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Impacts of the COVID-19 lockdown in China on new particle formation and particle number size distribution in three regional background sites in Asian continental outflow

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HIGHLIGHTS

• Particle number concentrations generally decreased during the lockdown period.
• New particle formation events were distinguished using an automatic method.
• Frequency of new particle formation reduced by 1%–7% during the lockdown period.
• H2SO4 proxy decreased in Anmyeon and Bongseong during the lockdown period.

ABSTRACT

Despite the curtailment of atmospheric condensing precursor gases during the Coronavirus disease 2019 (COVID-19) lockdown (LD) period, unexpected haze events via the formation of new particles and their subsequent growth have been identified. This study investigated the impact of emission reduction during the Chinese LD period on the new particle formation (NPF) frequency and corresponding particle number size distribution (PNSD) at three regional background atmospheric monitoring sites in the western coastal areas of the Korean Peninsula. During this duration, the number concentrations of the nucleation- (<25 nm) and accumulation-mode (>90 nm) particles significantly decreased in Baengryeong (BRY), showing decreases of 34% and 29%, respectively. Unlike BRY, the PNSD in Anmyeon (AMY), which is influenced by nearby industrial emissions, remained nearly unchanged during the LD period, possibly because the reduction in industrial emissions was not significant during the social distancing period enforced by Korea. Bongseong (BOS) showed a similar variation to that of BRY; however, the magnitude of the reduction was weaker because of its higher altitude compared to other sites. The cyclostationary empirical orthogonal function technique was applied to the measured PNSDs at the three sites to objectively classify NPF events. Because mode 1 of cyclostationary loading vectors commonly represented the typical diurnal variation of PNSD during regional NPF events at three sites, mode 1 of the corresponding principal component time series was used for NPF classification. The NPF frequency decreased by 7%, 1%, and 7% in BRY, AMY, and BOS, respectively, despite favorable meteorological conditions, such as increased temperature and insolation during the LD period. The diurnal variation in the sulfuric acid (H2SO4) proxy implied that the H2SO4 proxy acted as a determining factor for NPF events during the NPF occurrence time (8–12 local hours) in AMY and BOS; however, NPF occurrence in BRY was not connected to the H2SO4 proxy level.
1. Introduction

The Coronavirus disease 2019 (COVID-19) pandemic forced many countries to regulate anthropogenic activities in diverse fields, such as industrial processes and on-road mobile sources (Shrestha et al., 2020; Venter et al., 2020). China executed intensive lockdown (LD) measures against the spread of COVID-19 in 95 cities starting in late January 2020 (He et al., 2020). During the Chinese COVID-19 LD period, the concentration of ambient air pollutants showed significant national variation compared to previous years. For example, overall reductions in principal anthropogenic air pollutants, such as PM$_{2.5}$, NO$_2$, CO, and SO$_2$, were observed at many monitoring stations operated by the China National Environmental Monitoring Center (Shi and Brasseur, 2020). Bao and Zhang (2020) reported that the air quality index and the concentrations of five air pollutants (SO$_2$, PM$_{2.5}$, PM$_{10}$, NO$_2$, and CO) were significantly reduced in 44 cities in northern China because of the LD measures. Particularly, the severe reduction in NO$_2$ can be connected to the weakened titration effect of O$_3$ by NO, resulting in an increase in the O$_3$ concentration in many populated regions in China during the COVID-19 LD period (Wang et al., 2020; Meng et al., 2021). The impact of NO$_2$ reduction can be mitigated in rural regions by reinforcing the contribution of volatile organic compounds (VOCs) to the decrease in O$_3$ (Liu et al., 2021).

Even with the reduced emission of major gaseous air pollutants, unexpected haze events occurred owing to the efficient secondary formation of aerosols. Chang et al. (2020) analyzed the severe haze events induced by nitrogen chemistry and long-range transport during the COVID-19 LD period. Furthermore, several studies noted that the intensity of secondary pollution was nearly unchanged or even strengthened by the enhanced ambient oxidation capacity represented by increased O$_3$ concentrations, despite the primary emission of gaseous precursors during the COVID-19 LD period (Huang et al., 2021; Meng et al., 2021; Shen et al., 2021). In addition to anthropogenic emissions, changes in meteorological parameters during the lockdown period can also contribute to the properties of new particle formation (NPF; Bousiotis et al., 2021). Compared to the concentration of primarily emitted pollutants, NPF properties can be more complexly affected by emission curtailment during the lockdown according to changes in atmospheric chemistry or regional-scale meteorology (Kim et al., 2016).

The Korean Peninsula is located downwind of the Asian continent and can be influenced by changes in the emission of atmospheric gaseous and particulate pollutants during the LD of continental megacities (Kim et al., 2007; Lee et al., 2019). Moreover, the Korean government also attempted to manage the COVID-19 spread by enforcing social distancing from February 29, 2020 (Kwak et al., 2021). During the social distancing period, the mean concentrations of gaseous and particulate pollutants over the Korean Peninsula showed a significant decrease from February 29, 2020 (Kwak et al., 2021). During the Chinese COVID-19 LD period, the concentration of ambient air pollutants showed significant national variation compared to previous years. For example, overall reductions in principal anthropogenic air pollutants, such as PM$_{2.5}$, NO$_2$, CO, and SO$_2$, were observed at many monitoring stations operated by the China National Environmental Monitoring Center (Shi and Brasseur, 2020). Bao and Zhang (2020) reported that the air quality index and the concentrations of five air pollutants (SO$_2$, PM$_{2.5}$, PM$_{10}$, NO$_2$, and CO) were significantly reduced in 44 cities in northern China because of the LD measures. Particularly, the severe reduction in NO$_2$ can be connected to the weakened titration effect of O$_3$ by NO, resulting in an increase in the O$_3$ concentration in many populated regions in China during the COVID-19 LD period (Wang et al., 2020; Meng et al., 2021). The impact of NO$_2$ reduction can be mitigated in rural regions by reinforcing the contribution of volatile organic compounds (VOCs) to the decrease in O$_3$ (Liu et al., 2021).

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The PNSD was measured at three regional background sites (Baengryeong, Anmyeon, and Jeju) located on the west coast of the Korean Peninsula using a scanning mobility particle sizer (SMPS) from 2013 to 2020. The western regions of the Korean Peninsula are highly influenced by transported gaseous pollutants or particulate matter emitted from the Asian continent, especially in the winter and spring seasons (Kim et al., 2007; Park et al., 2021a).

The Baengryeong (BRY) comprehensive monitoring observatory (37.57°N, 124.37°E, 135 m above sea level (a.s.l.); Lim et al., 2018) is located around the northwestern tip of South Korea. BRY is a clean remote station with a small amount of local pollutant emissions; however, it can be affected by transboundary-transported pollutants under favorable meteorological conditions (Shim et al., 2022). The Korean Global Atmosphere Watch (GAW) Center in Anmyeon (AMY; 36.32°N, 126.19°E, 46 m a.s.l.) is located on the central west coast of South Korea (Lee et al., 2008; Kim et al., 2016). Although both stations have relatively low local air pollutant emissions compared to urban regions, the AMY site can be affected by adjacent industrial emissions (e.g., toluene) according to changes in atmospheric circulation (Simpson et al., 2020). The PNSD observed at the Bongseong (BOS) comprehensive monitoring observatory on Jeju Island (33.35°N, 126.39°E, 558 m a.s.l.; Fang et al., 2017) was also analyzed. While there are few emission sources near the BOS, the concentration of air pollutants at the BOS site can be influenced by daytime marine boundary layer development because of its high altitude.

SMPS produces the PNSD by estimating the particle number densities according to the diameters in 54 bins from 10.4 to 469.8 nm (Kim et al., 2021). The SMPS neutralizes the particles and assigns a known distribution of positive charges to aerosols. Aerosols are differentiated at the rod, namely the differential mobility size analyzer, according to their electrical mobility. Particle number densities were calculated at the condensation particle counter by growing particles to a measurable size using butanol (Hogrefe et al., 2006).

2. Materials and methods

2.1. Sites and measurements

The PNSD was measured at three regional background sites (Baengryeong, Anmyeon, and Jeju) located on the west coast of the Korean Peninsula using a scanning mobility particle sizer (SMPS) from 2013 to 2020. The western regions of the Korean Peninsula are highly influenced by transported gaseous pollutants or particulate matter emitted from the Asian continent, especially in the winter and spring seasons (Kim et al., 2007; Park et al., 2021a).

The cyclostationary empirical orthogonal function (CSEOF) technique

The cyclostationary empirical orthogonal function (CSEOF) technique was applied to the PNSD data observed at the three background stations. The CSEOF technique is an extended version of empirical orthogonal function analysis (Kim et al., 1996). In the CSEOF technique, the PNSD hourly data (T) are decomposed into different modes of CSEOF loading vectors (CSLVs) multiplied with principal component (PC) time series as follows (Kim et al., 2013):

\[ T(r,t) = \sum_n \text{CSLV}_n(r,t) \text{PC}_n(t) \]  

where \( n \) is the number of CSEOF modes. In this study, both PNSD and CSLVs are the function of the size bins \( r \) and time \( t \). Also, the CSLVs are periodic in the nested period \( d \), \( \text{CSLV}_n(r,t + d) = \text{CSLV}_n(r,t) \). Because NPF typically occurs during the daytime for a period of approximately one day, the nested period of the CSEOF analysis was set to 24 h. Accordingly, CSLVs display the temporal variation in daily PNSD, and PC time series denotes the amplitude of the pattern displayed by CSLVs (Kim et al., 2021; Park et al., 2021b).
3. Results and discussion

3.1. Characteristics of PNSD during the COVID-19 lockdown period in three background sites

The PNSD at the three background sites can be influenced not only by emissions from upwind continental regions but also by synoptic meteorological conditions in East Asia. Fig. 1 shows the spatial distribution of aerosol optical depth (AOD) and 850-hPa wind field during 2013–2019 and 2020. AOD is the monthly mean product estimated from Moderate Resolution Imaging Spectroradiometer (MODIS) Aqua platform, and wind vector was derived from NCEP/NCAR reanalysis 4-times daily data. Three sites located west of the Korean Peninsula were commonly affected by the long-range transport of aerosols from source regions in the Asian continent in 2013–2019 and 2020. The overall wind field during 2013–2019 was similar to that during 2020, except for the increased impact from the northerly wind in April. AOD levels in the source regions of the Asian continent from February to March generally decreased in 2020, implying a reduced transport of air pollutants toward the three sites (Zhang et al., 2021). In March 2020, the concentrations of air pollutants (e.g., PM$_{2.5}$, PM$_{10}$, NO$_2$, and CO) in the Korean Peninsula also distinctly decreased in 2020, implying a reduced transport of air pollutants toward the three sites (Zhang et al., 2021). In March 2020, the concentrations of air pollutants (e.g., PM$_{2.5}$, PM$_{10}$, NO$_2$, and CO) in the Korean Peninsula also distinctly decreased in accordance with the strengthened social distancing policy of the South Korean government (Ju et al., 2021). In other words, the impact of the COVID-19 outbreak LD was prominent from February to March 2020. Because the Chinese LD was enforced from almost the end of January and the concentrations of pollutants (e.g., PM, O$_3$, NO$_2$, SO$_2$) rebounded in April (Fan et al., 2021), the meaningful COVID-19 LD period was set from February to March in this study.

BRY showed a distinct decrease in the particle number concentration in both the nucleation and accumulation modes during the LD period. Although the PNSD according to diameter was not nearly unchanged, the number densities of aerosols decreased in all diameter ranges (Fig. 2b). Among them, the number concentration of accumulation-mode particles (>90 nm; Kulmala et al., 2004) in the measurable size range decreased more (~34%) than that of nucleation-mode particles (<25 nm; Dal Maso et al., 2005; ~29%). The overall number concentration level in AMY during the Pre-LD period was the highest among the three regional background stations. In AMY, there was a negligible change in the number densities of aerosols, except for a slight decrease in the accumulation-mode particle number concentration in the afternoon. This was possibly due to the impact of precursor gases, including VOC emissions from industrial regions in the vicinity of the AMY site. Simpson et al. (2020) captured not only abundant VOCs such as toluene and benzene, but also significant formaldehyde, which is a product formed via photolysis of VOCs, at the Daesan petrochemical complex near the AMY station based on observations from the Korea-United States Air Quality Study (KORUS-AQ) campaign in 2016. Furthermore, the decrease in PM$_{2.5}$ concentration in the cities in South Korea during the social distancing led by the South Korean government (from February 29, 2020) was positively correlated with the PM$_{2.5}$ emission fraction for mobile sources but negatively correlated with the combustion and industrial sources (Kwak et al., 2021). From these reports, we can deduce that the artificial reduction in industrial emissions was not significant during the period of social distancing in South Korea. The diurnal pattern of the PNSD at the BOS site had the characteristics of a high-altitude station. The number densities of particles were significantly lower than those at the BRY site during the nighttime because of the decay of the nocturnal marine boundary layer. The long-lasting enhancement of nucleation-mode particles (~16 local standard time (LST)) at the BOS site compared to that at the other sites was likely influenced by the transportation of gaseous precursors or nanoparticles following the daytime valley wind until the afternoon (Bei et al., 2018; Park et al., 2018). Furthermore, the impact of emission changes from the source regions in the Asian continent can be

Fig. 1. 850-hPa wind vector (arrow) from January to May during the period of 2013–2019 and 2020. The spatial distribution of AOD is represented as a colour scale. Green, blue, and black circles denote the location of BRY, AMY, and BOS sites, respectively.
Fig. 2. (a) Diurnal variation in mean PNSD and its standard deviation in three sites during the Pre-LD (left) and LD (right) periods. (b) Mean size distribution from 10 to 469.8 nm in the Pre-LD (green) and LD (red) periods.
weaker at the BOS because of its higher altitude. The PNSD in the BOS site over the LD period also experienced a decrease in number concentration in both the nucleation and accumulation modes; however, its magnitude (11% and 24% in the nucleation and accumulation modes, respectively) was lower than that of the BRY site.

3.2. Identification of NPF events

The CSEOF technique was applied to the daytime (7–16 LST) PNSD dataset measured at the three sites during the study period (Kim et al., 2013; Kim et al., 2021; Park et al., 2021a). All mode 1 CSLVs corresponded to typical NPF and subsequent growth, accounting for 34%, 27%, and 35% of the total variance of the PNSD measured at the BRY, AMY, and BOS sites, respectively (Fig. 3a). Particularly, mode 1 CSLVs at the BOS site also included a positive anomaly in the number concentration of accumulation-mode particles. Unlike the other two sea level sites, the daytime CSLV mode 1 at the BOS site can show a significant positive anomaly in the accumulation-mode as well as nucleation-mode size range under cloud-free clear weather conditions because it is a favorable condition for NPF as well as the development of the marine boundary layer (Hamed et al., 2007) or daytime valley breeze (Lehner et al., 2019). Specifically, when NPF occurs under the clear weather condition, not only the development of a marine boundary layer due to the intense solar heating can expand the well-mixed region up to the altitude of BOS, but also an active valley breeze can bring the aerosols up to the BOS as well.

Fig. 3. (a) Cyclostationary loading vector of first CSEOF mode (Mode 1 CSLVs) calculated from the PNSD obtained at the BRY, AMY, and BOS sites. (b) Cumulative frequency for the daytime (7–16 LST) CSEOF mode 1 PC amplitude for February–March during the study period (2013–2020). The zero line and a threshold value for distinguishing weak- and strong-NPFs are represented as solid red and dashed lines, respectively.
Since mode 1 CSLVs corresponded to the typical diurnal patterns during regional NPF events (Kerminen et al., 2018), NPF events were identified based on the CSEOF mode 1 PC amplitudes. NPF events were identified using the daytime PC amplitudes for February–March during the entire study period from 2013 to 2020. If the PC amplitudes were lower than zero, the day was classified as a non-NPF event. Moreover, strong- and weak-NPF events were distinguished based on the threshold value calculated by averaging the positive daytime PC amplitudes for each site (Fig. 3b; Kim et al., 2013).

An average PNSD profile of strong-, weak-, and non-NPF for the entire study period is presented in Fig. 4. The diurnal variation in PNSD during the strong-NPF events showed a distinct so-called ‘banana-plot’ at all sites. The clear banana shape on the plot of PNSD with time shows the features of regional NPF, which is distinguished from sub-regional NPF events that capture only a partial banana shape on the PNSD plot (Kerminen et al., 2018). During weak-NPF events, a relatively moderate increase in the number concentration of nucleation-mode aerosols was observed near noon. The number of aerosols in the growing mode was also significantly lower than that during strong-NPF events. Meanwhile, the enhancement of the nucleation-mode particles was nearly negligible during non-NPF events.

The CSEOF-based classification used in this study captured typical PNSD characteristics on NPF days. Fig. S1 shows an example of the classification result in February 2017 in BRY, where five days (February 1, 6, 11, 17, and 23) and three days (February 9, 20, and 21) were identified as strong-NPF and weak-NPF events, respectively. In particular, only February 9 and 11 were classified as weak- and strong-NPF events during 8–11 February. The CSEOF technique distinguished NPF days based on the objective numerical amplitude of diurnal PNSD evolution, even though this period was somewhat ambiguous in terms of NPF identification. The distinct increase in nucleation-mode particles near the lowest diameter (~10 nm) could be the most important for being classified as an NPF event based on the classification result. February 8 and 10 could be regarded as days influenced by the transport of slightly aged newly-formed fresh particles (~20–30 nm) from the vicinity of the station. The identification of NPF events using the CSEOF technique also has been validated in the previous study (Kim et al., 2013).

3.3. NPF events at three background sites during the COVID-19 LD period

The change in NPF events during the COVID-19 LD period was investigated in terms of frequency based on the previously shown NPF classification method. The frequencies of NPF (i.e., the sum of strong- and weak-NPF events) in BRY, AMY, and BOS during the Pre-LD period were 40%, 37%, and 46%, respectively (Fig. 5). The NPF frequencies estimated at the sea-level sites (BRY (40%) and AMY (37%)) in this study were lower than those determined by applying the method suggested by Dal Maso et al. (2005) to the PNSD observed at a rural background site located downwind of the Seoul metropolitan area during the spring season (52%; Lee et al., 2021). However, our results were closer to those estimated by applying the objective numerical value to the number concentration of 20-nm aerosols for NPF classification, which reported approximately 32% for March–April in AMY (Matsui et al., 2013). As BOS can be frequently located above the shallow marine boundary layer during the night, it can be in a clean environment in the early morning without aged particles, which play a role as the sink of newly-formed fresh particles, resulting in a relatively higher NPF frequency compared to other sites. Several previous studies have also reported a substantial frequency of NPF events associated with upslope valley winds in mountainous locations (Kerminen et al., 2018). Higher frequencies were observed than those in BOS at the high-altitude sites, such as Chacaltaya, during the dry season (64%; Rose et al., 2015) and the Storm Peak Laboratory (52%; Hallar et al., 2011, 2016).

During the LD in 2020, NPF frequency decreased by 7%, 1%, and 7% in BRY, AMY, and BOS, respectively. As a strict classification of NPF, it is also worth examining the change in strong-NPF frequency during the LD period. The decrease in strong-NPF events mainly led to a decline in the entire NPF frequency, with 6% (BRY), 2% (AMY), and 5% (BOS) decreases. The proportion of strong-NPF events among all NPF events also significantly decreased at all sites during the LD period (48% to 38% in BRY, 34% to

![Fig. 4. Averaged diurnal variation in PNSD during the classified strong- (left), weak- (middle), and non- (right) NPFs at the three sites.](image-url)
Although H$_2$O can participate in the formation of initial clusters promoting the occurrence of NPF and subsequent growth (Debevec et al., 2019), biogenic aerosol precursor vapors can be more actively emitted into the atmosphere with increased ambient temperature, (Kerminen et al., 2018), and ambient temperature can promote or suppress the occurrence of NPF events during the LD period, showing unfavorable changes (i.e., an increase in RH or decrease in WS) and a favorable change (an increase in ambient temperature) for NPF occurrence. However, several previous studies reported that the negative impact of increased RH on NPF occurrence becomes small in the range of RH at these sites (61% in BRY and 67% in GSN during the Pre-LD period; Wu et al., 2019; Fig. 4 in Bousiotis et al., 2021). Furthermore, Pushpawela et al. (2019) reported that the impact of WS change on the NPF frequency decreased in rural regions owing to the low number of pre-existing particles ventilated by the wind. Meanwhile, variations in insolation during the LD period were not statistically significant at any of the three sites, suggesting that photochemical reactions for NPF occurrence were not meaningfully promoted or suppressed by the insolation change. In other words, all three sites showed decreased NPF frequency under the reduced emission of precursor gases from the source regions in the Asian continent during the LD period, despite favorable meteorological conditions for NPF occurrence mainly developing by increased temperature.

Considering the substantial contribution of inorganic components to the particulate matter in East Asia (Jordan et al., 2020), the production of gaseous SO$_2$ plays a dominant role in the formation of H$_2$SO$_4$ via photochemical reactions in the presence of ultraviolet (UV) radiation, the UV multiplied by the SO$_2$ mixing ratio can be used as a surrogate for the H$_2$SO$_4$ concentration when this value is considered together with the condensation sink (CS) (Petäjä et al., 2009; Dall’Osto et al., 2018). In this study, hourly maximum solar radiation in the UV-B range (W m$^{-2}$) was used, considering that, UV-B mainly contributes to the photolysis of ozone and is ultimately related to the formation of hydroxyl radicals (OH). The condensation sink ($\text{s}^{-1}$), which can be an indicator of the condensation rate of non-volatile gaseous compounds on pre-existing large particles, was obtained following the method introduced by Kulmala et al. (2001). In this study, SO$_2$ mixing ratio was measured in BRY, Pado-ri located close to the AMY station (~8 km away from AMY), and GSN. Additionally, UV-B measurements were conducted in Incheon (INC; 37.48°N, 126.6249°E), AMY, and GSN. Owing to the absence of UV-B measurements in BRY, data measured at INC from February–March of 2018 (4272 hourly data) were used to represent the H$_2$SO$_4$ proxy in BRY despite the substantial distance, considering the relatively homogeneous spatial distribution compared to the properties of gaseous pollutants. Except for BRY, UV-B measured from February–March 2013 (11376 hourly data) was analyzed. SO$_2$ mixing ratio and UV-B intensity (in parenthesis) for the study period were 1.6 ± 0.9 ppb (0.0496 ± 0.0376 W m$^{-2}$), 2.2 ± 1.5 ppb (0.0449 ±

24% in AMY, and 31% to 24% in BOS). The less frequent occurrence of NPF events during the LD period is discussed later in this section.

Because meteorological conditions are known to significantly influence the occurrence of NPF events (Bousiotis et al., 2021), changes in meteorological conditions were also investigated during the NPF occurrence time (8–12 LST; Tang et al., 2021) of the LD period (Table 1) using data from the Automated Synoptic Observing System produced by the Korea Meteorological Administration. Although diverse physical processes associated with ambient temperature can promote or suppress the occurrence of NPF (Kerminen et al., 2018), biogenic aerosol precursor vapors can be more actively emitted into the atmosphere with increased ambient temperature, promoting the occurrence of NPF and subsequent growth (Debevec et al., 2018). Although H$_2$O can participate in the formation of initial clusters via binary or ternary nucleation (Yao et al., 2018), increased relative humidity (RH) reduces the probability of fresh particles surviving by producing a larger aerosol surface area (Li et al., 2019; Wu et al., 2019). A higher wind speed (WS) can create favorable conditions for the occurrence of NPF events by eliminating pre-existing particles (Pushpawela et al., 2019). Increased solar radiation can promote photochemical gas-phase reactions and ultimately contribute to the production of condensable vapors in NPF (Boy and Kulmala, 2002).

Because of the absence of solar radiation measurements in AMY and BOS, meteorological data measured in Seosan (SS; 36.78°N, 126.49°E) and Gosan (GSN; 33.29°N, 126.16°E) were used, respectively. In addition, it is worth noting that insolation measurement in BRY began in 2019. Generally, meteorological changes appeared at all three stations during the LD period, showing unfavorable changes (i.e., an increase in RH or decrease in WS) and a favorable change (an increase in ambient temperature) for NPF occurrence. However, several previous studies reported that the negative impact of increased RH on NPF occurrence becomes small in the range of RH at these sites (61% in BRY and 67% in GSN during the Pre-LD period; Wu et al., 2019; Fig. 4 in Bousiotis et al., 2021). Furthermore, Pushpawela et al. (2019) reported that the impact of WS change on the NPF frequency decreased in rural regions owing to the low number of pre-existing particles ventilated by the wind. Meanwhile, variations in insolation during the LD period were not statistically significant at any of the three sites, suggesting that photochemical reactions for NPF occurrence were not meaningfully promoted or suppressed by the insolation change. In other words, all three sites showed decreased NPF frequency under the reduced emission of precursor gases from the source regions in the Asian continent during the LD period, despite favorable meteorological conditions for NPF occurrence mainly developing by increased temperature.

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![Figure 5](image.png)

**Table 1** Variations in NPF occurrence time (8–12 LST) surface meteorological factors during the LD period at three sites. Values in the parenthesis represent the difference in each meteorological factor between the LD and pre-LD periods. When the difference's p-value is <0.05 (statistically significant difference), the impact of meteorological variation on NPF occurrence is reflected in the background colour in each compartment in the table.

|                   | BRY          | AMY (SS)     | BOS (GSN)    |
|-------------------|--------------|--------------|--------------|
| Temperature (°C)  | ↑ (+4.85)    | ↑ (+3.39)    | ↑ (+3.39)    |
| Wind Speed (m s$^{-1}$) | - (-0.09)   | ↓ (-0.29)    | ↓ (-0.76)    |
| RH (%)            | ↑ (+12.95)   | - (-2.35)    | ↑ (+6.41)    |
| Insolation (MJ m$^{-2}$) | - (-0.14)   | - (+0.08)    | - (-0.14)    |
0.0398 W m$^{-2}$), 0.7 ± 0.7 ppb (0.0496 ± 0.0461 W m$^{-2}$) in BRY, AMY, and BOS, respectively. Generally, UV-B intensity during the NPF events was significantly lower than that during the non-NPF events (p ≪ 0.05) at all three sites, but it was not the case for the SO$_2$ mixing ratio in the low-level stations (BRY and AMY). According to the brief sensitivity test for the NPF frequency with the increasing lower limit of the UV-B intensity from 0 to 0.15 W m$^{-2}$, NPF frequency increased from 36% to 50% in BRY, 37% to 49% in AMY, and 45% to 54% in BOS (Fig. S2), suggesting UV-B intensity is closely connected to the occurrence of NPF.

Fig. 6 shows the diurnal variation in the H$_2$SO$_4$ proxy ([UV-B radiation intensity] × [SO$_2$ mixing ratio]; ppb W m$^{-2}$) and CS during the NPF and non-NPF events over the Pre-LD and LD, respectively. The H$_2$SO$_4$ proxy was generally the highest near noon at all the three sites, implying a more significant contribution of inorganic compounds to the formation of new particles over time during the NPF occurrence time (8–12 LST). The H$_2$SO$_4$ proxy estimated at the three sites during the NPF occurrence time significantly decreased over the LD period. Under these circumstances, the H$_2$SO$_4$ proxy during 8–12 LST was higher during the NPF events compared to non-NPF events over both the Pre-LD and LD at the AMY and BOS sites. In particular, the H$_2$SO$_4$ proxy during 8–12 LST was higher during the NPF events compared to non-NPF events at the AMY and BOS sites. In particular, the H$_2$SO$_4$ proxy during the NPF occurrence time was estimated to be the lowest in BOS, showing only 7.546 ± 9.870 ppb W m$^{-2}$. Thus, the decrease in NPF frequency in BOS and AMY can be attributed to the reduced H$_2$SO$_4$ proxy during the LD period. However, BRY did not show a statistically significant difference in the H$_2$SO$_4$ proxy between the NPF and non-NPF events over both the Pre-LD and LD periods (p ≈ 0.12, pre-LD; p ≈ 0.60, LD; Fig. 6a). This indicates that the H$_2$SO$_4$ proxy can be saturated for the occurrence of NPF and is not a determining factor for NPF. The H$_2$SO$_4$ proxy relative to CS during the NPF occurrence time was the highest in BRY (13.684 ± 13.715 ppb W m$^{-2}$), followed by AMY (11.789 ± 16.657 ppb W m$^{-2}$).

The BRY region can be more susceptible to being influenced by the emission changes in hotspot regions in the Asian continent because of its lower altitude and the absence of local emissions compared to other sites. Although the altitude of AMY is lower than that of BRY, they are typically located in a vertically well-mixed marine boundary layer during February–March (Takegawa et al., 2020). Several works have reported that the emission changes in organic gaseous precursors for NPF are also significant. For example, Shen et al. (2021) reported that most VOCs measured in urban Beijing decreased during the Chinese LD period, except for benzene. Although the formation efficiencies of secondary aerosols increased during the LD period under the enhanced atmospheric oxidation capacity, a reduction in the mass concentration of organic aerosol factors during the LD period was also captured by Tian et al. (2021). The decrease in the NPF frequency estimated in BRY can be highly influenced by the

![Fig. 6](image-url)

Fig. 6. (a) Diurnal variation in H$_2$SO$_4$ proxy and (b) condensation sink (CS) at the BRY, AMY, and BOS sites during the Pre-LD (green) and LD (red) periods. Diurnal variation during the NPF (Strong- and Weak-NPF) events and non-NPF events are represented as solid and dashed lines, respectively. NPF occurrence time (8–12 LST) is the shaded area.
4. Conclusions

In this study, we analyzed the impact of the COVID-19 LD in Asian continental cities on the NPF frequency and corresponding PNSD at three regional background sites located west of the Korean Peninsula. The key findings are summarized as follows:

- Although the number concentrations of nucleation- and accumulation-mode particles decreased during the LD period (February to March 2020) in BRY, the PNSD of AMY was nearly unchanged under the impact of local emissions. The BOS experienced similar variations in the PNSD to those of BRY; however, the magnitude was smaller, showing a decrease of 11% in the nucleation-mode and 24% in the accumulation-mode.
- The CSEOF technique-based classification of NPF events showed that NPF frequency decreased at all sites, despite the generally more favorable meteorological conditions for the occurrence of NPF during the LD period. The H$_2$SO$_4$ proxy during 8-12 LST was a limiting factor for the occurrence of NPF in AMY and BOS during both the Pre-LD (February to March of 2013–2019) and LD periods; however, the difference between NPF and non-NPF events was not statistically significant in BRY. BRY showed the highest H$_2$SO$_4$ proxy relative to CS ([(H$_2$SO$_4$ proxy)/[CS]]) compared to the other sites, implying that the H$_2$SO$_4$ proxy could be saturated for determining NPF occurrence. The less frequent outbreak of NPF events in BRY during the LD period can be attributed to the influence of the reduced emission of organic vapors from the Asian continent. This is because BRY is more susceptible to emission changes from hotspot regions than other sites are.

Since the 2010s, anthropogenic pollutant emissions have steadily declined. For example, the decreasing trend of SO$_2$ and NO$_x$ emissions was identified, which was motivated by measures such as levying an environmental protection tax (Sun et al., 2018). Therefore, the sharp reduction in pollutant emissions due to LD measures provided an observation-based natural experiment for the investigation of the NPF properties in the future atmosphere under these trends of condensing vapors acting as precursors. Based on the results of this study, policy strategies would be more systematically established to prevent the future occurrence of puzzling haze events originating from NPF.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.159904.

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CRediT authorship contribution statement

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