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# Submicron aerosol pollution in Greater Cairo (Egypt): A new type of urban haze?

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**Abstract.** Greater Cairo, the largest megacity of the Middle East North Africa (MENA) region, is currently suffering from major aerosol pollution, posing a significant threat to public health. However, the main sources of pollution remain insufficiently characterized due to limited atmospheric observations. To bridge this knowledge gap, we conducted a continuous 2-month field study during the winter of 2019-2020 at an urban background site, documenting for the first time the chemical and physical properties of submicron (PM<sub>1</sub>) aerosols. Crustal material from both desert dust and road traffic dust resuspension contributed as much as 24% of the total PM<sub>1</sub> mass (rising to 66% during desert dust events), a figure not commonly observed in urban environments. Our observations showed significant decreases in black carbon concentrations and ammonium sulfate compared to data from 15 years ago, indicating an important reduction in both local and regional emissions as a result of effective mitigation measures. The diurnal variability of carbonaceous aerosols was attributed to emissions emanating from local traffic at rush hours and nighttime open biomass burning. Surprisingly, semi-volatile ammonium chloride (NH<sub>4</sub>Cl) originating from local open biomass and waste burning was found to be the main chemical species in PM<sub>1</sub> over Cairo. Its nighttime formation contributed to aerosol water uptake during morning hours, thereby playing a major role in the build-up of urban haze. While our results confirm the persistence of a significant dust reservoir over Cairo, they also unveil an additional source of highly hygroscopic (semi-volatile) inorganic salts, leading to a unique type of urban haze. This haze, with dominant contributors present in both submicron

(primarily as  $\text{PM}_{2.5}$ ), and deposition (largely as dust) modes, underscores the potential implications of heterogeneous chemical transformation of air pollutants in urban environments.

## 1. Introduction

The growth of urban populations and the consequent rise in anthropogenic activities lead to deteriorating air quality, which, in turn, has significant direct and indirect adverse impacts on public health and climate. According to the World Health Organization (WHO, 2021) and the European Environment Agency (EEA, 2023), short and long-term exposure to high concentrations of gaseous and particulate air pollutants is related to various health issues, such as irritated eyes, headaches, asthma, strokes and chronic diseases of the cardio-respiratory system (e.g., Lelieveld et al., 2019). Several epidemiological studies have demonstrated the impact of particulate matter (PM) on human health with increased mortality and morbidity (Ain & Qamar, 2021; Arias-Pérez et al., 2020; Dockery & Pope, 1996; Kelly & Fussell, 2012; Marey et al., 2010; Martinelli et al., 2013). All these effects are likely to be exacerbated in megacities, which concentrate both high pollution levels and high exposure levels to this pollution.

Monitoring and characterizing the sources and properties of particulate matter and gaseous pollutant levels in megacities is consequently critical for a better assessment of their health impacts and also for evaluating the efficiency of applied air pollution mitigation measures, including new strategies to control emissions from specific sources.

The WHO's revised air quality guidelines established more stringent annual ( $5 \mu\text{g m}^{-3}$ ) and 24-hour ( $15 \mu\text{g m}^{-3}$ ) limits for  $\text{PM}_{2.5}$  that reflect the evolving understanding of their health impacts at lower concentrations (WHO, 2021). Yet, there's a notable absence of  $\text{PM}_1$  regulations, highlighting a gap in public health policy. Addressing this gap by expanding research and regulations to include  $\text{PM}_1$  is crucial for protecting public health from air pollution.

The focus on  $\text{PM}_1$  has been limited, despite evidence pointing to its severe health effects, partly due to the scarcity of measurements. Emerging studies highlight the urgent need to address  $\text{PM}_1$  in epidemiological research, showing strong associations between fine particles and lung cancer mortality, which emphasizes the danger of even smaller particles (G. Chen et al., 2017; Franck et al., 2011; Lin et al., 2016).

The Eastern Mediterranean and Middle East (EMME) region has been identified as an air pollution hotspot (Osipov et al., 2022) with major influence of long-range transported air masses originating from three continents, Europe, Asia (Middle East), and Africa (Lelieveld et al., 2002), therefore comprising a mixture of natural and inadequately controlled anthropogenic emissions (Kanakidou et al., 2011; Lelieveld et al., 2002; Pikridas et al., 2018; Vrekoussis et al., 2005, 2022). The EMME experiences enhanced regional background pollution levels, particularly during summer, leading to exceedances of ozone and PM thresholds (Gerasopoulos et al., 2006; Kanakidou et al., 2011; Kleanthous et al., 2014). Therefore, air quality in EMME cities is not only impacted by local emissions but also by its regional background levels (e.g., Christodoulou et al., 2023) and intense pollution episodes emanating from distant sources. Eventually, the adverse health impact of air pollution in EMME cities may also be amplified by extreme weather events (e.g., heat waves). The latter is of significant importance as the EMME region is identified as a major hotspot for climate change (Mostafa et al., 2019; Zittis et al., 2019, 2022).

The Greater Cairo (GC) megacity, the capital of Egypt, stands as the largest city in the EMME region. Its population exceeds 22 million inhabitants according to the Central Agency for Public Mobilization and Statistics (CAPMAS), contributing to a national population of approximately 110 million. In such

gigacity regions, and from time to time, megacity air pollution can strongly influence the planetary boundary layer (PBL) (Ding et al., 2016; Huang et al., 2020; Z. Wang et al., 2018), resulting in the formation of extensive heat and pollution islands (Kulmala et al., 2021). A characteristic example of such conditions is the Indo-Gangetic Plain (IGP), where numerous observational studies focusing on surface PM concentrations have been conducted over India during the past decade. For instance, Gunthe et al. (2020 and references therein) reported persistently high chloride in Delhi and episode spikes in chloride concentrations in Chennai. These authors suggested that in the presence of excess ammonia in Delhi, high local emission of hydrochloric acid partitions into aerosol water. The highly water-absorbing and soluble chloride in the aqueous phase substantially enhances aerosol water uptake through co-condensation, which sustains particle growth, leading to haze and fog formation. Several rapidly urbanizing areas in Africa (like Cairo), are likely to undergo similar developments (G. Chen et al., 2020; Hoornweg & Pope, 2017; Ramaswami et al., 2016). However, detailed studies on the chemical composition of fine particles are missing at these gigacities.

According to the report by the World Bank (2012), Cairo is recognized among the most polluted cities globally, while Cheng et al. (2016) ranked Cairo as the second most heavily polluted city concerning  $PM_{2.5}$  levels among 45 megacities, following Delhi (India). Most of the studies conducted over the past decades, in Cairo have provided only partial information on the PM levels, composition, and sources (Abbass et al., 2020; Abu-Allaban et al., 2002, 2007; El-Metwally et al., 2008; Favez et al., 2008; Zakey et al., 2008) with very few documenting all the major PM chemical components such as carbon, ions, and trace metals (e.g. Abu-Allaban et al., 2002; 2007; Favez et al., 2008; Lowenthal et al., 2013). Moreover, most of these measurements were performed 10 to 20 years ago. Therefore, they can no longer account for the current air pollution in Cairo, and assess the efficiency of new environmental policies and the adoption of cleaner technologies, alongside the substantial growth in population over recent years (Heger et al., 2019).

To the best of our knowledge, there have been no studies conducted in Greater Cairo concerning the submicron aerosol ( $PM_1$ ), which is a crucial component of PM contributing to haze formation with severe health implications (G. Chen et al., 2017; Y. Hu et al., 2022; Gunthe et al., 2021). In addition, all PM measurements conducted so far in GC have used filters to collect PM, which are known to suffer from sampling artifacts (e.g., absorption/desorption of semi-volatile aerosol species) and lack of high-time resolution (hour or less) that can provide valuable insights on the diurnal variability of PM components and the dynamic of their emission sources.

To advance this limited knowledge on the actual chemical composition and sources of PM in GC and better assess its influence on air quality and health we conducted the field campaign POLCAIR (Air Pollution in Greater Cairo: sources and impact) project. This two-month intensive field campaign, spanning from November 28, 2019, to January 29, 2020 (prior to the COVID-19 lockdown), was carried out at a residential site representative of urban background conditions in GC. While, both online and offline measurements of the chemical and physical properties of aerosols were performed in this campaign, this work is more focused on the chemical composition and hygroscopicity of sub-micrometer aerosols in order to better assess their haze formation potential.

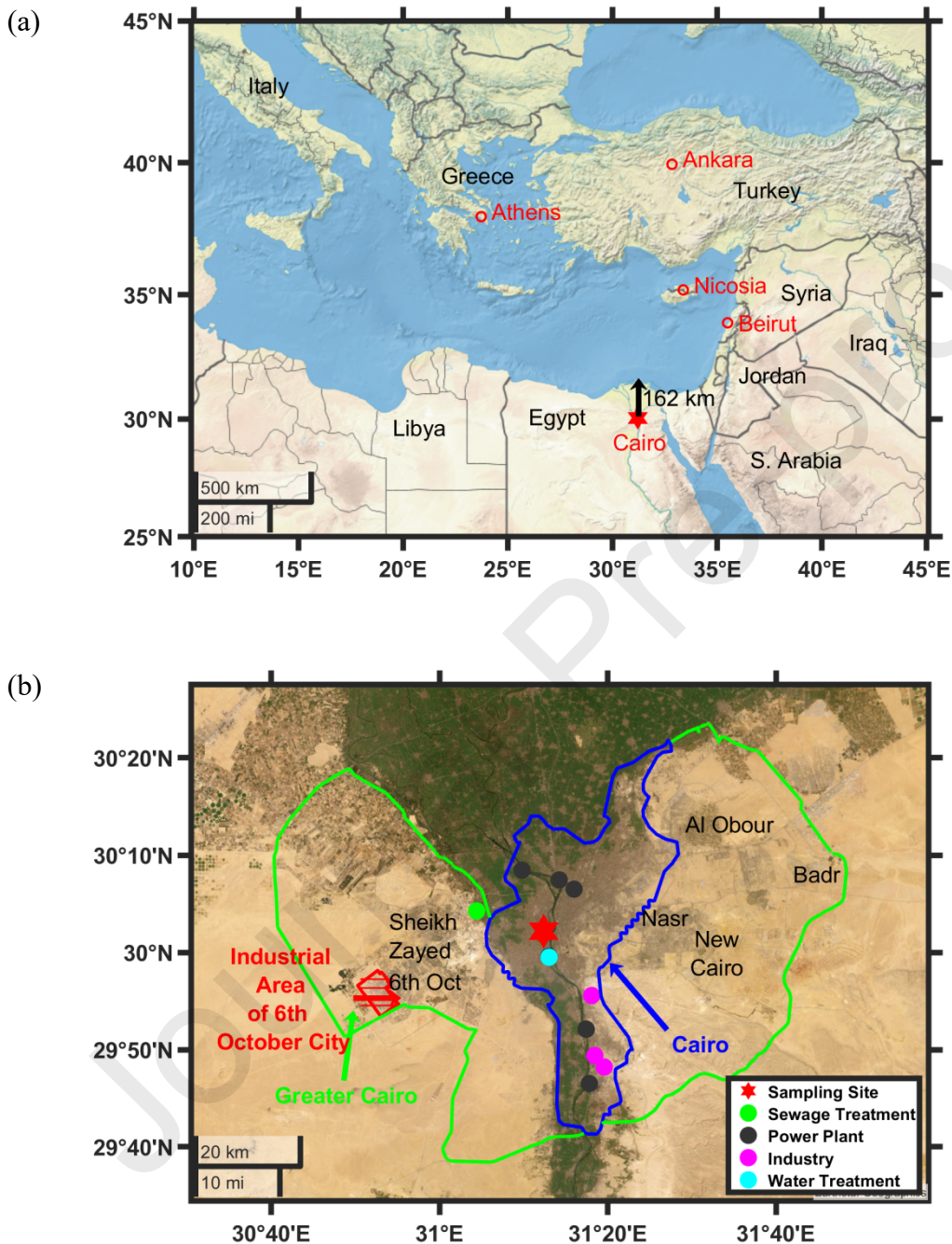
## 2. Methodology

### 2.1 Site Description

Atmospheric measurements in the frame of the POLCAIR project were performed in Cairo, the capital city of the Arab Republic of Egypt, located at the base of the Nile Delta, (Fig. 1a).

A comprehensive suite of state-of-the-art aerosol instrumentation was deployed at the premises of the National Research Centre of Egypt (NRC; 30.0358° N, 31.2051° E). The sampling site is located approximately 3.5 km southwest of the city center (Fig. 1b), 1 km west of the Nile river, and 1 km north

Of the same university, the latter being used as a sampling site by Farouk et al. (2008). The instruments were installed on the rooftop of a six-floor building with a clear (360°) unobstructed view. The site was chosen as representative of the Greater Cairo urban area (Fig. 1b), where urban-related emission sources can be observed with minimal local traffic contamination (the closest major roads being located at 250 m distance).



**Figure 1:** (a) Location of the Greater Cairo megacity in the Eastern, (b) Location of the measurement site (red star) within the Cairo (blue line) and Greater Cairo (green line) urban area. Location of major sewage and water treatment plants, power plants, and industries are also shown (Source: google maps)

The field campaign started on November 27<sup>th</sup>, 2019, and ended on January 29<sup>th</sup>, 2020, resulting in more than two consecutive months of continuous observations (80-95% coverage) before the COVID-19 lockdown (Spring of 2020) and Muslim Ramadan (23<sup>rd</sup> April to 23<sup>rd</sup> May 2020). The timing of the campaign, which mostly covered the winter season, was chosen as a period of the year with minimal influence of biomass burning, which occurs in the Nile Delta around October (Mahmoud et al., 2008; Marey et al., 2010; Prasad et al., 2010) and dust storms which typically occur in Spring (El-Metwally et al., 2011). Therefore, the observations are expected to enable an optimal characterization of local city emissions with potentially less interference from nearby emission sources.

### 2.3 Instrumentation

The instrumentation deployed during the campaign is described in Table 1 for each aerosol parameter, along with the related instrument/technique, averaged time resolution, and period of operation. Reactive and greenhouse gas measurements were also performed to identify their primary/secondary sources and will be further exploited in a separate publication.

**Table 1: Aerosol Measurements and Instruments at the sampling site.**

Parameter	Instrument	Time resolution	Measuring Period (data points /samples, data coverage)
Chemical Characterization of Non-Refractory PM <sub>1</sub>	Q-ACSM	29 min	28/11/2019 – 29/01/2020 (3019 data points, 100%)
Black Carbon (BC)	Aethalometer A33/	1 min	28/11/2019 – 29/01/2020 (89k data points, 100%)
PM <sub>1</sub> Filter sampling (EC/OC, Ions, Metals)	Leckel SEQ 47/50	6 h	02/12/2019 – 28/01/2020 (225 filter samples, 85%)
Submicron aerosol Hygroscopic properties	HTDMA	6 min	04/12/2019 - 28/01/2020 (19265 hygr. distributions / 5 dry mobility diameters, 60%)
Submicron particlemobility size distribution	SMPS	5 min	04/12/2019 - 28/01/2020 (12056 size distributions, 69%)

Meteorological Parameters	Vaisala Weather station WXT 536	5 min	01/12/2019 – 28/01/2020 (16k data points, 95%)
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**Submicron aerosol composition (NR-PM<sub>1</sub>: Non-refractory PM<sub>1</sub>).** Measurements were performed continuously using a quadrupole aerosol chemical speciation monitor (Q-ACSM, Aerodyne Research Inc., USA). The instrument provides quantitative information for non-refractory species such as organics, nitrate, sulfate, ammonium, and chloride in the size range of ca. 40–1300 nm; 90 – 650 nm with more than 50% penetration efficiency vacuum aerodynamic diameter; (Liu et al., 2007; Ng et al., 2011). Data were retrieved using ACSM local v.1.6.0.3 software, implemented in Igor Pro (v. 6.37, Wavemetrics Inc., USA). To account for incomplete detection caused by particle bounce, the mass concentrations were corrected using the chemical composition-dependent collection efficiency (CDCE) method (Middlebrook et al., 2012). The corresponding parameters, response factor (RF), and relative ionization efficiency (RIE) are reported in Table S1. More details about the Q-ACSM setup, operation, and QA/QC protocol used here can be found in Christodoulou et al. (2023).

**Black Carbon (BC):** Continuous black carbon (BC) mass-equivalent concentration measurements were obtained using a dual spot, 7- $\lambda$  Magee Scientific AE-33 aethalometer (Drinovec et al., 2015), operated at a flow rate of 3 L min<sup>-1</sup> and a 1-min time resolution. The “Aethalometer model” (Sandradewi et al., 2008) was used to allocate contributions of biomass burning (BC<sub>bb</sub>) and fossil fuel combustion (BC<sub>ff</sub>) to BC concentrations. The model calculates the fractional biomass burning contribution based on light absorption coefficients recorded at 470 and 950 nm and assuming instrument-default Ångström Absorption Exponents of 1.0 and 2.0 for fossil fuel combustion and biomass burning, respectively. The instrument was equipped with a PM<sub>1</sub> cut-off inlet (BGI Inc., USA) and a Nafion dryer.

**Aerosol number size distribution:** The aerosol number size distribution in the range of ca. 14–680 nm was measured by a commercially available SMPS system (S. C. Wang & Flagan, 1989), consisted of a long DMA (TSI Inc. model 3081), its control unit (TSI Inc. model 3080) and a condensation particle counter (CPC; TSI Inc. model 3025; Stolzenburg & McMurry, 1991). The system was operated at a sheath-to-aerosol ratio of 10:1 (i.e., 3 and 0.3 L min<sup>-1</sup>, respectively) while being equipped with a silica diffusion dryer and a soft X-ray neutralizer (TSI Inc. model 3088) to ensure adequate drying and neutralization of the sampled aerosols. The raw measurements were inverted into number-size distributions with a resolution of 64 channels/decade using the manufacturer’s software (TSI AIM ver. 9c), also used for multiple charge correction.

**Aerosol hygroscopicity:** A custom-made HTDMA (Rader & McMurry, 1986) system, similar to the one described in more detail in Salmatonidis et al. (2020), was employed for measuring the hygroscopic growth factors of monodisperse aerosols having electrical mobility diameters of 30, 60, 90, 120 and 160 nm (cf. supplement Section1 for more details). The sampling interval was 3 min for each monodisperse dry electrical mobility diameter, while the total measuring cycle for all 5 dry mobility diameters was 30 min (i.e., two consequent measurements for each dry mobility diameter).

The inverted size distributions were fitted with a custom-made algorithm (Bezantakos et al., 2013) to calculate the geometric mean electrical mobility diameter of the humidified particles, which in turn was used for deriving their hygroscopic growth factor (cf. supplement S1 for more details). In addition, the algorithm is capable of distinguishing between internally and externally mixed aerosol populations (pronounced by wider/bimodal distributions at elevated RH conditions).



The measured hygroscopic growth factors were then converted to the respective single hygroscopic parameters “ $\kappa$ ”. Note that in the case of externally mixed aerosols, more than one “ $\kappa$ ” value can be calculated, with each one denoting the hygroscopicity of each population. Sampled particles were conditioned and charged by the dryer and the neutralizer of the SMPS.

**PM<sub>1</sub> Filter collection for off-line chemical composition determination:** Off-line measurements were carried out to collect particles every 6 hours (4 samples per day from midnight local time) onto 47-mm diameter quartz filters (Pall Gellman, TISSUQUARTZ) using a low-volume sampler (Leckel SEQ47/50) operating at a flow rate of 2.3 m<sup>3</sup> h<sup>-1</sup>. During the 2-month campaign period, 225 filters were collected continuously. The elemental/organic carbon (EC/OC) content was analyzed using a Sunset OC/EC lab analyzer following the EUSAAR2 thermal protocol described in (Cavalli & Putaud, 2010). Major anions and cations were analyzed using ion chromatography (Bimenyimana et al., 2023; Sciare et al., 2008) after extraction of filter punches in ultra-pure water. PM<sub>1</sub> samples were additionally analyzed for trace elements (A, Fe, Ca, Zn, Pb, Cu, Ni, V, Cr, Mn, Cd, and As) by Inductively coupled plasma mass spectrometry (ICP-MS) based on the method described in Bimenyimana et al. (2023).

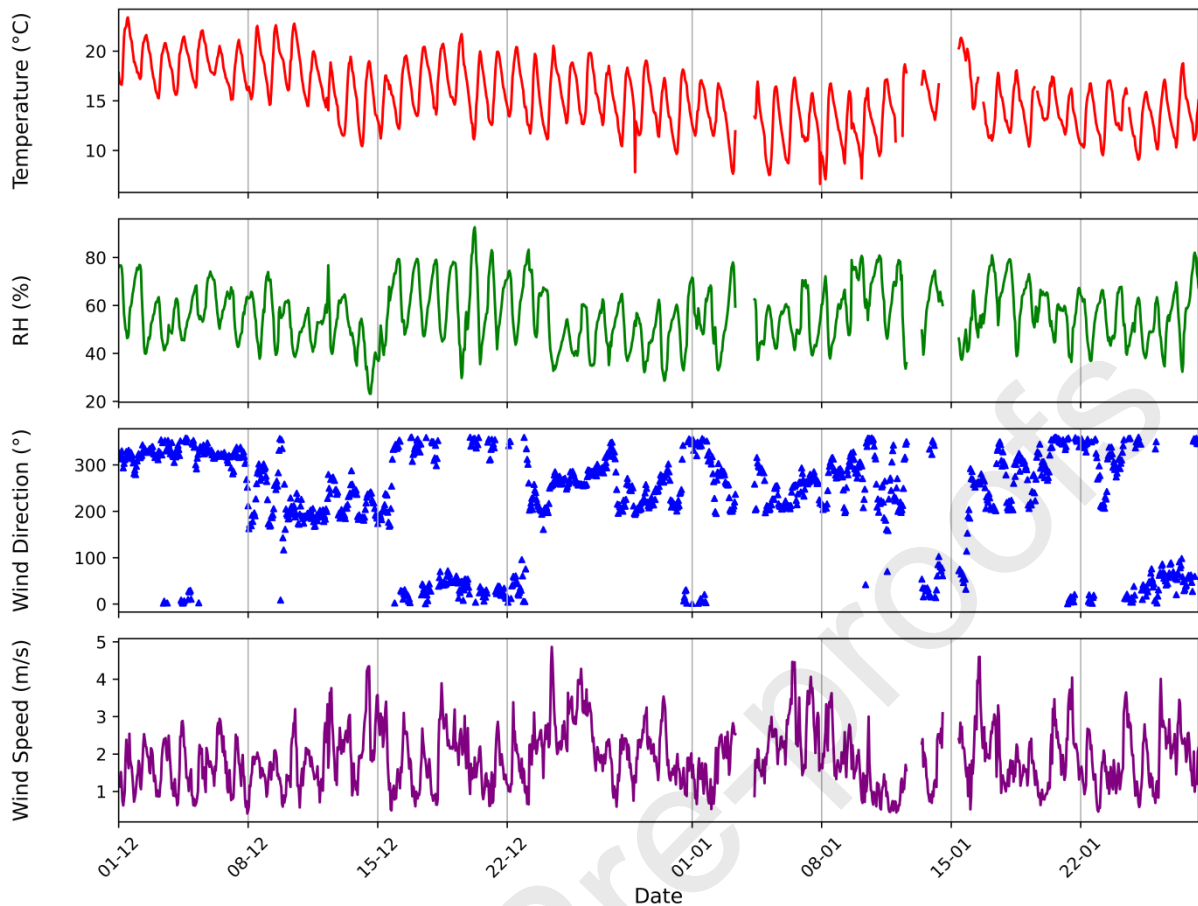
**Meteorological parameters:** Standard meteorological parameters (temperature, relative humidity, wind speed, and direction) were obtained from a meteorological station located near the sampling station.

**Air masses back-trajectory analysis:** Air mass 72-hour retroplumes were modeled using the FLEXible PARTicle (FLEXPART) dispersion model (Pisso et al., 2019) in backward mode. Particles (n=40,000) were released from 350 m a.g.l. and followed back in time for 3 days (Stohl et al., 2002) using as input three-dimensional forcing datasets from the NOAA Climate Forecast System (CFS) short-duration ( $t < 6$  h) forecasts (Saha et al., 2014). In this work, only the lower 100 m a.g.l., as representative of the interaction of the receptor site with local sources, were considered.

### 3. Results and Discussion

#### 3.1 Meteorological conditions

During the two months of the POLCAIR campaign, the meteorological conditions encountered were relatively stable from one day to another and variable within the day (see Fig. 2 & Fig. S1). Although the campaign was performed during winter, the sampling period was characterized by mild and typical temperatures for the period (on average at  $15.4 \pm 3.2$  °C) (The World Bank Group, 2021). The observed range of Relative Humidity (RH) varied from 45 to 75% over the day, being atypically high for a city located near desertic regions. However, this can be explained for Cairo by the presence of the Nile river and intense agriculture activities (incl. irrigation) in the Nile delta, which, together with elevated temperatures, promote intense evaporation. These humid conditions are likely to promote condensation of liquid water onto hygroscopic aerosols and, therefore, contribute to the degradation of visibility, which is often observed over Cairo over the first hours of the day (Fig. 3 & Fig. S1).



**Figure 2:** Temporal variability of temperature, RH, Wind speed, and Wind direction during the POLCAIR campaign.

The sector distribution of winds for the campaign duration, depicted in Fig. S2, clearly shows the predominance of NW winds during most days, followed by W and SW directions mainly during the night and until sunrise. Exceptions to this pattern occurred only on the few days when the wind direction was pointing from the sector between N and NE (Fig. 2).



**Figure 3:** South view of the sampling site in the morning and afternoon, highlighting the diurnal change of visibility along the day.

Figure S3 shows the seven clusters of back-trajectory classes calculated for the 2-month campaign. During this period, GC was mostly influenced by air masses originating from southeast Mediterranean Sea (cluster 1-2 with contribution 13-22% respectively) (Fig. S4a), northeast Mediterranean Sea (Aegean Sea, close to Turkish coastline, west of Turkey) (cluster 3-4 with respective contribution 22 – 14 %) (Fig. S4b) and the central Mediterranean Sea, south of Italy (cluster 7 contributing 9 %) (Fig. S4c). Although the major air masses arriving in Cairo originated from the Mediterranean Sea, two important clusters were identified originating from northwest Africa (clusters 5 and 6 with relative contribution of 11 and 9 %, respectively) and mostly covered by the Saharan Desert (Fig. S4d). In the following sections, the analysis of these two distinctive air mass episodes will further be explored.

Apart from the previously discussed seven clusters, it's worth noting that specific periods exhibited predominant air masses originating from the Middle East, encompassing countries like Lebanon, Syria, Iraq, Israel, Saudi Arabia, and Turkey (Fig. S4 a-d), and were associated with prevailing north-to-northeast winds passing over the Nile Delta region (Fig.2). These events will also be further discussed in the following.

### 3.3 Fine (PM<sub>1</sub>) aerosol chemical composition

#### 3.3.1 Comparison between online and offline measurements of individual chemical species

Ammonium, sulfate, nitrate, and chloride concentrations measured with the Q-ACSM (i.e., in the PM<sub>1</sub> size fraction) were averaged throughout the sampling duration of the off-line PM<sub>1</sub> (filter) measurements. Direct comparisons were made, resulting in correlations with more than 200 data points covering a wide concentration range of at least an order of magnitude (Fig. S5 a-e). Overall, all elements show a very strong correlation, including chloride, which is usually underestimated by filter sampling because of the volatility of NH<sub>4</sub>Cl (Budisulistiorini et al., 2014; Witz et al., 1990). The observed slopes were 1.13 with R<sup>2</sup> = 0.93 for SO<sub>4</sub><sup>2-</sup>, 1.03 with R<sup>2</sup> = 0.94 for NH<sub>4</sub><sup>+</sup>, 1.11 with R<sup>2</sup> = 0.96 for NO<sub>3</sub><sup>-</sup> and 1.04 with R<sup>2</sup> = 0.89 for Cl<sup>-</sup>. Measurements marked with a circle (sulfate in Fig. S5b and organics in Fig. S5a) indicate potential interference from dust concentrations and correspond to values where calcium concentrations exceed 6 µg m<sup>-3</sup>.

The concentrations of organic aerosols (OA) are also compared to the OC levels determined on PM<sub>1</sub> filters. An excellent agreement is also found here (R<sup>2</sup> = 0.89), with a slope of 1.6, similar to the one used to reconstruct chemically the PM<sub>1</sub> mass, and being close to the values reported by Christodoulou et al. (2023) for Nicosia (Cyprus) during wintertime and OM/OC calculations from AMS measurements in polluted environments (Saarikoski et al., 2012).

Given the excellent agreement between the Q-ACSM observations (NR-PM<sub>1</sub> components) and PM<sub>1</sub> filters, in the following discussion inorganic and OM levels derived from Q-ACSM will be used, while PM<sub>1</sub> filters will be used for the estimation of dust and trace elements.

#### *Dust calculation*

The calculation of fine mode crustal mass (dust) was done by using the trace elements levels determined on PM<sub>1</sub> filters (Table S2) and by implementing the IMPROVE formula (Malm et al., 1994), similar to other studies (e.g., Pant et al., 2015). This formula uses the oxides of the main soil elements such as Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, CaO, FeO, Fe<sub>2</sub>O<sub>3</sub>, and TiO<sub>2</sub> to calculate dust concentration as follows:

$$[\text{Dust}] = 2.20*[\text{Al}] + 2.49*[\text{Si}] + 1.63*[\text{Ca}] + 2.42*[\text{Fe}] + 1.94*[\text{Ti}] \text{ (Eq. 2)}$$

Simulation (Eq. 2) cannot be directly measured by filter and was estimated here using an  $\alpha$  of 0.45 (Simon et al., 2011). Figure 4 represents the reconstructed chemical composition in  $PM_1$ . On average, the fractions of major chemical species followed the order: ions (mainly sulfate, nitrate, ammonium, and chloride, i.e., SIA), OM, dust, EC, and Trace Elements (TE) accounting for 38 %, 30 %, 24 %, 6 %, 2 % respectively (Fig. 4 second subplot).

Intense dust episodes have the potential to temporarily influence PM concentrations and composition. An important dust event was observed during the campaign between 25-26/12, marked by a peak in  $PM_{1chem}$  concentration of almost  $150 \mu g m^{-3}$  (Figure 4). During this period, dust contributed to ca. two-thirds of the total  $PM_{1chem}$ . This result is noteworthy as it showcases that dust can become the predominant species in  $PM_1$  in Cairo, a fraction that otherwise is usually attributed to anthropogenic emissions.

However, removing the contribution of this ‘major’ dust event, the relative contribution of the various chemical species to  $PM_1$  is only marginally changed by 2-3%, with dust remaining the third contributor to  $PM_1$  load. This finding indicates that dust is likely to be ubiquitous in Cairo even in  $PM_1$ .

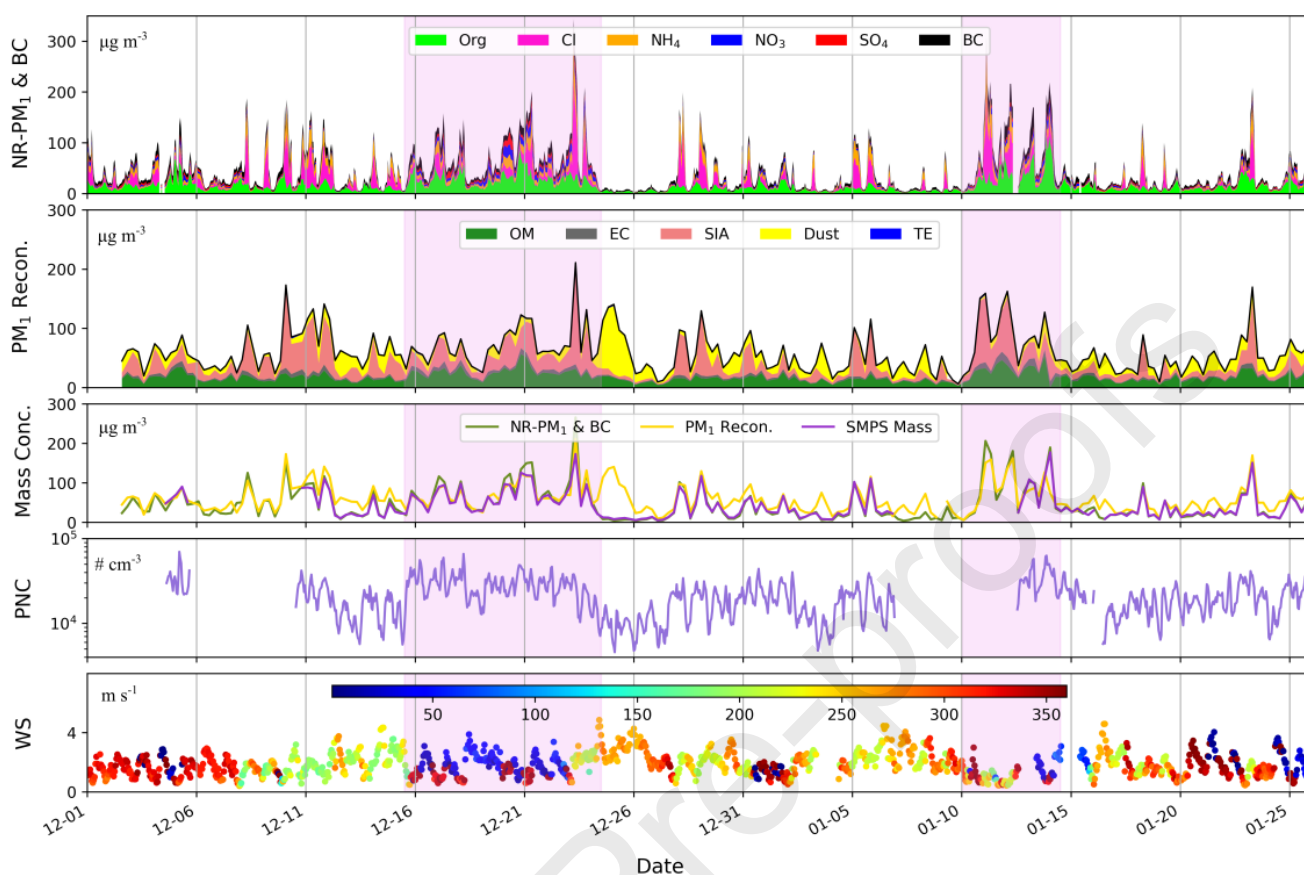
**Trace Elements:** Data on trace elements measured in this work are depicted in Table S2 and compared with the values reported in the literature in Table S3. It is important to note that due to the absence of previous research focusing on  $PM_1$  in Cairo, comparisons are constrained to studies based on filter  $PM_{2.5}$  and  $PM_{10}$  fractions. Nevertheless, for elements of anthropogenic origin, mainly confined in the fine fraction, such comparisons, despite potential limitations, can offer valuable insights. For instance, for lead (Pb) and cadmium (Cd), a comparison with literature data (Table S3) revealed a decrease of more than an order of magnitude, indicative of effective mitigation strategies applied in the last 20 years. As shown in Table S3, Pb has gradually and continuously decreased by a factor of five in 10 years, from ca.  $1000 ng m^{-3}$  in 2000-2001 to ca.  $200 ng m^{-3}$  in 2010 (Zakey et al., 2008; Lowenthal et al., 2013). These results suggest that Pb has further decreased between 2010 and 2019 (down to  $127 ng m^{-3}$ , this study) although at a slower pace. Levels of Pb, Cd, arsenic (As), and nickel (Ni) levels are comparable to the values reported for GC in the period of 2018 (Monged et al., 2022).

### 3.3.2 Reconstruction of online submicron $PM_1$

The sum of the concentrations of chemical components measured by the Q-ACSM and the BC concentrations measured by the Aethalometer cannot be compared directly with  $PM_{1chem}$  since any of these two online instruments measured dust, which has shown to account on average for 24% of  $PM_{1chem}$ . Alternatively, this sum was compared with PM mass concentration derived by the SMPS. This direct comparison is possible considering the size range of the SMPS (up to  $0.7 \mu m$  diameter) and that of the Q-ACSM, while accounting for the transmission efficiency of the aerodynamic lenses of the Q-ACSM (Liu et al., 2007) and expressing the electrical mobility diameters to vacuum aerodynamic ones. This comparison shows a strong correlation coefficient  $R^2 = 0.95$  and a slope very close to unity (1.09, see Fig. S5f), when an apparent density of the particles of  $1.6 g cm^{-3}$  is assumed, further validating the consistency of online chemical measurements. Additionally and even more importantly, both correlation coefficient and slope suggest that the PM fraction with diameters below  $0.7 \mu m$  does not contain a significant amount of dust, which can be detected by the SMPS if present within its measuring range. This outcome further implies that most of the dust in  $PM_1$  likely falls within the  $0.7-1 \mu m$  range, potentially as a submicron extension of the larger (supermicron) mode.

Figure 4 illustrates the time series of NR- $PM_1$  components (Org, Cl,  $NH_4$ ,  $NO_3$ ,  $SO_4$ ) from Q-ACSM, BC as measured by the aethalometer,  $PM_1$  chemical composition from filters, and the Particle Number

Concentration ( $\mu\text{g m}^{-3}$ ) measured by Q-ACSM, along with meteorological parameters (wind direction and speed) for the duration of the campaign.



**Figure 4:** Time series of 1-hour average NR-PM<sub>1</sub> species measured by the Q-ACSM (first subplot) and Black Carbon concentrations (1-hour average) measured by the aethalometer (black line, first subplot), Time series of PM<sub>1</sub> reconstructed mass measured by filters (6-h average) (second subplot), Mass concentration of NR-PM<sub>1</sub>&BC; PM<sub>1</sub> Reconstructed; SMPS deconvolved mass (third subplot), Particle Number Concentration (PNC) measured by SMPS (logarithmic y-axis, fourth subplot) and Wind Speed (WS) color-plotted with wind direction for the entire measuring period (fourth subplot). Purple-shaded areas refer to the two pollution episodes (PP1 & PP2) observed during the campaign.

### 3.3.3 Concentration of chemical species

The campaign's average for hourly-resolved NR-PM<sub>1</sub> is  $40.05 \pm 40.32 \mu\text{g m}^{-3}$ , ranging from 2 to  $324 \mu\text{g m}^{-3}$  with a median value of  $24.96 \mu\text{g m}^{-3}$  ( $\pm$  indicates the standard deviation reflecting the variability of the measurement over the averaged period). It is interesting to note that the sum of NR-PM<sub>1</sub> ( $40.05 \mu\text{g m}^{-3}$ ) with BC ( $7.25 \mu\text{g m}^{-3}$ ), filter-based dust ( $12.3 \mu\text{g m}^{-3}$ ), and trace elements ( $0.95 \mu\text{g m}^{-3}$ ) gives a total of  $60.6 \mu\text{g m}^{-3}$  which is very close to the  $59.6 \mu\text{g m}^{-3}$  calculated for the reconstructed PM<sub>1chem</sub> over the campaign, further cross-validating both online and off-line measurements.

The hourly maximum NR-PM<sub>1</sub> concentrations were observed on 23<sup>rd</sup> December ( $323 \mu\text{g m}^{-3}$ ). Overall, NR-PM<sub>1</sub> was dominated by organic aerosols (OA), which accounted for 42 % of the total NR-PM<sub>1</sub>, followed by chloride 25 %, ammonium 16.6 %, nitrate 9 %, and sulfate 6 % (Table S4). Table 2 showcases results from studies in other large cities, including those in the East Mediterranean. These studies incorporate both BC and Q-ACSM measurements, allowing for a direct comparison with the present work. The levels observed in our study significantly exceed the WHO guidelines for 24-hour PM<sub>2.5</sub> concentrations. While such high concentrations of PM<sub>1</sub> have been recorded in urban centers in China and

16. However, they are notably lower than those reported in previous studies from this region, as shown in Table 2.

As already emphasized for filter-based OC, the prevalence of submicron OA in Cairo aligns with findings from previous studies conducted worldwide (G. Chen et al., 2022; C. Sun et al., 2016; Zhou et al., 2018). However, reports on EC and OC concentrations in Cairo have been limited, with only two studies by Favez et al (2008) and Abu-Allaban et al. (2007), presenting results from collected filters at TSP and PM<sub>2.5</sub> fractions, respectively. In particular, the study by Favez et al. (2008) provides urban background measurements obtained close to the location of this study and conducted over extended periods, including annual and winter season data for three consecutive years. The findings reveal a distinct declining trend in organic carbon (OC) and elemental carbon (EC) concentrations over 15 years (i.e., by factors of 2 and 3, respectively). Conversely, the levels reported by Abu-Allaban (2007) although at different seasons indicate a significant decrease only for EC (factor of 2).

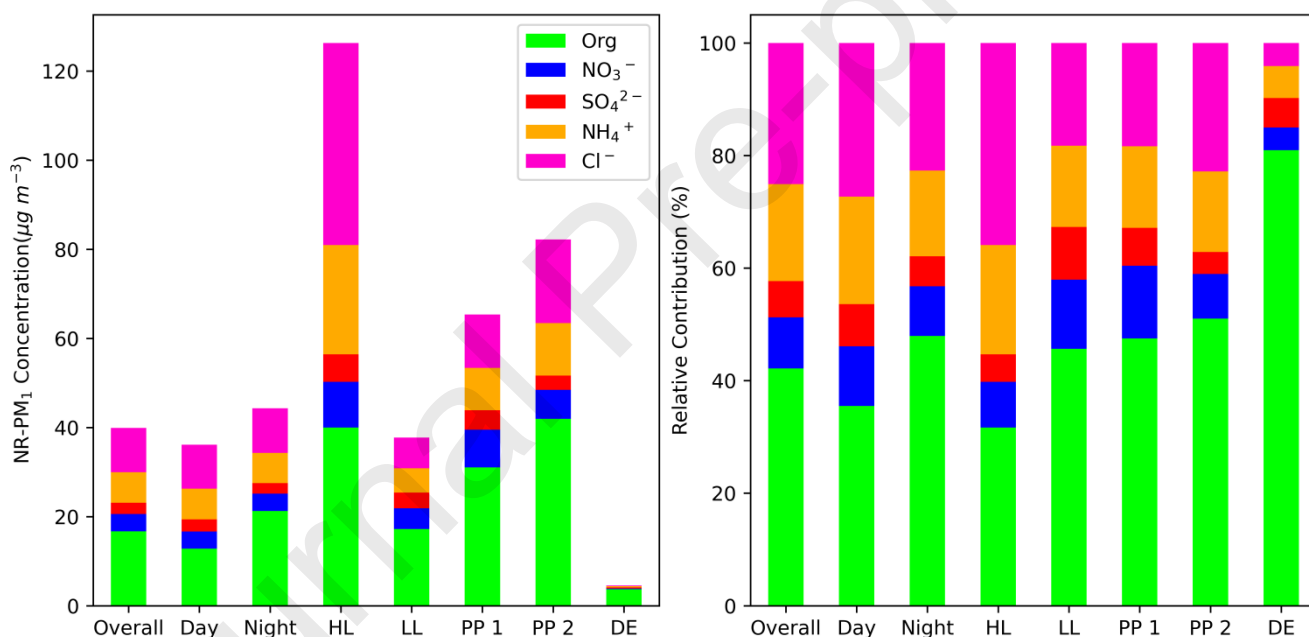
The sulfate concentrations are usually representative of a regional (than local) background. While the measured concentrations are a factor of 2 to 5 lower compared to those reported 20 years earlier in Cairo (Table S5), they are consistent with those observed recently in the Mediterranean (Kaskaoutis et al., 2023). However, the high contribution of NH<sub>4</sub><sup>+</sup> and specifically of Cl<sup>-</sup> is not often observed in the literature. Only a few limited studies in India have reported such high concentrations and contributions of chloride in NR-PM<sub>1</sub> (Gani et al., 2019; Reyes-Villegas et al., 2021) (Table 2). By considering the strong association of NH<sub>4</sub><sup>+</sup> with Cl<sup>-</sup> (R<sup>2</sup> = 0.93), NH<sub>4</sub>Cl contributes to almost half (41%) of the total NR-PM<sub>1</sub> mass, becoming thus the most abundant component even when compared to organics, which represents a huge number of compounds. In addition, the average Cl/Na ratio of 26.55 calculated over the entire measurement period is about 15 times greater than their typical ratio in seawater (1.8) and indicates significant contributions from anthropogenic sources. Interestingly, our non-sea-salt chloride concentrations are similar to those reported by Favez et al. (2008) (all year-round measurements) and Abu-Allaban (2007) for Cairo, suggesting no important decrease in emissions for this compound over the last two decades (see Table S5).

**Table 2:** Comparison of concentrations of the main submicron chemical species derived by ACSM for different cities in the region and the world. BC derived by Aethalometer measurements.

City (Country)	Type of site	Period	Mean [ $\mu\text{g m}^{-3}$ ]							Reference
			OA	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	Cl <sup>-</sup>	NR-PM <sub>1</sub>	BC	
Cairo (Egypt)	Urban	Dec-Jan	16.7	2.6	3.9	6.8	9.9	40.05	7.25	This Study
Beijing (China)	Urban	Jun-Aug	20	9	3.5	5.5	0.5	50.0	-	Sun et al., 2012
Hong-Kong	Urban	Sep-Dec	15	6	1.8	2.8	0.3	25.9	-	Sun et al., 2016

<b>Delhi (India)</b>	Urban	Feb	40	3.9	3.7	4.5	6	58.1	-	R.-Villegas et al., 2021
<b>Delhi (India)</b>	Urban	Winter	112	16	24	20	23	195	15	Gani et al., 2019
<b>Nicosia (Cyprus)</b>	Suburban	Winter	5.03	2.81	1.22	1.14	0.14	10.30	2.01	Christodoulou et al., 2023
<b>Athens (Greece)</b>	Urban	Winter	9	2.5	1.2	0.9	0.15		2.4	Stavroulas et al., 2019

### 3.3.4 Factors controlling the variability of online chemical species



**Figure 5:** NR-PM<sub>1</sub> concentration and relative contribution during different subsets/clusters of the campaign. HL, LL = High and Low Loading periods, PP1, PP2, DE = Pollution episode 1, Pollution episode 2, Dust event, respectively. (High Loading periods : NR-PM<sub>1</sub> mass > 100µg m<sup>-3</sup> & Low Loading periods : NR-PM<sub>1</sub> mass < 100µg m<sup>-3</sup>).

To further examine the influence of sources and chemical processes on the variability and composition of NR-PM<sub>1</sub>, we divided this large dataset into seven clusters using different criteria (Figure 5). The first two subsets of data accounted for day and night time conditions. Daytime, defined as the hours between 6:00 - 18:00 (LT), corresponding to periods of sunshine, while nighttime, from 19:00 to 05:00 LT, encompassed dark hours. Two more data clusters were created to explore the factors controlling the high and low loadings of NR-PM<sub>1</sub>, respectively. The first subset, named 'HL' (High Loading), included NR-PM<sub>1</sub> concentrations exceeding 100 µg m<sup>-3</sup>, while the second subset, 'LL' (Low Loading), included concentrations below 100 µg m<sup>-3</sup>. This differentiation was made due to the presence of shorter pollution episodes lasting only a few hours or up to a day, in addition to the two primary pollution episodes.

Following these, the three clusters/subsets refer to the three pollution episodes (PP1, PP2, DE), and the last subset corresponds to a period impacted by dust.

**DAY-NIGHT:** Daytime and nighttime averages are  $35.67 \mu\text{g m}^{-3}$  ( $\pm 41.89 \mu\text{g m}^{-3}$ ) and  $44.33 \mu\text{g m}^{-3}$  ( $\pm 37.85 \mu\text{g m}^{-3}$ ), respectively (Fig. 5). These datasets show slightly higher NR-PM<sub>1</sub> concentrations at night but the overall chemical composition remains with quite similar contributions of organics and NH<sub>4</sub>Cl (around 40%) and a slight increase of organics over the night.

**HL-LL:** During the periods with high loadings, NH<sub>4</sub>Cl appears as the dominant contributor, with more than half of NR-PM<sub>1</sub>. During the same periods, organics exhibit the lowest relative contribution of around 30%, suggesting that NH<sub>4</sub>Cl is a major driver of PM<sub>1</sub> pollution events in Cairo. Conversely, during periods with low loadings, organics are the dominant species (around 48%), suggesting a permanent (urban) background contribution of carbonaceous aerosols.

**DE:** The dust event shows the lowest NR-PM<sub>1</sub> concentrations among the 7 clusters, which can be partly explained by a higher ventilation rate (wind speed during the dust event was above average; Fig. 4) but also by lower RH, which is likely to play an important role on the formation of NH<sub>4</sub>Cl (see later on).

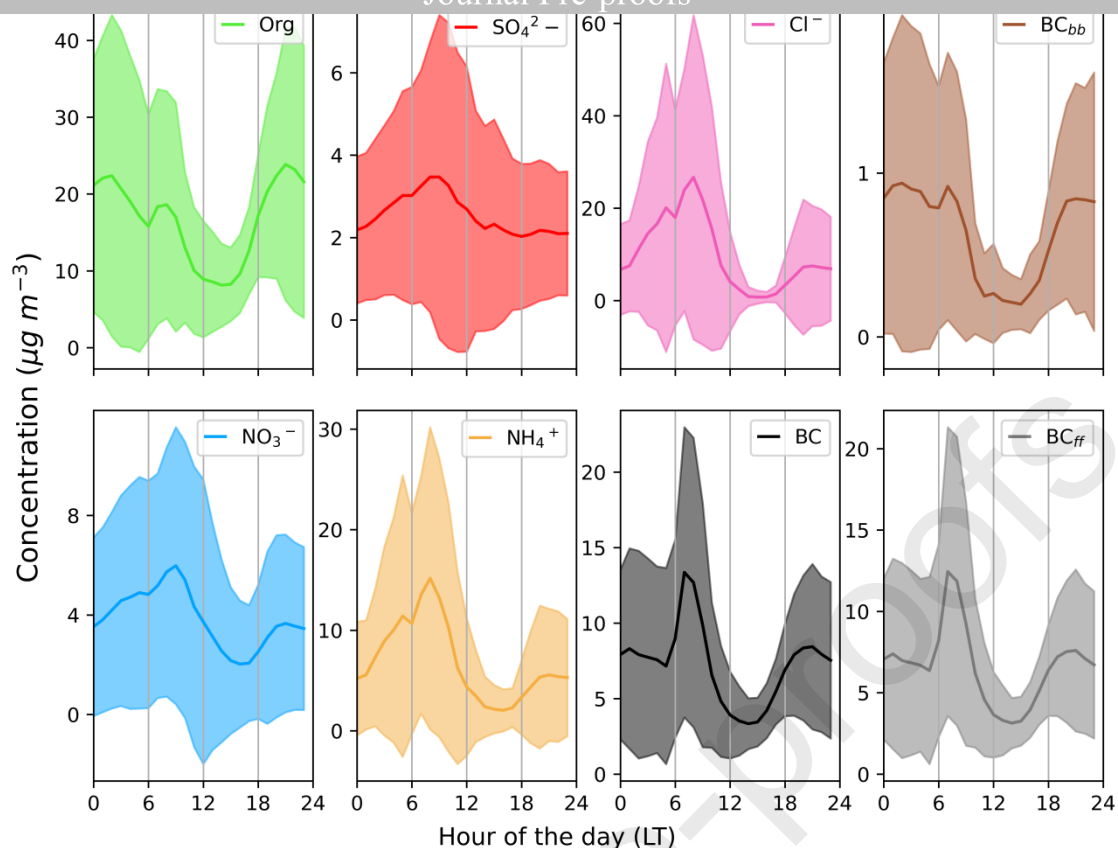
**PP1-PP2:** Two pollution periods (PP) were observed during the campaign and are associated with intense and long-lasting haze/smog events. Pollution period 1 (PP1) started on 15/12/2019 and ended on 25/12/2019 (first shaded area in Fig. 4), whereas pollution period 2 (PP2) started on 10/01/2020 and ended on 14/01/2020 (second shaded area in Fig. 4). Both episodes occurred under meteorological conditions when the prevailing winds had north to north-east origin (i.e., passing over the Nile delta) and corresponded to the two periods with air masses, primarily originating from the Middle East (See Fig. S4e-h). PP1 had an average value of NR-PM<sub>1</sub> of  $65.37 \mu\text{g m}^{-3}$  and recorded the highest hourly concentrations (above  $300 \mu\text{g m}^{-3}$ ) for NR-PM<sub>1</sub> during the campaign, with organics (32%) and chloride (19%) comprising its major fraction. During PP2, an average NR-PM<sub>1</sub> value of  $82 \mu\text{g m}^{-3}$  and a maximum hourly concentration during the first days of the episode to almost  $300 \mu\text{g m}^{-3}$  were measured. The average OA mass concentration ( $41.93 \mu\text{g m}^{-3}$ ) and contribution (almost 51 %) were higher in PP2 than in PP1. However, the percentage contribution of sulfate in PP2 decreased, as depicted in Figure 5, from 6.7 % in PP1 to 3.9 % in PP2, indicating the possible difference in the origin of the air masses. Contrary to sulfate, the contribution of chloride increased largely in PP2, from 18.3 % in PP1 to 22.9 % in PP2. The relative contribution of ammonium between the two periods was similar to 14-15 %. Thus, although an increase in organics is observed, NH<sub>4</sub>Cl contributes almost equally with organics in PP1 and dominates in PP2 (Figure 5).

Overall, independently of the air masses origin and wind direction NH<sub>4</sub>Cl is a great contributor to total NR-PM<sub>1</sub>.

### 3.3.5 Diurnal Variability of NR-PM1 species

The diurnal variations of NR-PM<sub>1</sub> species and BC during the entire campaign are shown in Figure 6.





**Figure 6:** NR-PM<sub>1</sub> species & BC (BC<sub>bb</sub>, BC<sub>ff</sub>) diurnal variability. The thick lines represent the means and the shaded areas span the standard deviation.

**Sulfate:** Sulfate is the only compound that does not display strong diurnal variations, with the highest concentrations observed during 06:00-12:00 LT and the lowest during 14:00 – 20:00 LT. The relatively flat pattern seen in the diurnal variability of sulfate is commonly reported in the literature and can be attributed to its regional origin, although we can distinguish higher concentrations in the late morning-midday that can be attributed to the photochemical (heterogeneous) processing of gas phase SO<sub>2</sub>, as reported by Khoder (2002) and Hassan et al. (2013) in one of the suburban areas of Cairo.

**Black Carbon:** Black Carbon shows a double peak in the morning and the evening. The highest concentration was observed early morning at 07:00 LT, reaching levels as high as  $13.4 \pm 9.6 \mu\text{g m}^{-3}$ , and the lowest one in the afternoon at 14:00 LT ( $3.3 \pm 1.7 \mu\text{g m}^{-3}$ ), probably combined with the dilution effect of the higher boundary layer during this time. This diurnal variability of BC is mainly dictated by its fossil fuel component (BC<sub>ff</sub>), which exhibits the typical double peak usually attributed to traffic at rush hours in the morning and evening. These observations are in line with the Cairo traffic congestion times, as reported by Nakat et al. (2013).

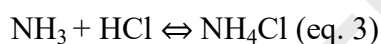
On the other hand, BC from biomass burning (BC<sub>bb</sub>) shows a different pattern with persistent high concentrations (around  $1 \mu\text{g m}^{-3}$ ) for almost the entire nighttime, when it usually shows an evening peak and low values over the night in many European cities during wintertime. While BC<sub>bb</sub> concentrations are much lower compared to BC<sub>ff</sub>, it is important to note that these two sources have very different OC/EC ratios, with (OC/EC)<sub>bb</sub> likely to be up to a factor of 10 higher compared to (OC/EC)<sub>ff</sub> over Cairo (Favez et al., 2008 and references therein). As such, while BC<sub>ff</sub> may be 4-5 times higher compared to BC<sub>bb</sub>, OC<sub>bb</sub> may be similar or even higher compared to OC<sub>ff</sub>.

**Organics:** Three peaks are observed in the diurnal variability of organic compounds: one at 2:00 (LT) ( $22.4 \pm 20.9 \mu\text{g m}^{-3}$ ) which is very rarely observed within urban environments, a second at 8:00 LT ( $18.6$

$\pm 1.10 \mu\text{g m}^{-3}$ ), and a value of  $2.100 \pm 0.10 \mu\text{g m}^{-3}$ . This observation supports the earlier assumption of a significant fraction of organics originating from biomass burning (in phase with the nighttime  $\text{BC}_{\text{bb}}$ ) and traffic-related sources (in phase with the double peaks of  $\text{BC}_{\text{ff}}$ ). The presence of three peaks in organic compounds is likely the result of a combination of these two major sources. It also demonstrates the value of high-time resolution measurements performed in our study to apportion the variability of combustion sources over Cairo.

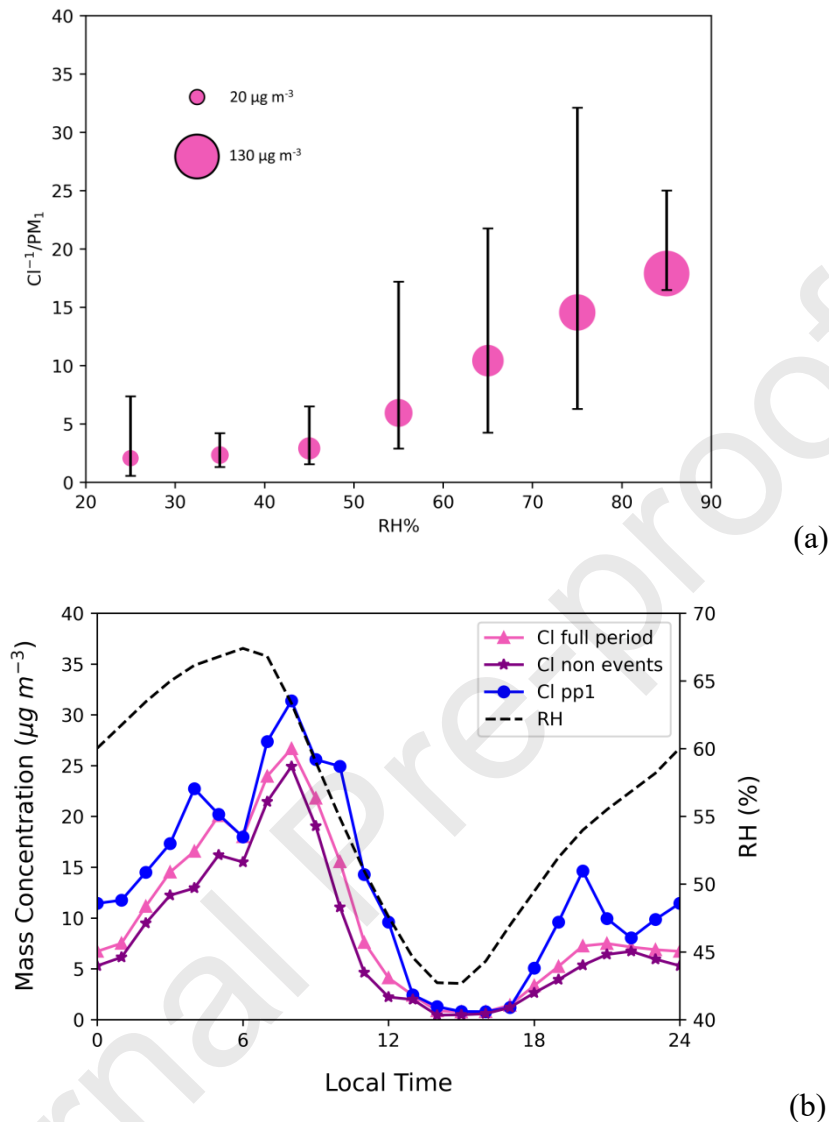
**Nitrate:** The diurnal variation of  $\text{NO}_3^-$  reflected optimal thermodynamic conditions to form ammonium nitrate and coincided with the traffic hours (with a 2-hour delay in the morning maximum at 09:00 LT and late evening at 22:00 LT). Being in the form of semi-volatile  $\text{NH}_4\text{NO}_3$ , condensation of nitrate is likely to be promoted by high relative humidity that will keep nitrate in the liquid phase and prevent its volatilization despite mild temperatures (e.g., the role of liquid water content on semi-volatile  $\text{NH}_4\text{NO}_3$  in Beijing (Sciare et al., 2007)).

**Chloride:** As for nitrate, chloride showed higher concentrations between 2:00 to 10:00 (LT). At 8:00 (LT), the chloride fraction reached its maximum, together with organics, with an hourly average of  $26.7 \pm 35.1 \mu\text{g m}^{-3}$ . At 15:00 (LT), chloride contributed very little to the total NR- $\text{PM}_1$  mass with diurnal average concentration below  $0.7 \pm 1.4 \mu\text{g m}^{-3}$ . Its variation is similar to that reported in New Delhi and other Indian cities in winter (Gunthe et al., 2021; Reyes-Villegas et al., 2021). Under favorable thermodynamic conditions, the formation of  $\text{NH}_4\text{Cl}$  results from the condensation of both  $\text{NH}_3$  and  $\text{HCl}$  as follows:



To better elucidate the factors affecting  $\text{NH}_4\text{Cl}$  formation and stabilization in the aerosol phase, its diurnal variability was plotted together with RH for selected conditions (i.e., full period, non-event, PP1; see Fig. 7b & Fig. S6). Chloride presented the same diurnal variability during all studied events, co-varying with RH, indicating stabilization in liquid phase (under elevated ambient RH conditions) in agreement with the thermodynamic behavior of  $\text{NH}_4\text{Cl}$  (Gunthe et al., 2021). The impact of ambient RH is also depicted

Figure 7a presenting the variability of the Cl mass fraction as a function of RH and shows a clear increase of the Cl contribution to  $PM_{10}$  mass for  $RH > 50\%$ .



**Figure 7:** Median chloride mass fraction in dry  $PM_{10}$  as a function of RH. The size of the circle indicates  $PM_{10}$  dry mass concentration. The error bars represent the 25th and 75th percentile (a). Diurnal profile of chloride concentrations in pink line for the entire period, purple for non-event periods, and blue for pollution episode 1 PPI, with a dashed line marking RH on the right y-axis.

The significant contribution of  $NH_4Cl$  to  $PM_{10}$  mass, especially at RH above 50%, together with its highly hygroscopic capacity, will substantially enhance aerosol water uptake through co-condensation, which will sustain particle growth, enhance aerosol condensation sink of reactive gases, therefore further leading to haze formation (Gunthe et al., 2021; Y. Chen et al., 2022).

### 3.4 Insights on the sources of dust and ammonium chloride

**Dust:** The calcium concentrations in  $PM_{10}$  in this study are remarkably higher (by a factor of 10) than the ones reported in Abu-Allaban et al. (2007) for  $PM_{2.5}$ . On the other hand, the calcium concentration in  $PM_{10}$  (this study) is consistently lower (by a factor of 3-4) than those reported for  $PM_{10}$  by Favez et al. (2008). As expected, calcium measured in this study correlates mainly with elements of crustal origin (Al, Fe, Mn, Ti; Fig. S7). This finding, coupled with the disparities in dust concentrations in  $PM_{10}/PM_{2.5}/PM_{10}$

mentioned above, underscores the need for further in-depth analysis of dust composition and sources in PM as it may not solely originate from natural (desert) sources. This is indicated by the fact that the average contribution of dust in PM<sub>1</sub> remains significant, comprising 21% of the total PM<sub>1</sub> mass concentration even when excluding the major dust event of the 25<sup>th</sup> of December. More specifically, dust characterization may warrant special attention since mitigation measures to decrease re-suspended dust from traffic may pose challenges to be implemented in Cairo, where natural dust deposition is expected to be substantial and washing out of the streets by rain may be particularly low. Equally important, the continuous high loading of dust in PM<sub>1</sub> indicates a much higher loading in PM<sub>2.5</sub> and PM<sub>10</sub>, thereby representing an important sink of water vapor and of a range of inorganic species as reported by Favez et al. (2008). In addition, coarse dust particles can act as a sink for sub-micrometer aerosols due to coagulation (Hinds, 1999), especially when the number concentration of the coarse mode is high, e.g. during a dust event (Jung et al., 2002). Coagulation of hygroscopic sub-micrometer aerosols on coarse dust particles can increase their water affinity (Tsai et al., 2015) and consequently their condensation sink potential. The reduction of the sub-micrometer particle number concentration (PNC; cf. Fig. 4) during the major dust event (DE) can be explained by their coagulation with the coarse dust particles in combination with the higher ventilation rate conditions prevailing during the DE (see section 3.3.4). Therefore, dust can be a significant contributor of urban haze observed over Cairo by acting as condensation and coagulation sink for atmospheric water, inorganic species and sub-micrometer aerosols.

**Ammonium chloride:** As discussed in Section 3.3.5, the high levels of NH<sub>4</sub>Cl (i.e., the most abundant chemical species in PM<sub>1</sub>) are attributed to the condensation of gas-phase ammonia and hydrochloric acid. The Nile Delta constitutes a significant agricultural ammonia hotspot, as confirmed by satellite observations (Clarisse et al., 2009; Van Damme et al., 2018), bringing to Cairo elevated levels of NH<sub>3</sub>. In their global emission inventory of HCl and particulate chloride from continental sources, Zhang et al. (2022) reported that emissions of HCl are mostly from open waste burning (38%), open biomass burning (19%), energy (19%), and residential (13%) sectors. The major contribution of open waste burning and open biomass burning on HCl was further assessed by D. Goetz et al. (2018) who reported emission factors for these specific combustion sources (open garbage burning and agricultural residue), highlighting large amounts of chloride from both sources but also high levels of organics and black carbon.

The concentration hotspots for chloride and ammonium shown in the polar plot (Fig. S8d-e) differ from those of other chemical species (Fig. S8a-c), as they point towards the south of the city. Although the hotspot aligns with a high concentration of chloride in the morning, this is also when thermodynamic conditions are favorable (high RH) for its formation, thus not implying a direct link to wind direction from the south. Instead, this suggests that waste burning activities, contributing to these concentrations, are widespread across the region, rather than being localized to the southwest (Fig S6.).

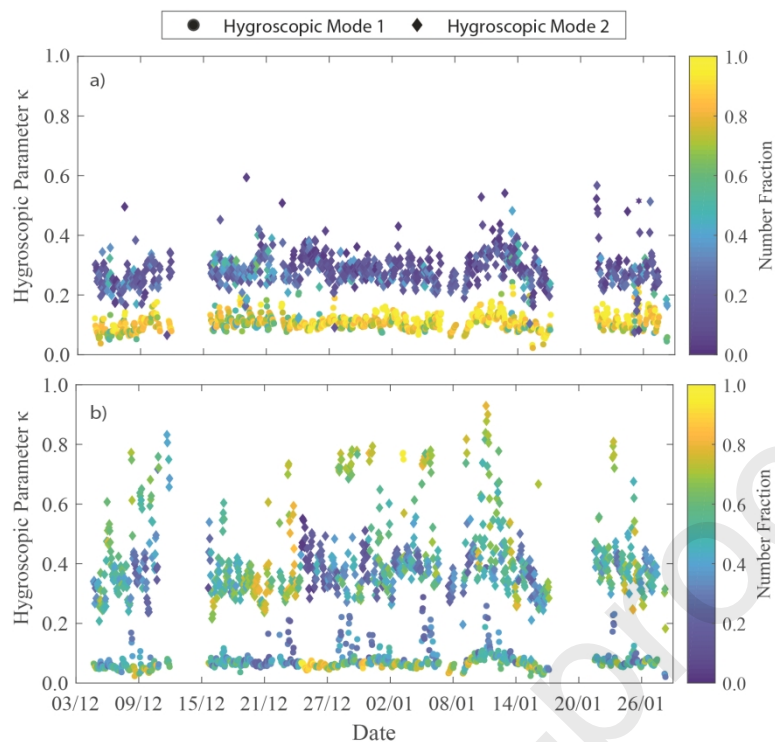
The potential role of open waste burning and open biomass burning over Cairo has been reported in the literature (e.g., Favez et al., 2009; Mahmoud et al., 2008). It is consistent with the statement that solid waste burning constitutes a major source of pollution in Greater Cairo (World Bank Group, 2013). The city generates 9.5 million tons of municipal solid waste annually, accounting for 47% of the municipal waste generated in Egypt, with collection coverage ranging between 40 and 90% in urban areas. A large amount of this waste (about 83%) ends up in large open dumps, and the remainder accumulates in streets or on illegal dumping sites. Five percent is being disposed of in sanitary landfills. As such, waste management seems inefficient and inadequate in Cairo, and self-ignition and open burning are regular patterns on these sites (ElSaid & Aghezzaf, 2018).

The significant correlation observed between NH<sub>4</sub>, Cl, and K (Fig. S7) (a well-known tracer of biomass burning) indicates that open burning practices of both municipal solid waste and biomass may happen at the same time and/or result from the mix of these two sources. Since, NH<sub>4</sub>Cl is a highly hygroscopic

species is theoretically predicted that the presence of water in the particle phase, at important levels as such observed in this work, will enhance aerosol hygroscopicity (Petters and Kreidenweis, 2007).

### 3.5 Submicron Particle Hygroscopicity (HTDMA)

Figure 8 shows the hourly averaged hygroscopic parameters derived by the HTDMA measurements (i.e., " $\kappa_{\text{HTDMA}}$ ") of aerosols having dry mobility diameters of 30 and 160 nm, while for rest of the sampled sizes are provided in Fig. S9. The majority of the sampled monodisperse aerosol populations (i.e., above 86% of the measured samples) exhibited an externally mixing state, resulting in two distinct hygroscopic modes after being exposed to elevated RH levels (i.e., ca. 89%), which is typical for particles sampled in urban areas (Swietlicki et al., 2008). For this reason, the number fraction of the particles residing in each hygroscopic mode is also denoted by different colors in Figure 8 and Fig. S9. Aggregated results showing the campaign mean values and standard deviations of the hygroscopic parameters and number fractions of each hygroscopic mode for each dry monodisperse aerosol population, together with the percentage of samples exhibiting externally mixed behavior, are given in Table 3. Overall, all examined monodisperse aerosol populations exhibited a hydrophobic/nearly hygroscopic mode with average  $\kappa_{\text{HTDMA}}$  values of ca. 0.1 and a more hygroscopic one with average  $\kappa_{\text{HTDMA}}$  values of more than ca. 0.3 (see Table 3). However, the number fraction of aerosols residing in each hygroscopic mode differed significantly for each monodisperse population measured. In more detail, the vast majority (i.e., number fraction of 83%) of the aerosols having a dry electrical mobility diameter of 30 nm resided in the hydrophobic/nearly hygroscopic mode, while only 52% of those with a dry electrical mobility diameter of 160 nm resided in the same hygroscopic mode. In addition, the hygroscopicity of the sampled aerosols classified as more hygroscopic increased as their dry electrical mobility diameters increased. For instance, the maximum hygroscopicity of the aerosols with dry electrical mobility diameters of 30 and 60 nm rarely exceeded the value of 0.6, while that of the aerosols with dry electrical mobility diameters of 160 nm exhibited maximum values above 0.9. This increasing trend is also reflected in the campaign average hygroscopic parameter values exhibited by the particles residing in the more hygroscopic mode (Table 3).



**Figure 8:** Hourly averaged hygroscopic parameter kappa values of particles having dry electrical mobility sizes of 30 (a), and 160 (b) nm. Kappa values were obtained from the respective hygroscopic growth factors (i.e., HTDMA measurements) accounting for the mixing state (i.e., internal or external) of the sampled aerosols. Different symbols represent the different observed hygroscopic modes (i.e., externally mixed aerosols), while the colors denote the number fraction of particles residing in each hygroscopic mode.

**Table 3:** Aggregate results of particle hygroscopic parameters  $\kappa$  and mixing state during the POLCAIR campaign for the different dry mobility diameters sampled by the HTDMA. For each sampled dry mobility diameter, the average values and standard deviations (in parenthesis) of the measured hygroscopic parameter and the number concentration fraction for each mode (i.e.,  $\kappa_{\text{mode1}}$ ,  $\kappa_{\text{mode2}}$  and  $N_{\text{f1}}$ ,  $N_{\text{f2}}$ , respectively) are provided. In addition, the percentage of samples where externally mixed particles were observed (i.e., with coexistence of two hygroscopic modes) is denoted with  $P_{\text{ex}}$ .

$D_{\text{dry}}$ (nm)	$\kappa_{\text{mode1}}$	$N_{\text{f1}}$	$\kappa_{\text{mode2}}$	$N_{\text{f2}}$	$P_{\text{ex}}$ (%)
30	0.11 (0.03)	0.83 (0.14)	0.28 (0.06)	0.18 (0.14)	86.4
60	0.08 (0.03)	0.62 (0.17)	0.30 (0.09)	0.38 (0.17)	98.9
90	0.08 (0.03)	0.54 (0.17)	0.34 (0.11)	0.46 (0.18)	99.8
120	0.07 (0.04)	0.53 (0.18)	0.37 (0.12)	0.47 (0.18)	99.9
160	0.08 (0.05)	0.52 (0.18)	0.41 (0.13)	0.48 (0.18)	99.3

Since the hygroscopic parameter of the sampled aerosols depends on their chemical composition (Petters and Kreidenweis, 2007), certain conclusions can be derived from the HTDMA measurements. The majority of the hydrophobic and less hygroscopic chemical species (i.e., BC and organics having  $\kappa$  values 0 and ca. 0.1, respectively) formed a distinct hygroscopic mode (i.e., hydrophobic/nearly hygroscopic) being externally mixed from other more hygroscopic chemical species with most of them residing in the lower sizes of the aerosol distribution (i.e., 30 and 60 nm exhibiting number fractions of 83 and 62% for the hydrophobic/nearly hygroscopic mode). While the same distinct mode (i.e., hydrophobic/nearly hygroscopic mode) with similarly low hygroscopicities was observed for aerosols of larger sizes (i.e., 90, 120, and 160 nm), only half of those resided in this mode. The other half of the population of these larger aerosols exhibited hygroscopicities indicative of mixtures of highly hygroscopic species (e.g., atmospheric salts like ammonium sulfate and/or chloride) with less hygroscopic species (i.e., BC and/or organics) at various proportions or even at certain cases hygroscopic properties related only to pure highly hygroscopic species (i.e.,  $\kappa_{\text{HTDMA}}$  values above 0.5; which is the value of pure ammonium sulfate). Most strikingly,  $\kappa_{\text{HTDMA}}$  values close to unity were observed for the largest particles sampled with the HTDMA, indicative of ammonium chloride having a theoretically derived  $\kappa$  value of ca. 0.95 according to the Extended AIM Aerosol Thermodynamics model (E-AIM model 4, Friese & Ebel, 2010; Wexler & Clegg, 2002) or in the order of 0.75, if the volatility and partial evaporation of the ammonium chloride aerosols inside the HTDMA apparatus is taken into account (D. Hu et al., 2011). This finding not only corroborates the existence of ammonium chloride, observed by the online and offline aerosol chemical composition measurements but also suggests that in certain instances such as during high loading periods and pollution events, ammonium chloride particles formed a distinct population (i.e., externally mixed) from other aerosols of the same size. In more detail, while the hygroscopicity of 30 nm aerosols remained rather unchanged throughout the different subsets (i.e., HL, LL, PP1, PP2, Day, Night; see section 3.3.4), the one exhibited by their bigger counterparts (i.e., 160 nm) classified in the more hygroscopic group, showed significant variations (see. Table 4 ). In addition, the number fraction of the 160-nm particles which were classified as more hygroscopic was found increased during periods of elevated chlorine mass concentrations (i.e., P1 and P2) and significantly increased during the HL period where  $\text{NH}_4\text{Cl}$  dominated  $\text{NR-PM}_{10}$  (see section 3.3.4). Overall, elevated chlorine mass concentrations are associated with an increase in the hygroscopicity of the bigger particles sampled by the HTDMA (e.g., those having dry mobility diameters of 160 nm) and were classified as more hygroscopic, as theoretically predicted and shown in Fig. S10.

Favorable conditions for the formation of particulate  $\text{NH}_4\text{Cl}$ , like elevated RH (see section 3.3.5), in combination with its very high water affinity can promote condensation of water vapor on the particles, favoring haze formation as demonstrated by Gunthe et al. (2021) and Chen et al. (2022). The possibility of haze formation under elevated ambient RH conditions is significantly higher when these highly hygroscopic aerosols are observed in high number concentrations (i.e., denoted by the number fraction of the more hygroscopic mode; e.g., during HL event). We should note, that the frequent observations of accumulation mode particles (i.e., 120 and 160 nm) exhibiting hygroscopicities close to that of pure  $\text{NH}_4\text{Cl}$ , is indicative of their local origin and distinct source, since these aerosols did not had enough time to coagulate or participate in atmospheric processing to be mixed with other species.

**Table 4:** Aggregate results of particle hygroscopic parameters  $\kappa$  and mixing state during the different subsets/events for particles having dry mobility diameters of 30 and 160 nm. For each sampled dry mobility diameter, the average values and standard deviations (in parenthesis) of the measured hygroscopic parameter and the number concentration fraction for each mode (i.e.,  $\kappa_{\text{mode1}}$ ,  $\kappa_{\text{mode2}}$  and  $N_{\text{fl}}$ ,  $N_{\text{E2}}$ , respectively) are provided. In addition, the

pl... modes) is denoted with  $P_{ex}$ .

$d_{m,dry}$	<i>Event</i>	$\kappa_{mode1}$	$N_{f1}$	$\kappa_{mode2}$	$N_{f2}$	$P_{ex}$ (%)
30 nm	HL	0.11 (0.02)	0.74 (0.15)	0.30 (0.05)	0.26 (0.15)	97
	LL	0.11 (0.03)	0.83 (0.14)	0.28 (0.06)	0.17 (0.14)	86
	PP1	0.11 (0.03)	0.75 (0.15)	0.30 (0.05)	0.25 (0.15)	96
	PP2	0.12 (0.03)	0.85 (0.14)	0.33 (0.06)	0.15 (0.14)	81
	Day	0.11 (0.03)	0.85 (0.13)	0.29 (0.07)	0.15 (0.13)	82
	Night	0.10 (0.02)	0.77 (0.15)	0.28 (0.05)	0.23 (0.15)	94
160 nm	HL	0.11 (0.06)	0.34 (0.11)	0.50 (0.21)	0.67(0.11)	100
	LL	0.07 (0.02)	0.54 (0.17)	0.39 (0.10)	0.46 (0.17)	100
	PP1	0.07 (0.02)	0.47 (0.22)	0.36 (0.09)	0.53 (0.22)	98
	PP2	0.09 (0.04)	0.47 (0.14)	0.46 (0.15)	0.53 (0.14)	100
	Day	0.08 (0.03)	0.52 (0.19)	0.41 (0.12)	0.48 (0.19)	99
	Night	0.07 (0.03)	0.51 (0.17)	0.39 (0.12)	0.49 (0.17)	100

#### 4. Conclusions

In the framework of the POLCAIR field campaign, a detailed characterization of the chemical and physical properties of submicron ( $PM_1$ ) aerosols at an urban background site of Greater Cairo (Egypt) was performed. Continuous observations, covering a 2-month period of the winter 2019-2020, were obtained using a comprehensive suite of quality controlled off-line and online atmospheric measurements.

The submicron particulate matter exhibited strong variability of its number concentration, with hourly averaged number concentrations reaching as high as  $70 \times 10^3 \text{ \#/cm}^3$  during intense pollution events. Analysis of aerosol hygroscopicity, which, depends on chemical composition, revealed that submicron aerosols in Cairo were externally mixed, exhibiting two modes: i) a hydrophobic/less hygroscopic mode indicating significant contributions of species such as organics and BC, and ii) a more hygroscopic one that is consistent with mixtures of less hygroscopic species (e.g., organics) with more hygroscopic ones



(e.g., ammonium chloride, sulfate, nitrate), including highly hygroscopic aerosols (i.e., more hygroscopic than pure ammonium sulfate), exhibiting hygroscopicities similar to that of ammonium chloride were observed in certain cases (e.g., during high loading periods and pollution episodes), forming a distinct population (i.e., externally mixed), accounting for 60% of the number concentration of the bigger particles (i.e., 120 – and 160 – nm; in dry conditions).

All these observations point to strong variability and heterogeneity of the observed sub-micron aerosols, indicative of the different sources and processes which were further assessed from a detailed characterization of submicron aerosol chemical properties.

Carbonaceous aerosols were measured as OC and EC from filter-based measurements, BC from biomass burning (BC<sub>ff</sub>) and fossil fuels (BC<sub>bb</sub>) from an aethalometer, and organics from Q-ACSM.

The most important findings of the study are the following:

- EC and OC were correlated, suggesting that combustion sources are the main drivers of their variability.
- The diurnal variability of BC<sub>ff</sub> showed an expected double peak (morning/evening) that corresponds to traffic rush hours. BC<sub>bb</sub> showed a nighttime maximum associated with (potentially unregulated) open biomass burning, which practice is known to occur during the night. Both fossil fuel and biomass burning sources were attributed to local activities within the Greater Cairo region.
- Dust has been found to be a significant component of PM<sub>1</sub>, accounting for an average of 24% over the 2-month duration of the campaign. During an intense dust event, PM<sub>1</sub> concentration peaked at 150  $\mu\text{g m}^{-3}$ , with dust comprising two-thirds of the PM<sub>1</sub> mass. This result highlights that natural aerosols should be carefully measured and accounted for when characterizing submicron aerosols in (semi-)arid urban environments.
- The most abundant chemical component of PM<sub>1</sub> was ammonium chloride (NH<sub>4</sub>Cl), corroborating the highly hygroscopic aerosols observed in certain cases (e.g., high loading periods, pollution events). Its diurnal variability revealed overnight formation due to biomass and waste burning and subsequent accumulation under favorable thermodynamic conditions with lower temperatures and higher relative humidity. The concurrent occurrence of biomass/waste burning is further supported by the strong correlation between NH<sub>4</sub>, Cl, and K (known as biomass burning tracer). The diurnal variability of NH<sub>4</sub>Cl is consistent with rapid volatilization around noon when RH drops below 40-50% and aerosol liquid water content diminishes. Conversely, high RH levels were found to promote the condensation of the highly hygroscopic NH<sub>4</sub>Cl in PM<sub>1</sub>, further amplifying the aerosol liquid water content. This finding, together with the significant number fraction of highly hygroscopic particles, highlights the critical role of NH<sub>4</sub>Cl as the principal factor behind the intense PM<sub>1</sub> pollution peaks and associated urban haze observed every morning over Cairo.

The persistent chloride concentrations over a two-decade span suggest minimal progress in mitigating the environmental impact of open biomass/waste burning. The latter emerges as a strong pollution source in Cairo and a major contributor to the city's urban haze. Overall, the findings of this study collectively suggest that Cairo may be experiencing a distinct type of urban haze stemming from the combination of chloride in the fine mode and dust in the coarse mode. Both elements could significantly influence the heterogeneous transformation of atmospheric pollutants, with further health implications to remain undetermined.

## 5. CRediT authorship contribution statement

**Aliki Christodoulou:** Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Spyros Bezantakos:** Methodology, Validation, Formal analysis, Investigation, Data curation, Writing – review

**Stavroulas:** Methodology, Investigation, Data curation. **Michael Pikridas:** Methodology, Investigation, Data curation, Visualization. **Konstantina Oikonomou, Minas Iakovides:** Data Curation. **Salwa K. Hassan:** Data curation, Writing – review & editing. **Mohamed Boraïy, Mostafa El-Nazer, Ali Wheida, Magdy Abdelwahab, Roland Sarda-Estèv, Martin Rigler:** Resources, Data Curation. **Giorgos Biskos:** Resources. **Charbel Afif:** Conceptualization, Project administration, Funding acquisition. **Agnes Borbon:** Conceptualization, Project administration, Funding acquisition, Writing – review & editing. **Mihalis Vrekoussis:** Supervision, Writing – review & editing. **Nikos Mihalopoulos:** Supervision, Writing – review & editing. **Stéphane Sauvage and Jean Sciare:** Conceptualization, Resources, Project administration, Supervision, Funding acquisition, Writing – review & editing.

## 6. 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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### CRediT authorship contribution statement

**Aliki Christodoulou:** Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Spyros Bezantakos:** Methodology, Validation, Formal analysis, Investigation, Data curation, Writing – review & editing, Visualization. **Efstratios Bourtsoukidis:** Writing – review & editing, Supervision. **Iasonas Stavroulas:** Methodology, Investigation, Data curation. **Michael Pikridas:** Methodology, Investigation, Data curation, Visualization. **Konstantina Oikonomou, Minas Iakovides:** Data Curation. **Salwa K. Hassan:** Data curation, Writing – review & editing. **Mohamed Boraiy, Mostafa El-Nazer, Ali Wheida, Magdy Abdelwahab, Roland Sarda-Estèv, Martin Rigler:** Resources, Data Curation. **Giorgos Biskos:** Resources. **Charbel Afif:** Conceptualization, Project administration, Funding acquisition. **Agnes Borbon:** Conceptualization, Project administration, Funding acquisition, Writing – review & editing. **Mihalis Vrekoussis:** Supervision, Writing – review & editing. **Nikos Mihalopoulos:** Supervision, Writing – review & editing. **Stéphane Sauvage and Jean Sciare:** Conceptualization, Resources, Project administration, Supervision, Funding acquisition, Writing – review & editing.

**Declaration of interests**

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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