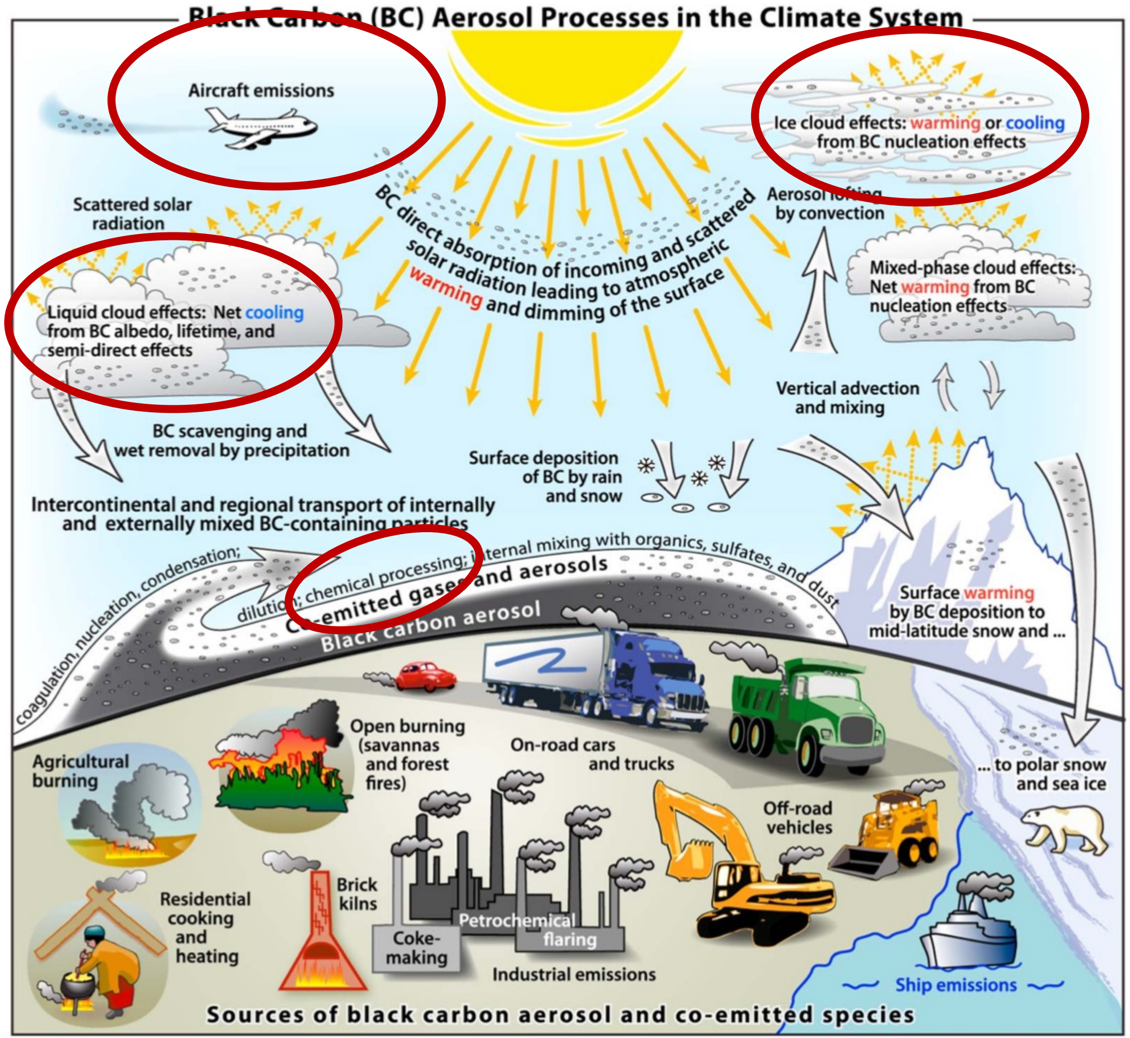
Future warming exacerbated by aged-soot effect on cloud formation

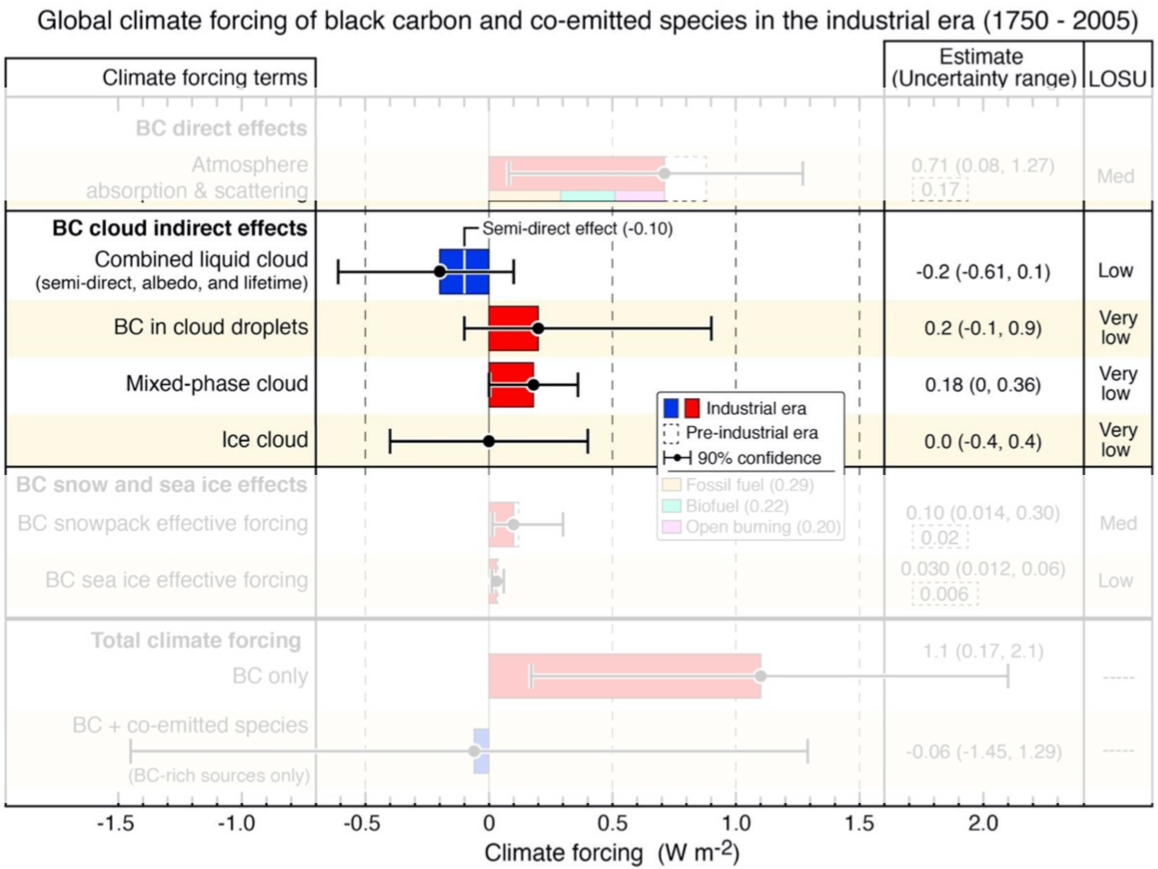
U. Lohmann, F. Friebel, Z. A. Kanji, F. Mahrt, A. A. Mensah and D. Neubauer

ETH zürich



Bond et al., 2013

Climate forcing of soot (1750-2005)



… uncertainties in net climate forcing from black-carbon-rich sources are substantial, largely due to a **lack of knowledge about** cloud interactions **with both** black carbon **and co-emitted organic carbon.**

Bond et al. (2013), JGR

Questions to be addressed in this talk

- Can soot particles act as cloud condensation nuclei (CCN) at atmospheric conditions and what is the impact on the present-day climate?
- Can soot particles act as ice nucleating particles (INPs) at atmospheric conditions?
- What is the impact of aged-soot via these mechanisms on cloud formation in a future climate?

Mechanisms of soot aging considered in this study

(Friebel et al. ACP, 2019)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Air


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Ozone

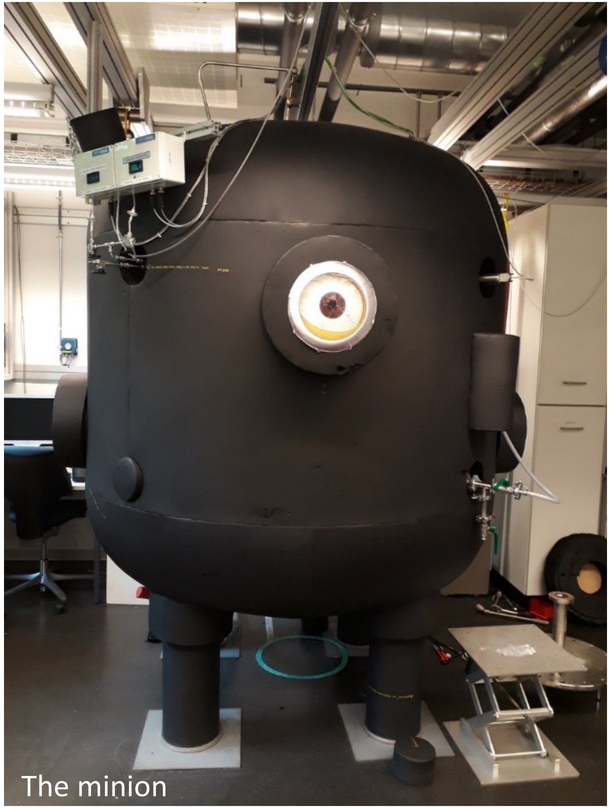
Continuous-flow Stirred Tank Reactor (CSTR)

- 100 nm soot particles
- 16 h aging time
- miniCAST brown (organic carbon rich soot)

Temperature
5 - 35 °C

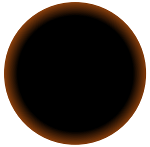
↔

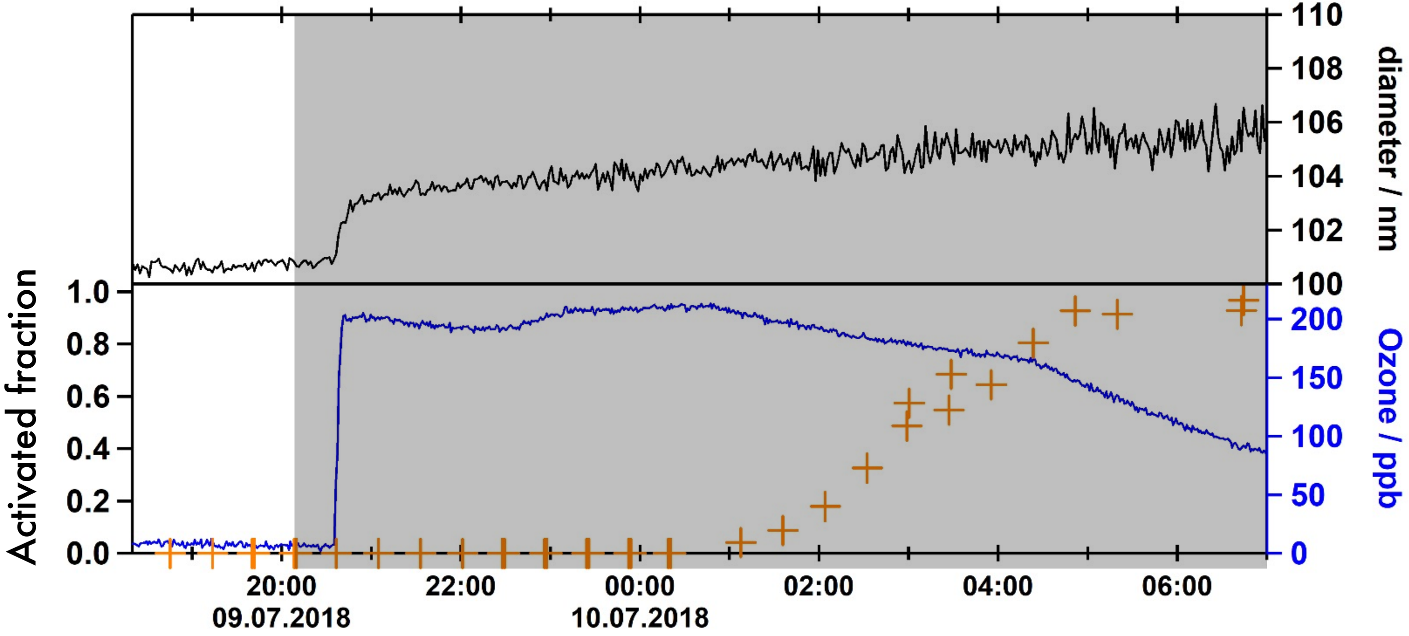
Ozone
0 - 200 ppb



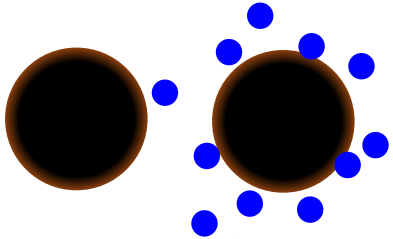
Friebel and Mensah, AMT, 2019

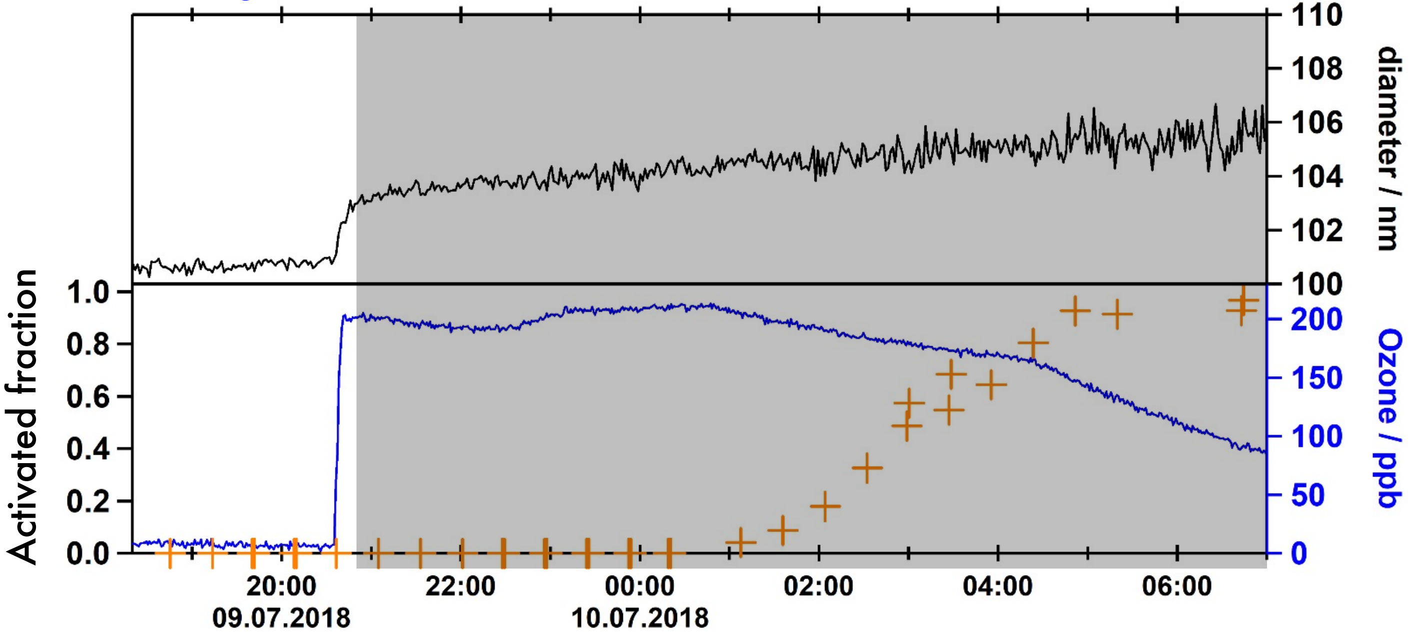
Ozone oxidation of 100 nm organic-rich soot



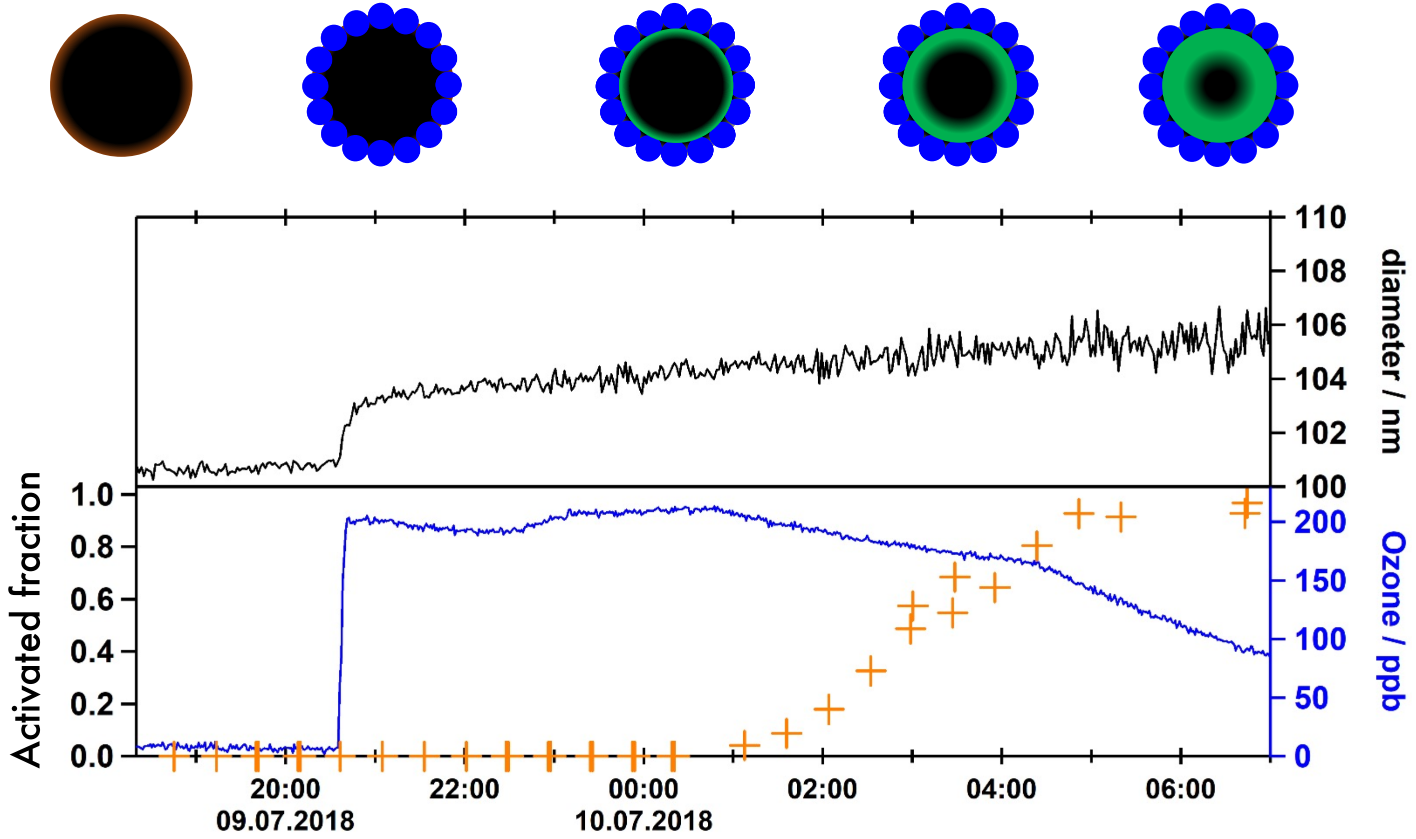


Adding 200 ppb ozone → adsorption

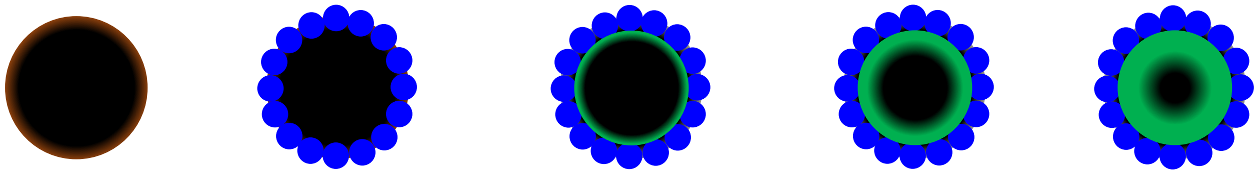




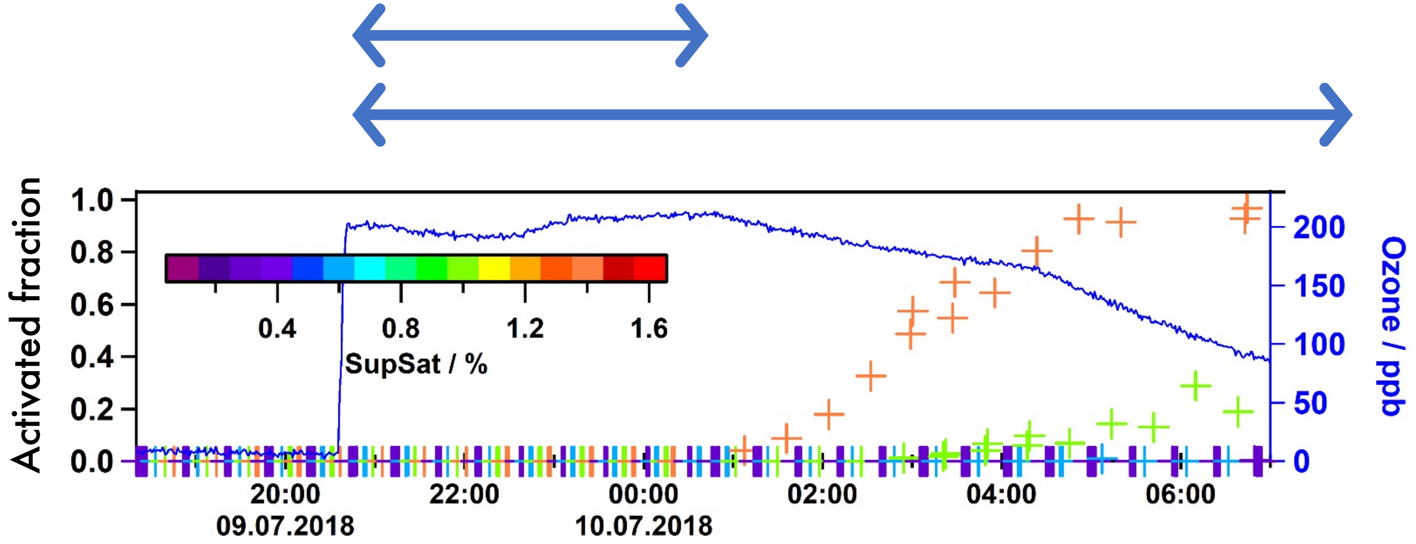
Continuous exposure to ozone → increase in CCN activity



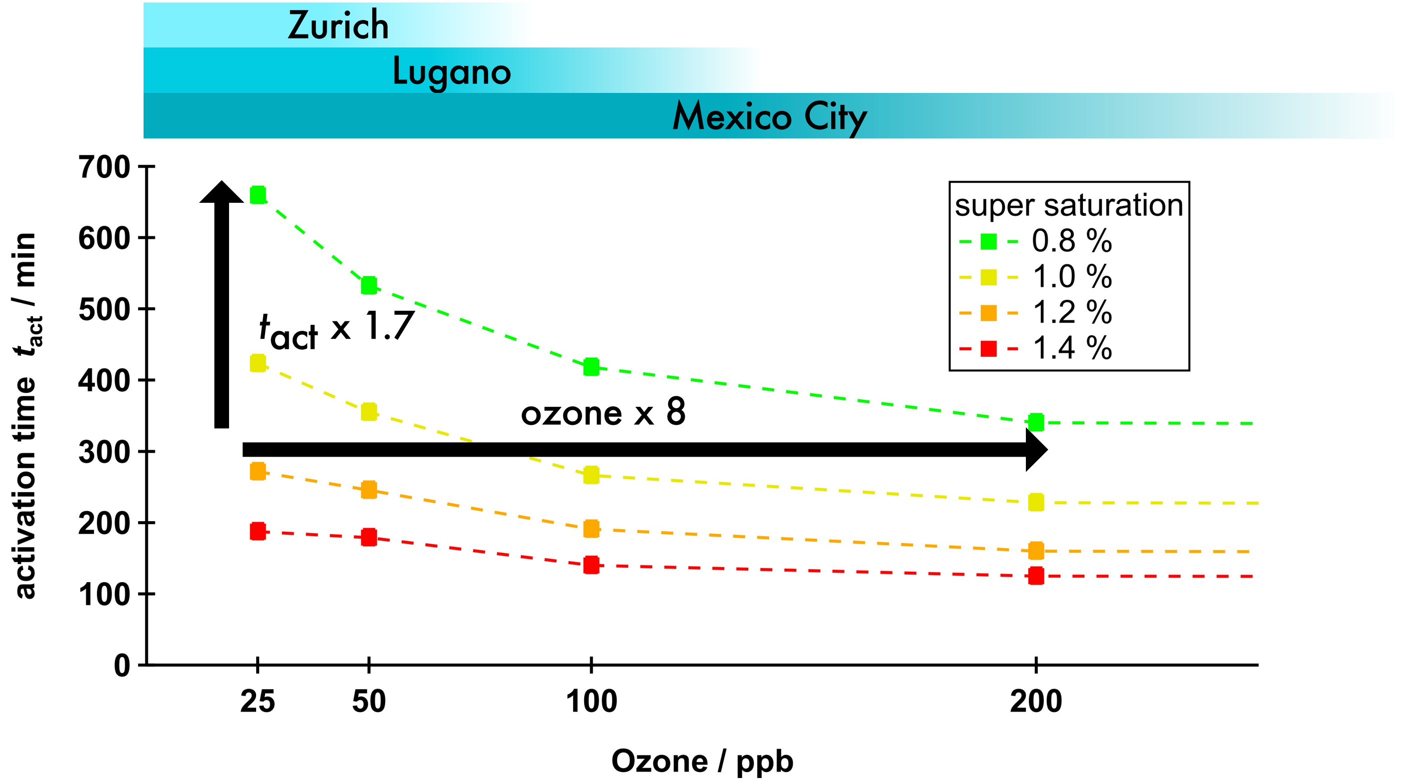
Activation time t_{act}: time until the particle is CCN-active



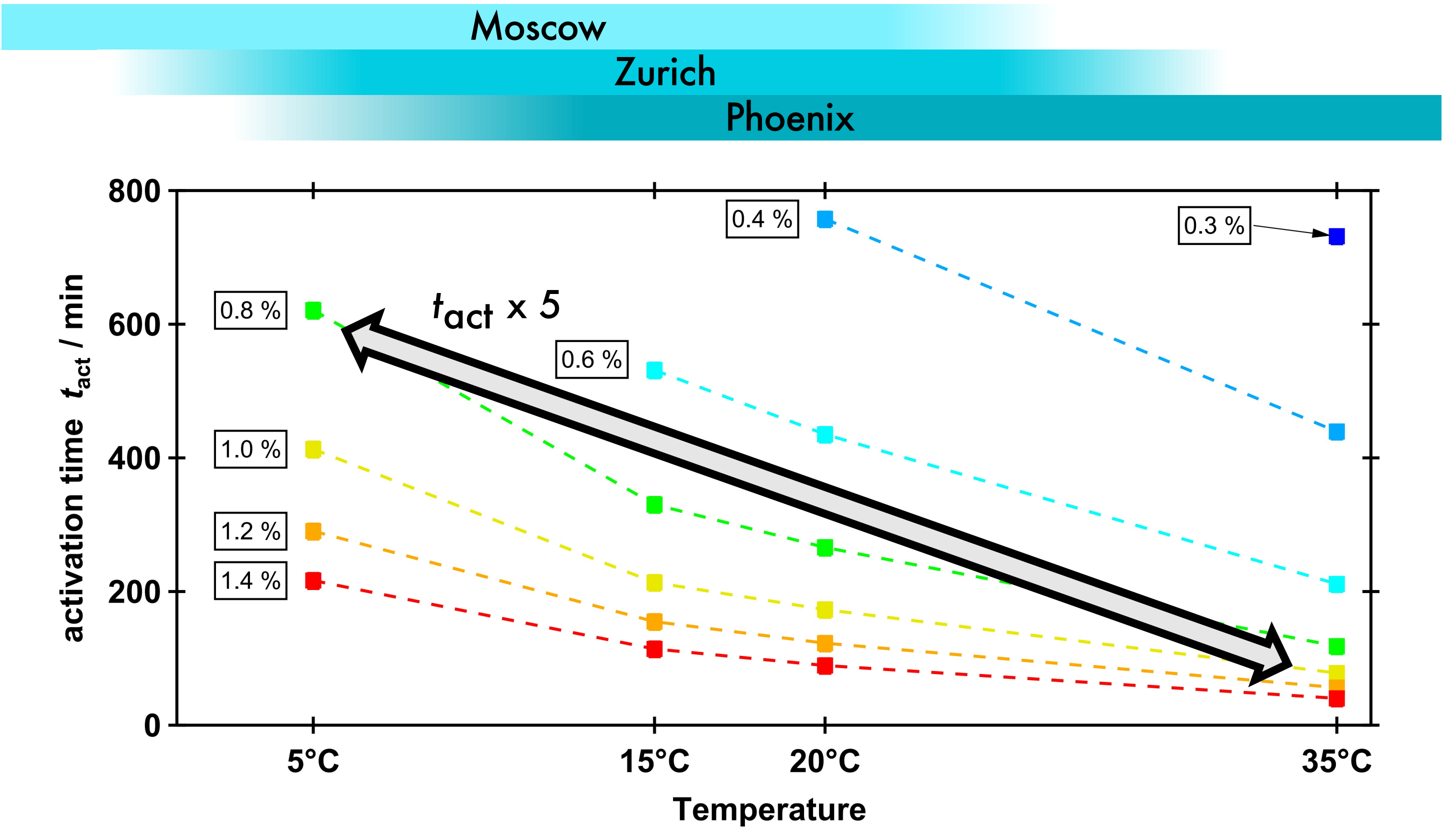
t_{act} = 3 - 12 h



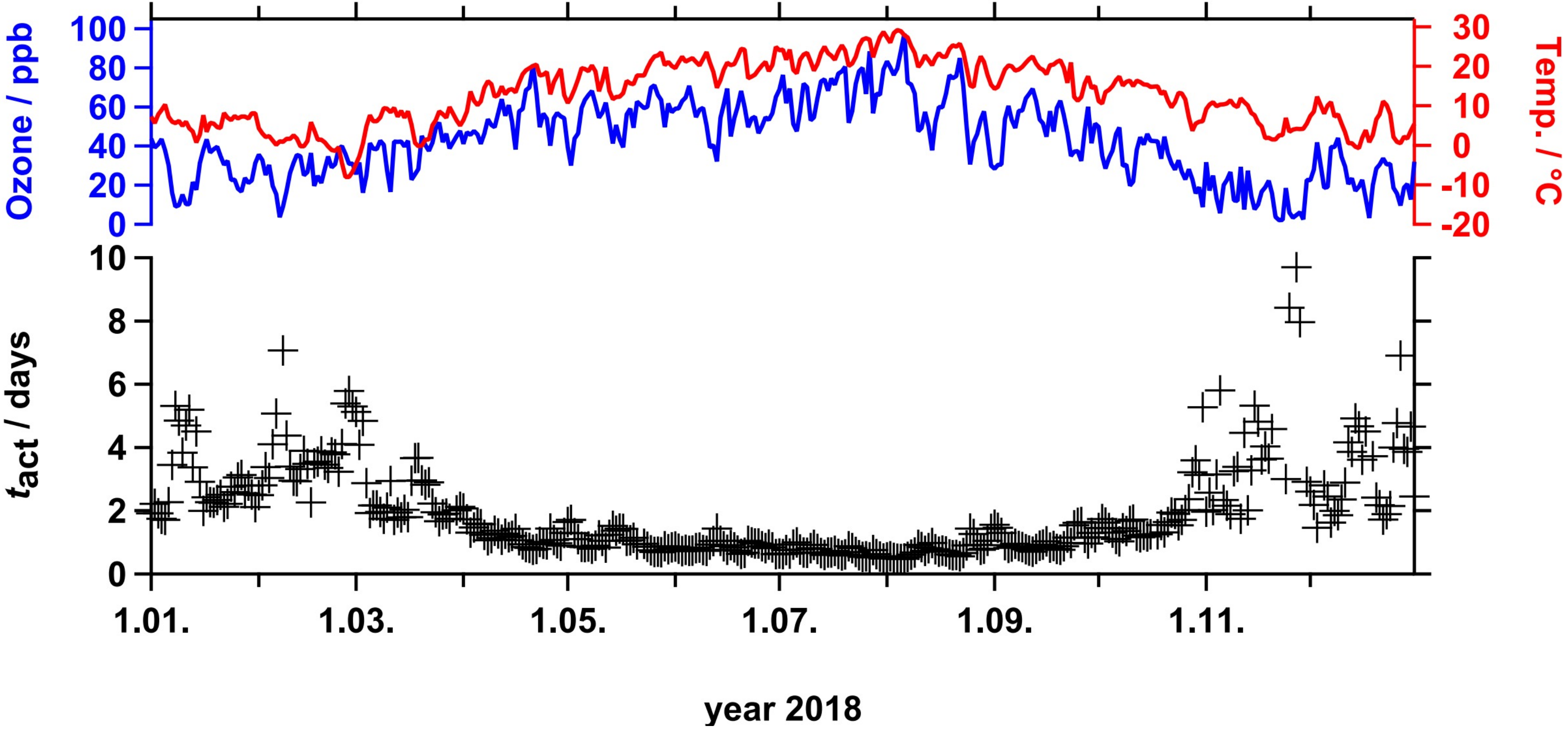
Activation time vs. ozone concentration at T=25 °C



Activation time vs. temperature at 200 ppb O_3

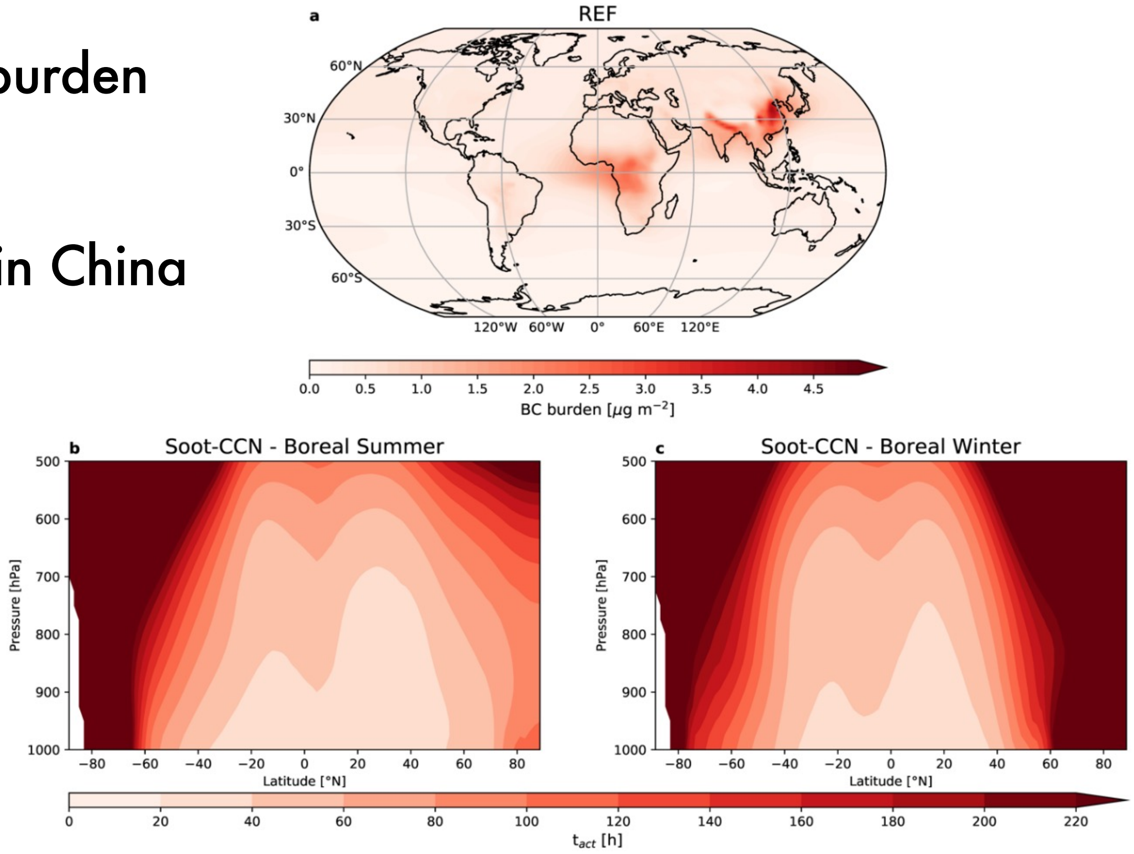


Activation times of soot in Zürich

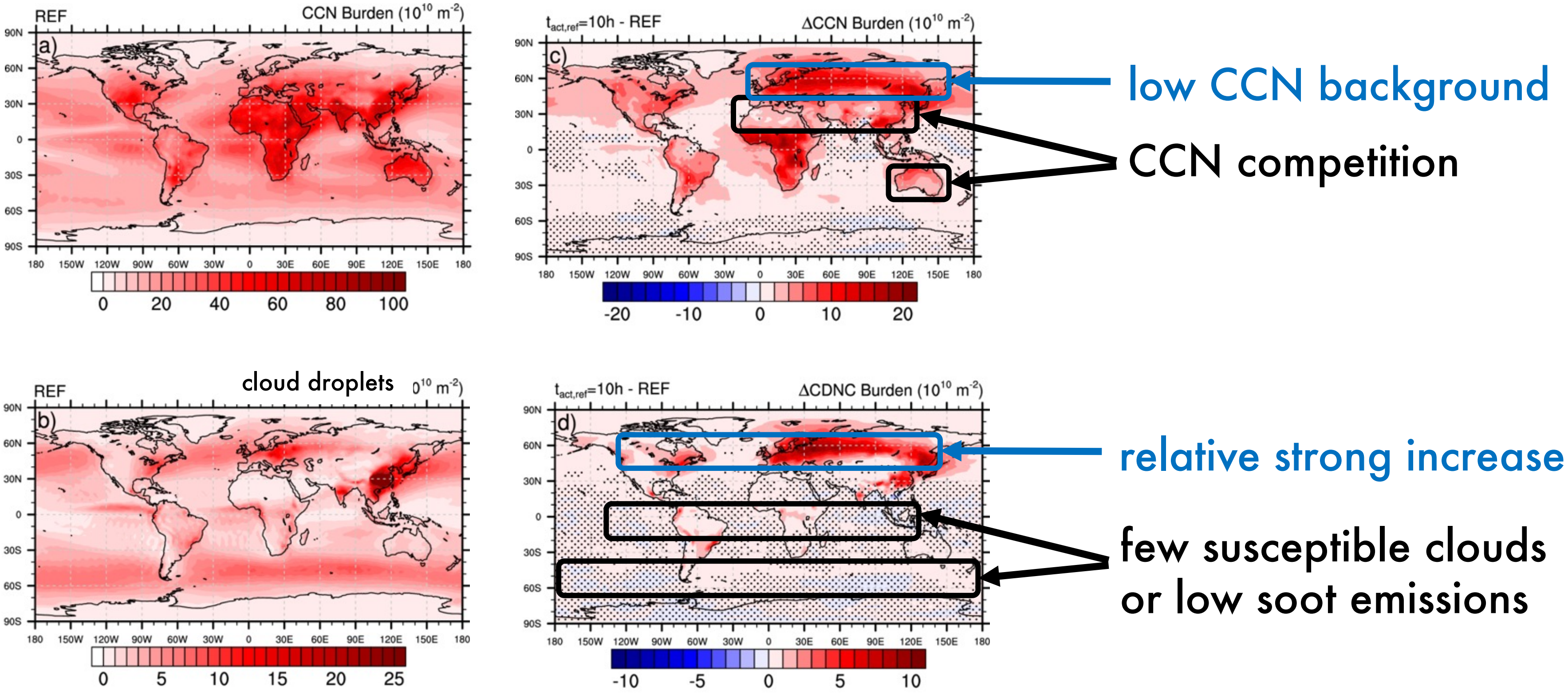


Annual mean black carbon burden and aging times

- High black carbon burden in China
- Short aging times in summer and winter in China



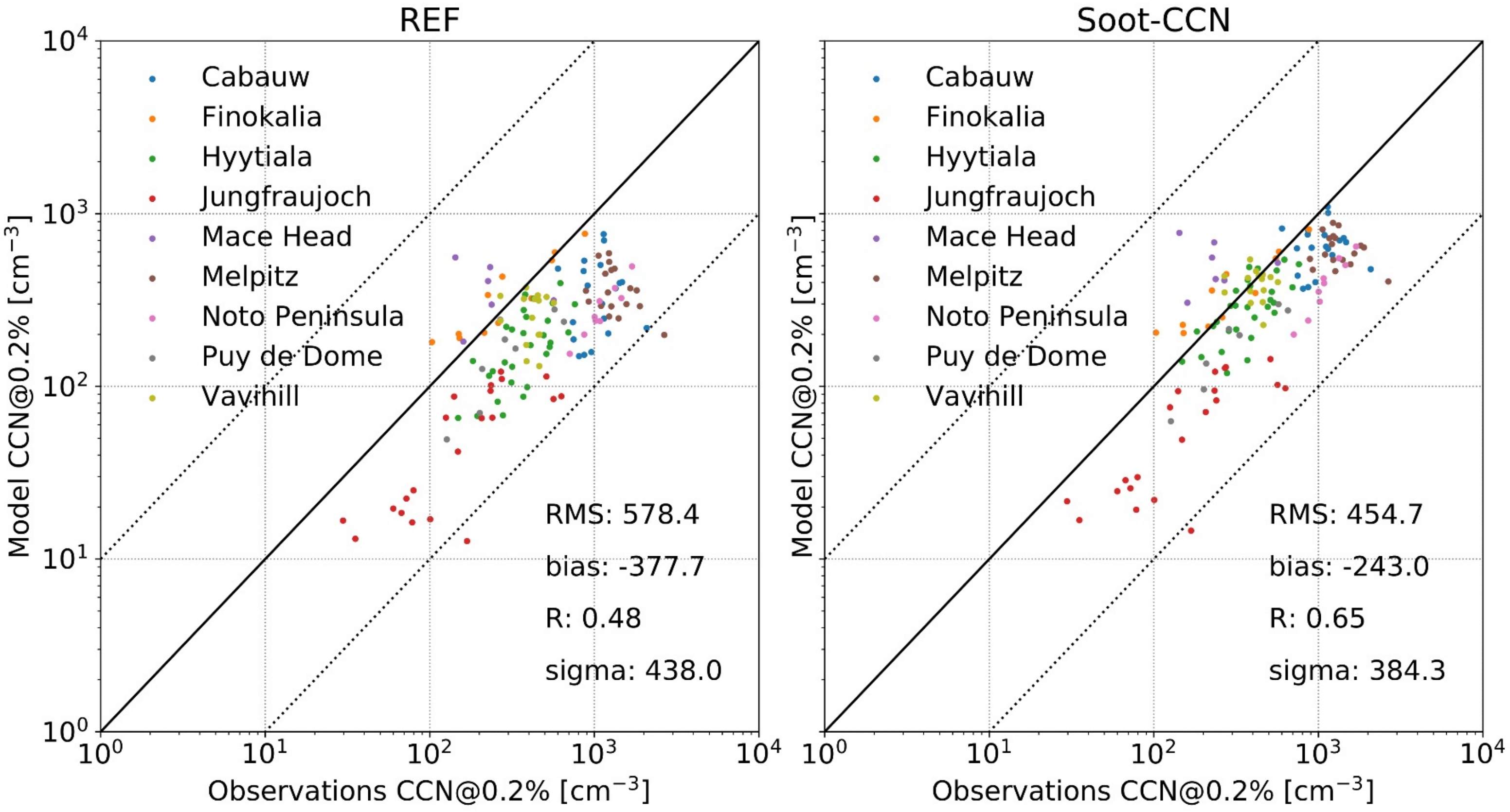
Climate impact of ozone-aged soot



→ 93% increase in cloud droplet burden north of 60 °N (t_{act} = 10h)

Friebel et al., ACP, 2019

Model validation



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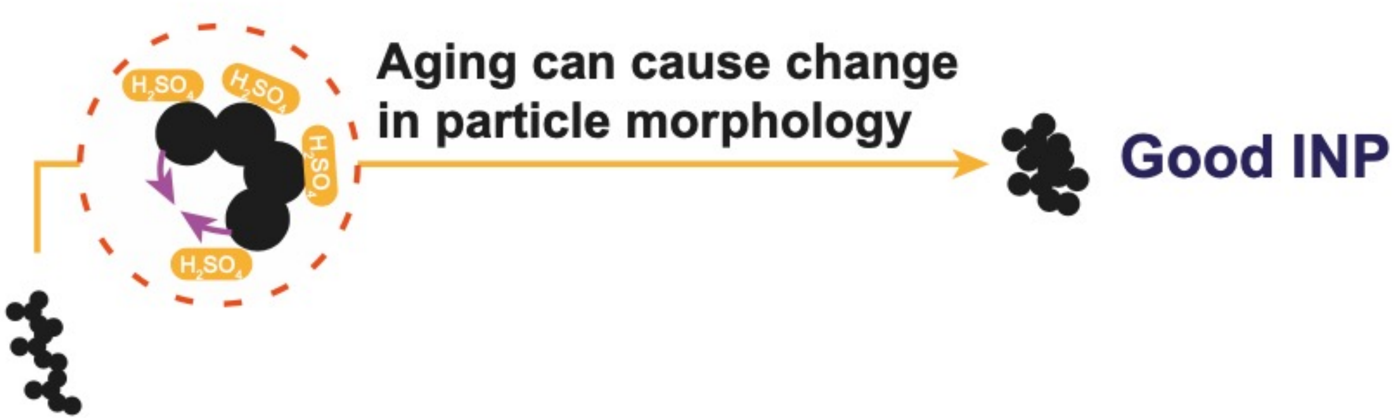
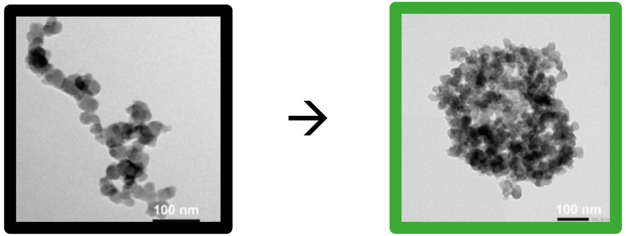
Mechanisms of soot aging considered in this study



(Mahrt et al. JGR/ESPI, 2020)

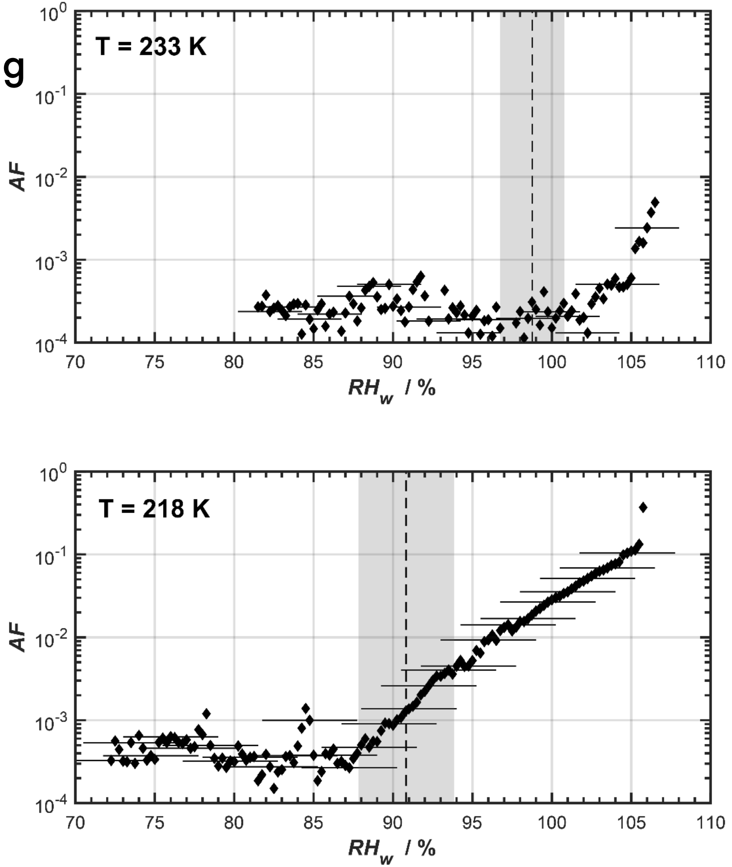


Ice nucleation activity of fresh and aged soot

d_m = 400 nm, miniCAST soot, Horizontal Ice Nucleation Chamber (HINC)

- - · Homogeneous freezing

♦ Unaged black



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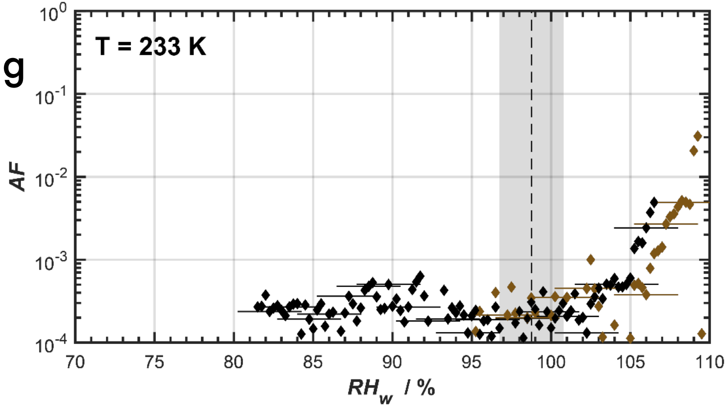
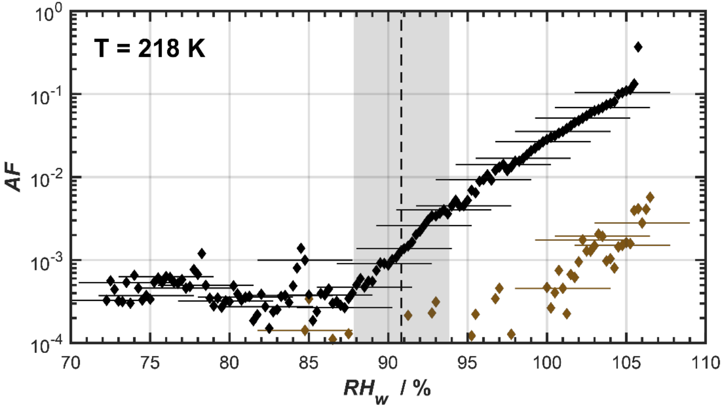


- OC rich soot nucleates ice worse.

◆ Unaged brown



Ice nucleation activity of fresh and aged soot

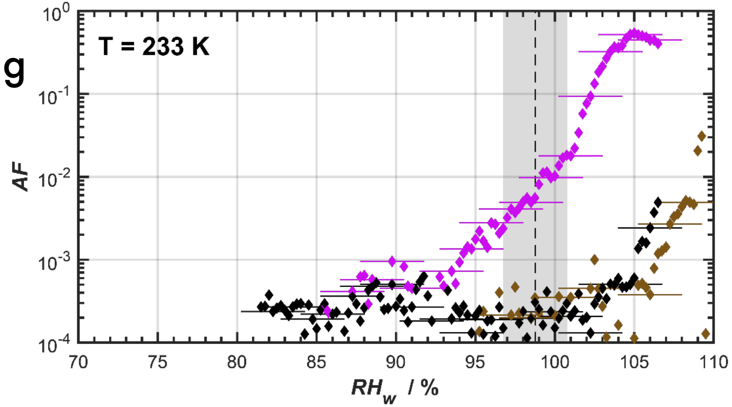
d_m = 400 nm, miniCAST soot, Horizontal Ice Nucleation Chamber (HINC)

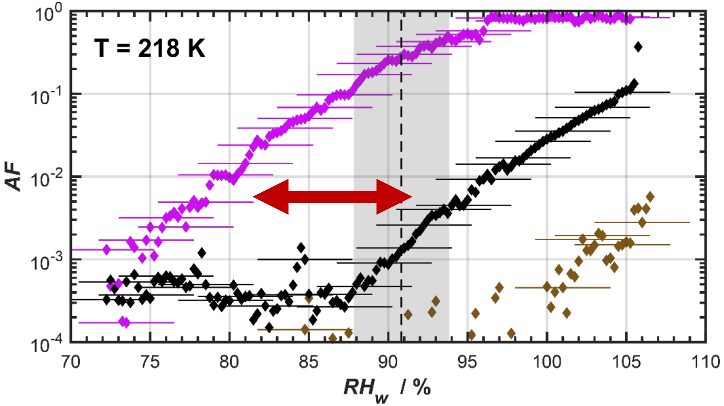
- - Homogeneous freezing

◆ Unaged black

◆ Acid aged black

◆ Unaged brown





- OC rich soot nucleates ice worse.
- Aqueous phase aging enhances ice nucleation ability.

Ice nucleation activity of fresh and aged soot

d_m = 400 nm, miniCAST soot, Horizontal Ice Nucleation Chamber (HINC)

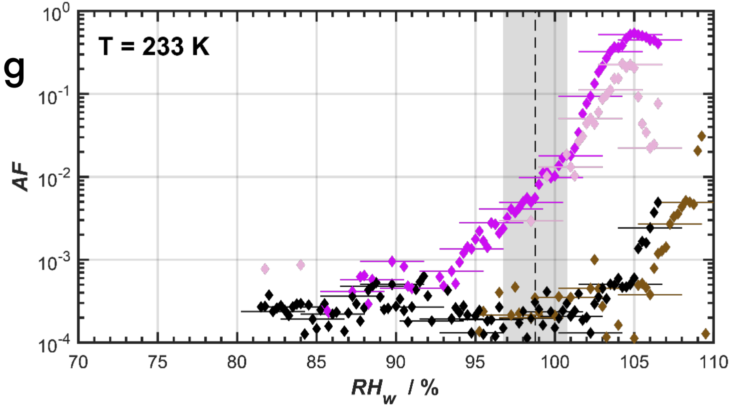
- - - Homogeneous freezing

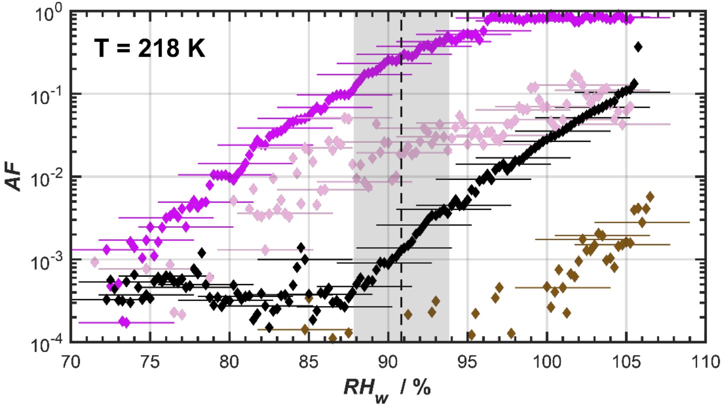
◆ Unaged black

◆ Acid aged black

◆ Unaged brown

◆ Acid aged brown



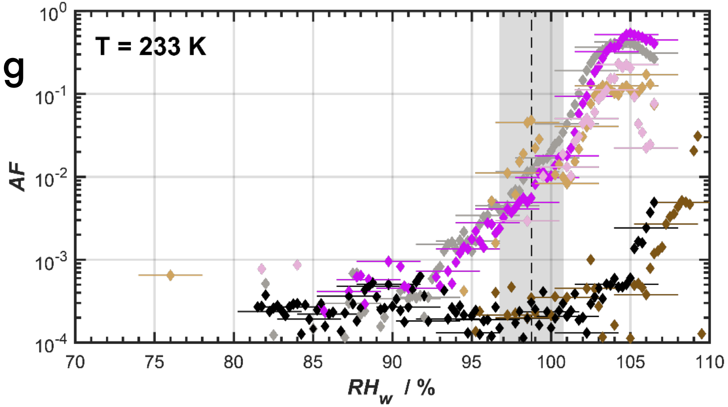


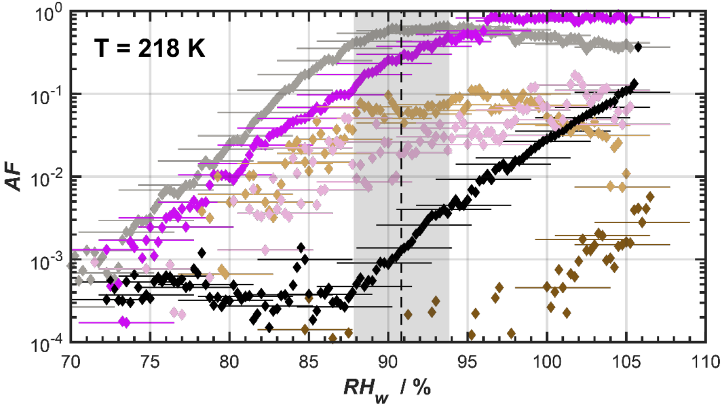
- OC rich soot nucleates ice worse.
- Aqueous phase aging enhances ice nucleation ability.
- Aging renders ice nucleation ability of soots *similar*.

Ice nucleation activity of fresh and aged soot

d_m = 400 nm, miniCAST soot, Horizontal Ice Nucleation Chamber (HINC)

- Homogeneous freezing (dashed line)
- Unaged black
- Acid aged black
- Water aged black
- Unaged brown
- Acid aged brown
- Water aged brown



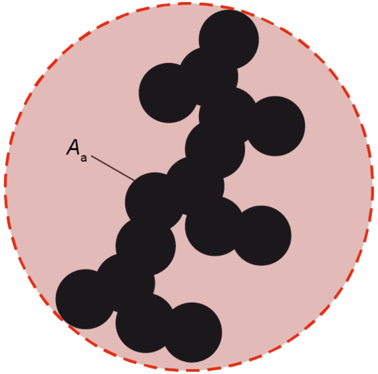


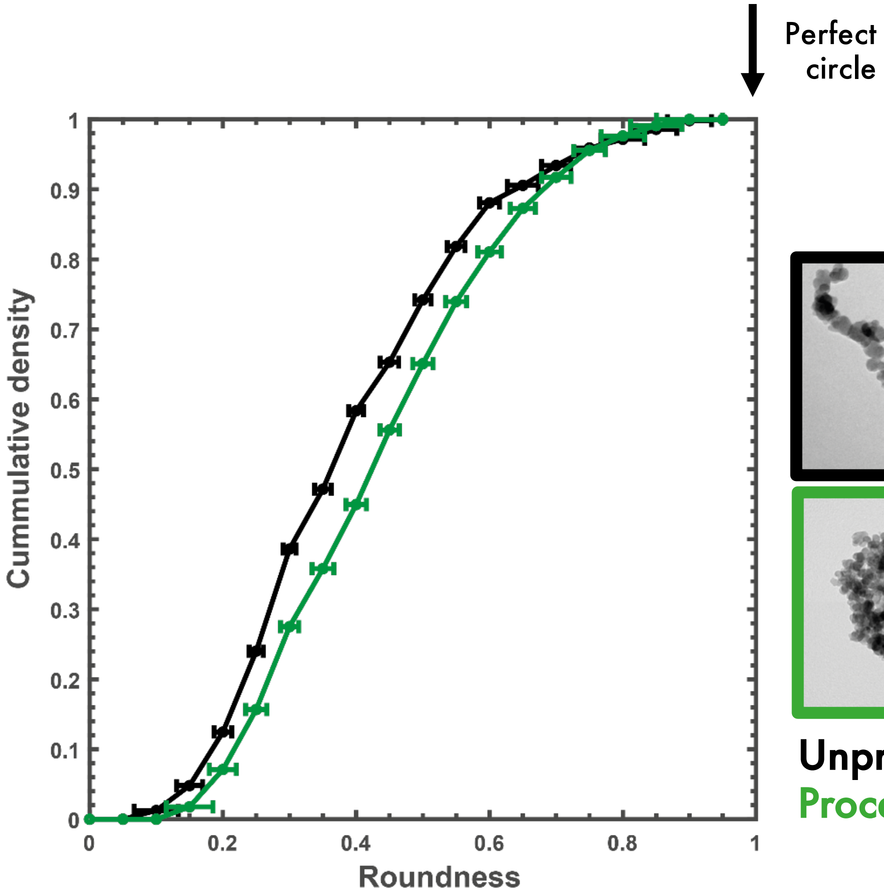
- OC rich soot nucleates ice worse.
- Aqueous phase aging enhances ice nucleation ability.
- Aging renders ice nucleation ability of soots *similar*.
- Ice nucleation ability is independent of aging type.

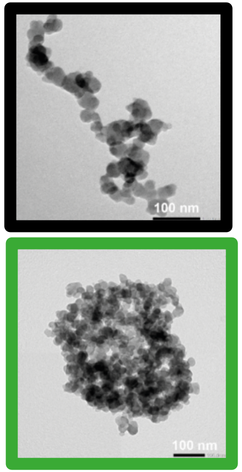
Mahrt et al. 2020

Hydrometeor formation compacts the soot particles

Compacted soot aggregates initiate ice formation via pore condensation & freezing







Unprocessed
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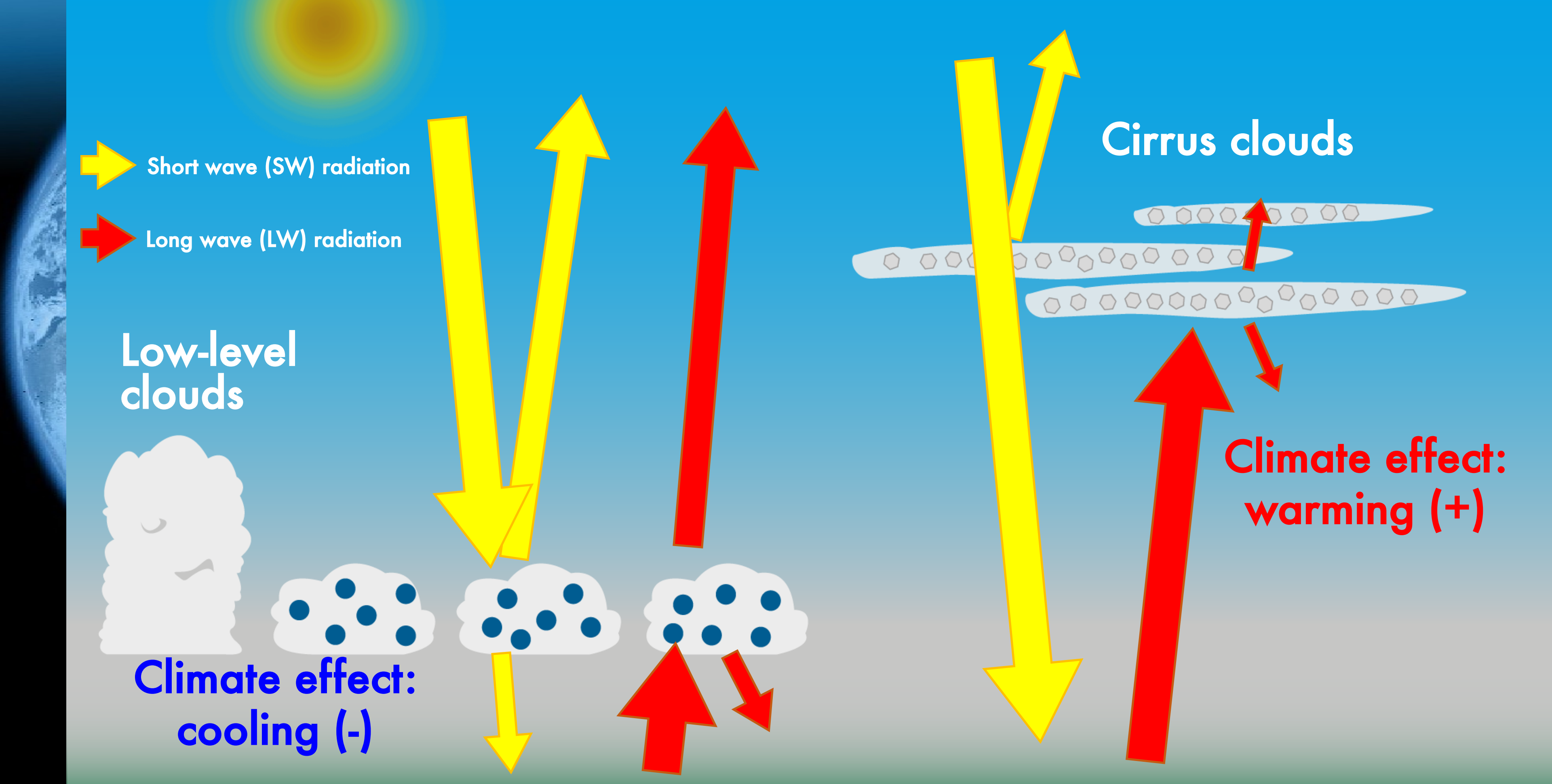
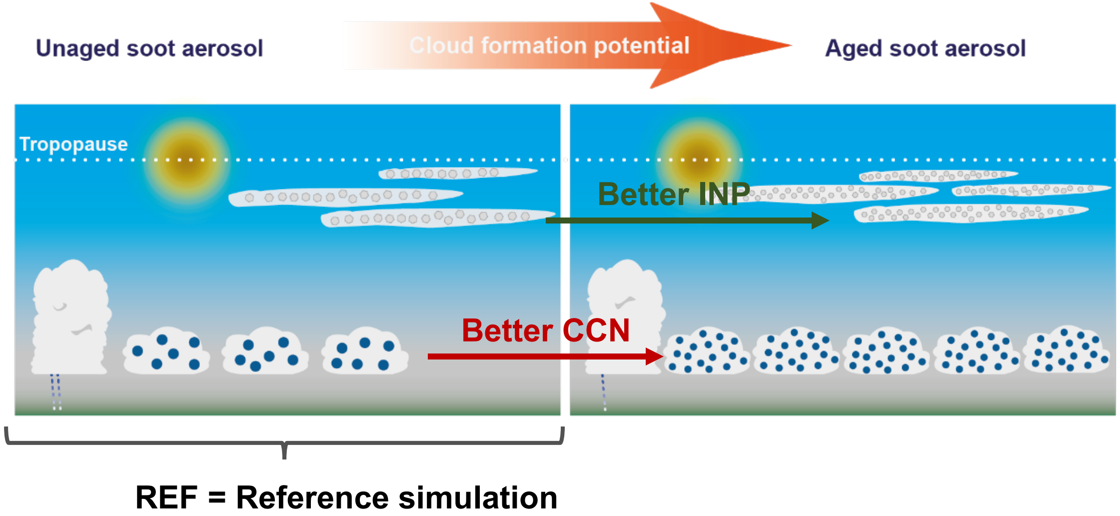
The impact of clouds on climate



Aging of soot changes their potential to form clouds

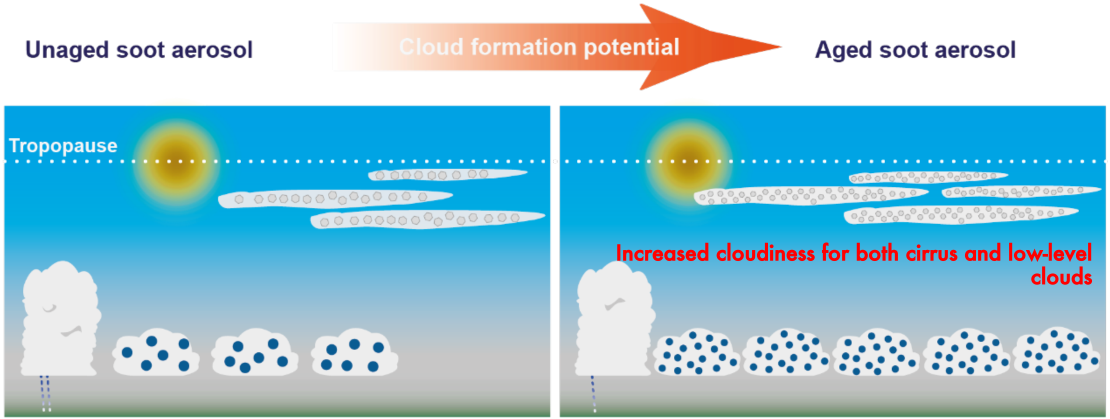


Aging mechanism considered here:

- Ozone aged soot particles show enhanced ability to act as cloud condensation nuclei (CCN). (from Friebel et al. 2019, ACP)
- Aging soot in aqueous sulfuric acid solutions enhances their ability to act as ice nucleating particles (INPs). (from Mahrt et al. 2020, ESPI)

Goal: Estimate the effect of aged soot on future climate.	Simulations of the future climate: Coupled atmospheric-mixed-layer ocean simulations, doubling CO_2 from pre-industrial (1850), running simulations into radiative equilibrium to obtain the global mean surface temperature response; the equilibrium climate sensitivity (ECS).

Future climate effects of aged soot



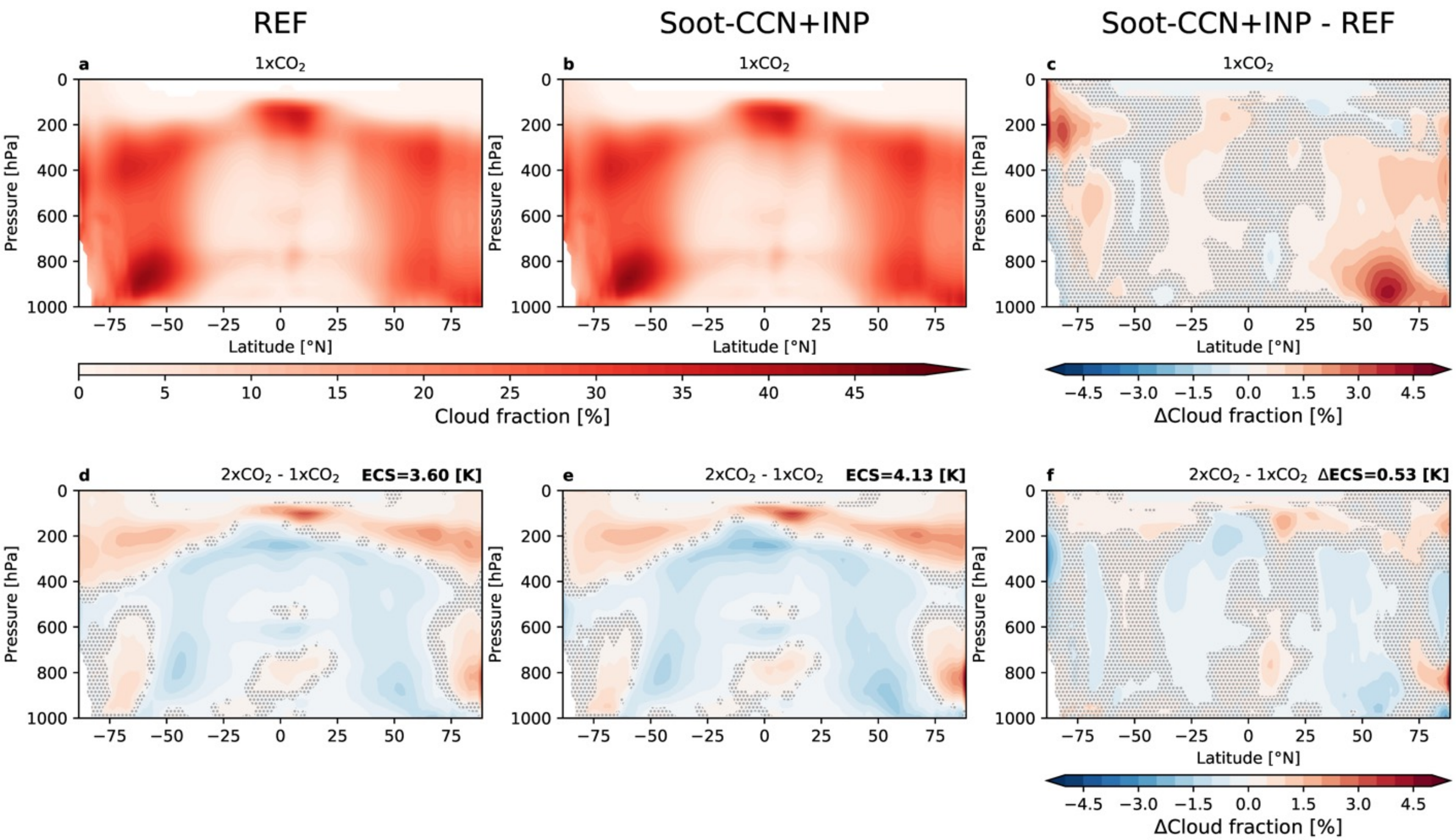
Effect of aged soot: Simulation Soot CCN+INP - REF:

How does cloud fraction change in warmer climate (2 x CO_2) and what is the impact on ECS?

REF 2 x CO_2:

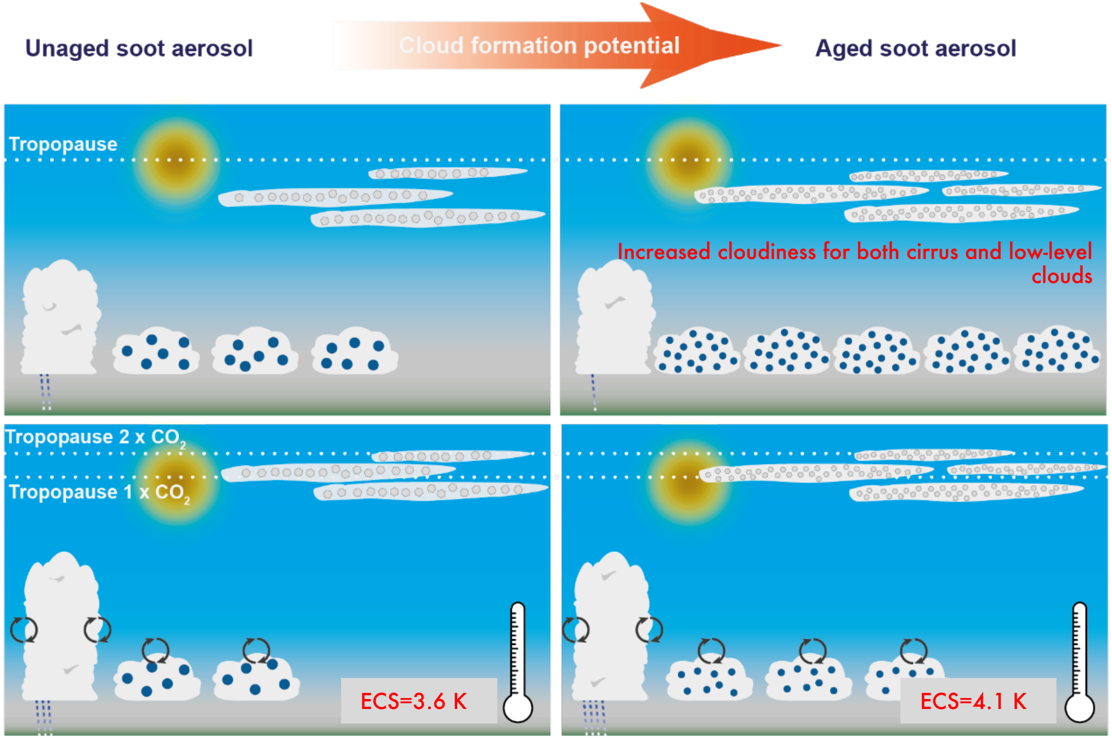
- Tropopause shifted to higher altitudes
- Reduced low-level cloudiness
- Increased entrainment of dry air

Changes in cloud cover from aged soot acting as CCN and INP: pre-industrial (1 x CO_2) and future (2 x CO_2)



0.53 K more warming due to aged soot acting as CCN and INPs.

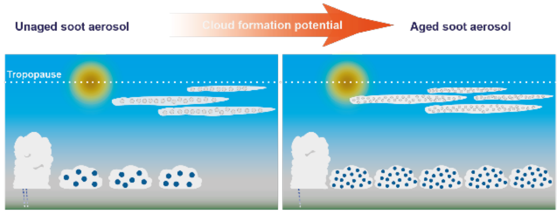
Future indirect climate effects of aged soot



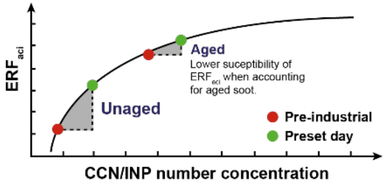
Soot-CCN+INP 2 x CO_2:

- Ice clouds become optically thicker, thus enhancing the positive cloud altitude feedback
- Further reduced low-level cloudiness due to increased entrainment of dry air caused by the enhanced cloud top cooling of the optically thicker clouds

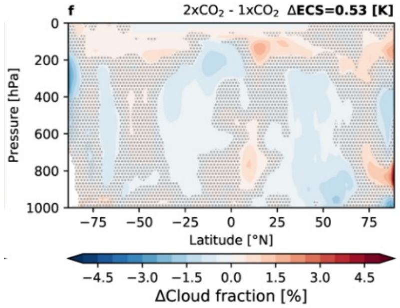
Summary and Conclusions



Soot particles can be aged chemically and physically and affect cloud properties and climate.



Smaller shortwave indirect aerosol effect (from pre-industrial times to the present day – not shown).



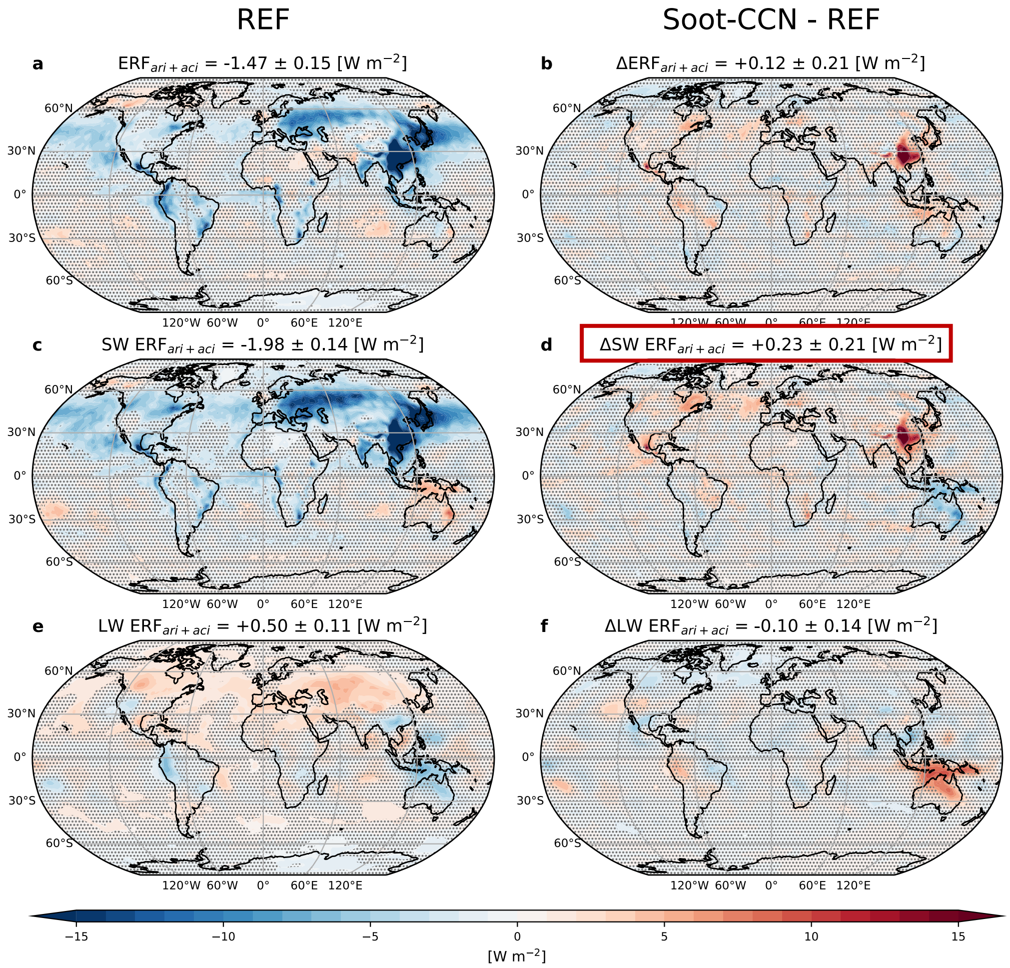
Further reduction in low-level clouds and enhanced high-altitude cirrus cloud thickness leads to exacerbated global mean surface warming by ~0.5 K.

Full reference: Lohmann et al. (2020): https://www.nature.com/articles/s41561-020-0631-0

Thanks a lot for your attention!

Questions?

Radiative impact of O_3-aged soot since pre-industrial times

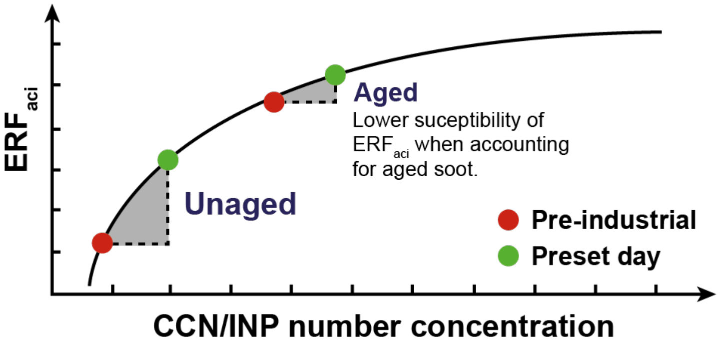


Simulation REF:

- Net ERF$_{ari+aci}$ dominated by SW radiation.
- Largest contribution by cloud effects (-1.64 W m^{-2}).

O_3 aged soot: Simulation Soot CCN - REF:

- SW anthropogenic aerosol effect reduced
- Due to increased pre-industrial CCN concentration, reducing relative impact of anthropogenic aerosols between pre-industrial and present-day.



Lohmann et al. (2020), Nat. Goesci.

Key climate impacts

Simulation	REF	Soot-CCN	Soot-CCN+INP	Soot-CCN+INP-ac
$ERF_{ari+aci}$ – SW [W m^{-2}]	-1.98 ± 0.14	-1.75 ± 0.15	-1.66 ± 0.17	-1.78 ± 0.18
$ERF_{ari+aci}$ – LW [W m^{-2}]	0.50 ± 0.11	0.40 ± 0.09	0.39 ± 0.10	0.42 ± 0.11
$ERF_{ari+aci}$ (net) [W m^{-2}]	-1.47 ± 0.15	-1.35 ± 0.15	-1.27 ± 0.16	-1.37 ± 0.17
IRF_{ari} (net) [W m^{-2}]	0.03 ± 0.07	0.00 ± 0.06	-0.01 ± 0.06	0.09 ± 0.06
Cloud effects (net) [W m^{-2}]	-1.64 ± 0.13	-1.46 ± 0.13	-1.32 ± 0.14	-1.64 ± 0.12
ECS [K]	3.60 ± 0.06	4.00 ± 0.05	4.13 ± 0.04	4.02 ± 0.06
Δ precipitation [mm d^{-1}]	0.194 ± 0.005	0.224 ± 0.006	0.238 ± 0.005	0.232 ± 0.006
hydrological sensitivity [% K^{-1}]	1.80 ± 0.06	1.89 ± 0.06	1.94 ± 0.05	1.94 ± 0.06

Changes in ice crystal number concentration: pre-industrial (1xCO_2) and future (2xCO_2)