Study of particle number concentrations and size distributions in the stratosphere using GEOS-Chem-APM

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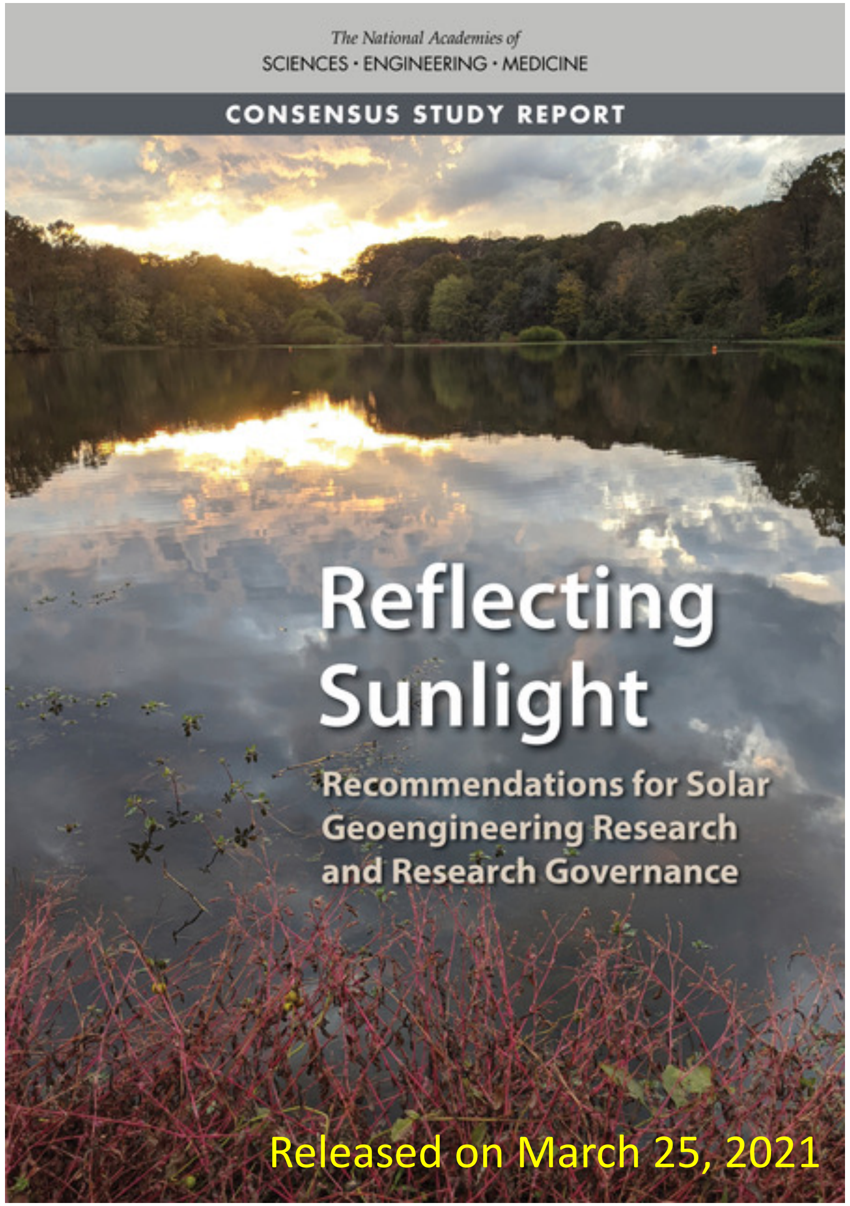
Understanding processes controlling particle size distributions in the stratosphere is important, especially for stratospheric aerosol injection (SAI)



Two quotes from a recent report by the National Academies of Sciences, Engineering and Medicine (NASEM)

> **"The overall magnitude and spatial distribution of the forcing produced by SAI depends strongly on the aerosol size distribution"**

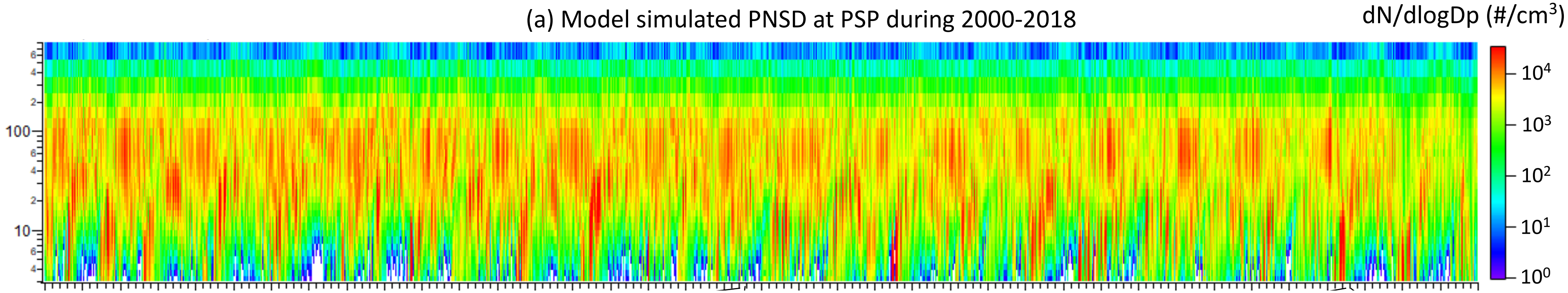
> **"One of the research priorities for SAI is thus to address critical gaps in knowledge about the evolution of the aerosol particle size distribution".**

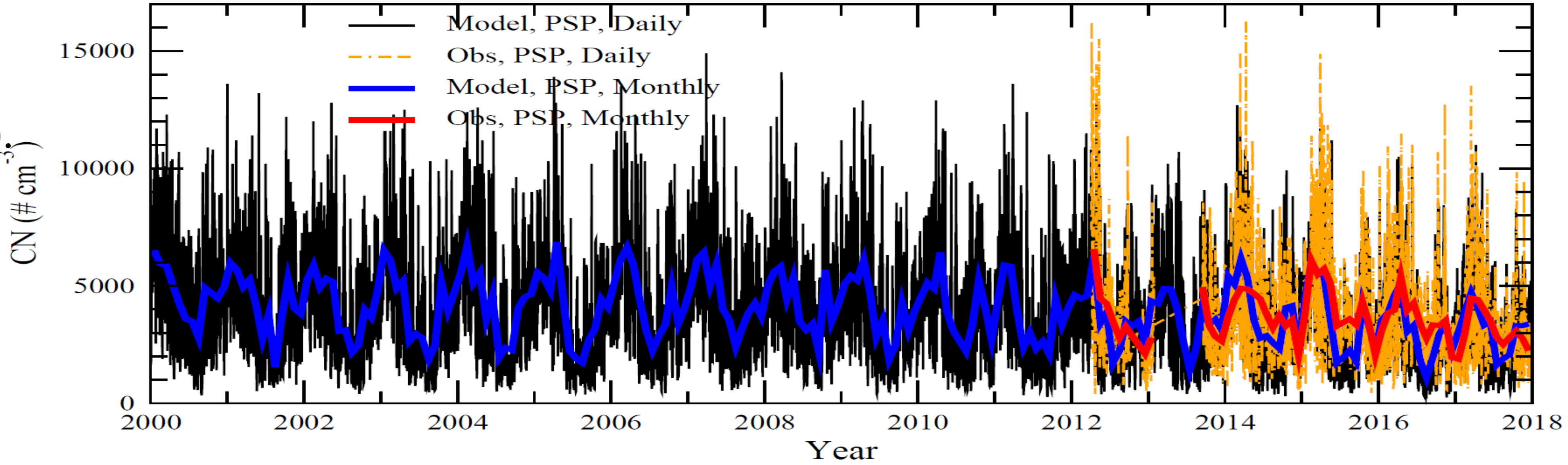
Objectives of this study:

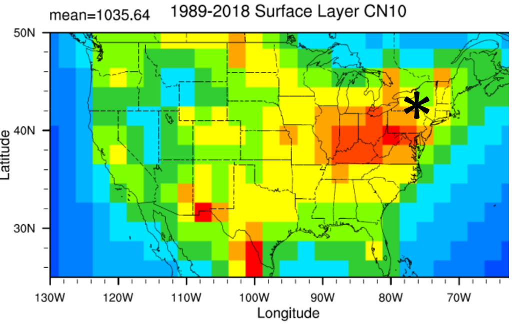
1. Integrate the size-resolved (sectional) Advanced Particle Microphysics (**APM**) package (Yu and Luo, 2009) with tropospheric–stratospheric unified chemistry extension (**UCX**; Eastham et al., 2014).
2. Employ GEOS-Chem-UCX-APM to study the processes controlling the evolution of particle size distributions (PSD) in the stratosphere under background conditions and perturbed scenarios.
3. Compare model simulations with Atmospheric Tomography Mission (**ATom**) in-situ observations of PSD down to 3 nm (Williamson et al., 2019, 2021)

Examples of APM simulation of PSDs in the troposphere

$2^o \times 2.5^o$, driven by MERRA2;
Simulation period: 1989-2018 (30 years).

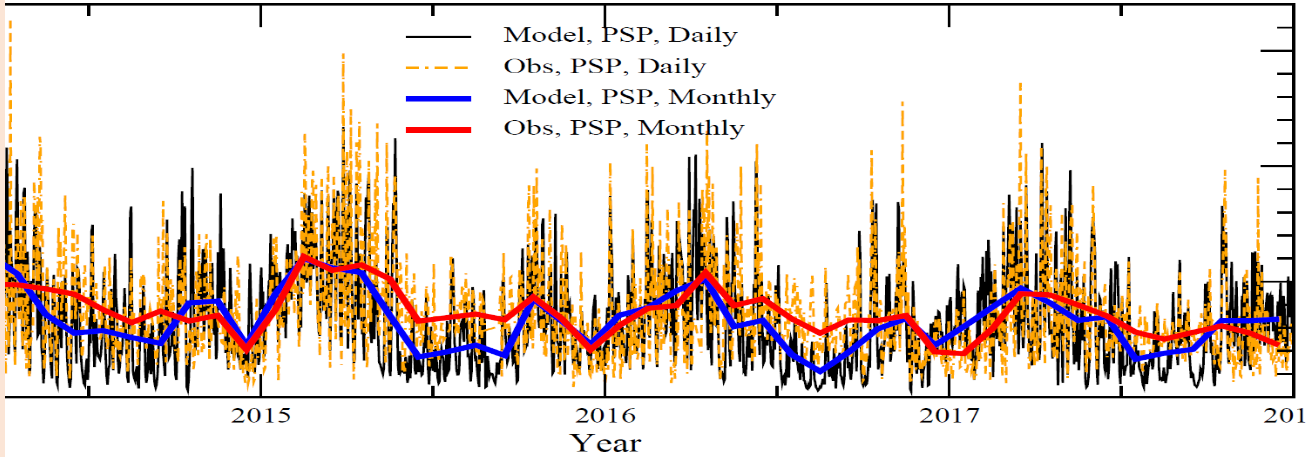




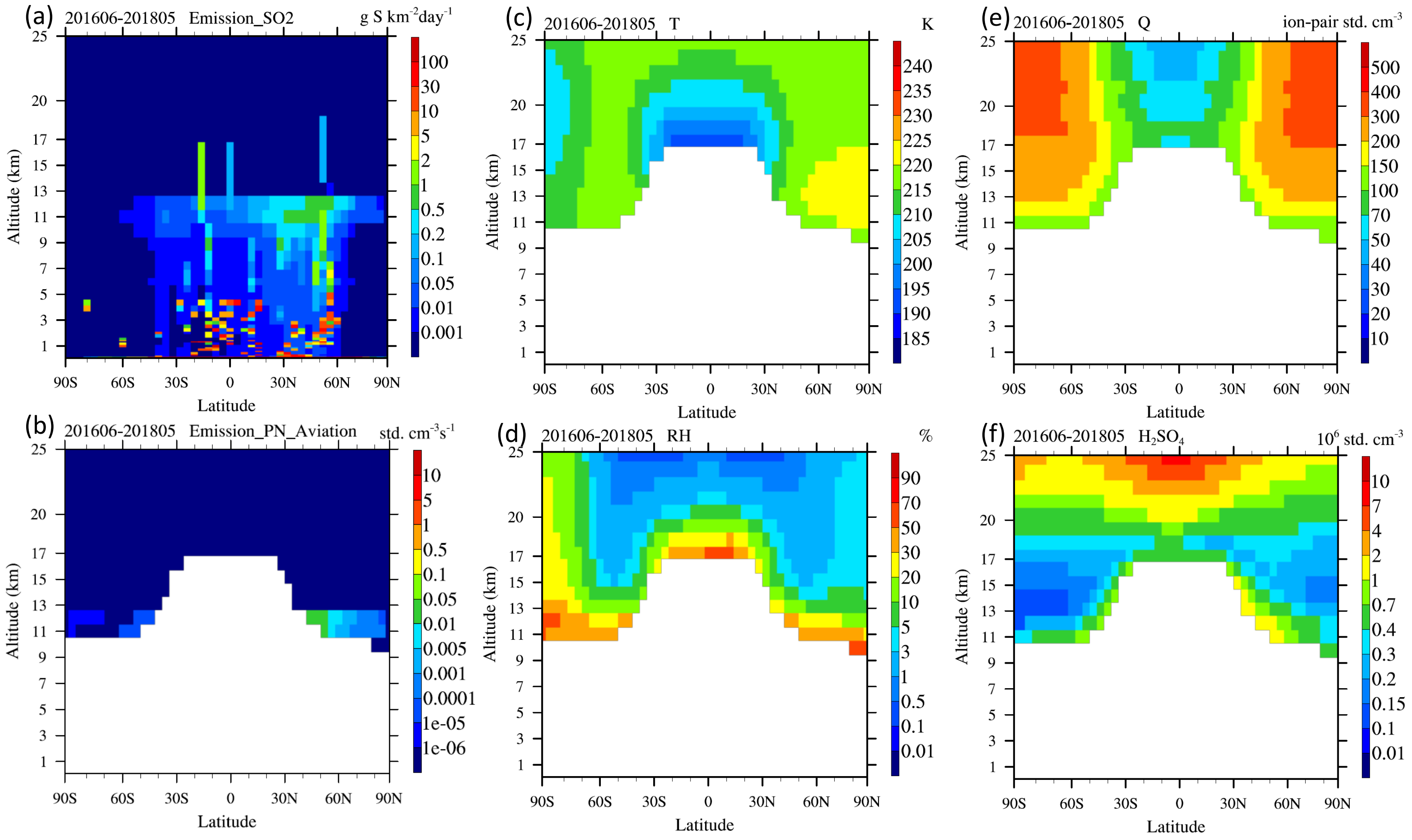


Outputs of GEOS-Chem-APM 30-year simulations have been used in machine learning studies to derive key aerosol properties (CCN, CN, surface area, extinction coefficients, etc.) for models with bulk aerosol scheme.

Thursday 10:50 am talk (Arshad Nair) :
GEOS-Chem-APM for physics-informed machine learning emulators and parameterizations



GEOS-Chem-UCX-APM simulation of PSDs in the stratosphere



Zonally mean SO_2 emission rate (a), direct emission of particle number due to aviation (b), temperature (c), relative humidity (d), cosmic ray ionization rate (e), and $[H_2SO_4]$ (f) averaged during a two-year periods (201606-201805) covering Atom 1-4. To focus on lower stratosphere, only the values of these variables (except for SO_2_emit) in the stratosphere (grid boxes with more than 50% time above tropopause) are shown.

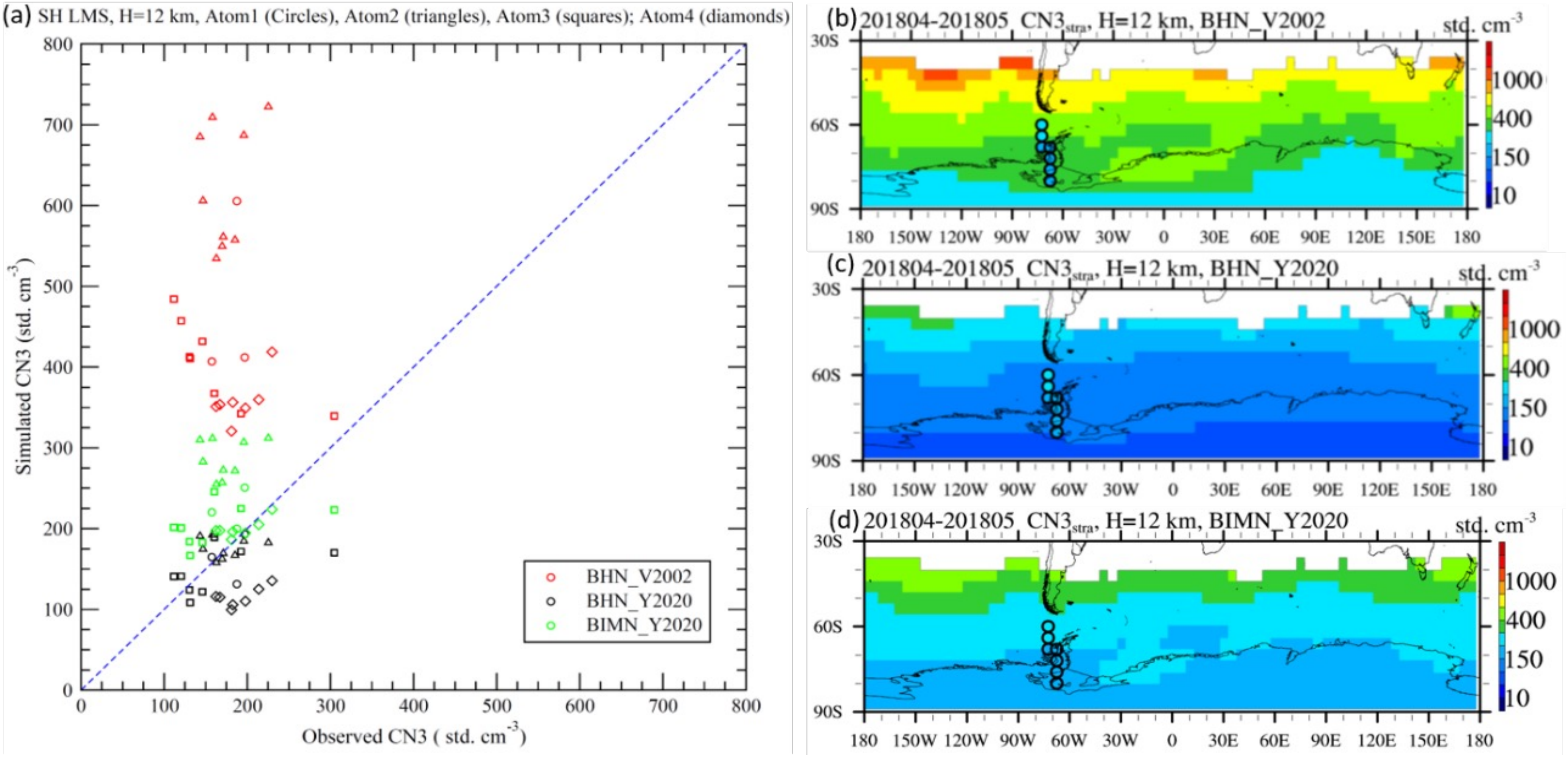
Nucleation schemes considered

(1) H_2SO_4–H_2O binary homogenous nucleation (BHN) scheme by Vehkamäki et al. (2002) (**BHN_V2002**),

(2) BHN of Yu et al. (2020) (**BHN_Y2020**), and

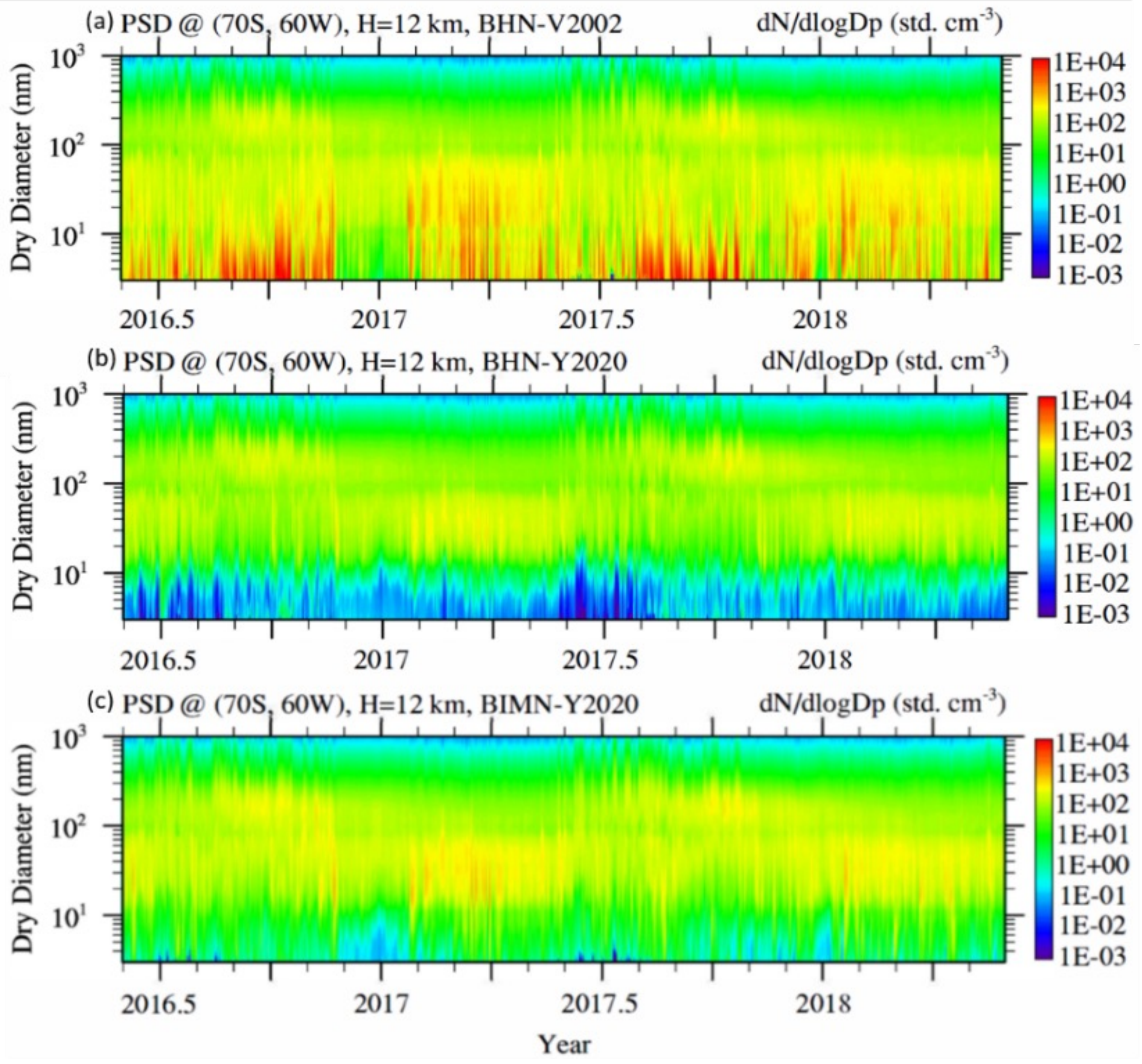
(3) Binary ion-mediated nucleation of Yu et al. (2020) (**BIMN_Y2020**)

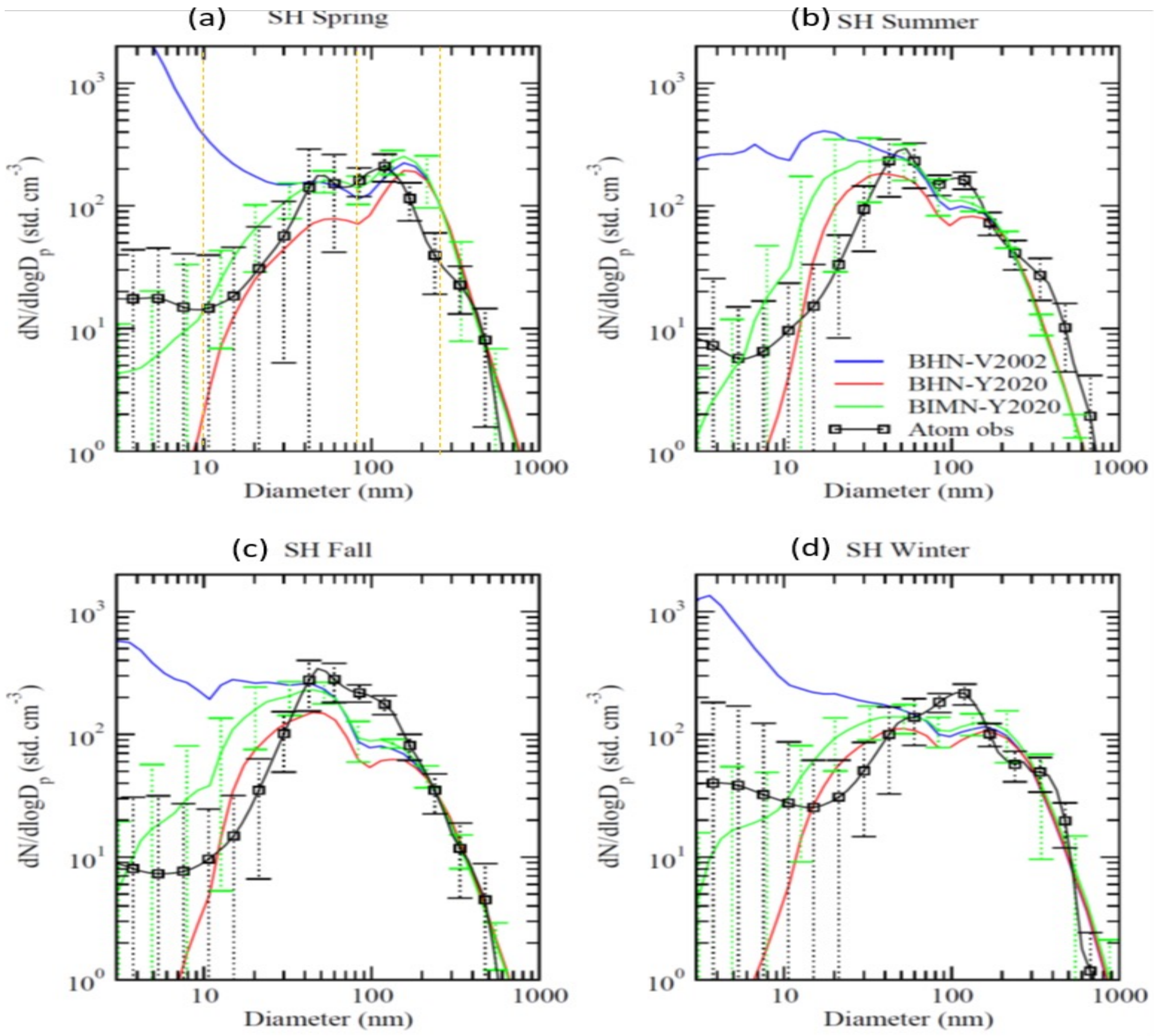
Comparisons with ATom measurements of condensation nuclei >3nm (CN3)



BHN_V2002 scheme overpredicted particle number concentrations in the background stratosphere by a factor ~2–4.

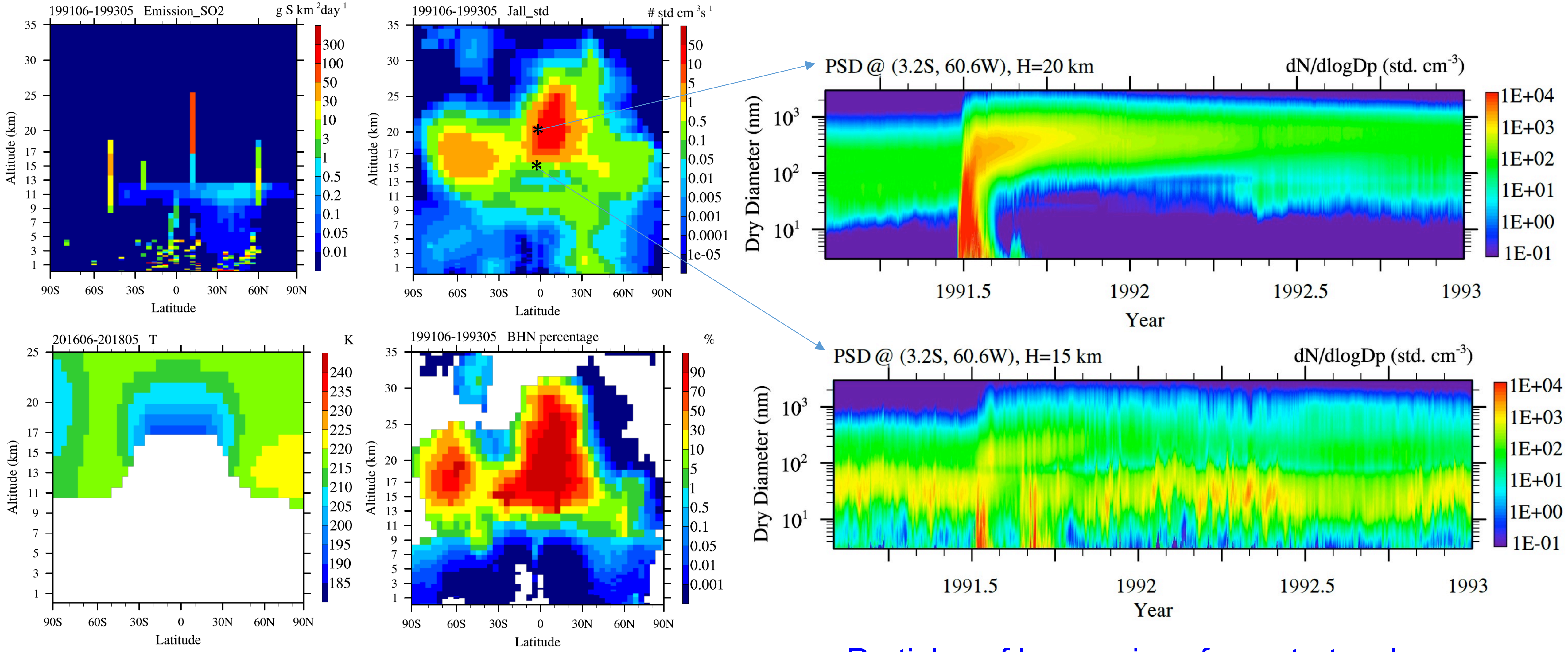
Comparisons with ATom PSD and implications





The model captures reasonably well the two modes (Aitken mode and the first accumulation mode) with the highest number concentrations and the size-dependent standard deviations. However, the model misses an apparent second accumulation mode peaking around 300–400 nm.

Pinatubo Volcano: Nucleation, PSD evolution, and Impact on Troposphere



In the regions affected by volcano plumes, both homogenous and ion nucleation can be important. The relative importance depends on injection location and strength.

Particles of larges sizes from stratosphere affect those in the upper troposphere.

Summary

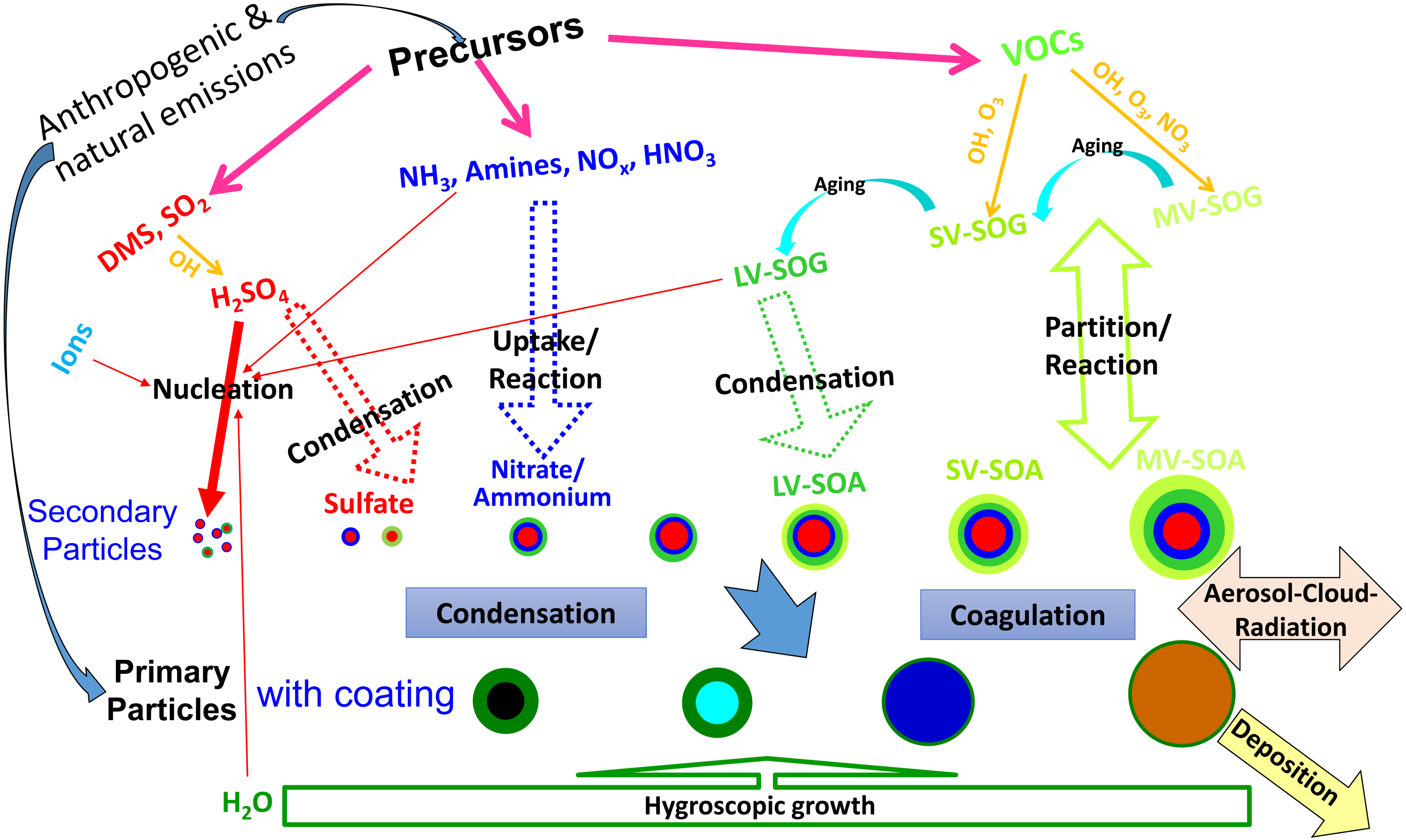
Questions? Please contact fyu@albany.edu

- APM has been integrated with UCX and the resulting GEOS-Chem-UCX-APM has been used to study the evolution of particle size distributions in the stratosphere
- Nucleation schemes have a strong effect on the model simulated particle number concentration and size distribution in the stratosphere. Both H_2SO_4-H_2O binary homogeneous nucleation (BHN) and binary ion-mediated nucleation (BIMN) occur in the stratosphere. BIMN rates are generally more than one order of magnitude higher than BHN rates and thus dominate nucleation in the background stratosphere. However, BHN can become dominant in volcano plumes, depending on locations and injection strength.
- Compared to ATom PSD measurements, GEOS-Chem-UCX/APM model misses an apparent second accumulation mode peaking around 300–400 nm. The possible reasons of the difference remain to be investigated.
- Pinatubo volcano eruption significantly enhances concentration of large particles in the upper troposphere. The impact of volcano eruptions and potential SAI on CCN and IN in the upper troposphere needs to be studied.

GEOS-Chem with APM

Size-resolved advanced particle microphysics (APM) model

(Yu and Luo, ACP, 2009)



New particle formation or nucleation in the stratosphere

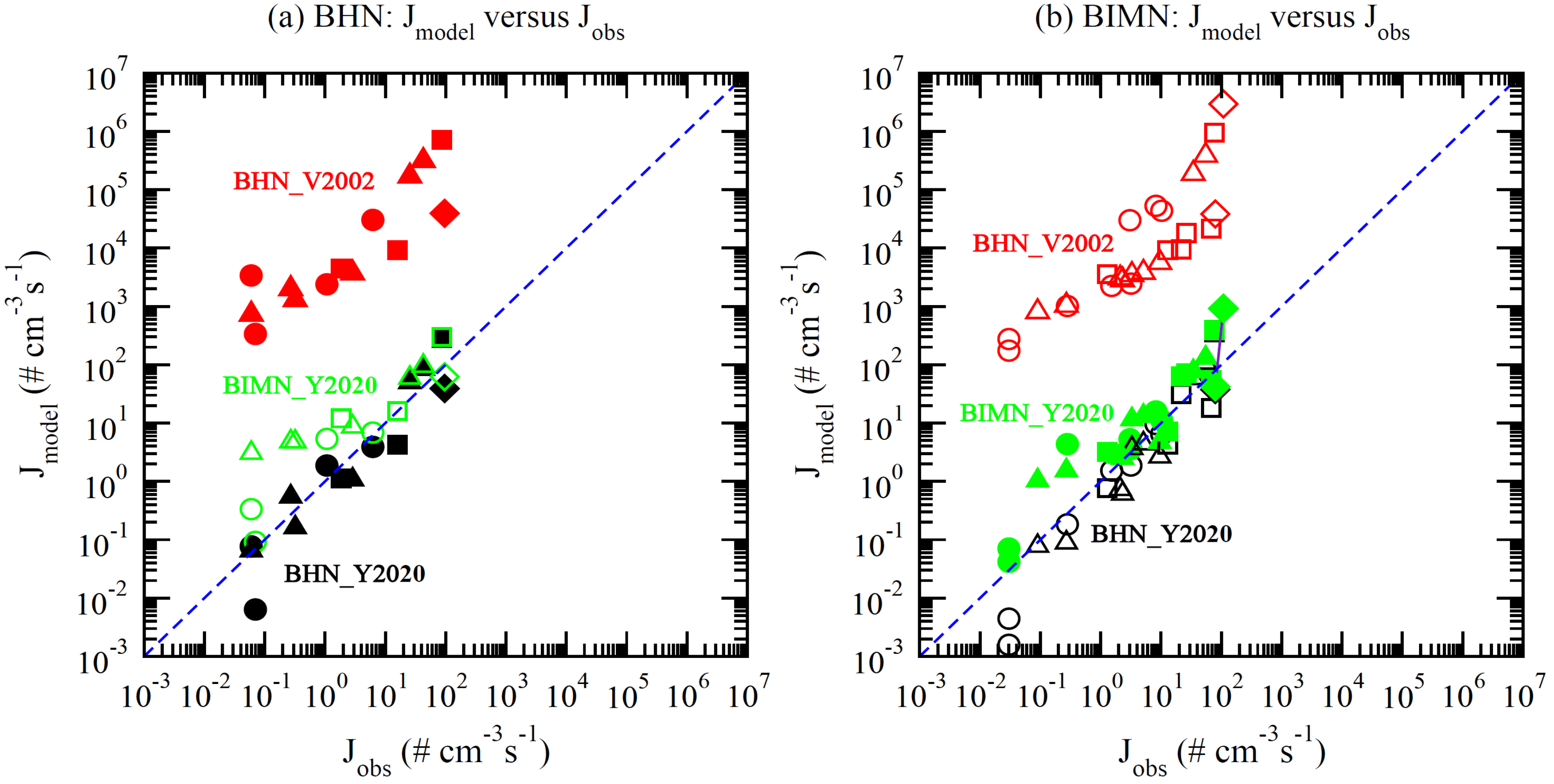
(1) H_2SO_4–H_2O binary homogenous nucleation (BHN) scheme by Vehkamäki et al. (2002) (**BHN_V2002**),
(2) BHN of Yu et al. (2020) (**BHN_Y2020**), and
(3) binary ion-mediated nucleation of Yu et al. (2020) (**BIMN_Y2020**)

Scientific questions to be addressed:

(1) When compared to CLOUD, BHN_V2002 overpredicts nucleation rates by 3-4 orders of magnitude (Yu et al., 2020). What's the implication to simulated PSD?

(2) Ionization rate is high in stratosphere, what's the relative importance of BHN versus binary ion-mediated nucleation (BIMN)

Comparison of nucleation rates (J_{model}) based on three different schemes with CLOUD measurements (J_{model}) under low T (205–223 K)



BHN of Vehkamäki et al. (2002) (**BHN_V2002**), BHN of Yu et al. (2020) (**BHN_Y2020**), and BIMN of Yu et al. (2020) (**BIMN_Y2020**).