TJWAPC 2024

Taiwan - Japan Workshop on Atmospheric Physics and Chemistry

ABSTRACT BOOK

📅 Date

May 15 ~ 18

中央研究院
ACADEMIA SINICA, TAIWAN

中央研究院　原子與分子科學研究所
Institute of Atomic and Molecular Sciences
Academia Sinica

TJWAPC 2024

Date May 15 ~ 18

Taiwan - Japan Workshop on Atmospheric Physics and Chemistry

2024 / 5 / 15 Wednesday	Time Start	2024 / 5 / 16 Thursday	Time Start	2024 / 5 / 17 Friday	2024 / 5 / 18 Saturday
Arrival Howard Civil Service International House Check in time after 15:00	08:20~	Registration	08:45~	Registration	Departure Howard Civil Service International House Check out time before 11:00
		Session A Chair : Dr. Jim Jr-Min Lin			
	08:50	Opening : Dr. Ching-Ming Wei		Session F Chair : Dr. Pei - Ling Luo	
	08:55	Opening Remarks: Dr. Yuan-Tseh Lee	09:00	Dr. Nanase Kohno	
	09:10	Opening talk : Dr. Hajime Akimoto			
	09:40	Dr. Tomoaki Okuda	09:40	Dr. Li-Hao Young	
	10:20	Coffee Break & Group Photo (20 mins)	10:20	Coffee Break (20 mins)	
		Session B Chair : Dr. Kei Sato		Session G Chair : Dr. Yuan - Pin Chang	
	10:40	Dr. Yuji Fujitani	10:40	Dr. Charles C.-K. Chou	
	11:20	Dr. Jim Jr-Min Lin	11:20	Dr. Satoshi Inomata	
	12:00	Mr. Mahiro Yamamoto	12:00	Mr. Daisuke Fukuyama	
	12:30	Lunch Break	12:30	Lunch Break	
		Session C Chair : Dr. Kaito Takahashi		Session H Chair : Dr. Yosuke Sakamoto	
	13:30	Dr. Pei-Ling Luo	13:30	Dr. Kei Sato	
	14:10	Dr. Yosuke Sakamoto	14:10	Dr. Yu Morino	
	14:50	Coffee Break (10 mins)	14:50	Coffee Break (10 mins)	
		Session D Chair : Dr. Satoshi Inomata		Session I Chair : Dr. Stephen Griffith	
	15:00	Dr. Mao-Chang Liang	15:00	Dr. Yoshiteru Iinuma	
	15:40	Dr. Shinichi Enami	15:40	Dr. Hui-Ming Hung	
	16:20	Coffee Break (10 mins)	16:20	Coffee Break (10 mins)	
		Session E Chair : Dr. Ta - Chih Hsiao		Session J Chair : Dr. Kaito Takahashi	
	16:30	Dr. Chia-Chen Wang	16:30	Dr. Satoko Kayaba	
	17:10	Dr. Masao Gen	17:00	Dr. Yu-Chieh Ting	
	17:50	Move to restaurant	17:40	Dr. Ta-Chih Hsiao	
Dinner (by invitation)	18:30~	Dinner (by invitation)	18:30~	Dinner (by invitation)	

TJWAPC 2024 Date May 15~18

Taiwan - Japan Workshop on Atmospheric Physics and Chemistry

May 16 (Thursday)

08:20~	Registration
	Session A Chair : Dr. Jim Jr-Min Lin (Academia Sinica)
08:50~08:55	Opening : Dr. Ching-Ming Wei (Academia Sinica)
08:55~09:10	Opening Remarks : Dr. Yuan-Tseh Lee (Academia Sinica)
09:10~09:40	Dr. Hajime Akimoto (National Institute for Environmental Studies) *Foci of Recent Atmospheric Chemistry and Physics Research*
09:40~10:20	Dr. Tomoaki Okuda (Keio University) *Chemical, Physical, and Biological Characteristics of Aerosol Particles related to Human Health: Particle Charging State and Cell Exposure Study*
10:20~10:40	Coffee Break & Group Photo (20 mins)
	Session B Chair : Dr. Kei Sato (National Institute for Environmental Studies)
10:40~11:20	Dr. Yuji Fujitani (National Institute for Environmental Studies) *Assessing the cellular oxidative stress induction by exposure to photochemical aging diesel exhaust particles using the air-liquid interface cell exposure method*
11:20~12:00	Dr. Jim Jr-Min Lin (Academia Sinica) *UV Spectroscopy and Reaction Kinetics of Criegee Intermediates*
12:00~12:30	Mr. Mahiro Yamamoto (Hokkaido University) *Kinetics of C9 Criegee intermediates formed from β-pinene*
12:30~13:30	Lunch Break
	Session C Chair : Dr. Kaito Takahashi (Academia Sinica)
13:30~14:10	Dr. Pei-Ling Luo (Academia Sinica) *Applications of high-resolution time-resolved dual-comb spectroscopy: from fundamental chemical physics to atmospheric chemistry*
14:10~14:50	Dr. Yosuke Sakamoto (National Institute for Environmental Studies) *Study Toward a Quantitative Description of Peroxy Radical Uptake by Aerosols in the Atmospheric HOx Chemistry*
14:50~15:00	Coffee Break (10 mins)
	Session D Chair : Dr. Satoshi Inomata (National Institute for Environmental Studies)
15:00~15:40	Dr. Mao-Chang Liang (Academia Sinica) *Heterogeneous reactions of ozone and airborne particulates*
15:40~16:20	Dr. Shinichi Enami (University of Tsukuba) *Atmospheric reactions in inhomogeneous solutions*
16:20~16:30	Coffee Break (10 mins)
	Session E Chair : Dr. Ta - Chih Hsiao (National Taiwan University)
16:30~17:10	Dr. Chia-Chen Wang (National Sun Yat-sen University) *Probing the Electronic Structures, Interfacial Properties and Chemical Activities of Aerosols via Aerosol VUV Photoelectron Spectroscopy*
17:10~17:50	Dr. Masao Gen (Tokyo Institute of Technology) *Single particle studies of deliquesced aerosols: multiphase reactions and surface tension*
17:50	Move to restaurant
18:30~	Dinner (by invitation)

May 17 (Friday)

08:45~	Registration
	Session F Chair : Dr. Pei-Ling Luo (Academia Sinica)
09:00~09:40	Dr. Nanase Kohno (Kindai University) *Observation of OH radical precursors in atmospheric aerosols by filter sampling*
09:40~10:20	Dr. Li-Hao Young (China Medical University) *Exploring atmospheric radicals and oxidants using a photochemical box model coupled with Master Chemical Mechanism*
10:20~10:40	Coffee Break (20 mins)
	Session G Chair : Dr. Yuan-Pin Chang (National Sun Yat-Sen University)
10:40~11:20	Dr. Charles C.-K. Chou (Academia Sinica) *An observation-based diagnosis for the air pollution in the Central-Western Taiwan*
11:20~12:00	Dr. Satoshi Inomata (National Institute for Environmental Studies) *Enhancement of the secondary organic aerosol yields and the formation of low- and extremely low-volatility organic compounds during the oxidations of a-pinene in the presence of acidic seeds*
12:00~12:30	Mr. Daisuke Fukuyama (Yokohama City University) *Functional group analysis of gas-phase oxidation products of monoterpenes using high-resolution collision- induced dissociation mass spectrometry*
12:30~13:30	Lunch Break
	Session H Chair : Dr. Yosuke Sakamoto (National Institute for Environmental Studies)
13:30~14:10	Dr. Kei Sato (National Institute for Environmental Studies) *A study of formation process of a multifunctional ring-opening product present in secondary aerosol from aromatic compounds*
14:10~14:50	Dr. Yu Morino (National Institute for Environmental Studies) *Atmospheric Simulation of Organic Aerosol Sources and Ozone Production Regime*
14:50~15:00	Coffee Break (10 mins)
	Session I Chair : Dr. Stephen Griffith (National Taiwan University)
15:00~15:40	Dr. Yoshiteru Iinuma (Okinawa Insitute of Science and Technology Graduate University) *Do typhoons have an impact on ground-level ozone and secondary organic aerosol formation?*
15:40~16:20	Dr. Hui-Ming Hung (National Taiwan University) *Impact of SO_2 on a-pinene Ozonolysis-derived Secondary Organic Aerosol Formation and its Implications*
16:20~16:30	Coffee Break (10 mins)
	Session J Chair : Dr. Kaito Takahashi (Academia Sinica)
16:30~17:00	Dr. Satoko Kayaba (National Institute for Environmental Studies) *Potential Impacts of Energy and Vehicle Transition Through 2050 on Oxidative Stress-Inducing $PM_{2.5}$ -Metal Concentrations in Japan*
17:00~17:40	Dr. Yu-Chieh Ting (National Taiwan University) *Seasonal and diurnal variations in the light-absorbing properties and molecular composition of soluble brown carbon in central Taiwan*
17:40~18:20	Dr. Ta-Chih Hsiao (National Taiwan University) *Elucidating the Dynamics of New Particle Formation in Central Taiwan : Observational Insights and Influencing Factors*
18:30~	Dinner (by invitation)

Foci of Recent Atmospheric Chemistry and Physics Research

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ABSTRACT

Climate change due to anthropogenic global warming is now a major focus of atmospheric research and social concern. As for climate change research, how chemistry can contribute to reduce large uncertainty in future climate is my immediate concern. I would like to propose the focus of this issue as follows.

1. How to quantify the effect of organic and inorganic aerosols as cloud condensation nuclei (CCN) based on theoretical molecular science? Similarly, how to quantify the ice nucleation effect of particles theoretically? Although these effects have been quantified by means of laboratory experiments, those should be quantified based on the theoretical molecular science in order to establish the phenomena as basic science. These are fundamental research on indirect effect of radiative forcing of atmospheric particulate matters.

In addition to climate change issue, tropospheric ozone pollution is still another major concern due to its impacts on human health/agriculture, and also on climate as a short-lived climate forcer. Although tropospheric ozone pollution research has been matured, there are still fundamental uncertainties.

2. The efficiency of RO_2/HO_2 cycle to reproduce OH as a function of NO_x concentration has not been established well, which brought inconsistency between the model calculation and observation of urban and regional ozone$^{1)}$. Heterogeneous reactions of HNO_3 to regenerate NO_x, NO_x + surface $H_2O \rightarrow HONO$, and homogeneous reactions of $RO_2 + NO_x \rightarrow$ products, needs to be established by means of laboratory experiments and field observation$^{2)}$.

References

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Chemical, Physical, and Biological Characteristics of Aerosol Particles related to Human Health: Particle Charging State and Cell Exposure Study

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ABSTRACT

Atmospheric aerosols are of serious concern for human health. However, the key factors of particulate matter responsible for the adverse health effects are not yet fully understood. Our lab tries to elucidate these factors of atmospheric aerosols, such as chemical, physical, and biological characteristics, by developing original measurement methods based on atmospheric chemistry and aerosol engineering (Fig. 1). Here I would like to present our recent achievements regarding aerosol particle charging state[1-5] and cyclonic collection of particles for cell exposure experiments[6-13].

Fig. 1 Research concept of our laboratory.

References

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Assessing the cellular oxidative stress induction by exposure to photochemical aging diesel exhaust particles using the air-liquid interface cell exposure method

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ABSTRACT

Atmospheric particulate matter (PM) is aged by reactions with oxidants during transport in the atmosphere. It has been suggested that the toxicity changes with aging, but little is known about these changes quantitatively. PM exposure method for toxicity evaluation using cells, the air-liquid interface (ALI) exposure method has been widely utilized in recent years. This enables toxicity evaluation by directly exposing cultured cells to PM and gaseous substances with properties similar to reality, but it has the problem that the PM deposition area on the deposition surface become ununiform, and the sensitivity of the entire exposure surface is reduced. The purpose of this study is to evaluate the toxicological changes caused by atmospheric aging of diesel exhaust particles through ALI cell exposure method. For the problem of ununiform on the exposure surface, an imaging method have established to visualize the oxidative stress response. For the imaging, a vector expressing NanoLuc® luciferase, which is highly sensitive to oxidative stress, was incorporated into rat-derived alveolar II epithelial cells (SV40-T2). After cell lysis, oxidative stress was evaluated by luminescence intensity and RNA was extracted to evaluate gene expression levels related to oxidative stress induction. Aging of the exhaust gas is performed by passing it through an OH radical reactor, and the conditions that the exhaust gas is subjected to in the real-world during summer and winter were defined as high aging and low aging conditions, respectively. After ALI exposure, although oxidative stress increased with low aging and decreased with high aging, and these aging particles showed greater oxidative stress than the freshly emitted particles. Results of the imaging method were compared to conventional methods of assessing by gene expression and luminescence results, and three methods were consistent. Imaging of oxidative stress intensity confirmed that the effect was relatively large in the central portion of the exposure chamber, consistent with the trend in the particle deposition distribution. This is expected to lead to more highly sensitive assay that is not averaged across the entire exposure deposition surface. This research shows that understanding aging particles is crucial for evaluating adverse health effect of PM.

UV Spectroscopy and Reaction Kinetics of Criegee Intermediates

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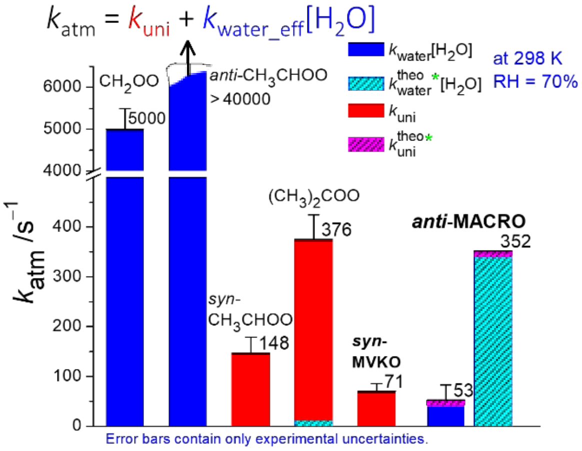
ABSTRACT

Criegee intermediates (CIs) are very reactive carbonyl oxides which are formed in the ozonolysis of alkenes. CIs are thought to be important in the oxidation of atmospheric molecules including SO_2, NO_2, inorganic and organic acids, and even water. To better assess the roles of CIs of various structures in atmospheric chemistry, it is desired to know the trend of their reactivity. Our group has measured the UV spectra of a few important CIs and determined their absolute absorption cross sections. The intense UV absorption of CIs allows scientists to probe their reaction kinetics. We found that there are strong structure dependences in the thermal decomposition reactions of CIs and in their reactions with water molecules. Furthermore, there is a strong synergy effect in the CI reactions with hydrogen-bonding molecules. Vinyl substituted CIs are more stable due to the extended conjugation of the double bonds, which also affect the reactivity. The fates of various CIs in the atmosphere can be estimated based on the trend of their reactivity. I will talk about the story of our research on CIs.



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Kinetics of C9 Criegee intermediates formed from β-pinene

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ABSTRACT

Stabilized Criegee intermediates (SCI) produced from ozonolysis of unsaturated organic compounds may contribute to secondary organic aerosol (SOA) formation through reactions with carboxylic acids to give low volatility products (1). β-Pinene (BP) is a monoterpene, which is a major biogenic volatile organic compound, and produces C_9SCI and C_1SCI through the ozonolysis. Because of the complex structure of C_9SCI, direct reaction rate measurements have not been performed, and the reaction rates have been estimated from structure-activity relationships (SARs) based on experiments and quantum chemical calculations for simpler SCIs (1,2). In this study, laboratory experiments were conducted to determine the ratio of the unimolecular rate coefficient (k_{uni}) of C_9SCI to the bimolecular rate coefficient (k_{FA}) of the reaction between C_9SCI and formic acid (HCOOH).

The experiments were conducted in a glass flow tube (FT) with an inner diameter of 4 cm and a length of 80 cm under atmospheric pressure conditions. Ozone, BP, and HCOOH were introduced into FT together with cyclohexane used as an OH scavenger. The reaction time in FT was about 45 s, and a portion of the gas from the FT outlet was introduced into a chemical ionization mass spectrometer to measure the signal intensity, I_{AAHP}, of ions attributed to α-acyloxyalkyl hydroperoxide (AAHP), which is produced in the reaction of C_9SCI with HCOOH.

Under dry conditions, I_{AAHP} was monitored as a function of HCOOH concentration, [HCOOH], and the results were fitted to the kinetic equation to obtain the relative rate coefficient, $k_{\mathrm{uni}}/k_{\mathrm{FA}}$. Because k_{uni} may be affected by reactions of C_9SCI with species produced from the BP ozonolysis, experiments were conducted at different initial concentrations of ozone, $[O_3]_0$. The $k_{\mathrm{uni}}/k_{\mathrm{FA}}$ was found to be independent of $[O_3]_0$, indicating that k_{uni} can be attributed to the unimolecular rate coefficient of C_9SCI. By weighted averaging of the $k_{\mathrm{uni}}/k_{\mathrm{FA}}$ values determined from the experiments at different $[O_3]_0$, it was determined to be $(3.7 \pm 0.5) \times 10^{11}$ molecules cm^{-3}. Using the values estimated by SARs from the previous studies (1,2), the $k_{\mathrm{uni}}/k_{\mathrm{FA}}$ value was calculated to be 1.2×10^{12} molecules cm^{-3}, which is three times larger than the value obtained in this study. Assuming an atmospheric [HCOOH] of 7.3 ppbv (3), approximately 23 % of C_9SCI is expected to react with HCOOH, which might play a significant role in SOA formation under dry conditions.

References

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Applications of high-resolution time-resolved dual-comb spectroscopy: from fundamental chemical physics to atmospheric chemistry

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ABSTRACT

Time-resolved infrared spectroscopic techniques are play significant roles in exploring transient processes of light-matter interactions as well as to study free radicals and reaction intermediates. Here, a new approach to time-resolved infrared laser spectroscopy based on dual-comb interferometry will be reported.[1,2] In our recent works, in addition to performing precision measurements of line strengths and pressure broadening coefficients of free radicals such as OH and HO_2 and the simplest Criegee intermediates,[3,4] we also investigated their reaction with key atmospheric pollutants.[5,6] By employing mid-infrared multifunctional dual-comb spectrometers, both reaction rate coefficients and product branching ratios can be accurately determined. Additionally, the atmospheric implications of the key reactions such as the reaction of Criegee intermediates with formaldehyde can also be evaluated by incorporating our results into a global 3-D chemistry-transport model.[6]

References

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Study Toward a Quantitative Description of Peroxy Radical Uptake by Aerosols in the Atmospheric HO_x Chemistry

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ABSTRACT

Tropospheric ozone, a major component of photochemical oxidants, is expected to be reduced as an air pollutant of concern for its effects on health and vegetation, and as a short-lived climate forcer affecting climate. Numerical simulations incorporating atmospheric chemistry models are used to predict changes in ozone concentrations due to precursor reductions. The accuracy of the assessment depends on the quantitative accuracy of the chemical model. However, it has been suggested that the uptake coefficients for heterogeneous uptake process of peroxy radicals (HO_2 and RO_2) onto aerosols and cloud droplet is not quantitatively understood, being potential sources of error in ozone prediction.

A laser-based radical reactivity measurement technique has been developed to measure the uptake coefficient of peroxy radicals onto aerosols with high temporal resolution. The technique has been applied to the measurement of uptake coefficients onto aerosols artificially produced in the laboratory[1] and onto real atmospheric aerosols in atmospheric observations[2,3]. We are also evaluating the impact on ozone prediction by incorporating the uptake process into atmospheric chemical model based on these observed uptake coefficients[2,3].

The results obtained from these studies showed that radical uptake by aerosols tends to contribute more to ozone production when the aerosol concentration is high and at the same time the radical and NO_x concentrations are not high, for example, in the afternoon when NO_x is consumed and in conditions of low solar radiation such as spring. The presentation will also include information on future attempts.

References

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Heterogeneous reactions of ozone and airborne particulates

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ABSTRACT

Ozone plays a fundamental role in the chemistry of the atmosphere, mediating oxidation reactions in phases and phase boundaries. Here, we investigate the least-explored solid-phase heterogeneous processes and report identification of a high reaction probability of O_3 with airborne aerosols. Using triple oxygen isotope ratios as tracer, we found that the reaction oxidizes organic particles efficiently and produces abundant carbon dioxide. We hypothesize that Criegee intermediates, formed by the reaction of unsaturated hydrocarbons and ozone and catalyzed by metal oxides, react with aldehyde/ketone-like organic compounds. Inclusion of the process in a chemistry-transport model yields a significant change in ozone budget and the O/C ratios of organic aerosols. The study shows the importance of ozone induced heterogeneous chemical reactions on aerosol surfaces occurring in polluted atmospheres.

Atmospheric reactions in inhomogeneous solutions

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ABSTRACT

Water is a major component of ocean and cloud droplets that influences global climate change. However, liquid water does not exist "homogeneously" on the earth. This inhomogeneity includes the air-water interface, where the density of water drops five orders of magnitude in ~1 nm, and the "domains" naturally formed with other components (e.g., organic solutes) in liquid phases (Fig. 1). Here, we demonstrate some examples of such molecular level inhomogeneity that potentially influences atmospheric physical/chemical processes.[1-4] For example, the decomposition rates of organic hydroperoxides formed by ozonolysis of a-pinene in water and acetonitrile mixture non-linearly increased as a function of molar fraction of water in the solution.[4] The implication is that a slight change of relative humidity unexpectedly (but dramatically) changes the lifetime of organic hydroperoxides in atmospheric condensed phases, that are inhomogeneous by nature.

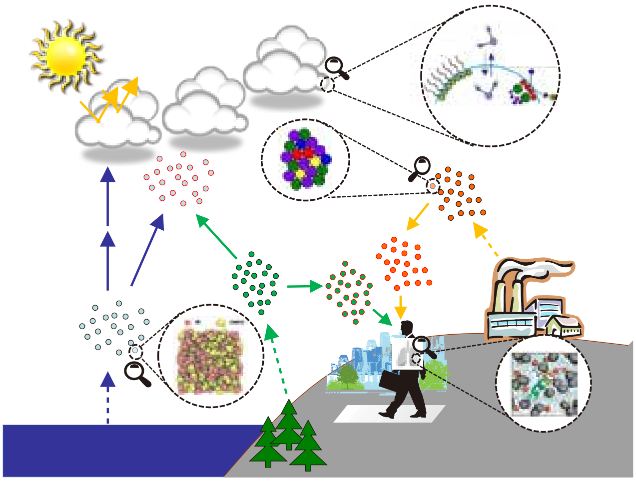

Fig.1 Schematic picture representing the importance of inhomogeneous physical/chemical processes associated with atmospheric condensed phases.

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Probing the Electronic Structures, Interfacial Properties and Chemical Activities of Aerosols via Aerosol VUV Photoelectron Spectroscopy

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ABSTRACT

Aerosols, ubiquitous in the environments, can be produced from a variety of sources, including natural and anthropogenic origins, which exhibit distinct chemical composition, formation mechanisms, evolution kinetic and dynamic properties, and physicochemical characteristics. In this talk, I will share our efforts in developing aerosol VUV photoelectron spectroscopy and utilizing it to gain insight into the pH-dependent electronic structures and interfacial properties of aqueous aerosols. Recently, we utilized the aerosol VUV photoelectron spectroscopy to study the valence electronic properties of limonene aerosols and secondary organic aerosols (SOAs) derived from limonene ozonolysis, with a specific focus on the effects of water in affecting the physico-chemical characteristics of Lim-SOAs [1]. Combining with DFT theoretical calculation, we show that water actively modifies the valence electronic structures of Lim-SOAs via intervening the CI chemistry by acting both as a catalyst and a reactant. The atmospheric implication is discussed. Additionally, I will also share our recent study to identify the chemical fingerprint of aerosols of various different origins, including secondary organic aerosols (SOAs) formed during ozonolysis of monoterpenes, and on-site, online field measurements of aerosols from harbour area, industrial districts in a harbour-industrial hybrid city, by applying mass spectrometry based approaches, including high-resolution liquid chromatography-electrospray ionization (LC-ESI) mass spectrometry and single particle mass spectrometry [2].

References

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Single particle studies of deliquesced aerosols: multiphase reactions and surface tension

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ABSTRACT

Atmospheric particulate matter or aerosol particles, directly and indirectly, impact climate, regional air quality, and human health. The impacts are closely associated with aerosol particles' chemical and physical evolution, which can be regulated by multiphase reactions and surface tension of deliquesced aerosol particles, respectively. However, in the extreme environment of aerosol particles, the behavior of the reaction kinetics and surface tension often deviates from our chemical intuition based on a beaker-scale understanding. My talk will introduce spectroscopic methods for single particle analysis to study reaction kinetics and surface tension of aerosol particles. The methods are based on inelastic (Raman) and quasi-elastic light scattering (QELS) techniques. Raman and QELS techniques will show dramatic differences in reaction kinetics[1] and surface tension[2] of deliquesced aerosol particles from beaker-scale (bulk) solutions. Our laboratory study would be helpful to improve our understanding of aerosol chemistry with broad implications for air quality and climate.

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Observation of OH radical precursors in atmospheric aerosols by filter sampling

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ABSTRACT

OH radical is very important chemical species because it is known to oxidize various compounds in atmospheric aerosols and potentially affect climate and air quality by changing aerosol size and properties. However, its behavior in aerosols is still unclear due to the difficulty of observation. Tong et al. have produced secondary organic aerosols by laboratory experiments and found that aerosols not only uptake radicals from the atmosphere and decomposition of hydrogen peroxide, but also generate OH radicals via the decomposition of organic peroxides in aerosols. In this study, we employed atmospheric aerosols collected by filters in Tokyo to observe the precursors of OH radicals in the aerosols and to investigate their relationship with the atmospheric trace species.

Atmospheric aerosols were collected three times every 48 hours using a high-volume air sampler (Shibata Kagaku, HV-1000F) in Tokyo in August 2023. Filters were stored in a freezer for about 3 months after collection and then extracted into a terephthalic acid solution. In this study, OH radicals formed in aqueous solution were detected by chemical reaction with terephthalic acid (TA), which reacts with OH radicals quickly to form hydroxyl terephthalic acid (HTA). Hydroxyl terephthalic acid absorbs UV light and emit fluorescence.

From the fluorescence spectrum observed in the TA extract solutions, the water-soluble OH radical precursors have been successfully detected throughout the entire observation period. Spectral intensities were slight varied depending on the observation period. To investigate the cause of the variation, we compared the concentrations of standard atmospheric trace species during the period from the database of the Ministry of the Environment. The results showed a positive correlation with the mass concentration of SPM in the air, indicating that the observed OH radicals originated from atmospheric aerosols. Additionally, fluorescence spectra of humic-like substances were also observed along with the OH radical precursors, and the origin of the OH radical precursors will be discussed by comparing with the fluorescence spectra.

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Exploring atmospheric radicals and oxidants using a photochemical box model coupled with Master Chemical Mechanism

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ABSTRACT

Atmospheric radicals and oxidants such as OH, O_3, and NO_3 are responsible for the oxidation and, hence, removal of primary air pollutants, including VOCs, CH_4, CO, SO_2, and NOx, leading to the formation of O_3 and secondary organic and inorganic aerosols (Ren et al., 2013; Ng et al., 2017). Due to the lack of direct measurements of short-lived, low-level radicals in Taiwan, this study aims to speciate and estimate the key radicals and oxidants during O_3 and $PM_{2.5}$ event and non-event days using a 0-dimensional photochemical box model coupled with the latest Master Chemical Mechanism (MCM v3.3.1) (Jenkin et al., 2015). The model was constrained with field measurements of 42 VOCs, O_3, and a suite of primary air pollutants. The preliminary results show that OH was the dominant radical (averaged 1.24×10^6 molec. cm^{-3}), peaking at noon and followed by comparable levels of HO_2 and RO_2 an hour later. On the other hand, the NO_3 peaked in the evening. The production of OH was mainly driven by the HOx cycling between HO_2 and NO, whereas the major loss of OH was due to its reaction with VOCs. The major contributor to the oxidation capacity by OH was CO, followed by aromatics and alkanes. NOx comprised the most significant fraction of the OH reactivity during daytime, whereas the aromatics became an important contributor at night. The O_3 concentration was strongly correlated with HO_2 concentration, by which the reaction with the peroxyacetyl radical ($CH_3C(O)O_2$) was a notable driver of O_3 formation. Nighttime chemistry of NO_3 and N_2O_5 hydrolysis were implicated in elevated $PM_{2.5}$ in the following morning hours. These modeling results raise interesting questions and hypotheses that may inform future research in deciphering the role of atmospheric radicals and oxidants in the formation of secondary pollutants in Taiwan.

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Impact of SO_2 on α-pinene Ozonolysis-derived Secondary Organic Aerosol Formation and its Implications

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ABSTRACT

The oxidation of biogenic volatile organic compounds (BVOCs), like α-pinene, by ozone is a significant pathway for secondary organic aerosol (SOA) formation. In this study, the interaction between α-pinene ozonolysis and SO_2, an anthropogenic emission, was investigated using a flow tube reactor. Results showed that SO_2 addition enhanced particle formation, likely due to interactions between ozonolysis products and H_2SO_4. The hygroscopicity (κ) of the formed SOA varied with particle size, indicating differences in aging processes. Modeling simulations will further elucidate the impact of SO_2 on SOA formation processes, providing insights into anthropogenic influences on the environment.

Enhancement of the secondary organic aerosol yields and the formation of low- and extremely low-volatility organic compounds during the oxidations of α-pinene in the presence of acidic seeds

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ABSTRACT

Secondary organic aerosols (SOAs) affect human health and climate change; however, the factors (e.g., temperature, acidity of pre-existing particles, and oxidants) influencing their formation are not sufficiently understood. The temperature and acidity dependence of SOA yields and chemical components of SOA from α-pinene ozonolysis[1] and α-pinene photooxidation under low NO_x conditions were systematically investigated under 278–298 K temperatures in the presence of neutral ($(NH_4)_2SO_4$) and acidic (H_2SO_4 + ($(NH_4)_2SO_4$)) seeds by using a compact chamber. The slight negative temperature dependence of the SOA yields was observed and the enthalpies of vaporization were estimated to be 25–48 kJ mo^{-1}, which is in agreement with the value of 40 kJ mol^{-1} applied in the CMAQ model. The SOA yields increased ~10–30 % with the increase in the acidity of seed particles ($[H^+]$ = 220 nmol m^{-3}) at low SOA mass loadings. Those increases were thought to be caused by the formation of low- and extremely low-volatility organic compounds such as dimer esters and organosulfates, which were measured by means of negative electrospray ionization liquid-chromatography time-of-flight mass spectrometry coupled to ion mobility spectrometry. To achieve better simulation of monoterpene SOA formation in the SOA module of the model, we propose that the conversion rate (τ^{-1}) of semi-volatile organic compounds (SVOCs) to non-volatile compounds (NVOCs) should be faster under acidic conditions compared with that under neutral conditions.

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Functional group analysis of gas-phase oxidation products of monoterpenes using high-resolution collision-induced dissociation mass spectrometry

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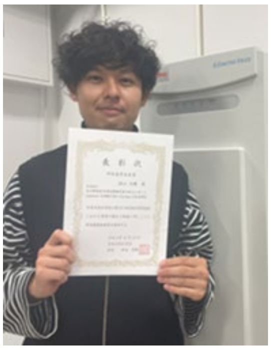

ABSTRACT

Forests emit large amounts of monoterpenes ($C_{10}H_{16}$) into the atmosphere. They are unsaturated hydrocarbons that are highly reactive with ozone (O_3), hydroxyl radicals, and NO_3 radicals in the atmosphere and can be converted into highly oxygenated organic molecules (HOMs), which includes multiple functional groups.1) HOMs are significantly involved in the formation of secondary organic aerosols (SOAs) that affect the earth's radiation balance. The potential of SOA formation depends on the reactivity of the functional groups of gas-phase oxidation products with OH radicals. The major oxidation products of α-pinene are known as pinonic acid and pinonaldehyde, although many isomers with multifunctional groups have not been identified experimentally.

In this study, we performed experiments using no-chromatography atmospheric pressure corona discharge ionization mass spectrometry (APCDI-MS)2) combined with collision-induced dissociation (CID) methods to analyze oxidation products. First, CID experiments for standards of α-pinene oxidation products were performed to investigate the relationship between functional groups and specific fragmentation pattern. It was found that the specific neutral loss in CID depends upon the type of functional group. Based on these relationships, we estimated the functional groups of various oxidation products originating from nine different monoterpenes. As an example, the results for oxidation products $C_{10}H_{16}O_3$ of nine monoterpenes are shown in Figure 1. The percentage of functional groups derived from the ion intensities of fragment ions were different for each monoterpene. From these results, we will discuss oxidation reactions of several monoterpenes in this workshop.

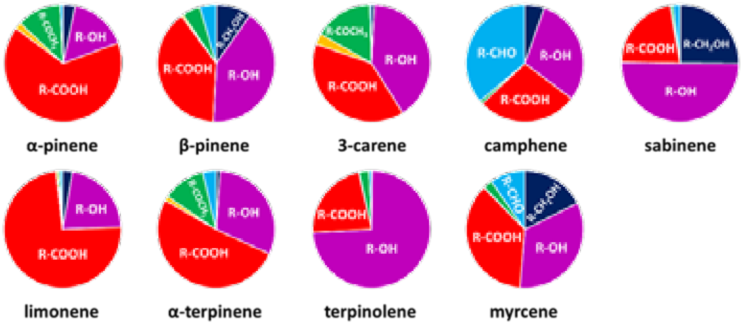

Figure 1 The ratios of functional group of oxidation products $C_{10}H_{16}O_3$ formed by nine monoterpenes.

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A study of formation process of a multifunctional ring-opening product present in secondary aerosol from aromatic compounds

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ABSTRACT

Multifunctional ring-opening products such as 2,3-dihydroxy-4-oxopentanoic acid (DHOPA), a secondary organic aerosol (SOA) tracer, are a major class of compounds present in SOA formed form the photooxidation of aromatic compounds. To provide fundamental knowledge related to the photooxidation mechanisms of aromatic compounds, we have studied the formation mechanism of DHOPA. In a previous study, we revealed that the ratio of DHOPA to total SOA formed from the photooxidation of toluene (f_{SOA}) is constant under various OH exposure conditions. These results suggest that DHOPA is formed by a reaction faster than SOA formation and stable under OH exposure. In this study, we performed a series of chamber experiments on the photooxidation of nine aromatic compounds. During chamber experiments, we measured the signal of gaseous products including ring-opening products (C4–C7 unsaturated dicarbonyls and C4–C5 furandiones), benzaldehyde derivatives, and phenol derivatives using a proton-transfer-reaction high-resolution mass spectrometer (PTR-MS). Using measured signal intensities and estimated sensitivities, the yields of total ring-opening products, benzaldehyde derivative, and phenol derivative formed from nine aromatic compounds were evaluated. The evaluated yields measured for nine aromatic compounds were compared with the corresponding f_{SOA} values measured by Al-Naiema et al. (2020) for DHOPA and the correlation coefficients were determined. The determined correlation coefficients were positive for total ring-opening products and phenol derivative, whereas it was negative for benzaldehyde derivative. The f_{SOA} of DHOPA is reported to be very low for the oxidation of phenol derivatives according to Al-Naiema et al. (2020). These results indicate that DHOPA is a second-generation product formed via ring-opening products and/or a first-generation product formed via bicyclic peroxy radicals, the precursor of ring-opening products.

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Atmospheric Simulation of Organic Aerosol Sources and Ozone Production Regime

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ABSTRACT

For the accurate understanding of the sources and atmospheric processes of atmospheric pollutants, including $PM_{2.5}$ and O_3, we conducted atmospheric simulations using a regional-scale chemical transport model.

Organic aerosol (OA), which is one of dominant components of $PM_{2.5}$, is composed of thousands of chemical compounds with a wide range of volatilities, and the sources and production pathways of OA are complicated. For accurate source apportionment of OA, we conduct field measurements of organic tracers at three sites and numerical simulations of forward and receptor models.[1] Cross-validation demonstrates that the source contributions estimated by the forward model generally agree with those of the receptor models for the major OA sources: mobile sources, biomass combustion, biogenic SOA, and anthropogenic SOA.

Understanding the atmospheric ozone (O_3) production regime is necessary for development of strategies to effectively control the amount of atmospheric O_3. We conduct smog-chamber experiments on the oxidation of nitrogen oxides (NOx) and volatile organic compounds (VOC) and use the instrument to directly evaluate the O_3 production regime.[2] The experimental results were compared with results of box model simulations with two chemical mechanisms to cross validate the measured and simulated O_3 production regimes. The O_3 production regimes estimated by the new instrument and by simulations are consistent with each other, and both the measured and simulated timing of the regime transition estimated from the sensitivity to injection of O_3 precursors NOx and VOC agree well with that estimated from the radical budget.

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Do typhoons have an impact on ground-level ozone and secondary organic aerosol formation?

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ABSTRACT

Ozone (O_3) is one of the most important tropospheric pollutants as it is related to a series of health issues for humans and plants, oxidation efficiency of the atmosphere, secondary organic aerosol (SOA) formation, and climate change (von Schneidemesser et al., 2015). A recent study showed that the typhoons in the North-western pacific region can increase regional O_3 levels by enhancing the interactions between biogenic and anthropogenic emissions (Wang et al., 2022). Typhoon Khanun in August 2023, that went along Okinawa, Japan, was not an exception, and it had a strong impact on local O_3 levels, BVOC emissions, and SOA formation at a sub-tropical forest site (Figure 1). Before the typhoon crossed Okinawa from 30th July to 6th August, the meteorology, $PM_{2.5}$, O_3, BVOC levels were steady, typical of this time of the year. Their levels rose sharply directly after the typhoon left the Okinawa islands, and they fell slowly during two weeks to the level of the pre-typhoon period. Their carbon oxidation states (OSc) also increased considerably after the event, showing that the typhoon enhanced SOA formation processes over the forest site. This presentation will also include chemical transport simulation results including the formation and loss processes of ozone during the prior and post typhoon periods.

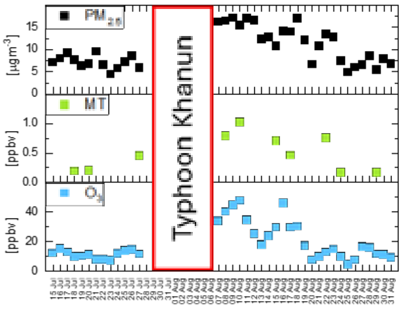

Figure 1. Time series of $PM_{2.5}$, monoterpenes, and O_3 between 15 July and 31 August 2023. $PM_{2.5}$ and O_3 data are obtained from Okinawa Prefecture Atmospheric Pollutant Monitoring System (https://okinawa-taiki.sakura.ne.jp/kyoku)

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An observation-based diagnosis for the air pollution in the Central-Western Taiwan

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ABSTRACT

Air pollution in Central-Western Taiwan (CWT) has seriously concerned the society for a long time. Despite the significant efforts made in the last couple years, the influences of various pollution sources remain unclear. Since the 2021, this research team has been dedicated to studies on the air quality issues in the CWT area. We setup an observation station at the downtown of Taichung City to host an array of in-situ instruments for measurements of specific air pollutants and meteorological parameters. In addition, a series of intensive campaigns were performed to collect $PM_{2.5}$ samples over the study area to investigate the spatial distribution of $PM_{2.5}$ and its composition. The results showed that the daily (24-h) mean of $PM_{2.5}$ mass concentration exhibited a significant positive correlation with the ambient level of ozone (O_3) in Taichung City, indicating that the air pollution in CWT was due mostly to the formation of photochemical smog. The investigations of $PM_{2.5}$ composition revealed that the formation of ammonium nitrate was the key process causing the drastic increase of $PM_{2.5}$ during a pollution episode. We looked further into the sources of the precursors of nitrate aerosols. It was found that nitrogen oxides (NOx) was contributed mostly from local sources, in particular the power plant and traffics, whereas the sources of OH and O_3 remain unclear. Nevertheless, our measurements of volatile organic compounds (VOCs) and analysis of the relevant ozone formation potential identified toluene, xylene, ethylene, and isoprene as the major species contributing to the photochemical formation of O_3. Except isoprene, all the other major VOCs were found emitted mostly from vehicles. Thus, according to the results of this study, mitigation of the emissions of photochemical pollutants from vehicles is the most critical strategy to improve air quality in the study area.

Key Words: Air pollution, Photochemical smog, Central Western Taiwan

Potential Impacts of Energy and Vehicle Transition Through 2050 on Oxidative Stress-Inducing $PM_{2.5}$-Metal Concentrations in Japan

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ABSTRACT

Oxidative stress is one of the important mechanisms leading to inflammation in the body. Metal components in $PM_{2.5}$ have high oxidative stress induction potential. In this study, the impacts of renewable energy shifting, passenger car electrification, and lightweighting through 2050 on the atmospheric concentrations of $PM_{2.5}$-total mass, -Fe, -Cu, and -Zn in Japan were evaluated using a regional meteorology–chemistry model. We estimated energy mix and passenger car mix in 2050 based on the Japanese government targets [1, 2], and also considered future vehicle lightweighting by technological innovation [3]. The passenger car electrification reduced the concentrations of Fe and Cu, relatively significantly (−8% and −13%, respectively) than $PM_{2.5}$-total mass and Zn (−2%, −3%, respectively). This is because the penetration of battery electric vehicles (BEVs) effectively reduces brake wear-derived metals by their regenerative brake system (RBS). Furthermore, future vehicle lightweighting enhanced this (−3% and −4% more for Fe and Cu). Several previous studies have pointed that the BEVs penetration does not bring benefits in terms of $PM_{2.5}$-total mass concentrations, that is the same result in this study (passenger car electrification reduced $PM_{2.5}$-total mass concentration by only 2% in urban). However, this study indicated that the concentrations of metals with high oxidative stress toxicity were more significantly reduced, which is a new perspective. This presentation is based on Kayaba and Kajino (2023) [4].

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Seasonal and diurnal variations in the light-absorbing properties and molecular composition of soluble brown carbon in central Taiwan

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ABSTRACT

In recent years, there has been growing interest in the impact of brown carbon (BrC) on climate change, which exhibits strong light absorbability in the ultraviolet to visible wavelength range, with its optical properties closely related to atmospheric radiative enhancement. To gain insight into the impact of BrC on regional climate, this study investigated the properties of BrC in central Taiwan by collecting daytime and nighttime $PM_{2.5}$ samples over a year, from April 22, 2022, to February 28, 2023. Water-soluble organic carbon (WSOC) and methanol-soluble organic carbon (MSOC) were investigated. The light absorption coefficients at 365 nm of dissolved organic carbon ($Abs_{365,\ WSOC}$ and $Abs_{365,\ MSOC}$) showed high correlation with the mass concentrations of WSOC and MSOC, respectively. Both WSOC and MSOC revealed seasonal variations with the highest in winter and the lowest in summer. Additionally, Abs_{365} was found to be highly correlated with primary organic carbon (POC), indicating the dominance of POC over the light absorbability of OC in Taichung. Over the sampling periods, $Abs_{365,\ MSOC}$ was about 2-3 times that of $Abs_{365,\ WSOC}$, attributed to more BrC chromophores extracted by methanol than water. The absorption Ångström exponent (AAE) ranges of WSOC and MSOC were 4.23 to 9.33 and 3.41 to 5.86, with average values of 5.8 ± 0.93 and 4.57 ± 0.52, respectively, indicating the presence of derivative BrC in Taichung. The analysis of $MAE_{365,\ WSOC}$ indicated that BrC with higher oxidation levels has weaker light absorption abilities, while organic aerosols with lower polarity and lower oxidation levels have stronger light-absorbing abilities. In addition, it was found that the formation of SOC resulted in the decrease of $MAE_{365,\ MSOC}$, in which the levels of SOC were predominantly affected by atmospheric oxidation capacity during the daytime, while the quantity of SOC during the nighttime could be attributed to the accumulation during the day. Fluorescence analysis showed that the biogenic sources contributed more significantly to WSOC in summer, while the condensed aromaticity of fluorescent groups was higher in winter, indicating more aged WSOC formed in winter. The results of the PARAFAC model indicated that WS-BrC is dominated by humic-like substances (HULIS), and the oxygen content of HULIS varies with the aging degree of aerosols in different seasons. This study provides a scope of the optical properties of BrC in central Taiwan, which underpin the further investigation in the qualification and quantification of BrC chromophores. More research on the influence of BrC on climate warming and the dominant sources of air pollution is needed.

Elucidating the Dynamics of New Particle Formation in Central Taiwan: Observational Insights and Influencing Factors

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ABSTRACT

New Particle Formation (NPF) is a critical mechanism that drives the transformation of gaseous precursors into aerosols, leading to a significant increase in aerosol number concentrations. Occurring with varied frequencies and intensities in the atmosphere, NPF begins with an initial event, subsequently enhanced by mechanisms such as vapor condensation among others, facilitating particle growth into larger sizes. The phenomenon, linked to both regional cloud condensation nuclei production and urban haze formation, serves as a crucial pathway for secondary aerosol formation. Highlighting the significance of NPF, this research comprehensively examines its occurrence and the influencing factors in central Taiwan.

From April 2022 to October 2023, extensive observations at Tunghai University in Taichung, Taiwan, captured the complete particle size distribution from 11.8 nm to 20 μm using the Scanning Mobility Particle Sizer (SMPS) and Aerodynamic Particle Sizer (APS). Using the $\Delta N_{12\text{-}25nm}$ rank analysis method proposed by Aliaga et al. (2023), the $\Delta N_{12\text{-}25nm}$ of each observation day was calculated and ranked to assess the intensity of NPF. The analysis indicated a significant increase in particle number concentration around 9 a.m. on days when $\Delta N_{12\text{-}25nm}$ exceeded the 80th percentile, accompanied by pronounced growth patterns. Moreover, the intensity of NPF events was directly correlated with the duration of particle growth, identifying such days as Strong-NPF events. Favorable conditions for Strong-NPF in central Taiwan included increased ultraviolet (UV) radiation levels, high temperatures, low particle number concentration, and low relative humidity. Notably, the summer months exhibited a higher frequency of Strong-NPF events (27.06% to 50.5% of days), accentuated by the prevailing wind direction that facilitated the transport of volatile organic carbon (VOC) from the south, thus triggering NPF.

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