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Low Hygroscopicity of Newly Formed Particles on the North China Plain and Its Implications for Nanoparticle Growth

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Key Points:

- Particles with sizes at 20–40 nm formed through new particle formation (NPF) are not very hygroscopic at rural North China Plain (NCP)
- The hygroscopicity parameter κ of the growing material varied from ~0 to ~0.1 among NPF events
- Precursors, oxidants and meteorological conditions may synergistically determine the chemical compounds responsible for particle growth

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract

The growth of newly formed particles through new particle formation (NPF) contributes a significant fraction to the cloud condensation nuclei, yet the driving mechanisms remain unclear, especially for polluted environments. To investigate the potential species contributing for nanoparticle growth in environments with significant anthropogenic influences, we measured the hygroscopicity of newly formed particles at 20–40 nm at a rural observational site in the North China Plain during winter 2018. Our results demonstrate that these particles were not very hygroscopic, with the mean hygroscopicity parameter κ of 0.13 ± 0.09. Clear differences in the inferred κ of the growing material responsible for the growth were observed among different events, indicating that even at the same region, the compounds driving particle growth may not be identical. This may be synergistically influenced by the NPF precursors, oxidants and meteorological conditions, suggesting complex mechanisms might co-exist behind nanoparticle growth in polluted environments.

Plain Language Summary

Particles newly formed through nucleation grow large enough to become cloud condensation nuclei which influences climate. However, the driving mechanisms and the key contributing species for nanoparticle growth are poorly understood. Here, we present direct measurements of the hygroscopicity of particles through new particle formation (NPF) that occurred in a rural environment in the North China Plain (NCP) during wintertime of 2018. Relatively low hygroscopicity of these newly formed particles were observed, suggesting less water-soluble compounds drove the growth there. We also found that the hygroscopicity of the growing material responsible for nanoparticle growth was not identical on different NPF days. Our results reveal that the compounds driving the growth varied with different NPF conditions, deepening our understanding of nanoparticle growth in polluted environments.

1. Introduction

New particle formation (NPF) is an important source of particles in the atmosphere and these newly formed particles must grow large enough to contribute to the global cloud condensation nuclei (CCN) (Dunne et al., 2016; Gordon et al., 2017; Kerminen et al., 2018). The contribution of atmospheric NPF to the CCN budgets varies significantly over different environments and temporal scales, resulting in the large uncertainties in radiative climate forcing (Reddington et al., 2017). This is largely due to the high uncertainty behind how these newly formed particles grow to CCN-relevant sizes (Pierce & Adams, 2007; Riipinen et al., 2011; Spracklen et al., 2008).

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In order to elucidate the driving mechanisms of nanoparticle growth, it is essential to obtain detailed information of the key contributing species that are involved in these processes. Previous studies concluded that sulfuric acid (H_2SO_4), amines, ammonia and organic compounds together drive the growth of nucleated particles (diameter <3 nm) to larger sizes (diameter >30 nm), which can act as CCNs, in terrestrial environments (Deng et al., 2020; Kulmala et al., 2021; Yao et al., 2018; Zha et al., 2023). However, all these findings are based on the approaches identifying molecules in the gas phase (Bianchi et al., 2016; Deng et al., 2020; Mohr et al., 2019; Qiao et al., 2021; Stolzenburg et al., 2020; Zha et al., 2023). Direct characterization of the chemical composition of newly formed particles is still a challenge (Li et al., 2022). This is largely due to the difficulties in analyzing chemical composition with low mass concentrations. It is still not clear which are the key driving species and what their relative contributions are to nanoparticle growth.

Indirect approaches, such as measurements of aerosol hygroscopicity or volatility, are a feasible alternative to monitor the evolution of the chemical species responsible for nanoparticle growth. Measurements conducted in a boreal forest environment suggest that more hygroscopic compounds drive nanoparticle growth there (Ehn et al., 2007; Ristovski et al., 2010). At an urban site in central Germany, a previous study (Wu et al., 2015) found that water soluble fractions that assumed to be $(NH_4)_2SO_4$-equivalent increase when newly formed particles grow up to 30 nm in sizes, whereas less hygroscopic organics start to play a role at the later stages of growth. However, most of these studies mainly focus on the variation of hygroscopicity of newly formed particles themselves and direct investigation in the changes of the chemical composition of species that contribute nanoparticle growth in various environments is still quite limited (Zheng et al., 2020). Or they assumed particles consist of only ammonium sulfate and an insoluble organic fraction and further calculate how the soluble fraction varies over the entire course of NPF. Such an assumption could differ largely from that of real ambient particles and result in large uncertainties in the estimation of the properties of contributing species for particle growth.

In this study, we measured the hygroscopicity of newly formed particles at a rural observational site in the NCP, which is strongly influenced by anthropogenic emissions, during the McFan (Multiphase chemistry in Fogs and Aerosols in the NCP) campaign in 2018. Based on the Zdanovskii-Stokes-Robinson (ZSR) mixing rule, hygroscopicity of the contributing species for nanoparticle growth was directly obtained by following the temporal evolution of aerosol hygroscopicity during particle growth, which allows a further estimation of the possible candidates for key chemical components responsible for the growth. Finally, our results show significant differences in the hygroscopicity of contributing species for particle growth and demonstrate the synergistic roles of distinct precursors as well as meteorological conditions in these processes on different NPF days.

2. Data and Methods

2.1. Measurements

The field measurements were conducted at Gucheng, a rural observational site (39.15° N, 115.74°E; 22 m a.s.l.) in the northern North China Plain, from 12 November to 24 December 2018 during the McFan (Multiphase chemistry in Fogs and Aerosols in the North China Plain) campaign. Detailed information about this site and campaign can be found in previous studies (Hong et al., 2023; Li et al., 2021). Ambient aerosols were sampled through a low-flow PM_{10} cyclone inlet with a Nafion drier where the sample RH was reduced to below 20%. The particle number size distribution ranging from 10 to 1,000 nm was measured with a scanning mobility particle sizer (SMPS, TSI model 3938). Aerosol hygroscopicity of selected particles was measured using a self-assembled hygroscopicity tandem differential mobility analyzer (HTDMA) (Hong et al., 2018). To be specific, the hygroscopic growth factor (HGF) was measured at the relative humidity (RH) of 87% for 20, 25, 30 and 40 nm particles. The instrument was calibrated, that is, RH calibration, by comparing the measured HGF of ammonium sulfate particles at 87% RH to the ones of previous studies. Further details of the HTDMA can be found in Shi et al. (2022).

At the GC site, meteorological parameters, including ambient temperature, RH, wind speed and direction, solar radiation, were continuously recorded during the McFan campaign (Li et al., 2021). Atmospheric non-methane hydrocarbons were collected by 3.2 L pre-evacuated stainless steel Suma canisters and further analyzed using an offline GC-MS/FID system (He et al., 2022). The concentration of conventional trace gases, including O_3, NO_X, CO and SO_2, were measured by a series of commercial analyzers from Thermo Environmental Instruments (Model 49C, 42C, 48iTLE and 43C).

2.2. Deriving Hygroscopicity Parameter κ

In this study, a single hygroscopicity parameter κ was derived from the hygroscopic growth factor (HGF) measured by our HTDMA system according to Petters and Kreidenweis (Petters & Kreidenweis, 2007) as:

$$\kappa = \left(\mathrm{HGF}^3 - 1\right)\left(\frac{K_e}{\mathrm{RH}} - 1\right). \tag{1}$$

RH is the relative humidity (ca. 87% RH) at the humidification unit inside our HTDMA system and K_e is the Kelvin term, which is calculated as:

$$K_e = exp\left(\frac{4M_W\sigma_W}{RT\rho_W D_p}\right). \tag{2}$$

M_W, σ_W and ρ_W are the molecular weight, surface tension and density of water, respectively. R is the ideal gas constant, T is the ambient temperature, D_P is particle diameter after humidification.

2.3. Estimation of the Hygroscopicity of Growing Material (GM)

Detailed classification of NPF events was based on the method proposed by Dal Maso et al. (2005) and can be found in Hong et al. (2023). Five NPF events were observed during the whole campaign. However, on the days of 6 and 23 December, hygroscopicity data was not available due to instrument failure. Therefore, only three NPF events on 18 November and 7–8 December 2018 can be used for further analysis.

As we focus mainly on NPF and their subsequent growth, only the hygroscopicity data representing newly formed particles during each NPF day were chosen for analysis. Specifically, taking the NPF event observed on 8 December 2018 (Figure 1a), as an example, the moment when the mode size of the measured particle number size distribution reached 20, 25, 30 and 40 nm, respectively, was denoted as t_1, t_2, t_3 and t_4. Then the hygroscopicity parameter data in a time window of 2 hr around each of these four moments was selected to represent the ones of newly formed particles at corresponding size.

Practically, any newly formed particles, for instance, 25 nm particles, were originated from the growth of relatively smaller ones, for example, 20 nm particles, by material (growing material, GM hereinafter) either through condensation or by particle-phase or multiphase reactions. According to the Zdanovskii-Stokes-Robinson (ZSR) relation (Stokes, 1965), the κ value of any internally mixed particles can be inferred from the hygroscopicity of different components within the particles with their corresponding volume fractions. Therefore, the hygroscopicity of the growing material within different size intervals can be estimated as:

$$\kappa_{i+1} = \varepsilon_i \cdot \kappa_i + (1 - \varepsilon_i) \cdot \kappa_{GM}, \tag{3}$$

where κ_{i+1} represents the hygroscopicity of particles at relatively larger sizes, for instance, κ_{25nm}, κ_{30nm} or κ_{40nm}, while κ_i represents the ones of relatively smaller particles, for example, κ_{20nm}, κ_{25nm} or κ_{30nm}; ε_i is the ratio of the volume of the smaller particles to that of the larger ones. Thus, we can calculate $\kappa_{GM,}$ the hygroscopicity parameter of the growing material that contributed to the growth for each size interval. It has to be noted that for the same chemical composition, the κ values of particles at different sizes, particularly in the range of 20–40 nm, may vary due to the nanosize effect (Cheng et al., 2015). The possible influence to the results will be discussed in the following section.

3. Results

3.1. Low Hygroscopicity of Newly Formed Particles

In this study, three NPF episodes on 18 November and 7–8 December 2018 were chosen for further analysis due to the availability of the hygroscopicity data set. These events showed clear particle bursts and continuous growth over several hours. On 18 November, particles only grew up to approximately 40 nm in size due to changing air masses that prematurely ended the growth event. On 7-8 December, we observed continuous growth to accumulation mode size particles, indicated by higher particle growth rate (GR) when the air mass remained. Detailed

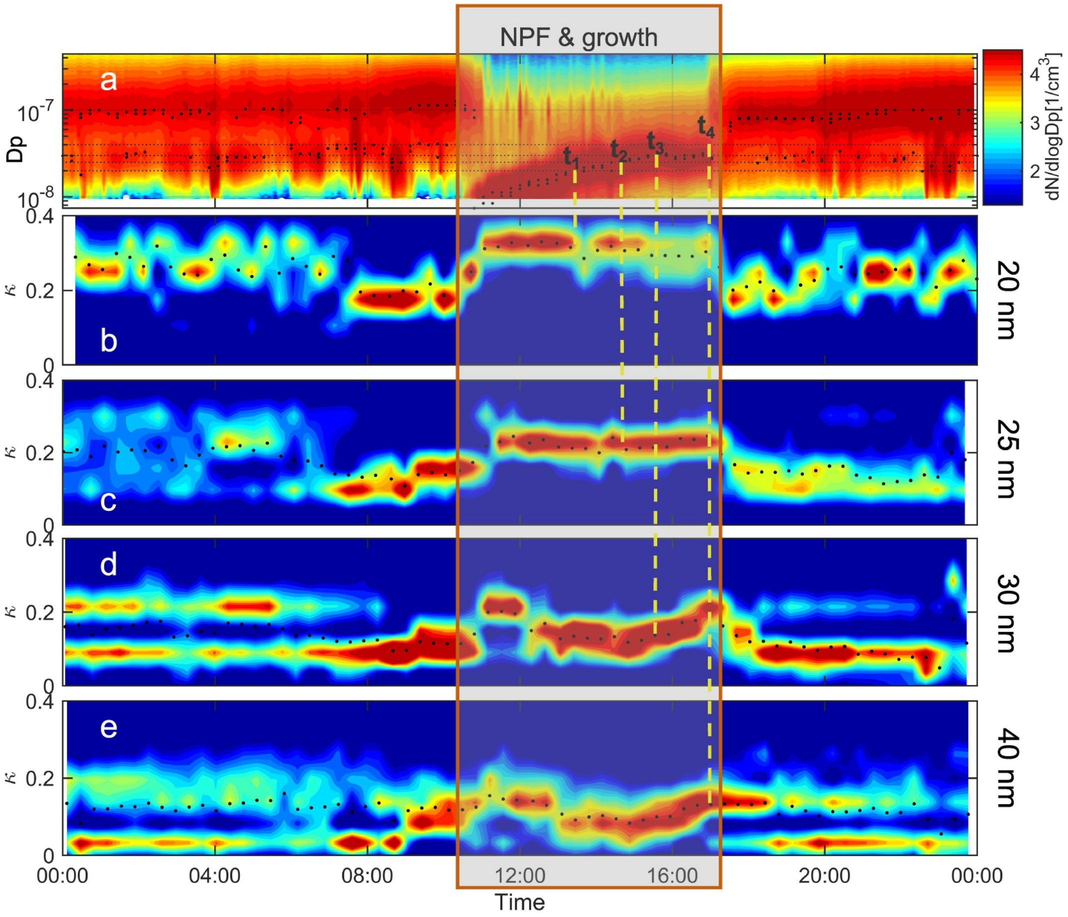

Figure 1. (a) Time series of particle number size distribution for a typical NPF event occurred on 18 November 2018 (b)-(e) Time series of the obtained κ-PDF (Probability Density Function) of particles at 20 nm, 25 nm, 30 nm and 40 nm during 18 November 2018.

characteristics regarding particle nucleation and subsequent growth for these events can be found in Hong et al. (2023).

For our GC site, we observed that the newly formed particles through NPF were not very hygroscopic, particularly for the two events in December. The mean κ obtained during this study was 0.13 ± 0.09, with the lowest ones approaching 0 for particles at 40 nm. Our results were in general lower than those in other relatively clean sites, such as the boreal forest in Hyytiälä of Finland with mean κ of 0.23 and a mixed deciduous forest in central Virginia, US with κ of 0.42 for 25 nm particles. The obtained κ in this study is still remarkably lower than other polluted environments, such as urban Beijing with κ of 0.21 for 40 nm particles. Many previous studies (Smith et al., 2010; Wu et al., 2013, 2017) suggested that water soluble organics, such as alkyl-ammonium-carboxylate salt and amines, may contribute significantly to nanoparticle growth. However, due to the lower hygroscopicity of newly formed particles of this study, the compounds that may explain current particle growth would be quite different, most likely being less water soluble.

A previous study considered that nanoparticles of identical chemical composition may exist in different phase-states depending on particle size (Cheng et al., 2015). As the solubility of chemical composition varies between different states, this will result in different aerosol hygroscopicity of the same composition, particularly for ~20-30 nm particles. However, we noticed that the difference in the obtained κ of ammonium sulfate particles at sizes between 20 and 40 nm was less than 1.7% (Biskos et al., 2006), which is within the uncertainties of our measurements and may not influence the main findings of this study. We are aware that this nanosize effect of ammonium sulfate particles might deviate from that of ambient aerosols, which are consisting of thousands of species and far more complex than single-component system. Therefore, this size dependency on aerosol

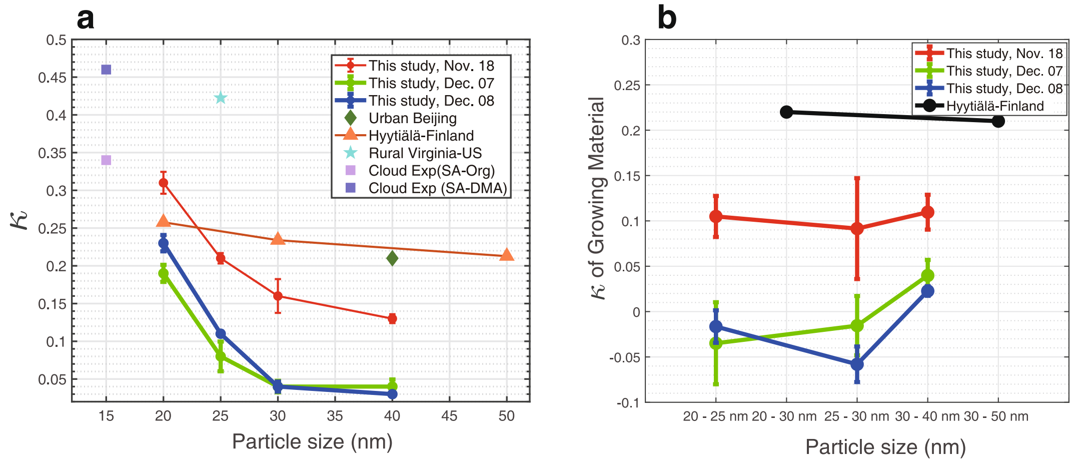

Figure 2. (a) Hygroscopicity of newly formed particles at a function of particles size obtained from different environments (Ehn et al., 2007; Kim et al., 2016; Liu et al., 2021; O'Halloran et al., 2009). The error bar of our data set represents one standard deviation of the data over the time window between 1 hour before the selected moment and 1 hour after that. (b) Hygroscopicity of contributing species for nanoparticle growth for our study as well as for the study conducted in Hyytiälä, South Finland. The error bar of our data set represents one standard deviation of the calculated distribution from Monte Carlo simulation, considering uncertainties of each relevant variable.

hygroscopicity cannot be fully addressed at the current stage for our study and would require comprehensive laboratory work covering a large variety of different compositions, from single component to highly complicated mixtures, in the future.

3.2. Different Hygroscopicity of Species That Contribute to the Growth

As shown in Figure 2a, the hygroscopicity parameter κ of newly formed particles decreased with increasing particle size during the events for our study. The κ values were always highest for the 20 nm particles, ranging from 0.19 to 0.31. This is plausibly explained by the fact that some hygroscopic or soluble compounds, for instance, H_2SO_4, were responsible for the early stage growth of new particles, causing the very small ones to be more hygroscopic. This is consistent with previous findings (Kulmala et al., 2001; Wu et al., 2013) and further supported by the coincidental rise in H_2SO_4 concentration during the start of NPF of our study (Fig. S1 in Supporting Information S1). Along with the subsequent growth of these particles to larger ones, for example, 25–40 nm particles, the κ values continued to decrease, suggesting that less hygroscopic material condenses on particles leading to their grow. Similar to our results, the measured κ of newly formed particles in Hyytiälä was also observed to decrease with increasing particles size, but with a much weaker size dependency.

To better probe the possible candidates of the compounds contributed to nanoparticle growth, we calculated the κ values of the growing material for different size intervals according to Equation 3 as described above. We found that the inferred hygroscopicity of the growing material during the NPF event in November at our GC site and at Hyytiälä (Ehn et al., 2007) showed no clear size-dependency. This indicates that for these events, compounds that contributed to particle growth at sizes beyond 20 nm until the "CCN-relevant" sizes may not vary significantly and chemical attributes such as hygroscopicity remained relatively constant. However, for the two NPF events in December in our study, the measured κ of newly formed particles displayed different patterns. A slight increasing trend with increasing particle size in the inferred hygroscopicity of the growing material was observed for the two events in December. This tells that for those cases, the compounds driving particle growth may not be identical at different growth stages, though their hygroscopicity were all extremely low.

On the other hand, we obtained negative values of κ of the growing material contributing to particle growth at sizes below 30 nm for the two events in December at GC. As suggested by previous studies (Wu et al., 2013), this could arise from the evaporation of the semi-volatile components in particles during drying, sizing or humidifying processes inside the HTDMA or during the increase of temperature as our measurement container was maintained at a higher temperature than the ambient. However, if the evaporation of semi-volatile compounds in particles could fully explain the negative κ values, we would obtain an even lower negative κ values of the growing material for particles at size interval between 30 and 40 nm compared to lower sizes. That is because as nanoparticles grow larger, the Kelvin effect becomes weaker and the condensation of semi-volatile compounds contributes more to particle growth, probably leading to more content of semi-volatile compounds in relatively larger particles compared to smaller ones (Tröstl et al., 2016). Furthermore, Gysel et al. (2007) calculated that 50%–60% of the volume of semi-volatile compounds evaporated in 50–100 nm particles during a residence time of around 60 s in their HTDMA. For our system, the residence time is less than 10 s, and the evaporation of semi-volatile compounds in particles should be much smaller. In order to verify this estimation, we measured the evaporation of 20–40 nm particles of ammonium nitrate, that is, a surrogate for semi-volatile compounds (Hong et al., 2017; Z. J. Wu et al., 2013), mixed with ammonium sulfate, which is considered as low volatile (Hong et al., 2017), with our HTDMA. We found that maximumly 15% of the volume of the selected particles evaporated, leading to less than 5% deviation in the measured HGF, which is still within the measurement uncertainties. Therefore, we believe that the influence of the losses of particulate semi-volatile compounds on the derived hygroscopicity of newly formed particles may not be significant and our main findings that the compounds contributed to nanoparticle growth at current site were not very hygroscopic still hold.

In this study we observed clear differences in the inferred κ of growing material for the same size intervals between the NPF events in November and December. Specifically, the growing material was more hygroscopic on 18 November, while for the two events in December, the material or compounds that contributed to nanoparticle growth were basically hydrophobic, with κ values of around 0. This demonstrates that at the same region, the compounds driving particle growth may be different. Nevertheless, these values were still much lower than those determined in Hyytiälä, with κ values of the growing material around 0.2. This is most probably associated with the remarkedly difference in atmospheric precursors, producing condensable compounds that varied largely between biogenic environments, such as Hyytiälä, and anthropogenic ones, such as ours. However, Liu et al. (2021) found that newly formed material responsible for particle growth to be very hygroscopic, with mean κ of 0.28, at another observational site in the NCP with potentially similar anthropogenic influences, that is, urban Beijing. They further suggested that these compounds were more likely to be organic acids, carboxylic organic amine and ammonia organic salts due to their similar water uptake ability. These compounds were not able to explain nanoparticle growth in our study.

Previous studies (Tröstl et al., 2016; Wang et al., 2020) concluded that compounds with low volatility dominantly drove nanoparticle growth, which have either large molecular weight or added functional groups. However, compounds with more functional groups were typically quite hygroscopic and their water uptake ability increased with increasing functionalities (Han et al., 2022). Therefore, these compounds with higher functionalities were not considered candidates for nanoparticle growth in this study. Owing to the low water uptake ability of the growing material, we speculate that these compounds might be organic compounds with large molecule weight, such as organic polymers or organosulfates (De Haan et al., 2019; Hansen et al., 2015; Petters et al., 2006; Rahman & Al-Abadleh, 2018). Indeed, direct molecular speciation is needed in the future to draw further conclusion regarding their specific chemical information.

3.3. Possible Roles of Precursors and Meteorological Conditions in Nanoparticle Growth

The contrasting κ of growing material between our GC site and Hyytiälä and between the three different NPF events of current study demonstrates that the condensable vapors contributing to nanoparticle growth may vary significantly between events. This may result in different chemical composition of particles and their corresponding hygroscopicity. One of the most plausible explanations for the contrast could arise from the distinct emissions of precursors during individual cases. Volatile organic compounds (VOCs), and in particular, highly oxygenated organic molecules (HOM), are considered one of the most important precursors to form condensable vapors that strongly contribute to nanoparticle growth (Ehn et al., 2014). These precursors mainly originated from biogenic sources at Hyytiälä, such as monoterpenes and isoprene. At our GC site, these precursors were dominated by anthropogenic ones, including benzene, toluene, naphthalene and ketones (Figure 3). Previous studies reported that oxidation products of anthropogenic VOCs are typically larger molecules or less oxidized (Garmash et al., 2020; Nie et al., 2022) and thus have quite low hygroscopicity compared to the ones for biogenic VOCs, which are normally more hygroscopic with κ values approaching to 0.2 (Kawana et al., 2017; Varutbangkul et al., 2006). This may possibly explain the much lower hygroscopicity of the growing material for current study in contrary to that of Hyytiälä.

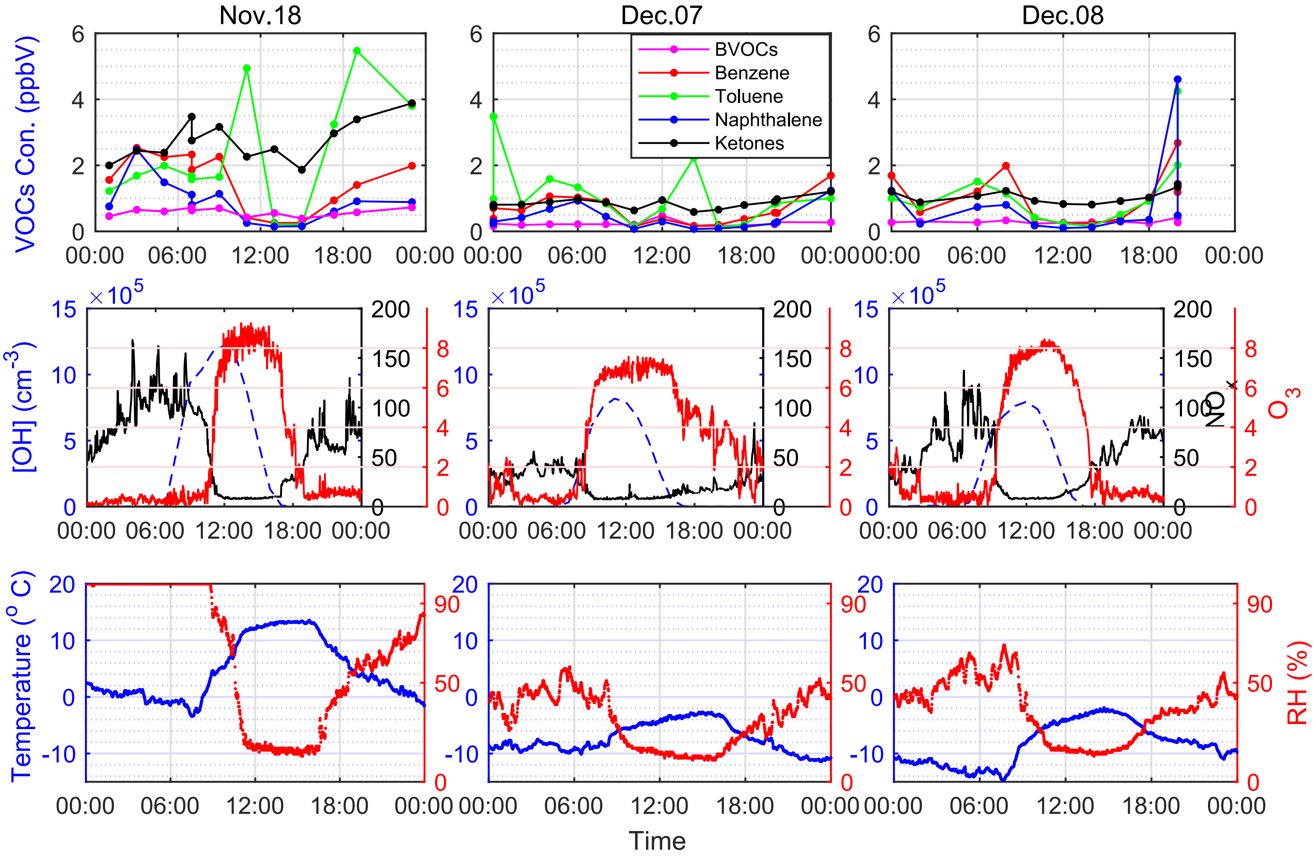

Figure 3. Diurnal variations of the concentration of typical VOCs, atmospheric oxidants, including OH radical, NO_x and ozone, relevant for NPF and subsequent growth and ambient temperature as well as RH during these three identified NPF days.

Moreover, a clear difference in the atmospheric level of these typical VOCs during the three event days of current study was observed. To be specific, on 18 November the concentration of these VOCs was significantly higher by a factor of 2–3 than that during 7–8 in December (see Figure 3, Figure S2 and Table S1 in Supporting Information S1). This change was more pronounced for toluene and ketones, specifically during late afternoon when the particle growth process was coincidentally observed. Associated with the content of precursors in the atmosphere, atmospheric oxidant concentration also affects the formation of HOMs and their temporal variation. We observed that the concentration of the calculated OH radical was about two times higher during 18 November than that on the other 2 days in December. A flow-tube study by Garmash et al. (2020) concluded that the precursor and OH concentrations had a strong impact on the formed HOMs composition as well as their yields. At higher OH concentrations, less dimers compared to monomers were identified from the HOMs spectrum during their flow-tube measurements. Dimers, typically having larger molecular weights, were expected to be less hygroscopic than monomers, which were relatively smaller molecules. This could be a potential reason for the higher hygroscopicity of growing material on 18 November due to the higher OH radical level in the atmosphere compared to the other 2 days in December. In addition to the OH concentration (see Figure S3 in Supporting Information S1), there were slight differences in the ambient concentration of other two oxidants, that is, NO_x and O_3. On 7 December, the concentration of NO_x was the lowest. Moreover, the concentration of O_3 on this day decreased slowly in the afternoon after reaching its maximum during noon time, which was in contrast to that of the other 2 days with a sharp drop in atmospheric O_3 level around 18:00 before evening. The differences in the ambient content of NO_x or O_3 did not exert apparent influence on the variation of the κ of growing material between different NPF days. This may indicate that at the GC site OH oxidation appeared to play a more important role in the formation of HOMs or other condensable vapors, which has been previously confirmed by other study that OH-initiated reactions dominated in the photooxidation of aromatics (Ji et al., 2017).

Besides the differences in available precursors and atmospheric oxidants, the meteorological parameters, such as temperature, relative humidity (RH), also varied between these 3 days. We observed higher temperature and relative humidity on 18 November, as shown in Figure 3. Simon et al. (2020) concluded that the HOM yields decreased with decreasing temperature based on their dedicated experiments conducted at the Cosmics Leaving OUtdoor Droplets (CLOUD) chamber. However, the volatility distribution of the formed HOMs could also be altered due to the reduction in their saturation vapor pressures at lower temperature. Nie et al. (2022) found that the volatility distribution of the produced HOMs shifted to a lower one from HongKong to Beijing owing to the substantial differences in ambient temperature between the two sites. The compounds that preferred to stay in the gaseous phase, such as semi-volatile organics (SVOCs), tended to partition into particle phase and may contribute more to the growth at colder environments. When newly formed particles contain more SVOCs due to the decreased ambient temperature, such as the two NPF days in December, aerosols would become less hygroscopic as these semi-volatile compounds were normally less-oxidized and thus having lower water uptake ability compared to low volatility compounds (Goulden et al., 2018).

A previous study found that the newly formed HOM species and their abundance did not change significantly at different RH conditions during their flow-tube measurements (Li et al., 2019). However, RH may exert a key role in other atmospheric processes, such as, heterogeneous reactions (Liu et al., 2018), which are believed to be another important mechanisms responsible for nanoparticle growth (Wang et al., 2010). At a more humid environment, newly formed particles may adsorb more water vapor on their surface, facilitating the partition or condensation of water-soluble gases. This may in turn promote nanoparticle growth and increase their corresponding hygroscopicity. The study by Kuang et al. (2020), which was part of our campaign, suggested that photochemical aqueous-phase reactions dominantly contributed to the SOA formation under high RH at our GC site. Though their results mainly focused on bulk aerosols rather than newly formed particles, it indirectly implies that heterogeneous reactions may help the nanoparticle growth and their hygroscopicity at GC as atmospheric chemistry might be approximate under similar conditions.

Overall, the differences in the concentration of precursors and atmospheric oxidants as well as meteorological factors, may synergistically govern the formation of condensable vapors, their condensation as well as other processes, such as heterogeneous reactions. This may together influence nanoparticle growth process and ultimately determine the chemical composition of newly formed particles and their water uptake abilities.

4. Discussions

Direct approaches (Bzdek et al., 2012; Vakkari et al., 2015; Zhang et al., 2011) had also been utilized over years to investigate the composition of the compounds responsible for particle growth, typically by an aerosol mass spectrometer (AMS) or an aerosol chemical speciation monitor (ACSM). However, as such instruments cannot detect particles smaller than 60 nm, the ones at or above 60 nm was sampled to typify newly formed particles. Or alternatively, the composition of particles at a certain size range, for example, 30 nm $< D_p <$ 60 nm, were integrated in order to obtain good signal level. Though these approaches provided quantitative estimates on how much of different compounds contributed nanoparticle growth, there still remained large gap as these detected particles may not well represent newly formed particles at different growth stages.

Hygroscopicity measurements of particles, being capable of covering a wide size range, from as small as 10 nm to a few hundred nm, provided direct observation of the hygroscopic properties of newly formed particles at different growing stages. As this approach depicte the evolution of the chemical composition of newly formed particles and more importantly, their growing material, it directly informed of any possible variations of the atmospheric chemistry driving nanoparticle growth and may hint the involvement of other atmospheric processes, such as heterogeneous reactions. For instance, at our GC site, the hygroscopicity of the growing material seemed to remain constant for particles at 20 nm until they grew to 40 nm (see Figure 2a). This suggested a very similar atmospheric chemistry throughout the growth of each event. However, temporal variation in the growing material may occur in other environments, indicating the alteration in atmospheric chemistry or the involvement of other processes during nanoparticle growth. Coupling with other relevant measurements, such as molecular identification in the gas phase, evidences proving other underlying mechanisms behind the growth can be explored, deepening our understanding of this phenomena. Moreover, quantitative analysis of the hygroscopicity for the growing material directly tells which group of candidates, for instance, hydrophobic or hydrophilic compounds, contribute to particle growth, which normally cannot be further differentiated by typical aerosol chemical composition measurements. This reduces the gap in the interpretation between gaseous and particle-phase measurements by narrowing down the candidates of potentially contributing species and may provide more accurate estimation of the pathways for this process.

Data Availability Statement

The measurement data of particle number size distribution, aerosol hygroscopicity, the concentration of the five typical VOCs, atmospheric oxidants, ambient temperature and RH as well as the calculated hygroscopicity of the growing material used in this manuscript is available at Hong (2023). The measurement data of aerosol hygroscopicity at other different environments, including the boreal forest in Hyytiälä of Finland, CLOUD chamber studies, urban Beijing as well as the mixed deciduous forest in central Virginia, US is available at Ehn et al. (2007), Kim et al. (2016), Liu et al. (2021), O'Halloran et al. (2009), respectively.

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