Global Modeling of Nucleation and Growth: TOMAS Status

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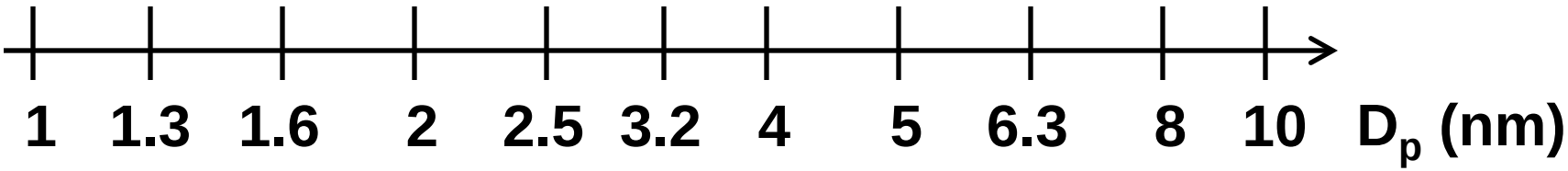
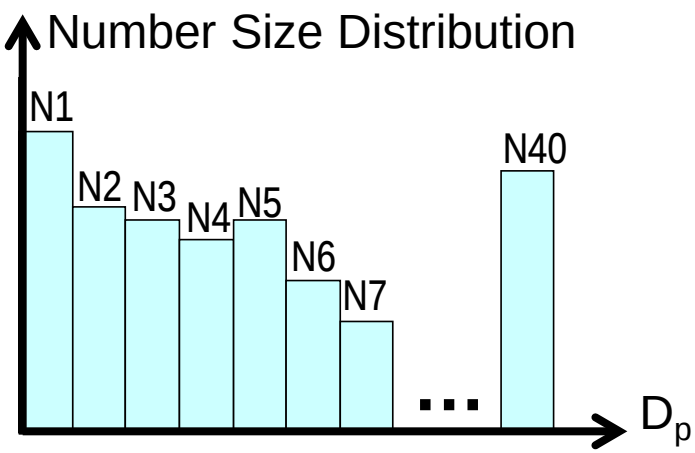
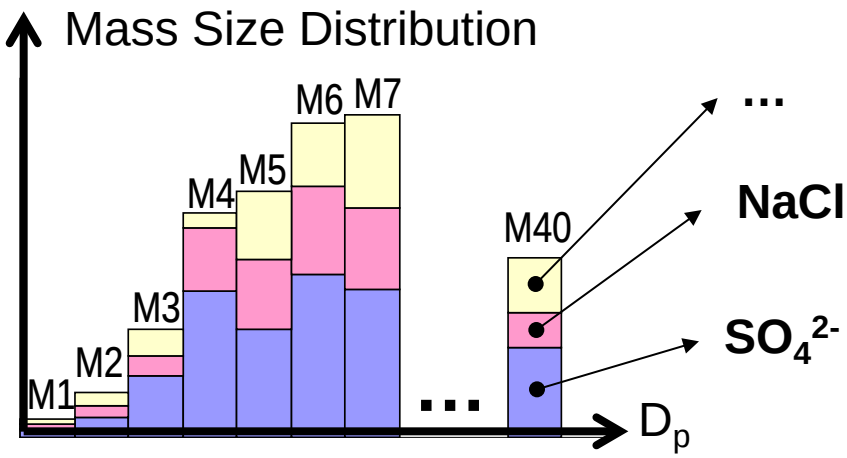
TOMAS Overview

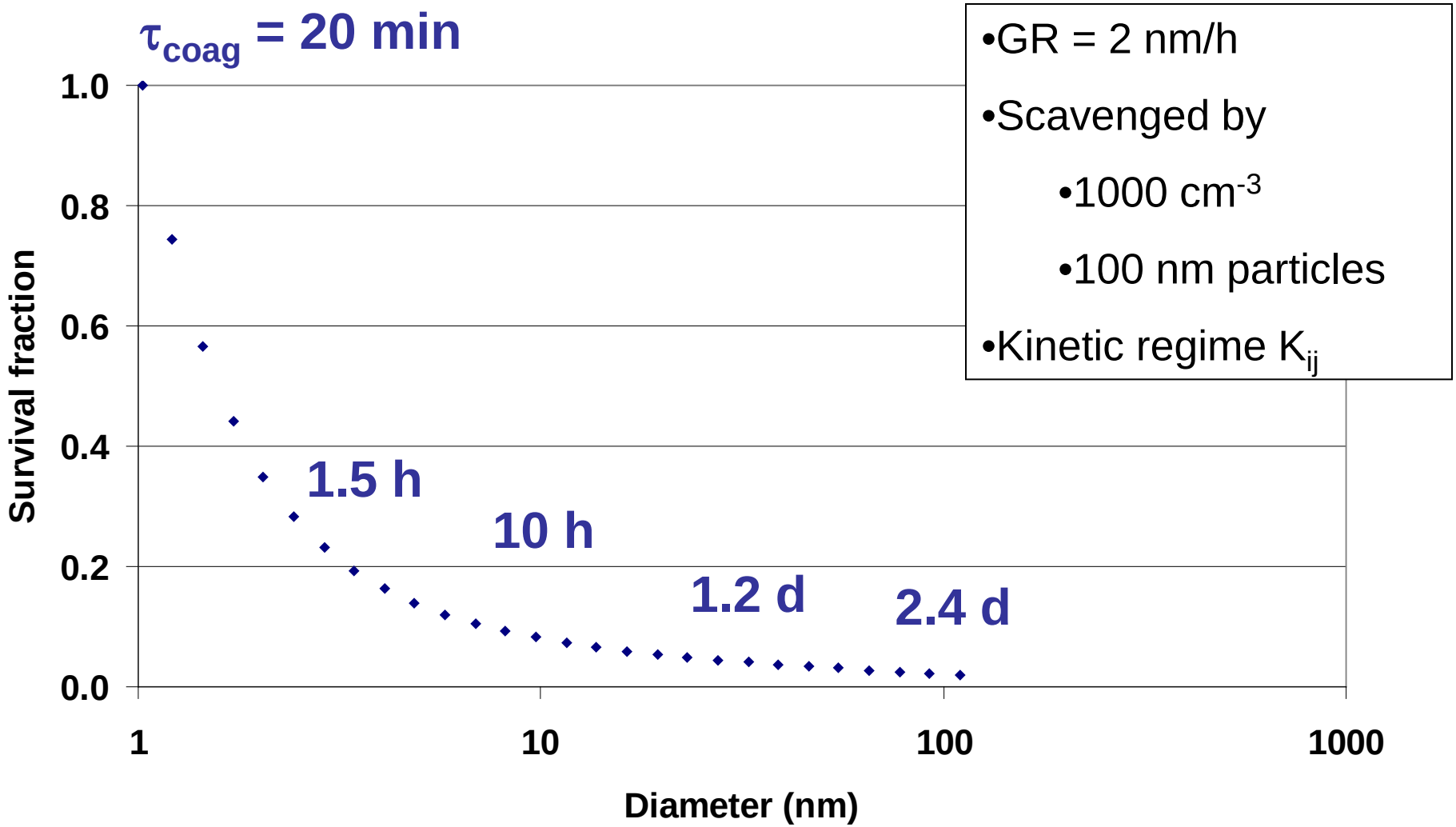
- TwO-Moment Aerosol Sectional algorithm
  - Solves general dynamic equation
  - Moments = 1) aerosol number and 2) aerosol mass
  - Size range usually 1 nm – 10 μm
  - 40 size sections (10 per decade of diameter)







Survival and Growth



- GR = 2 nm/h
- Scavenged by
  - 1000 cm^{-3}
  - 100 nm particles
- Kinetic regime K_{ij}

- Survival: 2% (1 nm to 100 nm); 45% (30 nm to 100 nm)
- Size resolution matters in modeling CCN formation

Model Status

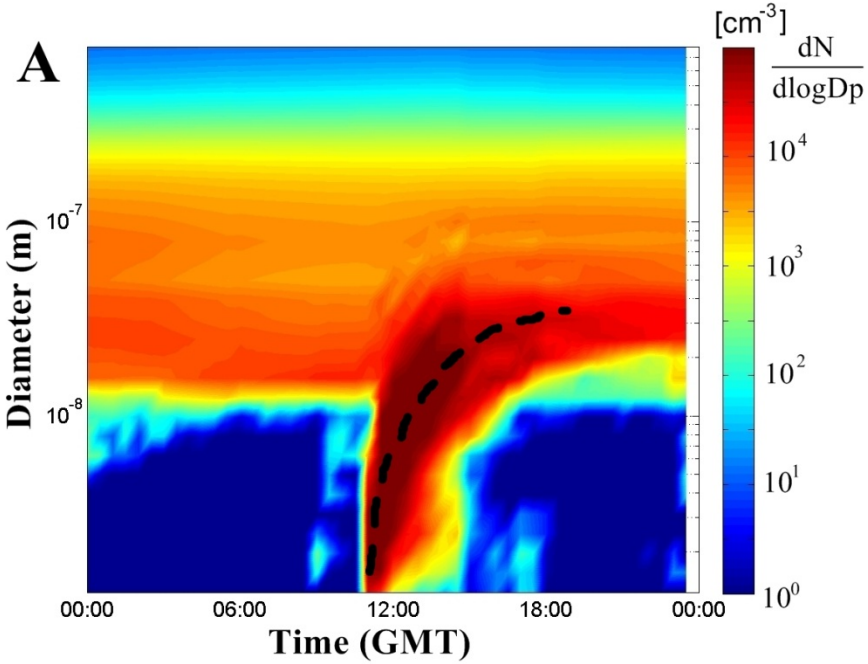
- 3D host models
  - Assimilated meteorology: GEOS-Chem, PMCAMx
  - Predicted meteorology: GISS GCM II / ModelE
  - GEOS-Chem is main global tool
    - potential to run at 0.25 x 0.3 degrees (~25 km)
    - recently parallelized, nested
- Nucleation parameterizations implemented
  - Binary (Vehkamaki, 2002)
  - Ternary (Napari, 2002; usually x 10^{-5})
  - Activation (Kulmala, 2006; Sihto, 2006)
  - Ion-mediated (Yu, 2010)
  - MODGIL ion-induced (Modgil, 2005)

Model Status

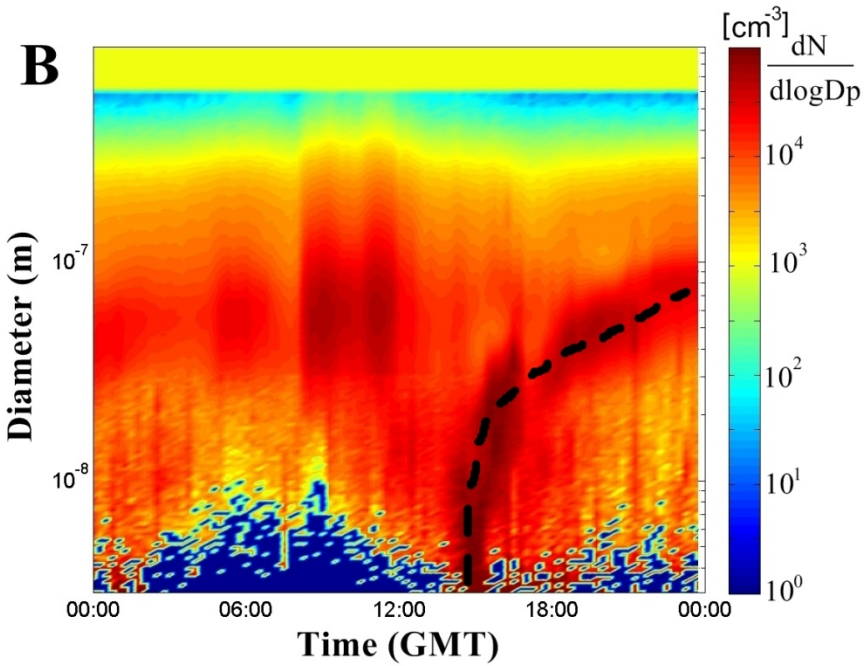
- Nuclei growth and organics
- Simple base case
  - 19 Tg/yr of SOA (biogenics)
  - assumed non-volatile
  - condenses to aerosol surface area
  - predicts reasonable growth rates
- Easy to do organic sensitivity studies
  - +100 Tg/yr “anthropogenically controlled” SOA
  - change condensation behavior
- H_2SO_4 condenses as well

Modeled and measured nucleation events

Model (GC-TOMAS)



Observations



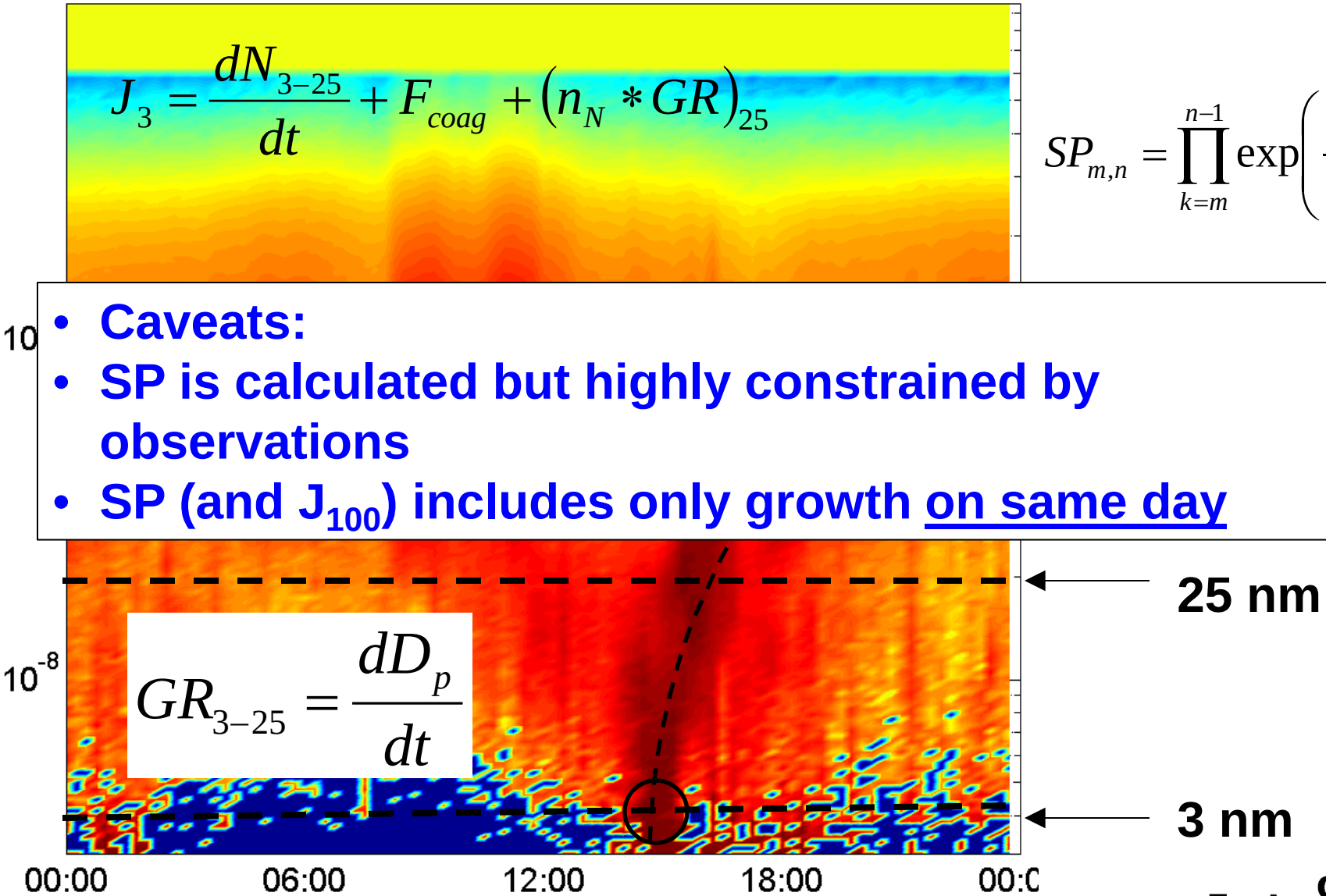
- Pittsburgh, April 16, 2002

CCN Budget Analysis

$$J_3 = \frac{dN_{3-25}}{dt} + F_{coag} + \left(n_N * GR\right)_{25}$$

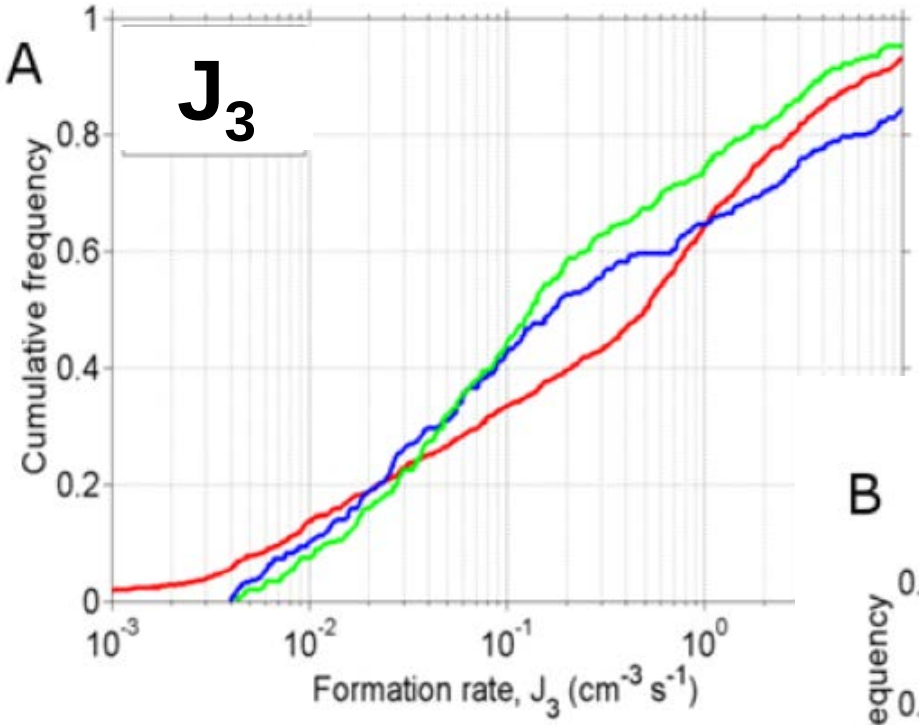
$$SP_{m,n} = \prod_{k=m}^{n-1} \exp\left(-\frac{\tau_{k,k+1}^{cond}}{\tau_{k,k+1}^{coag}}\right)$$

- **Caveats:**
- **SP is calculated but highly constrained by observations**
- **SP (and J_{100}) includes only growth <u>on same day</u>**

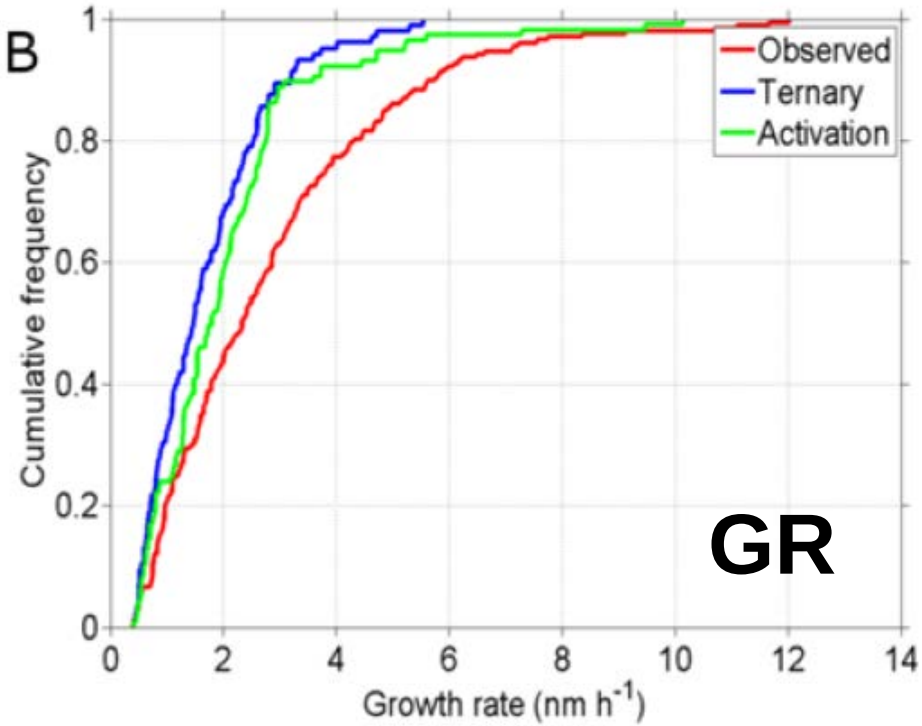


$$GR_{3-25} = \frac{dD_p}{dt}$$

Hyytiala Comparison (2007)



Observations
Ternary (model)
Activation (model)



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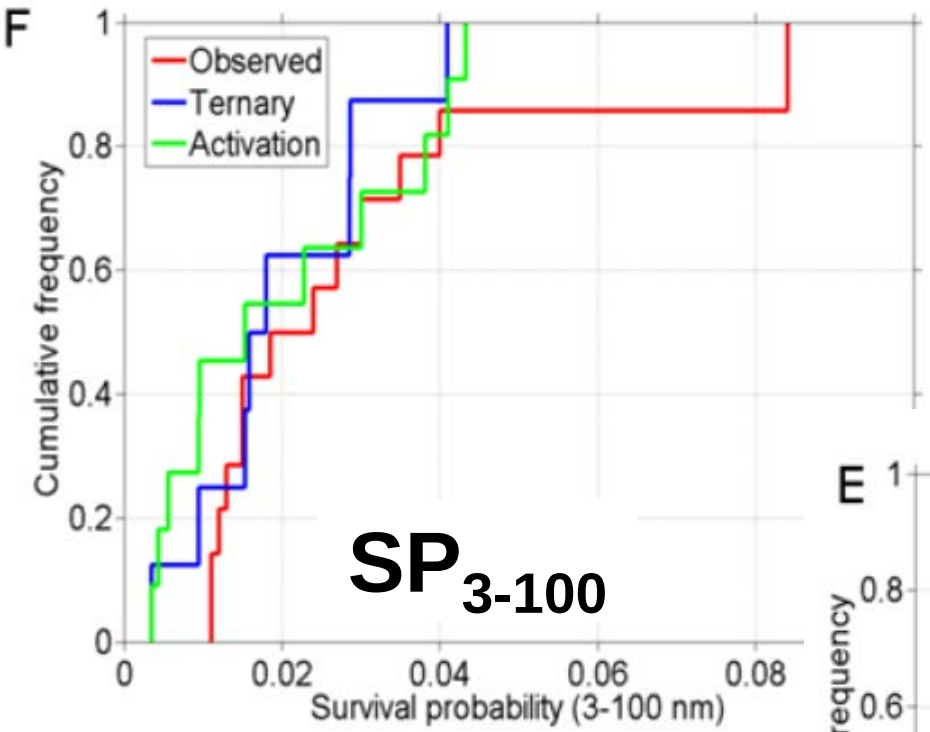

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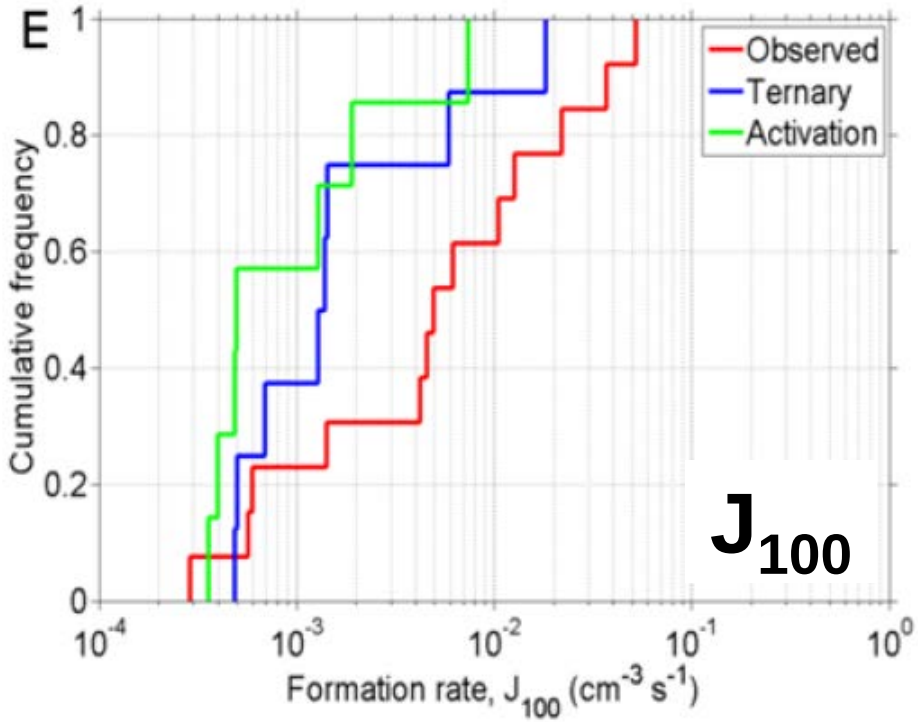
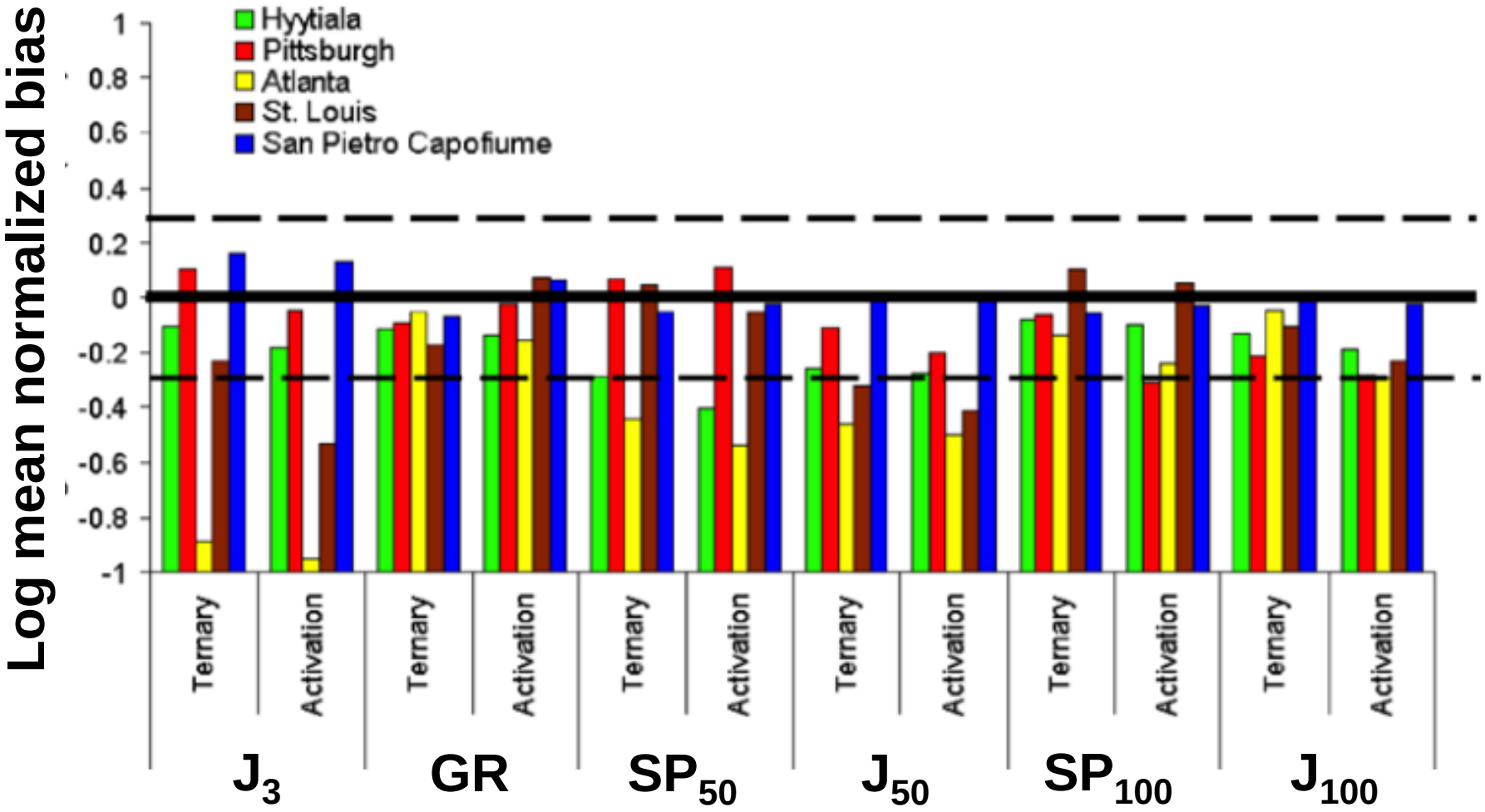
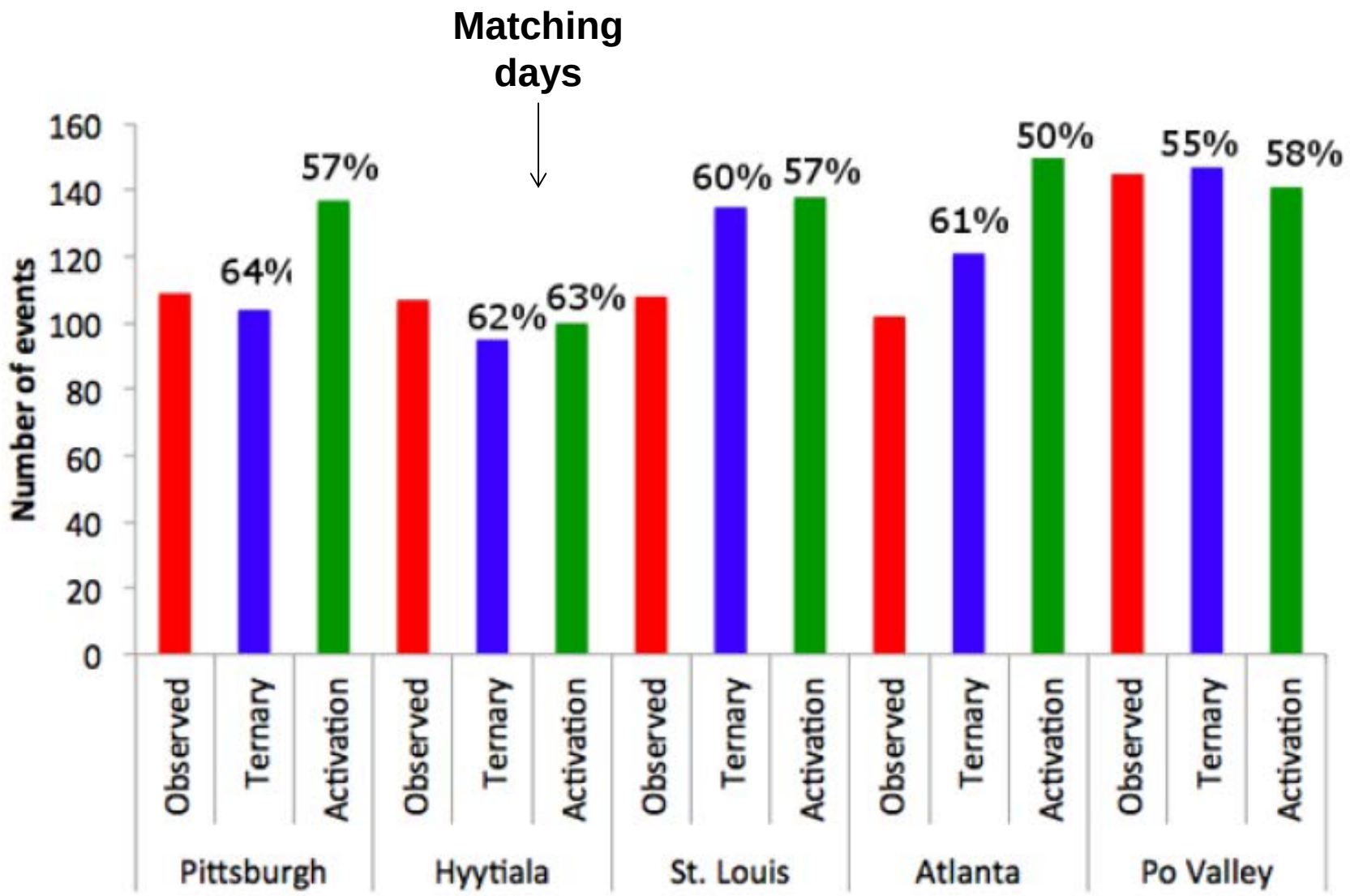


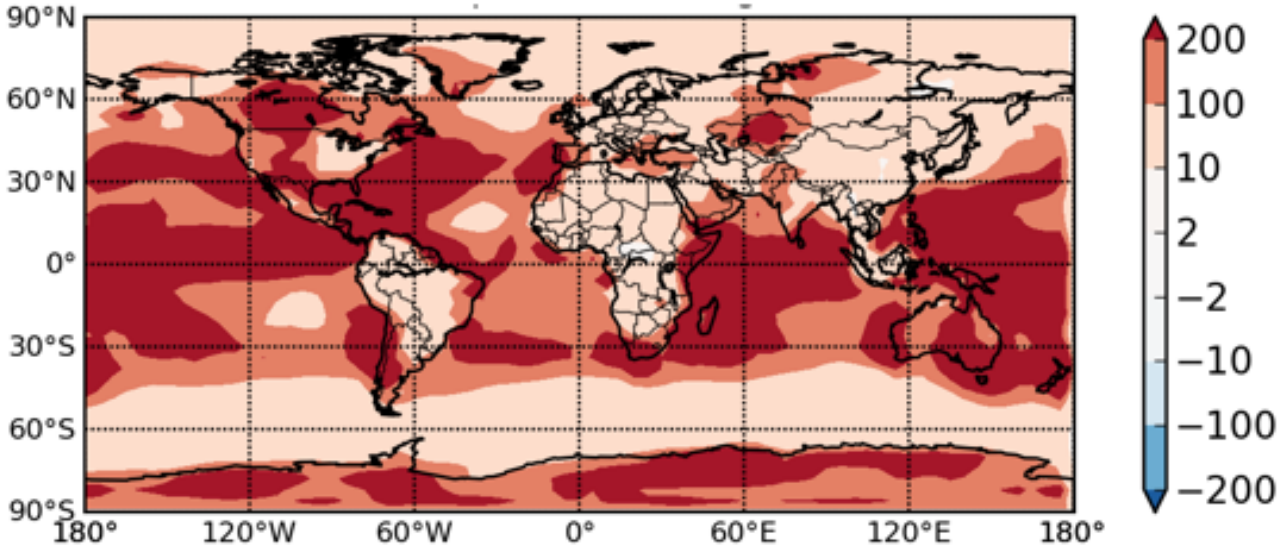
Evaluation Summary



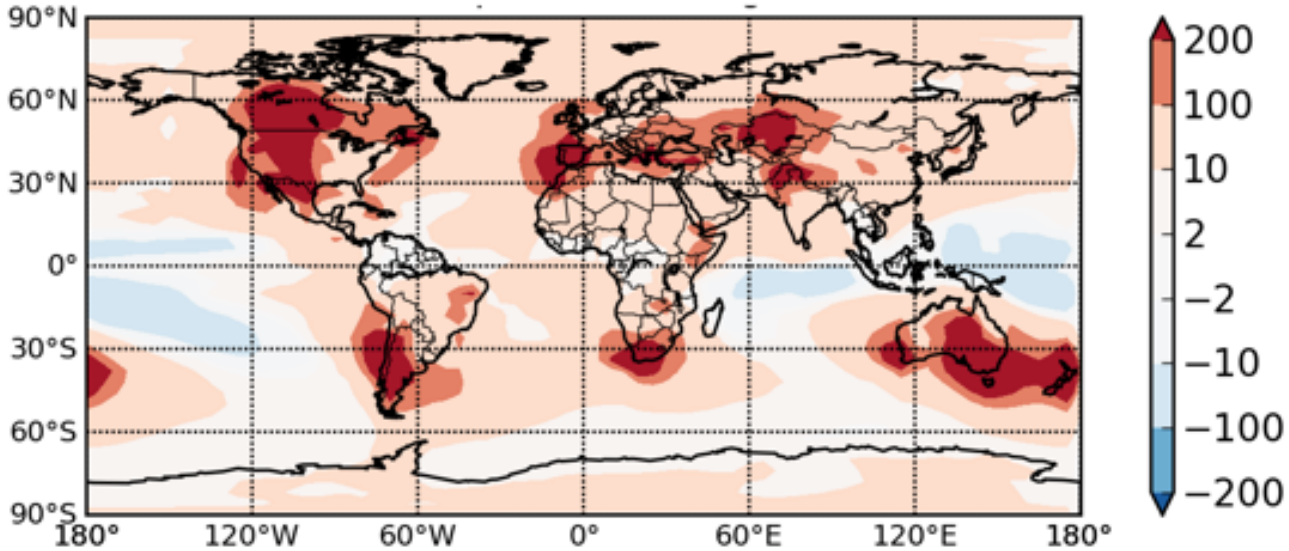
Event frequency



N10: Percent increase due to nucleation

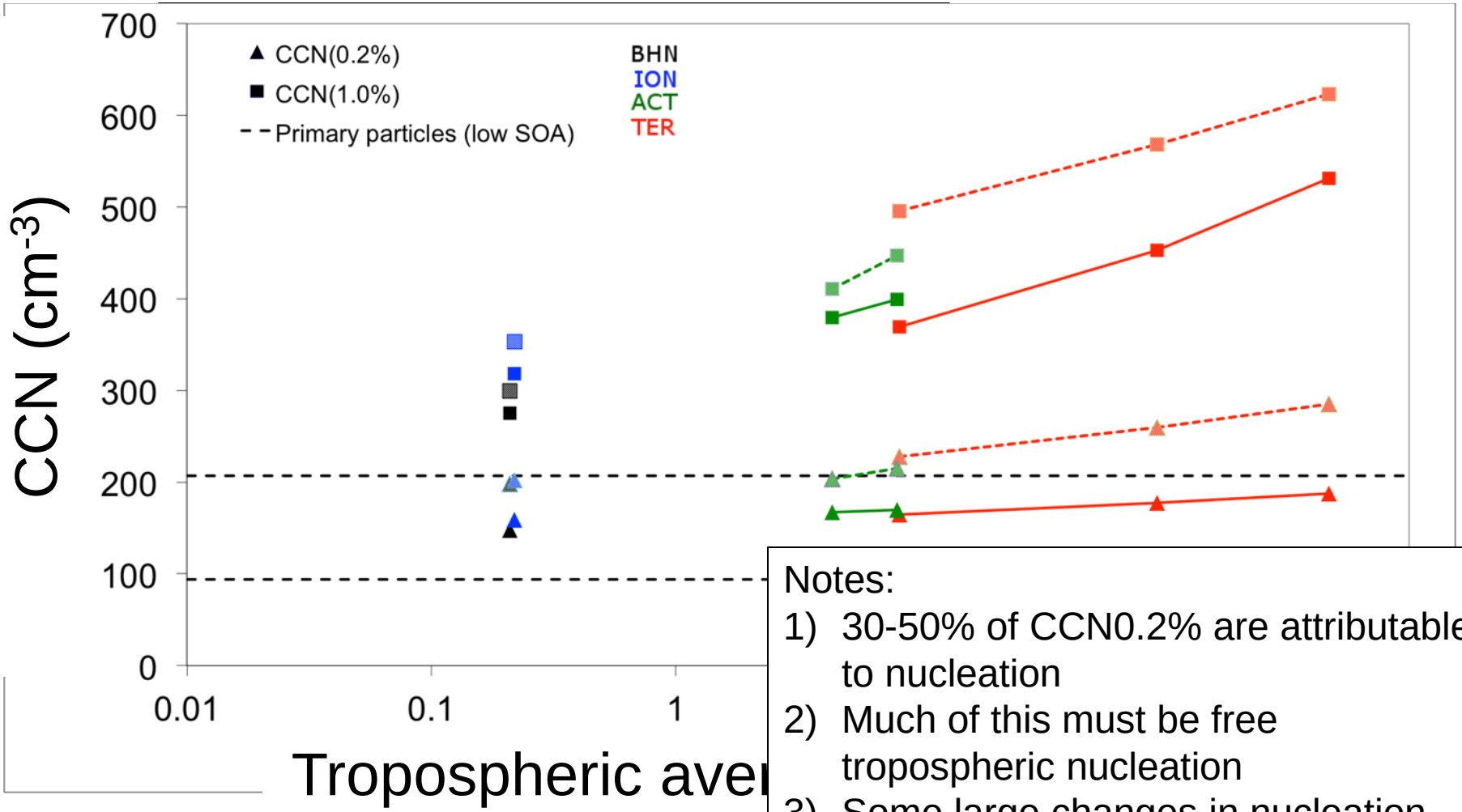


"Activation" nucleation
$A = 2x10^{-6}\ s^{-1}$



Napari ternary x 10^{-5}

Sensitivity Studies



Notes:

1) 30-50% of CCN0.2% are attributable to nucleation
2) Much of this must be free tropospheric nucleation
3) Some large changes in nucleation don't change CCN 0.2 much
4) Where and when nucleation happens matters

Next Steps

- Need to test latest mechanisms at global scale
  - Chen et al. (2012) Acid-base reaction model
  - CLOUD parameterization
- Size-dependent growth rates (Kuang et al., 2012)
  - can/should be implemented
  - has not been a problem for activation/ternary
  - recent mechanisms will require
- Amines
  - pretty limited data but could put together a simple representation in global model
- Evaluate poor predictions of day-to-day variability

Challenges

- Amines
  - how do they affect nucleation rates?
  - emissions / observations
- Organics
  - how do they affect nucleation rates?
  - which ones are responsible for early growth?
  - coupling organic thermodynamics to microphysics is non-trivial challenge
- Model reasonably sophisticated
  - (more limited by scientific knowledge than being a model per se)