Impact of carbon capture & storage technology alkanolamines on new particle formation in air

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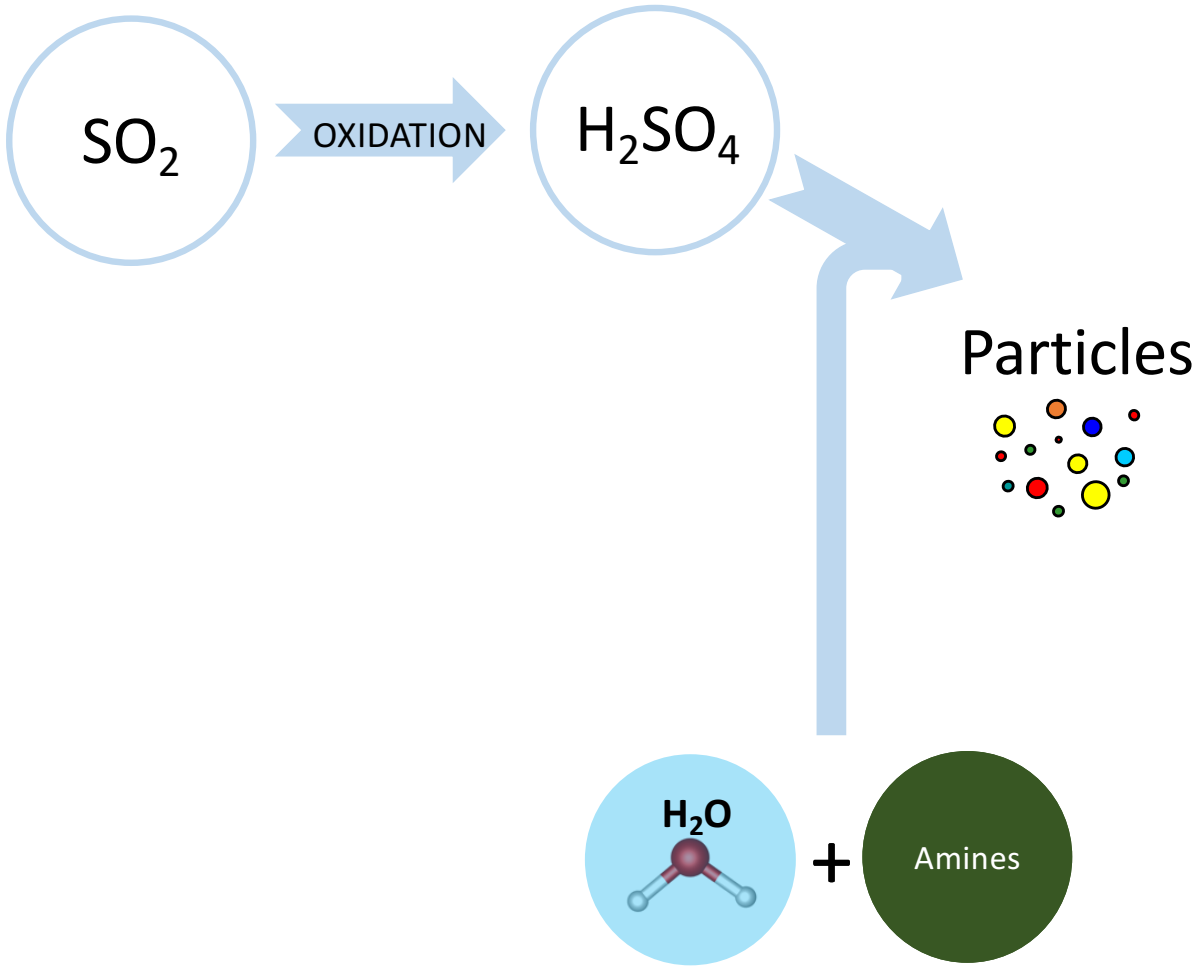
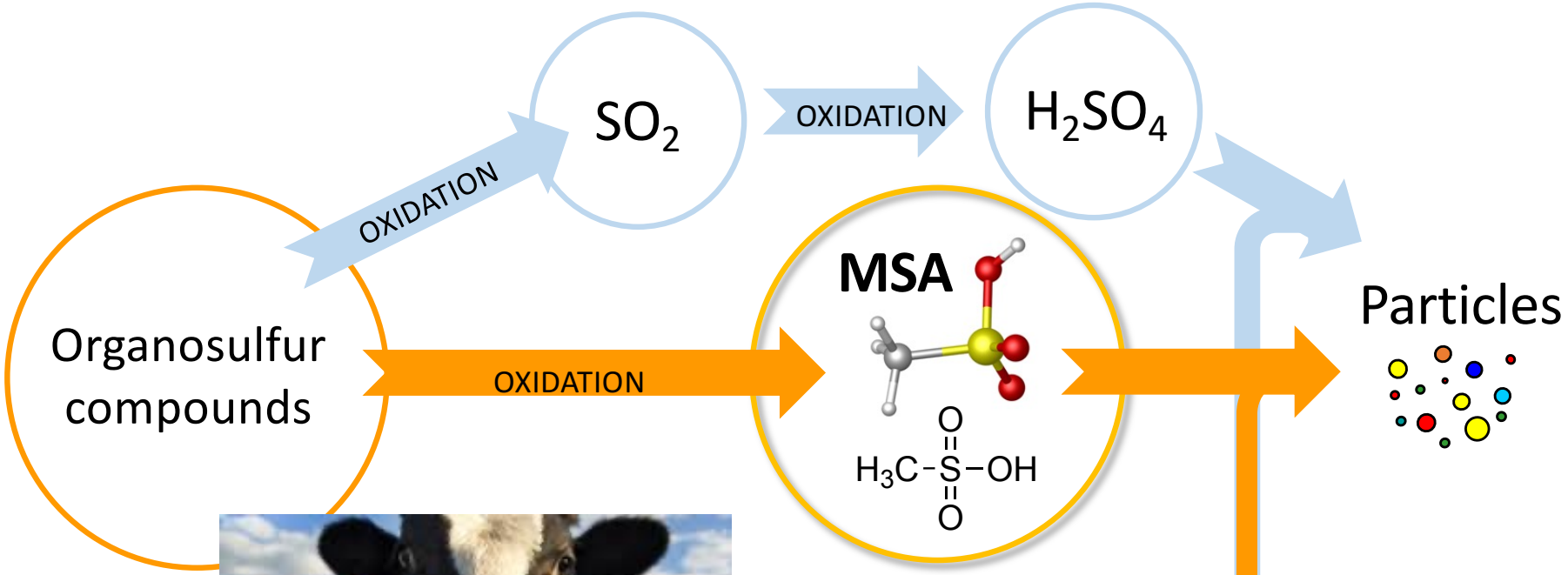
Research funded by

New particle formation in air



New particle formation in air



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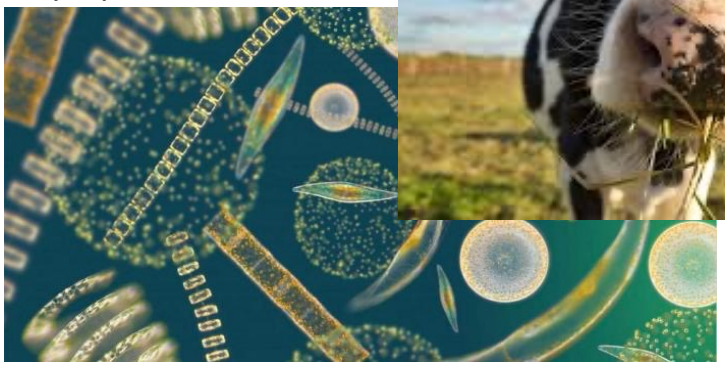
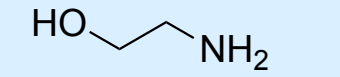
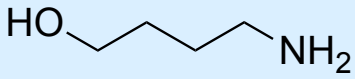


Agriculture

Monoethanolamine 

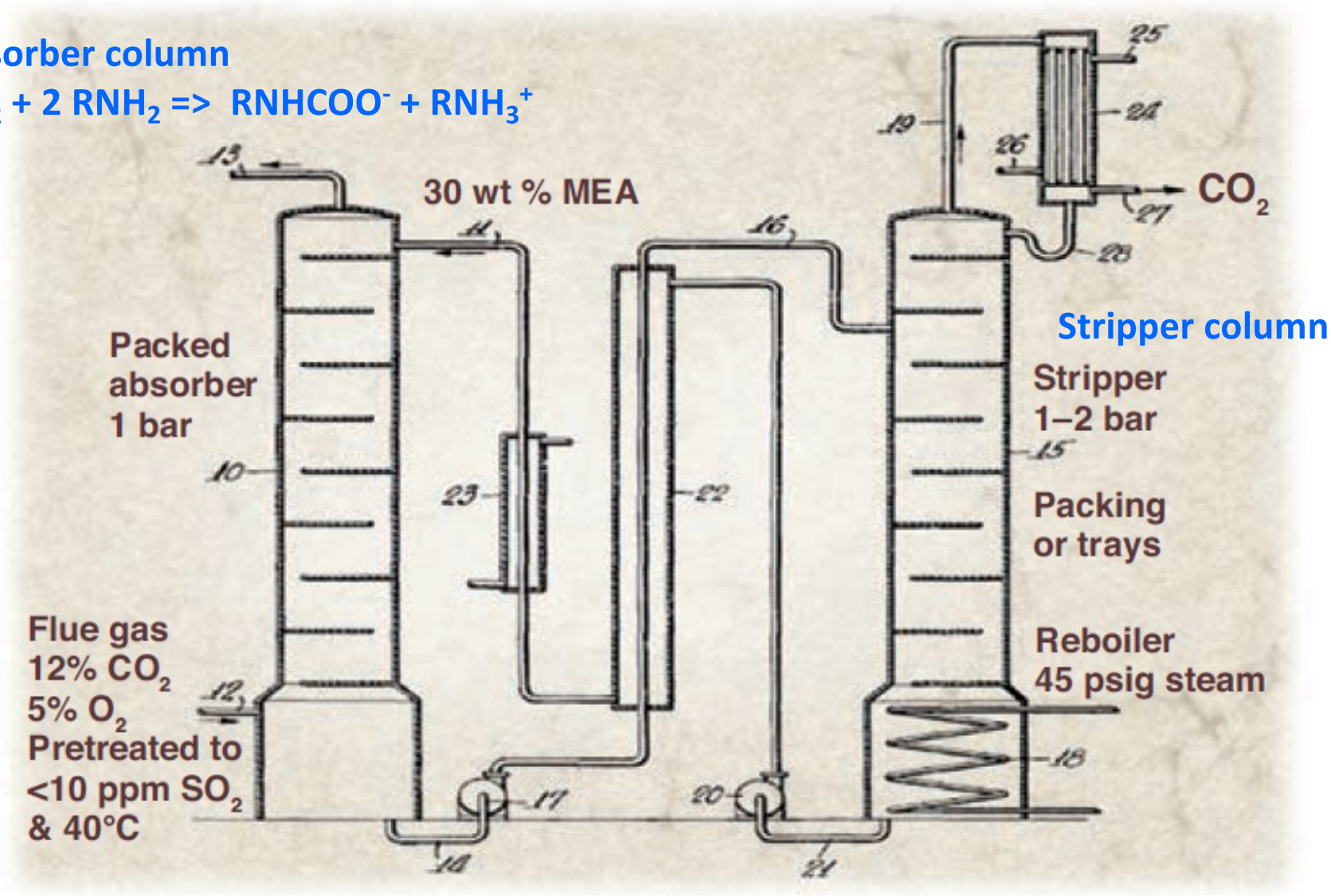
4-Amino-1-butanol 

Methylamine $CH_3\text{-}NH_2$

Carbon capture and storage technology

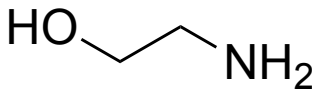
Adsorber column

$CO_2 + 2\ RNH_2 \Rightarrow RNHCOO^- + RNH_3^+$

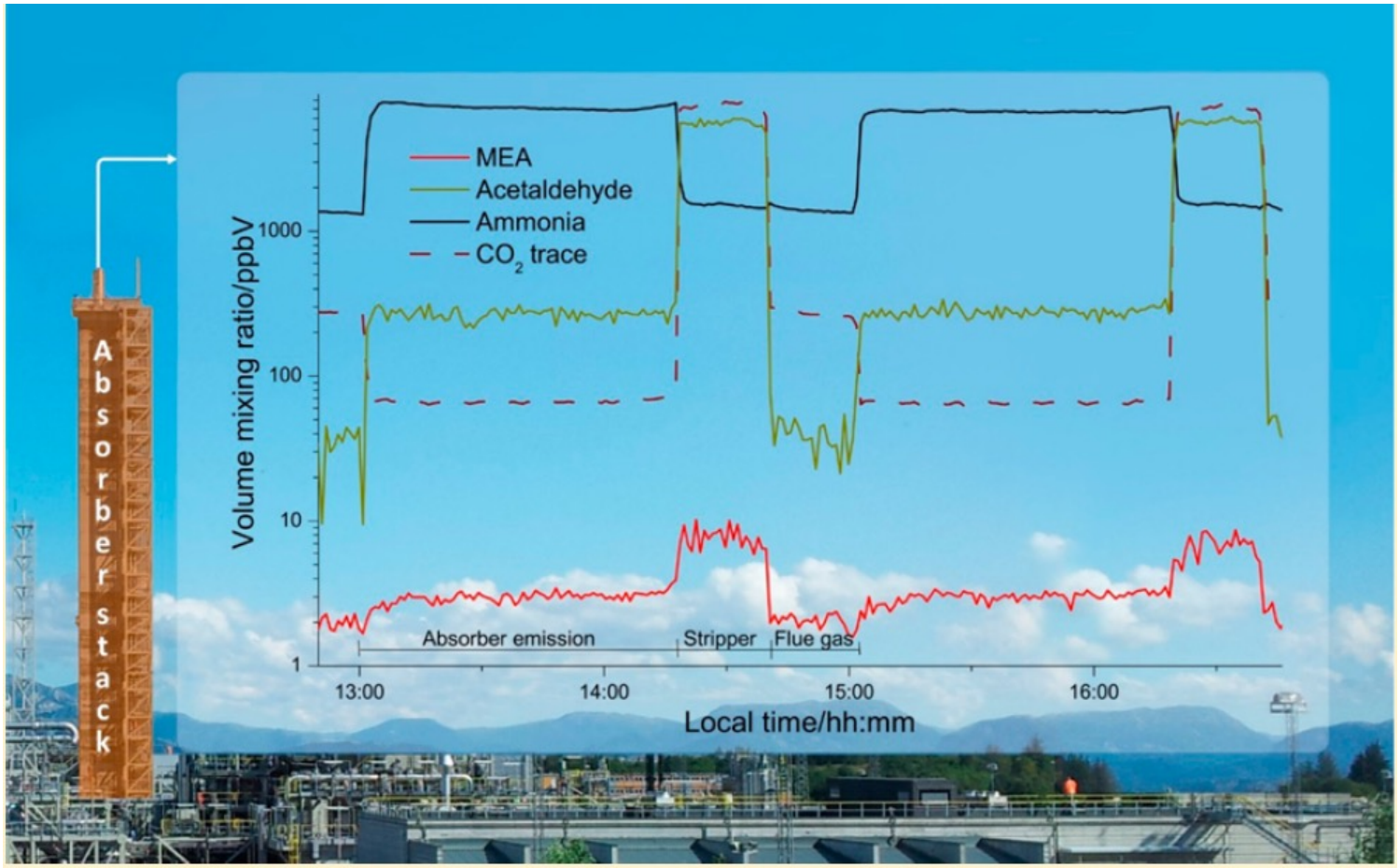


Rochelle et al. Science (2009)

Monoethanolamine (MEA)

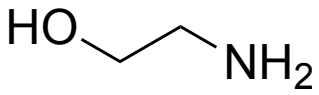


- MEA is currently the most used CCS technology and may be released into the atmosphere.
- MEA has been found in ambient particles *(Sullivan et al., 2020; Zhang et al., 2003)*

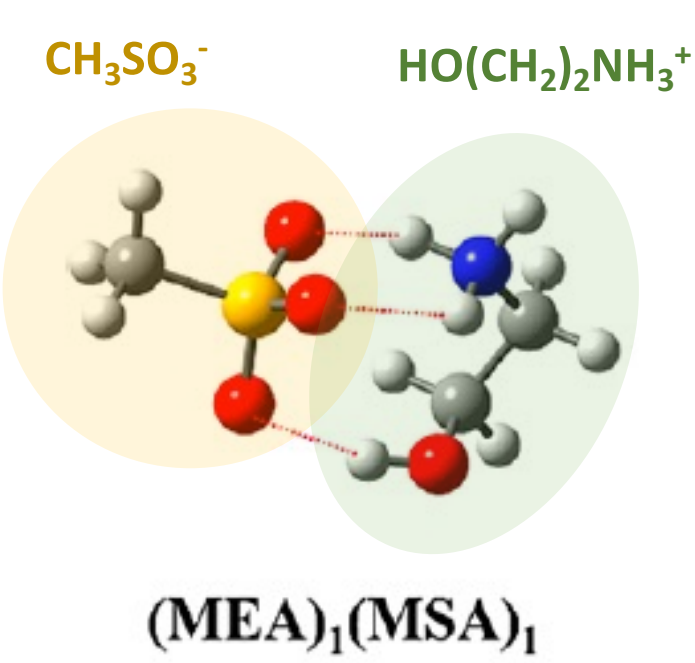


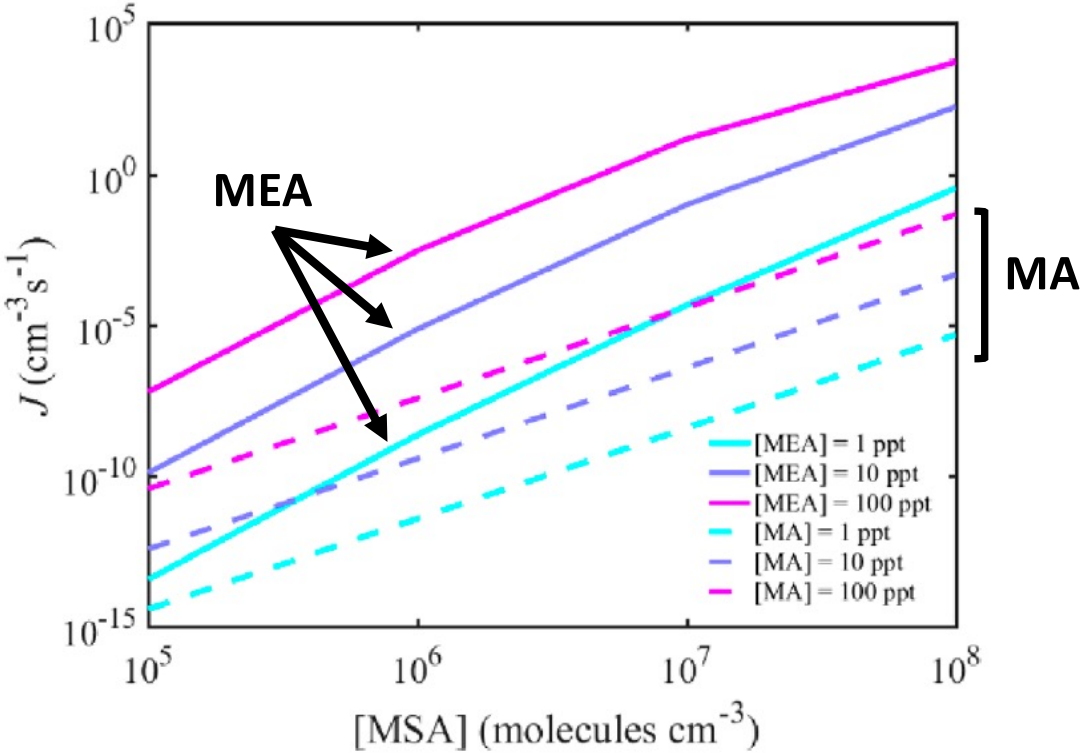
Zhu et al. EST (2013)

Monoethanolamine (MEA)



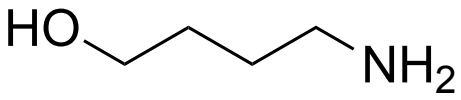
- MEA is currently the most used CCS technology and may be released into the atmosphere.
- MEA has been found in ambient particles *(Sullivan et al., 2020; Zhang et al., 2003)*
- MEA is an amine and thus will participate in acid-base reaction with strong acids in air such as H_2SO_4 *(Tian et al. 2022; Xie et al., 2017)* and MSA *(Shen et al. 2019)*.



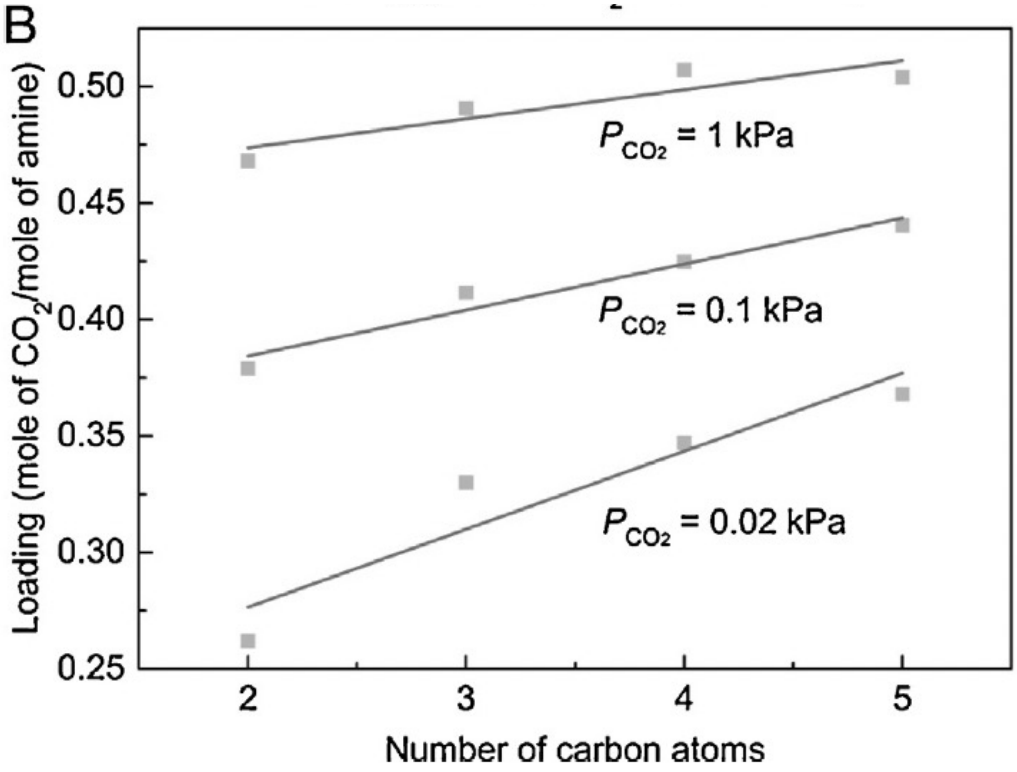


Shen et al. (2019)

4-Aminobutanol (4AB)

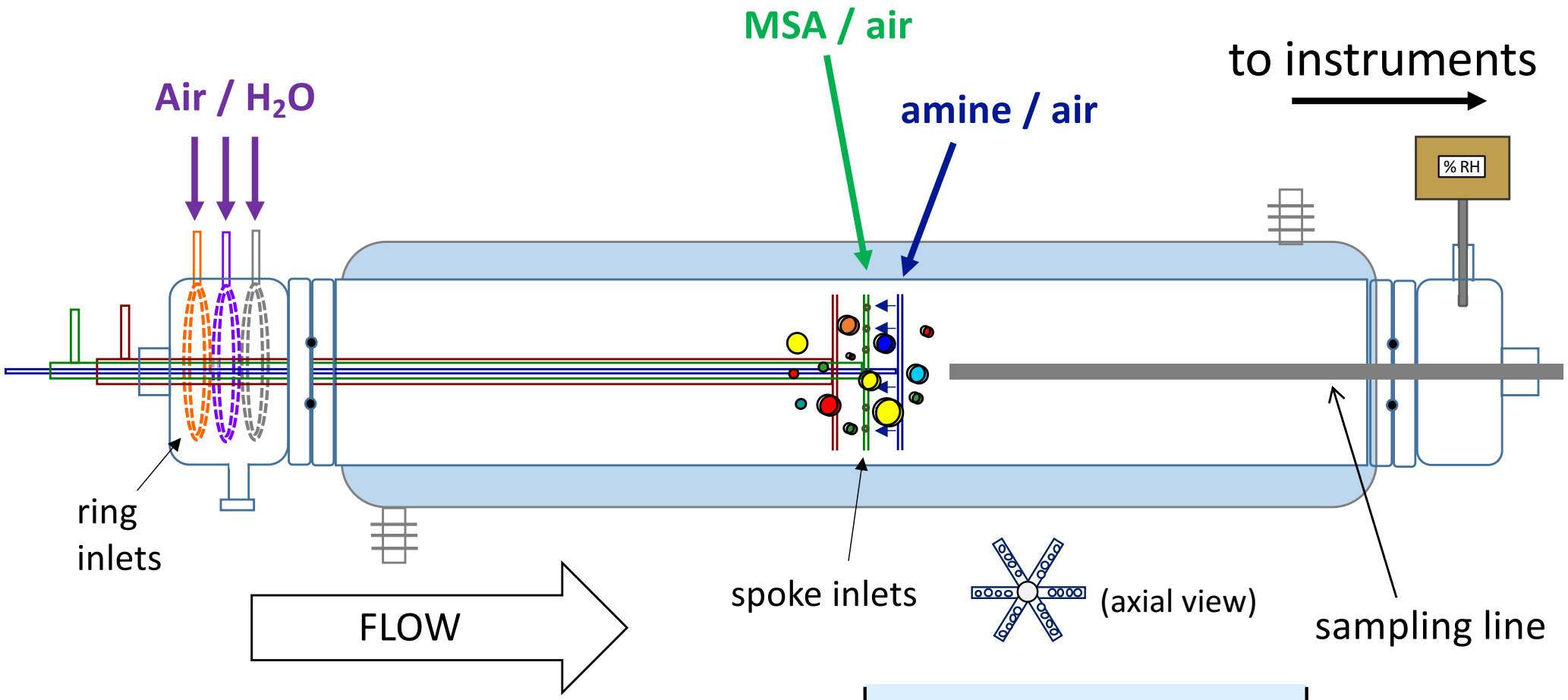


- 4AB is used in drug manufacturing and is a precursor to biodegradable polymers used for gene therapy delivery *(Prabowo et a. 2020).*
- 4AB has a higher CO_2 solubility than MEA hence may be a better CCS media *(Idris et al. 2015; Meng et al. 2022).*
- 4AB has a much higher gas phase basicity (932.1 kJ mol^{-1}) compared to MEA (896.8 kJ mol^{-1}).



(Idris et al. 2015)

Gas flow reactor studies

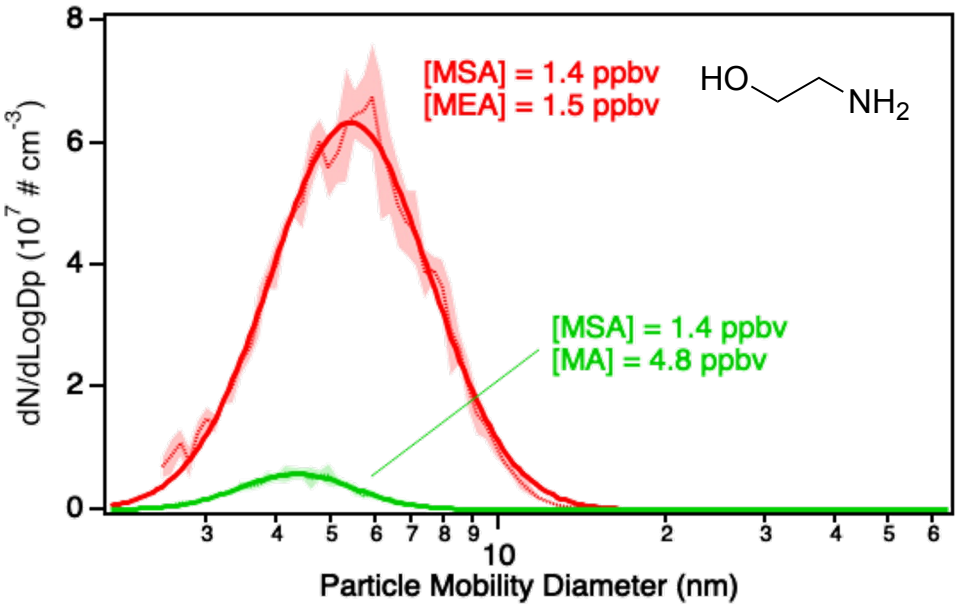


Size Distributions (SMPS)

Chemical composition (TDCIMS)

Results – MSA + alkanolamines (dry conditions)

MSA + MEA



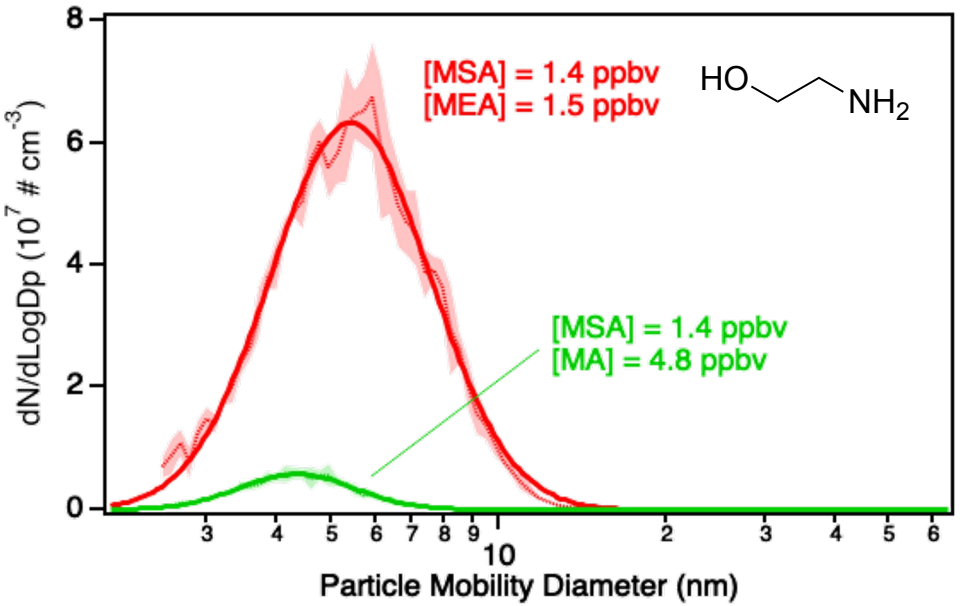
⇒ **MEA forms particles more efficiently with MSA than MA consistent with reported DFT calculations.**

⇒ **Consistent with higher gas phase basicity of MEA (896.8 kJ mol^{-1}) versus MA (864.5 kJ mol^{-1}).**

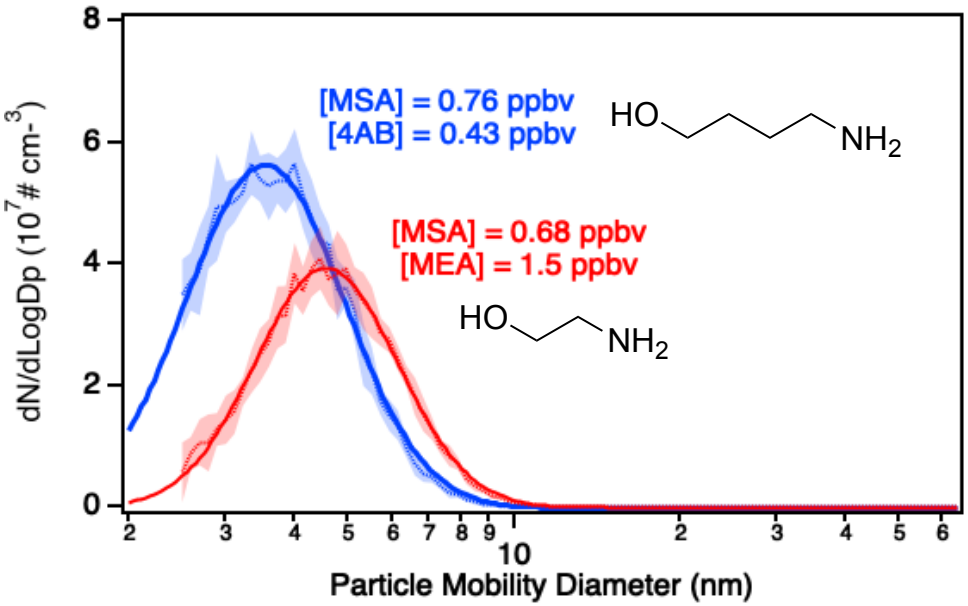
(Perraud et al. 2024)

Results – MSA + alkanolamines (dry conditions)

MSA + MEA



MSA + 4AB



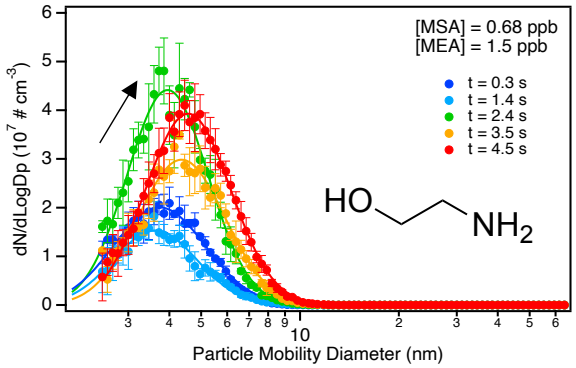
⇒ 4AB is even more efficient at nucleating particles with MSA.

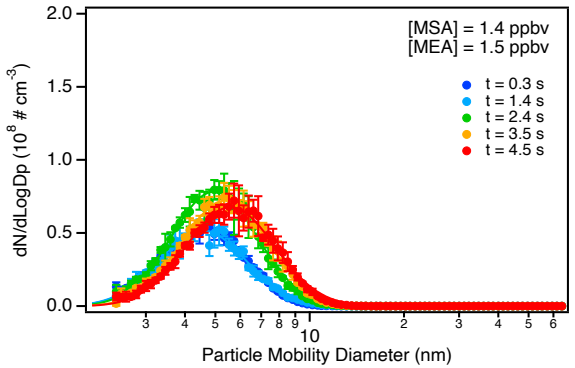
⇒ Consistent with higher gas phase basicity of 4AB (932.1 kJ mol^{-1}).

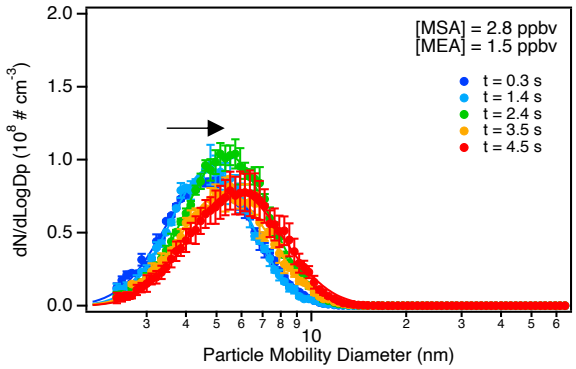
⇒ 4AB still forms detectable particles with MSA = 30 ppt.

Results – MSA + alkanolamines (dry conditions)

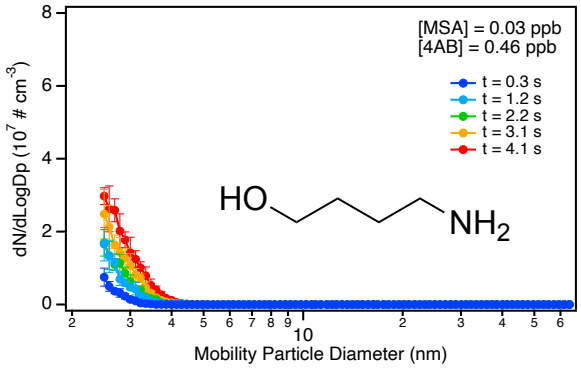
MSA + MEA

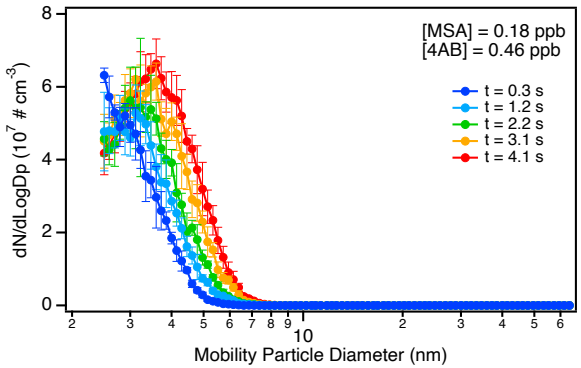


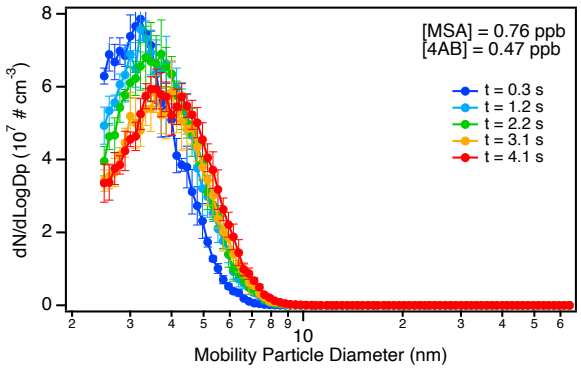




MSA + 4AB





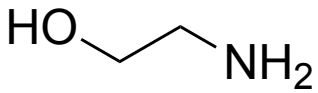


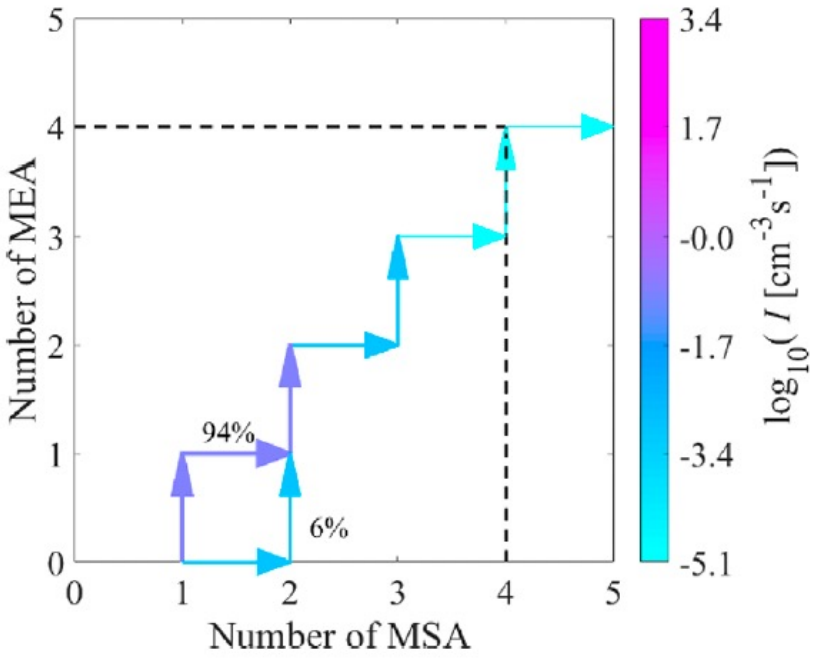
⇒ **Extremely efficient at forming particles even at very short reaction times.**

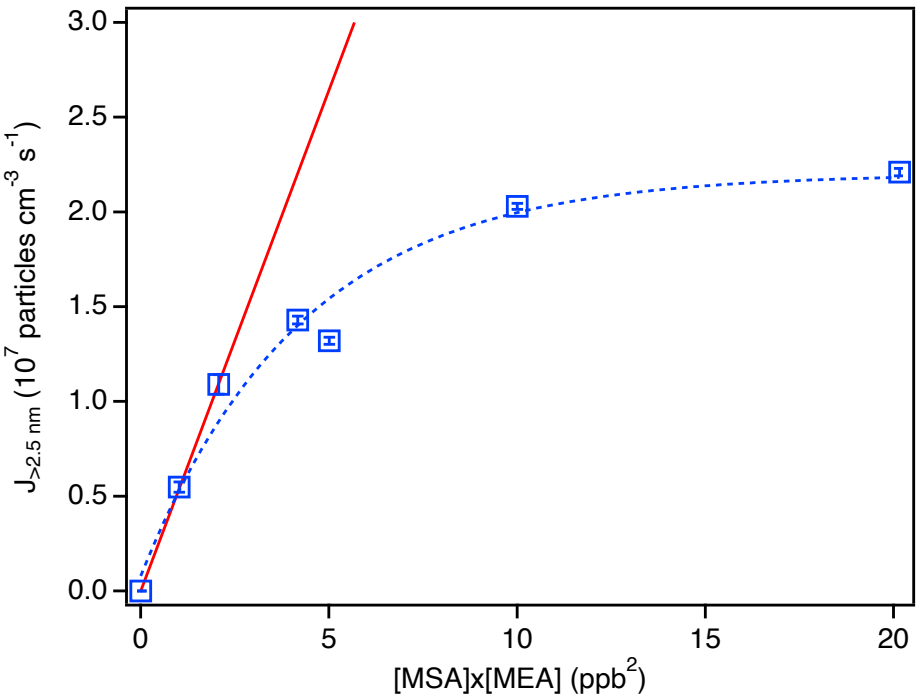
⇒ **Coagulation is observed at high concentration.**

(Perraud et al. 2024)

Results – MSA + MEA dry conditions







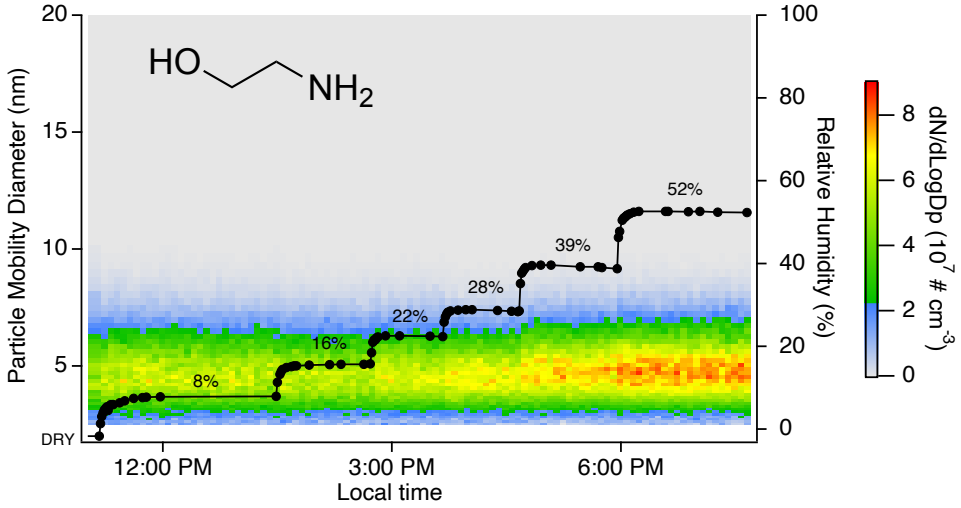
⇒ **DFT calculations suggests that the MSA_1-MEA_1 cluster is the initial step of the particle growth.**

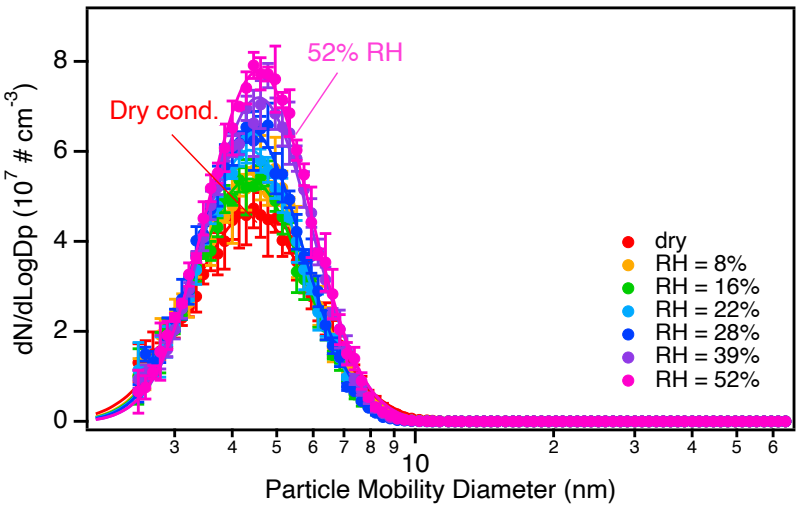
⇒ **Our kinetics plot shows that the particle formation rate follows an acid+base reaction till ~5 ppb^2; coagulation takes over at higher conc.**

(Shen et al., 2019; Perraud et al. 2024)

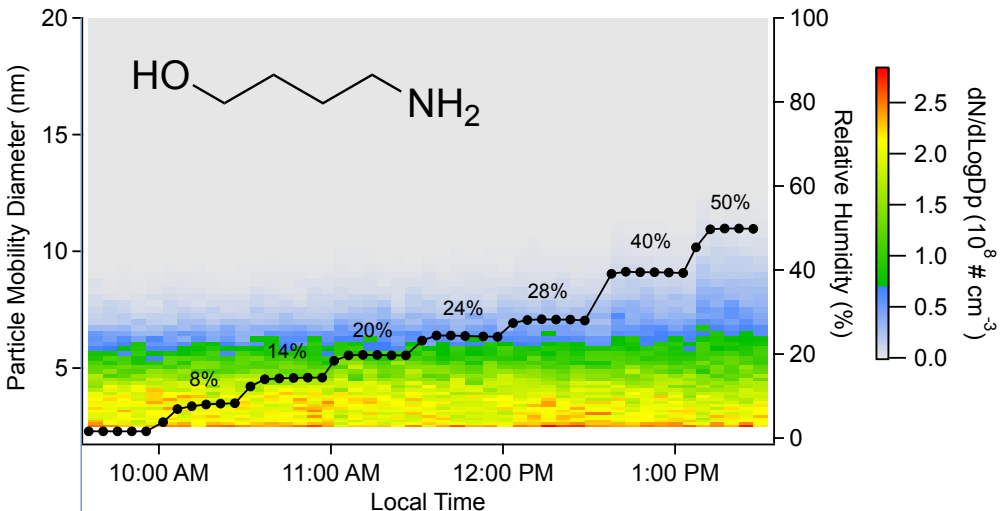
Results – MSA + alkanolamines (in presence of H_2O)

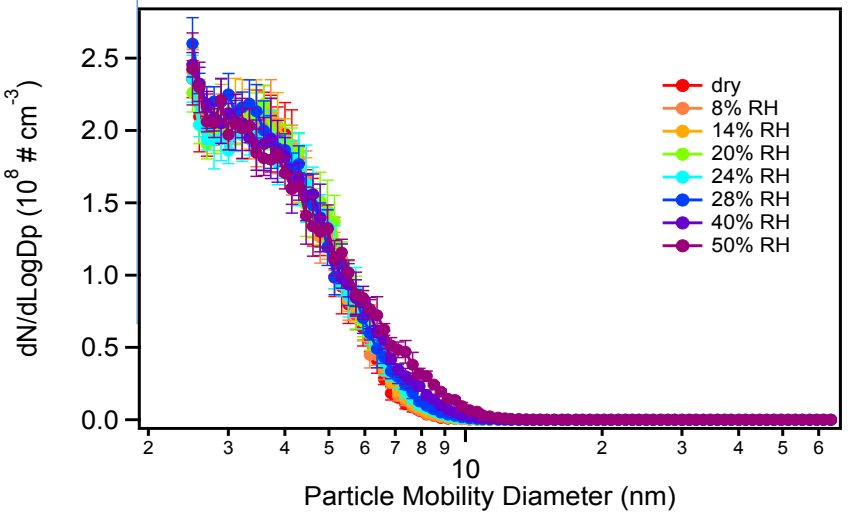
MSA (0.7 ppbv) + MEA (1.4 ppbv)





MSA (1.8 ppbv) + 4AB (0.5 ppbv)



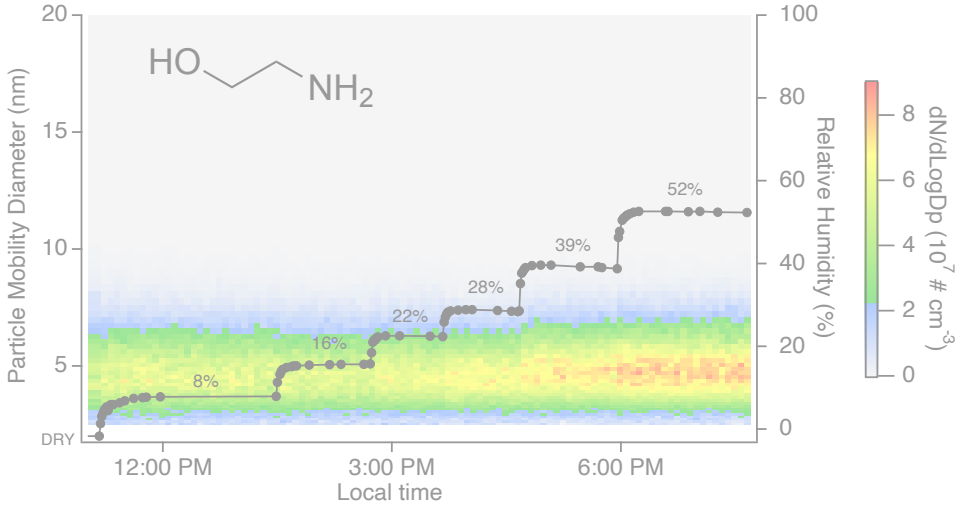


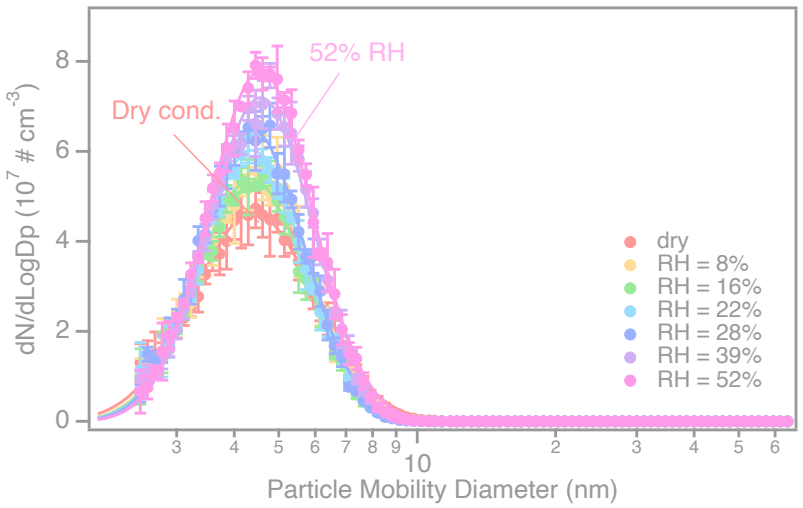
⇒ **Water has a modest effect on the MSA+MEA but has no effect on MSA+4AB.**

(Perraud et al. 2024)

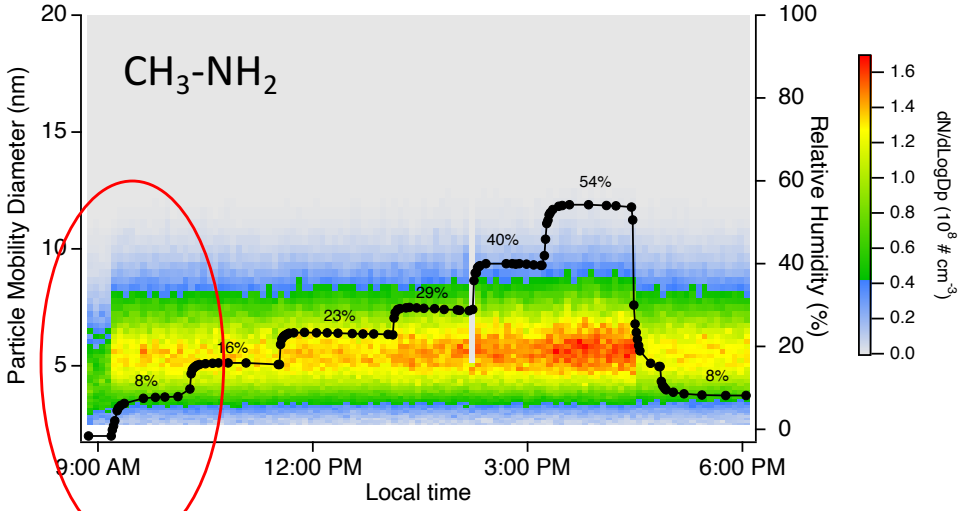
Results – Contrast with simple alkylamines

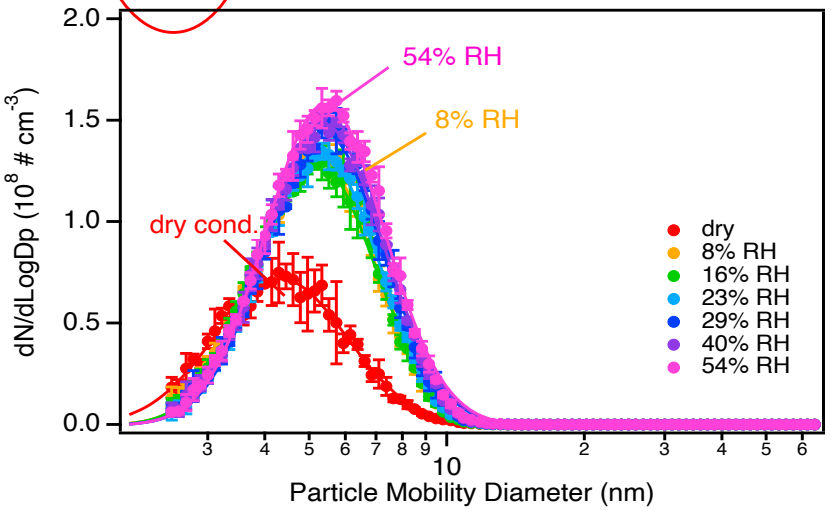
MSA (0.7 ppbv) + MEA (1.4 ppbv)





MSA (1.4 ppbv) + MA (10.8 ppbv)

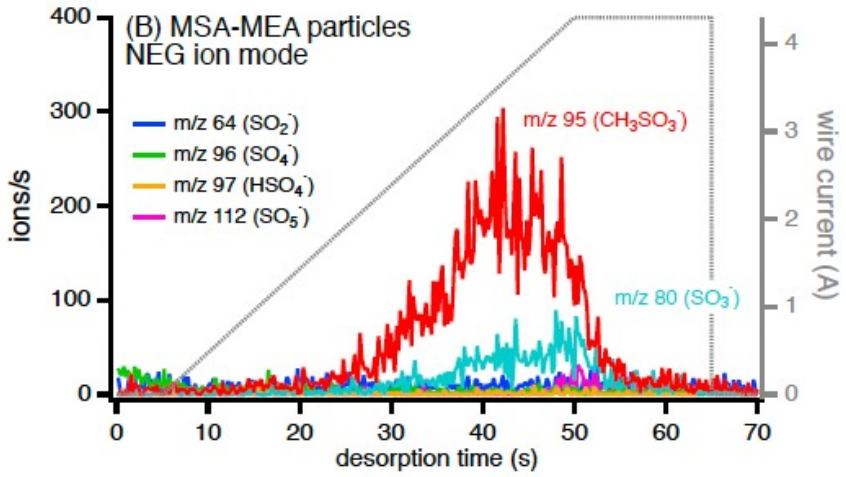


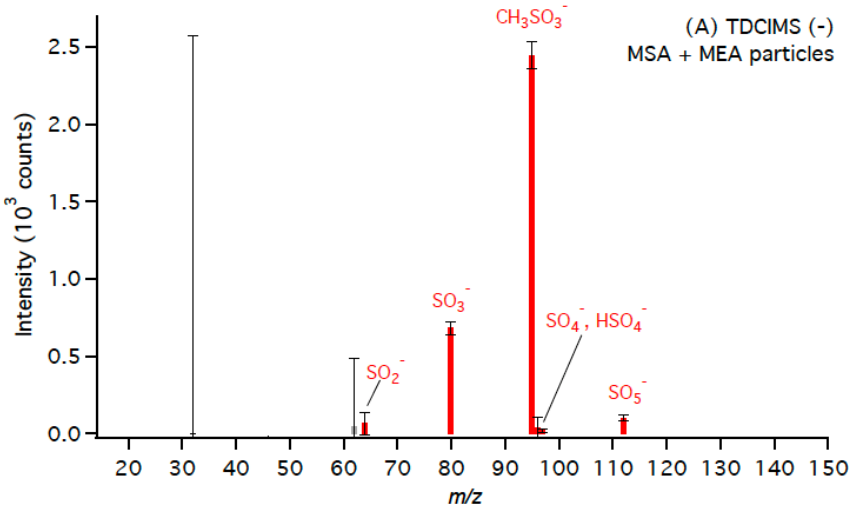


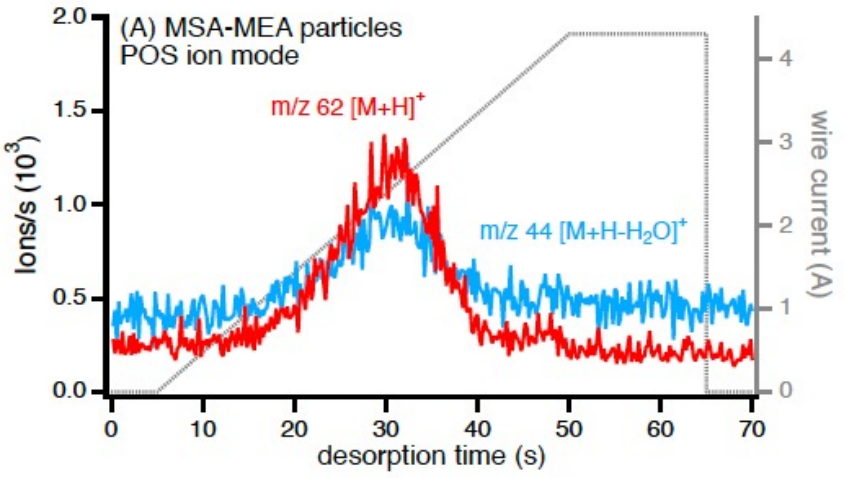
⇒ **In contrast, water has a drastic effect on simple alkylamines such as MA.**

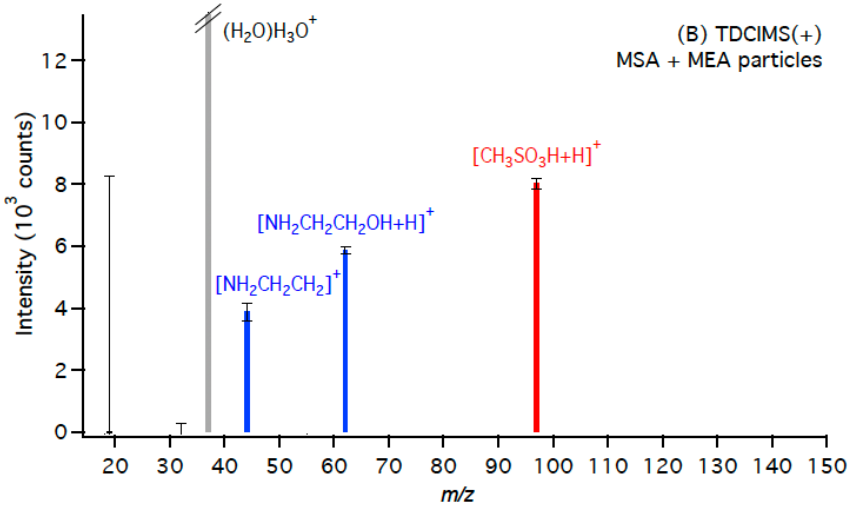
(Perraud et al. 2024)

Results – MSA + alkanolamines - TDCIMS









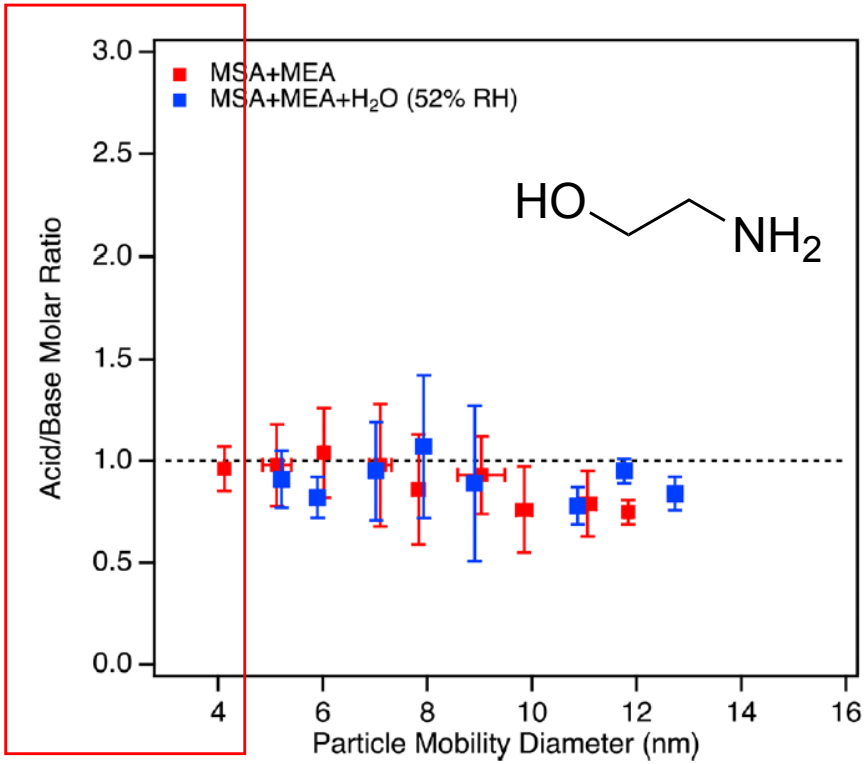
⇒ **TDCIMS measurement were possible down to 4 nm.**

⇒ **MSA is detected in NEG ion mode while MEA is detected in POS ion mode.**

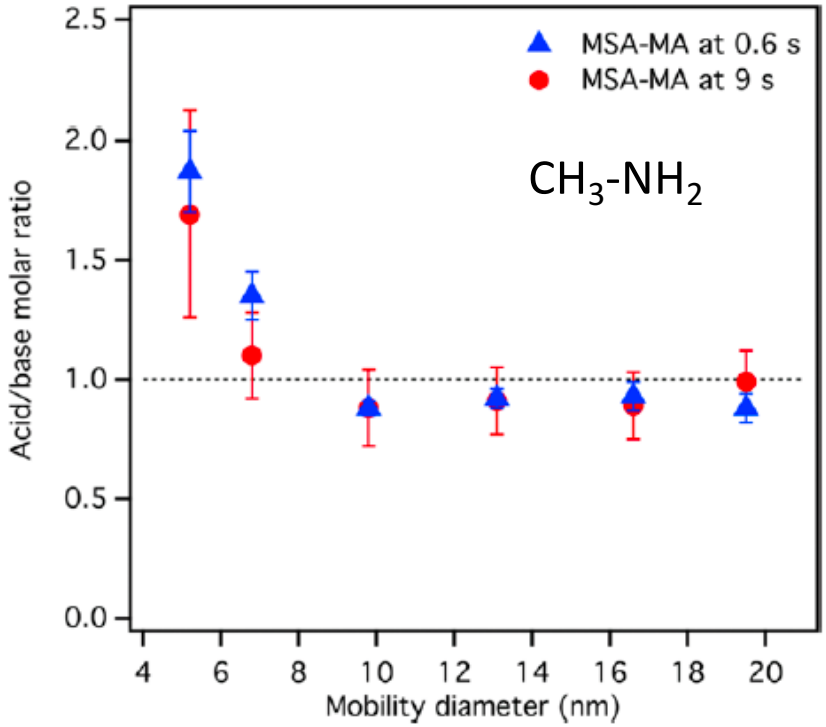
(Perraud et al. 2024)

Results – MSA + alkanolamines - TDCIMS

MSA + MEA



MSA + MA



⇒ **Particles from MSA + alkanolamines remain neutral at all diameters studied.**

⇒ **Formation and growth of the clusters is different from simple alkylamines.**

(Perraud et al. 2024; Perraud et al. 2020)

Summary

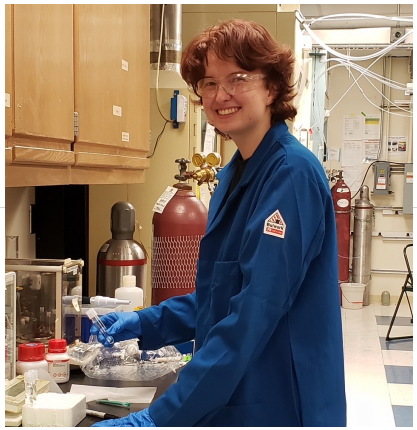
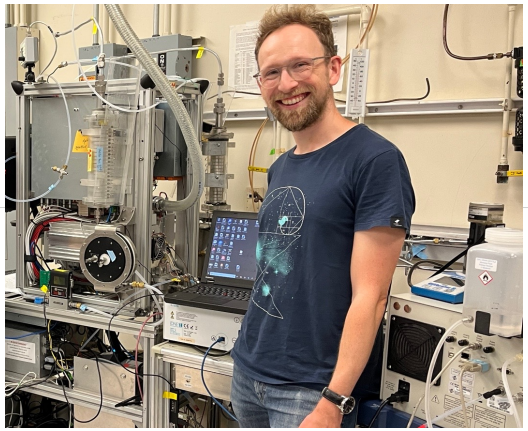
- MSA + 4AB > MSA + MEA > MSA + MA in terms of NPF potential, likely due to the extra H-bonding capabilities of the –OH and extra $–NH_2$ groups and the higher gas phase basicity of 4AB compared to MEA.

- Currently working on DFT calculations to try to explain the clusters stability & formation route in the MSA + 4AB system.

- Designed and tested a high flow DMA (half-mini-DMA; *de la Mora and Kozlowski, 2013*) to be able to scan and select diameters smaller than 4 nm to send to TDCIMS and study the cluster chemical composition at the smallest sizes.

- Future work also includes a structure study examining the NPF potential of a diamine (PUT, $NH_2(CH_2)_4NH_2$) and the C4 alkylamine butylamine.

Thanks!

The Finlayson-Pitts lab is recruiting two post-doctoral fellows for two projects

- Thermal Desorption Program (TPD) and Uptake Project
- MAIV-MS (*MAGIC*) Ionization Project