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Key factors explaining severe air pollution episodes in Hanoi during 2019 winter season

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Abstract: With the aim of identifying key factors explaining the severe pollution events that occur under the impact of the Northeast monsoon during the winter period in Hanoi, a two-month campaign was carried out (11 January to 22 March 2019). The local meteorological parameters and the origin of the air masses were investigated, notably using a Doppler Lidar, and the structure of the boundary layer was thoroughly studied using elastic Lidar measurements for the first time in Hanoi. Four heavily polluted events occurred during the campaign, which were associated with hazy days; the highest daily PM_{2.5} concentrations reached $\sim 130 \mu\text{g m}^{-3}$ while the mean concentration during the campaign was $45 \pm 25 \mu\text{g m}^{-3}$. These severe polluted events in Hanoi were mainly related to LRT coming from China besides local emission or LRT from other polluted regions of Vietnam. BLH proved to be one of the key factors controlling air pollution in Hanoi. Indeed, the elastic Lidar measurements showed that the daily maximum of the hourly BLH varied from 700m to 1200m during cloudy days, while it was always below 1000m during hazy days. The lower BLH on the hazy days weakened the vertical mixing and was directly related to an increase in aerosol concentrations at the surface.

Keywords: Air pollution; Boundary layer; Southeast Asia ; Haze; Lidar; PM_{2.5}

Abbreviations:

AQI: Air Quality Index

RH: Relative humidity

BL: Boundary layer

ROCS : Range and overlap corrected signal

BLH: Boundary layer heights

SLIM: Aerosol micro-pulse Lidar

BAM: Beta attenuation monitoring

SEM-EDX: Scanning Electron Microscopy coupled

with Energy Dispersive X-Ray analysis

CBL: Convective boundary layer

SBL: Stable boundary layer

DBS: Doppler Beam Swinging

T: Temperature

EPA: U.S. Environmental Protection Agency

TSP: Total Suspended Particles

GDAS: Global Data Assimilation System

TSV: Total spatial variance

GM: Gradient method

USE: US Embassy

HYSPLIT: Hybrid Single Particle Lagrangian
Integrated Trajectory

VAST: Vietnam Academy of Science and
Technology

LRT: Long-Range Transport

VMHA: Vietnam Meteorological and Hydrological
Administration

LLJ: Low-level jet

NAAPS: US Naval Research Laboratory

VC: Ventilation coefficient

NBL: Neutral boundary layer

WS: Wind speed

OS : Observation Site

WD: Wind direction

PM: Particulate matter

WAQI: World Air Quality Index

28 **1. Introduction**

29 A high level of particulate matter (PM), especially PM_{2.5} (particles with aerodynamic diameters
30 $\leq 2.5\mu\text{m}$), has been proven to adversely impact human health and life quality (Kelly and Fussell, 2012;
31 Pope et al., 2020). Moreover, PM_{2.5} has been considered as the main pollutant causing severe air
32 pollution issues in cities in Southeast Asia (Reddington et al., 2019; Janta et al., 2020) including Hanoi
33 (Cohen et al., 2010a, 2010b; Hai and Kim Oanh, 2013; Ly et al., 2020). Hanoi, the capital of Vietnam,
34 has experienced rapid growth and urbanization over the past two decades along with extreme changes
35 in weather such as atmospheric circulation, air temperature and downward solar radiation (Taylor,
36 2010; Nguyen et al., 2019), resulting in a severe increase of pollutants such as PM_{2.5}, NO_x and SO₂.

37 From 2010 to 2013, considering the EPA's Air Quality Index (AQI), the number of days with
38 unhealthy levels for sensitive groups (AQI = 101 - 200) accounted for 40 to 60% of the monitored days
39 at Hanoi, while 102 days and 20 days were associated with very unhealthy (AQI = 201 - 300) and
40 hazardous (AQI ≥ 300) levels, respectively (MONRE, 2013). Concerning PM, the diurnal particulate

41 concentrations fluctuate substantially along the year. Daily variations of fine (PM_{2.5}) and coarse (PM₁₀)
42 particulate matter concentrations in Hanoi varied from 26 to 143 $\mu\text{g m}^{-3}$ and from 37 to 165 $\mu\text{g m}^{-3}$
43 respectively (Hai and Kim Oanh, 2013). Furthermore, the number of days with levels of PM₁₀
44 exceeding the specified Vietnam's standard for ambient air quality in 24-h ($\geq 150 \mu\text{g m}^{-3}$) was
45 approximately 50 days per year (Hien et al., 2011) while PM_{2.5} concentrations exceeding standard
46 ($\geq 50 \mu\text{g m}^{-3}$) were also recorded regularly. As a result, air pollution has been identified as one of the
47 major environmental concerns in Hanoi. Northern Vietnam has two main seasons, which include
48 Winter (November - March) and Summer (May - September), and two transitory seasons Spring
49 including (April) and Autumn (October) (MONRE, 2007). Particularly, PM concentrations in Hanoi are
50 lower in summer, when the southeast monsoon directed by tropical maritime air from the Pacific and
51 Indian Oceans delivers hot weather and convective conditions. Conversely, high levels of PM are
52 recorded in winter, when northern Vietnam is affected by the northeast monsoon (Cohen et al., 2010a;
53 Hien et al., 2011; Huy and Kim Oanh, 2017). Moreover, a high-pressure ridge from polar air in Siberia
54 results in the northeast monsoon bringing continental cold air from this area either through inland
55 China (dry period) from November to January or via the Gulf of Tonkin (humid period) from January
56 to March. However, the synoptic motion of air masses is not the only parameter that can explain the
57 pollution levels encountered in Vietnam. A high level of pollutants was noticed in winter in Hanoi for
58 days under haze condition (Ly et al., 2018, 2020), so the humidity can play a notable role in the
59 variation of pollutants. Besides local emissions, long-range transport (LRT) was also considered as a
60 potential source to the high level of PM, especially in winter with a strong impact of the northeast
61 monsoon. Although there was still a debate, a few studies have stated a similar contribution of emission
62 originating from both local and LRT sources in Hanoi (Cohen et al., 2010a; Nguyen et al., 2020).

63 On the other hand, boundary layer heights (BLH) and vertical wind profiles also play an important
64 role in the vertical mixing and transport of pollutants (Stull, 1988). Solar radiation and weather
65 conditions are the main variables affecting the dynamics of the boundary layer (BL). Inland, BL may
66 undergo a typical daily evolution driven by surface heating, but it is also strongly dependent on local
67 meteorology (haze, sea-breeze) and fronts (Stull, 1988). Concerning wind profiles, horizontal and
68 vertical wind speeds can change quickly in time as well as in space, thus, monitoring these profiles lead
69 to improve our understanding of atmospheric dynamics as well as the evolution of the BL. Radiosonde
70 measurements are considered as the most reliable method for retrieving vertical profiles of
71 meteorological parameters as well as BLH. Unfortunately, soundings are usually released into the

72 atmosphere 2 - 3 times per day, so this method is not able to monitor continuously the BLH evolution.
73 Nowadays, Lidar becomes more suitable for analyzing the BL structure and determining BLH due to its
74 high spatio-temporal resolution. However, this method requires greater technical resources and it still
75 has some optical limits especially under cloudy weather. In a few recent decades, Lidar's application in
76 order to analyze the BLH is widely applied in many regions of the world like Europe and Asia (Menut
77 et al., 1999; Delbarre et al., 2005; Dieudonné et al., 2017; Liu et al., 2019; Pan et al., 2019). However,
78 similar studies in Southeast Asia, especially in Vietnam are still limited (Hai et al., 2013).

79 In this study, our goal is to identify the key factors explaining severe pollution events in Hanoi
80 during winter, which is the highest pollution period over the year. The impact of the local
81 meteorological factors and the origin of the air masses will be firstly investigated. In addition, the
82 structure of BL will be thoroughly studied by the Lidar measurements for the first time in Hanoi. The
83 impact of its evolution as well as of atmospheric dynamics on hourly $PM_{2.5}$ concentrations during
84 pollution episodes will be explored.

85 **2. Field campaign for a dynamical study of air pollution in Hanoi**

86 *2.1 Devices, location and availability*

87 The campaign was carried out from 11 January to 22 March 2019 in Hanoi. Three observation sites
88 (OS) located in the urban area were considered (Fig. 1): OS1 located at Vietnam Academy of Science
89 and Technology (VAST) ($21^{\circ} 2' N$, $105^{\circ} 47' E$) which is a residential and traffic mixed area including a
90 ring road and several busy roads, OS2 at US embassy (USE) building ($21^{\circ} 1' N$, $105^{\circ} 49' E$) and OS3 at
91 Vietnam Meteorological and Hydrological Administration (VMHA) ($21^{\circ} 1' N$, $105^{\circ} 48' E$). The
92 surrounding areas of sites OS2 and OS3 are also densely populated and experience busy traffic.

93 First of all, a Doppler Lidar (Windcube V2 from Leosphere/Vaisala) and a custom-made aerosol
94 micro-pulse Lidar (SLIM) were located at the site OS1 on the rooftop of a 15m high building. In
95 addition, an aerosol sampler was set up next to the Lidar devices. A meteorological station (Davis
96 Instruments, Hayward, CA; model Vantage Pro 2) was placed on the rooftop of a second building (50m
97 above ground level (a.g.l.), around 100m away from the Lidar location) to minimize the impact of high
98 buildings around the site OS1. Next, the site OS2 hosted a $PM_{2.5}$ analyzer, based on a Beta Attenuation
99 Monitoring (BAM) system, from an international monitoring network of the World Air Quality Index
100 (WAQI) at USE. $PM_{2.5}$ concentrations in Hanoi were retrieved from EPA (<https://www.airnow.gov/>).
101 Finally, the sounding profiles were extracted from the University of Wyoming website
102 (<http://weather.uwyo.edu/upperair/sounding.html>) and radiosondes are launched routinely from site

103 OS3 by VMHA twice per day, at 07:00 and 19:00 local time (LT) (i.e. 00:00 and 12:00 UTC). This
104 radiosonde generally offers profiles of temperature, pressure, relative humidity, wind speed and
105 direction with a vertical resolution of 5m.

106 *2.2 Remote sensing of boundary layer and wind vertical profile*

107 *2.2.1 Doppler Lidar (Windcube V2)*

108 The Windcube V2 from Leosphere/Vaisala is an eye-safe pulsed Doppler Lidar operating in the
109 infrared (1.543 μm) at low power (7mW laser). It measures the Doppler shift due to the particles
110 motion along the laser beam direction (radial wind speed), which is determined through heterodyne
111 detection followed by fast Fourier transform analysis. The profiles of the three components of the wind
112 are reconstructed using the Doppler Beam Swinging (DBS) technique originally proposed for Doppler
113 radars (Koscielny et al., 1984), here combining four slanted beams (28° from zenith) pointing in the
114 cardinal directions and a vertical beam. The accumulation time is 1s per beam so that a DBS cycle is
115 repeated every 5s; then, wind observations are averaged over 10-min periods. The laser pulse duration
116 and shape allow a 20m vertical resolution while the optics allow a minimal/maximal range of 40/280m
117 a.g.l. During the campaign, this device was set to retrieve the wind every 20m from 60m to 280m a.g.l.
118 (the 40m altitude was not used due to the interference of nearby higher buildings). Besides wind speed
119 (WS) and wind direction (WD) profiles, the vertical wind speed standard deviation (σ_w) was also
120 computed using the Doppler Lidar observations. This quantity provides a direct estimation of the
121 amount of turbulence, thus the vertical mixing state of the BL.

122 *2.2.2 Aerosol micro-pulse Lidar (SLIM)*

123 The SLIM was deployed during the campaign. It is a custom-made micro-pulse Lidar developed at
124 LATMOS in collaboration with CIMEL that operates at the edge of the visible spectrum (808nm) and
125 at low power (12 mW laser). The device was pointed at the zenith and recorded profiles of elastic
126 signal back-scattered by the atmosphere with a vertical resolution of 15m, averaged over 10-min, then
127 1-h. The overlap function was determined using the slope method applied to a 3-h average profile
128 recorded in a well-mixed afternoon BL. Then, the BLH was determined by the 1-h average profiles of
129 the Range and Overlap-Corrected Signal (ROCS), noted $P(z)$ with the altitude (z).

130 In cloud-free conditions, the Lidar signal yields a strong back-scattering from the particles within the
131 BL, then decreases through a transition zone and disperses completely in the free troposphere as back-
132 scattering from the molecules is much weaker, especially at 808nm. Thus, the first derivative $dP(z)/dz$
133 of the ROCS exhibits a strong negative peak at the height which is considered to be the BLH.

134 This well-known method for BLH determination is the Gradient Method (GM) (Flamant et al.,
135 1997; Cohn and Angevine, 2000). The GM is still valid when a thin cloud is capping the BL. Indeed,
136 that type of cloud is included in the BL, and the GM will then detect the strong decrease of signal
137 occurring when the laser beam exits the cloud. When a thick cloud is present just above the BL, it
138 rapidly extinguishes the laser beam, and the GM will return the extinction height, which is located
139 inside the cloud. This will cause a slight overestimation of the BLH because thick clouds are not
140 included in the BL, so the cloud base should be considered instead as BL top. However, this slight
141 overestimation is not a problem when the main point is to follow the BLH variability and its influence
142 over pollution, so a simple and more reliable algorithm was preferred. Table A.1 shows the SLIM data
143 availability during the campaign. Due to technical issues as well as problems with water condensation
144 in the optical head, the retrieval-signals in all the days was not always reliable and even if the data were
145 available, it did not mean that processing them to retrieve the aerosol optical properties was always
146 possible.

147 *2.3. Aerosol sampling and elemental analysis*

148 Total Suspended Particles (TSP) were collected at site OS1 by total filtration with 1-week period
149 on cellulose filter (Whatman® grade 41) with a flow rate of $1.2 \text{ m}^3 \text{ h}^{-1}$. The filters were changed every
150 Wednesday at 16:00 LT from 11/01 to 22/03, resulting in 10 TSP samples referenced from S₁ to S₁₀.

151 One fraction of each filter was dedicated to the analysis of the water soluble NO_3^- and SO_4^{2-} by
152 ionic chromatography, using an ICS 5000+ ion chromatograph from Thermo Scientific Dionex
153 equipped with an anionic CS 12A column (2x250 mm). After leaching of the sample in ultrapure water
154 ($18.2 \text{ M}\Omega \text{ cm}$) under ultrasonic shaking for 1-h, the obtained aqueous solution was filtered (IC
155 Millex®, Millipore) and analyzed using KOH solution as eluent. Atmospheric concentration of
156 ammonium is then deduced from nitrate and sulfate concentrations considering atmospheric nitric and
157 sulfuric acids as completely neutralized by ammoniac.

158 The other fraction of each filter was dedicated to SEM-EDX (Scanning Electron Microscopy
159 coupled with Energy Dispersive X-Ray) analysis using a JEOL™ JSM7100F electron microscope,
160 equipped with three ultrathin-window (30 mm^2) energy-dispersive X-ray detectors (Bruker™ XFlash
161 6/30). EDX spectra were acquired at an acceleration voltage of 15 kV, a probe current of 200 pA with a
162 time acquisition of 40s. 10 spectra per sample were acquired with large fields ($1600 \times 1100 \mu\text{m}^2$) and
163 summed to be then considered as one. Then, the elemental ratios of detected elements
164 (Na, Mg, Al, Si, S, Cl, K, Ca, Ti, Fe, Zn) were calculated using PhiRoZ quantification procedure from

165 the Esprit Software (Bruker Nano, Germany). The most significant error in the quantitative analyses is
166 due to the heterogeneity of particle deposition on the surface of the collection substrate. To assess this,
167 relative standard deviations associated with the 10 spectra per sample were calculated. They depend
168 strongly on the element considered, but are less than 10% for major elements and up to 20% for minor
169 elements. Atmospheric concentrations were then computed based on the atmospheric SO_4^{2-}
170 concentrations measured by ionic chromatography and S relative content in PM measured by EDX,
171 considering Al, Si, Ti, Fe and Zn as oxides and Ca as calcite. Though the particulate carbon fraction has
172 not been determined, the sum of these compounds represents a large part of the TSP and is highly
173 correlated with the online $\text{PM}_{2.5}$ measurements with BAM ($y = 1.22x$, $R^2 = 0.83$).

174 *2.4 HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) back-trajectories*

175 The PC Windows-based HYSPLIT model (Stein et al., 2015) was widely used for generating
176 backward trajectories to evaluate potential sources for LRT. The Global Data Assimilation System
177 (GDAS) was used as weather input to the HYSPLIT model. 72-h backward trajectories were computed,
178 ending at site OS1 every hour from 11/01 to 22/03/2019. Based on the observation of the radiosonde
179 profiles, four ending altitudes were used: 100m a.g.l. (daytime surface layer or stable layer),
180 600m a.g.l. (convective layer or residual layer), 1000m a.g.l. (transition layer) and 1500m a.g.l. (free
181 troposphere).

182 Single backward trajectory estimation by the HYSPLIT model has been considered to be definite,
183 but still maintain some computational uncertainty. For example, 10 to 20% of the travel distance was
184 estimated as the horizontal uncertainty of the trajectory measurements (Draxler and Hess, 1998), while
185 the bias of trajectories originated from wind fields considered to be typically of the order of 20% of the
186 travel distance (Stohl, 1998). However, the accuracy of this model can be improved by using a large
187 number of trajectories in statistical analysis (Cabello et al., 2008). Thus, control measures were applied
188 in our study to improve confidence in the HYSPLIT generated back-trajectories: Firstly, trajectories
189 were computed for four different heights to reduce topographic effects as well as to sample the different
190 dynamic layers. Secondly, an 1h integration time step, which is the shortest possible in HYSPLIT, was
191 used for the cluster analysis. Finally, the statistical analysis with a huge number of trajectories arriving
192 at the study site was performed to reduce the uncertainties of the cluster analysis. In total, almost 1700
193 backward trajectories were computed and classified using the clustering analysis of the HYSPLIT
194 software. These trajectories were assigned to clusters depending on their characteristics (speed and
195 direction) using a k-means clustering method, which has been considered as one of the simplest

196 unsupervised learning algorithms. The optimal number of clusters were chosen so as to minimize the
197 total spatial variance (TSV).

198 **3. Results and discussion**

199 *3.1 Overview of basic meteorological parameters and pollution during the campaign*

200 A summary of the meteorological conditions and their temporal variability for each month is shown
201 in Table 1. January was colder by $\sim 3^{\circ}\text{C}$ compared to February and March. Since the whole campaign
202 was carried out in the humid period, the RH was high. The visibility was at its lowest in January due to
203 frequent fog/haze episodes, and its highest in February. Moreover, monthly-mean $\text{PM}_{2.5}$ concentrations
204 were the highest in January, while the lowest concentrations were recorded in February, probably as a
205 result of the decrease of industrial activity and transport during the Lunar New Year (01 - 10/02). Figure 2
206 shows the mean $\text{PM}_{2.5}$ levels as a function of the WS and WD, using wind data from the Doppler Lidar at
207 100m a.g.l. as input. This height was chosen to be just above the highest buildings in the area (which
208 have 30 to 35 floors). Besides, the WS from the NE and SE were predominant during the three months,
209 which is typical of the winds observed in winter in Hanoi (Lasko et al., 2018). This figure also explicitly
210 demonstrates that the highest levels of $\text{PM}_{2.5}$ were mainly related to weak WS, so probably due to local
211 emissions that are not well dispersed. These local sources seem to be mainly located to the west of site
212 OS1 in January, to the Northeast in February and to the East and Southeast in March. Nevertheless, a
213 finer analysis of the atmospheric dynamics is necessary to draw reliable conclusions on the origin of this
214 pollution.

215 **Table 1:** Statistical summary (mean \pm one standard deviation) of meteorological parameters for each month.

| | <i>January</i> | <i>February</i> | <i>March</i> |
|--|-----------------|-----------------|-----------------|
| T ($^{\circ}\text{C}$) | 18.3 ± 2.5 | 21.9 ± 3.2 | 21.4 ± 2.8 |
| RH (%) | 83.1 ± 6.7 | 85.2 ± 4.9 | 85.9 ± 7.2 |
| WS (m s^{-1}) | 3.1 ± 1.2 | 3.9 ± 0.8 | 3.5 ± 0.87 |
| Visibility (km) | 5.4 ± 2.4 | 7.1 ± 1.6 | 6.5 ± 2.1 |
| $\text{PM}_{2.5}$ ($\mu\text{g m}^{-3}$) | 56.8 ± 33.2 | 36.4 ± 14.2 | 44.4 ± 23.5 |

216
217 Figure 3 displays time-series of the daily averages of some basic meteorological factors along with
218 the $\text{PM}_{2.5}$ concentrations during the campaign. Figure 3a shows the daily average temperature (T) in a
219 range of 12°C - 26°C . In addition, the weather in this period was under humid conditions with daily
220 relative humidity always higher than 60% (Fig. 3a). This humid condition might facilitate a
221 reinforcement in the level of $\text{PM}_{2.5}$ through the formation of secondary aerosols by heterogeneous

222 reactions and their hygroscopic growth in ambient air (Tang and Munkelwitz, 1994; Marris et al.,
223 2012). This is notably demonstrated by an increase of the sulfate to nitrate ratio retrieved from our
224 weekly PM samples as a function of relative humidity (RH) (Fig. A.1). Indeed, the sulfate can be
225 formed in aqueous phase reaction at high RH, while the nitrate is predominantly formed by gas-phase
226 or heterogeneous photo-chemical reaction (Seinfeld and Pandis, 2006). Figure 3b shows the result of
227 daily-average value of solar radiation (Q) and the daily maximum of the hourly BLH values (max
228 BLH) for each day during the study period. We can observe a relation between these two parameters,
229 but the correlation is very weak ($r^2 = 0.44$) because the weather is often cloudy during the campaign. In
230 conclusion, the solar radiation cannot be a good indicator of the maximum BLH at this period of the
231 year and the BLH need to be measured with a reliable method. Concerning $PM_{2.5}$, the daily values
232 varied from 9 to 129 $\mu\text{g m}^{-3}$ (Fig. 3f) with an average and standard deviation of $45 \pm 25 \mu\text{g m}^{-3}$, which
233 are typical values recorded in Hanoi within this period (Hien et al., 2011; Huy and Kim Oanh, 2017).
234 The highest daily level of $PM_{2.5}$ was recorded on 25 January ($129 \mu\text{g m}^{-3}$), while the lowest daily $PM_{2.5}$
235 concentrations ($\sim 25 \mu\text{g m}^{-3}$) were mainly recorded in February. During the campaign, four heavily
236 polluted events characterized by $PM_{2.5}$ higher than $50 \mu\text{g m}^{-3}$ for more than 48-h were observed (P1 -
237 P4 marked by grey areas), based on the specified Vietnam's standard for ambient air quality. Although
238 this is not always the case, the daily variation of $PM_{2.5}$ concentration is generally associated with local
239 WS: high WS are related to low concentrations of $PM_{2.5}$ and lower WS correspond to the high levels of
240 $PM_{2.5}$ (Fig. 3c and 3f).

241 In order to study the relation between PM concentration and meteorology, weather conditions were
242 classified into four distinct types typically observed in Hanoi from January to March: “clear”, “hazy”,
243 “foggy” and “cloudy” conditions (Fig. 3f). In this paper, hazy days should satisfy two criteria: a daily
244 visibility $\leq 5\text{km}$ and a daily RH $\leq 95\%$. Foggy days were discriminated only by RH $\geq 90\%$. The clear
245 days were selected by a daily visibility $\geq 10\text{km}$. The rest of the days were considered as cloudy days.
246 During the campaign, these cloudy days were observed in 41 days, which accounted for $\sim 59\%$ of total
247 days, so they were the most frequent type of weather condition. Afterward, the hazy days were the
248 second most common weather type during the campaign which were 20 days and made up for $\sim 29\%$ in
249 total. Then foggy and clear days accounted for $\sim 11\%$ and $\sim 1\%$ respectively.

250 As indicated in Figure 3(f), the daily RH values were very high ($\sim 90\%$) on both hazy and foggy
251 days. In fact, one of the parameters distinguishing these two types of day is the $PM_{2.5}$ concentration:
252 The hazy days are characterized by higher daily-average $PM_{2.5}$ concentrations ($75.8 \pm 23.1 \mu\text{g m}^{-3}$)

253 compared to foggy days, for which $\text{PM}_{2.5}$ concentrations were $33.4 \pm 9.5 \mu\text{g m}^{-3}$. Moreover, the 4
254 pollution events (P1-P4) are all associated with hazy days. This is explained by the fog's ability to wash
255 out the atmosphere and thus reduce air pollution. In the clear days, which appeared only one day during
256 the campaign, daily $\text{PM}_{2.5}$ concentrations were $19.1 \mu\text{g m}^{-3}$, while $\text{PM}_{2.5}$ concentrations in cloudy days
257 were $32.6 \pm 12.6 \mu\text{g m}^{-3}$. The daily-average $\text{PM}_{2.5}$ concentrations can be also controlled by WS
258 measured at 100m a.g.l.: clear days are characterized by high daily WS (5.6 m s^{-1}), while hazy days
259 were related to lower WS ($2.5 \pm 0.5 \text{ m s}^{-1}$). WS on cloudy and foggy days were $4.1 \pm 0.8 \text{ m s}^{-1}$ and
260 $3.3 \pm 0.6 \text{ m s}^{-1}$, respectively. In conclusion, a strong wind, which is favorable to the dispersion of
261 pollutants, generally induces lower concentrations of pollutants than a weak wind. Figure 3(c) indicates
262 that there was a significant difference between WS recorded at 50m a.g.l. by the weather station and the
263 Doppler Lidar observations at 280m. This could be explained by the complex structure of vertical wind
264 motion and the atmospheric dynamics which are not easy to be fully monitored by meteorological
265 ground-based devices. Thus, in order to clearly understand the role of the atmospheric dynamics on the
266 sudden change of $\text{PM}_{2.5}$, the vertical wind speeds need to be studied in further by using the Doppler
267 Lidar, which will be explored with the BLH in section 3.3.

268 The occurrence of cold surges in winter can also be a factor affecting $\text{PM}_{2.5}$ concentrations in
269 Hanoi. During our campaign, there were 5 cold surges marked with red triangles on the time axis in
270 Figure 3. The beginning of these cold surges was detected by (1) a significantly daily temperature drop
271 of more than 2°C along with (2) high wind coming from the NE. For example, a first cold surge
272 occurred in Hanoi from 14 to 17 January: the WD shifted from SE to NE (Fig. 3d), with a dramatically
273 increased of the WS and the temperature significantly dropped. After that, the 2nd, 3rd, 4th and 5th surges
274 were recorded on 21/01, 21/02, 05/03 and 14/03 respectively. Figure 3(f) shows the arrival of a cold
275 surge generally coincides with a sudden drop in $\text{PM}_{2.5}$ concentrations, followed by a gradual increase in
276 the following days when the temperature rises again. However, this decrease is not always of the same
277 magnitude, levels of $\text{PM}_{2.5}$ clearly decrease at the arrival of the first two and the last cold surges of the
278 campaign, but not really for the 3rd and the 4th cold surges. Thus, meteorological parameters associated
279 with cold surges cannot predict completely the variability in $\text{PM}_{2.5}$ levels. In more general terms,
280 conventional meteorological parameters recorded on the ground such as T, RH, WS and WD influence
281 pollution levels, but they do not seem to be the only determining parameters. It is in fact necessary to
282 take into account other factors to better understand the pollution events, such as the origin of air masses
283 notably described by backward trajectory data or the structure of the boundary layer.

284 3.2 Local against Long-Range Transport

285 The results of the HYSPLIT cluster analysis over the whole campaign are presented on Figure 4 to
286 further analyze the impact of LRT. During the scope of this study, pollution is considered from LRT
287 when the sources are located more than 50km from the OS1. The back-trajectories of air masses
288 arriving at 100m a.g.l. at Hanoi are classified among six clusters (Fig. 4a). For four clusters out of the
289 six (from clusters #1a to #4a), accounting for a total of 75% of time, the air masses arrived in Hanoi by
290 the Southeast while staying at low altitudes (i.e. inside the BL). Evidently, most of these air masses
291 passed through the northeastern coast of Vietnam (i.e. Quang Ninh, Hai Phong, etc.) and Red River
292 Delta (Fig. 1a) which were reported to have high number of pollution sources (Huy and Kim Oanh,
293 2017). Thermal power plants and industrial activities (i.e. mineral products, brick manufacturing and
294 cement factories) are notably significant PM sources located in these areas. Thus, the air quality in
295 several cities in these regions are highly impacted by local emissions, as demonstrated by the increase
296 of Pb-PM₁₀ in Hai Phong under NE monsoon (Chifflet et al., 2018) or PM_{2.5} in Quang Ninh in winter
297 (Hang and Kim Oanh, 2014). In this latter case, PM_{2.5} was composed of organic matter, water soluble
298 inorganic ions (NH₄⁺, SO₄²⁻, and NO₃⁻), soot and crustal elements which were essentially attributed to
299 pollution sources such as biomass burning, or to the formation of secondary inorganic aerosols. As a
300 result, these regional sources of pollution can contaminate the air masses arriving in Hanoi and this
301 LRT explains high PM concentrations. In addition, before reaching Vietnam, the air masses for
302 clusters #2 and #4 flew over eastern China where PM_{2.5} emissions are very high, especially on winter
303 haze days, as observed by many studies (Ding et al., 2019; Zhang et al., 2020). The major pollutions
304 transported from these regions include secondary sulfate-rich compounds originating from
305 automobiles, coal-fired power stations and industrial emissions (Cohen et al., 2010b; Hai and Kim
306 Oanh, 2013) and occasionally dust from deserts located a little more to the north (Cohen et al., 2010a).
307 By using the chemical transport model NAAPS (from the US Naval Research Laboratory), the
308 simulated mean of total optical depth over East Asia for the most polluted day of each pollution event
309 (P1-P4) confirms the presence of these polluted areas in China (Figure A.2).

310 The mean PM_{2.5} levels associated with each of these six air mass clusters is highly variable
311 (Fig. 4a). The highest PM_{2.5} concentrations were observed when the back-trajectories ending at 100m
312 belonged to clusters #2a ($59.6 \pm 34.3 \mu\text{g m}^{-3}$ in average) and #4a ($48.5 \pm 29.1 \mu\text{g m}^{-3}$ in average), as
313 these air masses likely accumulated pollution from eastern China and northeastern Vietnam. This
314 situation represented a total of 36% of time during the campaign. Lower PM_{2.5} concentrations were

315 observed when the air mass back-trajectories belonged to clusters #1a ($38.1 \pm 23.1 \mu\text{g m}^{-3}$ in average)
316 and #3a ($32.9 \pm 12.4 \mu\text{g m}^{-3}$ in average) which flew over the sea before reaching in Hanoi, with local
317 and Northeastern Vietnam's emissions predominantly contributing to these levels. This situation
318 represented a total of 40% of time during the campaign. Cluster #5a (21% of time) corresponded to air
319 masses coming at low altitude from the North, i.e. from inland China, an area that is less populated and
320 industrialized than the eastern coast, but still has large sources of pollution (the trajectory points
321 towards the Chongqing and Chengdu large agglomerations), thus leading to an intermediate $\text{PM}_{2.5}$
322 concentrations ($41.3 \pm 34.8 \mu\text{g m}^{-3}$ in average). Besides the dominated northeastern air masses during
323 this time, the westerly trajectories of cluster #6a rarely occurred (3% of time) and were associated with
324 higher levels of $\text{PM}_{2.5}$ concentrations ($51.2 \pm 27.6 \mu\text{g m}^{-3}$ in average). They flew over Laos and
325 northern Thailand, a forested area in which many burning spots were visible on MODIS thermal
326 anomaly images during the period of the campaign, especially in March (Fig. A.3). The altitude of
327 these trajectories is high, but still compatible with catching emissions from the ground, especially
328 smoke from large forest fires that rises in altitude due to the heat. For the back-trajectories arriving in
329 Hanoi at 600m, 1000m and 1500m a.g.l. (Fig. 4b, c and d respectively), HYSPLIT returned five
330 clusters similar to those found for an arriving height of 100m, only the cluster from inland China
331 (cluster #5a) did not exist for air masses arriving at high altitude (600m and higher). The fraction of the
332 westerly back-trajectories increased with altitude, representing 13% of time in total at 600m a.g.l.
333 (clusters #3b and #4b) against only 3% at 100m a.g.l. (cluster #6a). This point is even more marked at
334 higher altitudes, with 30% and 60% of westerly back-trajectories arriving at 1000m (clusters #3c and
335 #5c) and 1500m a.g.l. (clusters #2d, #4d and #5d), respectively. Conversely, the share of the
336 southeastern back-trajectories was maximal at an arriving height of 600m a.g.l. (87%) and then
337 decreased as the ending altitude increased; it still accounted for 49% of the back-trajectories at
338 1000m a.g.l. but only for 13% at 1500m a.g.l. Thus, there is a stronger impact of the northeast
339 monsoons at the heights of 100m and 600m than other altitudes. In the next parts, the relation of these
340 air masses and the pollution near the ground will be further studied.

341 In order to highlight a possible relationship between the chemical composition of aerosols and
342 potential source areas, the relative chemical composition of the weekly TSP samples and the origin of
343 associated air masses ending at 100m a.g.l. are presented in Figure 5(a) and 5(b). In general, the
344 relative contribution of sea-salt composed from Na and Cl were low (below 5% of total of TSP),
345 although it was slightly higher for samples associated with a strong contribution from marine air

346 masses (clusters #1a and #3a), i.e. for samples S5, S6 and S8. In the meantime, a high contribution of
347 minerals (Al, Si, Ca and Mg), metals (Fe, Zn) and secondary inorganic aerosols (sulfate, nitrate and
348 ammonium) was observed during all weeks, even for sample S6 for which clean oceanic air masses
349 were predominant. Therefore, these non seasalt aerosols have a local origin in this case, such as the Red
350 River Delta or the northeastern coast of Vietnam. The highest PM_{2.5} concentrations were observed for
351 weekly samples S2, S3 and S9 associated with air masses dominated by continental clusters #2a, #4a
352 and #5a, while the lowest level of PM_{2.5} were observed for samples S6 and S8 associated with the
353 domination of marine air masses from clusters #1a and #3a. Furthermore, a comparison of the TSP
354 composition between these samples indicates a similar chemical composition although there was a
355 huge different in TSP concentrations and origins between these weeks. This demonstrates a similarity
356 in the major chemical composition of local particulate emissions and those coming from LRT including
357 from other polluted regions of Vietnam and from China. This can be explained by the presence of the
358 same types of industrial and urban sources in these geographical areas.

359 At last, we focused at the four major pollution events (P1 - P4) in order to understand what are the
360 factors that explain these strong transient increases in PM_{2.5}. During the first episode (P1), Hanoi was
361 dominated by a high-pressure system and below a subsidence region (Fig. A.4a). This subsidence
362 generated a very strong temperature inversion above the BL, and the calm conditions at the surface were
363 favorable to the accumulation of pollutants, leading to widespread haze. The levels of PM_{2.5} started to
364 increase on 18/01, when the WD shifted from strong NE winds to weaker southwest and northwest winds
365 (Fig. 3d) and remained at high level until the arrival of a new cold surge. As expected, this one was
366 associated with an increase in the WS and precipitation on 20/01, which put an end to this pollution
367 event. Next, similar to the P1 event, Hanoi was also dominated by a high-pressure system during the
368 episode P2 from 23 - 28/01 (Fig. A.4b) and the event started when the WD shifted from strong NE winds
369 to weaker SE on 23/01. The daily PM_{2.5} concentrations tended to maintain at high pollution levels until
370 the beginning of the 28/01 with the replacement of a low-pressure system that normally brings wet and
371 windy conditions (Fig. A.4c). Afterward, the third polluted event (P3) occurred on day 26/02 when the
372 WD shifted from strong NE winds to weaker southwest winds along with the increase of PM_{2.5}
373 concentration. On 28/02, the appearance of low pressure system (Fig. A.4d) led to the reinforcement of
374 WS which ended this pollution event. Finally, the last pollution event (P4) was recorded between 11 and
375 14 March. The PM_{2.5} concentrations rose from 11/03 when the WD changed from NE to SE. Similar to
376 the first pollution event, the arrival of a new cold surge on day 14/03 with high WS put a termination to

377 this event. During the first three pollution events, the air masses originating from coastal or inland China
378 overwhelmed at lower heights, i.e. clusters #2a, #4a and #5a at 100m a.g.l. (Fig. 6a) and clusters #1b and
379 #2b at 600m a.g.l. (Fig. 6b). For the last pollution event P4, the appearance of cluster #6a from the West
380 (Fig. 6a) suggest an impact of transported biomass burning emission at elevated altitude. Moreover, the
381 total optical depth from simulated-model NAAPS shows clearly the impact of smoke from biomass
382 burning from the West on day 14/03 (P4 pollution event) (Fig. A2.d). This is consistent with scenarios in
383 which the NE monsoon air masses pass through eastern or inland China, the northeastern regions of
384 Vietnam and arrive in Hanoi, possibly carrying a large number of aerosols and gaseous pollutants.
385 Consequently, LRT from the Northeast includes a combination of essentially two main polluted
386 contributions, from the northeastern coast of Vietnam and from eastern or inland China. In addition, the
387 impact of biomass burning from forest burning and burning activities in Laos and Thailand in March is
388 also observed as mentioned by other studies (Huang et al., 2013; Reid et al., 2013).

389 However, the results shown in figure 6 does not fit with the results in figure 2, according to which
390 steady winds from the NE to SE were associated with very low $PM_{2.5}$ levels. This means that the local
391 WD measured by the Lidar was not always in agreement with the air mass origin, i.e. to the large-scale
392 WD data used by HYSPLIT to compute the back-trajectories. For instance, the WD on 18/01 and 19/01
393 (Fig. 3d) was southwest and northwest respectively while the direction of the dominant clusters on these
394 days was NE (cluster #5a) and SE (clusters #2a and #4a). This difference could be explained by external
395 local factors such as lake-breeze or topography which lead to a sudden change in the local wind. As a
396 result, models like HYSPLIT are likely more effective than local wind data in studying the LRT.

397 *3.3 Role of the boundary layer dynamics on $PM_{2.5}$ levels during pollution events*

398 In general, the evolution of the BL during a day can be described as follows: After the sunrise
399 (recorded at 07:00 LT), the convective boundary layer (CBL) grows steadily and often reaches its
400 maximum height from 14:00 to 16:00 LT. Afterward, the sensible solar radiation decreases and the CBL
401 gradually changes into neutral boundary layer (NBL) that has a short-lived state. Around the sunset
402 (recorded at 18:00 LT), the bottom part of the NBL is transformed into stable boundary layer (SBL)
403 driven by weaker and sporadic turbulence under stable conditions (Stull, 1988). However, the BLH
404 development is strongly impeded under haze conditions because a large part of the incoming solar
405 radiation is either absorbed or scattered by the fog, by clouds or by the pollution layer, thus reducing the
406 solar flux reaching the surface and leading to increased stability (Quan et al., 2013; Leng et al., 2016).

407 Lower BLH on hazy days weakens the vertical mixing and directly results in the accumulation of aerosol
408 at the surface, leading to an increase in aerosol concentration.

409 Figure 3(f) shows that the daily maximum of the hourly BLH ranged from 700m to 1200m during
410 cloudy days, depending on cloud properties (type, thickness and altitude), while it was typically below
411 1000m during hazy days or foggy days. Unfortunately, there are less days with maximum BLH on
412 figure 3(f) than days with data availability in Table S1; this is because it was not always possible to
413 retrieve the BLH due to the low-altitude clouds and precipitation. Furthermore, it is clear that the daily
414 concentration of PM_{2.5} is higher throughout a day when the maximum BLH is lower. For example, during
415 the event P2, a sudden drop of the maximum BLH occurred from day 24 to 25/01, leading to a dramatic
416 increase of daily PM_{2.5} level on day 25/01 (Fig. 3f). Figure 7 shows the time series of BLH measured by
417 the SLIM in Hanoi on 19/01, 24/01, 28/02 and 14/03, which corresponds to the most polluted day of each
418 pollution event (P1-P4). High back-scattering coefficient values were recorded at altitudes lower than the
419 BLH suggesting that the haze pollution was formed and evolved within the BL. Thus, a fine analysis of
420 the relation between the BLH and PM_{2.5} variations will be performed for these four pollution events.

421 3.3.1 Pollution event P1: 18 - 20/01

422 The analysis of the most polluted day of event P1 (19/01) indicates that the CBL grew constantly
423 after sunrise (around 07:00 LT) when the incoming solar radiation increased. The growth rate of the
424 boundary layer was weak ($\leq 50 \text{ m h}^{-1}$) in the morning but increased gradually in midday ($\sim 80 \text{ m h}^{-1}$)
425 but remained moderate as medium altitude clouds were present above the BL until 16:00 LT (Fig. 8a).
426 The maximum BLH on this day was 660m, observed at 16:00 LT before falling gradually. After the
427 sunset, the SBL maintained at heights around 400m to 450m until the next morning and it remained
428 capped by low-level clouds. Besides BLH, it is important to take into account the ventilation
429 coefficient (VC), an indicator which combines the effects of the vertical and horizontal dispersion of
430 aerosols. VC (m^2s^{-1}) is normally defined as the horizontal wind speed integrated over the whole BL
431 depth (Iyer and Raj, 2013; Sujatha et al., 2016); here, it was estimated as the product of hourly BLH
432 and the hourly mean wind speed (WS) at 200m a.g.l.

433 Regarding PM_{2.5}, the concentration was already high ($\sim 90 \mu\text{g m}^{-3}$) at the beginning of the day
434 which could be a consequence of the cumulative process from the day before. Besides, the vertical
435 mixing was weak (Fig. 9a) due to the influence of a high-pressure system associated with low SBL
436 height, enhancing the accumulation of pollutants near the surface. During the first 2 hours of the day,
437 PM_{2.5} concentrations increased to reach $106 \mu\text{g m}^{-3}$ at 02:00 LT. Afterward, the concentration of PM_{2.5}

438 fell gradually to $68 \mu\text{g m}^{-3}$ between 02:00 and 07:00 LT although there was no significant variation of
439 BLH at that moment. This decrease could be explained by the existence of a weak low-level jet (LLJ)
440 in very stable conditions that was recorded by the Doppler Lidar between 02:00 and 05:00 LT around
441 180 m altitude (Fig. 9a). The radiosonde profile recorded at 07:00 LT (Fig. A.5a) shows the presence of
442 another weak LLJ ($2 - 3 \text{ m s}^{-1}$) located beyond the Doppler Lidar range and just below the SBL top.
443 These jets were able to result in a sudden increase of vertical mixing at lower levels, leading to the
444 dispersion of pollutants near the surface (Mahrt, 2014; Wei et al., 2018).

445 After the sunrise, the concentration of $\text{PM}_{2.5}$ started to rise following the increase of emitting
446 activities, such as vehicles or cooking, during the rush hours, which occurred in a still slowly growing
447 BL. Around 16:00 LT, the $\text{PM}_{2.5}$ levels decreased slightly when the BLH reached its maximum height,
448 before rising once again from 18:00 LT. This evening increase in PM is likely due to the combination
449 between the depletion of BL and the second rush hour also starting from 18:00 LT. Interestingly, the
450 $\text{PM}_{2.5}$ concentration continued to rise rapidly despite a considerable reduction of vehicle activities and
451 others after the end of rush hour around 20:00 LT. This $\text{PM}_{2.5}$ increase was in fact associated with very
452 weak vertical mixing as indicated by the low VC observed at this time (Fig. 9a), leading to the poor
453 dispersion of particles and the accumulation of $\text{PM}_{2.5}$ near the surface.

454 3.3.2 Pollution event P2: 23 - 28/01

455 The analysis of 24/01 shows that the CBL also grew constantly after sunrise, along with the
456 increase of incoming solar radiation in the morning (Fig. 7b). However, the growth rate of the CBL
457 during daytime on 24/01 was faster ($\sim 100 \text{ m h}^{-1}$) than that on 19/01. The maximum height was 780m,
458 observed at 15:00 LT. Then, the BLH collapsed suddenly with a higher decay rate compared to day
459 19/01. At night, this layer maintained at a stable height of approximately 300m from 22:00 LT until the
460 next early morning.

461 At midnight, the $\text{PM}_{2.5}$ concentration was high ($\sim 90 \mu\text{g m}^{-3}$) then decreased gradually (Fig. 8b)
462 until sunrise (07:00 LT). After that, the concentration of $\text{PM}_{2.5}$ increased rapidly and reached a peak at
463 10:00 LT. This could be explained by enhanced transport activities in the first rush hour, associated
464 with the low BL height in the morning. Furthermore, the wind speeds during this period were weak,
465 with values $\leq 2 \text{ m s}^{-1}$, leading to low values of VC and mixing (Fig. 9b). After 10:00 LT, the $\text{PM}_{2.5}$
466 levels decreased, along with a rapid increase of BLH, which reached its maximum height at 15:00 LT.
467 The increase in VC in this period may be attributed to the fact that both BLH and WS show an
468 ascending trend, enhancing the air quality. From 16:00 LT, the $\text{PM}_{2.5}$ concentrations tended to increase

469 slightly once again and reached a peak at 20:00 LT. This trend was again the result of a combination of
470 the decrease of BLH and the second rush hour after the sunset. Afterward, the PM_{2.5} concentration fell
471 gradually but still remained at a high level ($\sim 70 \mu\text{g m}^{-3}$) at night due to the low height of the SBL.
472 Contrary to the 19/01, no LLJ was visible on this day within the SBL, either on the wind Lidar
473 observations (Fig. 9b) or on the radiosondes profiles (at 07:00 and 19:00 LT, Fig. A.5b). In general,
474 this result is consistent with low BLH and WS measured on day 24/01.

475 3.3.3 Pollution event P3: 26 - 28/02

476 Day 28/02 was one of the rare hazy days in February, and this day was also associated with the
477 third pollution event. After sunrise, the height of the BL started to increase with a weak growth rate
478 ($\leq 50 \text{ m h}^{-1}$) from 07:00 to 11:00 LT (Fig. 8c). From 11:00 to 14:00 LT, the BLH increased rapidly and
479 reached its maximum height of 850m at 14:00 LT (more than on 19/01 and 24/01). Then, the BL
480 decayed due to the decrease of the sensible solar radiation in the late afternoon.

481 Concerning PM_{2.5} concentration, it was high at the beginning of the day ($\sim 80 \mu\text{g m}^{-3}$) and remained
482 this way until 12:00 LT (Fig. 8c). This phenomenon might be due to the low height of the SBL at night
483 and the slow growth of CBL in the morning, which weakened the mixing (Fig. 9c), leading to the
484 accumulation of PM_{2.5} near the ground. During the campaign, the BLH retrieved from 00:00 to
485 11:00 LT was generally lower than what was observed on 19 and 24 January, as it mainly varied in the
486 range of 300 - 350 m. Afterward, the concentration of PM_{2.5} started to fall gradually with the
487 simultaneous increase of the BLH, which reached its maximum altitude at 15:00 LT, leading to the
488 increase of vertical mixing near the surface (Fig. 9c). In addition, an increase of VC was also recorded
489 at the moment. More generally, higher values of VC were observed on day 28/02 (Fig. 9c) due to the
490 higher BLH. Then, the concentration of PM_{2.5} declined more rapidly after 16:00 LT (from $\sim 77 \mu\text{g m}^{-3}$
491 to less than $35 \mu\text{g m}^{-3}$) due to the increase of WS under the impact of low-pressure system. Especially,
492 LLJ located just below the SBL top ($\sim 400\text{m}$) were recorded at 19:00 LT (Fig. A.5c). Then, these LLJ
493 combined with the wind shear (Fig. 9c) to form intermittent turbulence at lower levels that was
494 recorded by the Doppler Lidar during at night around 200m to 280m a.g.l, enhancing the mixing and
495 increasing the dispersion of PM_{2.5} near the surface.

496 3.3.4 Pollution event P4: 11 - 14/03

497 Day 14/03 was the last day and the most polluted day during the period P4. Concerning the
498 boundary layer, the heights of SBL on this day was commonly higher than these on other cases (with
499 heights $\geq 500\text{m}$ from 03:00 to 07:00 LT). Furthermore, CBL evolved slowly with a rate $\leq 40 \text{ m h}^{-1}$ after

500 the sunrise. And BLH reached the highest altitude at 890m at 14:00 LT (Fig. 8d) before decreasing over
501 time as a normal scenario for the depletion of the CBL in the late afternoon. After 23:00 LT, the BLH
502 could not be retrieved because of the precipitation.

503 At the beginning of the day, the $PM_{2.5}$ concentration was lower than during the other cases
504 ($\sim 60 \mu\text{g m}^{-3}$) but it was still considered a high concentration. This result is expected considering that
505 this day had the highest value of VC of the four case studies (Fig. 9d). Later on, the $PM_{2.5}$
506 concentration suddenly increased from 03:00 LT and after two hours, it reached $\sim 130 \mu\text{g m}^{-3}$ which
507 was twice as much as its value at the beginning of the day (Fig. 8d). The Doppler Lidar shows that a
508 LLJ probably existed from 04:00 to 09:00 LT (Fig. 9d), though the top of the jet was visible only on the
509 radiosonde profile at 07:00 LT (Fig. A.5d). This jet could have carried pollutants from regional sources
510 to Hanoi, leading to the increase of $PM_{2.5}$. Also, the increased wind shear below the jet reinforced the
511 vertical mixing leading to the irruption of pollutants at the surface. Afterward, the $PM_{2.5}$ concentration
512 went down slightly but it maintained at a high polluted level ($\geq 100 \mu\text{g m}^{-3}$) until 14:00 LT, as the
513 boundary layer only grew moderately after sunrise. After the BLH reached its maximum height of
514 910m at 14:00 LT, it tended to decrease gradually and the concentration of $PM_{2.5}$ began to increase
515 until 20:00 LT because of the depletion of the BL. Then, the concentration of $PM_{2.5}$ fell suddenly by
516 the arrival of a new cold surge, which resulted in a significant increase in WS.

517 *3.4 Discussion*

518 The main goal of our study was to identify key factors explaining the pollution events during the
519 winter season in Vietnam, by considering all phenomena that could contribute to air pollution, i.e. local
520 meteorological parameters, vertical wind profiles, BLH, air mass trajectories... At a large geographic
521 scale, our results indicate a significant impact of LRT on air quality during winter 2019, especially
522 during pollution events. The increase of $PM_{2.5}$ during these events corresponds mainly to large scale
523 pollutant transport from the Northeast, under the winter monsoon's impact, which is in agreement with
524 previous studies (Hien et al., 2004; Hai and Kim Oanh, 2013; Ly et al., 2020). In addition, the pollution
525 can sometimes come from high altitude LRT of biomass burning emissions from the West, as observed
526 for the event P4. Nevertheless, the LRT of pollution alone does not explain the temporal variability of
527 $PM_{2.5}$ levels in Hanoi. A complementary analysis of the local atmospheric dynamics is necessary.

528 The impact of meteorological factors (i.e. humidity, temperature) is known to influence the
529 variation of pollutant concentrations at the surface in winter (Hien et al., 2011; Nguyen et al., 2020).
530 However, while previous studies only considered WS and WD data from ground observations, the

531 hourly vertical structure of turbulence and vertical mixing profiles were measured precisely in this
532 study, and they revealed to be determinant parameters governing the hourly variability of PM_{2.5} during
533 pollution events. The vertical mixing measured by Doppler Lidar can vary significantly with the
534 evolution of incoming solar radiation during daytime, while it can be reinforced during nighttime when
535 a LLJ appears. These vertical mixing evolutions could lead to the accumulation or dispersion of PM_{2.5}
536 at the surface, as observed during the four pollution events in winter 2019 (Figure 9). The approach
537 proposed in this study brings a more accurate view for understanding the PM variations under stagnant
538 condition in Hanoi.

539 In addition, the dynamics of PM_{2.5} concentrations in the boundary layer is closely related to the
540 height of this layer (Stull, 1988; Cohn and Angevine, 2000; Boyouk et al., 2010; Altstädter et al.,
541 2018). Thus, the study of BLH here provides for the first time a complementary analysis on PM_{2.5}
542 variations during pollution events, while there is still limited information on the BLH evolution in
543 Southeast Asia, particularly in Vietnam. The maximum BLH in Hanoi ranges from 700m to 1200m
544 with values systematically below 1000m during hazy days. Besides, having simultaneous
545 measurements of the wind profile and BLH allows to determine the ventilation coefficient, a synthetic
546 parameter characterizing the pollutant dispersion which is of high interest for air pollution monitoring
547 and assessment. The high hourly PM_{2.5} concentration during hazy days is essentially consistent with the
548 slower growth of BLH and lower ventilation coefficient. Consequently, this analysis suggests that BLH
549 is an essential parameter to understand the mechanism connecting atmospheric dynamic factors and air
550 quality in Hanoi during the winter period.

551 **4. Summary**

552 To identify the factors affecting air pollution, this study analyzed the relationship between multi-
553 scale meteorological conditions and pollution events during the winter 2019. The mean PM_{2.5}
554 concentration during the campaign that took place in Hanoi from 11/01 to 22/03/2019 was
555 $45 \pm 25 \mu\text{g}\cdot\text{m}^{-3}$. The highest daily level of PM_{2.5} was recorded on 25/01 ($129 \mu\text{g}\cdot\text{m}^{-3}$), while lower
556 levels of PM_{2.5} were mainly observed in February.

557 Four severely pollution events were recorded during the campaign, associated with stagnant
558 conditions. The results of HYSPLIT backward trajectories showed that the three first pollution events
559 were mainly related to the occurrence of LRT from China and from the northeast coastal regions of
560 Vietnam. In addition, a mix of two LRT contributions was recorded during the fourth pollution event,
561 that included emissions from the Northeast and biomass burning from the West. However, the chemical

562 composition of TSP was pretty constant over the 10 weeks of sampling, which included both periods
563 associated with LRT aerosols and other periods with only local emissions. This indicates that locally
564 emitted particles had a major chemical composition which was relatively similar to those coming from
565 LRT.

566 Besides, the aerosol Lidar observations showed a large variability in the daytime BLH according to
567 the different weather conditions: the daily maximum of the hourly BLH varied from 700m up to 1200m
568 during cloudy days, while it was typically below 1000m during hazy days. In hazy days, the height of
569 the boundary layer was compressed and grew slowly within just a few hundred meters due to the strong
570 effect of haze. Because of the lower BLH, the accumulation of pollutants at the surface was reinforced,
571 leading to severe degradations of the air quality in Hanoi.

572 Our study provides a better understanding of the mechanisms linking meteorological parameters,
573 BLH and air quality during the winter period in Hanoi. The methods and results presented here can be
574 used to further assess the contents, types and controlling factors of atmospheric dynamics on polluted
575 events in Hanoi under different weather conditions and season, or in other cities in South-East Asia.

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601 References

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FIGURES:

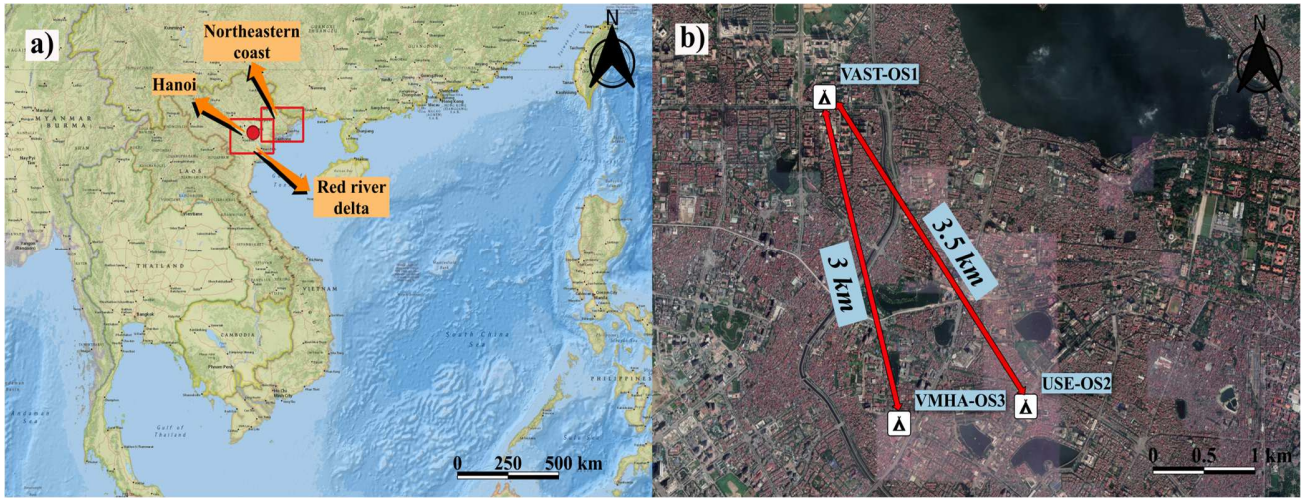


Figure 1: (a) Location of Hanoi, Red River Delta and northeastern coast of Vietnam and (b) three observation sites (VAST - OS1), (USE - OS2) and (VMHA - OS3).

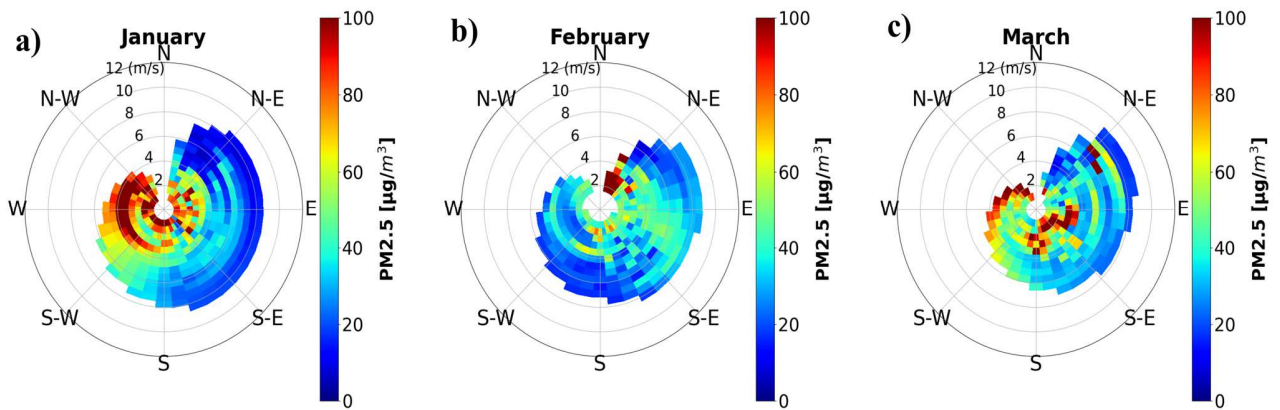


Figure 2: Bivariate polar plots of PM_{2.5} concentrations at site VAST - OS1 for each month. The radial and tangential axes represent the wind direction and speed in m s^{-1} at 100m a.g.l.

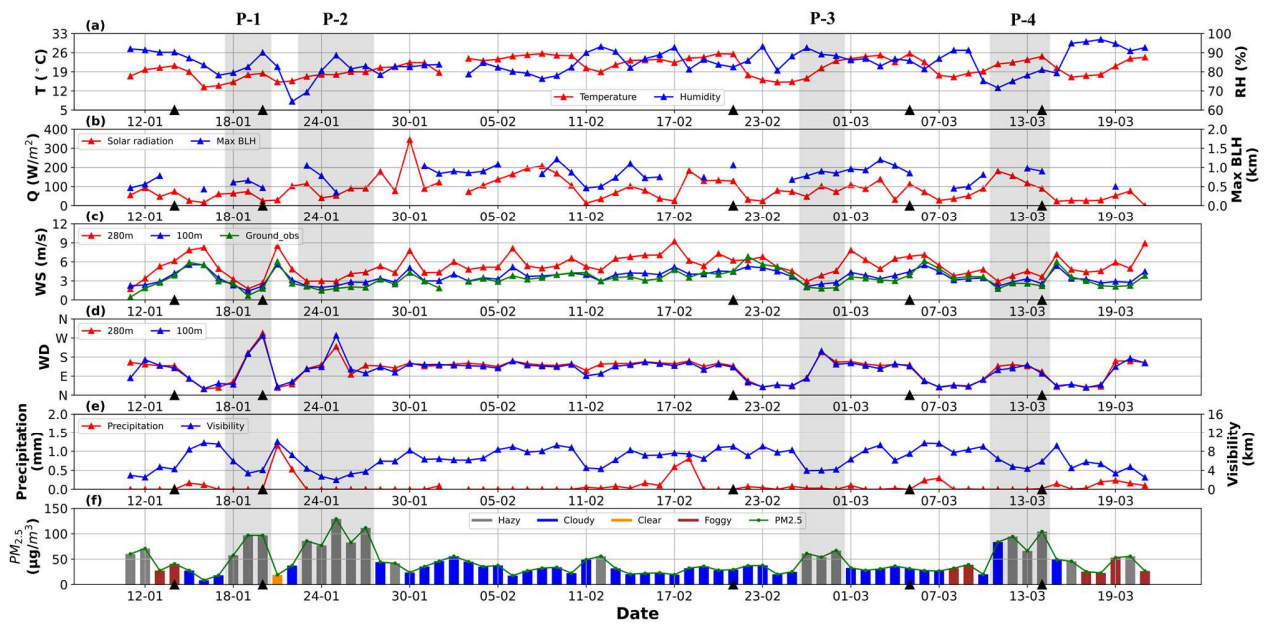


Figure 3: The daily average (a) air temperature (T) and humidity (RH), (b) Solar radiation (Q) and the daily maximum of the hourly BLH values (max BLH), (c) wind speeds (WS) at 100m, 280m a.g.l. and ground-based, (d) wind direction (WD) at 100m and 280m a.g.l., (e) precipitation and visibility, (f) PM_{2.5} concentrations. Black triangles on the horizontal axis mark the cold surges. P1 - P4 represent polluted events as grey areas.

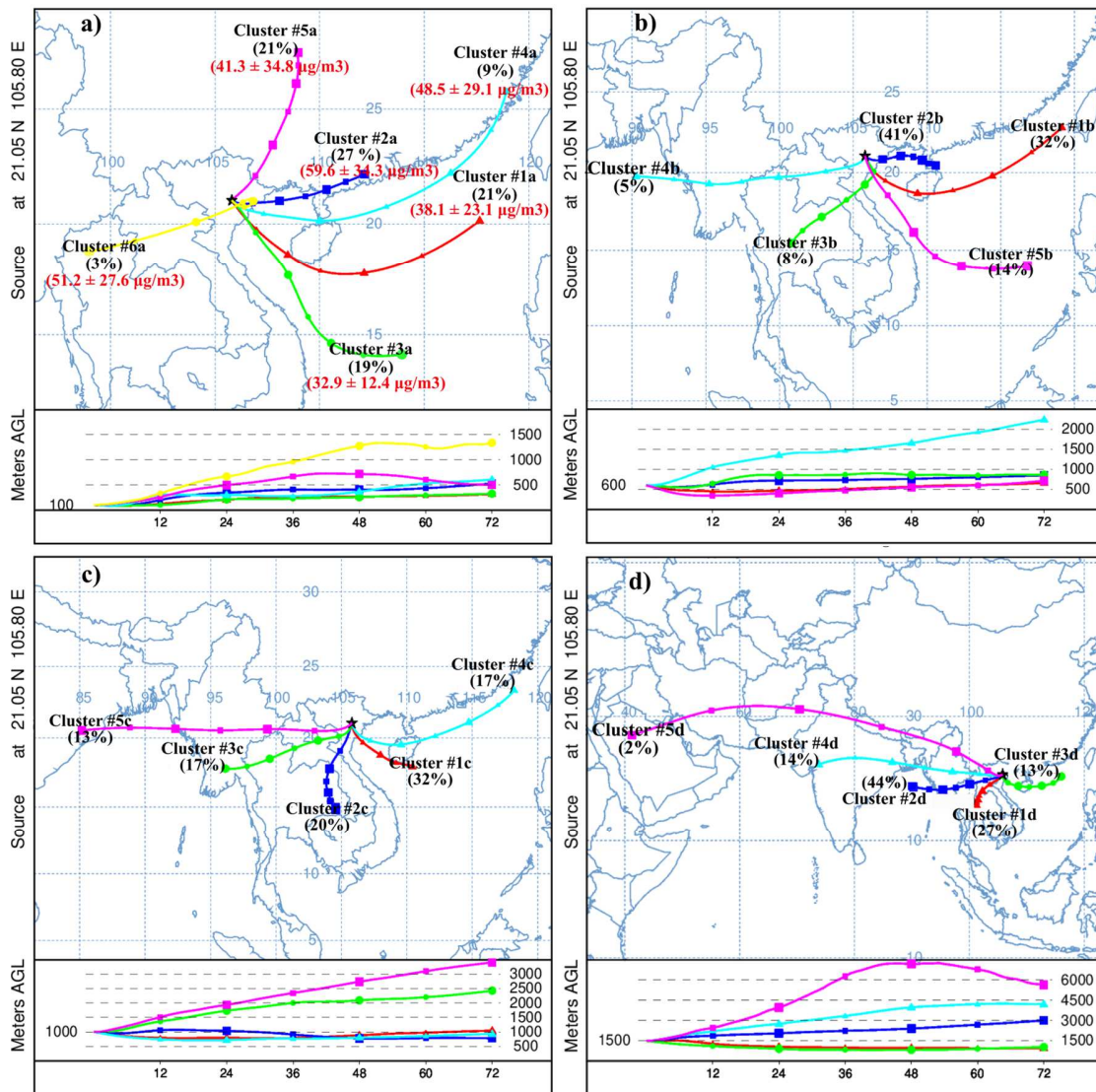


Figure 4: Cluster of HYPPLIT 72-hour back-trajectories arriving in Hanoi at heights of (a) 100 m, (b) 600 m, (c) 1000 m and (d) 1500 m a.g.l. from 11/01 to 21/03/2019.

Statistical summary of frequency and $\text{PM}_{2.5}$ concentration (mean \pm one standard deviation) as a function of clusters of air masses arriving in Hanoi ending at 100m a.g.l.

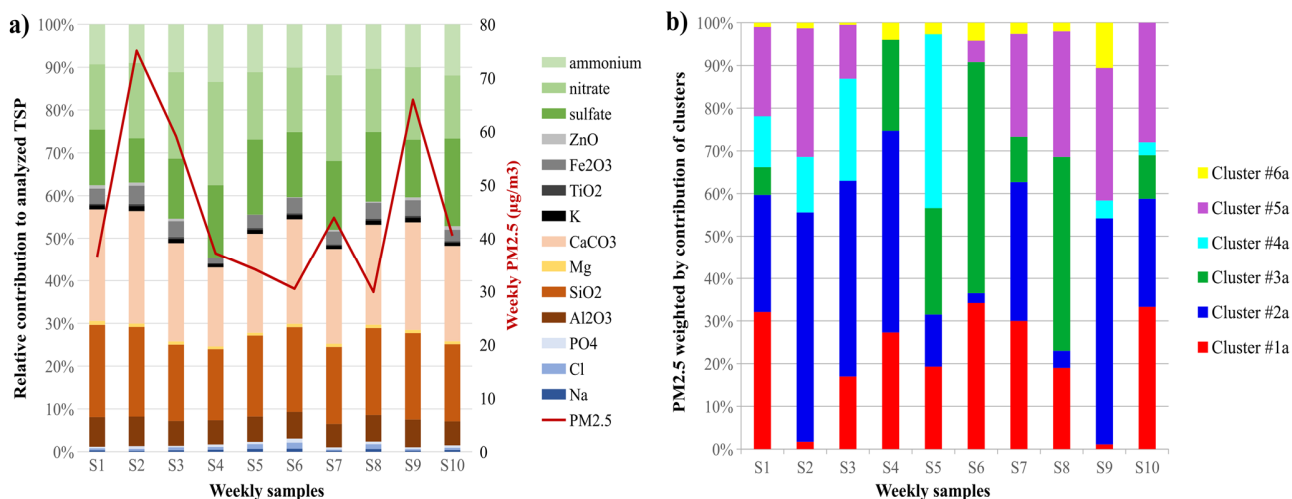


Figure 5: a) Relative chemical composition of the Total Suspended Particles (TSP) associated to the weekly samples collected at site OS1, along with the corresponding $PM_{2.5}$ concentrations ; b) relative contribution of the HYSPLIT clusters for back-trajectories ending at 100m a.g.l. in the ten weekly samples.

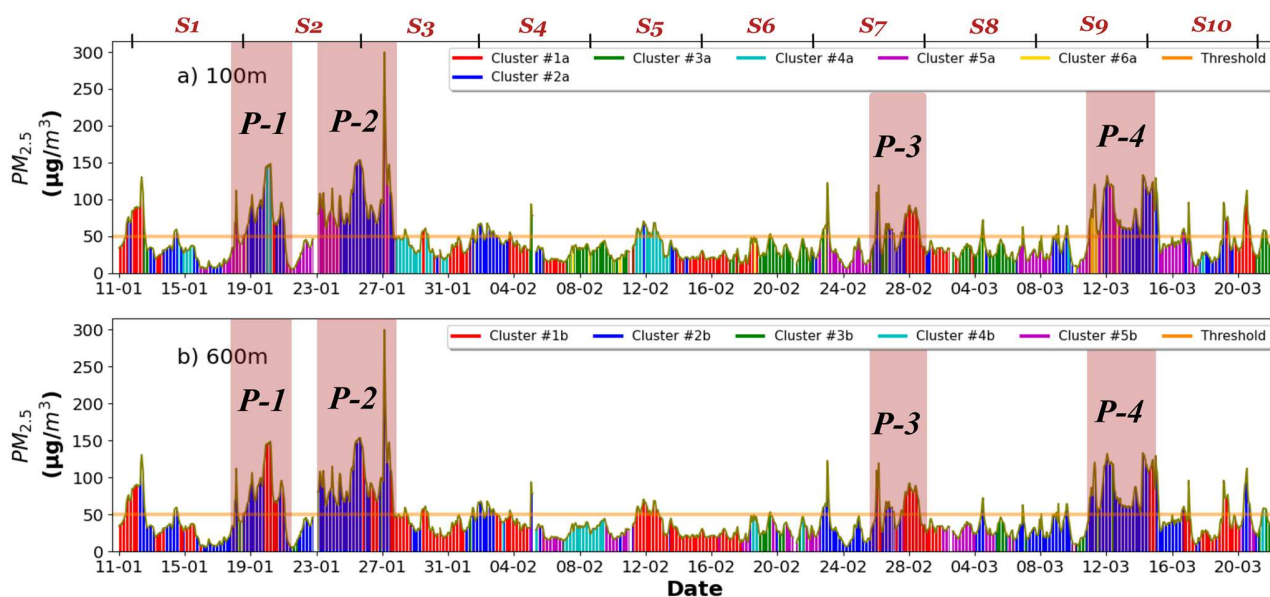


Figure 6: Hourly $PM_{2.5}$ concentrations colored as a function of the HYSPLIT cluster attribution for back-trajectories arriving at height of 100m and 600m a.g.l. Threshold (orange line) based on the specified Vietnam's standard for ambient air quality in 24-h ($\geq 50 \mu\text{g m}^{-3}$). $S_1 - S_{10}$ represent the periods of time of the 10 weekly filter samples.

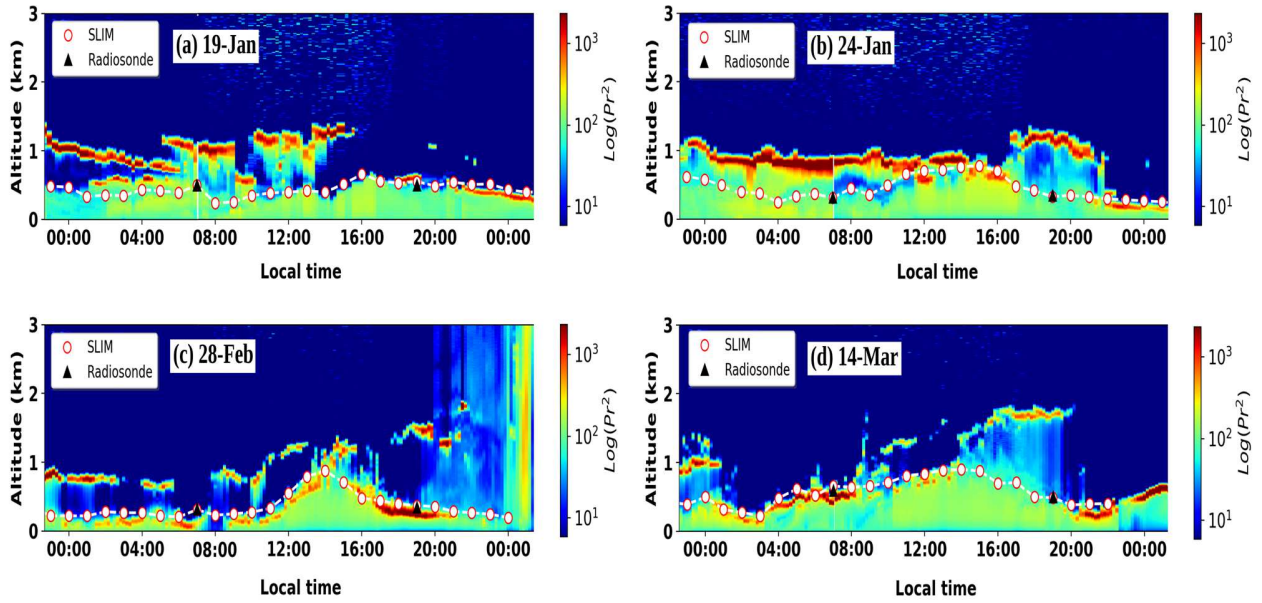


Figure 7: Time-height cross-sections of SLIM Lidar range-corrected signal (Pr^2) and time series of boundary-layer height (BLH) on (a) 19/01, (b) 24/01, (c) 28/02 and (d) 14/03. The BLH retrieved from SLIM (respectively radiosondes) profiles is represented as red circles (respectively black triangles).

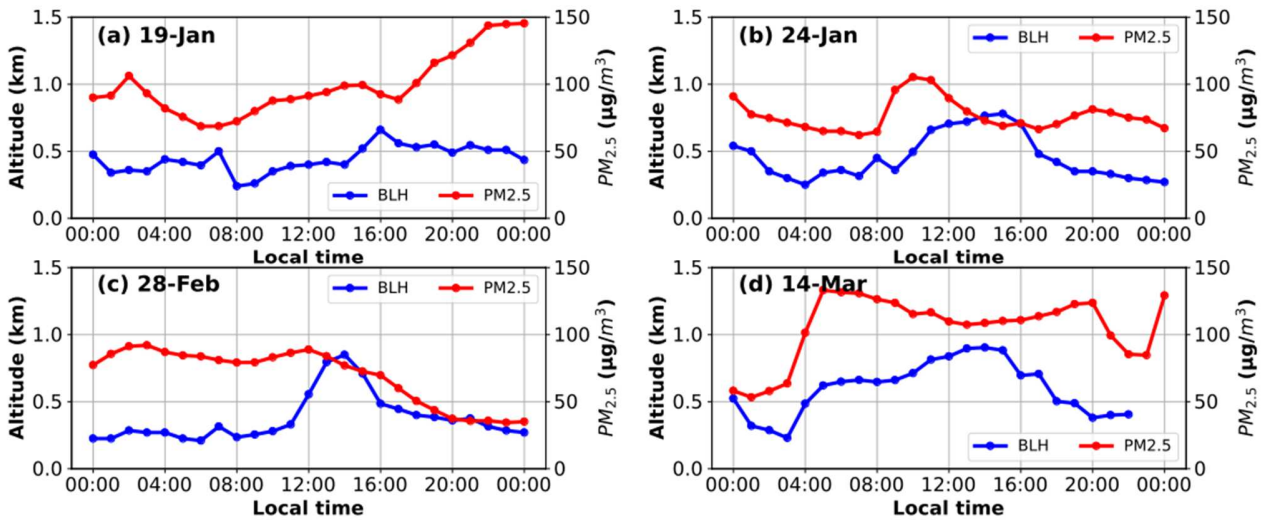


Figure 8: Hourly time series of boundary layer height (BLH) from the aerosol lidar and $PM_{2.5}$ concentrations on (a) 19/01, (b) 24/01, (c) 28/02 and (d) 14/03.

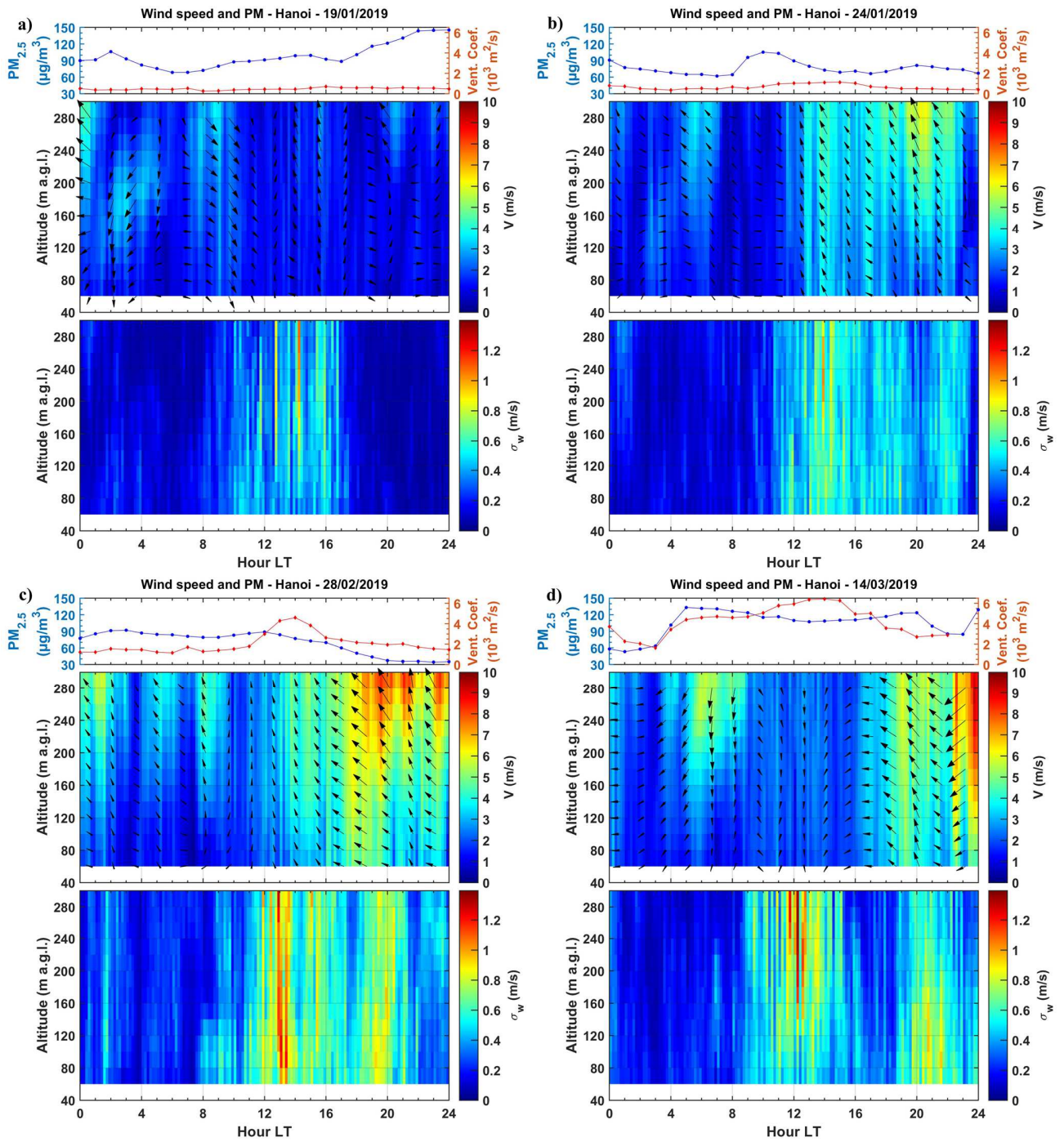


Figure 9: Time series of $PM_{2.5}$ concentration (upper) and time-height cross-sections of horizontal wind speeds (middle) and vertical mixing (below) measured by the Doppler Lidar on (a) 19/01, (b) 24/01, (c) 28/02 and (d) 14/03.

Key factors explaining severe air pollution episodes in Hanoi under the impact of the Northeast winter monsoon

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Graphical abstract

