A preliminary study on wind tunnel simulations of the explosive growth and dissipation of fine particulate matter in ambient air

Jingxin Xu, Fahua Zhu, Sheng Wang, Xiuyong Zhao, Ming Zhang, Xinlei Ge, Junfeng Wang, Wenxin Tian, Liwen Wang, Liu Yang, Li Ding, Xiaobo Lu, Xinxin Chen, Youfei Zheng, Zhaobing Guo

PII: S0169-8095(19)30123-1
DOI: https://doi.org/10.1016/j.atmosres.2019.104635
Article Number: 104635
Reference: ATMOS 104635

To appear in: *Atmospheric Research*

Received date: 28 January 2019
Revised date: 3 July 2019
Accepted date: 28 July 2019

Please cite this article as: J. Xu, F. Zhu, S. Wang, et al., A preliminary study on wind tunnel simulations of the explosive growth and dissipation of fine particulate matter in ambient air, Atmospheric Research, https://doi.org/10.1016/j.atmosres.2019.104635

A Preliminary Study on Wind Tunnel Simulations of the Explosive Growth and Dissipation of Fine Particulate Matter in Ambient Air

Jingxin Xu[1,2], Fahua Zhu[1,*] zhufahua@vip.sina.com, Sheng Wang[1], Xiuyong Zhao[1], Ming Zhang[1], Xinlei Ge[3], Junfeng Wang[3], Wenxin Tian[1], Liwen Wang[1], Liu Yang[4], Li Ding[1], Xiaobo Lu[5], Xinxin Chen[5], Youfei Zheng[2], Zhaobing Guo[2]

[1]State Environmental Protection Key Laboratory of Atmospheric Physical Modeling and Pollution Control, State Power Environmental Protection Research Institute, Nanjing 210031, China

[2]Key Laboratory of Meteorological Disaster, Ministry of Education (KLME)/Joint International Research Laboratory of Climate and Environment Change (ILCEC)/Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disasters (CIC-FEMD), Nanjing University of Information Science & Technology, Nanjing 210044, China

[3]Collaborative Innovation Center of Atmospheric Environment and Equipment Technology (AEET), School of Environmental Science and Engineering, Nanjing University of Information Science & Technology, Nanjing 210044, China

[4]Nanjing Normal University, Nanjing 210023, China

[5]Nanjing Environmental Monitoring Center, Nanjing 210013, China

[*]**Corresponding author.**

Abstract:

This study, for the first time, investigated the explosive growth and dissipation of fine particulate matter (PM) in Nanjing based on simulations using a closed-circuit boundary-layer wind tunnel. The effects of relative humidity (RH) and liquid water content (LWC) on $PM_{2.5}$ growth were examined. It was found that increasing moisture led to explosive growth of $PM_{2.5}$ mass concentrations (average growth rate of 12.35 μg/m^3·min) and the pollution levels. Winds were found to significantly aid the dissipation of fine PM, and high concentrations of fine PM only persisted for a very short time and dissipated after several hours. LWC was found to correlate more strongly with fine PM concentrations than did RH. This work underlines the effect of moisture on explosive growth in fine PM, and provides a new approach for the simulation of fine PM growth and dissipation in ambient air.

Keywords: Wind tunnel, fine particulate matter, explosive growth, dissipation, relative humidity, liquid water content

1. Introduction

The rapid urbanization and industrialization of China has led to an air pollution complex that varies from one region to another. Fine particulate matter ($PM_{2.5}$) is one of the most important elements of hazy, polluted atmospheric conditions (Molina et al., 2004; Kang et al., 2013). Previous studies have found that the physicochemical properties of fine PM in the atmosphere (such as its composition, particle size, hygroscopicity, and optical properties) can change through a series of chemical reactions (Jimenez et al.,

2009; Cappa et al., 2012; Huang et al., 2013; Liu et al., 2013; Li et al., 2017), changing its effects on air quality (Eliason et al., 2003; Rudich et al., 2007; Sun et al., 2011; Ge et al., 2017; Chen et al., 2018). Furthermore, central and eastern China have experienced severe haze pollution in recent years; these events are typically characterized by extremely high PM pollution intensities and long persistence times (Zhang et al., 2013; Wang et al., 2014), resulting in widespread concerns throughout China. Researchers have been studied this phenomenon through field observations, laboratory research, and model simulations. However, since the mechanisms of fine PM and haze formation are highly complex, there are inconsistencies regarding the impact of meteorological factors, such as relative humidity (RH), on the occurrence of haze pollution (Wu et al., 2011; Song et al., 2012; Ding and Liu, 2014; Wu et al., 2018). Nonetheless, it is known that a certain RH level is required for the occurrence of haze pollution (Wu, 2014). Liquid water is ubiquitous in the atmosphere and provides an important place for the chemical reactions of aerosols in the aqueous phase, but unfortunately, research in this area is quite limited (Ye et al., 2018). In addition, studies on the mechanisms of PM generation and growth under humid conditions are also not well understood (Hu et al., 2016). Therefore, it is not yet clear how best to quantitatively describe the factor(s) responsible for severe, large-scale occurrences of haze pollution in China.

Eastern China is well known frequently experiencing sever haze pollution, and the Yangtze River Delta is one of the most highly polluted haze regions (Hu et al., 2014). In this study, a closed-circuit boundary-layer wind tunnel was used to perform several experiments (i.e., control test, humidification with liquid nitrogen test, and atomization humidification test) to simulate, for the first time, the explosive growth and dissipation of fine PM in the ambient air of Nanjing, China. The effects of RH and liquid water content (LWC) on fine PM properties were also examined. Subsequently, a quantitative analysis was conducted on the factors that determine the occurrence and dissipation of severe haze pollution, which could in turn provide the means and basis for managing and preventing atmospheric pollution.

2. Materials and Methods

The study site was inside the State Power Environmental Protection Research Institute, which is located in Nanjing's Pukou District (32°1′ N, 118°7′ E), China. The experiment was conducted in late March to early April, 2018, and used a 24 × 4 m work section of a closed-circuit boundary-layer wind tunnel. The original work section was modified by removing the pneumatic balance in the center, then the main observation instruments were installed inside the observation house below the work section. The distance between the sampling tube and the front end of the work section was 5.5 m (see Fig. 1 for details).

The advantages of using the wind tunnel for this experiment were that: 1) it can reflect the actual atmospheric environment of the study site; 2) we can control the conditions and investigate the corresponding changes of aerosol properties; and 3) the air insider the work section is not affected by external changes in wind velocity and the boundary layer height when the fans are turned off.

To better understand the fine PM variations under different RH conditions, the experiment was performed under three different RH conditions: (i) control (CK), (ii) humidification with liquid nitrogen (EXP_M), and (iii) atomized humidification (EXP_N). The principles of the EXP_M and EXP_N conditions were to increase RH with temperature decrease and the addition of water vapor, respectively; the purpose of the CK condition was to compared with the other two conditions.

In the CK experiment, the wind tunnel fan was first switched on to allow external air to enter the work section, and then switched off once the air had been evenly mixed. Prior to the humidification tests, both ends of the work section were sealed using clean foam boards to create a relatively enclosed space. A standing fan was then placed at each end, with both fans facing each other, to periodically evenly mix the air. Data were collected continuously.

During the EXP_M experiment, a 165 L pressurized liquid nitrogen cylinder was placed outside the work section and connected via a stainless-steel tube to an inlet below the work section. By controlling the pressure valve of the liquid nitrogen cylinder, liquid nitrogen was uniformly sprayed into the work section until the cylinder was empty, which took approximately 60 min. The entirety of the liquid nitrogen spraying process and the restoration of the work section temperature (T) and RH to their initial, pre-humidification conditions took approximately 110 min in total.

During the EXP_N experiment, an atomizer (SX-SG-6500, Suzhou Suxin Cleanroom Equipment Factory) was used to spray mist to humidify the work section. During each test, the atomizer was filled with 4 L pure and deionized water and turned on, it was only switched off when it stopped emitting mist. The entirety of the mist-spraying process and the restoration of T and RH to its initial, pre-humidification value took approximately 215 min. The restoration of initial PM concentrations (as measured by the observation instruments) lasted from between 5 and15 hours. After the end of each experiment, the foam boards at the ends of the work section were removed. Subsequently, the two doors of the wind tunnel were opened and the fans were switched on to create a direct-flow wind tunnel; thus, ventilating the tunnel. The ventilation of the tunnel continued until the values (including T, RH, and the concentrations of gaseous pollutants and fine PM) reached the initial pre-humidifitcaiton conditions and stabilized. The doors were then closed, after which the fans were switched off and the two ends of the work section were sealed for the next experiment. Each humidification experiment was repeated three times.

In this study, the mass concentration, PM_1, and chemical components of the fine PM were measured using a $PM_{2.5}$ on-line monitor (BAM1020, Met One Instruments, USA) and an aerosol mass spectrometer (AMS) (SP-AMS, Aerodyne, USA). Air pressure, T, and RH were measured continuously using a small automatic weather station (PH-CJ-1, Wuhan Xinpuhui Technology Co., Ltd.), whereas SO_2, NO_2, O_3, and NH_3 were measured using an ultraviolet (UV) fluorescence SO_2 analyzer (TH-2002H, Wuhan Tianhong Environmental Protection Industry Co., Ltd.), chemiluminescence NO_x analyzer (TH-2001H, Wuhan Tianhong Environmental Protection Industry Co., Ltd.), UV photometer O_3 analyzer (EC9810, Ecotech, Australia), and NO-NO_2-NH_3 chemiluminescence analyzer (Model 17i, Thermo Fisher Scientific, USA),

respectively. The observation data were measured in 5-min cycles. A silica gel drying tube was added to the front of the AMS sampling section to reduce the impact of RH on the instrument's collection efficiency. Prior to the observations, the relative ionization efficiencies (RIEs) of the various components were calibrated using a scanning mobility particle sizer spectrometer (SMPS;3938L75, TSI, USA; Wang et al., 2016a). The RIEs of organic matter (Org), nitrates, sulfates, ammonium salts, and chlorides were 1.4, 1.05, 1.18, 3.15, and 1.4, respectively. Further information of the SP-AMS and its operating principles have been previously detailed (Onasch et al., 2012; Wang et al., 2016b). Unit mass resolution (UMR) and high-resolution (HR) AMS data were processed using SQUIRREL and PIKA (ToF_AMS_HRAnalysis_v1_16H), respectively, written in Igor Pro 6.37 (Wavemetrics, Lake Oswego, OR, USA).

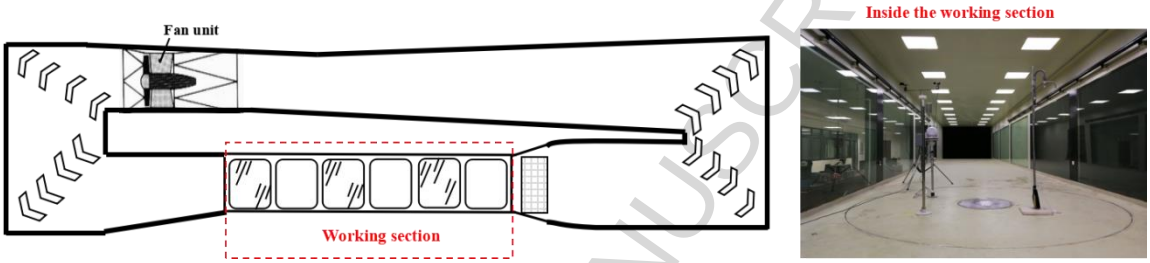


Fig. 1 Work section of a closed-circuit wind tunnel

3. Results and Discussion

3.1 Explosive growth and dissipation of atmospheric fine PM

The three rounds of explosive $PM_{2.5}$ growth and dissipation that occurred during the EXP_{N-1}, EXP_{N-2}, and EXP_{N-3} environmental wind tunnel simulations are shown in Fig. 2. After atomization humidification, $PM_{2.5}$ mass concentrations rapidly increased from approximately 40 μg/m^3 to 800 μg/m^3. The $PM_{2.5}$ growth rates in the three tests were 12.91, 11.21, and 12.93 μg/m^3·min (i.e., an average rate of 12.35 μg/m^3·min), respectively. Sun et al. (2019) investigated $PM_{2.5}$ mass concentrations growth events in Beijing and Shanghai and found average growth rates of ~7.1 and 6.6 μg/m^3 per hour, respectively; in our study, this would be deemed as "explosive growth". $PM_{2.5}$ mass concentrations began to decrease once the atomizer was turned off; further reductions took place after the fan in the wind tunnel was activated and the doors were opened. The restoration of the initial $PM_{2.5}$ concentration occurred around 14, 5, and 7 h after the three tests ended, respectively. These results show clearly that increasing moisture can lead to explosive growth in fine PM concentrations. The airflow significantly dissipated the fine PM, and the high concentrations of fine PM only persisted for a short time before dissipating after a few hours. This study has therefore provided a new method for simulating the growth and dissipation of fine PM (i.e., using the work section of a closed-circuit wind tunnel).

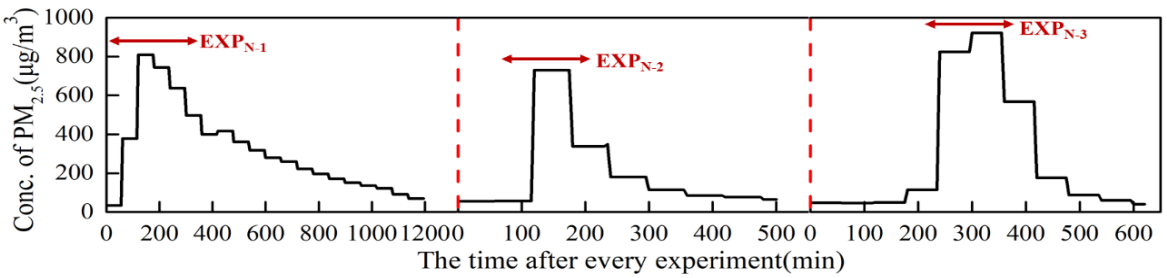


Fig. 2 Three rounds of $PM_{2.5}$ growth and dissipation, as simulated in an environmental wind tunnel (EXP_{N-1}–EXP_{N-3})

To obtain a better understanding of the above processes and to compare changes in fine PM concentrations with the CK and EXP_M conditions, we conducted a comparison between a randomly selected PM growth process in the CK experiment (48 hours) and the humidification periods of the EXP_M (110 min for each round) and EXP_N experiments (215 min for each round). The parameters of the EXP_M and EXP_N experiments were taken as the average of three tests. Fig. 3 illustrates how T, RH, $PM_{2.5}$, PM_1 (and its components), and the concentrations of gaseous pollutants varied under the CK, EXP_N, and EXP_M conditions. The CK conditions corresponded to the natural growth and dissipation of fine PM. By comparing the three test conditions, it was found that: (i) $PM_{2.5}$ concentrations peaked under CK at the lowest T and highest RH, whereas $PM_{2.5}$ concentrations only gradual decreased with increasing RH under the EXP_M conditions. $PM_{2.5}$ concentrations increased explosively under the EXP_N conditions when T was basically constant and RH was at its maximum; (ii) PM_1 concentration trends in all three conditions were generally the same as those of $PM_{2.5}$. The main component of PM_1 was organics; (iii) NO_2 and O_3 concentrations displayed diurnal trends under the CK conditions, whereas NO_2 concentrations decreased continuously under the EXP_M conditions. Furthermore, NO_2 and O_3 concentrations decreased to their minimum values under the EXP_N conditions.

On this basis, it can be concluded that the main pollutant in fine PM was Org. This is consistent with the findings of previous studies (Zhang et al., 2007; Huang et al., 2013). Fine PM concentration increase (including increasing explosively) with increasing RH under the CK condition and EXP_N conditions, or decreased slightly with increasing RH under the EXP_M condition. Therefore, RH should not be simply regarded as the main factor that determines the concentration of fine PM. This also explains why some studies have observed paradoxical relationships between RH and the occurrence of haze pollution. For example, Wu (2011) noted that high RH is a necessary condition for haze formation. After studying the weather in Nanjing, China, Mao and Liu (2012) concluded that high RH was needed for the occurrence of severe haze conditions. However, in a study regarding a severe haze incident in Jiangsu Province, Zhu et al. (2011) found that RH was usually around 45% when hazy weather conditions appeared in the Yangtze-Huai River area. Additionally, Song et al. (2012) proposed that the decline in RH in most parts of eastern China might be the primary cause of the increasing frequency of haze pollution during winter. Similarly, a study by Wu et al. (2016) found that increases in the number of hazy days in winter was significantly correlated with reductions in near-surface RH in the Yangtze-Huai River area and southern China. At the same time, other studies have concluded that aqueous-phase reactions in aerosols can

produce low-volatility organic compounds and secondary organic compounds (Ervens et al., 2004; Carlton et al., 2006). Therefore, we have deduced that changes in LWC might be a better factor that correlates generation of fine PM with RH.

A few interesting discoveries were also made in this study. For example, SO_4^{2-} concentrations did not fluctuate in the CK and EXP_M conditions, but a peak SO_4^{2-} concentration was observed under the EXP_N conditions. This indicates that the mechanism of SO_4^{2-} formation was different in each experiment, and this observation could be related to debated mechanism by which SO_2 is converted into SO_4^{2-} (Wang et al., 2016; Guo et al., 2017). As NO_2 and O_3 concentrations reached their minimum values under the EXP_N conditions, this may indicate that NO_2 and O_3 are involved in the relevant chemical reactions. Some previous studies have indicated that NO_2 and O_3 could participate in or promote the conversion of SO_2 to SO_4^{2-} (Shen et al., 2012; Liu et al., 2017). Additionally, NO_2 concentrations declined in the EXP_M-experiment, although the concentrations of the various components of PM_1 did not increase. This indicates that NO_2 may not have participated in a reaction related to atmospheric pollution. These results indicate that the impact of gaseous precursors on fine PM is not consistent.

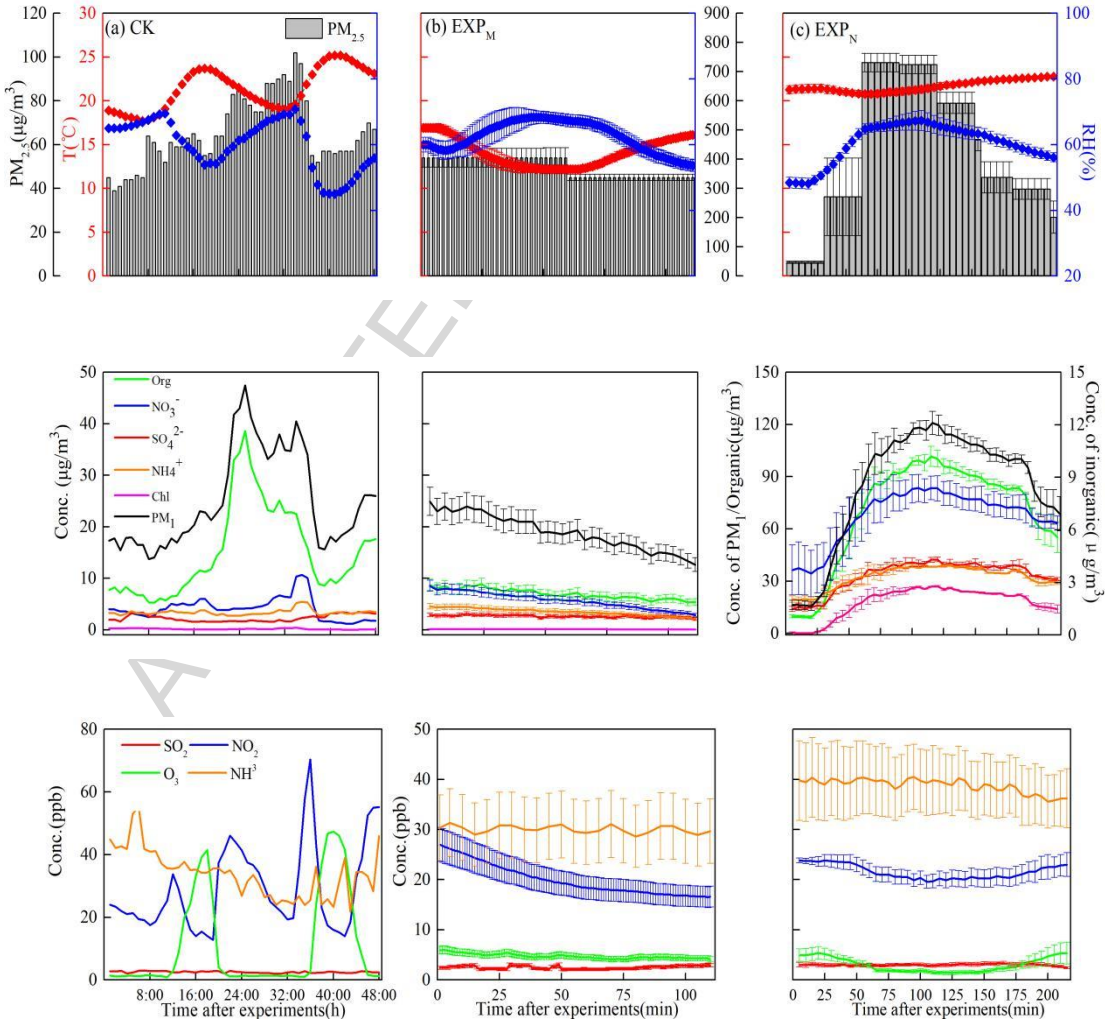


Fig. 3 Changes in T, RH, $PM_{2.5}$, PM_1 (and its components), and the concentration of gaseous pollutants in the CK, EXP_N, and EXP_M conditions

3.2 Relationship between fine PM and RH/LWC

The correlations between fine PM concentrations, RH, and LWC were analyzed (Fig. 4) to verify our hypothesis. Owing to a lack of instrumentation, LWC data were not collected directly; instead, we estimated the LWC. The average RH was ~60%(i.e., the air was not saturated). The steps for estimating LWC were as follows: 1) calculate the actual specific humidity (q) at each moment (i.e., mean value every 5 min), using the classical formula to calculate q:

$$q = 0.622 \times \frac{e_a}{p}, (g/kg) \quad (1)$$

$$e_s = 6.1078 \exp\left[\frac{17.269(T - 273.16)}{T - 35.86}\right] \quad (2)$$

$$e_a = e_s \times RH \times 100 \quad (3)$$

where e_a is the actual water vapor pressure (hPa), p is the air pressure (hPa), and e_s is the saturated water vapor pressure (hPa). The values of q differed significantly at each moment owing to temporal variations in T and RH.

2) Comparing adjacent moment values of q, the q of the next moment is subtracted from that of the preceding moment. If this subtraction yields a positive value, then condensation of liquid water must have occurred in the next moment, and the difference is the LWC. If the subtraction yields a value of 0 or a negative value, liquid water condensation did not occur in the next moment and the valve of LWC is zero.

3) Under the EXP_N condition, the value of LWC at each moment should include the amount of mist sprayed by the atomizer(0.091 L/min).

As shown in Fig. 4, $PM_{2.5}$ and PM_1 concentrations were positively correlated with RH under the CK and EXP_N conditions, with the correlation between PM_1 and RH being slightly higher (R^2 was above 0.39). On the other hand, there was no correlation between the fine PM concentrations and RH in EXP_M. However, LWC was significantly and positively correlated with $PM_{2.5}$ and PM_1 concentrations under all three experimental conditions. Under the CK and EXP_N conditions, the correlation between LWC and fine PM concentration was much stronger than that between RH and fine PM concentrations; the R^2 of the former was > 0.57, whereas the R^2 of the latter was only > 0.38. These results confirmed that LWC was more strongly correlated with changes in fine PM concentration than it was with RH. Some studies have

also noted that 80% of all particulate mass is gained through condensation on existing particles (Whitby, 1978). In some locations, the markers of secondary organic compounds are also more strongly correlated with LWC (Hennigan et al., 2009). Furthermore, it has been noted that increases in LWC lead to increases in the contribution of aqueous aerosols to fine PM concentrations (Ye et al., 2018).

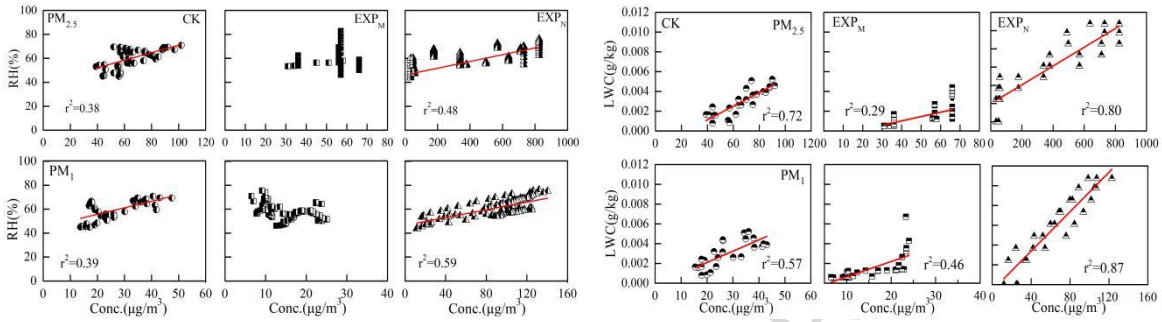


Fig. 4 Correlation between fine particulate matter (PM) concentration and relative humidity (RH; left) & liquid water content (LWC; right)

4. Conclusions

The explosive growth and dissipation of atmospheric fine PM was, for the first time, simulated via humidification tests in a closed-circuit boundary-layer wind tunnel using a suite of on-line measuring instruments. The findings indicate that increasing moisture can induce explosive growth in $PM_{2.5}$ mass concentrations and pollution levels. Wind has a significant effect on dissipating fine PM; the high concentrations of fine PM only persisted for very short periods of time, and dissipated after several hours. The wind tunnel therefore provides a new approach for the simulation of fine PM growth and dissipation in ambient air. By comparing the CK, EXP_N, and EXP_M experimental conditions, it was found that increasing moisture can lead to explosive growth in fine PM concentrations and LWC correlated more strongly with changes in fine PM concentration than did RH. In addition, Org was found to be the main component of PM_1. Based on these results, strong measures should be implement to control the emission of primary organic aerosols (POA) or volatile organic compounds. Furthermore, it was found that the formation of fine PM involves complex mechanisms, and the effects of gaseous pollutants on fine PM are not always the same; these issues and the aqueous-phase reactions of fine PM will be the major focus of our future research.

Acknowledgments: This study was supported by the National Natural Science Foundation of China (No.41771498), the group project of China Energy Investment Corporation (No. GJNR-18-31, H2018Y004), and the science and technology project of China Guodian Science and Technology Research Institute (NO.H2017Y001) and the State Environmental Protection Key Laboratory of Atmospheric Physical Modeling and Pollution Control of China. We would like to thank Jiangsu Province Nanjing Environmental MonitoringCenter for providing online monitoring instruments.

References

Cappa, C.D., Onasch, T.B., Massoli, P., et al., 2012. Radiative absorption enhancements due to the mixing state of atmospheric black carbon. Science. 337, 1078-1081.

Carlton, A.G., Turpin B J, Lim H J, et al., 2006. Link between isoprene and secondary organic aerosol (SOA): Pyruvic acid oxidation yields low volatility organic acids in clouds. Geophys. Res. Lett. 33, L06822.

Chen, Y., Ge, X., Chen, H., et al., 2018. Seasonal light absorption properties of water-soluble brown carbon in atmospheric fine particles in Nanjing, China. Atmos. Environ. 187, 230-240.

Ding, Y.H., Liu, Y.J., 2014. Analysis of long-term variations of fog and haze in China in recent 50 years and their relations with atmospheric humidity. Science China: Earth Sciences. 57, 36-46. (In Chinese)

Eliason, T.L., Aloisio, S., Donaldson, D.J., et al., 2003. Processing of unsaturated organic acid films and aerosols by ozone. Atmos Environ. 37, 2207-2219.

Ervens, B., Feingold, G., Frost, GJ., et al., 2004. A modeling study of aqueous production of dicarboxylic acids: 1. Chemical pathways and speciated organic mass production. J Geophys Res. 109, D15205.

Ge, X.; Li, L.; Chen, Y.; et al., 2017. Aerosol characteristics and sources in Yangzhou, China resolved by offline aerosol mass spectrometry and other techniques. Environ. Pollut. 225, 74-85.

Guo, H.Y., Weber, R.J., Nenes, A., et al., 2017. High levels of ammonia do not raise fine particle pH sufficiently to yield nitrogen oxide-dominated sulfate production. Scientific Reports. 7, 12109.

Hennigan, C.J., Bergin, M.H., Russell, A.G., et al., 2009. Gas/particle partitioning of water-soluble organic aerosol in Atlanta. Atmos. Chem. Phys. 9(1), 3613-3628.

Hu, J., Wang, Y., Ying, Q., et al., 2014. Spatial and temporal variability of $PM_{2.5}$ and PM_{10} over the North China plain and the Yangtze River delta, China. Atmos. Environ. 95, 598-609.

Hus, M., Shang, D.J., Guo, S., et al., 2016. Mechanism of new particle formation and growth as well as environmental effects under complex air pollution in China. Acta Chimica Sinica. 74, 385-391. (In Chinese)

Huang, X.F., Xue, L., Tian, X.D., et al., 2013. Highly time-resolved carbonaceous aerosol characterization in Yangtze River Delta of China: composition, mixing state and secondary formation. Atmos Environ. 64, 200-207.

Jimenez, J.L., Canagaratna, M.R., Donahue, N.M., et al., 2009. Evolution of organic aerosols in the atmosphere. Science. 326, 1525-1529.

Kang, H.Q., Zhu, B., Su, J.F, et al., 2013. Analysis of a long-lasting haze episode in Nanjing, China. Atmos Res. 120-121, 78-87.

Liu, D., Allan, J., Whitehead, J., et al., 2013. Ambient black carbon particle hygroscopic properties controlled by mixing state and composition. Atmos Chem Phys. 13, 2015-2029.

Liu, M.X., Song, Y., Zhou, T., et al., 2017. Fine particle pH during severe haze episodes in northern China. Geophysical Research Letters. 44, 5213-5221.

Mao, Y.Q., Liu, D.Q., 2012. The characteristic and relationship with meteorological elements of haze weather in nearly 3 years in Nanjing. The 9th annual proceedings of the Yangtze river delta meteorological science and technology. (In Chinese)

Molina, M.J., Molina, L.T., 2004. Megacities and Atmospheric Pollution. J Air Waste Manage Assoc. 54, 644-680.

Onasch, T. B., Trimborn, A., Fortner, E. C., et al., 2012. Soot particle aerosol mass spectrometer: Development, validation, and initial application. Aerosol Sci. Tech. 46(7), 804-817.

Rudich, Y., Donahue, Ne.M., Mentel, T.F., 2007. Aging of organic aerosol: bridging the gap between laboratory and field studies. Annu Rev Phys Chem. 58, 321-352.

Shen, X., Lee, T., Guo, J., et al., 2012. Aqueous phase sulfate production in clouds in eastern China. Atmos Environ. 62, 502-511.

Whitby, K.T., 1978. The physical characteristics of sulfur aerosols. Atmos. Environ. 12, 135-159.

Song, Y.F., Liu, Y.J., Ding, Y.H., 2012. A study of surface humidity changes in China during the recent 50 years. Acta Meteor Sinica. 26(5), 541-553.

Sun, J., Liang, M., Shi, Z., et al., 2019. Investigating the $PM_{2.5}$ mass concentration growth processes during 2013-2016 in Beijing and Shanghai. Chemosphere, 221, 452-463.

Sun, Y.L., Zhang, Q., Schwab, J.J., et al., 2011. A case study of aerosol processing and evolution in summer in New York City. Atmos Chem Phys. 11, 12737-12750.

Wang, G.H., Zhang, R.Y., Gomez, M.E., et al., 2016. Persistent sulfate formation from London fog to Chinese haze. PNAS. 29, 13630-13635.

Wang, J.F., Ge, X.L., Chen, Y.F., et al., 2016a. Highly time-resolved urban aerosol characteristics during springtime in Yangtze River Delta, China: insights from soot particle aerosol mass spectrometry. Atmos. Chem. Phys. 16, 9109-9127.

Wang, J., Onasch, T. B., Ge, X., et al., 2016b. Observation of fullerene soot in eastern China. Environ. Sci. Technol. Lett. 3 (4), 121-126.

Wang, Y.S., Yao, L., Wang, L.L., et al., 2014. Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China. Science China: Earth Sciences. 57, 14-25. (In Chinese)

Wu, D., 2011. Formation and evolution of haze weather. Environmental Science & Technology. 34(3),157-161. (In Chinese)

Wu, P., Ding, Y.H., Liu, Y.J., et al., 2016. Influence of the East Asian winter monsoon and atmospheric humidity on the winter time haze frequency over central-eastern China. Acta Meteorologica Sinica. 74(3), 352-366. (In Chinese)

Wu, Y., Ge, X., Wang, J., et al., 2018. Responses of secondary aerosols to relative humidity and photochemical activities in an industrialized environment during late winter, Atmos Environ. 193, 66-78.

Ye, Z.L., Qu, Z.X., Ma, S.S., et al., 2018. Secondary organic aerosol from aqueous reaction of aerosol water. Environmental Science. 39(8), 3954-3964. (In Chinese)

Zhang, Q., Jimenez, J.L., Worsnop, D.R., et al., 2007. A case study of urban particle acidity and its effect on secondary organic aerosol. Environ. Sci. Technol. 41, 3213-3219.

Zhang. X.Y., Sun, J.Y., Wang, Y.Q., et al., 2013. Factors contributing to haze and fog in China. Chin Sci Bull (Chin Ver). 58, 1178-1187. (In Chinese)

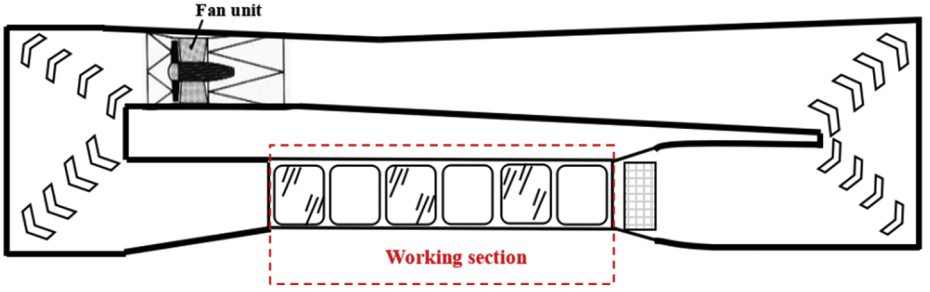
Zhu. J.L., Wang, T.J., Xing, L., et al., 2011. Analysis on the characteristics and mechanism of a heavy haze episode in Jiangsu Province. China Environmental Science. 31(12), 1943-1950. (In Chinese)

Highlights:

(1) This study used a closed-circuit boundary-layer wind tunnel.

(2) Increasing moisture led to the explosive growth of $PM_{2.5}$ mass concentrations.

(3) Liquid water content (LWC) was found to correlate more strongly with fine particulate matter (PM) concentrations than relative humidity (RH).



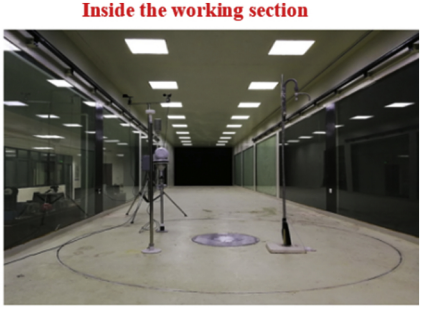


Figure 1

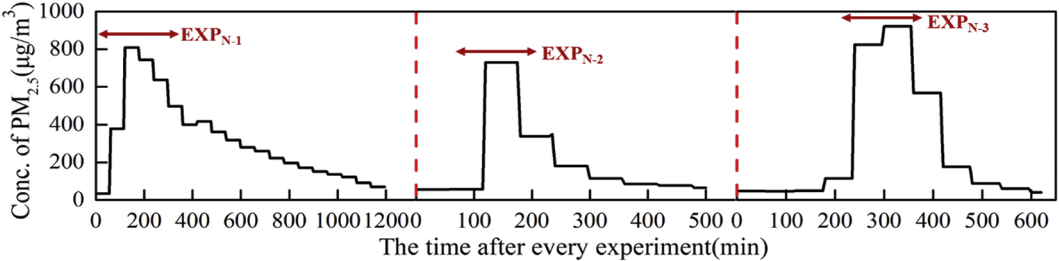

Conc. of PM2.5 (μg/m3)
1000
800
600
400
200
0
EXPN-1
EXPN-2
EXPN-3
0 200 400 600 800 1000 1200
0 100 200 300 400 500
0 100 200 300 400 500 600
The time after every experiment(min)

Figure 2

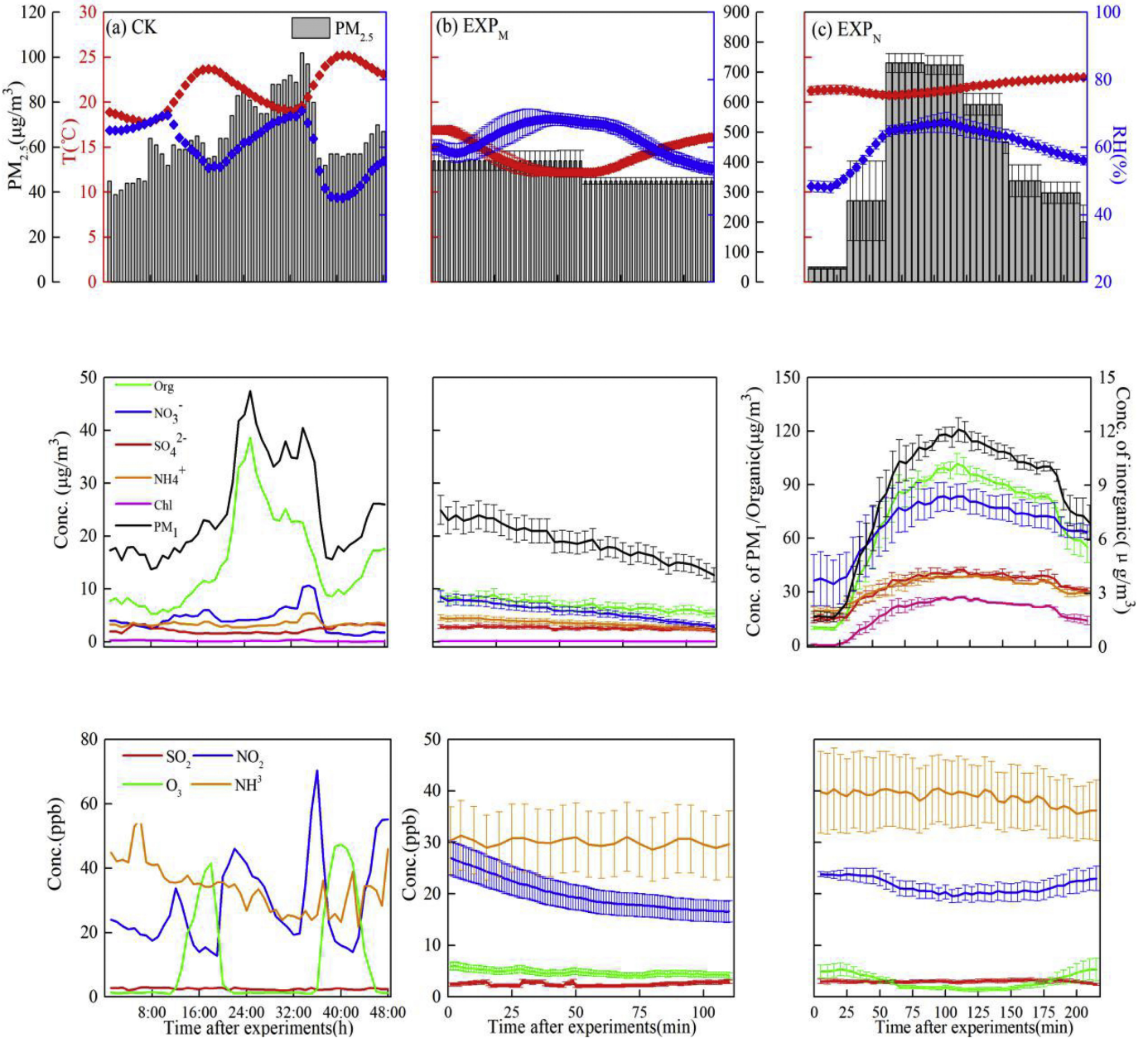

(a) CK
PM2.5
PM2.5(μg/m3)
T(℃)
(b) EXPM
(c) EXPN
RH(%)
Org
NO3-
SO42-
NH4+
Chl
PM1
Conc. (μg/m3)
Conc. of PM1/Organic(μg/m3)
Conc. of inorganic(μ g/m3)
SO2
NO2
O3
NH3
Conc.(ppb)
Time after experiments(h)
Time after experiments(min)

Figure 3

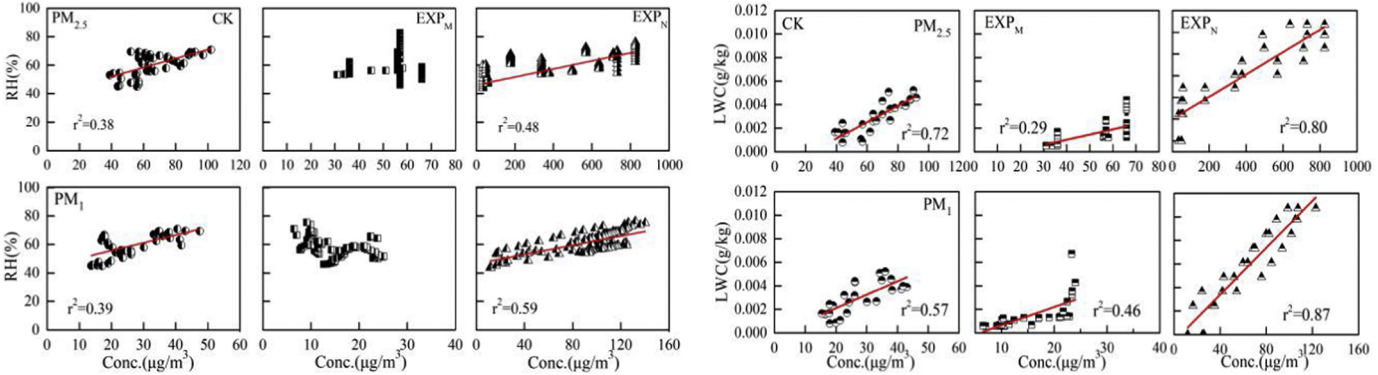

PM2.5
CK
EXPM
EXPN
PM1
RH(%)
LWC(g/kg)
r2=0.38
r2=0.48
r2=0.39
r2=0.59
r2=0.72
r2=0.29
r2=0.80
r2=0.57
r2=0.46
r2=0.87
Conc.(μg/m3)

Figure 4