A marked interannual variability of haze linked to particulate sources and meteorological conditions in Tehran (Iran), 1990-2020

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31 ABSTRACT

This research assessed for the first time the spatio-temporal changes of haze pollution (NHAZEs) 32 and its relationship with levels of gaseous pollutants and meteorological conditions over Tehran 33 34 metropolis (Iran) for 1990-2020. The results showed a significant decreasing trend of NHAZEs annually and in winter, along with a significant increasing trend in the horizontal visibility. 35 However, a marked interannual variability linked to changes in PM2.5 concentrations and the 36 influence of meteorological conditions was detected, which explained 65% and 30% of the 37 NHAZEs variances, respectively. We found that the increasing trend of wind speed annually and 38 in winter is the principal driver behind the decrease in NHAZEs and the increase in visibility; as 39 winds control the movement and dispersion of air pollution. In relation to gaseous pollutants, a 40 41 case study showed that the highest concentrations of PM2.5 and NHAZEs were recorded under 42 high levels of SO2, CO, and NO2, and low levels of O3, which mainly occurred under stable anticyclonic circulations. Spatially, the NHAZEs mostly affected the western, southwestern, 43 44 central, and some parts of the northern of Tehran metropolis, because of the location of industries, traffic, and lack of green areas. 45

46 KEY WORDS: Haze, visibility, trends, PM_{2.5}, meteorological influence, Tehran metropolis

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52 **1. Introduction**

Haze is a common synoptic phenomenon (World Meteorological Organization, 2019) primarily 53 caused by PM_{2.5}, in which visibility is less than 10 km, and relative humidity (RH) values below 54 55 90% (Yang et al., 2016; Shi et al., 2019). The cause of atmospheric turbidity when RH is less than 90% is the occurrence of haze as other meteorological phenomena such as rain, snow and 56 hail, which are atmospheric turbidity factors, usually occur when RH is higher than 90%. Haze 57 58 events are frequently related to particulate air pollution and low atmospheric visibility (Ding et al., 2009; Hyslop, 2009; He et al., 2018; An et al., 2019; Wei et al., 2020). Therefore, it causes 59 harmful effects such as increased deaths from cardiovascular and respiratory diseases (Kampa 60 and Castanas, 2008; Xingqin et al., 2015; Cohen et al., 2017; Liu et al., 2019), and reduced 61 safety of road, air, and rail transport (Wang et al. 2020), to name but a few. In recent years, the 62 study of haze has attracted the scientific interest due to its harmful effects on human health and 63 economic development, and many scientists around the world have studied various aspects of it, 64 including the characteristics and formation mechanisms (Li et al., 2017; Ye et al., 2019; He et al., 65 2021; Van et al., 2022), the role of anthropogenic emissions (Yang et al., 2016; Liu et al., 2022), 66 the influence of meteorological conditions (Yang et al., 2016; He et al., 2018; Shi et al., 2019; 67 Yin et al., 2021), and the impact on climate change and the effects of teleconnections (Shen et 68 al., 2018; Zhao et al., 2018; Wang et al., 2020; Yin et al., 2020; Zhao et al., 2022), among others. 69 These studies are mainly concerned about haze pollution in metropolises, with results that highly 70 depend on the features of the study area, large-scale atmospheric circulation and climate drivers 71 such as teleconnection patterns, and the proximity to pollutant emission sources. Therefore, site-72 specific studies to determine the spatio-temporal variability of haze in e.g. highly polluted 73 74 industrial cities such as Tehran (Iran), is strongly needed.

Tehran occupies a central political and economic role as Iran's capital which is one of the largest 75 and most populous cities in the world. Tehran is located between 35.35° to 35.48° N latitude and 76 51.17° to 51.33° E longitude, at a high altitude (1,200 m a.s.l.), and is surrounded by the Alborz 77 Mountain Range which strongly affects the dispersion pattern of pollutants (Safavi and Alijani, 78 2006; Ashrafi, 2012); Figure 1. The city has an area of about 730 km² with a population of 8 79 80 million (Statistical Center of Iran, 2022). According to the Köppen-Geiger climate classification, Tehran has a semi-arid climate (Raziei, 2017) and is a little cooler than other Middle Eastern 81 capitals due to its higher latitude and altitude. The air in Tehran is amongst the most polluted in 82 the world (World Bank staff based on data from WHO, 2016). Air pollution is a major problem 83 in Tehran (Shahbazi et al., 2016). Urbanization, rapid population growth, increasing fuel 84 consumption, and industrial development are pressure points for clean air in Tehran (Heger and 85 Sarraf, 2018). Tehran is one of the most polluted cities, and various studies have analyzed its 86 pollution due to the need of establishing regional pollution control strategies. According to the 87 findings of these studies, the natural conditions of the Tehran metropolis have a high impact on 88 the dispersion pattern of pollutants; e.g., local wind circulations are very effective in the 89 dispersion of air pollution (Safavi and Alijani, 2006; Ramezani et al., 2018; Pishdad et al., 2020). 90 91 Studies have shown an increase in PM_{2.5} in recent years in Tehran (Torbatian et al., 2019). Most of the studies have examined Tehran's pollution using qualitative air index data during short-term 92 93 periods (Hejazizadeh et al., 2017; Yasar et al., 2020; Ali-Taleshi et al., 2022). But to the best of 94 our knowledge, there has been no research examining the multidecadal spatio-temporal changes of haze pollution in Tehran. In view of this research gap, the overall aim of this study is to assess 95 96 for the first time the long-term changes and spatio-temporal variability of winter haze in Tehran

97 for a 31-yr 1990-2020 study period. Here we also analyze the relationship with levels of gaseous

98 pollutants and meteorological conditions.



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Figure 1: Elevation map of Tehran region with location of the meteorological stations and the average of $PM_{2.5}$ in the monitoring stations used in this study. Details for the air quality monitoring stations are given in Table 1.

104 **2. Data and methods**

105 **2.1.** The number of haze synop (NHAZEs)

Visibility, RH, and suspended particles are three attributes of haze (He et al., 2018), which cause 106 this phenomenon when visibility is less than 10 km and RH is less than 90% (Ding et al., 2014). 107 Specific codes are used in meteorological stations to express the current weather. During the 108 Coordinated Universal Time (UTC), these codes are sent to the Iran Meteorological Organization 109 (IRIMO, https://data.irimo.ir/; last accessed 27 November 2022). Code 05 is used to record the 110 haze phenomenon which occurs under the abovementioned conditions. Data for the current 111 weather code on the Synop scale (every 3 hours) for the stations analyzed in this study were 112 taken from the IRIMO. Haze events generally occur during the winter because of occurrence of 113 air temperature inversions. Air usually cools with altitude, but warm air settles above a layer of 114 cool air near the surface during an inversion. The warm air acts like a lid and traps pollutants 115 near the surface, especially in basins and valleys. Due to the maximum occurrence of haze in 116 Tehran in winter, we particularly focused on winter haze as also conducted previous studies 117 (Xiao et al., 2015; Shi et al., 2019; Zhao et al., 2020; Li et al., 2021; Zhang et al., 2021). Since 118 some stations do not report meteorological codes at night, we removed the data recorded at 06:00 119 PM, 09:00 PM, 12:00 AM, and 03:00 AM local time. Then, the haze synop (reported with code 120 05) was counted in the annual and seasonal (winter) scales for the 31-year 1990-2020 study 121 period, and the frequency (in synops) of haze was calculated. Boreal winter season is defined as 122 December, January and February. 123

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126 **2.2.** Observed air quality data

According to studies conducted around the world, haze is mainly caused by aerosol pollution (Li 127 et al., 2017; Gan et al., 2020; He et al., 2021). To explore the relationship between the variability 128 of haze and suspended particles, we received data on air pollutants from the Tehran Air Quality 129 Control Company (for locations see Fig 1. and details in Table 1). However, there is no long-130 term observation series of PM_{2.5} (in µg/m3) as the primary cause of haze (Li et al., 2017; Ye et 131 132 al., 2019) in Tehran because the PM_{2.5} concentrations have been observed and released since 2009 in Iran (only in a few stations). Since 2013, PM_{2.5} concentrations have been recorded in all 133 stations. The data is available for downloading at https://air.tehran.ir/ (last accessed 15 October 134 2022). 135

Table 1: Specifications of air quality monitoring stations used in Tehran. For locations see ID inFigure 1.

ID	Station name	Latitude (decimal degree)	Longitude (decimal degree)	Elevation (m a.s.l.)	Start year
1	Agdasiyeh	35.80	51.48	1 562	2000
2	Sharif	35.70	51.34	1,188	2012
2	Region 2	35.77	51.36	1.562	2012
3	Dorus	35.77	51.45	1,415	2010
4	Region 4	35.74	51.49	1,337	2009
5	Punak	35.76	51.33	1,467	2007
6	Tarbiat Modarres	35.72	51.38	1,287	2012
7	Setadbohran	35.67	51.30	1,162	2011
8	Golbarg	35.73	51.50	1,287	2008
9	Fath	35.67	51.33	1,137	2010
10	Region 10	35.69	51.35	1,137	2009
11	Region 11	35.67	51.38	1,137	2009
13	Piruzi	35.70	51.50	1,214	2011
14	Mahallati	35.66	51.46	1,137	2010
15	Masoudieh	35.63	51.50	1,162	2013
16	Region 16	35.64	51.40	1,093	2009
18	Shadabad	35.67	51.30	1,162	2011
19	Region 19	35.64	51.36	1,093	2009
21	Shahrrey	35.60	51.42	1,043	2005
22	Park roz	35.74	51.26	1,311	2017

138 2.3. Observed and reanalyzed meteorological data

Observed data from 4 synoptic meteorological stations in Tehran (for location names see Fig 1. 139 and details in Table 2), including three hour air temperature (T in °C), relative humidity (RH in 140 %), visibility (VIS, in m), wind speed (WS, in m s⁻¹), wind direction (WD, in °), and sea level 141 pressure (SLP in hPa) from 1990 to 2020 were obtained from IRIMO (https://data.irimo.ir/; last 142 accessed 27 November 2022). Furthermore, the European Centre for Medium-Range Weather 143 (ECMWF) ERA5 144 Forecasts reanalysis data (https://cds.climate.copernicus.eu/cdsapp/dataset/reanalysis-era5-land; last 27 145 accessed November 2022) for the 31 year period (1990-2020) were also used to apply a robust quality 146 control and homogenization protocol of the meteorological data (for details see section 2.4). 147 ERA5 is the fifth generation ECMWF atmospheric reanalysis of the global climate covering the 148 period from January 1950 to the present. The horizontal resolution of ECMWF ERA5 data is 149 $0.1^{\circ} \ge 0.1^{\circ}$, and the temporal resolution is hourly. 150

151 Table2: Specifications of the meteorological stations used in Tehran. For locations, see Fig. 1.

	Station ID	Station name	Station name Latitude		Elevation	Time period
			(decimal degree)	(decimal degree)	(m a.s.l.)	
1	99331	Tehran (Geophysic)	35.74	51.38	1,418	1991-2020
2	40751	Tehran (Shemiran)	35.79	51.48	1,549	1988-2020
3	40754	Tehran (Mehrabad Airport)	35.69	51.30	1,191	1951-2020
4	99320	Chitgar	35.73	51.16	1,305	1996-2020

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155 2.4. Homogenization of observed meteorological data

The homogenization method implemented in the R package CLIMATOL was used to 156 homogenize the meteorological data. This robust protocol detects sudden breakpoints using the 157 well-established relative Standard Normal Homogeneity Test (SNHT) (Alexandersson, 1986), 158 which has effectively been used in previous homogenization research (Azorin-Molina et al., 159 2014, 2017; to name but a few). The R Climatol homogenization package (Guijarro, 2018) 160 performs its process in three main stages, with much iteration within them. The first two are 161 devoted to removing unwanted outliers and splitting the series found most inhomogeneous into 162 two fragments at the term where the SNHT was more significant. Successive iterations refine the 163 process until no outlier nor are shift detected over preset thresholds. In the first stage, SNHT is 164 applied on stepped overlapping windows along with the series to diminish possible masking 165 problems when several shifts are present, while in the second stage, SNHT is applied over the 166 whole series, getting all the power of the test. These outlier rejections and shift detection are 167 performed over a series of differences (anomalies) between the observed data and a synthetic 168 reference series built from a number of nearby grid-points in ERA-5, in both cases in normalized 169 form. Normalization can be chosen within full standardization (the default), ratio to the mean, or 170 difference to the mean. The last third stage is dedicated to assigning their synthetic values to all 171 the missing data in all the series and sub-series that originated in the splitting process. The 172 number of detected and corrected break-points was 28 in the VIS data series, 17 in the WS data 173 series, 16 in the RH data series, and 9 in the T data series. WD data series did not pass any 174 homogenization protocol due to its complexity and were only used for analyzing a study case. 175 After completing the robust quality control and homogenization protocol, 4 homogeneous 176 meteorological series were used for 1990-2020. 177



Figure 2: Flow-chart of the Climatol protocol to quality control and homogenize meteorological
data, showing its nested iterative processes. SNHT is the abbreviation for the Standard Normal
Homogeneity Test.

202 **2.5. Trend analysis and statistics**

Sen's Slope Estimator test is recommended by the World Meteorological Organization as part of the trend detection in hydrometeorological data (WMO, 2019). The trend is supposed to be linear in this test and indicates the quantification of temporal change (Aditya et al., 2021). The Sen's Slope equation for a number of N data sample pairs is written as follows (Juraj et al., 2009):

$$207 \qquad \varrho = \frac{X_t - x_t}{t - s}$$

208 Where X_t and X_s are the observed data at times t and s, respectively. Calculate the C_a parameter 209 at the trust levels tested using the following formula:

210 $C_a = Z1 - \partial/2 \sqrt{Var(s)}$

Where z is the standard normal distribution statistic, this statistic is considered 1.96 and 2.58 for 95% and 99% confidence levels, respectively. Finally, the upper and lower confidence limits are calculated using the following formula:

214
$$M_1 = \frac{N^{\square} - C_a}{2}$$

$$215 \qquad M_2 = \frac{N + C_a}{2}$$

N is the number of slopes calculated in the first part. Previous studies have investigated hazy days to discover the existing trend of the haze phenomenon. But in this study, the unit intended for measuring haze is the synop, which is recorded every three hours.

The Mann-Kendall method is a non-parametric method that examines the presence or absence of change in parameters in a time series (Mann 1945; Kendal 1975). A factor that affects trend detection in a series is the presence of positive or negative autocorrelation (Yue *et al.* 2003; Novotny and Stefan, 2007). The statistics (S) is defined as:

223
$$S = \sum_{k=1}^{n-1} \sum_{j=k+1}^{n} sgn(x_j - x_k)$$

224 Where *N* is the number of data points. Assuming $(xj - xi) = \theta$, the value of sgn (θ) is computed as 225 follows:

226
$$\operatorname{Sgn}(\mathbf{x}) = \begin{cases} +1 & if \quad (x_i - x_k) > 0\\ 0 & if \quad (x_i - x_k) = 0\\ -1 & if \quad (x_i - x_k) < 0 \end{cases}$$

This statistic represents the number of positive differences minus the number of negative differences for all the differences considered. For large samples (N > 10), the test is conducted using a normal distribution, with mean and variance as follows:

230 Var =
$$\frac{n(n-1)(2n+5) - \sum_{i=1}^{m} t(t-1)(2t+5)}{18}$$

Here n is the number of tied (zero difference between compared values) groups, and t_k is the number of data points in the k_{th} tied group. The standard normal deviate (Z statistics) is then computed as:

234
$$Z = \begin{cases} \frac{S-1}{\sqrt{var(s)}} & if \quad S > 0\\ 0 & if \quad S = 0\\ \frac{S+1}{\sqrt{var(s)}} & if \quad S < 0 \end{cases}$$

Stepwise linear regressions were used for three-hour PM2.5 concentrations over Tehran to 235 236 estimate the relative contribution of each meteorological parameter to the variations in wintertime PM2.5 concentrations (as the primary cause of NHAZEs). Regressors include 237 normalized 3-h WS (m s⁻¹), T (c), RH (%), and SLP (hPa). 3-h wintertime PM_{2.5} concentrations 238 239 were used as the dependent variable in the regression model. Since the data measurement scale 240 was different, the Min-Max normalization method was used to change the scale of data and put 241 them in the numeric range of (0, 1). The min-max method can be expressed as follows (Zhao et 242 al., 2021):

243
$$x \text{ (normalized)} = \frac{x - \min(x)}{\max(x) - \min(x)}$$

244 Some other statistical methods are used in this study, including Pearson correlation analysis to

determine the correlation significance of the data (Zhao et al., 2020; Zhang et al., 2015) and a

246 10-year Gaussian low-pass filter to evaluate the interannual variability.

247 **2.6. Interpolation of NHAZEs and PM_{2.5}**

The spatial distribution of PM2.5, and NHAZES have been depicted using inverse distance 248 weighted (IDW) interpolation. The basic principle of IDW interpolation is to use a weighted 249 linear combination set of sample points, and it counts on both statistical and mathematical 250 methods to construct surfaces and predict unmeasured points (Khouni et al, 2021). Since IDW is 251 a weighted distance average, the average cannot be greater than the highest or less than the 252 lowest input (ArcGis 10.8 Desktop Help, 2021). The frequency of haze recorded at a station 253 includes a wide area (not just around the station) due to the large-scale occurrence of this 254 phenomenon. Previous research on air pollution and pollutant concentrations has used IDW 255 method (e.g., Ranjbar and Bahak, 2019). 256

257 **3. Results**

258 3.1. Temporal changes in NHAZEs, PM_{2.5} and visibility

The time series of annual and winter mean NHAZEs in Tehran are presented in Figure 3, showing a marked interannual variability of NHAZEs. The Gaussian low-pass filter clearly shows 5 phases for 1990-2020: (i) a significant decline with a rate of -50.7 synops year ⁻¹ on an annual scale (p<0.01) and -18.2 synops year ⁻¹ in winter (p<0.05) from 1990 to 1996; (ii) a

significant increase with a rate of +8 synops year $^{-1}$ on an annual scale (p<0.05) for 1997-2001, 263 and +4.6 synops year ⁻¹ in winter (p>0.05) from 1997 to 2005; (iii) a significant decrease with a 264 rate of -12.5 synops year $^{-1}$ on an annual scale(p < 0.05) for 2002-2012, and -3.6 synops year $^{-1}$ in 265 winter (p < 0.05) from 2006 to 2011; (iv) a non-significant increase with a rate of +12.7 synops 266 year ⁻¹ on an annual scale for 2013-2016, and +7.5 synops year ⁻¹ in winter (p < 0.01) for 2012-267 2015; and (v) a non-significant decrease with a rate of -5.06 synops year ⁻¹ on an annual scale, 268 and -6.06 synops year ⁻¹ in winter from 2016-2020. For the entire series, the winter NHAZEs 269 displayed an overall significant (p < 0.05) decreasing trend of -0.5 synops year ⁻¹, with an average 270 of 189 synops per season, and -3.5 synops year $^{-1}$ (p<0.05) on the annual scale, with an average 271 value of 386 synops per year. Since haze occurs more frequently in winter, the interannual 272 variations of annual NHAZEs resemble the winter NHAZEs. 273



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Figure 3: The time series of annual (black line) and winter (red line) NHAZEs over Tehran for
1990–2020. A 10-year Gaussian low-pass filter is drawn to highlight the interannual variability.

Figure 4 shows the annual and winter time series of average NHAZEs and $PM_{2.5}$ concentrations for 2013 – 2020. We found a strong and significant (p<0.05) positive relationship between both parameters in winter (R= 0.81) and annually (R= 0.55). This demonstrates that the interannual variability of both parameters was closely linked to each other (whit an exception in 2020). In particular, the lowest PM_{2.5} concentrations and NHAZEs occurred in 2014 and 2018 in winter, whereas the highest one took place in 2017.



Figure 4: Annual and winter time series of average NHAZEs and PM_{2.5} concentrations for 20132020.

Figure 5 presents the time series of observed annual and winter mean visibility and NHAZEs over Tehran for 1990-2020. Overall, the annual and winter mean visibility showed a significant increasing trend of +6.5 m year $^{-1}$ (*p*<0.05) and +29.2 m year $^{-1}$ (*p*<0.05) for 1990-2020, respectively. This increase in visibility is in line with the opposite decrease shown by NHAZES.

In addition, the average of visibility was estimated in 9,244.3 m year ⁻¹ (annual) and 7,834.1 m 290 year ⁻¹ (winter) for the entire 31-year period. Regarding the interannual variability, the winter 291 mean visibility clearly displayed an opposite trend behavior compared with NHAZES (R = -0.55; 292 p < 0.01), which experienced five changing phases: (i) an increase with a rate of +79.8 m year ⁻¹ 293 (non-significant) from 1990 to 1998; (ii) a decline with a rate of -172.98 m year ⁻¹ (non-294 significant) from 1999 to 2004; (iii) an increase with a rate of +228 m year $^{-1}$ (p<0.01) from 2005 295 to 2010; (iv) a decline with a rate of -6.5 m year $^{-1}$ (non-significant) from 2011 to 2015; and (v) 296 an increase with a rate of +345.28 m year ⁻¹ (p < 0.05) from 2016 to 2020. In the annual mean 297 visibility, the Gaussian low-pass filter clearly shows 3 phases for 1990-2020: (i) an increase with 298 a rate of +86.6 m year ⁻¹ (non-significant) from 1990 to 1994; (ii) a decline with a rate of -116.6 299 m vear $^{-1}$ (p<0.01) from 1995 to 2002; and (iii) an increase with a rate of +31.9 m year $^{-1}$ 300 (*p*<0.01) from 2003 to 2020. 301



302

303 Figure 5: The time series of annual and winter mean NHAZEs (black line) and visibility (blue

line) averaged over Tehran for 1990–2020. A 10-year Gaussian low-pass filter is drawn to
highlight the interannual variability.

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307 3.2. Spatial means in NHAZEs, PM_{2.5} and visibility

The spatial distribution of the annual and winter mean NHAZEs, PM2.5 concentrations and 308 visibility has been depicted in Figure 6. Figures 6a (annual) and 6b (winter) show that the mean 309 NHAZEs in Tehran reaches the highest value at the Mehrabad Airport station, in the western part 310 311 of the study area, and the lowest one in Shemiran station in the northeast of Tehran. In addition to the Mehrabad Airport station, with a winter and annual averages of 289 and 616 NHAZEs, 312 respectively, the Geophysics station also has a high frequency of this phenomenon with winter 313 314 mean of 230 and annual mean of 574 NHAZEs. As can be seen in the Figures 6c and 6d, the highest amount of visibility occurs in areas with low NHAZEs and vice versa. Figures 6e and 6f 315 show that the highest concentrations of PM2.5 were found over the west and southwest of the 316 region, where industries have developed quickly compared with other areas in Tehran. In 317 contrast, the high-elevation northern areas have low PM2.5 concentrations and high atmospheric 318 visibility. Overall, the areas with the highest frequency of NHAZEs and highest concentrations 319 of PM_{2.5} have the lowest atmospheric visibility. 320



Figure 6: Spatial distribution of annual and winter mean NHAZEs (synops) (a and b); visibility
(m) (c and d); and PM_{2.5} concentrations (µg/m3) (e and f) in Tehran averaged over 1990–2020
(except for PM_{2.5}, 2013-2020).

326 3.3. Spatial trends in winter NHAZEs and visibility

Figures 7 focuses on the spatial distribution of winter trends in NHAZEs and visibility for 1990-327 2020, which are no significant in 80% of the study area. A non-significant increasing trend in 328 NHAZEs (2 synops year ⁻¹) was observed in the northern parts of the region, while central areas 329 of Tehran showed a non-significant decreasing trend (-1.9 and -0.6 synops year ⁻¹). This strong 330 spatial variability in trends is shown e.g. by the non-significant decreasing trend at Mehrabad 331 Airport station (-1.4 synops year ⁻¹), while the Chitgar station (located west of the city, at 13 km 332 from the airport) showed a significant and highest increasing trend of NHAZEs (+6 synops year -333 ¹, p < 0.05). The rate of winter NHAZEs changes varies between -1.9 and +8.6 synops year ⁻¹ in 334 the measuring stations. In contrast to NHAZEs, spatial trends of horizontal visibility are positive 335 and significant in most parts of the study area; except for the Shemiran station located in the 336 northeast of Tehran, which displayed a non-significant decreasing trend. Overall, the rate of 337 changes for the winter visibility varies between -4.0 and +54.5 m year $^{-1}$ in different places. 338

339



Figure 7: Spatial distribution of the sign, statistical significance and trends of winter NHAZEsand visibility in Tehran for 1990-2020.

344 3.4. Meteorological conditions influencing winter NHAZEs, PM_{2.5} and visibility

To identify atmospheric conditions associated with winter NHAZEs and explain how these 345 conditions differ from non-NHAZEs, Figure 8 presents the meteorological parameters under both 346 situations. T in NHAZEs is higher on average than in non-NHAZEs, and it showed an increasing 347 trend for NHAZEs (+0.05°C year $^{-1}$; p<0.05) of a smaller magnitude than in non-NHAZEs 348 $(+0.06^{\circ}C \text{ year}^{-1}; p < 0.01)$ for 1990-2020. WS in NHAZEs is lower on average than in non-349 NHAZEs, and the increasing trend for NHAZEs (+0.04 m s⁻¹ year ⁻¹; p < 0.01) is higher than for 350 non-NHAZEs (+0.03 m s⁻¹ year ⁻¹; p<0.01), which has reduced NHAZEs in recent years. The RH 351 has the same behavior as WS, with lower values on average in NHAZEs than in non-NHAZEs. 352 The decrease in RH started from 1998, being the rate of change of this decrease in NHAZEs (-353 354 0.35% year ⁻¹; p < 0.01) smaller than in non-NHAZEs (-0.45% year ⁻¹; p < 0.01). SLP in NHAZEs

is higher on average than in non-NHAZEs, and it showed a non-significant decreasing trend for
NHAZEs (-0.01 hPa year ⁻¹), and a non-significant increase in non-NHAZEs (0.003 hPa year ⁻¹)
for 1990-2020. Overall, we found that meteorological parameters differ between NHAZEs and
non-NHAZEs, with high T, weak WS, low RH and high SLP dominating days with haze
pollution across Tehran in winter.



Figure 8. The mean winter air temperature (T), wind speed (WS), relative humidity (RH), and
sea level pressure (SLP) of NHAZEs and non-NHAZEs in Tehran for 1990-2020. A 10-year
Gaussian low-pass filter is drawn to highlight the interannual variability.

364	Table 3 summarizes the regression coefficients for the stepwise regression models. The
365	regression coefficient for WS was negative (-0.30), indicating that decreases in WS led to
366	increases in $PM_{2.5}$ concentrations. The T (+0.25) and SLP (+0.25) had a positive regression
367	coefficient, which indicates the direct relationship of these variables to the $PM_{2.5}$ concentrations;
368	that is, the increase in the T and SLP increased the $PM_{2.5}$ concentrations. The noteworthy point is
369	the low regression coefficient for the relative humidity (0.09), which shows the small effect of
370	this variable on NHAZEs. Therefore, high T and SLP, weak WS, and (almost low RH), enhance
371	high PM _{2.5} concentrations, resulting in strong haze events. Regression analysis showed that
372	meteorological variables explain 30% of $PM_{2.5}$ concentrations, with WS (R ² = -30%) having the
373	highest impact on NHAZEs in the long term, and $PM_{2.5}$ concentrations explain 65% of NHAZEs.
374	Table 3: Regression Coefficients for Stepwise Linear Regression Models Performed with

1 able 3: Regression Coefficients for Stepwise Linear Regression Models Performed with

375	Spatially	Averaged M	leteorological	Variables and PM _{2.5}	Concentrations	for 4	l synor	otic
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Regression Coefficients								
Meteorological Parameters	Chitgar (West of Tehran)	Geophysic (Center of Tehran)	Mehrabad Airport (Center of Tehran)	Shemiran (Northeast of Tehran)				
WS (ms ⁻¹)	-0.20	-0.17	-0.30	-0.15				
T (°C)	0.32	0.33	0.19	0.19				
RH (%)	0.14	0.27	0.23	0.06				
SLP (hPa)	0.24	0.14	0.21	0.22				

376 meteorological stations in Tehran

377

Lastly, to better understand the extent of the impact of meteorological conditions on the haze events in Tehran, we analyzed three time periods based on the temporal variations in NHAZEs (see Figure 3). We selected the periods with relatively low NHAZEs, that is, 2006-2011 (Period

1), large NHAZEs, that is, 2012-2016 (Period 2), and again low NHAZEs that is, 2017-2020 381 (Period 3) for comparison. There were noticeable differences in the means of the four 382 meteorological parameters (i.e., T, WS, RH, and SLP) among the periods (Table 4). Moreover, 383 the standard deviations of the four meteorological parameters were larger in Period 1 compared 384 to Periods 2 and 3, demonstrating that the meteorological conditions had larger variations during 385 the winters for 2006–2011. RH decreased significantly (p<0.05), whereas T increased 386 significantly in Period 2 compared to Period 1. RH was higher, whereas T was lower in Period 3 387 compared to Period 2. These changes may explain the increased role of climate and 388 meteorological conditions on haze pollution after 2013 in Tehran. 389

390 Table 4: The mean and standard deviations in the meteorological parameters for the different391 time periods in the Tehran metropolis

$\xrightarrow{\text{Time period}}$	Period 1 (2006-	-2011)	Period 2 (2012-2	2016)	Period 3 (2017-2020)		
Meteorological parameters	Mean	SD	Mean	SD	Mean	SD	
$\Psi_{WS (m/s)}$	1.62	0.18	1.78	0.02	1.77	0.14	
T (°C)	4.43	2.02	5.94	1.03	5.79	0.63	
RH (%)	59.30	4.05	54.77	1.27	56.30	2.12	
SLP	1019.76	2.29	1020.14	1.26	1019.46	1.03	

392

393 3.5. Winter haze case study: January 2017

According to Figure 3, the highest frequency of NHAZEs (during last decade), occurred in winter 2017, particularly in January. Figure 9 presents the daily series of $PM_{2.5}$ measurements, concentrations of gaseous pollutants (SO2, CO, NO2, and O3), and meteorological parameters

from the 1st till 30st January 2017 (For January 31, the data related to pollutants was not 397 recorded). PM_{2.5} concentrations were ranged from 46 to 177 µgm³. The highest value of PM_{2.5} 398 was recorded at 9:00 (i.e., rush hours in Tehran) on January 9 (177µg/m³), when RH was 30 % 399 and O3 was 11µg/m³. The T was high (10.8 °C), and WS was at 2 m s⁻¹ throughout that day. The 400 NO2 value was $45\mu g/m^3$, which was considerably higher than the average ($25.8\mu g/m^3$). On 401 402 January 9, the highest amount of PM_{2.5} was recorded when the amount of T, SLP, SO2, CO, and NO2 were high, and the WS, RH, and O3 were low. Conversely, the minimum value of PM_{2.5} 403 was at lower SO2 (10µg/m³), CO (1 mg/m³), and a higher concentration of O3 (24.0 µg/m³). We 404 also detected that CO levels rise a few hours after PM_{2.5} levels increase, reaching 10 mg/m3. 405 Since SO2, CO, and NO2 were primary emissions from similar sources such as coal combustion 406 and industrial processes, they showed similar variations in their concentrations. However, O3 407 showed the opposite trend. 408

409



Figure 9. Time series of daily means of $PM_{2.5}$ concentrations, gaseous pollutants (SO2, CO, NO2, and O3), T, SLP, RH, and WS (colored by wind direction, note 0, 90, 180, and 270 refer to northerly, easterly, southerly and westerly winds, respectively). Note that the gray rectangles represent the days with the highest $PM_{2.5}$ concentrations and NHAZEs (note that the synop associated with haze are marked with blue rectangles).

As part of our analysis of January's pollution, we identified three polluted periods with different 417 meteorological conditions (based on the criteria that the concentration of PM_{2.5} is higher than 120 418 419 ug/m3 and at least 4 out of 8 synop in a day have reported haze). Table 5 summarizes the average, minimum, and maximum values of pollutant gases and meteorological parameters for 420 the three polluted periods, the clean periods (which are recorded between polluted periods), and 421 the whole month of January. The average RH and WS in period 1 are low compared to periods 2 422 and 3. The highest T was also recorded in the period 1. There was no noticeable difference in 423 SLP during these periods. Except for O3, which experienced its maximum value in period 2, the 424

other pollutants had the highest values during period 1. The duration of haze was longer under low RH, weak WS, and high T (the first period lasted seven days). In periods 2 and 3, with the change in meteorological parameters (increased RH and WS and decreased T), the concentration of pollutants decreased, and these periods lasted shorter, indicating the key role played by meteorological parameters in controlling haze in Tehran.

- 430 Table 5: Comparison of major pollutants and meteorological parameters among three pollution
- 431 periods and the clean period in January 2017
- 432

	Polluted period1 (6 th 6:00 - 12 th 9:00)		Polluted period2 (16 th 6:00 - 20 th 18:00)		Polluted period3 (24 th 9:00 – 27 th 21:00)		Clean period			January (total)					
	Ave.	Min	Max	Ave.	Min	Max	Ave.	Min	Max	Ave.	Min	Max	Ave.	Min	Max
PM _{2.5} (ug/m3)	155.7	114.0	177.0	137.4	113.0	159.0	120.2	100.0	149.0	77.0	46.0	100.0	133.0	46.0	177.0
PM ₁₀ (ug/m3)	95.2	69.0	117.0	77.0	60.0	91.0	64.5	44.0	85.0	55.3	48.0	60.0	78.5	44.0	117.0
SO2 (ppb)	25.9	15.0	49.0	14.7	6.0	18.0	13.3	5.0	23.0	8.7	6.0	11.0	17.1	5.0	49.0
NO2 (ppb)	32.8	22.0	47.0	24.6	16.0	35.0	20.5	17.0	26.0	19.8	15.0	24.0	25.8	15.0	47.0
O3 (ppb)	17.3	11.0	26.0	19.3	13.0	28.0	15.7	10.0	20.0	22.8	18.0	29.0	18.7	10.0	29.0
CO (ppm)	3.1	2.0	5.0	2.9	2.0	4.0	2.5	2.0	3.0	2.2	1.0	3.0	2.7	1.0	5.0
T (°C)	7.0	0.5	14.6	5.7	1.0	10.8	5.1	0.4	9.9	4.4	-3.4	12.5	5.4	-3.4	14.6
RH (%)	44.2	21.0	70.0	50.2	26.0	77.0	51.5	36.0	77.0	51.6	15.0	93.0	49.5	15.0	93.0
WS (ms ⁻¹)	1.9	0.0	6.0	1.9	0.0	4.0	2.6	0.0	6.0	3.1	0.0	10.0	2.5	0.0	10.0
SLP (hPa)	1019	1009	1027	1021	1015	1024	1021	1010	1031	1018	1010	1032	1019	1009	1032

⁴³³

434 As mentioned above, January 9th 2017 recorded the peak of pollution in the Tehran metropolis.

435 The PM_{2.5} concentrations reached the highest value (177 μ g/m³), and all eight synops (once every

436 three hours) reported haze across all stations. Figure 10a displays the SLP on that day, showing a

high-pressure system (<1025 mb) located in the northwest of Iran, with the studied area 437 dominated by its clockwise currents so that the T (Figure 10b) of the high-pressure core reached 438 -4°C and varied between 4 and 8°C in Tehran. The dominance of high pressure conditions 439 caused the loss of humidity and increase in T due to the subsidence of the air to the lower layers 440 of the atmosphere. Combined maps of geopotential height and omega level at 850 and 700 hPa 441 442 (Figures 10c and 10d) indicate the formation of a low pressure over the Mediterranean. The study area was affected by the eastern part of this low pressure system, which moved warm air 443 from low latitudes to Iran driven by the counterclockwise movement. The positive omega values 444 indicate stability. Moreover, the combined maps of specific humidity and wind vector at 850 and 445 700 hPa (Figures 10e and 10f) also showed patches of low specific humidity (4 to 6 g/kg) around 446 Tehran. The dominance of weak southerly and southwesterly winds were also responsible of this 447 extreme pollution event, as suspended particles were trapped in Tehran. 448



Figure 10: Map of SLP (a) and T (surface) (b), geopotential height and omega at 850 (c) and 700 (d) hPa, and specific humidity and wind vector map at 850 (e) and 700 (f) hPa for January 9th 2017. Note that in the maps above, the measurement units are as follows: Slp (hPa), T (°C), omega (Pa/s), and specific humidity (g/kg). The black rectangle shows the location of Iran and the red dot shows the location of Tehran.

455 4. Discussion

Haze pollution is hazardous to human health and economic development (Hao et al., 2021; Gan et al., 2021; Feng and Yuan., 2021; Liu et al., 2019). Further research to better assess this phenomenon in highly polluted cities such as Tehran (Iran) is essential to develop regional pollution control strategies. Using observations of PM_{2.5} concentrations, NHAZEs based on

visibility and relative humidity and meteorological observations, this research investigated for 460 the first time the spatio-temporal variation of haze pollution, its relationship with gaseous 461 pollutants and meteorological conditions over Tehran for 1990-2020. The trend in NHAZEs 462 revealed significant (p < 0.05) decline of -3.5 synops year ⁻¹ annually and -0.5 synops year ⁻¹ in 463 winter (p < 0.05) across Tehran for 1990–2020. The long-term NHAZEs time series showed five 464 465 short phases of change, denoting a marked interannual variability. Considering that PM_{2.5} data was not recorded before 2013, a major constraint of this study was to quantify NHAZEs changes 466 in relation to PM_{2.5} for the entire period; we alternatively focused on the relationship between 467 NHAZEs and meteorological variables for investigating the drivers behind their variations for 468 the early decades. Moreover, the visibility and NHAZEs had a relatively high and negative 469 correlation coefficient (-0.55, p < 0.01), i.e. with an opposite trend behavior and showing five 470 opposite changing phases (Yang et al., 2016; Zhao et al., 2020). Overall, the annual and winter 471 mean visibility both showed a significant increasing trend from 1990 to 2020, with a rate of +6.5 472 m (p < 0.05) and +29.2 m (p < 0.05), respectively. 473

474 The investigation of the relationship between PM_{2.5}, the principal cause of haze, and NHAZEs evidenced a strong and positive correlation (R= 0.81, p<0.01) for 2013-2020. 475 However, we found a exception in 2020 as high PM2.5 concentrations and low NHAZEs 476 occurred, which could be explained by the changes in relative humidity or in the chemical 477 components of PM_{2.5} affecting aerosol optical characteristics (He et al., 2018). For this short 478 period, we found that the increasing tendency of PM_{2.5} concentrations is consistent with the 479 overall increase reported by the Environmental Research Institute - Air Pollution Research 480 Center (https://ier.tums.ac.ir ; Tehran Air Quality Control Company, 2021, https://air.tehran.ir/; 481 last accessed 15 October, 2022) 482

483 Examining the spatial variation of NHAZES, PM_{2.5} concentrations, and visibility, we revealed large differences in different regions across Tehran metropolis. The highest 484 485 concentrations of NHAZEs and PM_{2.5} were found at the west and southwest of the city, where industries have developed quickly over the last decades. Daytime traffic and population, car 486 traffic, a lack of green space, and the existence of passenger terminals are all factors that 487 488 contribute to high PM_{2.5} emissions in the region (International Communication and Relations Center of Tehran Municipality, 2019, https://ccia.tehran.ir/; last accessed 15 October, 2022; 489 Heydari et al., 2020; Ehsani and Bigdeli, 2021). Also, the north, east, and northeast regions had 490 the lowest concentrations of PM2.5. The northern and eastern areas of Tehran have cleaner air due 491 to more green spaces per capita and the use of new cars with fewer emissions (Alavi et al., 492 2020). However, the visibility varied according to the amount of PM_{2.5} concentrations and 493 NHAZEs in different parts of Tehran; that is, the regions with the highest PM_{2.5} emissions had 494 the lowest horizontal visibility. Since most industries have been established in the west of 495 496 Tehran, and the prevailing winds in Tehran are the westerlies, pollutants move towards the city center, resulting in the accumulation of primary and secondary pollutants in the west and central 497 parts of the city. 498

Exploring the role of meteorological parameters in the occurrence of NHAZEs in Tehran revealed that WS had a significant effect, with a regression coefficient of -0.30 (p<0.05). Over the entire 31 years, NHAZEs were observed when the T was high and the RH and WS were low. The intensity, duration, and prevalence of NHAZEs were high during the establishment of anticyclonic conditions, which caused the stability of the air. All meteorological variables played a role in the decreasing and increasing periods of NHAZEs, in which the WS had the greatest impact. As a result, in recent years, with the increase in wind speed, we observed a decline in 506 NHAZEs and an increase in visibility. This is consistent with the recent reversal of terrestrial 507 surface winds globally (Zeng et al. 2019) or in nearby countries like Saudi Arabia (Azorin-508 Molina et al. 2018), but there is a lack of research on changes of WS across Iran.

Simultaneous analysis of pollutant gases and PM_{2.5} concentrations (the primary cause of 509 haze) showed that the highest amount of PM2 5 was recorded when the amounts of RH, SO2, CO, 510 and NO2 were high but the WS and O3 were low. Conversely, the minimum value of PM2.5 was 511 512 at lower RH, SO2, and CO and a higher concentration of O3. Many previous studies have confirmed these events (Yang et al., 2007; Tai et al., 2010; Xu et al., 2011; Li et al., 2017). 513 Considering the high concentration of PM_{2.5} in recent years, it can be concluded that the reason 514 behind the reduction of NHAZEs is associated with the favorable meteorological conditions 515 (especially the increasing tendency of WS), which enhance the dispersion of pollutants. 516

In general, Tehran is a polluted city in Iran surrounded by the Alborz Mountain Range, which also affects the dispersion pattern of pollutants (Safavi and Alijani, 2006; Ashrafi, 2012). The government's primary concern should be to control pollution emissions effectively. Attempting to outlaw the use of fuels like mazut, kerosene, and diesel in all industries should be among the government's main focuses. In the future, the government and researchers must investigate human factors and large-scale atmospheric patterns affecting the haze and its characteristics and formation mechanisms in the Tehran metropolis.

- 525
- 526

528 5. Conclusions

529	The main	findings	of this	research	are as	follows:
		2)				

- A marked interannual variability in the number of haze synop (NHAZEs) is found in
 Tehran, with a significant decreasing trend for 1990-2020. On the contrary, visibility has
 improved as revealed by the significant increasing trend found in the last three decades.
- 533 2. Strong spatial differences in NHAZEs are observed across the region, with the highest
 534 NHAZEs detected in the western and central areas of the city, where industries have
 535 developed quickly over the years.
- The intensity, duration, and prevalence of NHAZEs were high under anticyclonic
 conditions. The meteorological variables can explain 30% of the interannual variability of
 NHAZEs and 65% of PM_{2.5} concentrations, with the WS exerting the highest impact.
- 4. Wind speed is increasing in Tehran for 1990-2020, which could partly explain the
 significant decline in NHAZEs and the significant increase in visibility.
- 541 5. The small regression coefficient of meteorological variables and the high correlation of 542 NHAZEs with PM_{2.5} concentrations indicate the relevance of investigating the influence 543 of human resources on haze events in Tehran; more pollution monitoring stations are 544 needed.
- 545

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