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UNIVERSITY OF CALIFORNIA SAN DIEGO

Application of an Electrical Low-Pressure Impactor for the High-Resolution Analysis of Respiratory Aerosols

A Dissertation submitted in partial satisfaction of the requirements
for the degree Doctor of Philosophy

in

Chemistry

by

Renee Niles

Committee in charge:

Professor Jonathan Slade, Chair
Professor Rommie Amaro
Professor Chantal Darquenne
Professor Clifford Kubiak

2024

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The Dissertation of Renee Niles is approved, and it is acceptable in quality and form for publication on microfilm and electronically.

University of California San Diego

2024

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PREFACE

In March 2024, Renee Niles passed away suddenly and unexpectedly. At that time, she was actively engaged in completing the following dissertation.

Members of her Ph.D. committee agreed that her work was of a quantity and quality that warranted our pursuing a posthumous degree. Thus, her committee chair, Jonathan Slade, assisted by Renee's fellow graduate students, friends, and family, assembled the following dissertation. The document here represents that work.

Renee's committee members unanimously agree that the dissertation is complete and acceptable. Renee's research primarily focused on quantifying the emissions of respiratory aerosols during human speech and analyzing aerosol physicochemical properties, which is critical for understanding the transmission of airborne pathogens. She successfully isolated how specific sounds and vocal behaviors during speech influence aerosol emissions – an incredibly challenging task due to the need for high-precision measurements at the scale of individual sounds. This was not achieved before due to instrument noise and background aerosol interference. To address these challenges, Renee dedicated significant effort to developing a clean room, a respiratory aerosol sampling apparatus, and a rigorous data analysis protocol that integrated aerosol measurements with vocal and respiratory airflow metrics. Her meticulous work allowed her to isolate respiratory aerosols within the 0.010-10 µm size range on a timescale of 100 ms, a groundbreaking achievement that will open new avenues of research.

Thanks to this measurement precision – made possible largely by Renee's contributions – we co-authored a paper on her methodology and findings in *Frontiers of Psychology*. This paper was the first to demonstrate how specific vocal behaviors affect aerosol emissions on the scale of individual sounds, with profound implications for understanding the relationship between language, respiratory emissions, and the transmission of airborne pathogens. Beyond this work, Renee played a pivotal role in two additional studies as a graduate student in the Slade lab. These focused on online viscoelasticity measurements of size-resolved sea spray aerosols and how chemical composition influences the viscosity of sea spray based on its size. She was the second author of one of these studies published in *Environmental Science and Technology*.

Academically, Renee was an outstanding scholar. She received the prestigious Graduate Research Fellowship from the National Science Foundation, which supported her graduate studies for three years. As an undergraduate at the University of Rochester, she was recognized as an REU and DAAD RISE Scholar. Beyond her role as a teaching assistant at UCSD, she was beloved by students as a co-instructor for the California State Summer School for Mathematics and Science (COSMOS). Renee was an exemplary team player and a highly valued member of the Slade lab. She mentored two undergraduate students, trained incoming graduate students on research equipment, and served as our lab safety coordinator – demonstrating her leadership, dedication, and generosity in supporting others.

As such, her work fulfills our department's standards for Ph.D.-level work in this field and exceeds those standards in the depth and insight of its scholarship.

We unanimously accept this dissertation as worthy of the award of a Ph.D. degree.

ACKNOWLEDGEMENTS

“Alongside my research, I am enthusiastic about sharing my knowledge with others. I enjoy teaching and inspiring future scientists to pursue STEM fields.” – Renee Niles

From her earliest days, Renee displayed an unquenchable thirst for knowledge and understanding of everything she encountered in life. She was fiercely independent and driven to succeed, striving for perfection in every endeavor.

Beyond academia, Renee was a true outdoor enthusiast. You could often find her hiking trails, seeking adventure in nature and catching waves while surfing. When looking for a moment of relaxation she turned to the piano as yet another way to tap into her creative genius.

We hope the world will remember Renee as a dedicated, determined and focused research scientist who generously shared her academic knowledge and love of nature with others.

This dissertation is dedicated to Renee’s partner David George, her parents and siblings, her UCSD team, the John Hart family, her beloved dog, Bella, and all those who loved and supported Renee during her lifetime.

Chapter 1, in full, is a reprint of the material as it appears in *Frontiers of Psychology*. Everett, C., Darquenne, C., Niles, R., Seifert, M., Tumminello, P. R., & Slade, J. H. (2023). Aerosols, airflow, and more: Examining the interaction of speech and the physical environment. *Frontiers in Psychology*, *14*, 1184054. The dissertation author was a primary researcher and co-authored this paper.

EPIGRAPH

“There is a place called ‘heaven’ where the good here unfinished is completed; and where the stories unwritten, and the hopes unfulfilled, are continued. We may laugh together yet.” – J. R. R. Tolkien

VITA

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PUBLICATIONS

Caleb Everett, Chantal Darquenne, Renee Niles, Marva Seifert, Paul R. Tumminello, and Jonathan H. Slade, "Aerosols, airflow, and more: Examining the interaction of speech and the physical environment." *Front. Psych.*, **2023** *14*, 1184054. DOI: 10.3389/fpsyg.2023.1184054

Paul R. Tumminello, Renee Niles, Vanessa Valdez, Chamika K. Madawala, Dilini K. Gamage, Ke'La A. Kimble, Raymond J. Leibensperger III, Chunxu Huang, Chathuri Kaluarachchi, Julie Dinasquet, Francesca Malfatti, Christopher Lee, Grant B. Deane, M. Dale Stokes, Elizabeth Stone, Alexei Tivanski, Kimberly A. Prather, Brandon E. Boor, and Jonathan H. Slade, "Size-dependent nascent sea spray aerosol bounce fractions and estimated viscosity: the role of divalent cation enrichment, surface tension, and the Kelvin effect," *Environ. Sci. Technol.* **2024** *58* (44), 19666-19678, DOI: 10.1021/acs.est.4c04312

CONFERENCE ABSTRACTS

Paul R. Tumminello, Renee Niles, Vanessa Valdez, Raymond Leibensperger III, Chunxu Huang, Christopher Lee, Grant Deane, Dale Stokes, Kimberly Prather, Brandon Boor, and Jonathan H. Slade, "Size-resolved phase state of nascent sea spray aerosol using the particle bounce method," American Chemical Society, Spring Meeting, Indianapolis, IN, USA, 2023. Oral.

Paul R. Tumminello, Renee Niles, Vanessa Valdez, Raymond Leibensperger III, Chunxu Huang, Christopher Lee, Grant Deane, Dale Stokes, Kimberly Prather, Brandon Boor, and Jonathan H. Slade, "Size-resolved analysis of the phase states and effective densities of nascent sea spray aerosol using the particle bounce method," American Chemical Society, Fall Meeting, San Francisco, CA, USA, 2023. Poster.

ABSTRACT OF THE DISSERTATION

Application of an Electrical Low-Pressure Impactor for the High-Resolution Analysis of Respiratory Aerosols

by

Renee Niles

Doctor of Philosophy in Chemistry

University of California San Diego, 2024

Professor Jonathan Slade, Chair

The recent COVID-19 pandemic, which was spread mainly by airborne transmission, changed how society functioned across the globe. A significant reason for the virus's rapid spread was human interactions through speaking and the release of tiny respiratory aerosols. My projects encompass studying how certain physiological aspects of speech, including vocal amplitude (loudness), associated airflow, and different sound types, affect respiratory aerosol size, emission rates, and concentrations during speech. I am optimizing a technique to measure particles as small as viruses and at a timescale coherent with individual sounds. This resolution will allow me to

better understand respiratory aerosol emissions on a scale unattainable previously, marking an important advancement in potentially better predicting how viruses spread when people communicate verbally. Respiratory aerosols are expected to transform in their physical state and chemical composition upon emission from the mouth and into the surrounding air. Water condensation and evaporation from the particles, depending on the temperature and relative humidity of the air, will cause them to alter their phase and viscosity. Aerosols become less viscous when they take up water, similar to how it's easier to clean dried toothpaste from the sink basin when wetting it. However, the growth rates and changes in respiratory aerosol phase and viscosity following water uptake have not been studied. Understanding how their physical state evolves in the air is critical in understanding aerosol lifetimes and the potential survivability of viruses in respiratory aerosols. My research is designed to fill these important gaps due to their implications for public health, as airborne disease transmission greatly depends on respiratory aerosol emission characteristics and particle properties. Knowing and predicting which pathway(s) certain diseases may spread will enable more comprehensive models to be made. These more complete models will, in turn, allow leadership to provide the public with more effective instructions on how to behave and the safety measures to limit the spread of disease and ultimately decrease the chances of epidemics or pandemics occurring.

INTRODUCTION

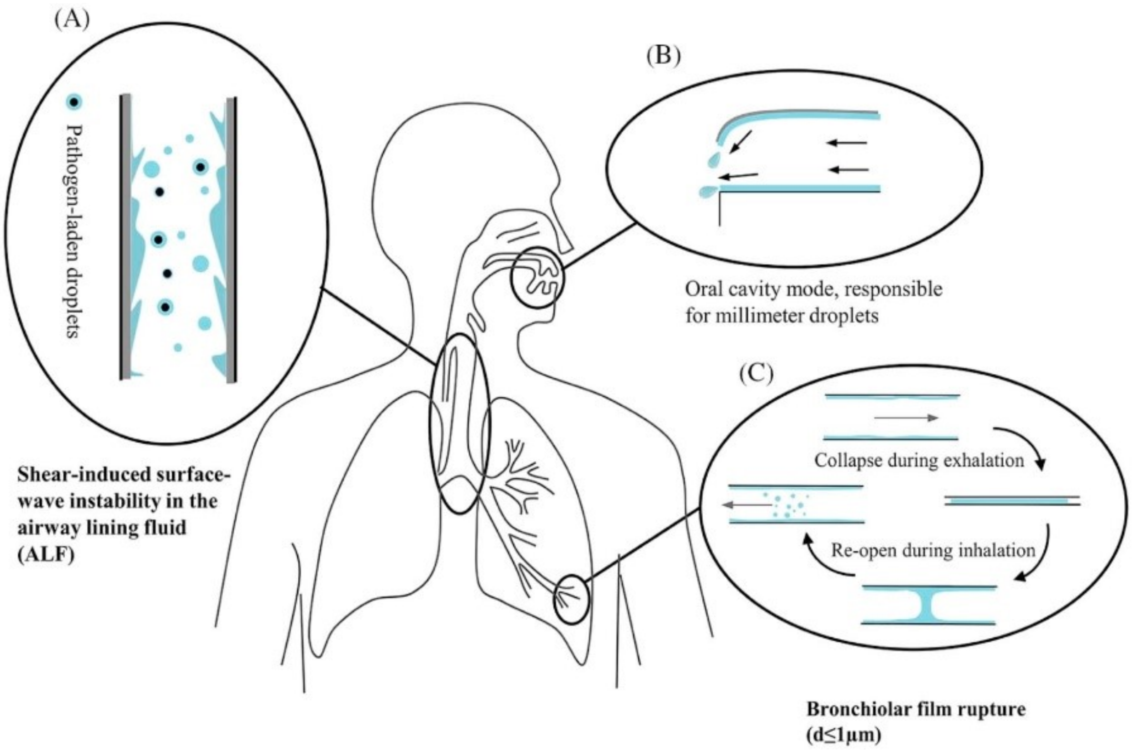

Figure 0.1 Diagram illustrating the different particle generation mechanisms that generate different size modes for respiratory aerosols (Wei et al., 2016).

The importance of respiratory aerosols in transmitting infectious diseases was highlighted during the recent COVID-19 pandemic.[1–3] Respiratory aerosols are emitted from various respiratory activities, including talking, whispering, and singing.[1–4] Several studies have characterized respiratory aerosols' emission rates and size distributions from various speech activities.[5–10] These respiratory aerosol particles can range in size from that of single viruses (sub-100 nm) to 100s of microns in diameter.[9–11] Particle size is critical as it affects aerosol buoyancy and depositional lifetime,[12,13] multiphase chemistry,[14] and the capacity to serve as a vector for airborne viruses.[3,6,15,16] The size of emitted respiratory aerosol varies as a function of specific respiratory activity and the ambient relative humidity and temperature,[17] along with physiological factors,[10,18,19] including vocal amplitude,[20] age, gender, and body mass index.[21–23] The ambient relative humidity effects on aerosol growth are dictated by the aerosol's hygroscopicity,[24] which is unknown for respiratory aerosol from human subjects.

There have been many investigations into respiratory aerosol production that have utilized various standard aerosol measurement instruments, including the aerodynamic particle sizer (APS),[10,20,25–27] Scanning Mobility Particle Sizer (SMPS),[18] and time-resolved planar particle image velocimetry (PIV).[28,29] Many studies have reported particle number concentrations <100 particles per cubic centimeter during respiratory activities.[20,25,26] However, only a few studies have measured respiratory aerosols smaller than 500 nm in diameter.[30] This is a critical size range due to their ability to still act as suitable hosts for pathogens, some of which are as small as 90 nm[15] and have greater buoyancy, thus having a higher likelihood of being inhaled and infecting others.[31,32] This highlights the importance of adding to the limited data on the existence of these smaller aerosols in respiratory emissions to gain a more complete understanding of disease transmission.

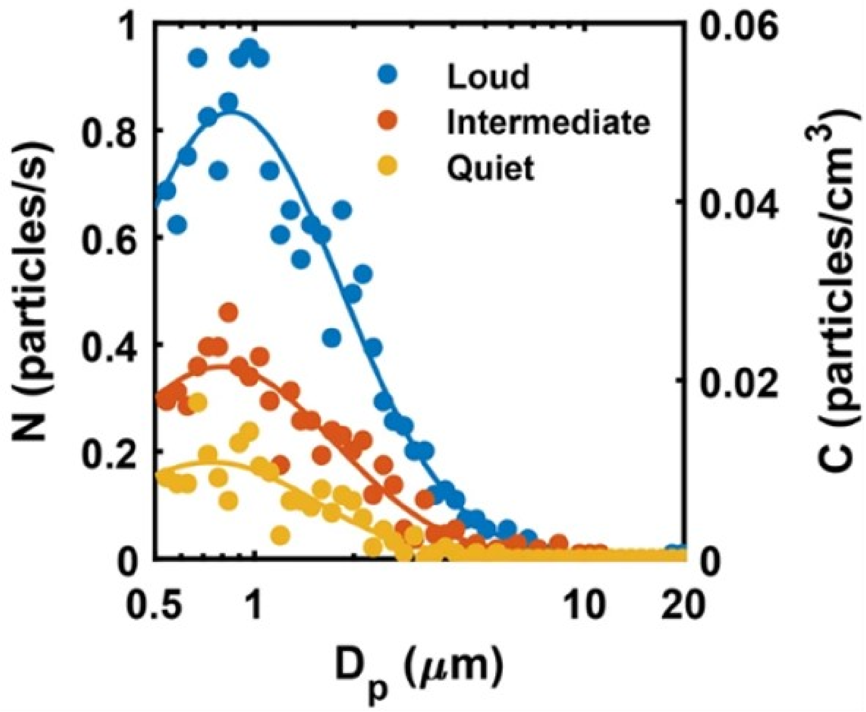

Figure 0.2 Size distributions and concentrations for one subject at different vocal amplitudes (Asadi et al., 2019).

Another understudied aspect of respiratory aerosols is the relationship between their emission, sites of origin, and the effects of airflow. Reports suggest that respiratory particle size is related to the individual's vocalization behaviors and speech sounds.[33,34] Three main size distribution modes are linked to distinct generation mechanisms, as illustrated in Figure 1. The first mode originates from film-bursting that occurs in the bronchioles during breathing. The second originates in the larynx and vocal cords, involving film-bursting with vocal cord vibrations and shear forces on the airway mucus layer in the larynx due to high airflow. The third mode originates in the oral cavity through film-bursting of lips, teeth, and tongue movements.[11] These different aerosolization processes generate different size modes and concentrations.[11,34] This previous study of these modes, however, was size-limited, with only particles >0.1 μm being measured, despite the possible presence of smaller sizes that may occur when the bronchial mode involving the deep lung area is the origin site. Particle deposition in the lungs that will be subsequently emitted during respiratory activities is driven by flow velocities and particle size, such that larger particles are deposited earlier in the respiratory tract by impaction, and only the smaller particles can travel into the deep lung by diffusion.[35] Thus, excluding this size range could severely limit our understanding of the different size modes. These studies of particle origin have led to subsequent investigations, some of which have found that particle generation is positively correlated with vocal loudness due to the associated natural slight increase in pitch and, therefore, in the frequency of vocal fold opening and closure, which leads to an increased number of film-bursting events (see Figure 2 from Asadi et al., 2019).[20] However, this conclusion has the caveat that it does not explore the possible additional influences of airflow. This may be significant as the number of particles produced via the shear force mechanism is directly related to airflow rates.[34] Furthermore, at faster-exhaled airflows, a greater volume of air will be introduced to the instrument's inlet over the duration of the activity, carrying a greater absolute number of respiratory aerosols. Thus, the exhaled air volume alone affects the measured aerosol number concentrations, leading to potential misconceptions regarding vocal amplitude and volume flow. Disentangling the influences of vocal amplitude and airflow rates on particle generation and size distributions is a major factor to consider for developing models of an individual's probability to serve as a vector. Studies on the SARS-CoV-2 virus have shown that some individuals, coined "super emitters," have higher particle production rates than most of the population.[36,37] If a relationship between airflow and particle emission rates does indeed exist, this could start to explain the "super emitter" phenomenon. If a person naturally generates a higher airflow than the average population while performing a certain word or sound, they would produce more total aerosol particles when speaking and could have a greater chance of infecting others.

Words and sounds are typically articulated within short timeframes, with syllables lasting around 50-400 milliseconds for American English.[38] Thus, to compare the effects of generation mechanisms, such as vocal loudness and airflows, on emissions, observing the evolution of the full particle size distributions at the same temporal resolution that speech behaviors occur is essential. It is also key to measure aerosols, airflow rates, and the acoustics in tandem for each measurement since, as previously discussed, it is well-known that particle emissions are highly variable between people since they often depend on many environmental and individual physiological variables.

To fill in some of these knowledge gaps, I have been working on integrating a measurement approach that employs the Electrical Low-Pressure Impactor (ELPI, Dekati Ltd) for respiratory aerosol size distribution analysis with simultaneous volume flow and acoustic recordings during human speech. The ELPI measures a broader range of aerosol sizes, from 6 nm to 10 μm, than commonly employed aerosol measurement techniques, including the APS and SMPS. Particles within this size range are detected at a rate of 0.1 s, on the same timescales as individual sounds and words in English. The ELPI's relatively larger size range and high temporal resolution could make it a key approach to gaining a more in-depth understanding of respiratory aerosol emissions. However, it has not commonly been applied to such studies; thus, optimizing it for these conditions is critical. So far, only one study of respiratory aerosol has been done with the ELPI.[30] The reported respiratory aerosol number concentrations were much greater, by a factor of 10 to 100 than in other reports.[4,16,20,39] The reason for this is unknown, highlighting the need to optimize this instrument for respiratory aerosol detection to obtain meaningful results.

My Aim 1 is to determine the viability of the ELPI for respiratory aerosol detection and optimize it for future studies. This is being conducted through instrument intercomparisons and testing the ELPI response to transient changes in the relative humidity, particle size distributions, and airflow rates at the instrument's inlet during speech. I plan to develop a methods-focused paper from this work. Aim 2 will focus on applying the ELPI in an integrated respiratory aerosol, airflow, and acoustics measurement system to determine the effects of exhaled volume airflow rates on respiratory aerosol size distributions and emissions rates during controlled speech. I co-wrote a publication in the *Frontiers of Psychology* that highlighted the applicability of this integrated approach.[40] A future publication concerns disentangling the effects of volume flow rate and vocal amplitude during speech. Aim 3 will characterize, on an individual particle basis, the hygroscopic growth and changing phase states of respiratory aerosol as a function of relative humidity employing atomic force microscopy. I am currently collaborating with Dr. Vicki Grassian's laboratory on using their atomic force microscope. I plan to publish how different components in respiratory aerosol and where they are generated in the respiratory tract influence their hygroscopic growth and phase states, essential for understanding the fate of virus-laden particles in the environment. This will be performed using model respiratory aerosol mixtures generated in the lab and real respiratory aerosol particles sampled from human participants. More details regarding each of these aims are discussed in the following.

Chapter 1 AEROSOLS, AIRFLOW, AND MORE: EXAMINING THE INTERACTION OF SPEECH AND THE PHYSICAL ENVIRONMENT

frontiers | Frontiers in Psychology

TYPE Original Research
PUBLISHED 15 May 2023
DOI 10.3389/fpsyg.2023.1184054

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OPEN ACCESS

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RECEIVED 10 March 2023
ACCEPTED 28 April 2023
PUBLISHED 15 May 2023

CITATION
Everett C, Darquenne C, Niles R, Seifert M, Tumminello PR and Slade JH (2023) Aerosols, airflow, and more: examining the interaction of speech and the physical environment.
Front. Psychol. 14:1184054.
doi: 10.3389/fpsyg.2023.1184054

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Aerosols, airflow, and more: examining the interaction of speech and the physical environment

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We describe ongoing efforts to better understand the interaction of spoken languages and their physical environments. We begin by briefly surveying research suggesting that languages evolve in ways that are influenced by the physical characteristics of their environments, however the primary focus is on the converse issue: how speech affects the physical environment. We discuss the speech-based production of airflow and aerosol particles that are buoyant in ambient air, based on some of the results in the literature. Most critically, we demonstrate a novel method used to capture aerosol, airflow, and acoustic data simultaneously. This method captures airflow data via a pneumotachograph and aerosol data via an electrical particle impactor. The data are collected underneath a laminar flow hood while participants breathe pure air, thereby eliminating background aerosol particles and isolating those produced during speech. Given the capabilities of the electrical particle impactor, which has not previously been used to analyze speech-based aerosols, the method allows for the detection of aerosol particles at temporal and physical resolutions exceeding those evident in the literature, even enabling the isolation of the role of individual sound types in the production of aerosols. The aerosols detected via this method range in size from 70 nanometers to 10 micrometers in diameter. Such aerosol particles are capable of hosting airborne pathogens. We discuss how this approach could ultimately yield data that are relevant to airborne disease transmission and offer preliminary results that illustrate such relevance. The method described can help uncover the actual articulatory gestures that generate aerosol emissions, as exemplified here through a discussion focused on plosive aspiration and vocal cord vibration. The results we describe illustrate in new ways the unseen and unheard ways in which spoken languages interact with their physical environments.

KEYWORDS
phonetics, environment, aerosols, airflow, adaptation, acoustic, respiratory

1. Background: effects of the environment on speech, and of speech on the environment

While our understanding of language and linguistic diversity continues to evolve, one area of research that remains underexplored is the interaction of speech and the physical environment. Like other facets of human behavior, languages are affected over the long-term by external physical factors (Bentz et al., 2018). Conversely, however, languages themselves might affect the immediate physical environments of their speakers, and this impact could in turn affect other individuals in those environments. In this paper, we dedicate some of our attention to exploring the way in which two articulatory gestures in languages appear to impact the physical environments of their speakers via differences in airflow and generation of aerosol particles. One of these gestures, vocal cord vibration, is critical to all spoken languages. The second, aspiration, is found in about a fifth of the world's languages, including English. The exploration of the aerosol generation characteristics of these articulatory gestures is preliminary, serving primarily to illustrate a novel method we have developed for simultaneously capturing airflow, acoustic, and aerosol data. First, we briefly survey some of the research suggesting that languages are themselves affected by the physical environments in which they are spoken.

It is becoming increasingly clear that languages evolve in ways that are sensitive to the typical characteristics of their speakers' environments. To cite one relatively obvious example, the frequency with which people discuss particular weather phenomena varies in accordance with environmental factors (Kemp et al., 2018). Less obviously, urban and industrialized environments yield an increased likelihood that certain colors are foregrounded and discussed, yielding an apparent influence on the development and usage of some color terms. Evidence suggests that languages spoken by industrialized groups tend to develop more precise color terms for brightly colored hues associated with modern techniques of dying and coloring (Gibson et al., 2017). Given that agriculture and industrialization are not stochastically associated with environment types, such factors hint at indirect environmental influences on speech. The kinds of spatial language speakers employ are impacted more directly by the environments in which they are embedded, as evidenced for instance by experimental research in virtual environments (Nölle et al., 2018). Combinations of certain lifestyle types in particular ecologies may also impact the likelihood that speakers come to use robust sets of abstract terms for odors (Majid et al., 2018). These are just some of the ways in which environmental factors appear to influence lexical phenomena.

With respect to phonetic and phonological phenomena, research suggests that the diet types characteristic of particular cultures can impact the likelihood that the members of those cultures use particular sound types. Languages spoken by people with softer diets are more likely to rely on labiodental consonants, presumably because the softer diet yields characteristic overbite and overjet dental configurations in adults (Blasi et al., 2019). These configurations, in turn, yield a greater ease of articulation of labiodental consonants. Given that softer diets are largely a byproduct of agriculture of particular kinds, this fact hints at a long-term probabilistic yet indirect effect of physical environments on speech [Of course, the degree to which cultures rely on agriculture is due to a complex interaction of factors including environment and cultural transmission patterns (Vilela et al., 2020)]. The fact that labiodental consonants are associated with particular bite types has now been supported by a range of findings, including biomechanical modeling, diachronic trends, phonological typology, the frequency of sounds in wordlists worldwide, and the observation of the phonetic tendencies of individuals with divergent bite types (Blasi et al., 2019; Everett and Chen, 2021).

Related research has also suggested that the ambient characteristics of given cultures impact in more direct ways, though subtle and gradual ones, the extent to which their languages rely on certain kinds of sounds. More specifically, it has been hypothesized that extremely arid climates, most notably those in very cold regions with typically low specific humidity, place pressures on the ease of articulation of certain laryngeal gestures required for complex tonality and vowel production (Everett et al., 2015; Everett, 2017). While more direct, these putative environmental effects would nevertheless surface crosslinguistically via well-established diachronic and sociolinguistic phenomena (Everett, 2021). The central claim in such work is that some phonetic phenomena might be triggered at slightly different rates due to very minor variations in the ease and precision of vocal cord vibration, owing to the effects of aridity on the vocal cords' viscosity (Leydon et al., 2009). Ease of articulation is already well known to impact the rate at which certain sound types occur in speech and in phoneme inventories worldwide, so the central mechanism at the heart of this hypothesis is itself uncontroversial. Nevertheless, it is unclear whether environmental factors like extreme aridity impact ease of production of the relevant articulatory gestures, at least to the extent that they subtly influence diachronic sound changes, and some objections have been raised to this hypothesis (e.g., Collins, 2016). In short, while correlational data are broadly consistent with the possibility of a direct ecological effect, the likelihood of this possibility is contested. Setting aside these particular debates about direct long-term ecological effects on sound use, there is growing consensus that languages are affected indirectly and directly by environmental factors in ways that have only recently been considered (Bentz et al., 2018).

While environmental factors may impact the way that languages evolve over the long-term, speech can conversely impact the immediate environment in invisible and inaudible ways. As people speak, they do not simply emit energy via the propagation of sound waves. They also emit air molecules and particles, including aerosolized particles. Aerosol particles are suspended in the air and often defined as ranging in size from 10 nm to 5 μm in diameter. Particles larger than this (i.e., droplets) are also generated during speech, as described in the literature (e.g., Stadnytskyi et al., 2020). Although 5 μm is often used as a cut-off to distinguish aerosols from droplets, a size of ~100 μm should be considered as an alternative cut-off as this figure denotes the largest particle size that can remain suspended in still air for more than 5 s from a height of 1.5 m (Wang et al., 2021; Darquenne et al., 2022). Our focus here is on the airflow and aerosol particles generated during speech. In the following section, we describe a new method developed for simultaneously capturing acoustic, airflow, and aerosol particle data during speech. In the remainder of this section, we offer some relevant background from the literature on the production of airflow and aerosols.

Humans produce air molecules, including carbon dioxide, oxygen, and nitrogen, during expiratory activities like speaking and singing. These molecules are only a fraction of nanometers in size, but are exhaled in tremendous volume with airflow. There are numerous findings in phonetics and biomedicine demonstrating how certain kinds of articulations yield varying amounts of airflow. We focus here on the airflow findings related to consonants in English, as this is relevant to our subsequent discussion of aerosol particles. Vowels typically have limited peak airflow, and there is little variation in peak airflow between vowels (Baken and Orlikoff, 2000, chapter 9). More specifically, we focus on key results in the literature related to the peak airflow of word-initial and word-final consonants, as measured in mL/s. It is important to note that airflow varies substantially according to body size and lung capacity, at least in the case of egressive

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pulmonic consonants. Stathopoulous (1980) examined the airflow associated with consonant production in English-speaking adults, teenagers, and children. Adults were found to produce significantly greater airflow across the same consonant types, with teenagers producing greater airflow than younger children. The findings were based on word-initial and word-final consonants, and clear patterns also emerged across consonant types. Nasal consonants were not included in the analysis, which focused on oral airflow. The consonants associated with the lowest peak airflow were word-final voiced stops and fricatives. Voiceless plosives and fricatives, particularly in word-initial contexts, were associated with greater peak airflow. The reduced airflow associated with voiced consonants is due in part to the blockage of the airstream at the glottis during vocal cord vibration, which limits peak egressive airflow. This same factor limits the peak airflow of vowels.

In Figure 1, we offer a visualization of peak airflow across key English consonants, based on relevant data in Stathopoulous (1980). In the figure, the greater peak airflow associated with word-initial voiceless consonants, in particular word-initial aspirated plosives, is readily apparent. These data are based on averages for 10 adults (five male), 10 teenagers (five male), and 10 children (five male). Note that the aspirated consonants of adults yield peak airflow up to three times greater than that evident in other consonants tested, with the mean peak airflow exceeding 1,700 mL/s. Given the average adult male lung vital capacity is roughly 6 L; this suggests that a significant portion of pulmonic air can be used during the production of aspirated consonants. The anomalous nature of aspirated consonants is also evident in our airflow data, some of which are presented below. It is worth noting that, while common in English, aspirated consonants are not particularly frequent cross-linguistically. This is supported by an inspection of PHOIBLE, the most extensive database on phoneme inventories worldwide (Moran and McCloy, 2019). Judging from the 3,183 phoneme inventories represented in PHOIBLE, [t^h] is found in fewer than one fifth of the world's languages. It is found in 17% of inventories, while [p^h] and [k^h] are slightly more prevalent, each occurring in roughly 20% of inventories. [p^h] is found in 592, while [k^h] is slightly more common, being documented in 605. While not particularly frequent cross linguistically, these sounds are hardly typological rarities either. Intriguingly, it has been speculated that aspirated consonants may be associated with greater likelihood of airborne pathogen transmission during speech (Inouye, 2003). While this remains a speculation, the approach we present in the next section allows for the detection of both airflow and aerosol particles, which can potentially host pathogens, offering a less speculative route to the future exploration of this and other related issues.

While research on the airflow associated with the production of speech dates back decades, only in the last few years have studies begun to emerge that address the aerosol particles produced during speech. New devices allow for the detection of aerosol particles, though such devices are generally applied to nonlinguistic phenomena. They can, however, be adapted to explore the production of aerosols during speech. Speech-based aerosols have received increased attention in the last several years due to the advent of such devices and associated instrumental adaptation, and also due to the fact that it became increasingly clear that such speech-based particles were relevant to the transmission of the SARS-CoV-2 virus among asymptomatic individuals (Abkarian et al., 2020; Fennelly, 2020; Meselson, 2020). Case studies demonstrated early in 2020 that speakers and singers could transmit this virus, yielding a push to better understand the mechanisms through which humans produce viral-laden particles during speech (Hamner et al., 2020; Bahl et al., 2021). That push remains underway, and a variety of methods are being deployed to better illuminate how exactly aerosols are generated during the articulation of sounds. These methods include the utilization of laser sheets and aerodynamic particle sizers to isolate the size distribution of miniscule particles produced during specific articulatory gestures (Stadnytskyi et al., 2020). Work relying on an aerodynamic particle sizer (APS) has suggested, for instance, that the high front vowel /i/ yields an inordinate number of aerosol particles when contrasted to other phonemes in English (Asadi et al., 2020).

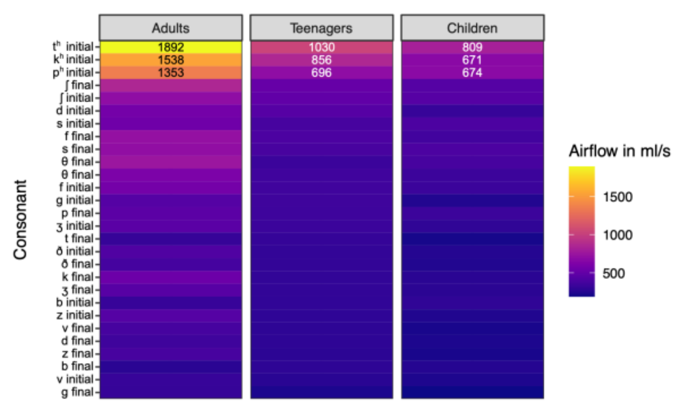

FIGURE 1
Heatmap of peak airflow associated with plosives and fricatives in English. Based on data in Stathopoulous (1980). The top three airflow values for each speaker category are provided on each of the appropriate bars.

More critically, APS-based research has suggested that the volume of aerosol particles produced during speech is a function, at least in part, of the amplitude at which the vocal cords vibrate (Asadi et al., 2019). Judging from such work, vocal cord vibration appears to be the chief mechanism through which aerosols are produced during speech. There are two key caveats to this conclusion, however. First, work to date has not simultaneously examined airflow, aerosol, and acoustic data. Instead, the conclusion has been based on research demonstrating an association between increased amplitude of vocal cord vibration and aerosol production. Given that increased amplitude of vocal cord vibration is achieved partially through greater airflow through the glottis, such an approach makes it difficult to disentangle the relative contributions of amplitude and airflow. The approach we outline below allows for such disentanglement since it includes simultaneous measures of airflow, aerosol, and acoustic data. A second caveat associated with the relevant conclusions in the literature, vis-à-vis the association of sounds like /i/ and increased aerosols, is that they rely on a method with limited temporal resolution. The APS used in such studies samples air once per second. Since words, syllables and in particular phonemes typically last less than 1 s, this means that the method requires the repetition of stimuli over a particular duration, during which time the total number of aerosols is measured (Greenberg et al., 2003; Asadi et al., 2020). This number of aerosols is then correlated with the number of particular sound types, for instance /i/, in a given set of phonetic stimuli. Thus, testing aerosols once per second does not allow for the direct observation of the production of aerosols during specific articulatory gestures. In part for this reason, we developed an approach with greater temporal resolution, one that allows us to sample air 10 times per second, to more confidently make assessments regarding the role of individual articulatory gestures in aerosol production. Such heightened physical resolution is critical to better isolating the extent to which vocal cord vibration or alternate mechanisms actually produce aerosols. We return to this point below. Our approach also allows for a greater physical resolution, with the potential to observe aerosols with diameters as small as 70 nm, or about the size of some airborne viruses. Previous approaches generally allow only for the isolation of those particles greater than 500 nm in diameter (Morawska et al., 2009; Asadi et al., 2020). Some airborne virions, which are infective forms of viruses, can be hosted by particles as small as 90 nm in diameter, so capturing particles in this size range is potentially relevant to speech-based viral transmission (Lee, 2020).

More broadly, the approach we describe could eventually help to impact public health guidance related to speech during future airborne pandemics. Some widely disseminated guidance in 2021 suggested that people should reduce vocal cord vibration via whispering, in order to reduce the risk of transmitting the SARS-CoV-2 virus (Thompson, 2020). As we will see below, further work is needed to support such guidance and some of our preliminary findings are inconsistent with this suggestion. Relatedly, there has been some speculation in prominent venues like The Lancet that consonant aspiration could help to transmit airborne viruses (Inouye, 2003). We avoid such speculations here, though we return to aspiration below as our preliminary results suggest that it produces a greater number of aerosols alongside the increase in peak airflow. Such results, while quite preliminary and requiring caution to interpret, demonstrate that exploration of this understudied topic could help to elucidate our understanding of airborne disease transmission during speech. While air molecules do not transport pathogens, aerosol particles that can do so are suspended within that airflow (Wang et al., 2021). Characterizing these aerosolized particles is key to quantifying and modeling respiratory pathogen transmission risk, especially since small particles (<3 μm) penetrate deeper into the lung and infection in the lower respiratory tract requires fewer numbers of pathogens to produce lethal infection in animal models (Thomas, 2013). Additionally, depending on the primary mode of transmission of an infectious respiratory pathogen, understanding the size of particles produced during speech can have significant implications on use and effectiveness of non-pharmaceutical interventions for transmission mitigation in an outbreak setting (Leung, 2021). The first step in this elucidation is, in our view, to illuminate in greater detail the actual articulatory mechanisms through which airflow and aerosols are produced. Regardless of its potential eventual influence on our understanding of airborne pathogen transmission, however, this illumination will allow us to better understand the invisible effects of speech on the proximate physical environment. In the following section, we discuss this new approach, illustrating how it allows for the isolation of the aerosols produced by both aspiration and vocal cord vibration.

2. Examining the phonetic production of airflow and aerosols via a new approach

In this section, we first offer some new data on airflow, which is relevant to contextualizing our approach. We then describe the method being used to analyze airflow, aerosol, and acoustic data simultaneously. Finally, we offer some very preliminary data with this approach, based on the speech of two of the authors. These preliminary data demonstrate how the method allows for the isolation of the role of individual articulatory gestures in the production of aerosols. Further, the preliminary data suggest that aspiration produces an inordinate number of aerosol particles below the threshold of detection of previous methods.

We analyzed the airflow of 12 fluent English speakers (six male), to better contextualize our examinations of aerosol production. To do so, speakers wore a mask connected to a pneumotachograph (Fleisch no. 1, OEM Medical, Richmond, VA, United States) to record flow as they sang "happy birthday," but also as they whispered "happy birthday" and as they spoke the words to the song, at a normal amplitude and at a loud amplitude. Mean flow rate and exhaled volume were averaged over four repetitions of the song for each modality. During normal speech, speakers produced an average of 150 mL/s of airflow and exhaled an average of 1.2 L of air throughout "happy birthday," though there was variation across speakers as we might expect. Mean airflow and exhaled volume across speakers was 157 ± 42 mL/s and 1,204 ± 339 mL [average ± standard deviation (SD), $N = 12$]. In Figure 2, we present the normalized mean airflow and exhaled volume across modalities. In the figure, each speaker's normal speech airflow and exhaled volume are set to one and the other modalities are presented as a ratio of the airflow and exhaled volume to that of normal speech, respectively. Four of the speakers exhibited a pronounced increase in airflow and exhaled volume of air during whispering, with one speaker producing nine times the flow rate and eight times the exhaled volume as he did while speaking at a normal

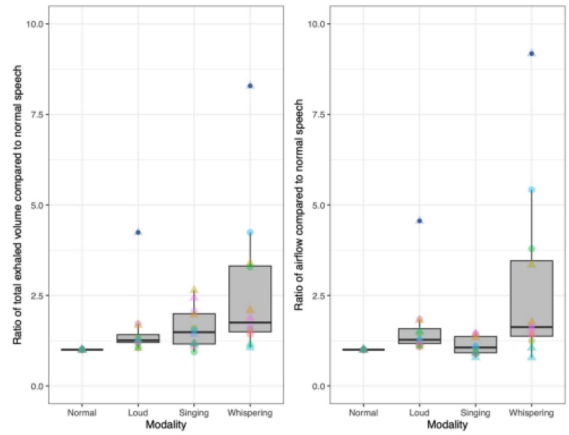

FIGURE 2
Normalized exhaled volumes **(left)** and airflow **(right)** across modalities for 12 speakers (six male), based on each speaker's exhaled volume/airflow as a ratio of their mean exhaled volume/airflow during normal speech. Triangles represent male speakers. Each color corresponds to an individual.

volume. Another subject produced five times the flow rate and four times the exhaled volume during whispering when compared with normal speech. Whispering involves a constricted glottis without vibrating vocal folds, so airflow is not regularly blocked as it is with sounds like vowels (Sundberg et al., 2010). This point is relevant to the production of aerosol particles. There are several potential mechanisms for the production of such particles in the respiratory tract. Two of these are particularly relevant to this discussion. One involves a fluid-film burst in the bronchioles, which creates aerosols that can then be emitted. The larger the exhaled volume is the greater the number of exhaled aerosols and thus the greater the concentration in the surrounding environment. Aerosols originating deep in the respiratory tract via this mechanism may have a greater likelihood of transmitting viral pathogens (Lindsley et al., 2016). A second relevant mechanism for aerosol generation is the vibration of the vocal cords, the viscous covering of which can burst into particles including tiny aerosol particles. The higher the exhaled flow rate is, the higher the shear stress and the greater the aerosol generation. This mechanism is presumably responsible for the increased aerosols associated with vowels, particularly loud vowels, in the literature (Asadi et al., 2019). However, as noted above most studies in the literature did not detect particles smaller than 500 nm in diameter.

For this background airflow analysis, we also recorded the speakers as they produced individual words and two vowels, [a] and [i], at a normal amplitude. Three pairs of words were recorded: (1) "spar" and "par," (2) "star," and "tar," and (3) "scar" and "car." For each of these pairs, the first word includes an aspirated plosive while the second includes a non-aspirated version of the same voiceless plosive, i.e., made at the same place of articulation. As apparent in Figure 3, the peak airflow associated with aspirated voiceless plosives was noticeably greater than that associated with non-aspirated plosives, consistent with Figure 1. This increase was observed across all 12 speakers and at each place of articulation. The mean peak airflow

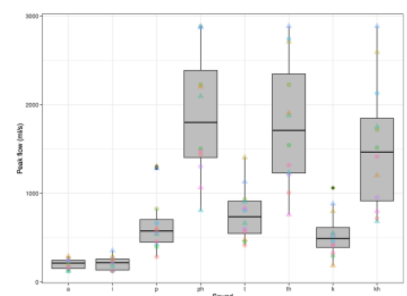

FIGURE 3
Peak airflow associated with two English vowels and six voiceless plosives, for 12 adults (six male). Triangles represent male speakers. Each color corresponds to an individual.

across all speakers was greatest for the aspirated voiceless bilabial stop, with a mean that exceeded 1,800 mL/s. The two vowels tested produced negligible peak airflow (means <100 mL/s).

This context on the airflow associated with whispering and aspiration is useful to our ongoing exploration of the aerosols produced during speech. Since the airflow associated with whispering and aspiration is pulmonic and since neither whispering nor aspiration entail voicing, it is expected that any aerosols detected during such speech activities are due to the fluid-film burst mechanism, originating from deep within the respiratory tract. Further, the quantification of the airflow associated with voiced sounds like [a] and [i] helps to illuminate the extent to which aerosols observed during the production of such sounds are due directly to vocal cord vibration, or potentially due to the increased airflow associated with greater amplitude of vocal cord vibration. As observed in Figure 2, there is typically an increase in the mean airflow for loud speech, when compared to speech at a normal amplitude. As noted above, this complicates the interpretation of the results in the literature suggesting that the aerosol increase associated with loud vowels is due in a straightforward manner to the increase in the amplitude of vocal cord vibration as opposed to airflow carrying aerosols from deeper within the respiratory tract.

This background on airflow associated with both aspiration and vocal cord vibration serves as critical contextualization of our discussion of the aerosol production owing to these key articulatory gestures. Here we focus on these gestures to illustrate our new method for simultaneously capturing aerosol, airflow, and acoustic data. Ongoing research utilizing the method is exploring aerosol production with a large number of speakers in the lab of the last author. Previous work has simultaneously examined airflow and acoustic data (e.g., Yu et al., 2022), but no studies to date have illustrated a method capturing these data alongside aerosol data. The method we have developed is described schematically in Figure 4. Experiments proceed as follows: Participants sit alone in a mini clean room surrounded by a downward laminar flow of HEPA-filtered air, which creates an environment that is nearly free of background aerosols. They then read prepared stimuli off of a screen, into a rubber mask that is attached to their mouths. The rubber mask leads directly into a custom-built stainless steel particle sampling manifold, which curves gently into an electrical low-pressure particle impactor (ELPI+, Dekati Ltd.) that measures aerosols from 70 nm to 10 μm in size (Järvinen et al., 2014). Details of this particular ELPI+ are provided in Tumminello et al. (2021). Pure air is fed into the manifold at a rate of 11 L per minute. A flow meter detects fluctuations in this airflow resulting from the incoming airflow generated by the speakers. Above the facemask, there is a microphone

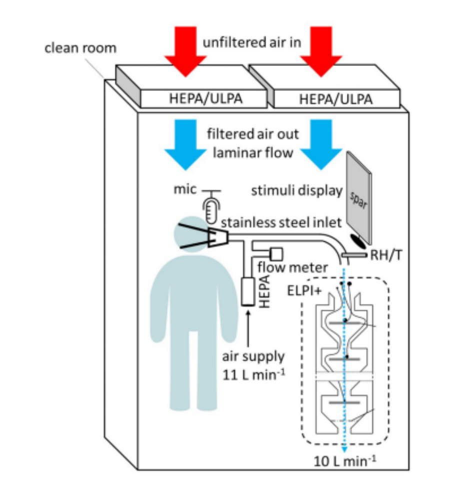

FIGURE 4
Schematic of new method for simultaneously capturing data on airflow, sound types, aerosols, temperature, and humidity.

which records audio stimuli directly to a laptop computer at 44.1 kHZ, via PRAAT (Boersma and Weenink, 2023). As the vacuum pump necessary for ELPI+ operation is not quiet, the resultant waveforms and spectrograms do include some background noise. Given that our present focus requires only coarse acoustic data to interpret key articulatory gestures, this does not present an issue, particularly given that the airflow data yield clear signatures for vocal cord vibration and aspiration (see Figure 5). For future analyses with more acoustic detail required, we aim to use sound proofing materials in the setup. It is also worth noting that the relative humidity and temperature of the air leading into the ELPI+ is measured, allowing us to test the effect of humidity on the number distribution of aerosol particle sizes. Humidity is well known to affect the ways that speech-generated particles interact with the surrounding air (De Oliveira et al., 2021).

Upon entering the ELPI+ inlet, the speech aerosol particles are initially charged with a positive corona charger before traveling down through the impactor. The unipolarly charged particles are then collected at each impactor stage on high surface area sintered plates, which are coated with a thin layer of high viscosity vacuum grease to maximize collection efficiency. Particles are size segregated by their aerodynamic diameter over 14 stages, ranging from 10 μm at the inlet to 5 nm at the bottom stage of the impactor. Particle collection is measured by sensitive electrometers (fAmp sensitivity) on each stage at a sampling rate of 10 Hz. The resulting currents are converted to number concentrations based on particle size.

Across both speakers whose aerosols have been measured without background particles (both males), we have found that aspiration is associated with an increase in the production of submicron particles. Given that we have only tested two speakers with this method, we stress that these results are meant only to illustrate the enhanced physical and temporal resolution of our method. In Figure 6, the physical resolution of the method is demonstrated. Based on averages of five iterations each of the words "spar" and "par," we see that the word "par," beginning with a voiceless aspirated bilabial plosive, is associated with an increase in aerosol particles with diameters of around 300–500 nm. Note that such particles were not detectable in most previous studies relying on an APS, which is limited to particles greater than 500 nm. Further, we see in Figure 6 that speech produces dozens of aerosol particles in the case of both words, while the background particles are nearly nonexistent or below the instrumental detection limit in the clean room environment. Nevertheless, there are some background particles and these fluctuate slightly under the laminar hood. This is evidenced by the slight differences in the red lines for panels A and B in Figure 6. Note also that there is some variation in the number of larger particles (diameter > 1 μm) produced during the words "spar" and "par" in these instances. These variations could be due to slightly louder productions of the vowel in the word "par," or to random fluctuations for these particular instances of these words. We stress that these results are preliminary and that we aim to run these tests with many individuals and sound stimuli prior to drawing conclusions about the associations between particular sound types and their associated aerosols. This will be necessary to reduce the effect of noise in the data, but also to reduce the undue influence of idiosyncratic findings associated with individual speakers.

The method offers a more critical advantage for exploring the invisible effects of speech on the environment: It allows for fine-grained temporal resolution given the 10 Hz sampling capacity of the ELPI+. In Figure 5, this temporal resolution is illustrated via an

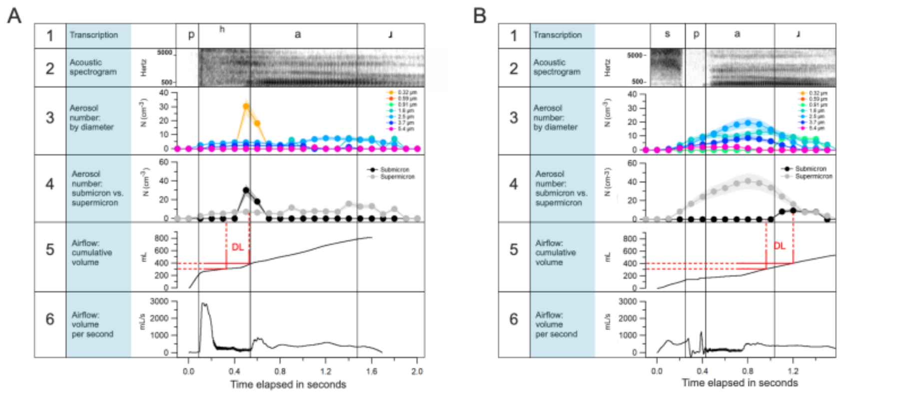

FIGURE 5
Temporal resolution allowed by the method. Aerosols as naturally produced during a speaker's articulation of "par" **(A)** and "spar" **(B)** with spectrogram, airflow, and aerosol data offered simultaneously. In A5 and B5, "DL" refers to the approximate range of cumulative volume of air over which air from the "deep lung" gets emitted and corresponds to the increase in submicron respiratory aerosol concentration in A4 and B4, respectively.

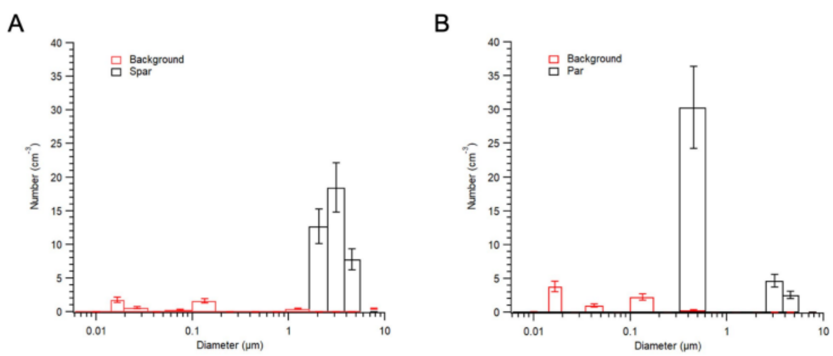

FIGURE 6
Physical resolution of the method, as evidenced by aerosols detected during one speaker's articulation of "spar" **(A)** and "par" **(B)**. Note that during the speaker's productions of the word "par" there was a particularly pronounced increase in submicron particles, likely due to the aspiration of the first sound of the word. All submicron aerosol data in red (background) are below instrumental detection limit and cannot be attributed to aerosol.

analysis of the first author's deliberate articulation of two words, "par" and "spar." As evident in panel A of the figure, there is a peak in submicron aerosols immediately after the burst of airflow owing to the aspirated bilabial plosive in "par." This aerosol burst coincides with the point at which the cumulative exhaled volume exceeds 300–400 mL, which is consistent with work suggesting that tiny aerosols generated deep in the lungs are emitted from volumetric depths beyond the anatomical dead space (i.e., volume of air in airways down to the respiratory bronchioles) during expiratory activities (Gebhart et al., 1988). A similar pattern is observed in panel B, but note that the 400 mL threshold is achieved much later in the word due to the lack of aspiration in the word "spar." In panels A and B, we observe that larger aerosol particles, greater than 1 µm in diameter, are generated shortly after the vocal cords begin to vibrate, as evident in the alignment with the spectrogram. This is consistent with the literature that has focused on vocal cord vibration as a source of larger aerosol particles. Our preliminary results suggest, then, that the two aforementioned potential loci of the origination of speech-generated aerosols, the vocal cords and the bronchioles, are detectable and isolated via our method. That is, it appears we are able to detect when aerosols are generated at the glottis during vocal cord vibration, and when they are generated deep within the respiratory tract and emitted alongside airflow such as that characteristic of aspiration. Of course, we need much more data before offering any conclusions on the role that individual articulatory gestures play in aerosol production. To that end, future work will test dozens of English speakers to more carefully isolate the roles that consonant aspiration and vocal cord vibration play in generating aerosol particles during speech.

Finally, while we think this method represents a step forward in terms of how we might investigate the precise mechanisms through which speech generates aerosols, we also recognize that the approach has limitations and should be complemented by other approaches. One limitation is that speakers must wear a tight-fitting mask during the tests and must face the same direction during the whole test. Similarly, the equipment used is not quiet, so speakers may compensate by increasing their loudness to more clearly hear themselves speak. In short, while the method offers advances it does not allow us to test the aerosols produced in natural conversation-like settings. No method available to date allows this. We should also mention that this work is limited in that we are only examining English speakers at present. In the future we hope to test speakers of other languages.

3. Conclusion

We began this paper by discussing some of the proposed invisible effects of the environment on how people speak. We then focused our discussion on the converse issue that has received even less attention in language research: the invisible and inaudible effects of speech on the immediate environment. This topic offers two key gains, when contrasted to the exploration of the ways in which languages are affected by their environments. First, the topic can be addressed more directly via experimentation, though that experimentation presents a number of challenges and requires costly equipment. Second, exploration of this topic has the potential to do more than shed light on the nature of language and its relationship to the physical environment. Such exploration may ultimately yield health guidance related to speech that is firmly founded on a clearer understanding of how sounds generate potentially viral laden aerosol particles. In short, the issue has potential relevance not just to our understanding of speech, but perhaps to contemporary medicine as well. The precise articulatory mechanisms that help transmit pathogens during conversations are still not fully understood, but hopefully that will change in the coming years. Here we have described a new method that could assist in the elucidation of those mechanisms.

Data availability statement

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding author.

Ethics statement

The studies involving human participants were reviewed and approved by UCSD IRB. The patients/participants provided their written informed consent to participate in this study.

Author contributions

CE, MS, CD, and JS conceptualized, funded, and supervised the study. CE wrote the original manuscript draft and analyzed the acoustic data. CE, MS, RN, PT, CD, and JS reviewed and edited the manuscript. PT, RN, and JS performed the aerosol measurements and analyzed the aerosol data. CD performed the volume flow measurements and analyzed the volume flow data. CD, CE, MS, JS, PT, and RN collected the data. All authors contributed to the article and approved the submitted version.

Funding

CD acknowledges funding by National Institute of Health grant U01 ES028669. PT and JS acknowledge funding support by the National Science Foundation Center for Aerosol Impacts on Chemistry of the Environment under Grant CHE-1801971. RN acknowledges support by a National Science Foundation Graduate Research Fellowship.

Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Acknowledgements

Chapter 1, in full, is a reprint of the material as it appears in *Frontiers of Psychology*. Everett, C., Darquenne, C., Niles, R., Seifert, M., Tumminello, P. R., & Slade, J. H. (2023). Aerosols, airflow, and more: Examining the interaction of speech and the physical environment. *Frontiers in Psychology*, *14*, 1184054. The dissertation author was a primary researcher and co-authored this paper.

Chapter 2 FUTURE WORK

Aim 1: Optimize the ELPI for respiratory aerosol measurements.

A previous study used the ELPI to detect respiratory aerosol while participants coughed. Particle number concentrations were reported on the order of 100s to ~1000 cm^{-3}, significantly above any previous measurement reported.[30] This disparity indicates possible artifacts in measuring respiratory aerosol with this approach, which must be addressed. The ELPI is a cascade impactor with fourteen stages that operates at a nominal flow rate of 10 L/min, and its operating principle can be seen in Figure 3. The bottom stage controls the flow rate, serving as a critical orifice. The particles are first charged at the inlet with a unipolar corona charger to their maximum

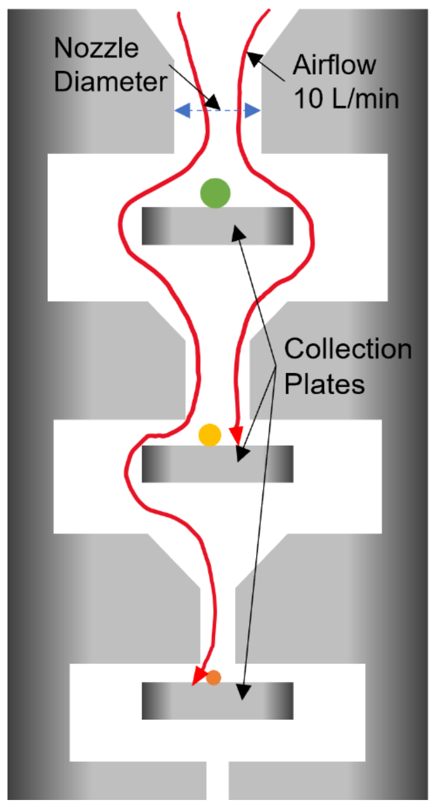

Figure 2.1 Cascade impactor operating principle. The colored circles represent aerosol particles.

charge capacity, dependent on their Stokes diameter.[41] The charged particles get collected as they cascade down to the filter stage. The size segregation is achieved at each of the 14 stages within the device. The stages are a collectively stacked series of jet orifices with progressively smaller nozzle sizes, which determine the particle sizes allowed to pass through each, coupled with collection plates at each stage between each orifice. The collection plates and nozzles are positioned according to the mechanical mobility of particles with certain aerodynamic diameters. The flow rate is constant at 10 L/min, which ensures that only particles with the desired diameter are collected on their respective plates. The particles smaller than the desired diameter at that plate can continue following the airflow stream down the impactor until reaching their specific nozzle and plate combination. The quantification of the particles is measured by taking the difference between the current, I, before a collection plate and the current after the collection plate, measured in femtoamperes (fA) with sensitive electrometers. This ΔI is then converted into number concentrations for the sizes of each collection plate with a correction factor (Eq. 1), where N is the number concentration in particles cm^{-3} and X is the charging efficiency, which is dependent on the particle's Stokes diameter.[42,43]

$$N = \frac{1}{X} * Dilution \qquad (1)$$

It is well-known that the electrometers responsible for producing the current readings are susceptible to vibrational noise and induced currents caused by rapidly changing particle concentrations at the inlet.[44] However, I have found that more factors produce artifacts in the readings that have not been previously reported.

One such factor that leads to false readings is pressure changes at the ELPI inlet. Figure 4 shows the ELPI response (ΔI in gray) during controlled injections of aerosol-free air at different airflow rates and "bursts" in relative humidity. The first four conditions in Figure 4 illustrate that even a few millibars increase in pressure above ambient at the inlet (ΔP in green), in the absence of particles or changing relative humidity (Δ%RH, blue), produces a notable increase in the ELPI signal relative to the background. This indicates a direct correlation between inlet pressure change and current, likely due to piezoelectric behaviors exhibited by the insulators positioned between each collection plate in response to the physical force they experience.[45] This can be easily mitigated by increasing the inner diameter of the tubing that transports the particles from the subject to the ELPI, as shown for the last two conditions in Figure 4. These tests illustrate the necessity to account for pressure changes, which otherwise would lead to overestimations in respiratory aerosol concentrations.

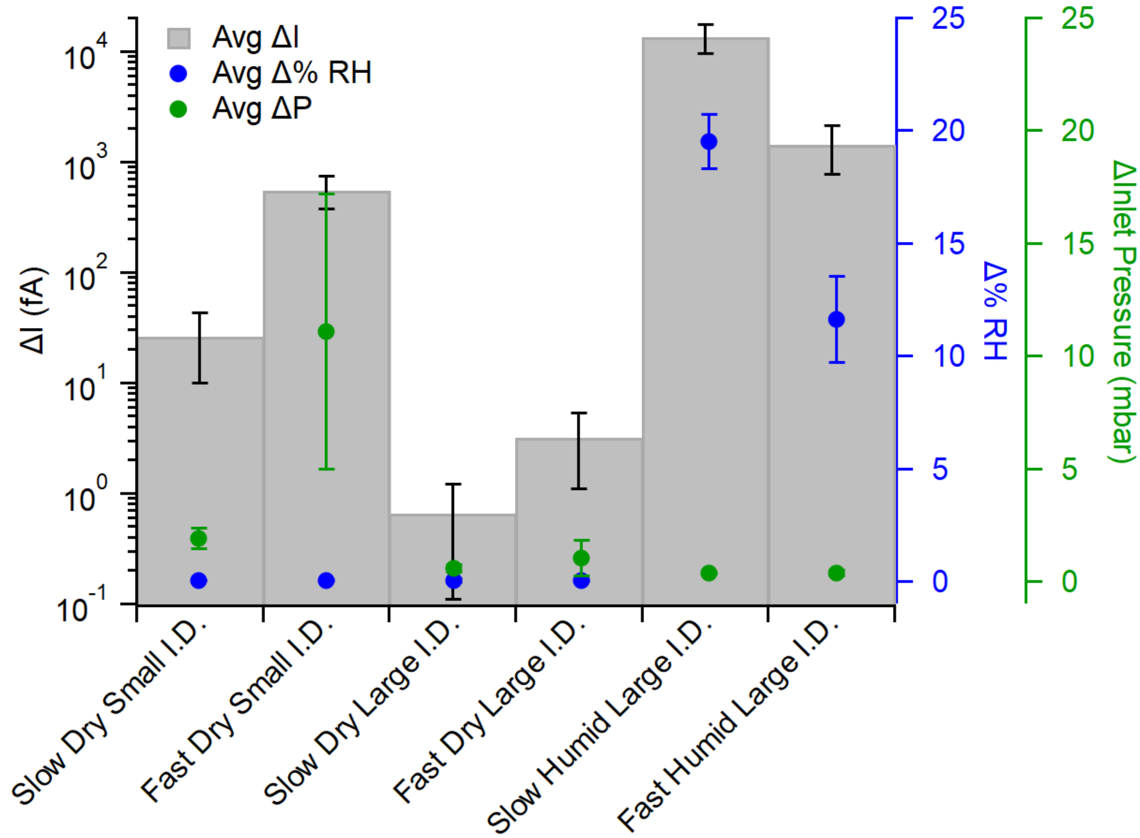

Figure 2.2 Plots showing the effects of inlet pressure changes and relative humidity changes on ELPI current responses.

Another condition during respiratory aerosol measurements that we found produces a false positive signal is Δ%RH, induced by the exhalation of high-humidity air in human breath. This effect is illustrated in the last two conditions presented in Fig. 4. While the pressure did not change upon injection, the increasing RH caused a significant increase in the detected current (i.e., particle counts). Since the air emitted from respiratory activities typically exits at RH ~90%[46] and ambient RH typically sits around 30-80%,[47] there may be large RH changes during the measurements if not adequately compensated for via a drying agent installed inline before the ELPI inlet. This effect may be due to condensation within the impactor and on the insulators since the RH within the instrument is closer to the ambient RH, typically much lower than the exhaled RH. Thus, finding a suitable desiccant safe for participants when incorporated into the setup is an ongoing pursuit for optimizing the ELPI for these measurements.

Aim 2: Develop an integrated setup for simultaneous *in-situ* measurements of acoustics, airflow, and particle distributions produced by respiratory activities.

The uncertainties in the effects of vocal amplitude (loudness) versus airflow rates on aerosol emissions have motivated the development of a method to measure aerosol, airflow, and acoustics during speech simultaneously. Currently, I have designed a setup that is enclosed in a "clean room" respiratory aerosol chamber (Terra Universal Product, CleanBooth™ Laminar Flow Station), as seen in Figure 5 (Everett et al., 2023). The clean room supplies a continuous laminar flow of HEPA/ULPA-filtered air from the top, creating a positive pressure differential preventing any ambient particles from entering the space. This is essential to the setup since the

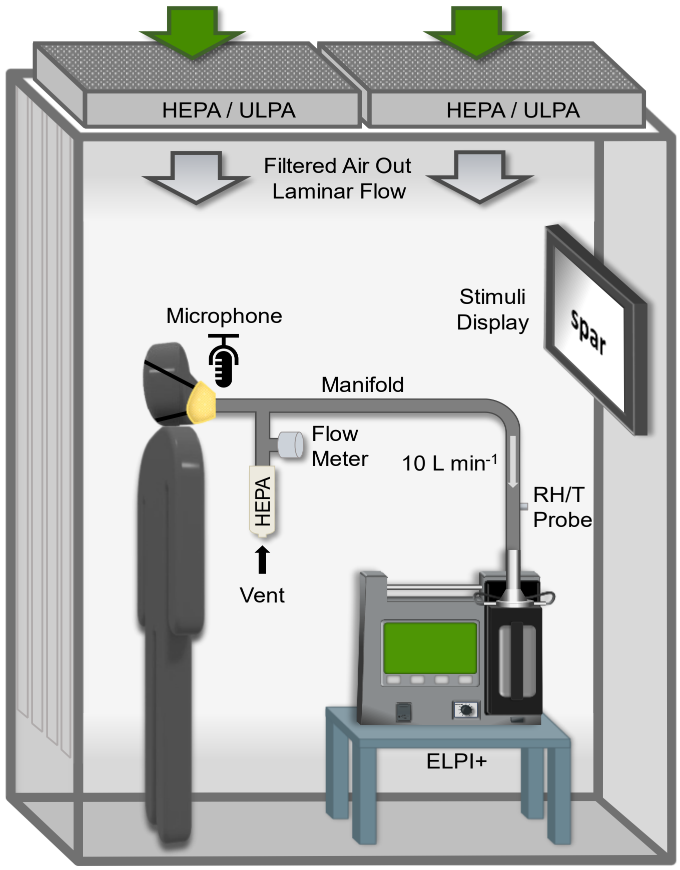

Figure 2.3 Diagram of the setup for integrated in-situ particle, airflow, and acoustics measurements.

concentrations of speech aerosols are very low, and any leakage of room aerosol into the setup would artificially enhance the measured aerosol concentrations. As shown in Figure 6, the background aerosol concentrations are below the ELPI detection limit and significantly below the measured respiratory aerosol concentration for one individual's iteration of the word "spar."

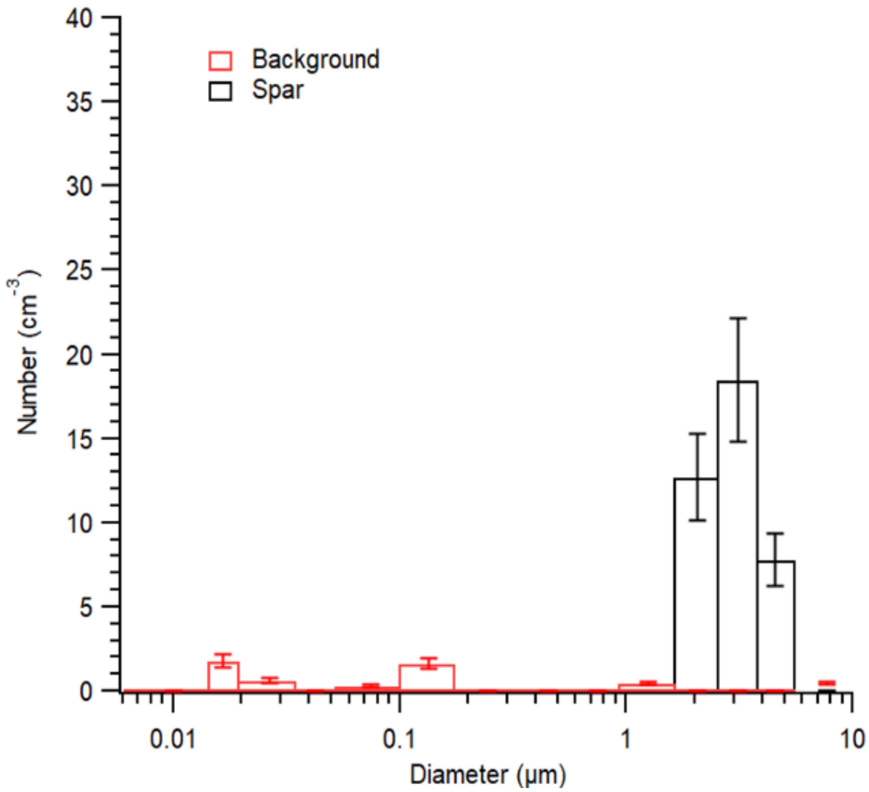

Figure 2.4 Size distribution and total number concentrations measured by the ELPI of a subject saying the word "spar" versus the background concentrations.

The other integrated measurements include a microphone and a pneumotachograph (Fleisch no. 1, OEM Medical, Richmond, VA, United States). The pneumotachograph measures the airflows at a rate of 1000 Hz, on the timescale of vocal fold vibrations. This device operates by measuring a thin diaphragm's positive and negative displacement from its resting position, which requires a closed system up to the device for accurate flow measurements. To create this closed system, the setup incorporates a silicone mask that straps onto the subject's head and creates a seal on the face. An airtight manifold connects it directly to the ELPI inlet and pneumotachograph. The acoustics are measured with a microphone placed directly in front of the subject.[40]

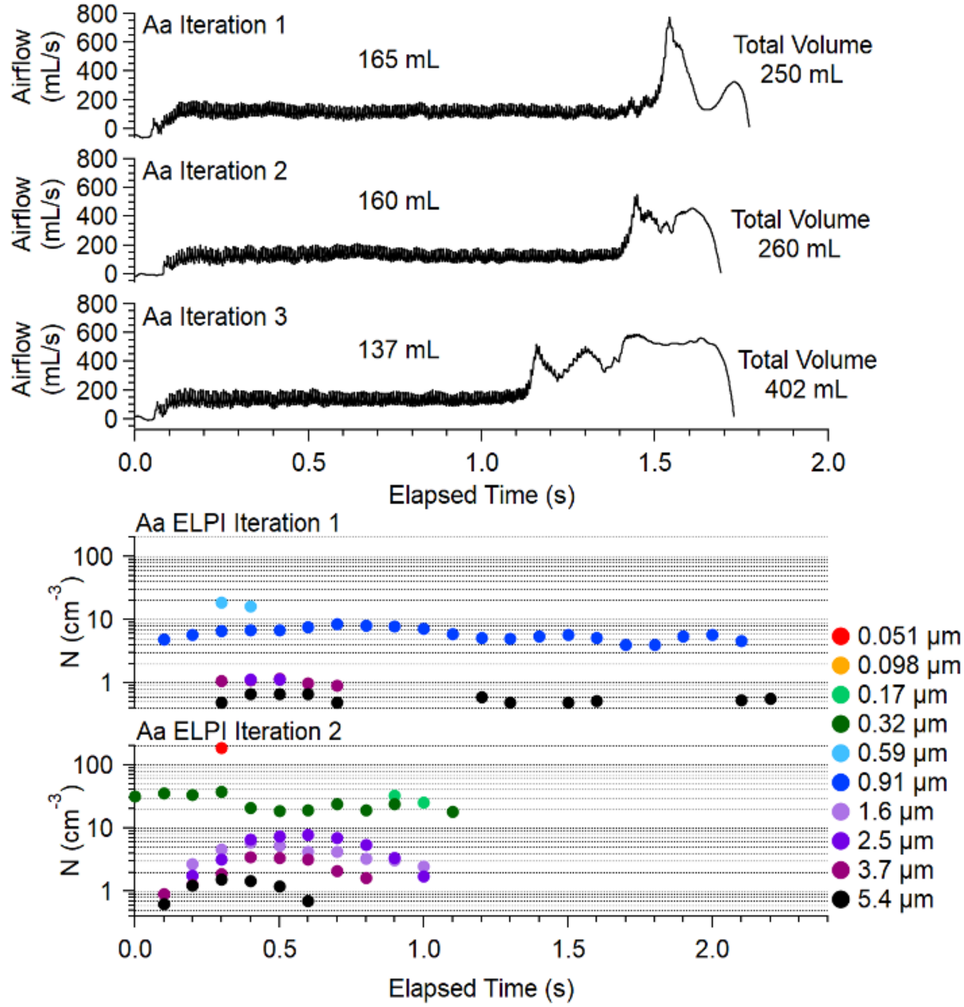

Figure 2.5 Airflow and particle distribution timeseries of one individual doing isolated repetitions of the sound "aa".

In these initial tests, I discovered a high variability in an individual's performance of the same respiratory task regarding airflow and size distribution. Figure 7 shows this variability with the same subject saying the sound "aa" multiple times, giving varied airflows and size distributions. This highlights the need for an integrated system to measure the effects of vocal cord amplitude and airflows on particle generation and emission. All three data types need to be correctly time-aligned to disentangle the degree of influence that each aspect has on aerosol production by isolating certain respiratory activities of interest.

My future work will focus on isolating the effects of airflow rate and vocal loudness on aerosol emissions during speech. The aerosol concentrations and airflow rates during vocal and non-vocal cord vibrations can be isolated by having participants speak an individual sound in replicate at different loudnesses, from whispering to shouting. I will measure the aerosol concentration as a function of vocal amplitude (measured in decibels, dB), and volume flow. It is hypothesized that an increase in loudness will increase the integrated volume of air emitted and, thus, the total particles that get sampled. This integrated approach will isolate the effect of vocal amplitude on aerosol emissions during speech, which has not previously been done.

Aim 3: Characterize the hygroscopic growth and phase states of substrate-collected respiratory aerosol particles using atomic force microscopy.

Physicochemical properties such as hygroscopicity, phase state (i.e., viscosity), and aerosol morphology significantly impact aerosol interactions and atmospheric lifetimes.[48] These properties are also expected to impact the ability of respiratory aerosols to host and transport virions.[49] Their composition influences the hygroscopicity of particles. One study investigated hygroscopic growth using model lung-origin respiratory aerosols composed of 87% mass fraction of sodium, potassium, calcium, and magnesium salts and 13% mass fraction of the organic compounds: ascorbic acid, uric acid, glutathione, albumin, cysteine, dipalmitoylphosphatidylcholine, glycine, and mucin. They suspended single particles with a linear quadrupole electrodynamic balance and modulated the RH while monitoring the particle's size with Mie resonance spectroscopy. To obtain the hygroscopic growth factor (HGF), they calculated the hydrated particle radius ratio to the dry particle's radius. This study found that the particles' HGF was closer to that of the inorganic salts than the organic compounds.[50] As previously mentioned, size is a major factor in the airborne lifetimes of particles. The uptake or loss of water will modulate the aerosol size depending on the hygroscopicity, thus affecting its duration in the air, which influences infection potential. Water loss and uptake also impact virion survivability on the surfaces of particles.[51,52] Studies have shown a negative relationship between virus viability and particle water content attributed to decreased surface activation or salt toxicity.[51] The drying process that particles undergo after leaving the mouth, typically drying until thermodynamic equilibrium with the surrounding air, may also influence the lifetime of a virus on the particle through morphological changes. Phase separation may occur during dehydration, which can lead to a configuration in which the organic matter within the originally heterogeneously mixed hydrated particle, such as mucins, separates from the hygroscopic salts and forms a core-shell structure with a hydrophobic shell. The shell would then be a protective layer for viruses embedded in the particle bulk.[53]

Due to their relatively lower solubility in water than inorganic salts, the organic matter in particles generally promotes more viscous phase states.[54] When a particle phase separates, e.g., leaving an inorganic core and a hydrophobic organic shell, the particle will be more viscous at the interface than if it were homogeneously mixed. Phase transitions as a function of RH under sub-saturated conditions depend on the particle's viscosity. Particles with high viscosity tend to respond slower to changes in RH due to the outer low hygroscopicity organic shell, decreasing diffusion rates, and water uptake or loss rates. Lower viscosity particles respond more rapidly to environmental changes as they likely have a higher fraction of compounds with greater hygroscopicity, allowing diffusion and water uptake or loss to occur more readily.[55] This, in turn, impacts particle equilibration timescales, with less viscous particles having bulk diffusion timescales on the order of nanoseconds to seconds and more viscous particle timescales on the order of days to years.[56] Phase state also impacts the particle's reactivity, affecting diffusion rates of reactants between the particle surface and bulk.[56] Reactions between oxidants and pathogens hosted by particles can affect virion survivability or inhibit/deactivate the pathogens by modifying proteins or disrupting lipid bilayers.[56–59]

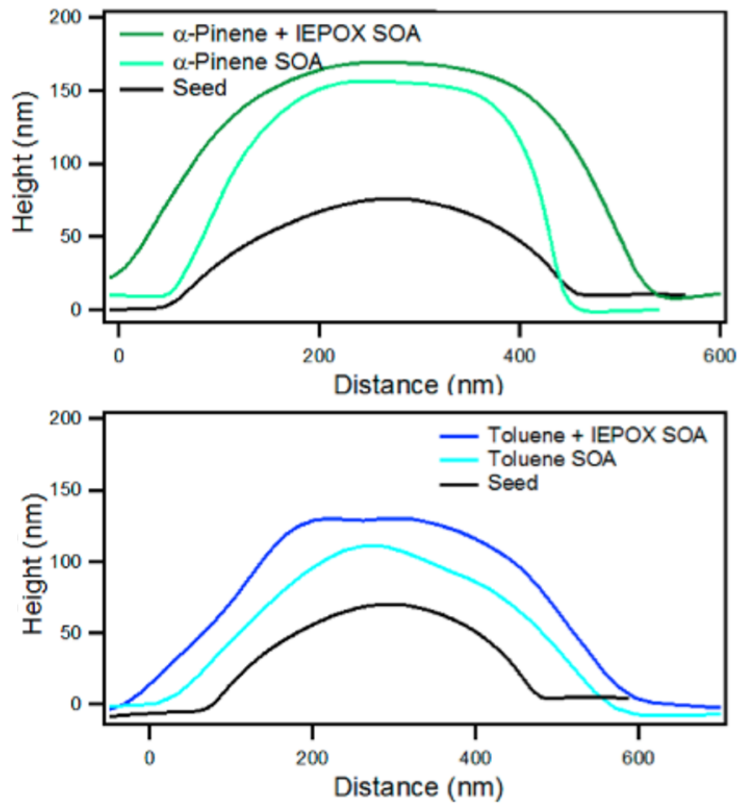

Figure 2.6 Example of spreading height images measured by AFM of two variations of aerosol particles comparing their profiles before and after modification which are used to calculate their spreading ratios. (Olson et al., 2019).

The phase states and hygroscopic growth of respiratory aerosol have remained insufficiently explored. I propose that respiratory aerosols that form via the film-bursting mechanisms that occur due to vocal fold vibrations will be more viscous and less hygroscopic than those formed at the bronchioles in the deep lung region due to the higher ratio of organic matter, such as mucins, to inorganic salts found in the laryngeal area.[50] The composition of the bronchial mucus includes hygroscopic salts, mucins, and surfactants. The greater hygroscopicity of the salts and surface tension depression from the surfactants could render the particles generated from the bronchial mucus less viscous. However, laryngeal-specific mucus is not so well documented in the literature other than it also contains mucins but lacks surfactants, so this will also be evaluated. One approach I plan to use to strengthen my understanding of these properties involves Atomic Force Microscopy-Infrared Spectroscopy (AFM-IR). AFM is a technique that can measure a particle's morphology at nanometer-level resolution by recording the displacement of a cantilever as it interacts with a particle's surface,[60] while IR can be used to gain insight into the particle's composition. I am collaborating with the Grassian lab to optimize their AFM-IR for detecting substrate-collected respiratory aerosols under controlled RH conditions in an environmental cell. I plan to utilize the ELPI to collect respiratory aerosols onto hydrophobic substrates. Aerosols will be prepared systematically in the lab by atomizing representative components identified in human respiratory aerosol, including mixtures with different mole ratios of inorganic salts, mucins, proteins, and organic molecules.[50] I will test the hypothesis that an increasing fraction of mucins in the particles will limit hygroscopic growth and promote more viscous phases. Tests on the lab-generated aerosol will serve as control experiments for the collected human respiratory aerosol to determine how the different components impact the particle hygroscopic growth and phase state. A subset of substrate-deposited human respiratory aerosol samples from Aim 2, generated from specific sound types to elicit respiratory aerosol emissions from a specific site in the respiratory tract, will be analyzed via AFM-IR. The substrates will be stored in nitrogen-purged (air-free) and sealed petri dishes immediately after collection. The substrates will be placed in the environmental chamber and subsequently hydrated and dehydrated while measuring their morphological and phase changes in response to these varying conditions.[61] As shown in other studies,[61–64] AFM height images and traces of individual particles can be used to calculate particle spreading ratios from Eq. 2,[64]

$$spreading\ ratio = \frac{r_p}{h_p} \qquad (2)$$

where r_p is the radius of the particle and h_p is its height. This value is a direct measurement of the phase state of individual particles as particles with higher viscosities will spread out less and thus appear taller than less viscous particles of comparable size (see Figure 8 from Olson et al., 2019). Hygroscopic growth factors (HGF) will be calculated from Eq. 3,[61]

$$\mathrm{HGF(RH)} = \frac{\mathrm{D}_{p,RH}}{\mathrm{D}_{p,0}} \qquad (3)$$

where RH is the chosen relative humidity (up to ~90%) in the environmental cell, $D_{p,0}$ is the volume equivalent diameter of the collected respiratory aerosol measured under dry conditions (RH~0%), and $D_{p,RH}$ is the particle diameter at the chosen RH, as measured through AFM analysis. The HGFs will be measured as a function of increasing organic carbon fraction in the lab-generated samples and contrasted with the human samples. I will test how the HGFs change as a function of particle size by analyzing substrates from different collection stages of the impactor. The composition determined from integrated IR absorption spectra of the lab-generated samples will be compared with the aerosols generated by the subjects. Specifically, I will investigate the characteristic vibrational modes that are associated with mucins (amide bands and carbohydrate bands), immunoglobins (aromatic amino acids, S–H and S–S bands), surfactants (PO_2^- and trimethylamine bands), and salts (carbonate and ammonium bands). The model aerosols will be made with different mole fractions of the above classes of molecules and their IR spectra will be compared to that of the collected human samples to determine the presence and relative amounts or absence of the different classes. Each compound's intensity will be plotted against sound type, spreading ratio, and HGF. This analysis can help confirm the hypothesis that a relationship exists between the site of respiratory origin and particle physicochemical properties.

Proposed Timeline

Winter 2024 – Complete optimizing the setup for ELPI measurements of respiratory aerosols.

Spring 2024 – Ensure the ability to reproducibly collect respiratory aerosol, airflow, and acoustic data from real subjects utilizing the integrated setup.

Summer and Fall 2024 – Collect data across several demographics to compare the results to the listed hypotheses.

Winter 2025 – Prepare the model respiratory fluids and complete hygroscopic growth calculations and phase measurements with the AFM.

Spring and Summer 2025 – Carry out AFM measurements on the aerosols collected from participants and compare them to model aerosol results to evaluate the listed hypotheses.

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