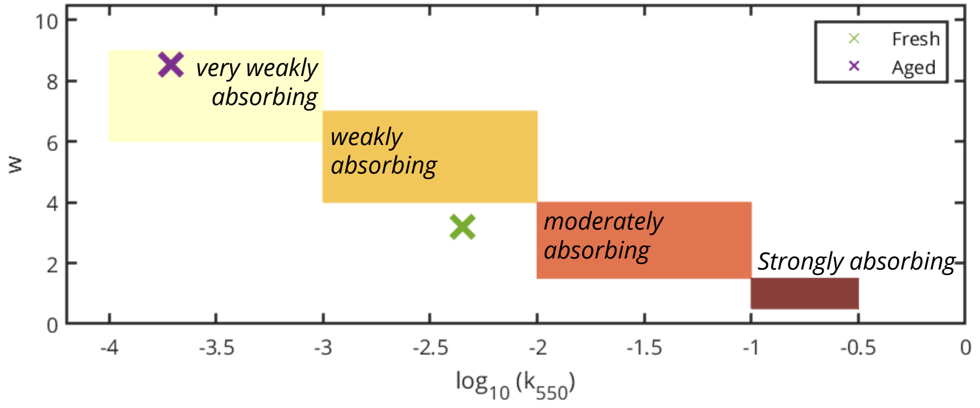
(*UV-VIS Spectrometer*, mean ± SD, n=4 for fresh, 3 for aged exhausts)

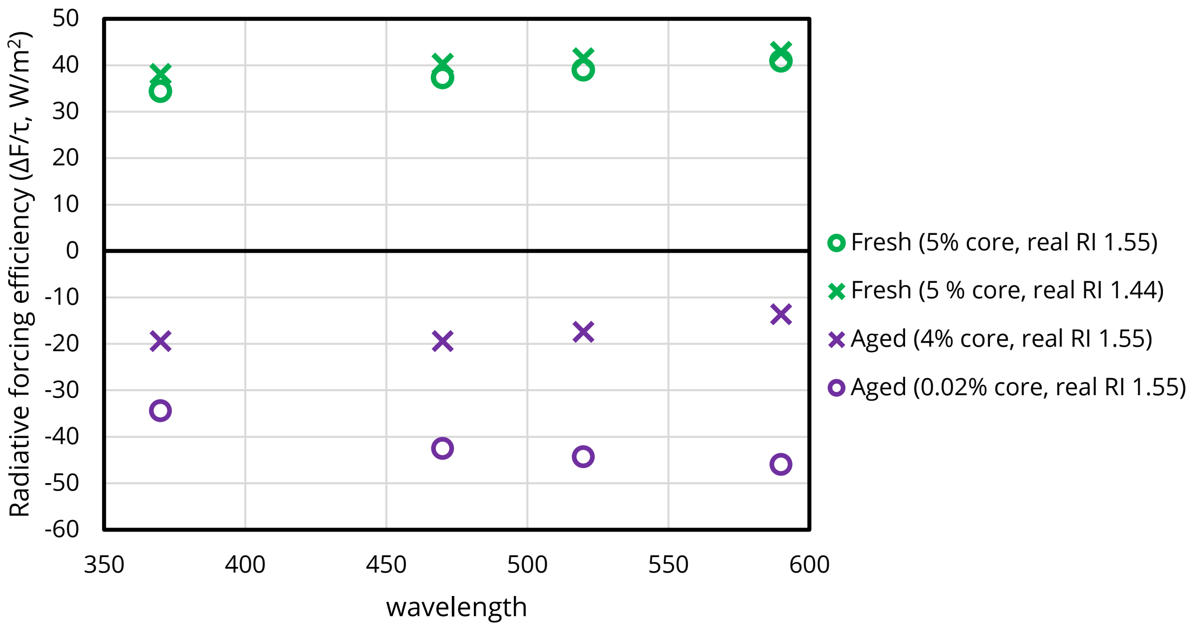
Fresh exhaust has warming, aged exhaust cooling impact on climate



Weakly (fresh) or very weakly (aged) absorbing brown carbon [5]

$$k_\lambda = k_{550}\left(\frac{550nm}{\lambda}\right)^w$$





[5] Saleh, R. 2020. Curr Pollution Rep., 6, 90–104. DOI: 10.1007/s40726-020-00139-3

Conclusions

- Photochemical aging led to notable formation of secondary organic aerosol: **×300** increase in particulate mass
  - ➢ should be considered when assessing health impacts of aviation
- Continuous transformation of particle chemical composition, similarly to other organic aerosol sources
- Formation of weakly absorbing organic aerosol
  - ➢ shift from warming to cooling climate impact upon photochemical aging

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Thank you for your attention!

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ULtrafine particles from TRansportation - Health Assessment of Sources

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