# Upper Tropospheric Particle Formation from Anthropogenic Pollution

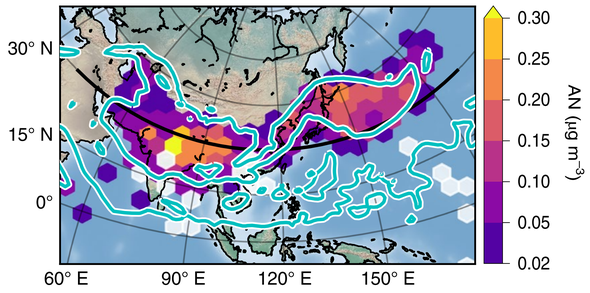
**Mingyi Wang**, Assistant Professor

*Department of the Geophysical Sciences, University of Chicago*

## 1. Motivation

### Enhanced aerosol layer over the tropics

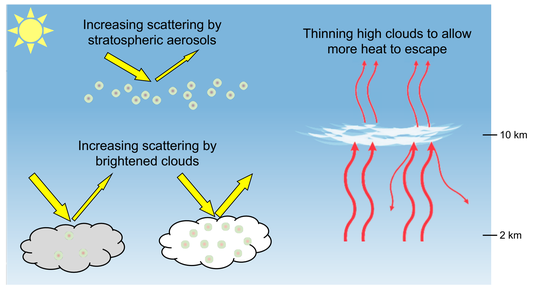
- The satellite has observed an enhanced layer of $NH_4NO_3$ particles at altitudes of 14-18 km over the Asian monsoon region;
- $NH_4NO_3$ particles may originate from the reaction between abundant $HNO_3$ from lightning and $NH_3$ from upward transport of ground level emissions.
- **Q: What mechanisms drive the consistent upper atmospheric particle formation?**




Adapted from Höpfner *et al.*, Nat. Geo. (2019)

### Microphysics driven climate impacts

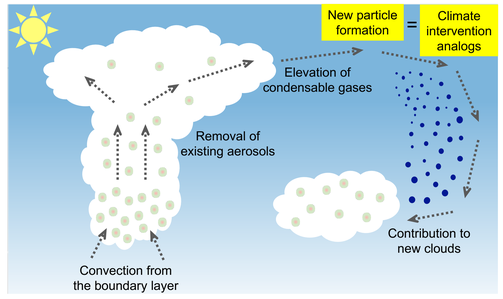
- Aerosols of a few hundred nm dia. scatter the incoming sunlight, ~ stratospheric aerosol injection (SAI);
- Aerosols > 50-100 nm dia. increase cloud albedo & lifetime, ~ marine cloud brightening (MCB);
- Solid aerosols > a few hundred nm dia. reduce homogeneous ice formation, ~ cirrus cloud thinning (CCT).




## 2. Problem setup

### Convective overshoots as natural analogs

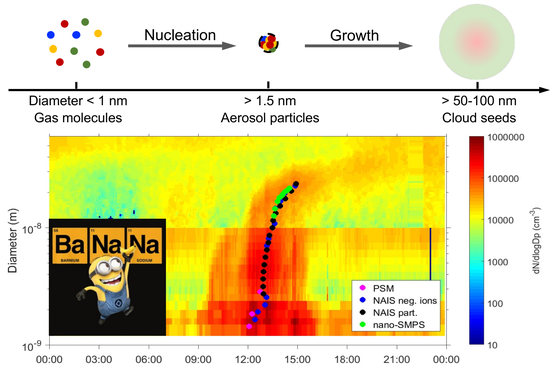
- Deep convective clouds efficiently remove pre-existing particles, reducing condensation sink…
- … and carry condensable gases aloft into the cold upper atmosphere for particle formation by entrainment or cloud glaciation;
- Many newly formed particles may grow on their descent and contribute to cloud condensation nuclei at lower levels.




Adapted from Williamson *et al.*, Nature (2019)

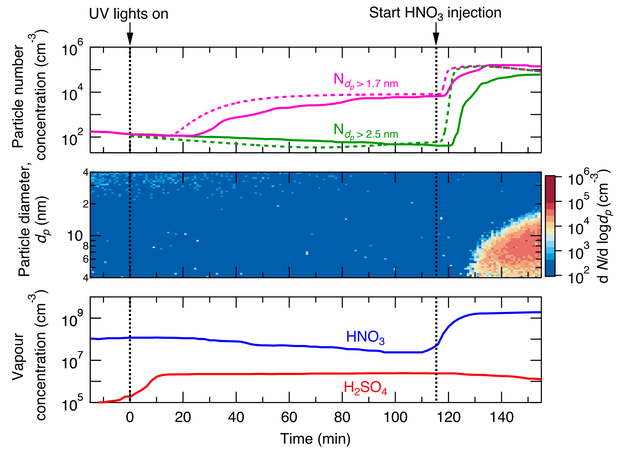
### Missing processes in models

- Aerosol nucleation: Gas-to-particle conversion can be parameterized by the first nucleation theorem $J = [X]^m * [Y]^n * [Z]^o * \cdots$;
- Aerosol growth: Saturation ratio determines the thermodynamic driving force for condensation;
- However, mechanistic understanding is still lacking at compositional, spatial, and temporal scales; ambient observations and laboratory experiments are critical.



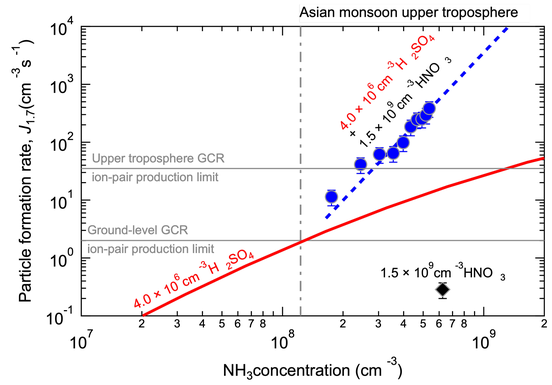

Adapted from Yao *et al.*, Science (2018)

## 3. Results and conclusions



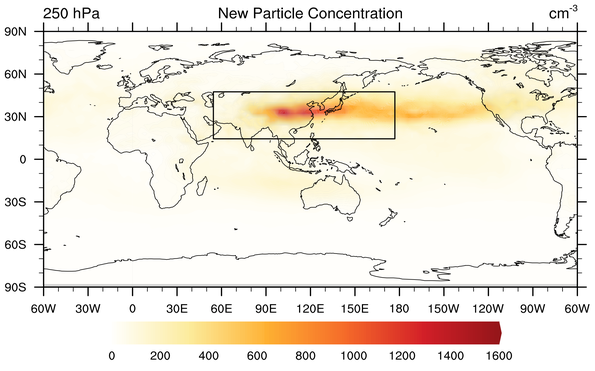

### Particle formation experiments at 223 K

- $H_2SO_4$ and $NH_3$ alone form 1.7 nm particles as expected, but these particles did not reach 2.5 nm in 2 hours due to a slow growth rate;
- Adding $HNO_3$ to the mix increases the particle number concentration by 1000 times, with much faster particle growth to 20 nm;
- Two other experiments, holding another pair of vapors constant while adding the third, show increases in particle number concentration of several orders of magnitude consistently.




### Rapid $HNO_3$-$H_2SO_4$-$NH_3$ particle formation

- At 223 K, $HNO_3$ and $NH_3$ can nucleate particles, but at a much slower rate than $H_2SO_4$ and $NH_3$;
- $HNO_3$-$H_2SO_4$-$NH_3$ nucleation is even faster than the sum of $HNO_3$-$NH_3$ and $H_2SO_4$-$NH_3$ nucleation;
- $H_2SO_4$-$NH_3$ nucleation may dominate in regions with low $NH_3$, while $HNO_3$-$H_2SO_4$-$NH_3$ nucleation may dominate at higher $NH_3$ levels (> $10^8$ cm$^{-3}$) typical of the Asian monsoon upper troposphere.



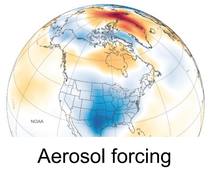

### 5x increase in regional particle population

- $HNO_3$-$H_2SO_4$-$NH_3$ nucleation increases particle population by a factor of 3-5, compared to the same model with only $H_2SO_4$-$NH_3$ nucleation;
- Although these particles are formed over Asia, they can influence cloud formation across the mid-latitude Northern Hemisphere;
- With potentially increasing $NH_3$ emissions from agriculture, this mechanism is likely to become progressively more important in the future.

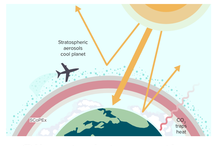
Wang *et al.*, Nature, 605, 483-489 (2022)

## 4. Broader impacts


Science-informed policymaking



Aerosol forcing prediction



Climate intervention assessment

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