



# **Opportunities for advancing aerosol** predictability in Earth system models using ARM aerosol measurements

## **Po-Lun Ma**

Atmospheric, Climate, and Earth Sciences **Pacific Northwest National Laboratory** 

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# **Evaluation: E3SM\_diags and ARM Diagnostics**

### E3SM\_diags

### **Diagnostic Plot Types and Currently Available Analysis Datasets**

The software provides support for diagnostics based on seasonal or annual climatology data, including:

- Latitude-Longitude contour maps
- Polar contour maps
- Zonal mean line plots
- Pressure-Latitude zonal mean contour
- CloudTopHeight-Tau joint histograms
- Tables summarizing metrics
- Taylor Diagrams for spatial variability

A basic set of analysis datasets are established for earth system

- GPCP for Precipitation
- CERES\_EBAF for radiation
- · Hadley Center data for sea surface temperature
- WHOI-OAFlux for ocean surface latent and sensible heat fluxes.
- ERA-Interim and MERRA2 for reanalysis datasets
- ISCCP, MISR and MODIS for Satellite simulator datasets

More datasets and variables can be easily extended based on scientists' requests.



- Incorporating ARM Diagnostics •
  - Zhang et al (2020), BAMS
  - Mostly on cloud, precipitation, radiation
  - SGP AOD is included •



## **ARM Diagnostics**



Figure 5. Probability density plots for aerosol number concentration (x-axis) versus CCN number concentration (y-axis) at 0.2% supersaturation level at the ARM SGP (left) and ENA site (right).

- Updates to the ARM Diagnostics (Zheng et al, 2022, DOE **Tech Report**)
- Aerosol physical/chemical properties and CCN datasets measured at SGP and ENA have been quality controlled and processed.
- The statistics metrics on the aerosol physicochemical • properties and CCN number concentrations annual cycles
- The process-oriented diagnostics metrics on the aerosol-to-• CCN activation assessments at different supersaturation levels.





## **Evaluation: EAGLES Diagnostics**



- al, TBS
- series, size distribution)



### Tang et al (2022, 2023), GMD; Christensen et al (2022), ACP; Varble et al (2023), ACP, Huang et

Aerosol state (composition, distribution, time



# **Evaluation: EAGLES Diagnostics**





Tang et al (2022, 2023), GMD; Christensen et al (2022), ACP; Varble et al (2023), ACP, Huang et

Aerosol state (composition, distribution, time ACI process-oriented diagnostics (Nd-CCN, LWP-Nd), but more research is needed to pinpoint



# **Evaluation: EAGLES Diagnostics**







- Tang et al (2022, 2023), GMD; Christensen et al (2022), ACP; Varble et al (2023), ACP, Huang et
- Aerosol state (composition, distribution, time
- ACI process-oriented diagnostics (Nd-CCN,
- LWP-Nd), but more research is needed to pinpoint
- Reconcile differences between satellite and

# What do these diagnostics tell us?

Pacific Northwest

N.C.









• Model produces too much aerosol

### Identify critical deficiencies in ESMs and motivate innovation Pacific Northwest



- Model produces too much aerosol
- Seems to be related to overly weak below-cloud scavenging



### Identify critical deficiencies in ESMs and motivate innovation Pacific Northwest



- Model produces too much aerosol
- Seems to be related to overly weak below-cloud scavenging
- Current parameterization was developed in 1974.
- It has been reported that the scavenging computed by this parameterization is 2-3 orders of magnitude lower than observations.

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SHOUT COEFFICIENTS FOR POLYDISPERSE AEROSOL

M. Terry Dana and J. M. Haler

ific Northwest Laboratoria ichland, WA 9935

MASTER

ormed by Battelle, Pacific Northwest Laboratories, Richland, Washington roy Commission under Contract AT(45.1).1830



## **Size distribution**





## **Identify critical deficiencies in ESMs** Pacific Northwest and motivate innovation

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### ATMOSPHERIC PARTICLES

### New particle formation in the free troposphere: A question of chemistry and timing

P. Blandel, <sup>1124</sup> J. Stylel, <sup>1</sup>M. Junniner,<sup>1</sup> C. Frege,<sup>1</sup> S. Henne,<sup>4</sup> C. R. Hayle,<sup>114</sup> U. Michael, E. Harrman,<sup>4</sup> A. Adamov, <sup>1</sup>M. Bukarski,<sup>1</sup>X. Chen,<sup>4</sup> J. Digiling,<sup>4</sup> M. Hyal,<sup>1</sup> M. Hitzhi,<sup>1</sup>J. J. Engelstone,<sup>1</sup> J. K. Stotkaev,<sup>4</sup> K. K. Kinner,<sup>4</sup> K. Misner,<sup>4</sup> K. M. Stotkaev,<sup>4</sup> K. Stotkaev,<sup>4</sup> K. K. Stotkaev,<sup>4</sup> J. Stotkaev,<sup>4</sup> J. Stotkaev,<sup>4</sup> J. K. Stotka

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### High concentration of ultrafine particles in the free troposphere produced by organic new particle formation

### Bin Zhao<sup>n 1</sup>, Manish Shrivastava<sup>4</sup>0, Neil M. Donahue<sup>8,064</sup>0, Kamish Gordon<sup>5/</sup>0, Meredith Sche John E. Shilling\*, Rahui A. Zaverl\*O., Jan Wang<sup>e</sup>, Meinrat O. Andreae<sup>512</sup>O. Chun Zhao<sup>53</sup>, Brian G liven Fan"0, and Jeromo D. Fast"

<sup>1</sup>Amogeheric Sciences and Ocdard Durings Davatas, Fachic No-Tworld National Liberatory, Rotkins, Web 19850, <sup>1</sup>Center fur Ja Greene Mitterin University, Thomas Markan, No. 1987, "Development and the Antonia Liberatory, Rotkins, Web 19850, <sup>1</sup>Center fur Ja Greene Mitterin University, Thomas Barray, No. 1987, "Development and the Antonia University, Rotkins, Web 1985, "Center fur Ja Rothardy, No. 312, "Development and Involvement of Development Antonia University, Rotkins, University, Rotkins, Database, No. 1997, "Development and Involvement of Development Antonia University in St. Loss, St. Lauk, Md. 41389, "Development Department of Energy, Development and Involvement (Jack Centers), "Stephent Contexplay, No. 312, No. 314, Md. 41389, "Development Department of Development and Centers, Mol Development Contexplay, No. 314, Md. 41389, "Development Contexplay, No. 3149, Md. 3149, Tol. 2015, St. Lauk, Md. 41389, "Development Contexplay, No. 3149, Md. 3149, Tol. 2015, Development Contexplay, D

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Etited by Summer C. Writig, Interval University, contribution, M. et al. Spectra 2000 Invalued for motion April 10. The large concentrations of university, contribution particle in the properties at an annual particle screen density within the particle interval in annual particle in the interval in a difference of density of the annual to density. The density of the same particles in the interval in a difference of density of the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the annual to density of the same particles in the sa their roles in NPI processes. We find that pure-organic NPI driven anotherizans. A roccur study by William ther route in the processes. We tool that pure-organic for it over a constraint of the form of the processes of the soft of the constraints in the appropriate topological activity of the constraints in the appropriate conditions, while the mark of the constraints while the mark of the constraints while the mark of the constraints and appropriate conditions in the large constraints and appropriate conditions in the terms and the constraints and appropriate conditions and the terms and the constraints and the terms and terms and the terms and terms and the terms and the terms and the terms and the t from docmoused volatility of organics and increased MP4 efficiency at low temperatures, somewhat counterbalanced by a reduced

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represents a major source of cloud condensation mudel globally, the prevailing view is that new particle formation rarely occurs, in nemote marine boundary layer over open nora Here we present evidence of the regular and 'request occurrence of new particle forms the upper pw. o' remote marine boundary layer following cold front passages. If particle formation is facilitated by a combination of efficient removal of existing p by precipitation, cold air temperatures, vertical transport of reactive grant fro ocean surface, and high actinic fluxes in a bro-con cloud field. The newly formed pa subsequently grow and contribute substantially to cloud condensation nuclei in the marine boundary layer and thereby import marine, my clouds,

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### Atmospheric new particle formation from the CERN CLOUD experiment

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Acrosol particles in the atmosphere profoundly influence public health and climate. Ultrafine particles enter the body through the lungs and can translocate to essentially all organs, and they represent a major yet poorly understood health risk. Human activities have considerably increased perosols and cloudiness since preindustrial times, but they remain persistently uncertain and underrepresented in global climate models. Here we present a synthesis of the current understanding of attrospheric new particle formation derived from laboratory measurements at the CERN CLOUD chamber. Whereas the importance of sulfuric acid has long been recognized, condensable vanours such as highly ovygenated organics. and iodine oxoocids also play key roles, together with stabilizers such as ammoria, amines and ions from galactic cosmic rays. We discuss how insights from CLOUD experiments are helping to interpret new particle. formation in different atmospheric environments, and to provide a mechanistic foundation for air quality and climate models

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### Global variability in atmospheric new particle formation mechanisms

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Article

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A key challenge in acrosol pollution studies and climate change assessment is to understand how atmospheric aerosol particles are initially formed<sup>12</sup>. Although new particle formation (NPF) mechanisms have been described at specific sites11, in most regions, such mechanisms remain uncertain to a large extent because of the limited ability of atmospheric models to simulate critical NPF processes12. Here we sonthesize molecular-level experiments to develop comprehensive representations of 11 NPH mechanisms and the complex chemical transformation of procursor gases in a fully coupled global climate model. Combined simulations and observations show that the dominant NPF mechanisms are distinct worldwide and vary with region and abitude. Previously neglected or underrepresented mechanisms involving organics. amines, lodine oxoacids and HNO, probably dominate NPF in most regions with high concentrations of aerosols or large aerosol radiative forcing; such regions include oceanic and human-polluted continental boundary layers, as well as the upper troposphere over rainforests and Asian monsoon regions. These underrepresented nechanisms also play notable roles in other areas, such as the upper troposphere of the Pacific and Atlantic oceans. Accordingly: NPF accounts for different fractions (10-80%) of the nuclei on which cloud forms at 0.5% supersaturation over various regions in the lower troposphere. The comprehensive simulation of global NPF mechanisms can help improve estimation and source attribution of the climate effects of aerosols.

to compulable sources of primary particles and gases.

Atmospheric acrosol particles cause more than three million premature atmospheric models - tools indispensable for understanding th deaths worldwide every year' and act as a key modulator of Farth's mechanisms and impacts of NPF on plobal and regional scales-lack oranne novelenoe every yname aan er a kan yn moanter o'r chef dan merstal Comman. Niff moe nodernade gan moleculei is the fundiamerstal source of most zanoopheris particles". The subsequere growth of these particles is thought to contribute approximately haf of the global mather of cloud condensation nuclei (CCV<sup>®</sup>, subsamility affecting sia DAU, and owe", which undergroet both NPF rate and die Source of the subsequere growth of the subsequere growth of sia DAU, and owe", which undergroet both NPF rate and die Sou, ammo-sia DAU, and owe", which undergroet both NPF rate and die Source of the subsequere growth of the subsequere growth of sind DAU and owe". Which undergroet both NPF rate and die Source of the subsequere growth of the subsequere growth of sind DAU and owe". Which undergroet both NPF rate and die name in access consequences in the state of including amine-H.SO, excleation<sup>14/8</sup> synergistic HNO,-H.SO,-NH Despite its annoyheric importance, NFF has long been among mucleation<sup>1</sup> and iodine oxoacids nucleation<sup>1,2,4</sup>, are important at or tain locations<sup>1</sup> int<sup>2</sup>, but such mechanisms have soldom. If ever, her observational studies have revealed NPF mechanisms at specific sites incorporated in atmospheric models<sup>11,10,10</sup> Another complicated mecha through direct detection of molecular clusters intermediates for par-ticle formation)<sup>1,4</sup>. However, NPF mechanisms in most regions and EXTEMPT (INTERPORT INTERPORT at most altitudes remain a mystery. This is largely because current respectively) with saturation vapour concentration (C\*) of less than

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## **Identify critical deficiencies in ESMs** Pacific Northwest and motivate innovation







## **Identify critical deficiencies in ESMs** Pacific Northwest and motivate innovation

We utilized the data set of QUEST 2 (Quantification of Aerosol Nucleation in the European Boundary Layer) campaign that was held at the SMEAR II station in Hyytiälä, Finland, in March-April 2003. The QUEST 2 data set is quite unique in the sense that during the campaign a large number of events was observed: of the total of 23 measurement days (from 18 March to 9 April 2003) 20 were new particle formation days. During QUEST 2 campaign a large number of different quantities were measured; here we describe only the measurements relevant to this study.

The measurement station SMEAR II (Station for Measuring Forest Ecosystem - Atmosphere Relations) is located in Southern Finland (61°51' N, 24°17' E, 181 m a.s.l.) in a rural region with large areas of forested land. The conditions at the station are most of the time relatively clean, even though polluted continental air arrives occasionally from the southeast to south-west directions. Also the nearest city, Tampere, located 60 km south-west from the station, can influence the local air quality. More information about the station and the measurement equipment can be found in Hari and Kulmala (2005) and at http://www.atm.helsinki.fi/SMEAR/.





Sihto et al (2006)

## From model evaluation to model improvement Northwest

### **Evaluation of state**



- Providing critical information regarding aerosol state, including
  - Mass •

Pacific

- Number
- Composition
- Size distribution
- Spatial distribution
- Time evolution
- CCN at different SS

**Evaluation of processes** 





- Bridging knowledge gaps with process-oriented diagnostics, including
  - CCN-Nd joint histogram
  - Nd-LWP joint histogram
  - GCCN effects
  - BC aging and absorption
  - Aerosol water uptake
  - Wet scavenging
  - Inferred ERFaci



### **Improvement of** understanding & predictability

### New/more observations needed for developing better process representations