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Effect of restricted emissions during COVID-19 on atmospheric aerosol chemistry in a Greater Cairo suburb: Characterization and enhancement of secondary inorganic aerosol production

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ABSTRACT

To prevent the rapid spreading of the COVID-19 pandemic, the Egyptian government had imposed partial lockdown restrictions which led emissions reduction. This served as ideal conditions for a natural experiment, for study the effect of partial lockdown on the atmospheric aerosol chemistry and the enhanced secondary inorganic aerosol production in a semi-desert climate area like Egypt. To achieve this objective, SO2, NO2, and PM2.5 and their chemical compositions were measured during the pre-COVID, COVID partial lockdown, and post-COVID periods in 2020 in a suburb of Greater Cairo, Egypt. Our results show that the SO2, NO2, PM2.5 and anthropogenic elements concentrations follow the pattern pre-COVID < post-COVID > COVID partial lockdown. SO2 and NO2 reductions were high compared with their secondary products during the COVID partial lockdown compared with pre-COVID. Although, PM2.5, anthropogenic elements, NO2, SO2, SO42−, NO3−, and NH4+ decreased by 39%, 38–55%, 38%, 32.9%, 9%, 14%, and 4.3%, respectively, during the COVID partial lockdown compared with pre-COVID, with the secondary inorganic ions (SO42−, NO3−, and NH4+) being the dominant components in PM2.5 during the COVID partial lockdown. Moreover, the enhancement of NO3− and SO42− formation during the COVID partial lockdown was high compared with pre-COVID. SO42− and NO3− formation enhancements were significantly positive correlated with PM2.5 concentration. Chemical forms of SO42− and NO3− were identified in PM2.5 based on their NH4+/SO42− molar ratio and correlation between NH4+ and both NO3− and SO42−. The particles during the COVID partial lockdown were more acidic than those in pre-COVID.

1. Introduction

Aerosol, especially particulate matter (PM) with an aerodynamic diameter of less than 2.5 μm (PM2.5), is considered one of the most important atmospheric pollutants since it has become a severe problem due to its adverse effects on the environment and human health (Tao et al., 2017; Cohen et al., 2017; Zhou et al., 2017; Wang et al., 2017; Wright et al., 2018; EEA, 2018; Othman et al., 2020). Natural and anthropogenic emissions as well as the atmospheric conversion of primary air pollutants into secondary particles are the main atmospheric PM sources (Byrd et al., 2010; Im et al., 2012; Police et al., 2016; Hassan and Khoder, 2017; Marchetti et al., 2019). Due to this diversity in PM sources, its size, shape, and chemical composition depend on their releasing sources and atmospheric chemical reactions, and they vary spatially and temporally (Ghio et al., 2012; Kim et al., 2018; Hao et al., 2019).

PM has diverse chemical and biological characteristics (Pope and Dockery, 2006; Naimabadi et al., 2016) since it contains a mixture of different hazardous pollutants like organic and inorganic chemical components (Di Vaio et al., 2016; Huang et al., 2018). This diversity in the chemical characterization of PM controls its toxicity (Atkinson et al., 2015). Prior studies (Dao et al., 2014; Kong et al., 2014; Cheng et al., 2016; Cao et al., 2013; Hassan et al., 2013; Hassan and Khoder, 2017; Luong et al., 2022) have shown that water-soluble inorganic ions (WSIs) were accounted for about one-third, or more, of the PM mass, and play an important role in increasing PM concentrations in the

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atmosphere. The characteristics of WSIs in PM associate with the types of pollution sources and meteorological status (Huang et al., 2016; Kawamura et al., 2010). Not only can WSIs provide information on the sources and meteorological status (Huang et al., 2016; Huang et al., 2021; Chen et al., 2020a,b; Zhang et al., 2021; Lv et al., 2020; Dutheil et al., 2020; Collivignarelli et al., 2020; Venter et al., 2020; Zhang et al., 2020; Muhammad et al., 2020; Dutheil et al., 2020; Collivignarelli et al., 2020; Venter et al., 2020; Parra and Espinoza, 2020; Zambrano-Monserrat and Ruano, 2020; Hashim et al., 2020; He et al., 2021a,b; He et al., 2021a,b). Although the complete and partial lockdown set up to limit the spread of this disease, such as stay-at-home orders via application of social distancing, perfect house ventilation, and environment cleanliness are useful in reducing the diffusion of this virus (Liu et al., 2020a).

The COVID-19 pandemic has led the governments of world countries to take strict measures, which are unparalleled in recent history, to limit the spread of this disease, such as stay-at-home orders via application of complete or partial lockdown. This led to a decrease in the transportation sector and industrial activities, and the consequent sudden decrease in the emission of air pollutants to the atmosphere (Forster et al., 2020; Venter et al., 2020; He et al., 2021a,b; Naqvi et al., 2021; Lv et al., 2022). Although the complete and partial lockdown set up to limit the spread of the COVID-19 pandemic led to an unprecedented and fast reduction in economic and social movements, they give a unique scientific chance to understand the effects of anthropogenic emissions, from regional to global, on the earth’s atmosphere (Forster et al., 2020; He et al., 2020; Kroll et al., 2020; Le Que’r et al., 2020; Liu et al., 2020b) as well as on the climate and earth system (Duffenbaugh et al., 2020; Phillips et al., 2020; Raymond et al., 2020). In particular, a significant reduction in air pollution levels during COVID-19 lockdowns or partial lockdowns, especially for NOx and PM, was noticed in numerous countries around the world (Shrestha et al., 2020; Tobías et al., 2020; Wang and Su, 2020; Zhang et al., 2020; Muhammad et al., 2020; Dutheil et al., 2020; Collivignarelli et al., 2020; Venter et al., 2020; Parra and Espinoza, 2020; Zambrano-Monserrat and Ruano, 2020; Hashim et al., 2020; He et al., 2021a,b; He et al., 2021a,b). However, there are no data concerning the effect of partial lockdown on the chemical characterization of atmospheric aerosol and the secondary inorganic aerosol production in a semi-desert climate area like Egypt. Therefore, this study is the first in-depth investigation of the effect of the COVID-19 partial lockdown on the atmospheric aerosol chemistry and enhanced secondary inorganic aerosol production and its effect on the chemical characterization of aerosol in a suburb of Greater Cairo (GC), Egypt. Finally, this study is focused on the side effects of intense anthropogenic emissions reduction, especially NOx, and the nonlinear response of chemical composition and characterization of PM2.5 to help us implement control strategies for secondary SO2 and NOx pollution, and consequently decrease atmospheric aerosol levels.

Both atmospheric SO2 and NO2 are produced from the oxidation of SO2 and NO2 which are arising from various anthropogenic sources like power plants, industry, motor vehicles, biomass burning, through atmospheric gas phase, and droplet phase reactions (Tsitouridou et al., 2005; Khoder, 2002; Hassan et al., 2013; Hassan and Khoder, 2017). The reaction of the gaseous acid species (e.g., H2SO4, HNO3, peracids, and oxides of nitrogen) with photochemical processes in the atmosphere with ammonia gas (NH3) that is released from both agricultural and non-agricultural sources in the atmosphere is also a source of ammonium salts (Lazaridis et al., 2006; Querol et al., 2001; Seinfeld and Pandis, 2012; Zhu et al., 2015). Therefore, it is very important to study the factors affecting the concentration, chemical composition, and characterization of atmospheric PM, especially PM2.5, in order to develop suitable control strategies. The first-time detection of the novel coronavirus-19 (COVID-19) infectious disease was recorded in December 2019 in the Wuhan province, China (Huang et al., 2021; Chen et al., 2020a,b; Zhang et al., 2021; Aify et al., 2021), and later spread to other countries, then became a pandemic (Bai et al., 2020; Sohrabi et al., 2020; Lai et al., 2020). Previous studies (Contini and Costabile, 2020; Liu et al., 2020a) stated that exposure to high levels of air pollution for a long time may lead to an increase in vulnerability and mortality rates due to COVID-19. However, the factors that influence the fast spread of the COVID-19 virus and mortality rates, like air pollution and PM, are still under debate (Conticini et al., 2020; Yao et al., 2020). Although there is no confirmation in ozone concentrations, as a secondary air pollutant, was observed in numerous studies (Nault et al., 2018; Shah et al., 2018; Laughner and Cohen, 2019; Womack et al., 2019). Therefore, COVID-19 lockdowns provide a unique chance to understand the sensitivity of atmospheric secondary pollutants formation and production as well as their effect on the characterization of PM during primary emission changes.

Egypt was not excluded from the hit of the COVID-19 pandemic. As one of the most densely populated countries in the world, the Egyptian government carried out a set of measures to prevent the fast spread of the COVID-19 virus. These strict measures included placing travel bans, locking the international borders, quarantine for infected residential areas, limitation in social movement and public ingathering (including religious activities in mosques, sports clubs, coffee shops, restaurants, and beaches) as well as implementing a partial lockdown, which included the suspension of nighttime transportation, schools, universities and educational institutions, shopping centers, and suspension of public transportations during Eid festival days. This partial lockdown led to a reduction in the emissions of air pollutants and consequently changed the atmospheric chemistry regimes. Power plants and industrial activities were not affected by strict measures taken during the COVID partial lockdown in Egypt.

Recently, air pollution became the most serious environmental problem in Greater Cairo (GC) which has suffer from high PM pollution levels. PM emits directly from natural and anthropogenic sources, and/or formed from conversion of gases into secondary particles through chemical reactions (Byrd et al., 2010; Im et al., 2012; Police et al., 2016; Hassan and Khoder, 2017; Marchetti et al., 2019). The existence of intensive anthropogenic activities from industrial activities, big electrical power stations and vehicles running in the streets of the GC city led to an increase in the emission of O3 precursors (NOx and VOCs) beside PM. The general climate of GC is cold, moist and rainy in winter; however, in summer it is characterized by high temperature, high solar radiation, and clear sky and rainless. So, the problem of air pollution in GC has been shifted towards the so called secondary photochemical pollutants, and their formation is facilitated by the local climatic conditions (high temperature, intense solar radiation, and clear sky), especially in the summer season (Khoder, 2002; Hassan et al., 2013). One of the most important secondary air pollutants that formed from the photochemical reactions is SO2 and NO2 aerosol. This lead to an increase in the concentration of atmospheric PM. Therefore, understanding the factors affecting the chemical characterization, formation of secondary inorganic pollutants and atmospheric aerosol chemistry in GC atmosphere are very important. Previous research articles were focused on the status of air pollution, aerosol chemical composition, sources and secondary pollutants like O3 before and during the COVID-19 lockdown period in different countries around the world (e.g. Liu et al., 2021; Chang et al., 2020; Huang et al., 2021; Yang et al., 2022; Shrestha et al., 2020; Tobías et al., 2020; Wang and Su, 2020; Zhang et al., 2020; Muhammad et al., 2020; Dutheil et al., 2020; Collivignarelli et al., 2020; Venter et al., 2020; Parra and Espinoza, 2020; Zambrano-Monserrat and Ruano, 2020; Hashim et al., 2020; He et al., 2021a,b; He et al., 2021a,b).
2. Materials and methods

2.1. Study area and sampling period

Greater Cairo (Cairo, Giza, and Qalyubia, Fig. 1) is one of the largest megacities in the world. It includes about 22% of Egypt’s population (approximately 15–20 million inhabitants) with a density of 13,107 inhabitants/km² (Mostafa et al., 2018; Shaltout et al., 2020). Industrial activities are concentrated in the northern (Shoubra El Kheima) and southern (Helwan) regions of the city. Traffic density and congestion is a considerable problem in GC, since more than 3.4 million vehicles, fueled by unleaded gasoline, natural gas, and diesel fuels, are operating on city streets (Parry and Timilsina, 2015; Shaltout et al., 2018; Abbass et al., 2020). Generally, rubbish burning, and traffic and industrial emissions are the major sources of air pollution in GC (Parry and Timilsina, 2015; Shaltout et al., 2018, 2019; Abbass et al., 2020).

Gaseous NO₂ and SO₂ as well as aerosol PM_{2.5} samples were collected on the roof of a private building, located in a suburban area (Kafr Tohormos) of GC (Fig. 1), at a height of ~15 m above ground level. Traffic emissions represent the main sources of air pollution in the study area. Based on the timeline of controlling and prevention of the spreading of the COVID-19 pandemic released by the Egyptian government, which imposed the strictest measures, the first-level emergency response was immediately imposed by all governorates in Egypt. On 29 March, partial lockdown measures were issued by the Egyptian government, which lasted until 14 June. Therefore, the observation period for the study of the gaseous pollutants and characterization of PM_{2.5} is divided into three periods: pre-COVID (2 February–22 March 2020), COVID partial lockdown (29 March–31 May), and post-COVID (28 June - 16 August). Samples were collected on a daily basis (24 h, from 7.00 a.m. to 7.00 a.m. on the following day) during the three different study periods.

Our focus objective, in the present study, is primarily on the effect of the COVID partial lockdown on the decreasing of in air pollutant concentrations and its effect on the chemistry of the atmosphere. Given that the period pre-COVID pandemic, which included the late winter (February) and early spring (March), and the period of COVID-partial lockdown during the COVID pandemic, which also included the rest of the spring months (April and May), the climatic conditions almost do not undergo major changes and are almost similar. Moreover, our rationale for choosing the comparison days during the two periods, before and during the partial lockdown, was based on strict criteria such as excluding the abnormal days like rainy days or dust storm days and using only the normal days with sunny skies during the two periods (before and during the partial lockdown). This will help us to investigate the effect of the COVID-partial lockdown on the decreasing of in air pollutant concentrations by comparing the status of air pollution before and during the partial lockdown periods. On the other hand, the post-COVID period during the summer season (28 June - 16 August), which is characterized already by high solar radiation with high dispersion rate for air pollutants, was used to find out whether the canceling of the partial lockdown during the COVID period will increase the primary pollutants such as NO₂ and SO₂ that are emitted directly from their sources or not. It’s well known that most of these pollutant are decreased in Cairo’s atmosphere during the summer season due to high dispersion and their conversion to the secondary pollutants (Khoder, 2002; Hassan et al., 2013).

2.2. Sampling and analysis of primary gaseous air pollutants

Ambient NO₂ gas was collected by bubbling air, at a flow rate of 0.5 L/min, through a sodium hydroxide–sodium arsenite solution to produce a stable sodium nitrite solution according to Harrison and Perry (1986). The nitrite ion that formed during the sampling time was reacted...
with phosphoric acid, sulfuranilide, and N-(1-naphthyl)-ethylene-
amic dihydrochloride to produce an azo dye. Thereafter, the absorb-
bance of the collected sample versus the reagent blank was measured at 540 nm spectrophotometrically. The calibration standard curve was prepared from an external standard nitrite solution. Based on the volume of air (m³) during the sampling period (24 h) and the standard curve, the ambient NO₂ concentration (μg/m³) was calculated. The method detection limit (MDL) for NO₂ was 3 μg/m³ for 24 h sampling. The analytical precision of measured NO₂ estimated from the standard deviation of repeated analyses of the standard solution was 3.5%.

The West and Gaek (colorimetric) method (Harrison and Perry, 1986) was used for the determination of atmospheric SO₂ gas in the present study. It is essentially specific for the determination of SO₂ since the exposed samples are relatively stable after collection (Harrison and Perry, 1986). Moreover, the West and Gaek method has been vastly used as a reference method and is covered by an international standard (UNEP/WHO, 1994). Ambient SO₂ gas was collected by bubbling air, at a flow rate of 0.5 L/min, through a solution of potassium tetrachloromercurate to form a stable non-volatile dichlorosulfotomercurate ion. Subsequently, solutions of purified, acid-bleached pararosanilin and formaldehyde were added to the exposed absorbing solution to form intensely colored pararosaniline methyl sulphonic acid, and thereafter the absorbance of the collected sample versus the reagent blank was measured at 560 nm spectrophotometrically. The concentration of SO₂ was determined from the prepared calibration standard curve, and then its concentration (μg/m³) in ambient air was calculated based on the volume of air (m³). The method detection limit (MDL) for SO₂ was 5 μg/m³ for 24 h sampling. The analytical precision of the measured SO₂ estimated from the standard deviation of repeated analyses of the standard was 3%.

2.3. PM₂.₅ sampling and its mass calculation

Ambient air PM₂.₅ samples were collected only during the periods of gaseous SO₂ and NO₂ sample collection. Two SKC IMPACT samplers (SKC Inc. USA), designed for the collection of PM₂.₅ in ambient air at a flow rate of 10 L/min, were used for the sampling of PM₂.₅. One set of PM₂.₅ samples was used for the elemental determination, whereas the other set of samples was utilized for water-soluble inorganic ion species (WSIs) analysis. PM₂.₅ samples were collected on quartz filters (diameter 47- mm). For the gravimetric analysis of PM₂.₅ in air, quartz filters were placed in controlled temperature and relative humidity desiccators for 24 h before and after sampling to remove any moisture. The difference in the weight of filter masses before and after sampling time represented the mass of PM₂.₅ dust on the filter, then the concentration of PM₂.₅ in the air was calculated from the dust mass on the filter and volume of air (m³) during the sampling time.

2.4. Chemical analysis of atmospheric aerosol PM₂.₅

2.4.1. Elemental analysis

Aerosol loaded on the quartz filters of the first set of PM₂.₅ samples was used for the elemental measurement using a digestion method. To prevent the contamination of PM₂.₅ samples during the steps of digestion processes and analysis of elemental concentrations, all equipment, glassware, and filters were acid-washed with HNO₃ solution, rinsed with double-distilled water, and then oven-dried. Moreover, all chemicals used for sampling digestion and analysis were obtained from Merck Company. The digestion step of PM₂.₅ samples for their elements (Zn, Pb, Cr, Cu, V, Cd, As, Co, Ni, Mn, and Fe) analysis was carried out according to Method 10-3.1 (1999) Hassan et al. (2013) and Hassan and Khoder (2017). To determine the concentrations of these elements, quartz filters were transferred into cleaned conical flasks of 50 ml volume. Thereafter, ten (10) ml of aqua regia (HCl: HNO₃ 3:1) was added to the conical flasks and left to digest overnight in the fume hood. Then, the samples were placed on a hot plate for 2 h at 90 °C till evaporated near to dryness, and subsequently, 5 ml of double-distilled water was added and heated at 100 °C on the hot plate till near to dryness. Afterwards, 25 ml of double-distilled water was added to the samples, then cooled at room temperature and filtered through Whatman filter paper (No. 42) using double-distilled water for washing. Finally, the digested samples were stored at 4 °C in pre-cleaned polyethylene bottles until analysis.

Inductively Coupled Plasma Optical Emission Spectrometry (ICP – OES 5100, Thermo Fisher Scientific, MA, USA) was used to analyze the elementals in the digested PM₂.₅ samples. Calibration curves, for the measured elements, were prepared from stock standard solutions, and subsequently, the elemental concentrations in air based on the air volume (M³) were calculated as μg/m³. Quality assurance and quality control (QA/QC), including PM₂.₅ digestion processes and analyses of field, laboratory and reagent blanks, analytical standards, ICP – OES 5100 calibration and detection limits, were carried out continuously. The ICP – OES 5100 was calibrated using standard solutions prepared from the multi-element stock standard. The external elements contamination from the field blanks, as well as laboratory blanks, HCl, HNO₃, and the double-distilled water used in the digestion process, were determined by the same used analytical procedures. All the blanks, acids, and double-distilled water were lower than the method detection limits (MDLs), and the measured element concentrations data of the PM₂.₅ samples were corrected by these blanks. Standard solutions of the target elements were run as a sample after analyzing ten (10) digested PM₂.₅ samples to realize the precision and accuracy of ICP OES 5100 as well as to minimize the error. The analytical precisions of measured elements estimated from the standard deviation of repeated measurements of standards were lower than 3%. MDLs for measured elements were specified from replicate measurements of low concentration and their standard deviations. MDLs for all measured elements ranged from 0.004 ng/m³ to 0.013 ng/m³.

2.4.2. Water-soluble inorganic ion species (WSIs) analysis

Double-distilled water (20 ml) was used to extract aerosol loaded on the quartz filters of the second set of PM₂.₅ samples, in 50 ml polypropylene tubes using an ultrasonic bath for 40 min, then filtered through a filter paper (Whatman No. 42) and refrigerated in pre-cleaned polyethylene bottles at 4 °C until WSIs analysis. The target anions (NO₃⁻, SO₄²⁻ and Cl⁻) and cations (NH₄⁺, Ca²⁺, Mg²⁺, Na⁺, and K⁺) species were analyzed by ion chromatography (Thermo IC-5000). An Ion Pac ASRS-4 suppressor and an Ion Pac CS11-HC × 250 mm analytical column were used for the detection of anions, whereas an Ion Pac CRS-S4 suppressor and an Ion Pac CS12A × 250 mm analytical column were used for cations detection. The eluents of 10 mmol/L Na₂CO₃ and 10 mmol/L CH₃O₂S were used for anions and cations analysis, respectively. A 10 μL loop was used for the sample’s injection. QA/QC, including field blank, laboratory and reagent blanks, analytical standards, ion chromatography calibration, and detection limits, was carried out continuously. Calibration standard curves were prepared from external standard solutions. The Ion Chromatography was calibrated using the prepared standard solutions. Field blanks, laboratory blanks, and the double-distilled water used in the extraction process were analyzed through similar procedures with the loaded filter to evaluate the external WSIs contamination and for quality control. All the blanks and double-distilled water were lower than the method detection limits (MDLs), and the measured WSIs concentrations data of the PM₂.₅ samples were corrected by these blanks. The analytical precisions of measured WSIs estimated from the standard deviation of repeated measurements of standards were lower than 4.5%. MDLs for measured WSIs were specified from replicate measurements of low concentration and their standard deviations. MDLs for measured WSIs were specified from replicate measurements of low concentration and their standard deviations.
2.4.3. Enhancement of secondary transformation

The sulfur conversion (oxidation) ratio ($F_{\text{SCR}}$) and nitrogen conversion (oxidation) ratio ($F_{\text{NCR}}$) are widely used to get a deep insight into the enhancement of secondary transformation, i.e., the production rates of $SO_2^+$ and $NO_3^-$ and the degree of conversion of $SO_2$ and $NO_2$ to $SO_2^+$ and $NO_3^-$ in the atmosphere (Sicard et al., 2020; Yuan et al., 2015; Hassan et al., 2013). The $F_{\text{SCR}}$ and $F_{\text{NCR}}$ have been calculated using the following formulas (Khoder, 2002):

$$F_{\text{SCR}} = [SO_2^+]/([SO_2] + [SO_2^-])$$

(1) $$F_{\text{NCR}} = [NO_2^-]/([NO_2]^+ + [NO_3^-])$$

(2) Where $SO_2^+$ is the sulfate concentration, as $SO_2$, $\mu g/m^3$; $NO_3^-$ is the gas–phase $SO_2$ concentration, $\mu g/m^2$; $NO_2^-$ is the nitrate concentration, as $NO_2$, $\mu g/m^3$; and $NO_2$ is the gas–phase $NO_2$ concentration, $\mu g/m^3$.

2.4.4. Acid-base balance of atmospheric aerosol PM$_{2.5}$

To comprehend the acid-base balance of atmospheric PM, ion balance using the sum of cations ($\sum A$) and anions ($\sum C$) microequivalents concentrations needs to be evaluated according to previous studies (Shen et al., 2011; Wang et al., 2013; Gao et al., 2015; Hassan and Khoder, 2017). This will help us understand the acidity of atmospheric PM and the ways to limit it since the acidity is an important key that affects the hygroscopic growth, toxicity, and heterogeneous reactions of this PM (Sun et al., 2010; Cheng et al., 2014). PM is in acidic status when the ratio of the sum of the microequivalents concentrations of anions (A) to cations (C) ($A/C$) is higher than one, and the increase in the value of $A/C$ ratio suggests an increase in particle acidity. Values of $A/C$ ratios close to one indicate particulate in ionic equilibrium state. In the present study, the $A/C$ and $C$ in the PM$_{2.5}$ were calculated as follows:

$$\sum A = \sum SO_2^+/48 + NO_3^-/62 + Cl^-/35.5$$

(3) $$\sum C = NH_4^+/18 + Ca^{2+}/20 + K^+/39 + Mg^{2+}/12 + Na^+/23$$

(4)

2.4.5. Neutralization of atmospheric aerosol PM$_{2.5}$

The calculation of neutralization factors (NFs) gives an insight into the roles of $NH_3$ gas and crustal components such as $Ca^{2+}$ and $Mg^{2+}$ in the neutralization of acidic components of atmospheric PM$_{2.5}$. NFs have been calculated using the following formula (Saxena et al., 1996; Hassan et al., 2015; Khoder and Hassan, 2008):

$$NF_{\text{NH}_4} = [NH_3]/[2[SO_2^+] + [NO_3^-]]$$

(5) $$NF_{\text{Mg}_{2+}} = [Mg^{2+}]/([SO_2^-] + 2[NO_3^-])$$

(6) $$NF_{\text{Ca}_{2+}} = [Ca^{2+}]/([SO_2^-] + 2[NO_3^-])$$

(7)

2.4.6. Aerosol acidity estimation using thermodynamic model

The E–AIM-IV model (http://www.aim.env.uea.ac.uk/aim) was used in the present study for estimating the aerosol acidity (Gong et al., 2021). Based on the E–AIM-IV model, particle total acidity ($[H^+]_t$) was firstly calculated by eq. (8) with using the field measurement data:

$$[H^+]_t = 2[SO_2^+] + [NO_3^-] + [Cl^-] - [NH_4^+] - [Na^+] - [K^+]$$

(8) where $[H^+]_t$, $[SO_2^+]$, $[NO_3^-]$, $[Cl^-]$, $[NH_4^+]$, $[Na^+]$, and $[K^+]$ are the ambient air concentrations of aerosol-bound ions in mol/L. After that, based on the state of the thermodynamic equilibrium system of $H^+-NH_4^+-Na^+-SO_2^+-NO_3^-+Cl^-+H_2O$ with temperature in the range of 263.15–330 K (Behera et al., 2015), in situ aerosol acidity was estimated using the following equation:

in situ $pH = - \log q_{H^+} = - \log \{[H^+]_t \times 1000 \times p/m\}$

(9) where $q_{H^+}$ is activity of $H^+$ in mol/L in the aqueous phase on the particle, $\gamma_{H^+}$ is the activity coefficient of the aqueous phase $H^+$, and $[H^+]_t$ is the free $H^+$ concentration of the aqueous phase in unit of mol per m$^{-3}$ of air, $p$ is the aerosol aqueous phase density in g/cm$^3$, and $m$ is the air concentration in g/m$^3$ in the aqueous phase. The parameters, $p$, $\gamma_{H^+}$, and $[H^+]_t$ and the concentration of aerosol water content (H$_2$O) were derived from the E-AIM-IV model.

2.5. Meteorological parameters

Meteorological data, including the relative humidity, ambient temperature, and wind speed, over the three study periods were taken from Weather Underground (www.wunderground.com).

3. Results and discussion

3.1. Temporal variations of PM$_{2.5}$, $NO_2$, and $SO_2$ concentrations

The temporal variations in the daily concentrations of PM$_{2.5}$, $NO_2$, and $SO_2$ as well as the values of meteorological parameters (temperature, relative humidity, and wind speed) during the pre-COVID, COVID partial lockdown, and post-COVID periods are graphically presented in Fig. S1. In this figure, it can be seen that the daily concentrations of PM$_{2.5}$, $NO_2$, and $SO_2$ pollutants maintained at a relatively high level during the pre-COVID period, then decreased dramatically during the COVID partial lockdown and the concentrations increased gradually again during the post-COVID period. These temporal variations in daily concentrations during the different study periods generally coincide with the effect of the increase or decrease in their emission sources during these times. Although we could not obtain any data regarding the rate of decreasing in the number of cars during the covid-19 period, the decreasing in traffic flow during COVID partial lockdown period due to closing of the schools and colleges and the stopping the traffic flow during nighttime was clear. Therefore, the decline of PM$_{2.5}$ and $NO_2$ might be attributed to the reduction of transportation activity, while this could not explain the drop of $SO_2$, the sources of $SO_2$ were mainly industrial activities and power plants, not vehicle emissions.

Table 1 presented the average concentrations of PM$_{2.5}$, $SO_2$, and $NO_2$ as well as the temperature, relative humidity, and wind speed values during the three study periods. The highest average temperature (28.8 °C) was recorded in the post-COVID period, whereas the lowest value was found in the pre-COVID period (16.1 °C). The average wind speeds were 10.1 km/h in the pre-COVID period, 10.4 km/h in the COVID partial lockdown period, and 8.8 m/h in the post-COVID period. As for relative humidity, its average values ranged from 46.6% in the COVID partial lockdown period to 57.4% in the pre-COVID period. Insignificant correlation coefficients ($P < 0.001$) were found between the temperature and the concentrations of $SO_2$ ($r = 0.23–0.39$), $NO_2$ ($r = 0.25–0.49$) and PM$_{2.5}$ ($r = 0.30–0.45$) and between the relative humidity and $SO_2$ ($r = -0.24–0.37$), $NO_2$ ($r = 0.21–0.38$) and PM$_{2.5}$ ($r = 0.20–0.31$) concentrations, and between the wind speeds and the concentrations of $SO_2$ ($r = 0.23–0.39$), $NO_2$ ($r = 0.18–0.30$) and PM$_{2.5}$ ($r = 0.27–0.46$) during the thee periods of study. These results indicate that the concentrations of measured air pollutants during pre-COVID period and their decreasing (COVID partial lockdown period)/increasing (post-COVID period) were not mainly depending on the meteorological conditions but fundamentally depending on the strength of local emission sources, especially traffic emissions. As reported in Table 1, the average concentrations of PM$_{2.5}$, $SO_2$, and $NO_2$ were 60.2, 89.4, and 38.5 μg/m$^3$ in the pre-COVID period, 56.7, 55.3, and 39.2 μg/m$^3$ in the COVID partial lockdown period, and 54.3, 65.2, and 45.9 μg/m$^3$ in the post-COVID period, respectively. Compared with the mean concentrations of PM$_{2.5}$, $NO_2$, and $SO_2$ during the pre-COVID period, PM$_{2.5}$ had the largest reduction of 39.0%, followed by $NO_2$ at 38.1%, and $SO_2$ at 32.9% during the COVID partial lockdown. On the other hand, the change in PM$_{2.5}$, $NO_2$, and $SO_2$ mean concentrations during the COVID partial
lockdown to the post-COVID period were +32.4%, +15.2%, and +14.6%, respectively. In Egypt, the COVID partial lockdown included mainly closing schools and universities, and the strictest measures that suspended traffic flow during nighttime, which is normally characterized by traffic congestion until the early hours of the morning on normal days. Consequently, the sharp decrease in traffic volume during the COVID partial lockdown period led to a sharp reduction in the concentration levels of PM$_{2.5}$ and SO$_2$, and NO$_x$. However some studies report an increase in secondary air pollution like PM$_{2.5}$ in China (Chen et al., 2020a,b) and France (Shai et al., 2021a) and Morocco in north Africa (Shai et al., 2022). Because photochemistry can be a significant contributor to PM formation especially under high ozone levels because strong oxidizing capacity could accelerate the formation of OH radicals in the atmosphere in the presence of humidity; OH radicals react with VOC and BVOC and lead to the formation of secondary aerosols which represent a significant fraction of PM$_{2.5}$ (Ortega et al., 2016; Sbai and Dutheil, 2020). In the present study, the relatively lower reduction rate of SO$_2$ than NO$_x$ suggests that COVID restrictions had a much more significant influence on traffic emissions than on power plants and industrial activities that are the main sources of SO$_2$. When restrictions were lifted in the post-COVID period, all the measured pollutants rebounded and there was a progressive increase in their concentrations, especially PM$_{2.5}$.

### Table 1

Average atmospheric aerosol PM$_{2.5}$, NO$_x$ and SO$_2$ concentrations, as well as temperature, relative humidity and wind speed values during pre-COVID, COVID partial lockdown and post-COVID periods.

| Parameter | Pre-COVID | COVID partial lockdown | Post-COVID |
|-----------|-----------|------------------------|------------|
| PM$_{2.5}$ | N | Range | Mean | SD | N | Range | Mean | SD | N | Range | Mean | SD |
| NO$_x$ | 15 | 40.4-75.7 | 60.2 | 11.1 | 23 | 16.3-56.5 | 36.7 | 11.4 | 15 | 41.9-64.7 | 54.3 | 7.4 |
| SO$_2$ | 15 | 67.3-106.1 | 89.4 | 12.1 | 23 | 39.5-66.9 | 55.3 | 7.1 | 15 | 47.7-78.9 | 65.2 | 8.9 |
| Temperature (°C) | 15 | 42.6-69.8 | 58.5 | 7.7 | 23 | 27.6-47.4 | 39.2 | 6.2 | 15 | 34.9-55.1 | 45.9 | 5.9 |
| Relative humidity (%) | 15 | 12.2-19.6 | 16.1 | 2 | 23 | 19.4-30.0 | 23.7 | 2.9 | 15 | 23.0-31.3 | 28.8 | 1.8 |
| Wind Speed (km/h) | 15 | 28.9-74.7 | 57.4 | 11.7 | 23 | 24.0-60.2 | 46.6 | 9.6 | 15 | 30.6-64.8 | 53.9 | 9.2 |

N: number of daily samples. SD: standard deviation.

Dutheil et al., 2020). In the present study, the relatively lower reduction rate of SO$_2$ than NO$_x$ suggests that COVID restrictions had a much more significant influence on traffic emissions than on power plants and industrial activities that are the main sources of SO$_2$. When restrictions were lifted in the post-COVID period, all the measured pollutants rebounded and there was a progressive increase in their concentrations, especially PM$_{2.5}$.

#### 3.2. Chemical composition of atmospheric aerosol PM$_{2.5}$

##### 3.2.1. Elemental compositions of atmospheric aerosol PM$_{2.5}$

The measured element concentrations in PM$_{2.5}$ during the pre-COVID, COVID partial lockdown, and post-COVID periods are illustrated in Fig. 2. Based on the average element concentrations, Fe and Pb were the most dominant elements in PM$_{2.5}$, accounting for about 70.14% and 11.90% during the pre-COVID period, 78.14% and 7.97% during the COVID partial lockdown period, and 76.53% and 8.58% during the post-COVID period of the total measured elements concentrations, respectively. The total mass concentrations of elements in PM$_{2.5}$ followed the order pre-COVID (1639.5 ng/m$^3$) > COVID (1475.5 ng/m$^3$) > post-COVID (1223.6 ng/m$^3$). The total mass of elements in PM$_{2.5}$ during the COVID partial lockdown when compared with that of the pre-COVID period was reduced by about 25.4%. Compared with those during the pre-COVID period, Ni had the largest reduction of 55%, followed by Zn (52%), Pb (50%), Cr (48%), Cu (45%), V (44%), Cd (43%), As (41%), Co (38%), Mn (19%), and Fe (16.7%) during the COVID partial lockdown. On the other hand, the element concentrations increased gradually after the COVID partial lockdown period. The change in concentrations from the COVID partial lockdown period to the post-COVID period was +43.7% for As, +38% for Ni, +37% for Co, +35% for Cr, +35% for Cu, +34.4% for V, +30.1% for Zn, +30% for Pb, +29% for Cd, +19% for Mn, and +18% for Fe. Previous studies reported that elements like Cu, Pb, As, and Zn are emitted from non-exhaust sources, such as tire and brake wear (Alves et al., 2018; Jeong et al., 2019; Denier van der Gon et al., 2007). Traffic emissions, common usage of anti-corrosive galvanized car accessories, use of leaded gasoline, and lubricating oil leakages are the

![Fig. 2. Variation of the elemental concentration in atmospheric aerosol PM$_{2.5}$ during Pre-COVID, COVID partial lockdown and post-COVID periods.](image-url)
anthropogenic sources widely related to the emission of Cd, Cu, Pb, As, and Zn (Zhao et al., 2021a, 2021b; Patel and Jain, 2022; Wang et al., 2019; Kumari et al., 2021). It is therefore supposed that the decreasing of the anthropogenic emission sources of elements in PM, like industrial emissions and traffic density, contributes to the reduction of these element concentrations. So, the effect of decreased traffic density during the COVID partial lockdown did not only cause low exhaust particulate emissions, but also reduced emissions generated from tire wear and re-suspension of street dust, which include some elements. As a result, the substantial decrease in the concentrations of Ni, Zn, Pb, Cr, Cu, V, Cd, As, and Co during the COVID partial lockdown could be due to the restrictions on traffic activities during this period. This is consistent with Nguyen et al. (2021) who found that the concentrations of Cd, Se, As, Sr, Ba, Cu, Mn, Pb, K, Zn, Ca, Al, and Mg in Hanoi (Vietnam) within the partial lockdown period were lower than those prior to the partial lockdown. Li et al. (2020) and Kammiah et al. (2020) reported that the reduction of air pollution was mainly related to the restrictions on traffic and industrial activities. In the present study, a small reduction in Fe and Mn (mainly crustal elements) concentrations compared with other measured elements was recorded during the COVID partial lockdown, since they emit mainly from the re-suspension of road dust (Chang et al., 2009; Lee and Hieu, 2011; Lim et al., 2010).

3.2.2. Water-soluble inorganic ion species (WSIs) in atmospheric aerosol PM$_{2.5}$

3.2.2.1. Concentrations and distribution of WSIs. The average concentrations of the WSIs in PM$_{2.5}$ during the pre-COVID, COVID partial lockdown, and post-COVID periods are shown in Fig. 3. The PM$_{2.5}$ ions mass concentrations followed the order $\text{SO}_4^{2-} > \text{NO}_3^- > \text{NH}_4^+ > \text{Ca}^{2+} > \text{Cl}^- > \text{Na}^+ > \text{K}^+ > \text{Mg}^{2+}$ during pre-COVID and the COVID partial lockdown, and $\text{SO}_4^{2-} > \text{NO}_3^- > \text{Ca}^{2+} > \text{NH}_4^+ > \text{Cl}^- > \text{Na}^+ > \text{K}^+ > \text{Mg}^{2+}$ during post-COVID. Despite the relatively higher decline of gaseous NO$_2$ (38.1%) and SO$_2$ (32.9%) concentrations during the COVID partial lockdown period compared with the pre-COVID period, the decline in the concentrations of particulate SO$_4^{2-}$ (8.99%) and NO$_3$ (14.2%) was relatively lower compared with their precursor’s (NO$_2$ and SO$_2$) during the same time. This means that the transformations of NO$_2$ to NO$_3$ and SO$_2$ to SO$_4^{2-}$ were improved during the COVID partial lockdown. Yang et al. (2022) reported that the disproportionate drop of particulate NO$_3$ compared to NO$_2$ gas concentration (60%) implied the improved conversion of NO$_2$ during the COVID lockdown period. Compared with those during the pre-COVID period, Cl$^-$, NH$_4^+$, Ca$^{2+}$, K$^+$, Mg$^{2+}$, and Na$^+$ concentration reductions were 28.79%, 4.24%, 47.93%, 21.03%, 36.10%, and 27.36%, respectively, during the COVID partial lockdown in this study. After the COVID partial lockdown period, the changes in mean concentrations were +41.43% for NO$_3^-$, +32.00% for SO$_4^{2-}$, +29.71% for Cl$^-$, +26.63% for NH$_4^+$, +45.60% for Ca$^{2+}$, +25.45% for K$^+$, +34.02% for Mg$^{2+}$, and +31.90% for Na$^+$ during post-COVID. The average mass concentrations of crustal-mineral species (Ca$^{2+}$, K$^+$, Mg$^{2+}$ and Na$^+$) in PM$_{2.5}$ were 3.37 µg/m$^3$ (pre-COVID), 2.07 µg/m$^3$ (COVID partial lockdown), and 3.35 µg/m$^3$ (post-COVID). The relatively higher levels of crustal–mineral species in the pre and post-COVID periods than in the COVID partial lockdown period resulted from the effect of local and regional man-made activities on the production of crustal–mineral dust, like the re-suspension of road dust during traffic flow during the pre and post-COVID periods.

NO$_3$/SO$_4^{2-}$ mass concentration ratio can be used to distinguish between the relative importance of vehicular traffic versus stationary sources of sulfur and nitrogen in the atmosphere (Li et al., 2016). High values of this ratio reveal that the predominance of vehicular traffic sources is higher than stationary sources of these pollutants. In the present study, the mean NO$_3$/SO$_4^{2-}$ ratios in PM$_{2.5}$ during the pre-COVID, COVID partial lockdown, and post-COVID periods were lower than one, they were 0.52, 0.49, and 0.57, respectively. These ratios indicate the predominance of stationary sources over vehicular sources during the three different periods of study. These results are sensible since the power plant and heavy industries were still working during the COVID-19 pandemic period. The differences in the values of reduction of SO$_2$ compared with NO$_2$ during the COVID partial lockdown further support this argument. The relatively higher NO$_3$/SO$_4^{2-}$ ratio in both the pre and post-COVID periods may be attributed to the relatively higher traffic density during these times.

Among all WSIs determined in PM$_{2.5}$, SO$_4^{2-}$ is the most abundant chemical species in the pre-COVID, COVID partial lockdown, and post-COVID periods, accounting for about 8.49%, 12.01%, and 12.01% of the PM$_{2.5}$ mass, and 37.97%, 42.18%, and 40.54% of the total WSIs, respectively (Fig. 4). The sum mass concentration of the secondary inorganic ions (SNA (SO$_4^{2-}$, NO$_3^-$ and NH$_4^+$) accounts for about 14.81%, 21.27%, and 22.01% of the PM$_{2.5}$ mass, and 66.98%, 74.13%, and 73.42% of the total WSIs in the pre-COVID, COVID partial lockdown, and post-COVID periods, respectively. This result indicates that the formation of secondary pollutants (SNA) from the oxidation of its precursors during the COVID partial lockdown and post-COVID periods was enhanced. The concentrations of the total WSIs in PM$_{2.5}$ were 12.80 µg/m$^3$, 10.44 µg/m$^3$, and 15.97 µg/m$^3$, accounting for 21.51%, 28.65%, and 29.95% of the PM$_{2.5}$ mass during the pre-COVID, COVID partial lockdown, and post-COVID periods, respectively. The relatively higher percentages of WSIs in both the COVID partial lockdown and post-COVID periods than in the pre-COVID period indicate that WSIs form more PM$_{2.5}$ under COVID partial lockdown and summertime (post-COVID period) conditions.

3.2.2.2. Enhancement of secondary transformation. As seen in Fig. 5, an increase in the F$_{\text{SCR}}$ and F$_{\text{NCR}}$ values occurred during both the COVID partial lockdown and post-COVID periods compared with the pre-COVID period. The values of F$_{\text{SCR}}$ were 3.91% in pre-COVID, 4.53% in the COVID partial lockdown, and 7.51% in post-COVID. Compared with the values of F$_{\text{SCR}}$ in pre-COVID, F$_{\text{SCR}}$ values in the COVID partial lockdown and post-COVID periods were about 1.16 and 1.92 times, respectively, higher than F$_{\text{SCR}}$ values in the COVID partial lockdown and post-COVID periods. However, the values of F$_{\text{SCR}}$ were 8.75%, 12.83%, and 13.75% during the pre-COVID, COVID partial lockdown, and post-COVID periods, respectively. F$_{\text{SCR}}$ values in the COVID partial lockdown and post-COVID periods were about 1.47 and 1.57 times, respectively, higher than F$_{\text{SCR}}$ values in the pre-COVID period. The relatively higher values of F$_{\text{SCR}}$ and F$_{\text{NCR}}$ during the post-COVID period (summertime) led to an increase in SO$_2$ and NO$_3$ concentrations, indicating that the atmospheric oxidation of SO$_2$ and NO$_2$ to SO$_4^{2-}$ and NO$_3$ and their conversion are based on the atmospheric photochemical oxidation reactions. Consistent with previous studies (Khoder, 2002; Li et al., 2006; Chiwa, 2010; Hassan et al., 2013), the highest F$_{\text{SCR}}$ and F$_{\text{NCR}}$ were found in the summer season. Atmospheric NO$_2$ is formed from HNO$_3$, which is produced from the reactions of NO$_2$ with OH radicals during nighttime and with NO$_3$ radicals during nighttime (Seinfeld and Pandis, 2016). Moreover, SO$_4^{2-}$ is produced from the oxidation of SO$_2$ through both heterogeneous and homogeneous
reactions in the presence of relative humidity (Rattigan et al., 2002; Fahey et al., 2005). As a result, the concentrations of NO\textsubscript{3} and SO\textsubscript{4}\textsuperscript{2-} in PM\textsubscript{2.5} depend on the concentrations of NO\textsubscript{2} and SO\textsubscript{2} and their conversion rates to NO\textsubscript{3} and SO\textsubscript{4}\textsuperscript{2-}.

In the present study, despite the decline of NO\textsubscript{2} (38.1%), SO\textsubscript{2} (32.9%), SO\textsubscript{4}\textsuperscript{2-} (8.99%), and NO\textsubscript{3} (14.2%) in the COVID partial lockdown compared with those in the pre-COVID period, the F\textsubscript{SCR} and F\textsubscript{NCR} values in the COVID partial lockdown were about 1.47 and 1.16 times, respectively, higher than F\textsubscript{SCR} and F\textsubscript{NCR} values in the pre-COVID period. These results indicate that the relatively enhanced production rate of NO\textsubscript{3} and SO\textsubscript{4}\textsuperscript{2-} during the COVID partial lockdown did not enhance SO\textsubscript{4}\textsuperscript{2-} and NO\textsubscript{3} concentrations. This may be due to the reduction of NO\textsubscript{2} and SO\textsubscript{2} concentrations, which are the precursors of SO\textsubscript{4}\textsuperscript{2-} and NO\textsubscript{3}, during the same period. As a result, the concentrations of both NO\textsubscript{2} and SO\textsubscript{4}\textsuperscript{2-} in PM\textsubscript{2.5} still decreased, although their secondary production rates increased. Consistent with previous studies (Chang et al., 2020; Huang et al., 2021; Yang et al., 2022), the NO\textsubscript{3} formation was enhanced relative to the reduction of NO\textsubscript{2} emission during the COVID-lockdown period in Beijing, Zhengzhou, and Shanghai. In China (Yangtze River Delta region), nitrogen oxide (NOx) reductions enhanced O\textsubscript{3} accumulation and therefore increased secondary aerosol formation (Wang et al., 2021). Up to 90% reduction of certain emissions during the city-lockdown was observed, whilst O\textsubscript{3} enhancement and further increase in the atmospheric oxidizing capacity and secondary aerosol formation in urban areas were occurred (Le et al., 2020). The oxidation rates of SO\textsubscript{2} to SO\textsubscript{4}\textsuperscript{2-} and NO\textsubscript{2} to NO\textsubscript{3} during the COVID lockdown were higher than those during the pre-COVID period (Liu et al., 2021). Sulfur and nitrogen conversion ratios during the COVID lockdown period were higher than those during the pre-lockdown period (Yang et al., 2022). In the present study, the relatively higher values of F\textsubscript{SCR} and F\textsubscript{NCR} in the COVID partial lockdown suggest that the oxidizing capacity of the atmosphere was significantly enhanced during the COVID partial lockdown period, and consequently, the oxidation of NO\textsubscript{2} to SO\textsubscript{4}\textsuperscript{2-} and NO\textsubscript{2} to NO\textsubscript{3} increased. The increase of oxidizing capacity means enhancements in the production of O\textsubscript{3}, OH radical, and H\textsubscript{2}O\textsubscript{2}, which lead to an increase in the oxidation and the rate of conversion of SO\textsubscript{2} to SO\textsubscript{4}\textsuperscript{2-} and NO\textsubscript{2} to NO\textsubscript{3}. Although the concentration of O\textsubscript{3}, which is considered a key precursor of OH that controls the oxidizing power of the atmosphere, was not measured in the present study, previous studies reported that the concentration of O\textsubscript{3} increased during the COVID lockdown period due to a significant reduction in NO\textsubscript{x} emissions and a decrease in the O\textsubscript{3} consumption via NO titration (Dantas et al., 2020; Kerimray et al., 2020; Li et al., 2020; Nakada and Urban, 2020; Tobias et al., 2020; Sharma et al., 2020; Chu et al., 2020; Duan et al., 2021). Significant reduction in NO\textsubscript{x} and enhancement in O\textsubscript{3} has been found in India regions during the lockdown (Singh et al., 2020). By comparing the difference in O\textsubscript{3} concentrations during the three periods (before controlling, during controlling and after controlling) of COVID-19 in three important cities in the region (Wuhan, Changsha, and Nanchang) (Zhao et al., 2021c), it is found that after controlling the human activities, the O\textsubscript{3} concentration increased by 111.83%. NO\textsubscript{x} reductions can increase O\textsubscript{3} levels by decreasing the NO\textsubscript{x} titration (through NO + O\textsubscript{3} and also via nocturnal NO\textsubscript{x} + O\textsubscript{3} reactions) (Leung et al., 2020). The elevated O\textsubscript{3} concentration was favorable for the formation of NO\textsubscript{3} despite the drastic decrease of NO\textsubscript{2} during the COVID lockdown period (Chang et al., 2020; Huang et al., 2021; Lv et al., 2020; Xu et al., 2020; Yang et al., 2022). An increase in the oxidation capacity (O\textsubscript{3}) of the atmosphere led to the enhancement of sulfur oxidation ratio, and then SO\textsubscript{4}\textsuperscript{2-} formation during the COVID lockdown (Duan et al., 2021). Ren et al. (2021) reported that a 26–61% reduction of NO\textsubscript{x} emissions only lowered surface HNO\textsubscript{3} by 2–3% and enhanced the NO\textsubscript{3} formation in the presence of a high concentration of NH\textsubscript{3} reduction in China during the COVID-19 pandemic due to the enhanced atmospheric oxidizing capacity.

Relative humidity (RH) is considered an important meteorological factor that affects the chemical compositions and characterization of PM in the atmosphere (Sun et al., 2013; Tian et al., 2014; Hassan and Khoder, 2017). Therefore, the impact of RH on the F\textsubscript{SCR} and F\textsubscript{NCR} was...
investigated in the present study. Significant positive correlation coefficients ($p < 0.001$) were found between the RH and $F_{\text{SCR}}$ in the three different periods of study (Fig. S2). The correlation coefficients ($r$) were 0.86, 0.83, and 0.86 in the pre-COVID, COVID partial lockdown, and post-COVID periods, respectively. This result suggests that the RH is a significant agent for the oxidation processes of $SO_2$ to $SO_4^{2-}$, and enhancement of the conversion rate needs high relative humidity besides the presence of gas-phase reactions. On the other hand, the insignificant correlation coefficients between relative humidity and $F_{\text{NCR}}$ during the pre-COVID, COVID partial lockdown, and post-COVID periods (Fig. S2) indicate that the gas-phase reactions are important and predominant for the oxidation processes of $NO_2$ to $NO_3$. Previous findings showed that both droplet phase and gas-phase reactions are significant for the conversion of $SO_2$ to $SO_4^{2-}$, whereas the gas-phase reactions are the predominant for the conversion of $NO_2$ to $NO_3$ (Khoder, 2002; Hassan et al., 2013). In the present study, the correlation coefficients between wind speeds and the $F_{\text{SCR}}$ ($r = 0.26$–0.34) and $F_{\text{NCR}}$ ($r = 0.20$–0.31) were insignificant ($p < 0.001$) during the three periods of study. Moreover, statistically significant correlation coefficients ($p < 0.001$) were found between temperature and the $F_{\text{SCR}}$ ($r = 0.75$–0.84) and $F_{\text{NCR}}$ ($r = 0.79$–0.88) during pre-COVID, COVID partial lockdown, and post-COVID periods. This indicates that the increase of temperature that related with increase of solar radiation led to an increase in ozone concentration (Vukovich and Sherwell 2003; Ribas and Penuelas 2004; Garcia et al., 2005) which enhance the conversion of $SO_2$ to $SO_4^{2-}$ and $NO_2$ to $NO_3$.

![Relationship between the atmospheric aerosol PM$_{2.5}$ concentration and sulfur conversion ratio ($F_{\text{SCR}}$) and nitrogen conversion ratio ($F_{\text{NCR}}$) during pre-COVID, COVID partial lockdown and post-COVID periods.](image)

Fig. 6. Relationship between the atmospheric aerosol PM$_{2.5}$ concentration and sulfur conversion ratio ($F_{\text{SCR}}$) and nitrogen conversion ratio ($F_{\text{NCR}}$) during pre-COVID, COVID partial lockdown and post-COVID periods.
To investigate the effect of enhancement of conversion rate of primary pollutants (SO$_2$ and NO$_2$) into secondary aerosol (NO$_3^-$ and SO$_4^{2-}$) on the concentrations of atmospheric PM$_{2.5}$, correlation coefficients ($r$) between F$_{SCR}$ and PM$_{2.5}$ and F$_{NCR}$ and PM$_{2.5}$ were calculated during the three different periods of study (Fig. 6). Significant positive correlation coefficients were found between F$_{SCR}$ and PM$_{2.5}$ and between F$_{NCR}$ and PM$_{2.5}$. These correlation coefficients were 0.80, 0.85, and 0.87 for F$_{NCR}$ and PM$_{2.5}$, and 0.85, 0.92, and 0.85 for F$_{SCR}$ and PM$_{2.5}$, during the pre-COVID, COVID partial lockdown, and post-COVID periods, respectively. These results suggest that the enhancement of secondary aerosol

Fig. 7. Scatter plot of NH$_4^+$ and SO$_4^{2-}$ and NO$_3^-$ concentrations during pre-COVID, COVID partial lockdown and post-COVID periods.
formation, from the oxidation of gaseous NO\textsubscript{2} and SO\textsubscript{2}, is important for increasing the atmospheric PM\textsubscript{2.5}.

### 3.2.2.3. Chemical forms of SO\textsubscript{4}\textsuperscript{2−} and NO\textsubscript{3} in atmospheric aerosol PM\textsubscript{2.5}

The relationships and concentration ratios of SO\textsubscript{4}\textsuperscript{2−}, NO\textsubscript{3}, and NH\textsubscript{4} in the PM\textsubscript{2.5} can be used to get insight into the chemical forms of these secondary pollutants as well as the chemical characteristics of PM\textsubscript{2.5}. Significant positive correlation coefficients ($p < 0.001$) were found between the concentrations of NH\textsubscript{4} as a neutralizing ion and the sum of acidifying ions ($\sum$SO\textsubscript{4}\textsuperscript{2−} and NO\textsubscript{3}) in PM\textsubscript{2.5} during pre-COVID ($r = 0.85$), COVID partial lockdown ($r = 0.78$) and post-COVID ($r = 0.77$).

Moreover, the correlation coefficients between the concentrations of NH\textsubscript{4} and both SO\textsubscript{4}\textsuperscript{2−} and NO\textsubscript{3} during the pre-COVID, COVID partial lockdown, and post-COVID periods are illustrated in Fig. 7. A significant positive correlation coefficient was found between the concentrations of NH\textsubscript{4} and NO\textsubscript{3} in PM\textsubscript{2.5} during pre-COVID ($r = 0.75$, $p < 0.001$), clearly suggesting that NO\textsubscript{3} in PM\textsubscript{2.5} may have been present as NH\textsubscript{4}NO\textsubscript{3} during the pre-COVID period, which is characterized by low atmospheric temperature. On the contrary, under the effect of relatively higher temperature during the COVID partial lockdown and post-COVID periods, a portion of NH\textsubscript{4}NO\textsubscript{3} is volatilized (Liu et al., 2017), forming nitric acid (HNO\textsubscript{3}) and ammonia (NH\textsubscript{3}) gas. Then, a fraction of HNO\textsubscript{3} forms...
particulate NO\(_3^-\) through the gas-to-particle conversion in the presence of the dust particles with alkaline components (Tang et al., 2017; Wang et al., 2018). Consequently, particulate NO\(_3^-\) other than NH\(_4\)NO\(_3\) probably existed. NO\(_3^-\) can be associated with other minerals to form Mg (NO\(_3\)_2), KNO\(_3\), and Ca (NO\(_3\)_2) (Qiao et al., 2019). Acidic nitrogen species can react with alkaline ions (Ca\(^{2+}\), Mg\(^{2+}\), and K\(^+\)) in particles (Hayami and Carmichael, 1998; Bourrot et al., 2007; Hassan et al., 2013; Wang et al., 2017). This is confirmed by the insignificant positive correlation coefficients between the concentrations of NO\(_3^-\) and NH\(_4^+\), and the significant positive correlations between the concentrations of NO\(_3^-\) and Ca\(^{2+}\), K\(^+\), and Mg\(^{2+}\) in the COVID partial lockdown and post-COVID periods. The concentrations of NH\(_4^+\) and SO\(_4^{2-}\) in PM\(_{2.5}\) were significant (p < 0.001) positive correlated during the three different study periods (Fig. 7), the correlation coefficients (r) were 0.82, 0.82, and 0.77 during the pre-COVID, COVID partial lockdown, and post-COVID periods, respectively. Moreover, significant positive correlation coefficients (p < 0.001) were found between SO\(_4^{2-}\) and Ca\(^{2+}\) concentrations during post COVID (r = 0.83), COVID partial lockdown (r = 0.86), and post-COVID (r = 0.87). These results indicate that both NH\(_4^+\) and Ca\(^{2+}\) play important roles in the neutralization of sulfate, and (NH\(_4\)_2)SO\(_4\) and/or NH\(_4\)HSO\(_4\) and CaSO\(_4\) are sulfate forms existent in the PM\(_{2.5}\); this is consistent with previous studies (Tang et al., 2004, 2017; Wang et al., 2017).

The value of NH\(_4^+\)/SO\(_4^{2-}\) molar ratio is considered the borderline to describe the PM system in poor or rich NH\(_4^+\) systems (Veire-Filho et al., 2016). With the excess of atmospheric NH\(_4^+\) gas, PM is fully neutralized to produce (NH\(_4\)_2)SO\(_4\), and the NH\(_4^+\)/SO\(_4^{2-}\) molar ratio exceeds 2 (Walker et al., 2004; Huang et al., 2011). In the presence of CaCO\(_3\), (NH\(_4\)_2)SO\(_4\) can be converted to (NH\(_4\)_2)SO\(_4\).CaSO\(_4\).2H\(_2\)O and further to CaSO\(_4\) within 2–6 days (Mori et al., 1998). Based on the X-ray diffraction, (NH\(_4\)_2)SO\(_4\).CaSO\(_4\).2H\(_2\)O existed when NH\(_4^+\)/SO\(_4^{2-}\) molar ratio was close to 1 (Querol et al., 1998). Particulate SO\(_4^{2-}\) existed as (NH\(_4\)_2)SO\(_4\), (NH\(_4\)_2)SO\(_4\).CaSO\(_4\).2H\(_2\)O, and CaSO\(_4\) when the NH\(_4^+\)/SO\(_4^{2-}\) molar ratios lie between 1:1 and 2:1 theoretical lines (Duan et al., 2003). In the present study, the NH\(_4^+\)/SO\(_4^{2-}\) molar ratios ranged from 1.08 to 1.71 (with a mean value of 1.32) during pre-COVID, 1.06 to 2.83 during the COVID partial lockdown (with a mean value of 1.44), and 0.98 to 1.81 (with a mean value of 1.28) during post-COVID. Fig. 8 illustrates the reference theoretical molar ratio for NH\(_4\)HSO\(_4\) (1:1) and (NH\(_4\)_2)SO\(_4\) (2:1), as well as the scatter plot of NH\(_4^+\) and SO\(_4^{2-}\) concentrations (μmol/m\(^3\)) during the pre-COVID, COVID partial lockdown, and post-COVID periods. It can be observed that, from this figure, the NH\(_4^+\)/SO\(_4^{2-}\) molar ratios in PM\(_{2.5}\) samples during the three periods of study scatter between 1:1 and 2:1 theoretical lines, indicating that SO\(_4^{2-}\) in PM\(_{2.5}\) during the three periods of study may be present as (NH\(_4\)_2)SO\(_4\), (NH\(_4\)_2)SO\(_4\).CaSO\(_4\).2H\(_2\)O, and CaSO\(_4\). This is consistent with the observation of previous investigators (Querol et al., 1998; Mori et al., 1998; Duan et al., 2003; Khoder and Hassan, 2008; Hassan et al., 2013) who suggested that SO\(_4^{2-}\) in PM may exist as (NH\(_4\)_2)SO\(_4\), (NH\(_4\)_2)SO\(_4\).CaSO\(_4\).2H\(_2\)O, and CaSO\(_4\) when NH\(_4^+\)/SO\(_4^{2-}\) molar ratios scatter between 1:1 and 2:1 theoretical lines. The results in this study displayed the high contribution of NH\(_4^+\) salts in the PM\(_{2.5}\) (Andrada et al., 2012; Albuquerque et al., 2012).

### 3.2.2.4. Acid-base balance of atmospheric aerosol PM\(_{2.5}\)

The scatter plot of ∑A and ∑C microequivalents values during the pre-COVID, COVID partial lockdown, and post-COVID periods are illustrated in Fig. 9. It can be seen that all PM\(_{2.5}\) samples during the pre-COVID period plotted below the reference theoretical ∑A/∑C ratio of 1:1 line. The values ∑A/∑C ratios ranged from 0.62 to 0.98 (with an average of 0.72), suggesting that the pre-COVID PM\(_{2.5}\) samples were alkaline. In the post-COVID and COVID partial lockdown periods, all PM\(_{2.5}\) samples plotted around the reference theoretical line 1:1 (∑A/∑C). The values of ∑A/∑C ratios for PM\(_{2.5}\) samples ranged from 0.83 to 1.19 (with an average of 0.99) during the COVID partial lockdown period and from 0.92 to 1.41 (with an average of 1.01) during the post-COVID period, which indicates that those PM\(_{2.5}\) samples were nearly neutral. On the other hand, significant (p < 0.001) positive correlation coefficients were found between ∑A and ∑C microequivalent concentrations. The correlation coefficients (r) were 0.70 during pre-COVID, 0.95 during the COVID partial lockdown, and 0.90 during post-COVID. Furthermore, the ∑A/∑C ratios during the COVID partial lockdown and post-COVID periods were 1.38 and 1.40 times higher than those during the pre-COVID period, which suggests that the particles during the COVID partial lockdown and post-COVID periods were more acidic than those during the pre-COVID period. This is due to the promotion of the formation of secondary inorganic ions (NO\(_3^-\) and SO\(_4^{2-}\) ), which could enhance the acidity of particles (Tian et al., 2020). In the present study, the unmeasured anions, such as bicarbonate, organic ions (formate and acetate), F\(^-\), NO\(_2^-\), PO\(_4^{3-}\), and Br\(^-\), lead to deficiency of the total anions (∑A), and consequently increase the deviation of ion balances to the lower side of the theoretical line (1:1) during the three periods of study. As a result, the situation of ion balance may be changed to the above reference theoretical 1:1 (∑A: ∑C) line (acidity) when the ∑A includes these unmeasured anions.

### 3.2.2.5. Acidity of atmospheric aerosol PM\(_{2.5}\)

Based on the E–AIM-IV model used in the present study for estimating the aerosol acidity, the mean values of the total acidity (HI\(^+\)) during the three sampling periods showed significant variations, with higher values during both the
post-COVID (0.09 mmol/L), COVID-partial lockdown (0.07 mmol/L) than pre-COVID (0.04 mmol/L) period. On the other hand, the mean values of in situ pH estimated from the E-AIM model were following the pattern: pre-COVID (2.03) > COVID-partial lockdown (1.61) > post-COVID (1.53), which suggested that both the COVID-partial lockdown and post-COVID aerosols were more acidic than the pre-COVID aerosol. Moreover, the higher levels of [H+] during both the post-COVID and COVID-partial lockdown could be explained based on the enhanced levels of SO$_4^{2-}$ and NO$_3^-$; The total acidic nature of the aerosols depends on the levels of SO$_4^{2-}$ and NO$_3^-$ species (Bherera et al., 2013; Xue et al., 2011). The total acidity [H+] is influenced by emission sources of SO$_2^+$ and ambient atmospheric chemistry (Nenes et al., 1998; Xue et al., 2011).

The estimated values of in situ pH ranged from 1.54 to 2.03, in the present study, were comparable to those observed in the Mediterranean and eastern parts of European countries (1–4, Kakavas et al., 2021), in a rural site in southeastern US (0–2, Battaglia et al., 2019; Weber et al. (2016)) but lower than those observed in North China Plain (4.2, 2016), in Central and northern European countries (3–5, Kakavas et al., 2021) and in a suburban site of Nanjing (2–4, Gong et al. (2021)). Based on the finding of the present field study and the other field studies around the world, policymakers across the world need to set up measures for decreasing the emission sources that are responsible for acid deposition (pH < 5.6) from the atmosphere in wet or dry forms influencing the ecosystem.

3.2.2.6. Factors affecting the neutralization of atmospheric aerosol PM$_{2.5}$. Fig. 3 shows the NFs for NH$_4^+$, Ca$_{2+}$, and Mg$_{2+}$ during the pre-COVID, COVID partial lockdown, and post-COVID periods. NH$_4^+$ has the highest NF followed by Ca$_{2+}$ and Mg$_{2+}$. NFs were 0.47, 0.46, and 0.44 for NH$_4^+$, 0.34, 0.18, and 0.23 for Ca$_{2+}$, and 0.14, 0.09, and 0.1 for Mg$_{2+}$ during the COVID partial lockdown, and post-COVID periods, respectively. This indicates that NH$_4^+$ is the principal component of the neutralization of acidic components in atmospheric PM$_{2.5}$ during the three different study periods. Moreover, the relatively lower NFs for Ca$_{2+}$ and Mg$_{2+}$ during the COVID partial lockdown period than those during the pre-COVID period may be attributed to the decline in their concentrations in PM$_{2.5}$ during the COVID partial lockdown period.

4. Conclusions and recommendations

In this study, the influence of the partial lockdown due to COVID-19 on the chemical characterization of atmospheric PM$_{2.5}$ in a Greater Cairo suburb was investigated. Our results reveal that reductions in vehicular emissions were responsible for the decline of PM$_{2.5}$ (39.0%), elements (38–55%), NO$_2$ (38.1%), and SO$_2$ (32.9%) during the COVID partial lockdown. Relatively lower reductions in the secondary inorganic ions (SO$_4^{2-}$ (9%) and NO$_3^-$ (14%)) were observed compared with their precursors (NO$_2$ and SO$_2$) in the COVID partial lockdown. Although the concentrations of particulate NO$_3$ and SO$_4^{2-}$ in PM$_{2.5}$ decreased during the COVID partial lockdown compared with the pre-COVID period, their production rates were higher during the COVID partial lockdown compared with the pre-COVID period. Moreover, the secondary inorganic ions (SNA) were the main dominant components in PM$_{2.5}$ during the COVID partial lockdown. These results suggest that the secondary aerosol productions were enhanced during the COVID partial lockdown compared with the pre-COVID period. This enhancement, which partially offset the reduction in PM$_{2.5}$, may be a result of the increased oxidizing capacity of the atmosphere under the effect of reduced emissions. On the other hand, the relatively higher production rates of NO$_x$ and SO$_2^+$ during the COVID partial lockdown confirmed the occurrence of high photochemical reactions and oxidants formation that increase the atmospheric oxidizing capacity. The enhancement of the conversion rate of SO$_2$ to SO$_4^{2-}$ needs high relative humidity besides the presence of gas-phase reactions, whereas the gas-phase reactions are predominant for the oxidation processes of NO$_x$ to NO$_3^+$. The enhancement of secondary aerosol formation is important for increasing the atmospheric PM$_{2.5}$; SO$_4^{2-}$ existed as (NH$_4$)$_2$SO$_4$, (NH$_4$)$_2$SO$_4$.CaSO$_4$.2H$_2$O, and CaSO$_4$ during the three periods of study, whereas NO$_x$ existed as NH$_x$NO$_3$ in the pre-COVID period and was present in other different forms like Mg(NO$_3$)$_2$, KNO$_3$, and Ca(NO$_3$)$_2$ during the COVID partial lockdown and post-COVID periods. PM$_{2.5}$ in the COVID partial lockdown and post-COVID periods was more acidic than that in the pre-COVID period, and NH$_4^+$ was the main neutralizing agent of acidic components in atmospheric PM$_{2.5}$. Finally, this study focused on the side effects of intense anthropogenic emissions reduction, especially NO$_x$, and the nonlinear response of chemical composition and characterization of PM$_{2.5}$. Control strategies for secondary SO$_4^{2-}$ and NO$_3^-$ pollution need not only reduction of NO$_x$, but also reduction of the key (oxidants) that leads to an increase in atmospheric oxidation capacity, which is involved in the formation of these secondary pollutants. Therefore, developing coordinated control strategies for multi-type pollutants is needed to minimize the side effects caused by emission reduction.

Credit author statement

Salwa K. Hassan: Provide the idea of the study and designed research, validation, project administration, supervision, discussed the results and writing-reviewing and editing; Mansour A. Alghamdi: investigation, validation, interpreted the results, discussed the results and writing-reviewing and editing; Mamdouh I. khodr: Provide the idea of the study and designed research, investigation, validation, interpreted the results, discussed the results and writing- original draft preparation.

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.apr.2022.101587.

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