Understanding New Particle Formation Events in GEOS-Chem by Comparing The Lagrangian and Eulerian Perspectives

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1. Overview

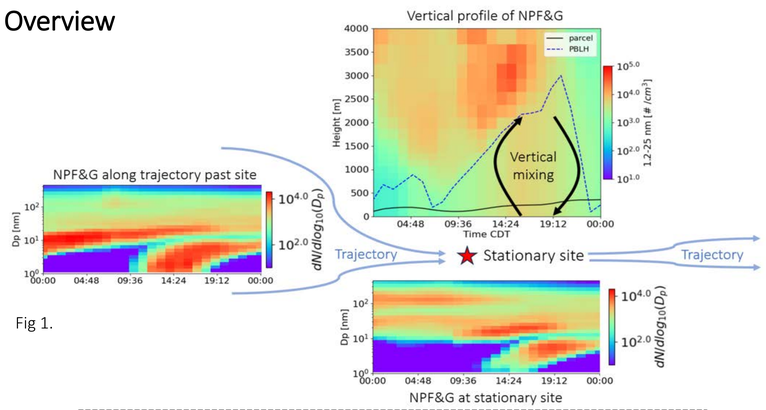


Fig 1.

Key Points:

- New particle formation (NPF) observed at a stationary site is often intermittent or discontinuous;
- Undefined NPF events are often localized NPF advecting past or away from a stationary site;
- Smoke observed at a stationary site along with NPF can make interpreting NPF statistics difficult.

- **Background:**
  - Aerosol particles exert considerable influence on global climate through direct and indirect radiative forcing.
  - NPF and growth is estimated to contribute a substantial fraction of the global average CCN.
- **The issue:**
  - NPF events are analyzed to understand the underlying microphysics and chemistry.
  - NPF events are assumed to be regionally homogeneous; however, this assumption is often not reasonable.
- **Goal:**
  - Understand how horizontal advection of particles and NPF precursors influences what is observed at a stationary site.
  - Understand how observations of NPF at a stationary site may be inadequate.

2. Observations of NPF

- Data are taken from the Southern Great Plains (SGP) observatory in North-central Oklahoma (US).

- Measurements used:
  - SMPS (11-460 nm)
  - ACSM: speciated aerosol mass

Below are several peculiar size distributions from the SGP site that will be used to demonstrate our analysis method.

Particles appear (similar to a typical NPF event), but quickly disappear.

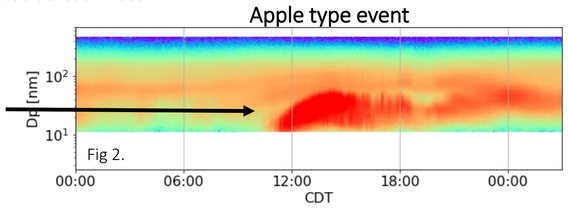


Fig 2.

Particles appear already grown, indicating NPF and growth may have occurred elsewhere in the atmosphere.

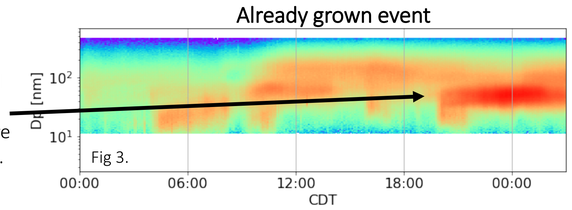


Fig 3.

Biomass burning plumes and/or NPF can complicate analysis of NPF events.

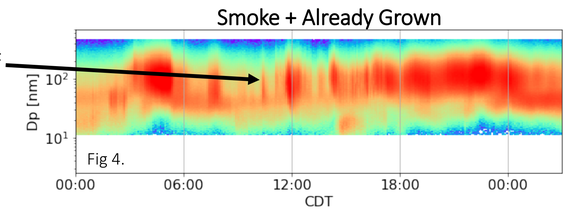


Fig 4.

3. GEOS-Chem TOMAS

- **GEOS-Chem:**
  - Simulation period: April – September 2019
  - 0.25x0.3125° grid over central United States
  - Full chemistry, 47 layer, with TOMAS aerosol microphysics
  - QFED fire emissions inventory and online biogenic VOCs

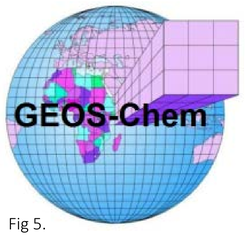


Fig 5.

- **TOMAS:**
  - 40 or 15 size bins spanning ~1.0 nm to 10 μm range.
  - Key processes:
    1. Nucleation (updated)
    2. Coagulation
    3. Irreversible size-dependent condensation

Updated NPF Mechanisms

- The inorganic NPF rates are from Dunne et al. (2016):
  - Neutral: Sulfuric Acid + Water; Sulfuric Acid + Water + Ammonia
  - Ion induced: Sulfuric Acid + Water; Sulfuric Acid + Water + Ammonia
- Organic NPF rates are from Riccobono et al. (2014):
  - Organic: Organics + Sulfuric Acid

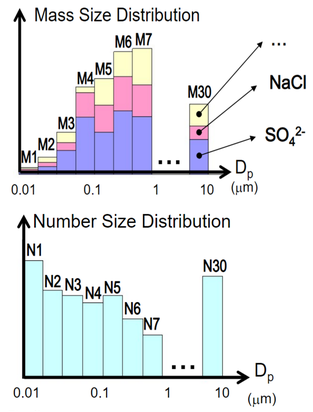


Fig 6.

4. Tool for getting the Lagrangian Perspective in GEOS-Chem:

- **HYSPLIT:**
  - HYSPLIT is a Lagrangian parcel model that can run forward and backward parcel trajectories from a single point
  - Run using MERRA-2 meteorology (would like to run with GEOS-FP in the future)
  - Trajectories pass over stationary ground site every 3 hours
  - Aerosol size distributions along trajectories are derived from a 3-D linear interpolation between adjacent grid points and vertical layers
  - The framework is easily altered to investigate gas-phase species or other processes

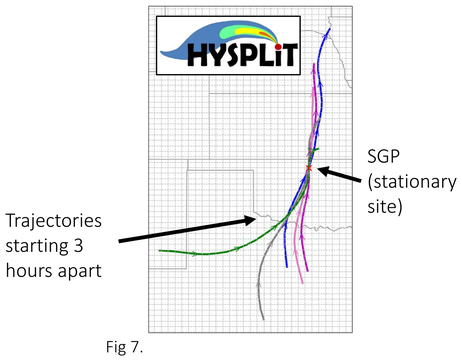


Fig 7.

5. Lagrangian versus Eulerian perspectives of NPF events

The model predicts an apple type NPF event at the SGP site

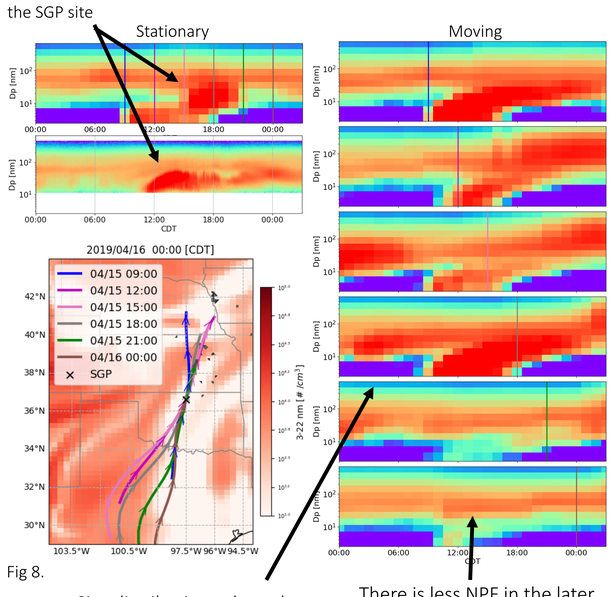


Fig 8.

Size distributions along the first several trajectories show smooth (typical) NPF events

There is less NPF in the later trajectories leading to the apple type event at the stationary site

The model and observations show a mode appearing that is already grown

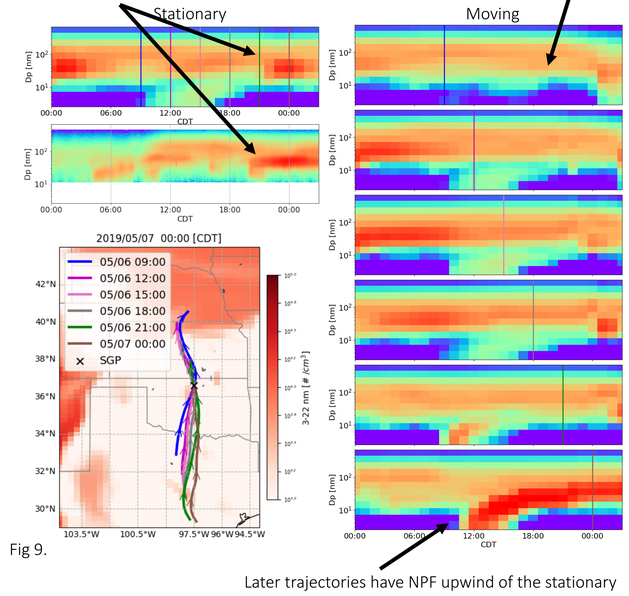


Fig 9.

Early trajectories have little NPF along their path

Later trajectories have NPF upwind of the stationary site, leading to particles to appear grown

The model and observations have smoke and NPF influence

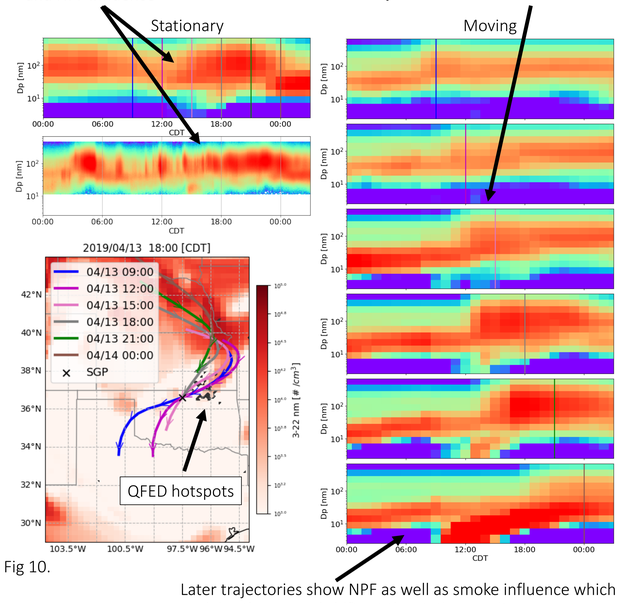


Fig 10.

Trajectories pick up smoke during the day (shift in particle size)

Later trajectories show NPF as well as smoke influence which explains the aerosol shifts at the stationary site

6. Event classification comparison

Model Lagrangian \ Model stationary	No	Undef.	NPF
NPF	11	35	5
Undef.	6	9	0
No	69	39	0
	86	83	5

51 NPF events when looking along trajectories!

51

Only 15 undefined NPF events along trajectories!

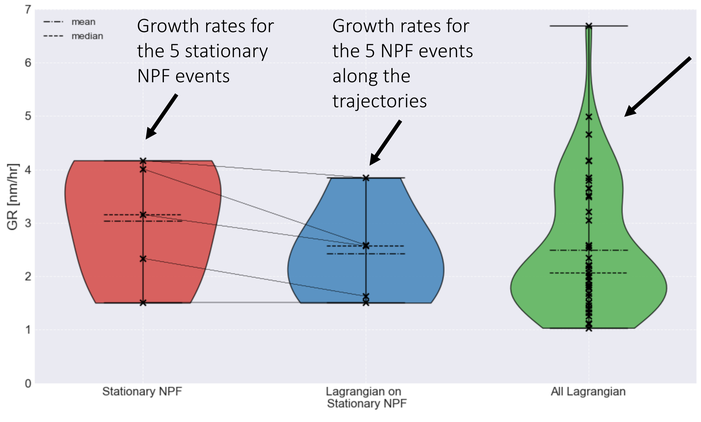
15

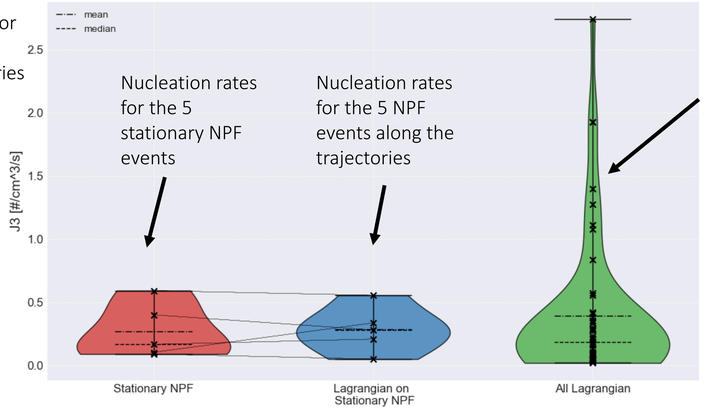
108

More non-events when looking along trajectories.

****Looking along the most NPF looking trajectory. ****

7. Growth and Nucleation rates for stationary and Lagrangian NPF events





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