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A Study of Characteristics, Origins, and Fates of Ultrafine Particles Over Taichung City, Taiwan

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Abstract

Ultrafine particles (UFPs, <0.1 μm) pose serious health risks and remain challenging to accurately monitor and model due to complex atmospheric processes. This study investigates UFP dynamics over Taichung City, Taiwan, utilizing April 2021 observational data from a Scanning Mobility Particle Sizer (SMPS) and simulations from the GEOS-Chem-TOMAS model. Observations included particle number size distributions (analyzed from 11.8 to 593.5 nm), sulfuric acid concentrations, and meteorological variables. The model employed 15 size bins and evaluated five nucleation schemes (Basecase, Binary, Binary ion, Ternary, Ternary ion) to assess new particle formation (NPF) sensitivity. Results indicate that Basecase, Ternary, and Ternary ion schemes simulated sporadic nucleation bursts; Binary schemes showed minimal activity. Despite visual coincidence with sulfuric acid peaks, simulated nucleation rates exhibited a weak linear correlation (Pearson $R \approx -0.04$), underscoring that NPF is a complex, non-linear process influenced by multiple atmospheric parameters beyond sulfuric acid, including ammonia, amines, and organic vapors, The model consistently and substantially underestimated observed particle number concentrations across all modes (NMBs ranging from -8.5% to -90.9%; correlation coefficients (absolute values) from 0.06 to 0.21) except for accumulation mode that the model display relatively good agreement of magnitude. The current model struggled to capture the overall magnitude and variability of particle number size distribution, underscoring the need for improving our understanding of the emission inventory as well as the boundary layer meteorology. With regard to new particle formation, crucially no clear classical "banana-shaped" NPF events were observed Importantly, no clear classical "banana-shaped" NPF events were observed in either the observational data or the model simulations throughout the study period. These findings underscore the critical need for improved nucleation and growth parameterizations, higher-resolution meteorological data, and refined local emission inventories to enhance the accuracy of urban aerosol modeling in subtropical environments.

Keywords : Ultrafine Particles; GEOS-Chem-TOMAS; Nucleation; Air Quality; Taichung; Atmospheric Modeling

Introduction

Ultrafine particles (UFPs), defined as particulate matter with diameters less than 0.1 μm, are of growing concern due to their adverse health effects [1, 2]. These particles can penetrate deep into the lungs, translocate into the bloodstream, and induce oxidative stress, potentially leading to cardiovascular, respiratory, and neurological diseases [3, 4]. Mounting evidence from toxicological and epidemiological studies supported by organizations like the World Health Organization (WHO) and the Health Effects Institute (HEI) suggests UFPs can trigger systemic inflammation and other adverse outcomes even at relatively low ambient concentrations. This is particularly concerning given the lack of official regulatory standards for UFP number concentrations in most countries, which contrasts with the mass-based metrics used for $PM_{2.5}$ and PM_{10}. Recent studies emphasize particle number concentration (PNC) as a more suitable metric for evaluating UFP exposure and informing public health policy [5].

New particle formation (NPF) is a significant source of UFPs in the atmosphere. This process involves the conversion of precursor gases into new particles, with key roles played by species such as sulfuric acid, ammonia, and organic compounds. These chemical reactions are highly sensitive to meteorological conditions, including solar radiation, temperature, and relative humidity, which influence the formation of condensable vapors and the removal of nascent particles [6, 7].

As with any air quality monitoring, spatial coverage is limited but air quality model can help supplement these gaps. Advanced chemical transport models, such as GEOS-Chem coupled with the TwO-Moment Aerosol Sectional (TOMAS) microphysics module, offer a robust framework for simulating aerosol number and mass size distributions [8]. Comparing model output against high-resolution observational data is essential for enhancing predictive accuracy and improving our understanding of UFP behavior.

This study investigates the characteristics, origins, and atmospheric evolution of UFPs over Taichung City, Taiwan, by integrating ground-based measurements with GEOS-Chem-TOMAS simulations. We aim to identify key processes driving new particle formation (NPF), evaluate various nucleation schemes, and assess the model's ability to replicate observed UFP dynamics. Findings are expected to enhance UFP representation in regional air quality models and provide insights for future regulatory and public health strategies.

Methodology

This study investigates ultrafine particle (UFP) behavior in Taichung, Taiwan, using both observational data and regional-scale modeling for the period April 2021. This specific month was selected due to the high completeness and quality of the observational data, particularly from the Scanning Mobility Particle Sizer (SMPS) measurements, which exhibited the fewest missing (NaN) values during this period, thus ensuring a robust dataset for detailed analysis of new particle formation events and reliable model-measurement comparison. Particle number size distribution (PNSD) measured by the Scanning Mobility Particle Sizer (SMPS), covering a size range of 11.8–593.5 nm, at the Tunghai University monitoring site in collaboration with National Taiwan University (NTU). Sulfuric acid and other meteorological variables, including solar radiation, relative humidity, and $PM_{2.5}$ concentrations, were monitored to support the analysis of atmospheric conditions influencing UFP formation. It should be noted that the SMPS instrument's lower detection limit of 11.8 nm inherently limits the observation of nascent NPF stages (particles typically form at sub-10 nm sizes). SMPS observations were averaged to hourly resolution before comparison with hourly model outputs.

For the modeling component, the GEOS-Chem chemical transport model [9] coupled with the TwO-Moment Aerosol Sectional (TOMAS) microphysics module was employed, following the configurations described by Trivitayanurak et al. (2008) [8]. The simulations were conducted using GEOS-FP assimilated meteorological data from NASA's Global Modeling and Assimilation Office, at a nested horizontal resolution of 0.25° × 0.3125°, encompassing the Taiwan region. The GEOS-Chem-TOMAS framework calculates aerosol number and mass concentrations across 15 discrete size bins ranging from 3 nm to 10 μm. To investigate the sensitivity of UFP formation to different nucleation pathways, five nucleation

schemes were evaluated: Basecase, Binary, Binary ion, Ternary, and Ternary ion.

The model's emissions inventory relies on a combination of global and regional datasets. For Asia, the MIX inventory is commonly used, which provides anthropogenic emissions on a regional scale. While this inventory offers a good overall picture, its spatial and temporal resolution may not fully capture local, highly variable sources like on-road traffic or small-scale industrial activities in a densely populated urban area like Taichung. This can lead to discrepancies between modeled and observed concentrations, especially for fresh, small particles.

The five nucleation schemes represent different theoretical pathways for new particle formation:

- Basecase: A standard kinetic nucleation scheme that represents the collision of precursor molecules to form a stable cluster.
- Binary Homogeneous Nucleation (BHN): Describes the formation of new particles solely from the condensation of sulfuric acid and water vapor. This is often less effective in the lower troposphere due to the high vapor pressure of water.
- Binary Ion-Mediated Nucleation (BIMN): Similar to BHN, but the process is catalyzed by atmospheric ions, which can significantly enhance nucleation rates by stabilizing the particle clusters.
- Ternary Homogeneous Nucleation (THN): An extension of BHN that includes a third component, typically ammonia, which acts as a stabilizing agent to lower the energy barrier for nucleation, making the process more efficient.
- Ternary Ion-Mediated Nucleation (TIMN): The most complex scheme, incorporating the effects of both ammonia and atmospheric ions to promote nucleation.

New particle formation (NPF) events were identified based on observed particle growth in the 11–20 nm range, with growth rates determined from changes in geometric mean diameter (Dpg), in line with Dal Maso et al. (2005) [10]. The classification of NPF events was conducted through visual inspection of PNSD surface plots and analysis of nucleation-mode particle evolution, consistent with methodologies outlined by Wang et al. (2023) [7].

Model diagnostics, including nucleation, condensation, and coagulation rates, were used to interpret observed events and assess the capability of each nucleation scheme in reproducing measured UFP trends. These insights are critical for understanding regional-scale UFP dynamics and improving model performance in simulating fine-scale aerosol processes.

Results and Discussions

The GEOS-Chem model simulation for April 2021, shown in Figure 1, illustrated the spatial distribution of total particle number concentrations (PNC) over Taiwan, revealing elevated levels in northeastern regions consistent with monsoonal winds and inland advection.

To understand observed particle dynamics and atmospheric conditions, Figure 2 presents the contour plots of hourly particle number size distribution (PNSD) for the study site at Taichung City, selected for the period of 20-25 April 2021 for both the observation and the simulated results. The PNSD is plotted in units of dN/dlogDp (#/cm³). My analysis found similar features of the contour plots for the rest of April 2021 thus they are not shown here. Due to the measurement limitation where the SMPS begins detection at 11.8 nm, the comparison of particles smaller than 11 nm is not possible. Despite this limitation, the observation data revealed there was no evidence of a classical "banana-shaped" new particle formation (NPF) events throughout the study period, as depicted in Figure 2 a) as an example.

A classical "banana-shaped" NPF event is a distinct pattern on a PNSD contour plot, characterized by a sharp, vertical increase in the number of particles in the nucleation mode, typically between 3–25 nm, followed by a clear, sustained diagonal upward shift as these newly formed particles grow into the Aitken mode (25–100 nm) over several hours. The plot resembles a "banana" as the particle size increases with time.

For the simulation, there is no NPF occurred during the study period as well, as shown in Figure 2b) as an example. The plot

depicts particle growth behavior but none with the feature of a major NPF event that is typically followed by growth that lasts over several hours.

Upon comparison of the PNSD in Figure 2, the measured data was consistently higher and also with more variability. The lower temporal variability in modeled PNSD is attributed to the nature of the model grid representing averaged concentrations over a grid area, which may not fully resolve the fluctuations occurring at a spot site. The modeled PNSD allows an investigation down to 3 nm and demonstrates a peak of the nucleation mode particles occurring every day at regular hours typical of the morning traffic rush hours. The observed PNSD, on the other hand, shows a higher variation with multiple peaks at morning traffic rush hours, midday, and another in the evening. The absence of midday and evening peaks in the model highlights gaps in emission inventories that may not readily reflect real-world fluctuations and limitations in the representation of evening boundary layer meteorology. Another important feature of the simulated PNSD is the persistent concentrations above 10 nm with a major peak at 20 nm and a fewer occurrence of another peak at 100 nm.

Analysis of meteorological parameters revealed that persistently high relative humidity (often exceeding 70-80% during daytime), solar radiation in the range of 500-800 W/m², and low SO_2 concentrations (typically below 1 ppb) likely inhibited NPF. Furthermore, the environment was characterized by elevated background aerosol concentrations and generally low levels of critical precursor gases. These elevated background particles contribute to a substantial condensation sink, efficiently scavenging low-volatility vapors and newly formed clusters, thereby suppressing nucleation. Combined with non-optimal meteorological conditions, these factors likely explain the observed absence of classical NPF events in Taichung.

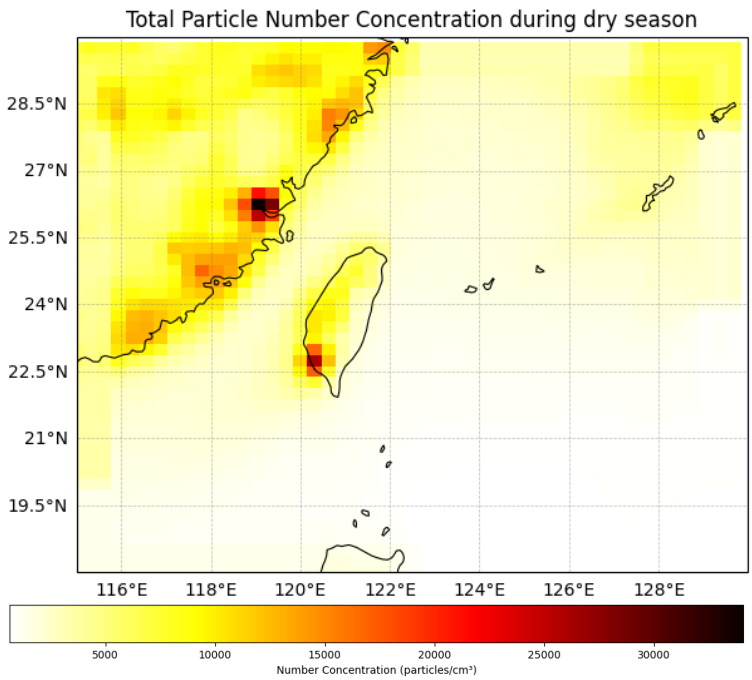

Figure 1 Spatial distribution of total particle number concentrations (PNC) over Taiwan, simulated by the GEOS-Chem model averaged for April 2021 period

a)

b)

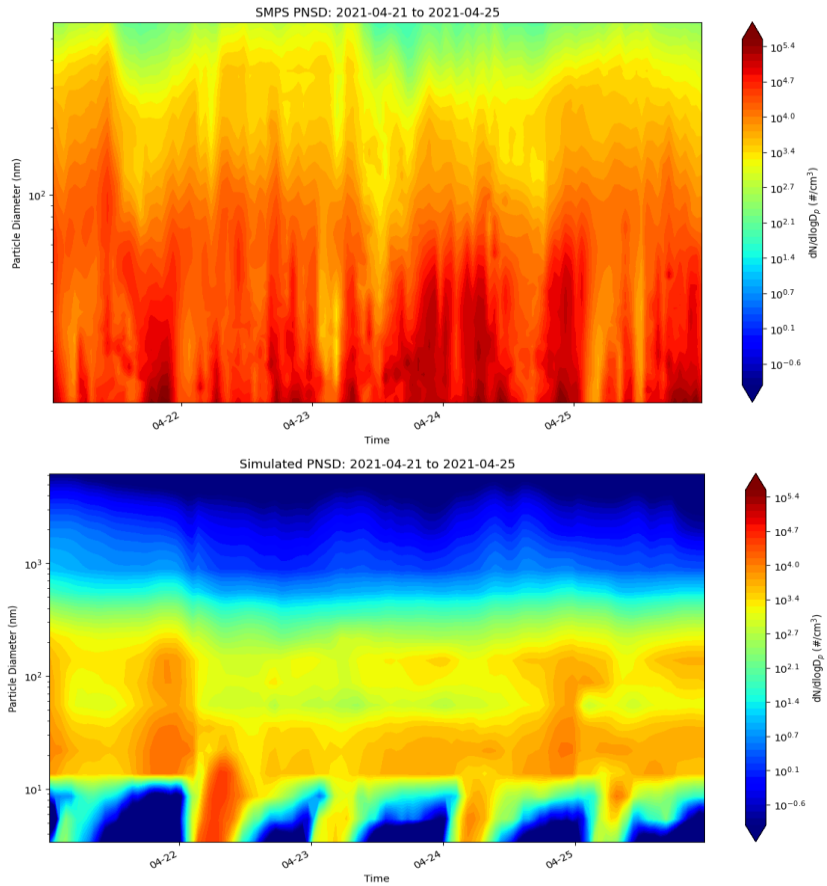

Figure 2 PNSD Contour Plot at Tunghai University, Taichung City, during 21-25 April 2021, including a) measurement by SMPS and b) simulated by GEOS-Chem-TOMAS model. The measured PNSD covers sizes of 11.8-593.5 nm while the simulated PNSD covers 3 nm to 10 microns

To better understand the particle dynamics, it is important to distinguish between the three primary particle modes:

- Nucleation Mode (<25 nm): Dominated by particles freshly formed from gas-to-particle conversion (new particle formation). These are highly sensitive to precursor gas concentrations and photochemical activity.
- Aitken Mode (25-100 nm): Formed primarily by the growth of nucleation mode particles through coagulation and condensation of semi-volatile vapors.
- Accumulation Mode (100-1,000 nm): Formed by the coagulation and condensation of smaller particles. These particles have a longer atmospheric lifetime and are less sensitive to rapid, local processes.

To quantitatively assess the model's performance, Figure 3 and Figure 4 compare simulated and observed particle number concentrations (PNCs) as time-series plot and scatter plots, respectively, for total PNC (≤50 nm) and individual modes (nucleation, Aitken, and accumulation). The model consistently and substantially underestimated observed particle concentrations across all modes except the accumulation mode. The accumulation mode provides better agreement because these larger particles are less dependent on rapid, local processes like primary emissions or nucleation, which are poorly resolved by the model. Their concentrations are more influenced by regional

transport and long-term accumulation, which the model is better equipped to simulate. The time-series plot in Figure 3 d) and the scatter plot in Figure 4 c) display significantly better agreement between modeled and observed PNC compared to other size modes. For total PNC (≤50 nm), the model showed a Normalized Mean Bias (NMB) of -90.88% and a Root Mean Square Error (RMSE) of 37,006.79 #/cm³. Mode-specific analysis revealed NMBs ranging from -8.49% for accumulation mode to -89.22% for Aitken mode, and -86.31% for nucleation mode. Correlation coefficients (R) were notably low across most modes, with R^2 values ranging from 0.0036 (Total PNC) to 0.04 (Nucleation Mode) and 0.03 (Aitken Mode). However, the accumulation mode exhibited a relatively strong correlation (R^2 = 0.78). This indicates that the model largely fails to reproduce the observed temporal variability and synchronous changes in particle concentrations, highlighting a significant challenge in capturing UFP dynamics at local scales.

The time-averaged PNSD for both simulated and observed data showed that while the model qualitatively captures the general shape, it systematically underestimates mean concentrations and exhibits higher variability than observed, particularly at the smallest (<13 nm) and largest (>600 nm) particle sizes. The notably wide standard deviation in the sub-13 nm range suggests that the model simulates sporadic and intense bursts of new particle formation or very rapid initial growth/loss, which, even if short-lived, lead to significant variability around the mean.

Diurnal variations were also examined in Figure 5; the model fails to capture the variations exhibited by observed data for all size modes. Only in the accumulation mode did the modeled data display comparable values but the morning peak at 7:00 and 10:00 were also missing underscoring the challenges in emission representation and possibly in the photochemical reactions simulated in the model relative to the real ambient environment [11].

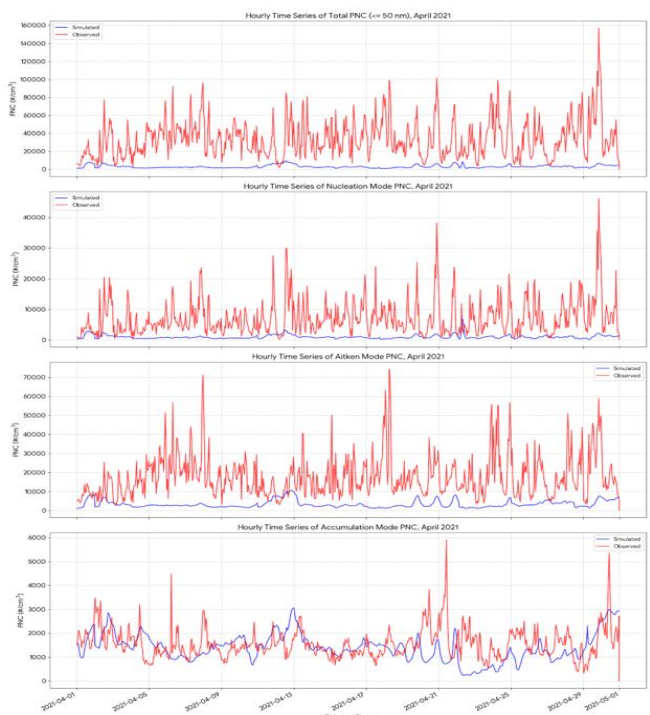

Figure 3 Hourly Time Series of Simulated (Blue) and Observed (Red) PNC for April 2021 Presented as a) Total PNC, b) Nucleation Mode, c) Aitken Mode, and d) Accumulation Mode PNC

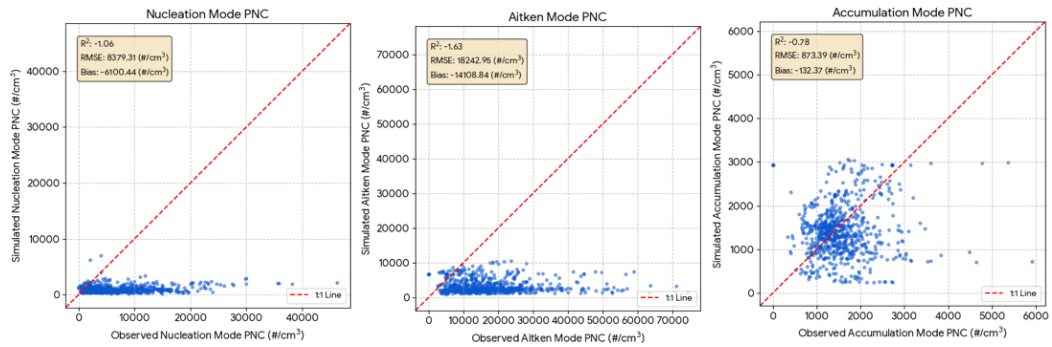

Figure 4 Scatter Plots Comparing Simulated and Observed Particle Number Concentrations in Taichung City over the April 2021 period for a) nucleation mode, b) Aitken mode, and c) accumulation mode PNC.

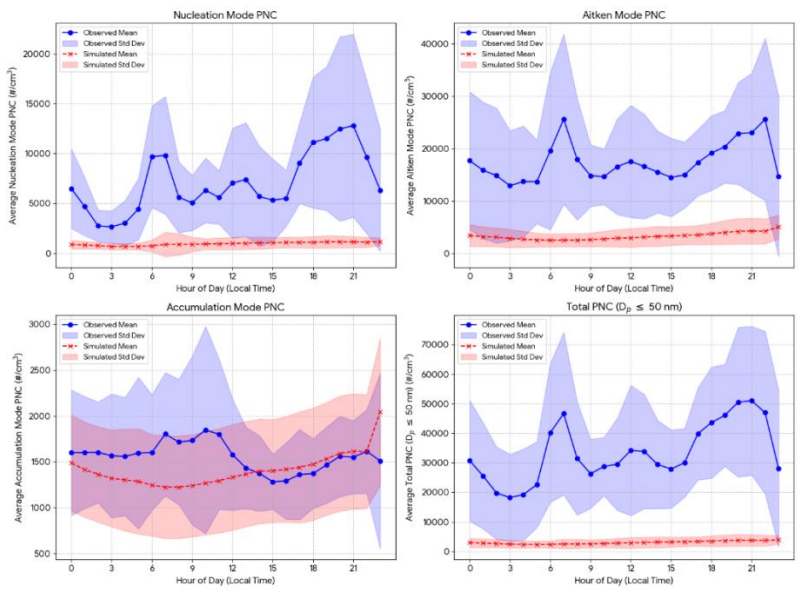

Figure 5 Diurnal Variation of Simulated vs. Observed Mode-Specific Particle Number Concentrations in Taichung City, April 2021

To investigate the sensitivity of particle formation processes, five nucleation schemes were evaluated: Basecase, Binary, Binary ion, Ternary, and Ternary ion. The Basecase, Ternary, and Ternary ion schemes demonstrated comparable behavior, effectively simulating sporadic temporal nucleation burst events, with peak values reaching approximately 1.17 #/cm³/s. However, as confirmed by the simulated PNSD contour plots (Figure 2), these simulated bursts did not consistently lead to sustained growth characteristic of classical NPF events. In contrast, the Binary and Binary ion schemes exhibited negligible activity, suggesting that binary nucleation mechanisms were not thermodynamically favored under the ambient conditions in Taichung. Elevated H_2SO_4 concentrations visually corresponded with the timing of nucleation bursts in the active schemes, highlighting its role as a key precursor. However, quantitative analysis revealed a weak statistical relationship (Pearson R ≈ -0.04), indicating that nucleation is a non-linear, multi-factorial process influenced by other variables beyond just sulfuric acid, including ammonia, amines, and organic vapors.

The consistent underestimation of UFP concentrations and the model's limited ability to capture sustained, classical NPF events highlight several contributing factors. These include Model Resolution Constraints, here the 0.25° × 0.3125° resolution may be insufficient to fully resolve sharp concentration gradients and localized emissions, leading to underestimation and weakened temporal correlations. Uncertainties in Emission Inventories are also a factor, as discrepancies could arise from underestimations in primary particle emissions or precursor gas emissions (e.g., SO_2, VOCs, NH_3); emission inventories often rely on averaged data and may not fully capture temporal or spatial heterogeneity.

Furthermore, limitations in the representation of nucleation and growth mechanisms in the model's schemes to accurately represent complex chemical pathways and environmental conditions can lead to discrepancies in timing and magnitude of NPF events, with the underestimation of particle growth suggesting inadequate capture of condensable vapors or the efficiency of condensational growth and coagulation processes. Meteorological input accuracy also plays a role, as inherent uncertainties or resolution limitations in assimilated meteorological fields can propagate into the chemical transport simulation, leading to misrepresentation of dilution, transport, and removal processes. Finally, measurement uncertainty, where the SMPS instrument begins detection at approximately 11 nm while the model resolves particles down to 3.41 nm, can contribute to the observed underestimation of nucleation-mode particles, as a significant fraction of newly formed particles can exist in the sub-10 nm range.

Conclusions

This study investigated the spatial and temporal characteristics of ultrafine particle (UFP) number concentrations and size distributions over Taichung City, Taiwan, utilizing GEOS-Chem-TOMAS simulations and ground-based SMPS observations. The model successfully captured broad spatial patterns of particle number concentration (PNC), including an inland shift of concentration hotspots influenced by the northeastern monsoon and regional advection.

In terms of temporal behavior, the model reproduced some general trends in total UFP number concentration (≤50 nm). However, it substantially underestimated the overall magnitude compared to observations. The model also exhibited lower temporal variability than the observed SMPS data with sharp and frequent fluctuations, highlighting challenges in capturing UFP dynamics at local scales.

Correlation analysis further underscored these limitations. An R value of 0.06 (R^2 value of 0.0036) and a Normalized Mean Bias (NMB) of -90.88% indicated poor agreement between simulated and observed concentrations. These discrepancies likely stem from uncertainties in nucleation parameterizations, underrepresentation of localized emissions, and limited resolution of meteorological processes.

Contour plots of particle size distributions (PNSDs) showed no distinct clear, classical "banana-shaped" new particle formation (NPF) events in either the simulation or the observations for April 2021. This consistent absence could be attributed to unfavorable atmospheric conditions during the study period or limitations in both model sensitivity and observational resolution. Further analysis by segregating particle size modes reveals that while accumulation mode PNC showed relatively good agreement for magnitudes, other size modes demonstrate underprediction. The model currently cannot capture the peak concentrations appearing at different times of day as shown in the observations.

While the World Health Organization (WHO) provides air quality guidelines for particle mass concentrations (e.g., $PM_{2.5}$), no such standards exist for UFP number concentrations. Given the potential health impacts associated with UFP exposure, model underestimations pose a risk of misinforming exposure assessments and policy decisions if not properly accounted for. Overall, these findings highlight the potential of GEOS-Chem-TOMAS for regional air quality analysis but also reveal key areas needing improvement. Future work should prioritize refining nucleation schemes (such as incorporating organic aerosol effects),

enhancing the resolution of local emissions and meteorology, and expanding validation datasets, particularly for size-resolved comparisons. Addressing the model's persistent underestimation and weak agreement with observations will be critical to improving its utility for both scientific analysis and public health applications.

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