

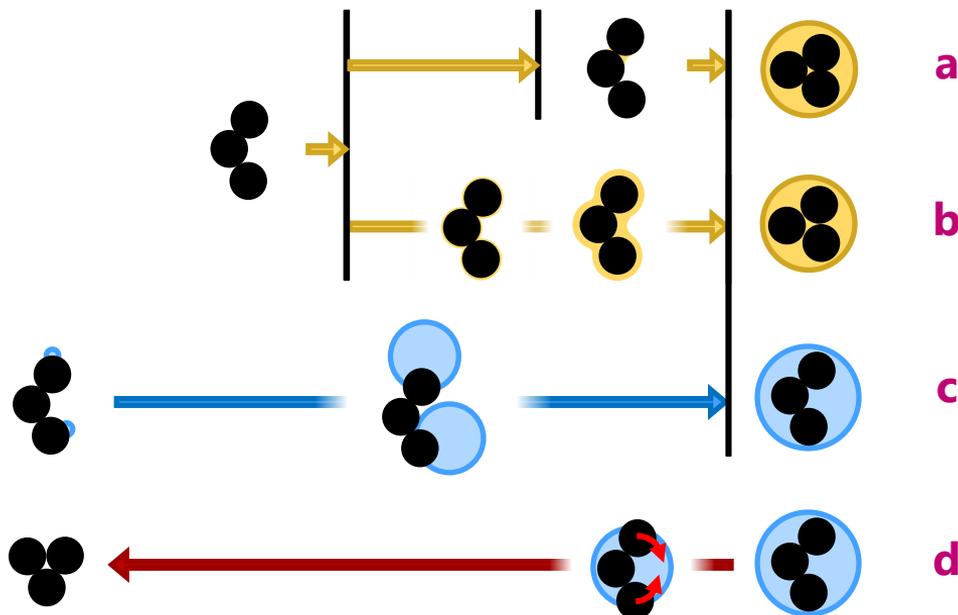
# Mechanisms of soot restructuring and compaction

Which one happens in the atmosphere?

Joel C. Corbin<sup>1</sup>  
 Robin L. Modini<sup>2</sup>  
 Martin Gysel-Beer<sup>2</sup>

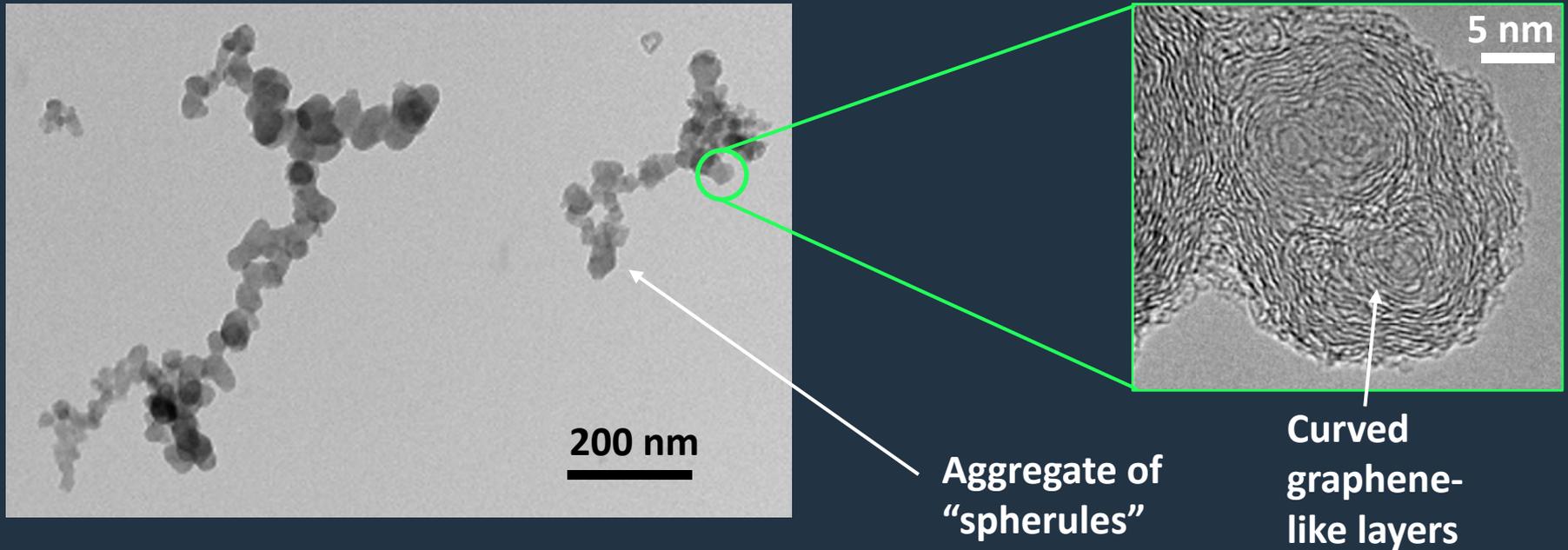
<sup>1</sup> National Research Council, Canada  
<sup>2</sup> Paul Scherrer Institute, Switzerland

June 2022



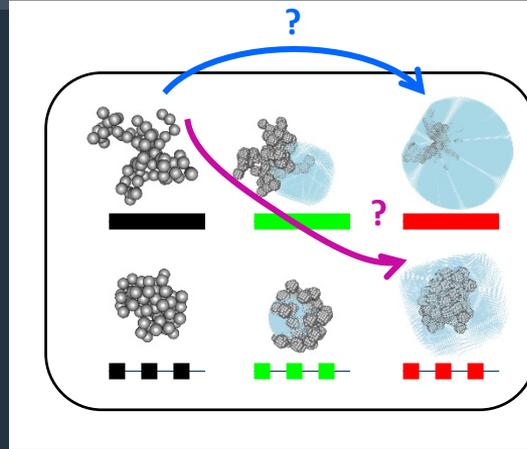
# Typical Soot BC (black carbon)

**Definition: flame-synthesized nano-aggregates of nearly-graphitic carbon spherules.**

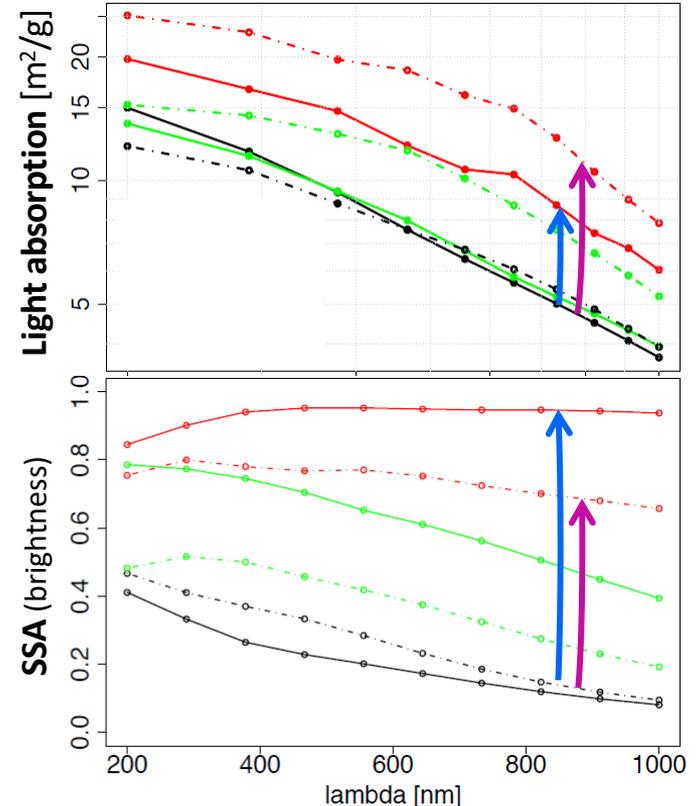


# Soot compaction affects its climate forcing

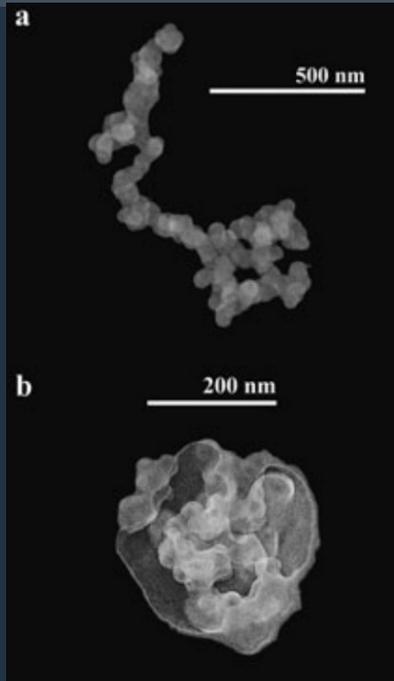
Soot climate warming comparable to that of CO<sub>2</sub> [1]



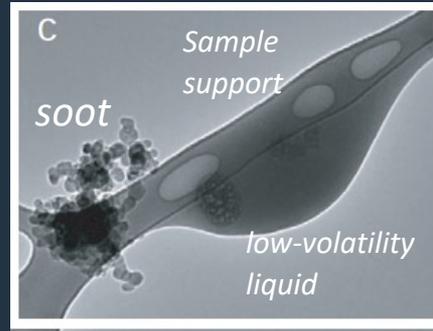
Structure can change SSA ("brightness") by 0.2; light absorption by 30%. [2]



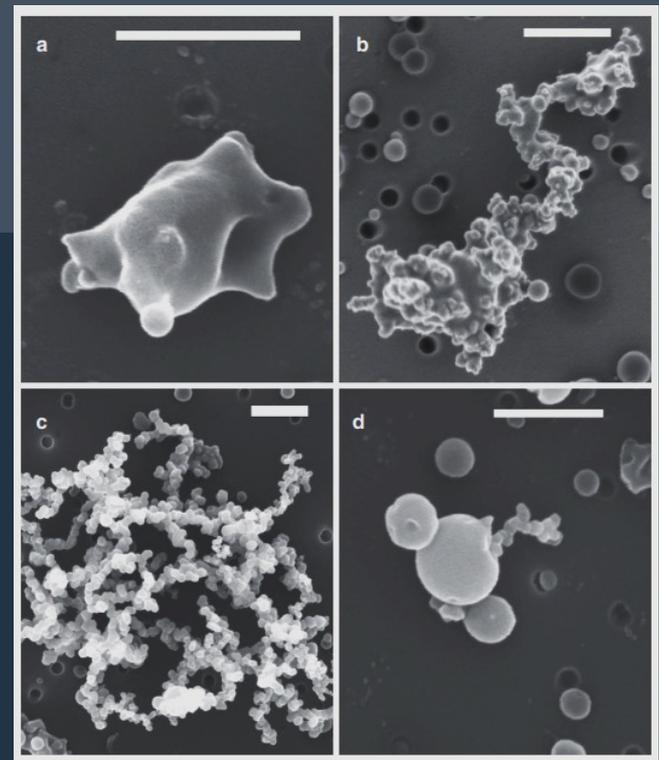
# Atmospherically-coated and evaporated soot



Near India,  
Pt-coated



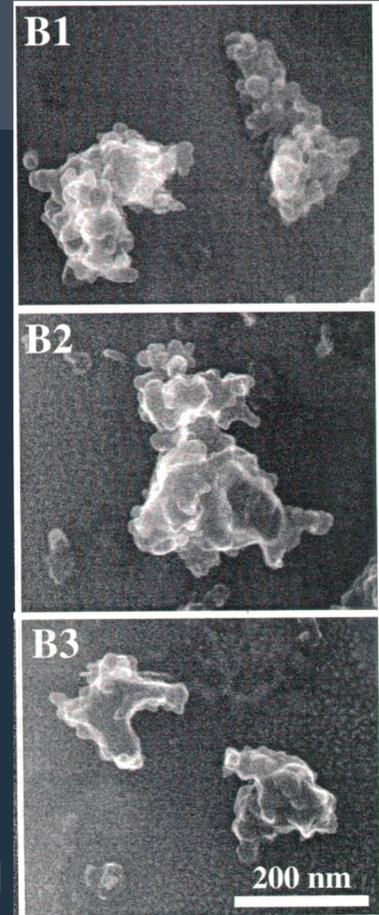
Biomass burning  
in Mexico City



Wildfires

[Wildfires] Swarup China et al., Nature Comm. 2013  
[Pt-coated] Coz and Leck, Tellus B 2011  
[Mexico City] Adachi and Buseck, ACP 2008

# Soot compacted in engine exhaust

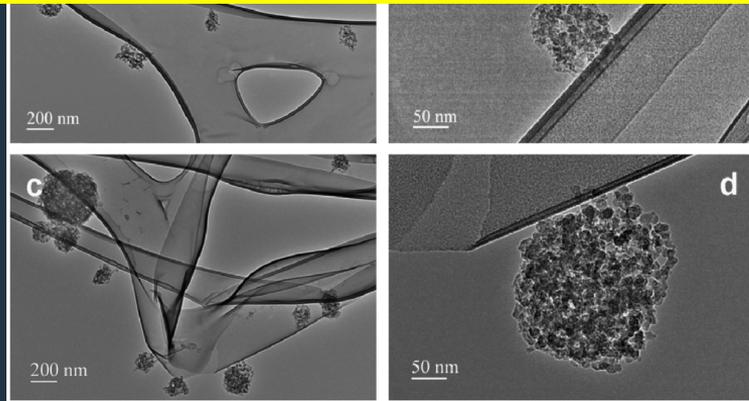


Diesel engine,  
S-enriched fuel

# Laboratory-coated and evaporated soot

Water-coated soot was directly injected into water ... the structure was uncollapsed. Soot restructuring [...] occurs during evaporation, not condensation.

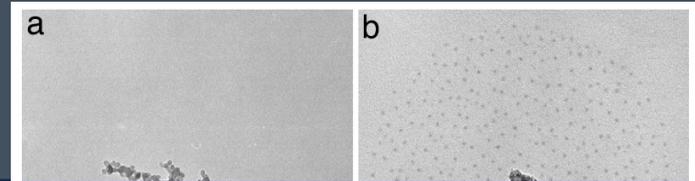
[Ma, ... Zachariah, JAS 2013]



Flame soot + water

[Oleic] Ghazi and Olfert, AST 2013  
[Sulfuric] Zhang et al PNAS 2008  
[Water] Ma, Zachariah, et al. JAS 2013

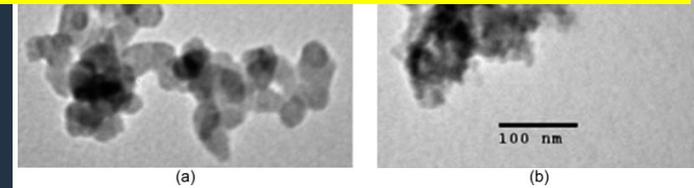
## Soot + sulfuric acid



“Our results show that the particles are restructured (become compact) when coated.”  
[Bambha, Dansson, Schrader, Michelsen, 2013]

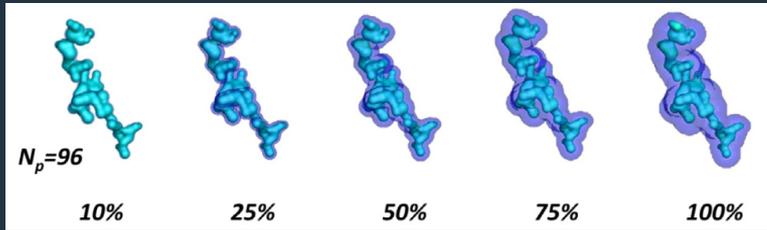
minor restructuring occurs already at zero vapor supersaturation due to capillary condensation  
[Enekwizu, Hasani, Khalizov, 2021]

...and many others



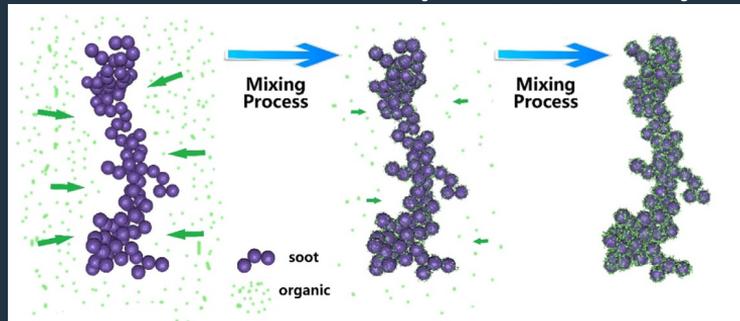
Flame soot  
+ oleic acid

# No consensus on how to model coated soot

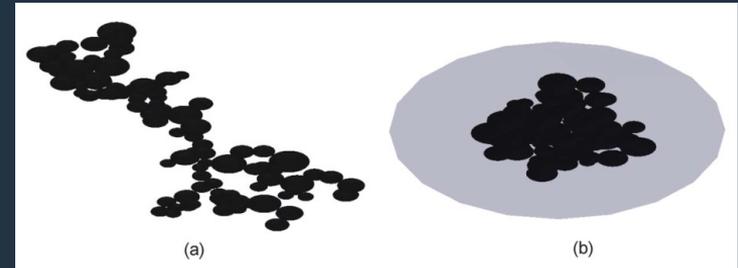


“no compaction”  
(Liu, 2017)

“no compaction”  
(Yu Wu, 2017)

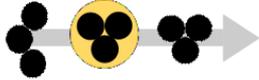
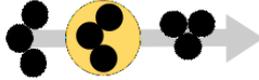


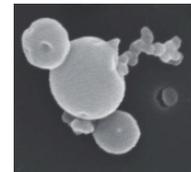
“compaction”  
(Yu Wu, 2015)



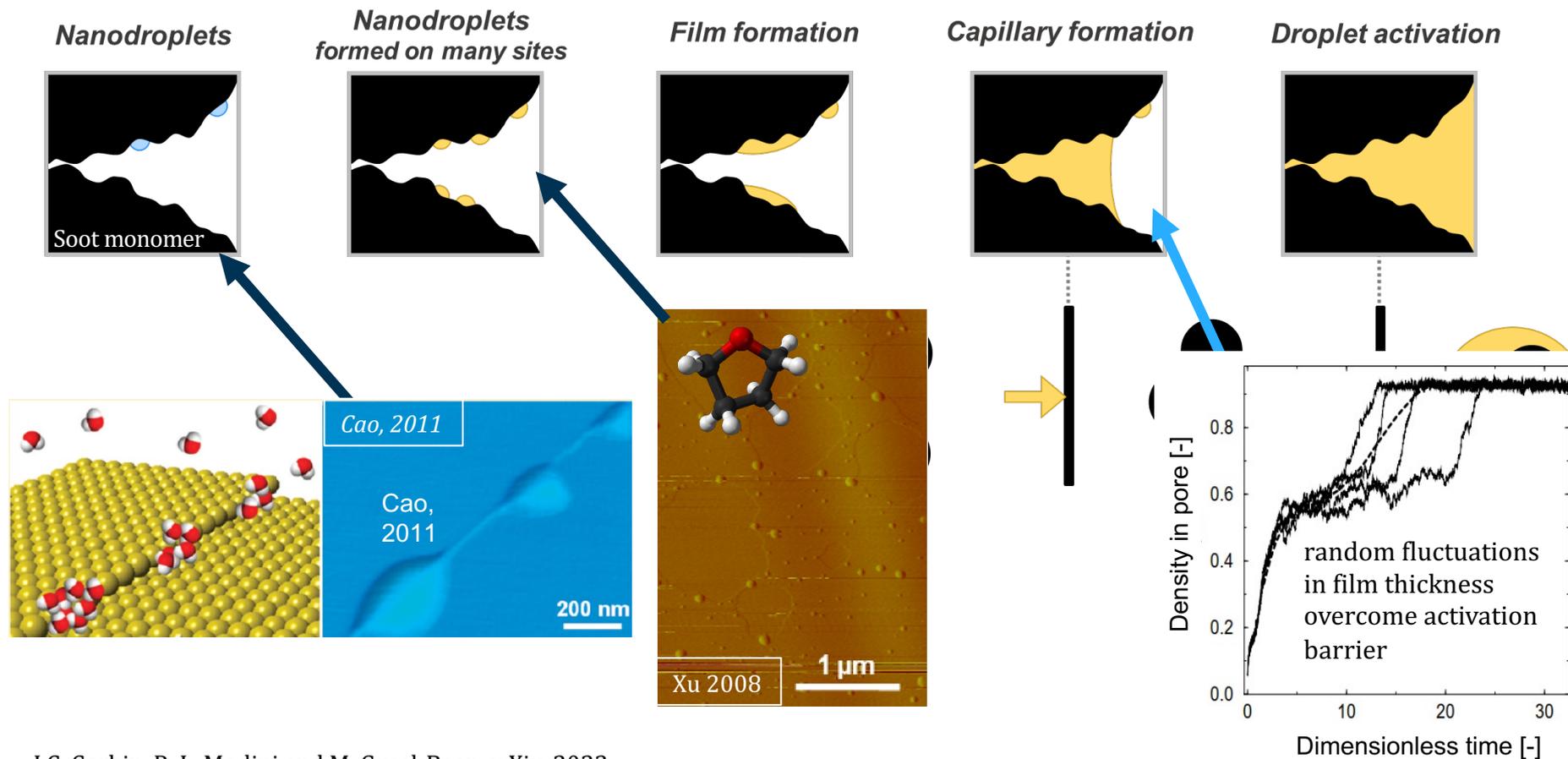
# Summary of soot-compaction literature

Review: J.C. Corbin et al, arXiv, 2022.

- We reviewed 57 studies reporting soot compaction.
- Most assumed  without confirmation.
- 1 of 57 demonstrated  directly.
- Some (underappreciated!) studies demonstrated  directly:
  1. Leung et al. (2017):  $d_{mob}$  decreased for liq. but not solid SOA
  2. Miljevic et al. (2010): compacted soot by bubbling through organics (**S<1**)
  3. Chen et al. (2016, 2018): tiny amounts of PAH caused (capillary) compaction
- Some (overappreciated!) microscopy studies observed exceptions. Statistical representativeness not demonstrated.

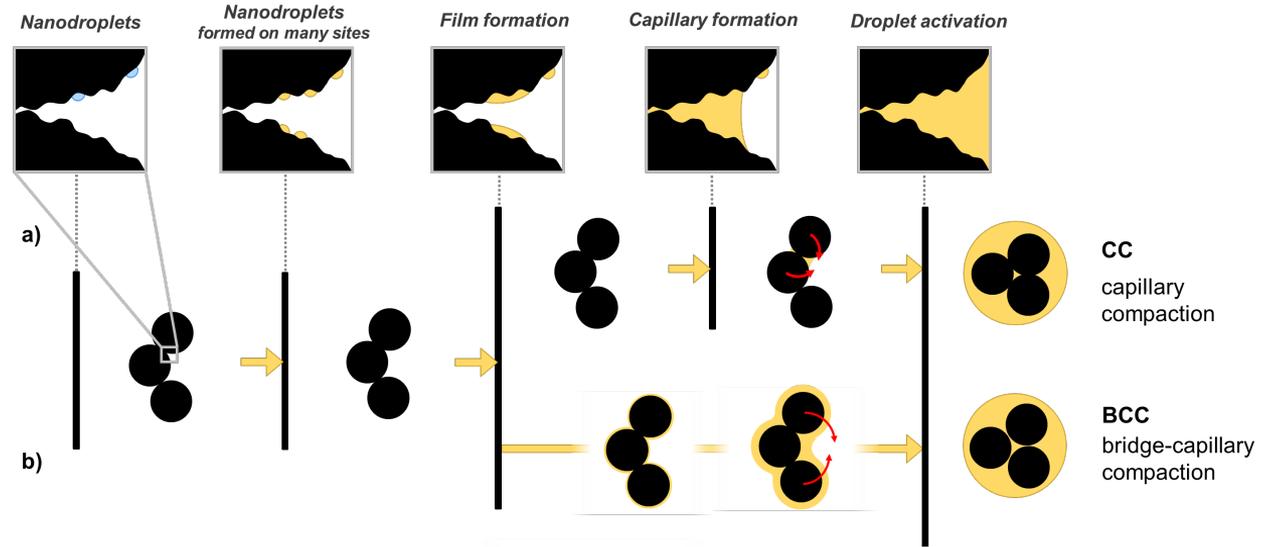


# Mechanisms of compaction: summary of lit. review & experiments

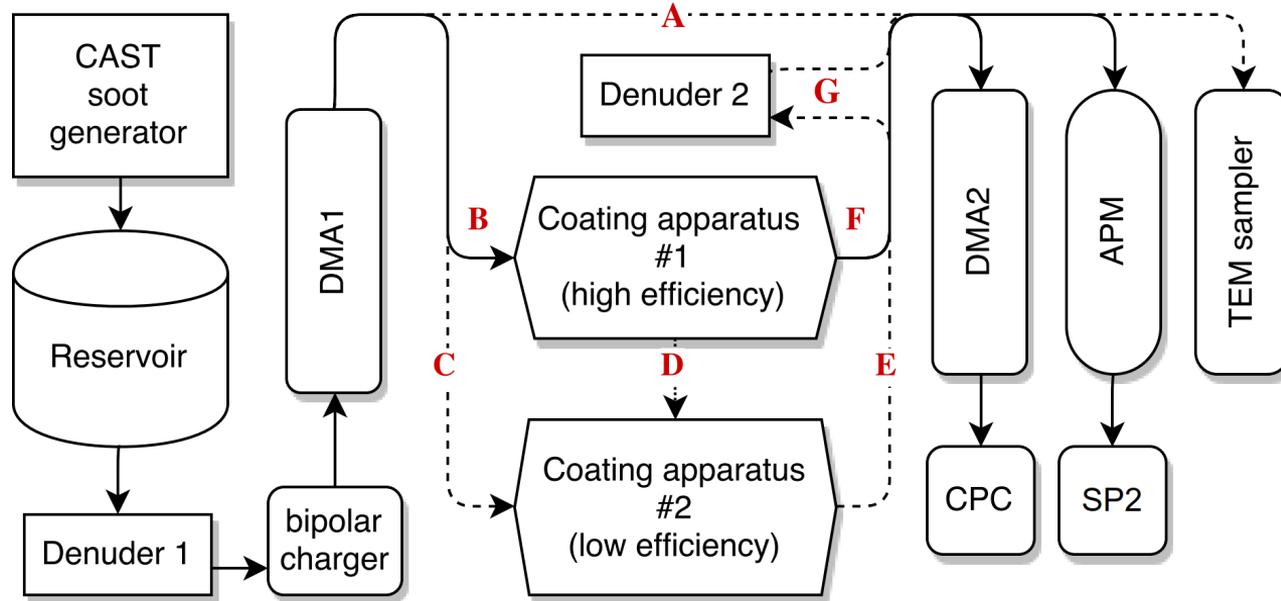


# Mechanisms of compaction: summary of lit. review & experiments

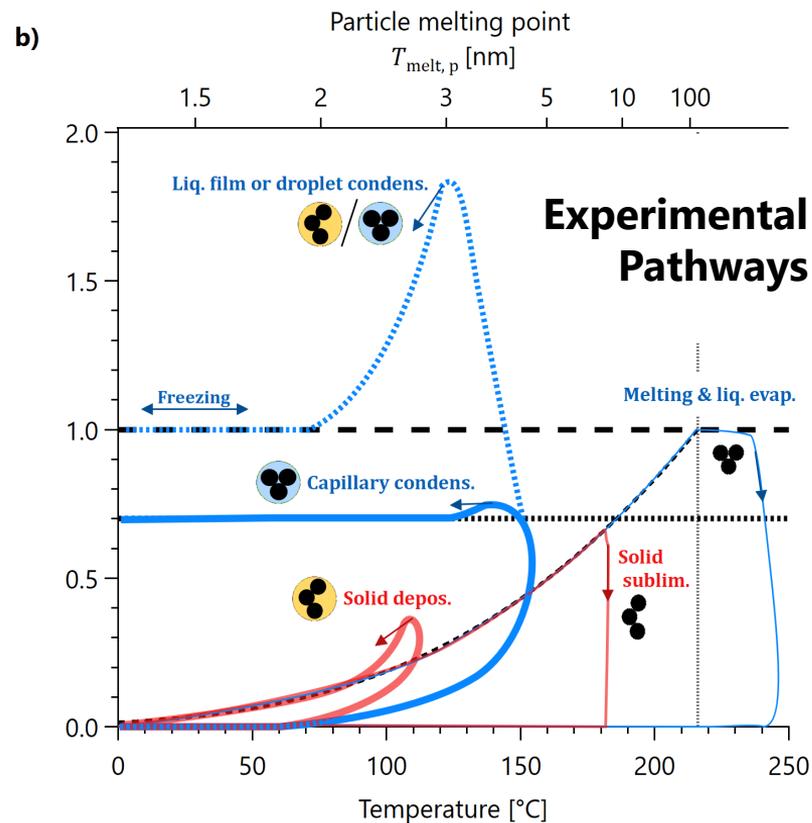
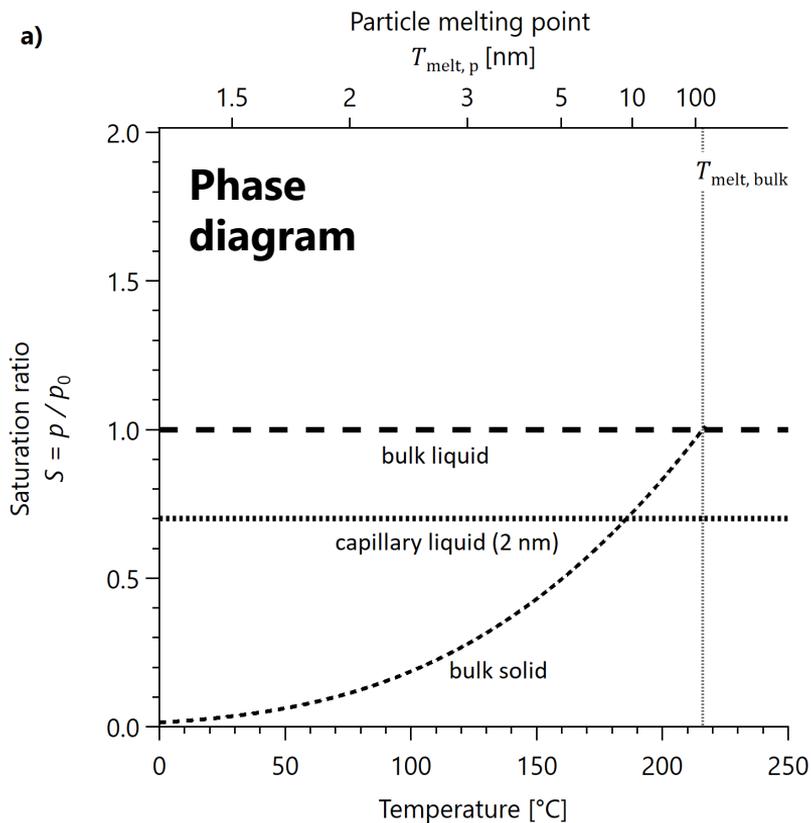
1. New phases face energy barriers |
2. We identified 4 mechanisms from surface-science literature (*right*)
3. Only 1 of 57 reviewed soot-structure studies observed path (c), via **NDAC**: high  $\theta$  and RH > 120%.
4. Next, we demonstrated these mechanisms in the laboratory...



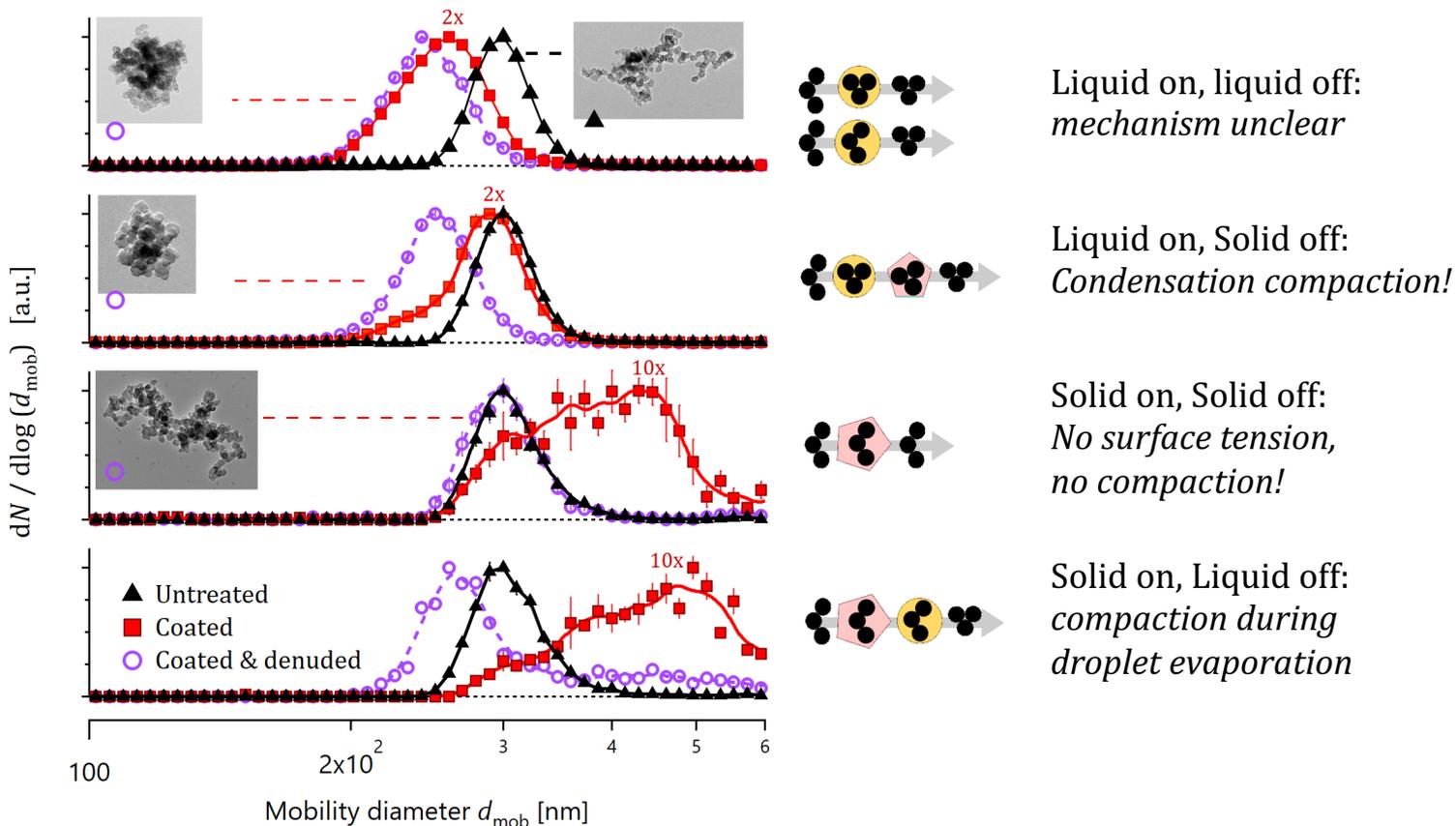
# Experimental setup



# When does soot restructure? Experiment



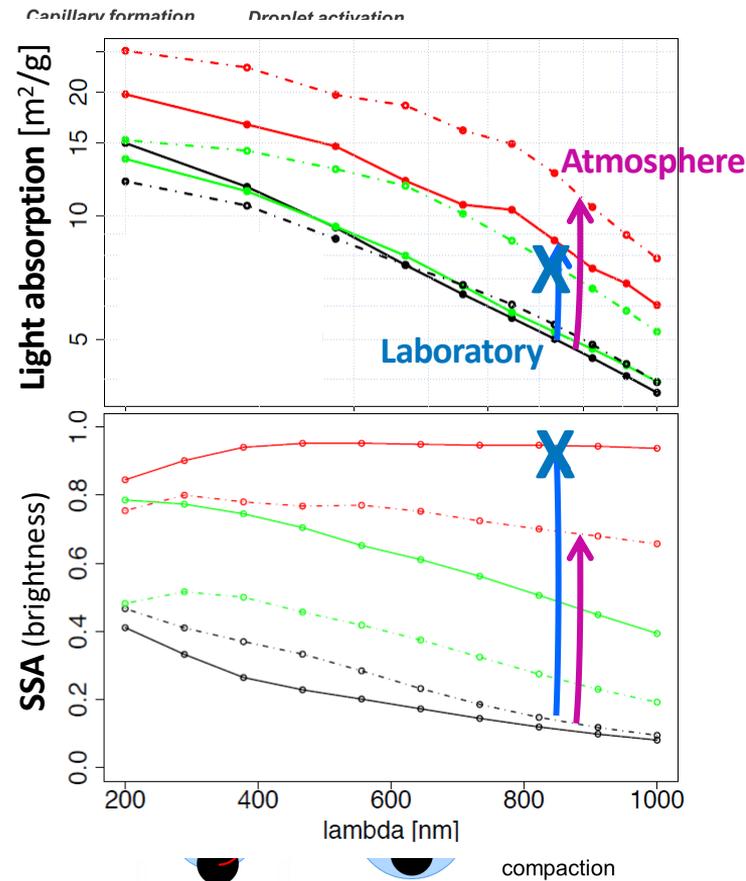
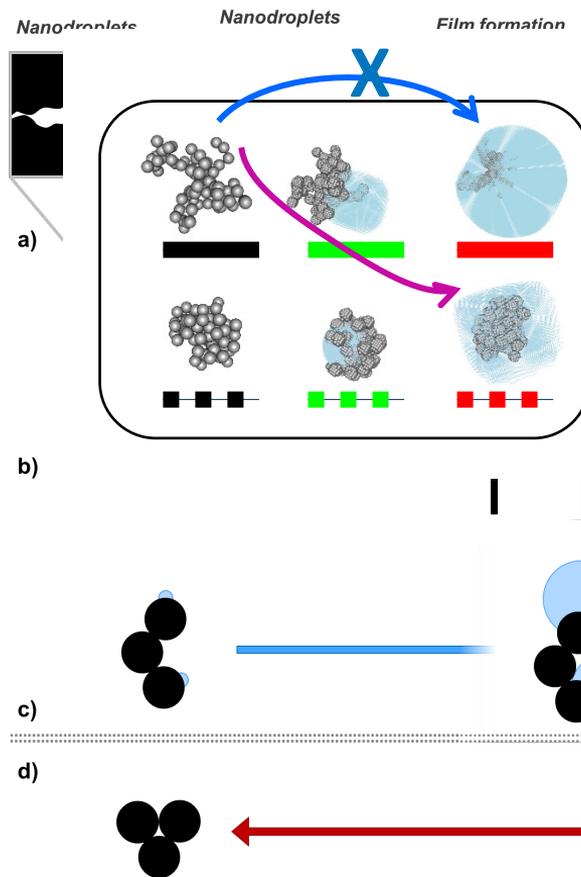
# Results: Both condens. and evap. cause compaction





# Mechanisms of compaction, Summary

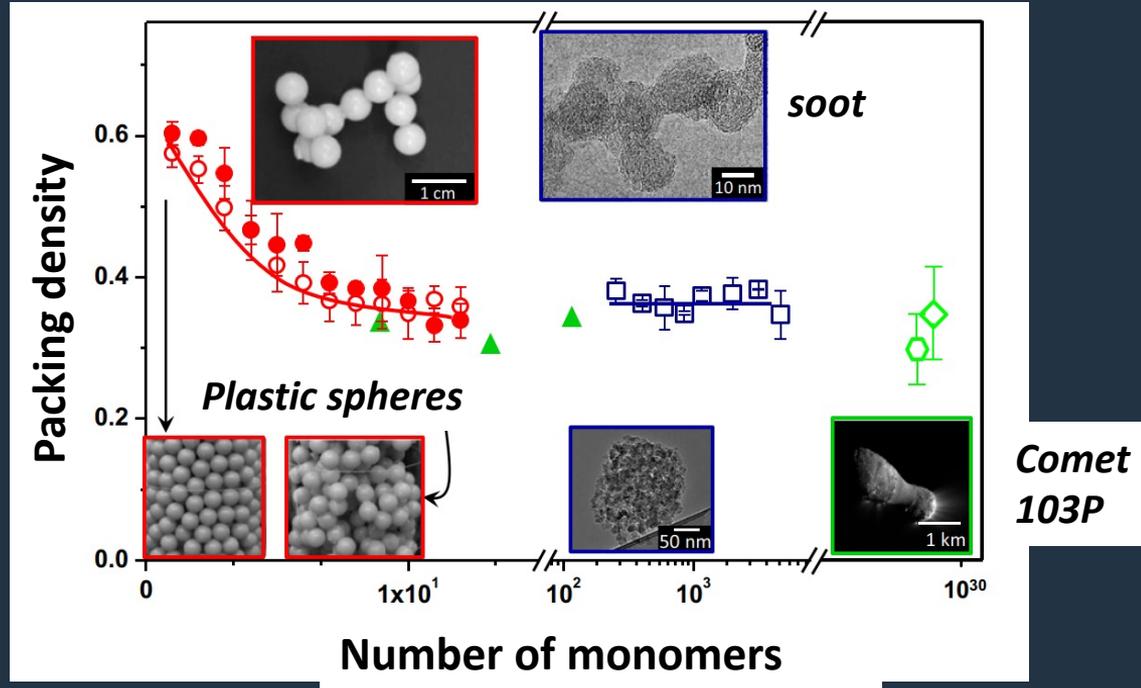
1. We propose a theoretical framework, connecting **compaction mechanisms** to **phase nucleation**.
2. Soot is compacted by **condensation and evaporation**, as shown in lab.
3. **Condensation-compaction** will occur in **engines & atmosphere**. For aged soot, likely also in **human lungs**.



Joel.Corbin@nrc.ca

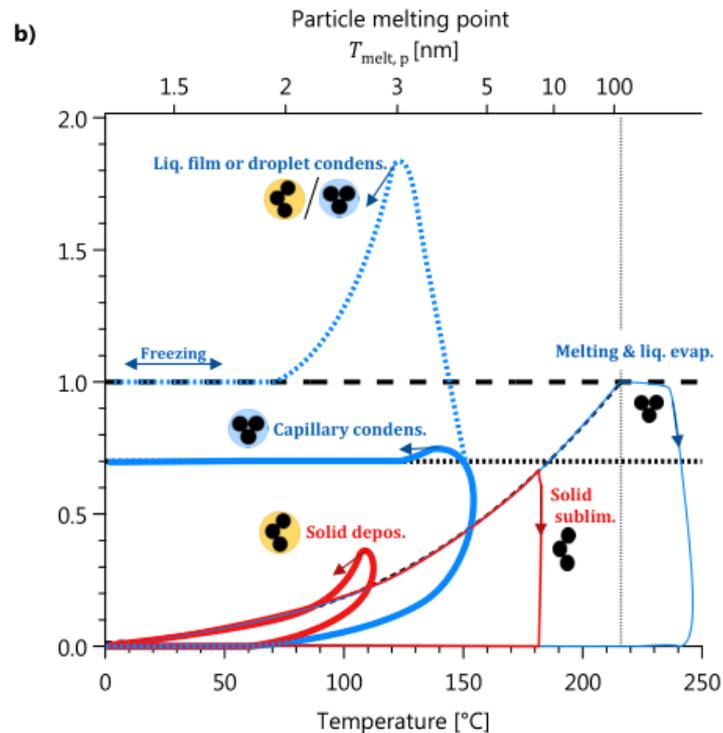
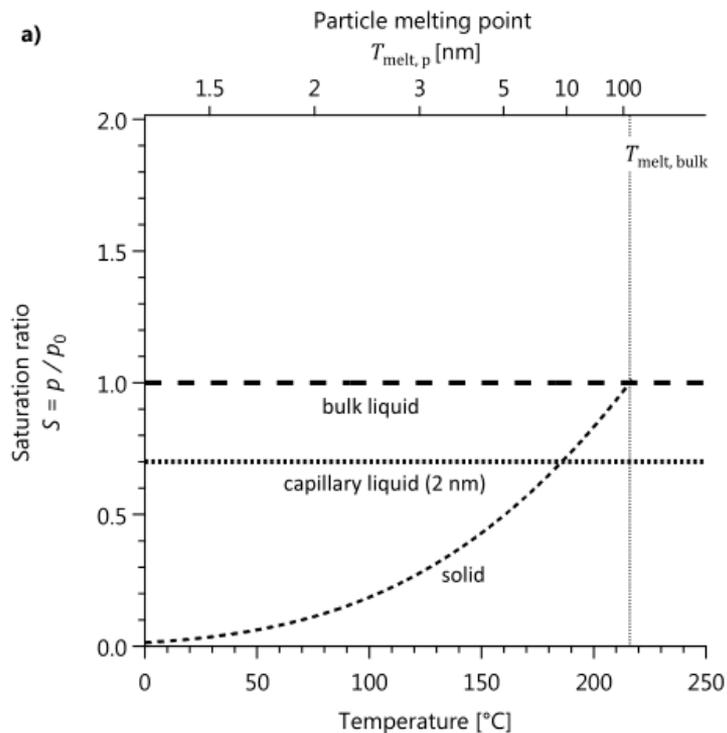
**FIN**

# Compact aggregates are universally observed

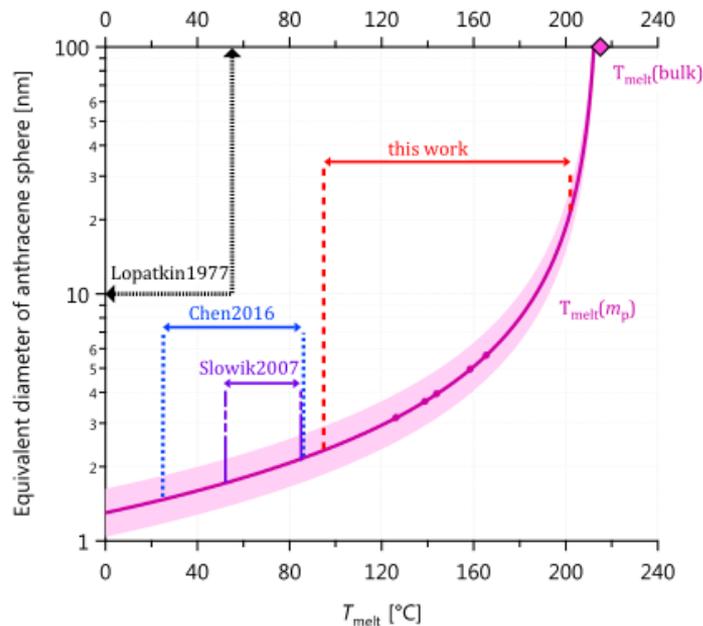
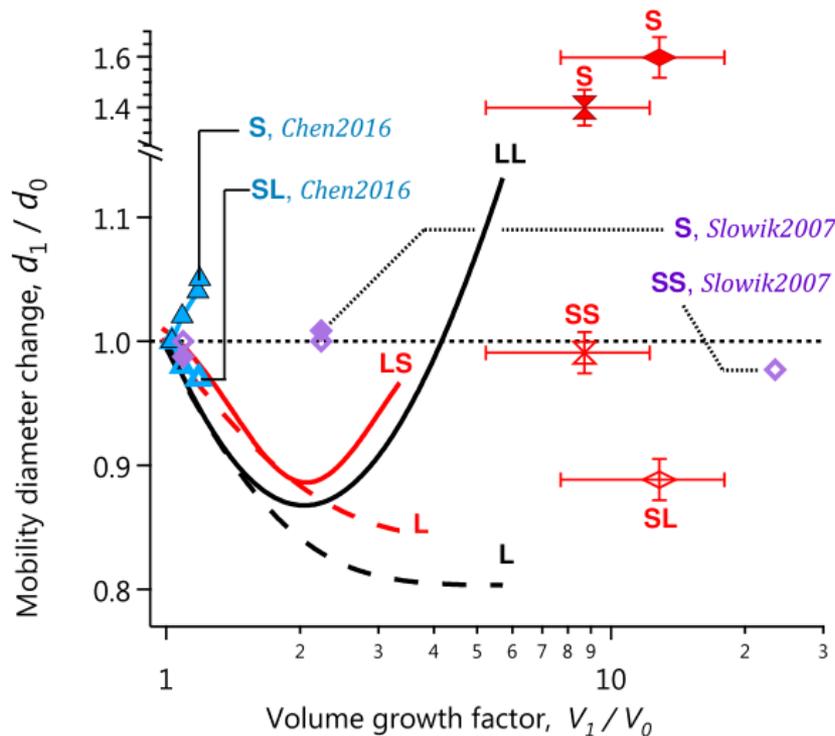


# When does soot restructure? Experimental design

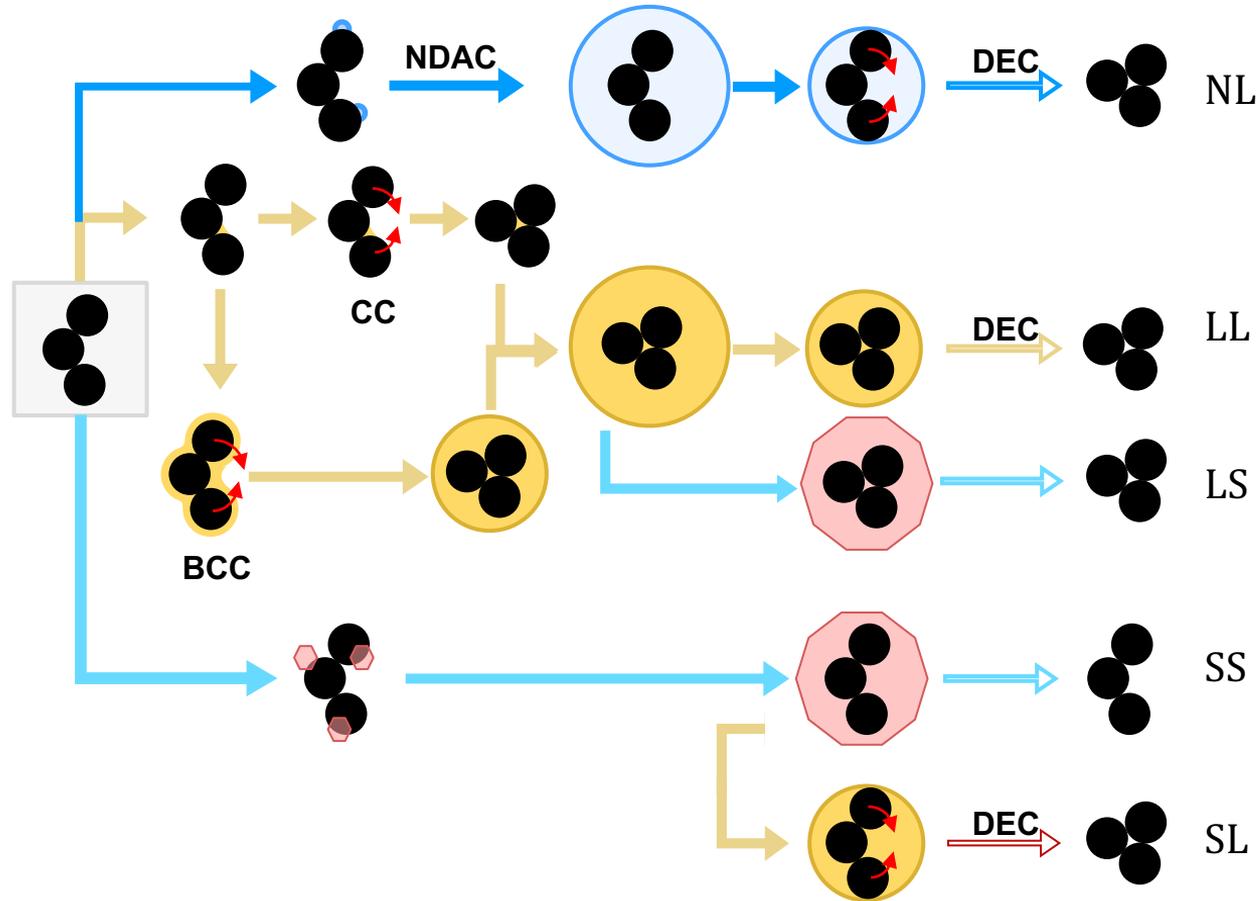
## Coating phase controlled by travelling across phase diagram



# Results: corroborated by two previous literature studies

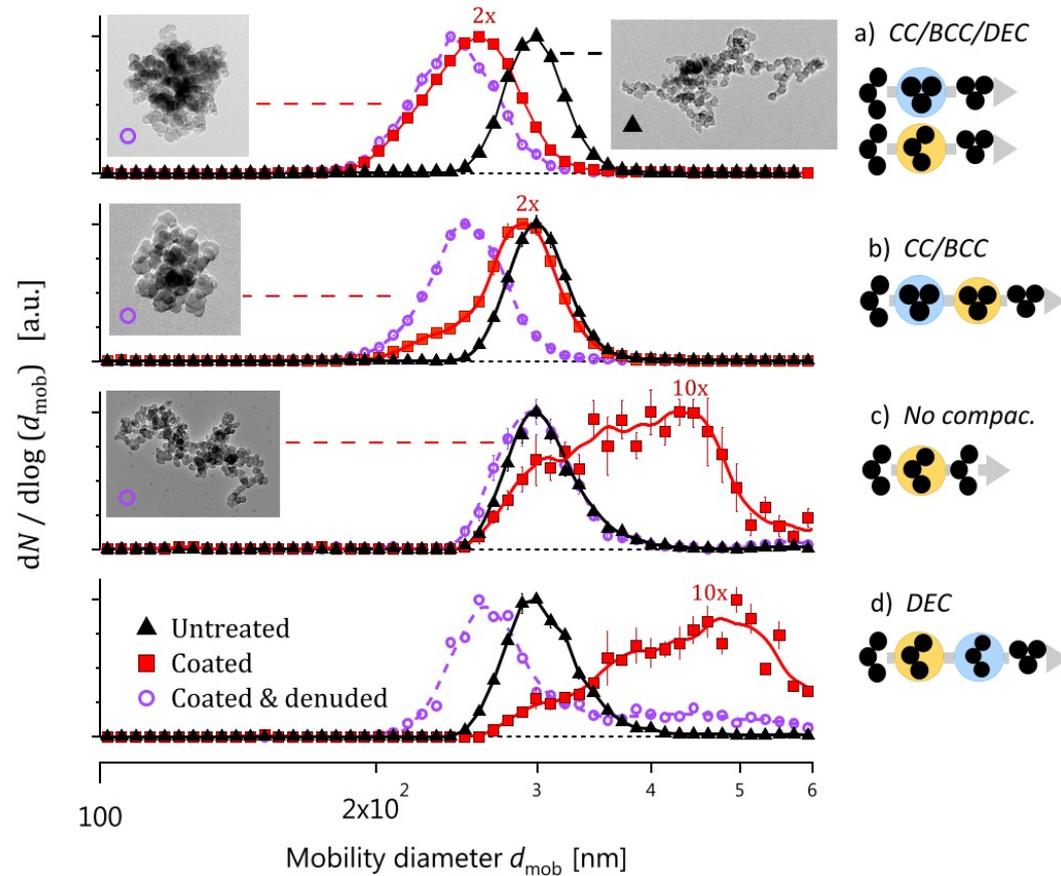


# Discussion: Coating pathways

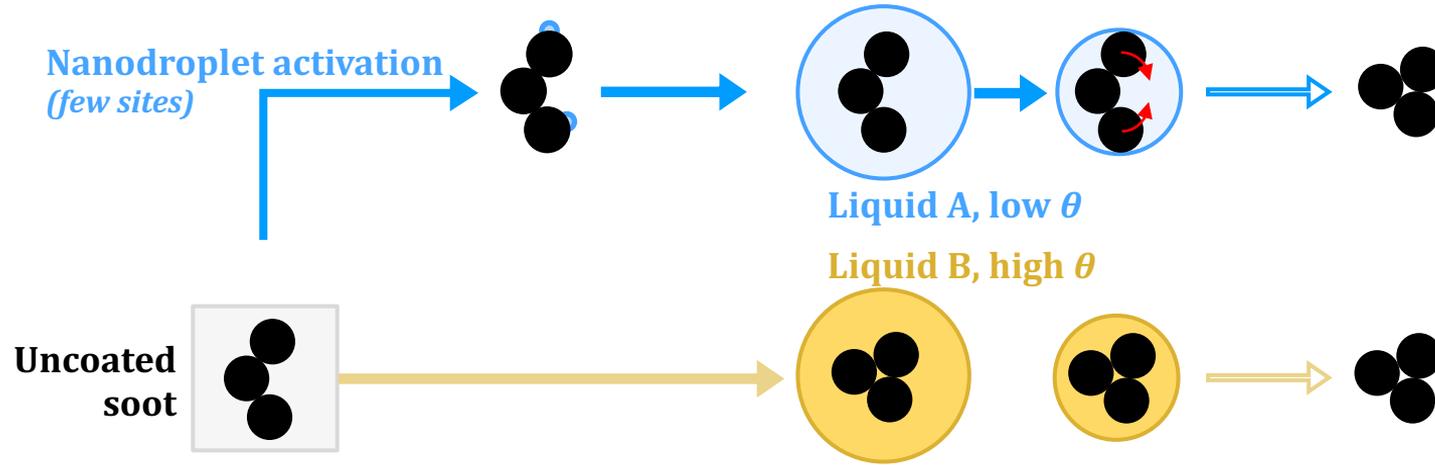


# Conclusions

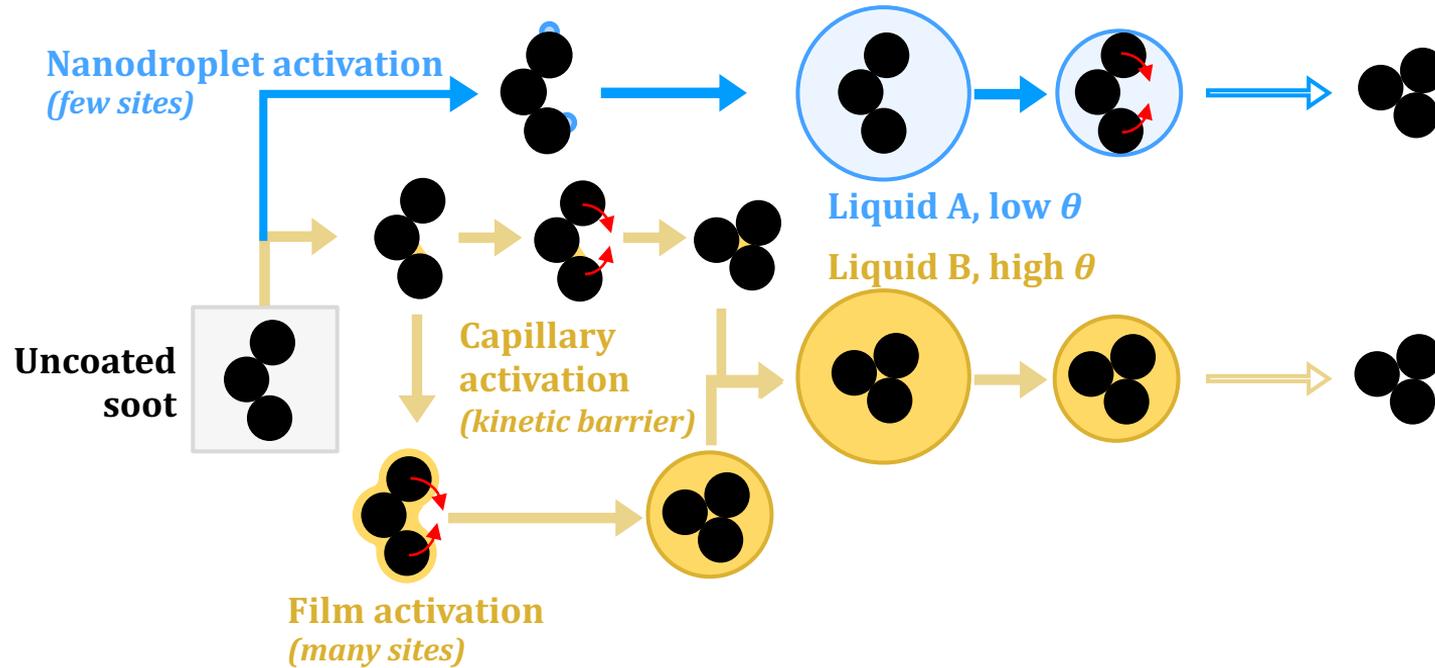
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# Research question: When does soot restructure?



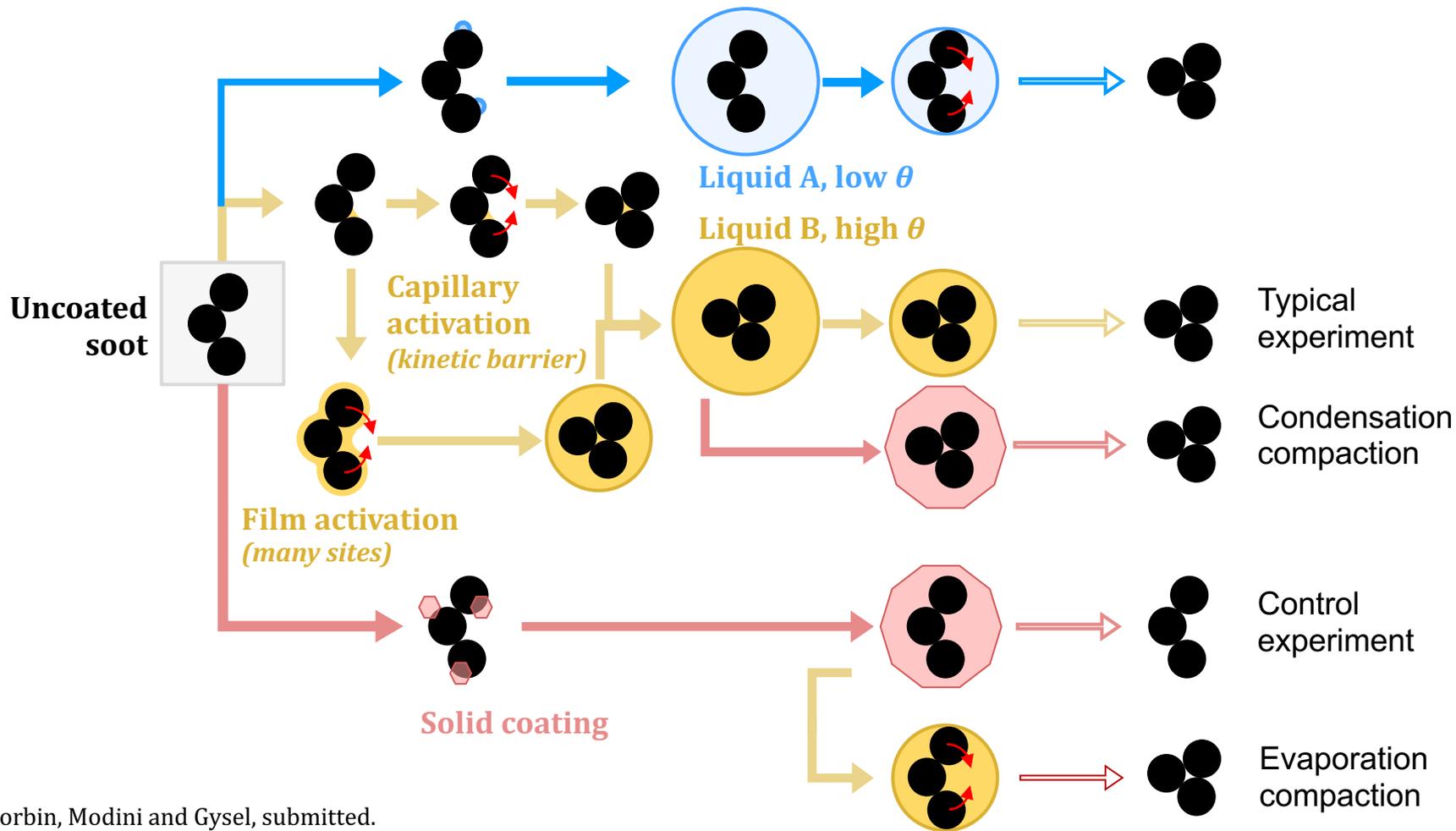
# When does soot restructure? Mechanisms from surface science



Activation mechanisms: Laaksonen 2020, Xu et al 2010, Cao et al 2011, Restagno 2008, and others

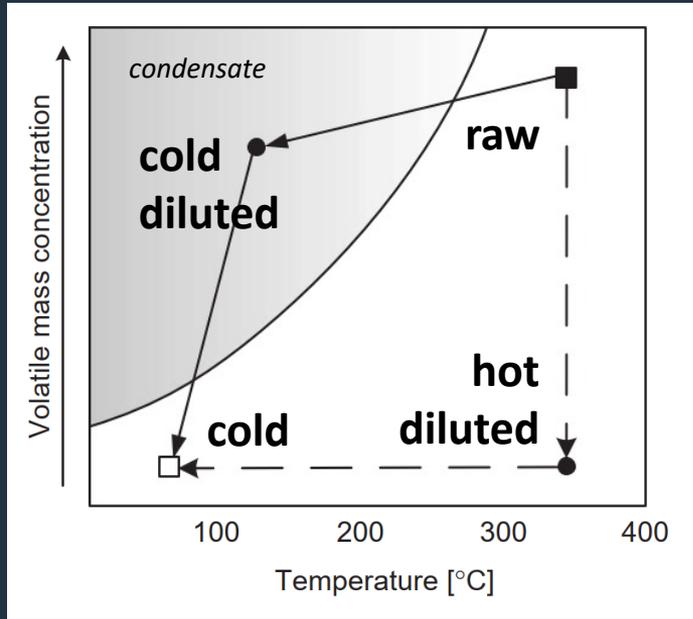
\*Corbin, Modini and Gysel, submitted.

# When does soot restructure? Experiments

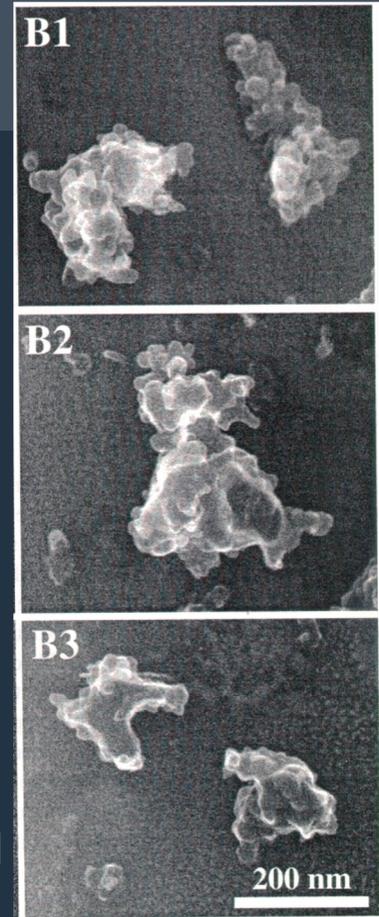


\*Corbin, Modini and Gysel, submitted.

# Soot compaction may also occur in engines



Burtscher, J. Aerosol Sci 1997



Diesel engine,  
S-enriched fuel  
[Weingartner]