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Biological particles and aerosol-cloud interactions in the Southern Great Plains

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1.0 Abstract

The United States Southern Great Plains (SGP) is the genesis for much of the warm season precipitation in the central and eastern United States. While atmospheric thermodynamics and large-scale dynamics play an important role in formation of precipitation, precipitation mechanisms are also sensitive to aerosols. Aerosols can suppress precipitation as cloud condensation nuclei (CCN) or act as ice nucleating particles (INP) in deep convective clouds. This project focused on understanding the role of primary biological aerosol particles (PBAP) in the region and its influence on cloud formation. Specifically, we focused on biological aerosol in the form of pollen, one type of PBAP that is emitted in large yet variable quantities from vegetation in the mid latitudes. Field observations provide evidence of pollen in the planetary boundary layer and pollen components in cloud droplets and fine particulate matter. Further, pollen grains can easily rupture when wet, forming smaller, sub pollen particles with sizes less than one micron. We evaluated the potential for PBAP events at the Department of Energy (DoE) SGP Atmospheric Radiation Measurement (ARM) research facility due to the rich dataset available. We also developed model simulations that accounted for pollen emission and the generation of sub pollen particles, which have been shown to be both cloud condensation nuclei and ice nucleating particles. The proposed work was designed to address the following questions:

1. What are the physical and chemical signatures of biological aerosol such as pollen in SGP ARM measurements?
2. What is the role of pollen-derived particles on deep convection and precipitation in the Central US?

Using measurements from the DoE SGP ARM site and recent airborne campaigns, we evaluated the signatures of pollen and pollen-derived aerosols on optical properties, cloud properties and precipitation (Subba et al., 2021). We identified pollen-driven events over the data record and used the Weather Research and Forecasting Model with fully coupled chemistry (WRF-Chem) to conduct chemically realistic simulations of aerosols during and summer mesoscale convective events in the Southern Great Plains (Subba et al., in review). This work improved our understanding of the role of biological aerosol on clouds and precipitation in the Central United States and placed these results in context with anthropogenically-driven processes.

2.0. Project Activities

2.1 Major Project Goals and Approach

Our two main project goals were designed to address the two science questions presented in the abstract and include (1) determining methods to identify signatures of PBAP events from existing observations, and (2) developing a modeling framework that can simulate pollen emissions, their transport, rupture and removal from the atmosphere, and their impact on cloud and precipitation processes.

To identify the physical and chemical signatures of PBAP, we used two strategies. The first was to mine the SGP ARM data to identify the number of pollen and fungal spore events, described in Section 2.2. Additionally, we also continued and developed collaborations with scientists at the DOE Pacific Northwest National Laboratory (PNNL) Environmental Molecular Sciences Laboratory (EMSL) to collect, process and analyze ambient aerosol samples for PBAP signatures, described in Section 2.4. Finally, we initiated the evaluation of model runs with flight data from the DOE HI-SCALE campaign (Fast et al., 2020) to identify airborne PBAP signatures convective events (Section 2.5).

For the second goal, we developed a new modeling framework (Subba et al., in review) within a regional coupled meteorology-atmosphere model (WRF-Chem). This included the integration of the pollen emissions model (PECM; Wozniak and Steiner, 2017; Zhang and Steiner, 2022) into WRF-Chem, and the development of several different pollen rupture mechanisms to understand when and where pollen rupture may occur (Subba et al., in review). Model simulations were conducted over the SGP region to allow for additional evaluation with the SGP data (see Section 2.3).

2.2 Activity 1: Identifying signatures of PBAP events at the DOE ARM site (Subba et al., 2021)

We use 10 years of data from the Department of Energy (DoE) Atmospheric Radiation Measurements (ARM) United States Southern Great Plains (SGP) site with nearby regional pollen and fungal spore measurements to indirectly estimate the seasonal influence of these two primary biological aerosol particles (PBAP). We estimate possible primary emissions of larger PBAP and PBAP rupture events, which form submicron organic aerosol during precipitation or high relative humidity. High pollen counts at two urban stations near SGP occur during late winter/early spring (day of year (DOY) 50–120) and late summer (DOY 240–310). Around 4–19 days per year show possible pollen events (PPE) when near-surface lidar observations of daily linear particle depolarization ratio >0.1 are coincident with high organic aerosol fraction. For PPE days with rainfall, aerosol size distribution observations show enhanced submicron particle concentrations consistent with pollen rupture events. For fungal spores, high fungal spore counts occur during late spring/early summer (DOY 110–195) and late summer/autumn (DOY 220–340). Based on size distribution observations, up to 7% of days have possible fungal spore rupture events (PFE) with higher aerosol number count specifically over the range expected for fungal spore fragment mobility diameter (20–50 nm). These short-lived PFE correlate with rainfall or occur after prolonged exposure to rainfall (e.g., >10 h). While the SGP site lacks direct measurements of bioaerosol and large particle sizes, this analysis suggests that PBAP primary emissions and rupture events could occur about 32 days per year, representing an important component of the aerosol budget during seasonal emissions.

2.3 Activity 2: Modeling of pollen emissions and rupture over the SGP site (Subba et al., in review)

Pollen, one type of primary biological aerosol particle (PBAP), is emitted from the terrestrial biosphere and can undergo physical changes in the atmosphere via particle rupture. To examine the fate of pollen and its atmospheric processing, a pollen emission and transport scheme is coupled to the Weather Research and Forecasting Model with Chemistry (WRF-Chem). We simulate the emission of pollen and its impacts on the cloud properties and precipitation in the Southern Great Plains from 12- 19 April 2013, a period with both high pollen emissions and convective activity. We conduct a suite of ensemble runs that simulate primary pollen and three different pollen rupture mechanisms that generate secondary pollen, including (1) high humidity-induced surface rupture, (2) in-atmosphere plus surface rupture, and (3) lightning-induced rupture, where in-cloud and cloud-to-ground lightning strikes trigger pollen rupture events. When relative humidity is high (>80%), coarse primary pollen (~1 µg m^{-3}) is converted into fine secondary pollen (~$1.2e^{-4}$ µg m^{-3}), which produces 80% more secondary pollen than lightning-induced rupture. Secondary pollen is primarily produced by the in-atmosphere humidity-driven rupture, which is further enhanced during a frontal thunderstorm. During strong convection, vertical updrafts lift primary and secondary ruptured pollen (~$0.5e^{-4}$ µg m^{-3}) to the upper troposphere (~12 km) and laterally transports the ruptured pollen in the anvil top outflow. In regions of high pollen and strong convection, ruptured pollen can influence warm cloud formation by decreasing low cloud (< 4 km) cloud water mixing ratios and increasing ice phase hydrometeors aloft. Figure 1 identifies the type of transport that we can observe for primary pollen (pink contours), ruptured pollen (color contours) during a deep convection event.

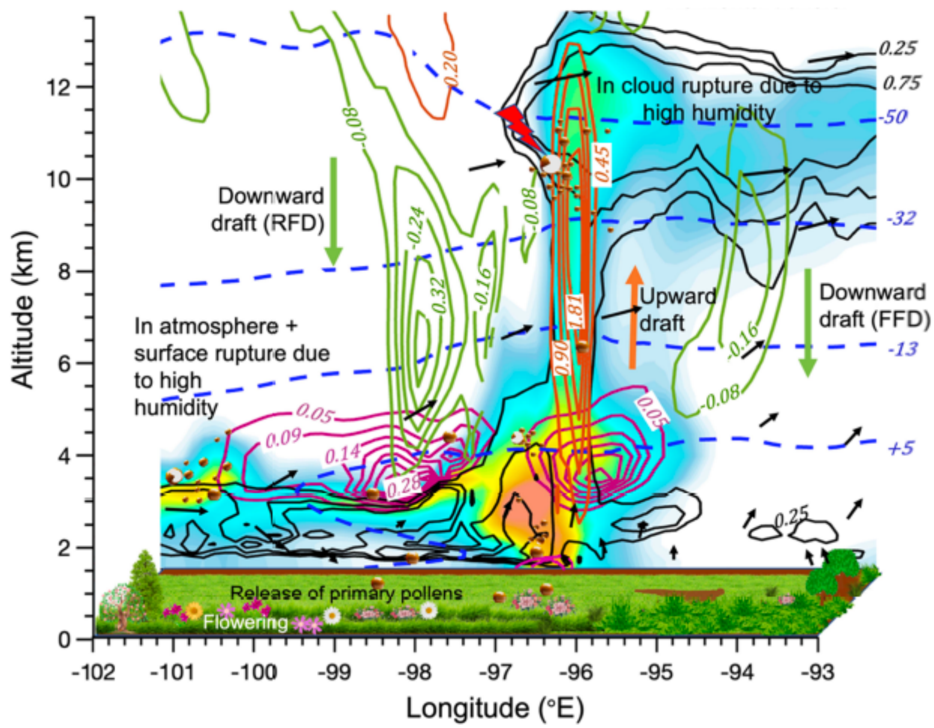

Figure 1. From Subba et al. (in review), demonstrating the pollen rupture, enhancement and transport of pollen during a convective event.

While this study only included the warm effects of pollen on precipitation processes, additional work in the research group has been including the impacts of pollen on cold-phase cloud processes. Under separate funding from the National Science Foundation, graduate student Yingxiao Zhang worked with postdoctoral fellow Tamanna Subba (funded on this project) to include pollen as an ice nucleating particle with in the WRF-Chem cloud microphysics. We presented this work at the AGU Fall 2022 meeting and anticipate this manuscript will be submitted in early 2023, and postdoc Subba will be a co-author on this work and the manuscript will include funding acknowledgement from this project.

2.4 Activity 3: Leveraging new techniques in observations to interpret PBAP in aerosol samples (Rivas-Ubach et al., 2019; Rivas-Ubach et al., 2021)

Partnering with the DoE Environmental Molecular Sciences Laboratory (EMSL), we have leveraged the data analysis and modeling of PBAP from this proposal to understand and develop new techniques to detect signatures of PBAP in the atmosphere. This has been supported through a prior EMSL grant (2016-2017) titled "Using metabolomics to understand the fate of pollen in the atmosphere," which resulted in two publications that we developed during the ASR grant period (Rivas Ubach et al., 2021; Rivas Ubach et al. 2019). These two publications have developed a pathway to use advanced instrumentation and data analysis to identify biological particles in the atmosphere. We have a current EMSL Facilities Integrating Collaborations for User Science (FICUS) grant in 2022 ("Characterizing primary biological aerosol particles during TRACER"), that will continue to advance the connection between atmospheric observations and our modeling of WRF-Chem with pollen. The 2022 FICUS proposal will integrate experimental and modeling study of biological particles using the multi-modal micro-spectroscopy, advanced mass spectrometry, and high-performance computing platforms available at EMSL. We will use a suite of EMSL instrumentation to understand the particle composition, mixing state and morphology of particles to identify both PBAP and ruptured PBAP throughout the atmospheric column. Observations from this project will be implemented in a modeling framework to improve the representation of PBAP emission, fate, and cloud processing and understand the role of PBAP on regional aerosol-cloud interactions.

2.5. Activity 4: Preliminary analysis with HI-SCALE data

Ongoing work includes the investigation of biological aerosols using HI-SCALE observations, with the goal of understanding and evaluating model output to understand the role of biological aerosols and their role in the aerosol budget and cloud formation processes. HI-SCALE was a field campaign conducted near the DoE SGP site (Fast et al., 2019), and we evaluated data from the first Intensive Operating Period (3-10 May 2016). We selected three days with a focus on pollen, including 6-8 May due to the relatively high organics on those days. During the flights on these days, we have information from aloft on the composition and size of aerosols, in comparison with our prior evaluations which were based on ground-based observations only.

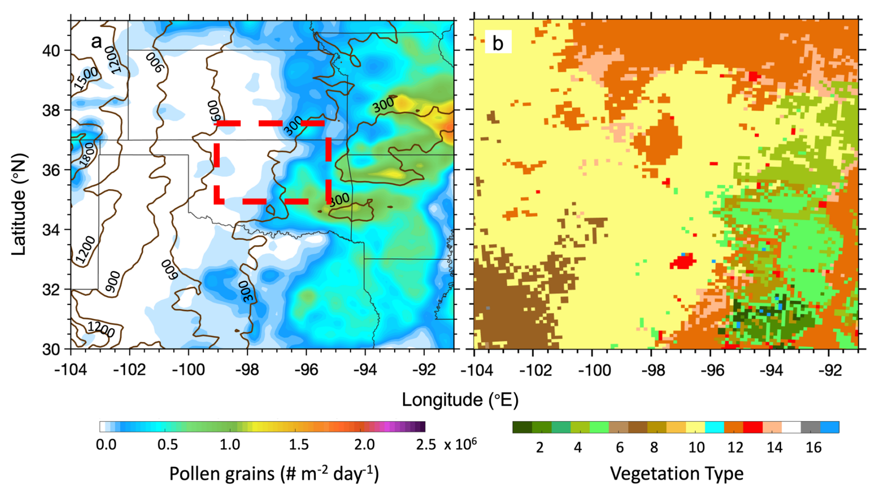

Figure 2. (left) WRF-Chem model domain with pollen grain fluxes as color contours, and the HI-SCALE campaign focus area outlined in red, and (right) land cover for WRF simulations highlighted forested areas to the east.

We conducted additional model simulations at 4km resolution for the HI-SCALE domain (Figure 2) and identified that the greatest regional pollen emissions are to the east of the flight paths. We have started to evaluate these simulations with the HI-SCALE cloud data for both meteorological and chemical conditions. Preliminary results suggest that this is a relatively low location for primary biological aerosol emissions, leading to a relatively small contribution to the total coarse mass (Figure 3). Further work will investigate if certain ACSM marker compounds can be used to analyze particle composition aloft, though we acknowledge it may be difficult to observe at this location during this IOP.

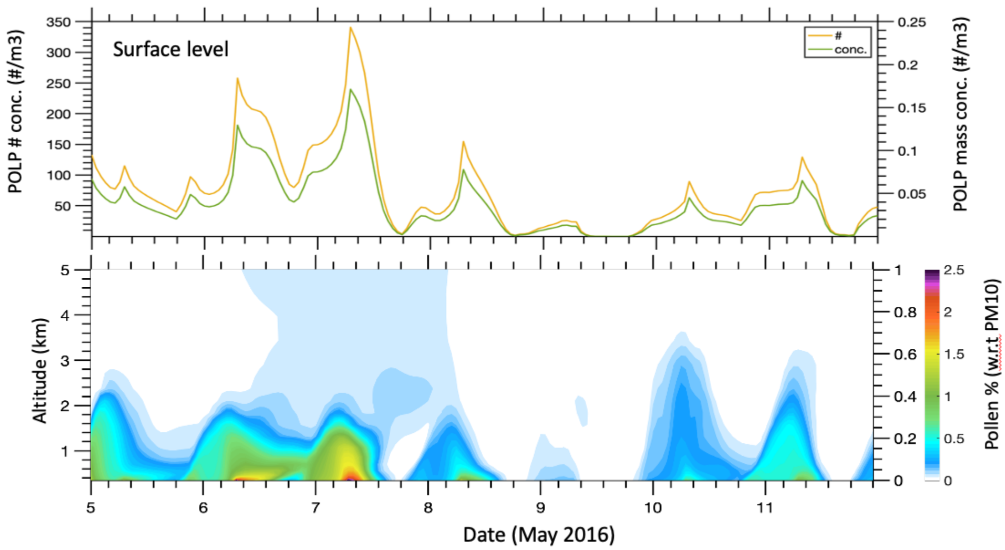

Figure 3. (top) Simulated pollen number and mass concentration at the surface over the HI-SCALE domain (e.g., red box in Figure 2), and (bottom) vertical profiles of the pollen percent contribution to simulated total PM_{10} mass.

3.0 Products

3.1. Journal articles

1. Subba, T., Y. Zhang and A.L. Steiner, Simulating the transport and rupture of pollen in the atmosphere, submitted to *Journal of Advances in Modeling Earth Systems*, submitted 29 July 2022, in revision with minor revisions requested.
2. Subba, T., M.J. Lawler and A.L. Steiner, Estimation of Possible Primary Biological Particle Emissions and Rupture Events at the Southern Great Plains ARM Site, *JGR-Atmospheres*, 126, 16, https://doi.org/10.1029/2021JD034679, 2021.
3. Rivas-Ubach, A., B. Stanfill, S. China, L. Pasa-Tolic, A. Guenther and A.L. Steiner, Deciphering the source of primary biological aerosol particles: A pollen case study, *ACS Earth Space Chem*, 5, 4, 969-979, 2021.
4. Steiner, A.L., Role of the terrestrial biosphere in atmospheric chemistry and climate, *Accounts of Chemical Research*, 53,7,1260-1268, doi: 10.1021/acs.accounts.0c00116, 2020.
5. Rivas-Ubach, A., Y. Liu, A.L. Steiner, J. Sardans, M.M. Tfaily, G. Kulkarni, Y-M. Kim, E. Bourrianne, L. Pasa-Tolic, J. Penuelas, A. Guenther, Atmo-ecometabolomics: A novel atmospheric particle chemical characterization methodology for ecosystem research,

Environmental Monitoring and Assessment, 191, 78, doi: 10.1007/s10661-019-7205-x, 2019.

3.2 Conference presentations

1. Subba, T., Y. Zhang, A.L. Steiner, Modeling of pollen rupture mechanisms and their role in cloud formation, American Meteorological Society Annual Meeting, Abstract 13.3, 27 January 2022.
2. Zhang, Yingxiao, T. Subba and A.L. Steiner, Simulating pollen-meteorology interactions and evaluating the impacts of pollen on cold cloud formation processes, American Geophysical Union Fall Meeting, Abstract A25Q-1883, 14 December 2021.
3. Subba, Tamanna, Y. Zhang, and A.L. Steiner, Modeling of pollen rupture mechanisms during a thunderstorm event, American Geophysical Union Fall Meeting, Abstract A25Q-1884, 14 December 2021.
4. Steiner, A.L., Tamanna Subba and Yingxiao Zhang, Biological aerosols and cloud-aerosol interactions in the Southern Great Plains, Department of Energy Atmospheric Science Research Annual Meeting, 21 June 2021.
5. Subba, Tamanna and A.L. Steiner, Identification of potential primary biological particle emissions and rupture events at the Southern Great Plains ARM site, American Geophysical Union Fall Meeting, Abstract A092-0007, 10 December 2020.
6. A.L. Steiner, Y. Zhang and M.Z Wozniak, Developing regional to global pollen inventories, Global Emissions Initiative (GEIA) conference (online), 23 June 2020.
7. A.L. Steiner, Biological particles and aerosol-cloud interactions in the Southern Great Plains, Department of Energy Atmospheric Science Research Conference, 10 June 2019.

3.3 Software

The pollen emissions model (PECM; Zhang and Steiner, 2022) and modifications to WRF-Chem code are available via the Steiner research group GitHub (github.com/steiner-lab). We have also shared WRF-Chem code with the NOAA RAP-Chem experimental forecast, as a potential means of incorporating pollen forecasts in conjunction with air quality forecasts. After publication of Subba et al. (in review), we will make this code available on GitHub.