Environment International 156 (2021) 106732

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

The state of science on severe air pollution episodes: Quantitative and qualitative analysis

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ARTICLE INFO

Keywords:
Severe air pollution events
Pollution episodes
Urban air pollution
Pollution emissions
Formation of secondary pollutants
Mitigating air pollutants

ABSTRACT

Severe episodic air pollution blankets entire cities and regions and have a profound impact on humans and their activities. We compiled daily fine particle ($PM_{2.5}$) data from 100 cities in five continents, investigated the trends of number, frequency, and duration of pollution episodes, and compared these with the baseline trend in air pollution. We showed that the factors contributing to these events are complex; however, long-term measures to abate emissions from all anthropogenic sources at all times is also the most efficient way to reduce the occurrence of severe air pollution events. In the short term, accurate forecasting systems of such events based on the meteorological conditions favouring their occurrence, together with effective emergency mitigation of

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https://doi.org/10.1016/j.envint.2021.106732
Received 2 March 2021; Received in revised form 27 May 2021; Accepted 21 June 2021
Available online 28 June 2021

anthropogenic sources, may lessen their magnitude and/or duration. However, there is no clear way of preventing events caused by natural sources affected by climate change, such as wildfires and desert dust outbreaks.

1. Background

Questions regarding causes underlying severe air pollution events and our capacity to prevent them have arisen since these events began to occur on a large scale during the advanced stages of the industrial revolution. The answer depends not only on the knowledge of the respondent, but also on their interests or agendas. Winston Churchill, Prime Minister of Great Britain, asserted that the infamous smog event of December 1952 in London – one of the earliest and best-known events of this kind – was an act of God, thereby implying that it was not avoidable (Brimblecombe, 2012; Cohen et al., 2017). Yet it has been argued that it was related to the location and density of power plants burning low grade (high sulphur) coal within the city of London as decided by a previous government, and was therefore caused by humans (Brimblecombe, 2012).

Our understanding of severe air pollution events has significantly improved over the last decade, revealing an incredible complexity of causes and a myriad of factors governing their severity, for e.g. (Doherty et al., 2017; Mazzeo et al., 2018; Millán, 2014; Wu et al., 2018). In addition, the importance of preventing such events and not only mitigating baseline air pollution has been fully recognized: for example, reanalysis of data from the 1952 London smog event showed that it caused 12,000 additional deaths and immense economic costs due to disruption of the city's operations (Bell and Davis, 2001; Davis, 2002). Thereafter, both short- and long-term exposure guidelines were developed and promulgated (WHO, 2006), and countries around the world have since implemented prevention and mitigation policies and curbing measures.

Unfortunately, severe air pollution events are not a memory from the past, but still occur around the world. While it was expected that the mitigation measures implemented by the countries would gradually lower the burden of severe air pollution events, whether this is indeed the case is not known. How have the frequency, intensity and duration of these events evolved? To answer these questions, the objectives of our work were: (a) to evaluate the trends in severe air pollution in a representative number of cities around the world; and (b) to compare the trends in severe air pollution with the trends in baseline air pollution of the studied cities. Here we attempted only a general qualitative overview of the likely main drivers, while detailed quantitative analysis of the drivers of change must be done at the local scale while considering all the relevant local parameters and factors that contribute to the change.

The World Health Organization (WHO) concluded that $PM_{2.5}$ (particles with aerodynamic diameter <2.5 μm), is the air pollutant with the most severe health impacts (WHO, 2013a, 2013b), and therefore we selected $PM_{2.5}$ as the pollutant for our evaluation. To address our objectives, we first need to define what constitutes a severe $PM_{2.5}$ pollution event, and then summarize the scientific understanding of the various mechanisms of the formation of these events.

Note that ambient particulate matter can be characterised by many other metrics in addition to mass concentration ($PM_{2.5}$), including particle number concentration / size distribution, surface area, composition (content of metals, elemental and organic carbon content, other elements or compounds); however, none of these are routinely monitored, and hence are not available for global comparisons.

2. What is a severe air pollution event?

One basic manifestation of severe air pollution is low visibility. Reduction in visibility is caused by an increased concentration of airborne particles; therefore, the severity of the event is often linked to the concentration of particulate matter (PM) in the air, expressed as $PM_{2.5}$ However, air pollution events can occur without any immediate change to visibility. High ozone (O_3) concentrations occur in spring and summer, when the presence of its precursors, nitrogen oxides ($NO_x = NO_2 + NO$) and volatile organic compounds (VOCs), and favourable meteorological conditions, including high insolation, do not initially cause reduced visibility. The second stage of the process, when particles form in the air (Zhang et al., 2015), is when visibility decreases.

There are many definitions of severe air pollution events, most commonly provided by the authorities responsible for controlling them. For example, the definition of 'severe' could be associated with the EU Directive 2008/50/EC, which requires member states to inform or alert their population if alert thresholds from Annex XIV are exceeded or an exceedance is forecast. Specific actions are required when alert thresholds are exceeded to inform the public about health risks and recommended personal behavior to minimize exposure (UNION, 2008). However, these alert thresholds have only been set for nitrogen dioxide (NO_2), O_3 and sulphur dioxide (SO_2). Furthermore, Annex II reports the limit values for NO_2, PM_{10} and $PM_{2.5}$, with only NO_2 and PM_{10} having a short-term limit value. Implementation of measures is also suggested to avoid exceedances of the NO_2 and PM_{10} limit values. By contrast, in China, severe air pollution events are related to $PM_{2.5}$, and in other countries, they are related to air quality indices including $PM_{2.5}$, O_3 and other pollutants; sometimes one pollutant drives the index, but not always. Several examples of severe air pollution events in different countries are provided in Supplement 1. The substantial differences in severe air pollution definitions and alert limit values between countries are due to differences in the economic and political situations of the countries, which lead to differences in countries' approaches to the mitigation of air pollution.

It is important to note that the notion of 'severity' is relative rather than absolute. There are cities where baseline air quality is relatively good (baseline is defined here as the trend component of concentration with the influence of seasonal variation and pollution events removed; see the Methods section for details), and therefore a 'severe' increase over the baseline could be of much smaller absolute magnitude than the baseline in polluted cities. There is evidence, however, that despite overall local air quality being relatively good, such pollution events still have a measurable acute impact on health (Di et al., 2017; Milojevic et al., 2014; O'Connor et al., 2008).

A severe air pollution event is sometimes termed 'haze'. Haze is a meteorological term (referring to visibility-reducing liquid aerosols), so to avoid confusion, it is not used in this paper. As a point of interest, the word for haze in some Latin languages is *bruma,* which is a poetic word for mist and fog.

3. Mechanisms behind severe air pollution events

Severe episodic air pollution events are caused by local, regional, or transboundary (anthropogenic or natural) emissions of air pollutants, but specific meteorological conditions often favor the development of such events and influence their severity. In particular, forest fires, desert dust storms, crop burning, high insolation, or intense anticyclonic conditions might favour the transport, formation, or accumulation of pollution that might yield high levels of pollutants. Briefly, the causes and contributors include: (1) occurrence of meteorological conditions leading to efficient formation and rapid growth of secondary pollutants; (2) a rapid increase in the intensity of source operation intensity of source operation (e.g., increase in residential space heating using coal or wood during cold spells) or accidental emissions; and (3) occurrence of meteorological conditions favourable for stagnant air, trapping and accumulating pollution. Synergetic effects across these three mechanisms can further amplify the severity of events. The causes of and contributors to the specific formation mechanisms of severe PM air pollution events, as described above, are schematically presented in Fig. 1, and are further discussed below, in the context of the results of the data analysis. We highlight this as being consistent with a study by Amato et al. (2016), who reported that most of the PM mass concentration is driven by the secondary formation of particles (vapor condensation).

4. Methods

4.1. Selection of cities

We selected a non-probability purposive sample of 100 cities based on the availability of $PM_{2.5}$ monitoring station data, which optimised global geographic coverage. We expected that there would be greater differences in the trends of $PM_{2.5}$ concentrations for locations in different regions (e.g., with different sources, economic development or government policies) versus between cities in the same regions. We

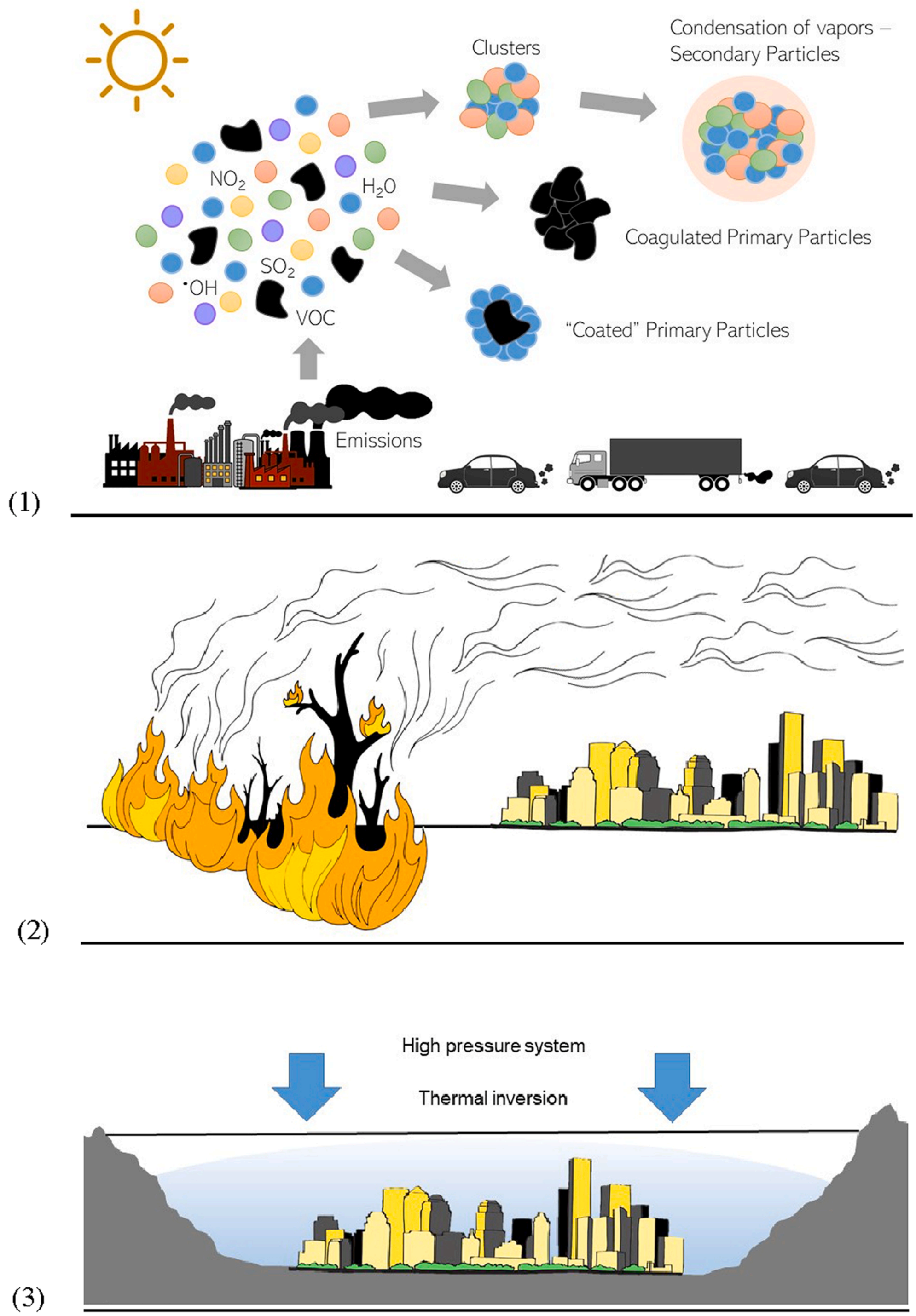

Fig. 1. Formation mechanisms of severe air pollution events: (1) occurrence of meteorological conditions leading to efficient formation and rapid growth (vapor condensation) of secondary pollutants; (2) a rapid increase in the intensity of source operation or accidental emissions (fire as an example); and (3) occurrence of meteorological conditions favourable for stagnant air, trapping and accumulating pollution.

focused on the period of 2013 to 2017, because this is when the standards were introduced in China, leading to substantial decreases in baseline $PM_{2.5}$ concentrations in the country (Wang et al., 2017a).

The specific criteria that we used for inclusion of a city were: (1) a population of at least 20,000 by the year of 2017 according to United Nations statistics (https://datahub.io/core/population-city#data); (2) at least one $PM_{2.5}$ monitoring station in the downtown area of the city; and (3) $PM_{2.5}$ daily average monitoring data in the city with a coverage of at least 80% from 2013 to 2017. In cases with more than one monitoring station in a city, we chose the station that is most representative of the city's general urban air quality. For cities whose monitoring data are hourly averages, we calculated the daily average as long as at least 20 h of data were valid on that day. We also checked to ensure that the missing data of each city's dataset did not concentrate on any specific season. This study did not take into account the differences among the methods nor the instruments used for measuring $PM_{2.5}$ in different cities. An assumption was made that the organizations operating the instruments (most of them the government) followed appropriate calibration procedures. Regardless, because the emphasis of the study was on temporal trends within each city, any inter-city differences in absolute values due to the instruments would not influence the findings.

4.2. Data preparation

Depending on the sources (discussed below), the format of the $PM_{2.5}$ data differed. In order to make a common processing program work for every city's data, we first read in the data and then wrote the data to a prescribed format. This process was repeated for every city, and no data filtering was applied during this process. All data processing was done via RStudio (Team, 2017).

4.3. Analysis of the trends of the events: overall, frequency, magnitude and duration

Initially we intended to use the local definitions of severity used in the cities (countries) included in the analysis and not to restrict it to a pollutant. Because the differences between the definitions of severity in different countries proved to be impossible to reconcile for the purposes of this work, we decided to base the analysis on the actual concentration values of $PM_{2.5}$.

We considered three indices to describe the $PM_{2.5}$ pollution events occurring in each city, including: (1) the annual frequency of pollution events (denoted as I_f, unit: events per year); (2) the annual average pollution event magnitude (denoted as I_m, unit: percentage over the criterion value, which is derived by first calculating the percentage by which the event's concentration surpasses the criterion, then calculating the mean percentage of all the pollution events occurring within 1 year); and (3) the annual average pollution duration (the mean of the duration of all the pollution events occurring within 1 year, denoted as I_d, unit: days per event). Obviously, the value of all the three indices will vary, depending on the method used for the identification of severe air pollution events. The method that we used was based on the theory of STL (seasonal-trend decomposition procedure based on Loess). STL is a useful methodology for "decomposing a time series into trend, seasonal and remainder components" (Cleveland et al., 1990), and it allows the trend and seasonal component to be removed from the time series $PM_{2.5}$ data. Normalization and linear regression have been performed on individual indices for further analysis. Other studies used 95th percentile to identify pollution events (Johnston et al., 2011), but we consider that our method is more appropriate because this method implies that 5% of the days are grouped as pollution events, which cannot be true for all the cities. A detailed reasoning for our choice of analysis methods is given in Supplement 3.

As the next step, we calculated the standard deviation (SD) of the remainder component and used 3*SD as the criterion for pollution events (Hirata et al., 2005). This means that if a value in the remainder component is higher than 3*SD, then its corresponding date is classified as one day with a pollution event. This approach helped us to focus on the variation rather than the absolute magnitude of the concentrations. Because different cities had different remainder components, we were able to make a direct comparison between severely polluted and moderately polluted cities, which would not be practical if we chose a universal pollution event value for all the cities.

To comprehensively consider all the three indices, we used principal component analysis (PCA) as a method of rating, which is useful for identifying the directions in which data have the most variance (Vasilyeva et al., 2018). First, we applied PCA to recognize the direction that is constituted by the linear combination of I_m and I_d and where most of the variance in I_m and I_d concentrate. Then we calculated the geometric mean of the recognized direction and the I_f. I_f is not included in the PCA because it shows only a weak correlation with the other two indices. The results can be interpreted as the rating of pollution events for each city in each year, and the value of the rating (denoted as R_{PE}, no unit) is related to the severity of pollution events: a higher R_{PE} means higher severity. Normalization is performed on R_{PE} through equation (1) to make its meaning clearer.

$$R_{PE,norm} = \frac{R_{PE} - R_{PE,min}}{R_{PE,max} - R_{PE,min}} * 100 \tag{1}$$

In this way, $R_{PE,norm}$ has a value from 0 to 100, where 0 represents the lowest pollution event level among all the cities during 2013–2017, and 100 represents the highest. Through the slope of the linear regression of $R_{PE,norm}$ (denoted as $S_{PE,norm}$), we can judge how a city's rating, or the severity of air pollution events has changed within the period investigated. If $S_{PE,norm}$ is positive, then the severity of a city's pollution events has become more severe over the 5 years, and vice versa.

4.4. Separation of baseline from severe air pollution events

Because the trend component (long-term trend) identified from the STL decomposition process represents the concentration level with the influence of seasonal variation and pollution events removed, it is regarded as the baseline $PM_{2.5}$ concentration of a city.

It is important to compare between the trend of $R_{PE,norm}$ and the long-term air pollution trend for the same city. This is because their relationship can reflect the effectiveness of mitigation measures: whether measures to reduce baseline $PM_{2.5}$ are also effective in reducing the frequency and/or severity of pollution events. The trend of $R_{PE,norm}$ was calculated through linear regression over its 5-year values, and the long-term trend was calculated through linear regression of the baseline concentration.

4.5. Comparison of trends in the events versus baseline trends

We compared the trend of $R_{PE,norm}$ and the baseline trend for every city during 2013–2017 and classified the cities into four groups based on the results:

Group 1. The trend of $R_{PE,norm}$ is positive while the baseline term trend is negative
Group 2. The trend of $R_{PE,norm}$ is negative while the baseline term trend is positive
Group 3. The trend of $R_{PE,norm}$ and the baseline trend are both positive
Group 4. The trend of $R_{PE,norm}$ and the baseline trend are both negative.

This classification is a consequence of the analysis method we adopted here, taken as the basis for comparison of the actual concentrations of $PM_{2.5}$ and then separating the events from the baseline pollution. The division would likely be different if a fixed value had been used to judge severe air pollution events, which is the approach most countries take in practice. However, because the baseline is different in every country, using a fixed value would have masked the trends actually occurring.

5. Results

(i) *Results: Quantitative analysis*

We collected $PM_{2.5}$ data from a total of 100 cities in 29 countries on five continents (listed in Table S2, together with the data sources). The data met our criteria and no seasonality was observed in the dates with missing data for individual cities. While we endeavoured to optimize the global coverage, there are many blank areas around the globe, for which no data that would meet our criteria were available; this was particularly the case for Africa, with data available only for South Africa, as it does not appear that there is any regulatory monitoring conducted in other African countries. Other countries, for example Russia, conduct monitoring, but the data are not made public, and are not available on request.

The severity of the $PM_{2.5}$ pollution events was considered according to three indices: (1) the annual frequency of pollution events (I_f); (2) the annual average pollution event magnitude (I_m); and (3) the annual average pollution event duration (I_d). The yearly average values of these three indices are listed in Table S3.1, as well as the slopes of their normalized values. Additionally, we introduced an overall rating of the severity of pollution events for each city in each year, considering all three indices ($R_{PE,norm}$). Significantly, we used the magnitude of standard deviation as a key parameter in the calculation of $R_{PE,norm}$. Thus, this metric describes the severity of events in a relative sense, rather than in an absolute sense for any given city. A full description of the indices and how they were computed is presented in the Methods section. In general, neither the severity, nor its elements, including frequency, magnitude or duration, are experienced equally by all the regions, as can be seen from Figs. 2–5. Qualitative analysis of the possible reasons for this and the suggested drivers is presented below.

Overall, the highest mean baseline $PM_{2.5}$ concentration over the 5 years was observed in Shijiazhuang, China (112 μg/m^3), while the lowest was recorded in Halifax, Canada (5.3 μg/m^3). The highest mean annual severe pollution frequency over the 5 years was observed in Cagliari, Italy (4.6 per year), while the lowest was recorded in Nicosia, Cyprus (0.4 per year). Comparing larger regions, the mean frequency (I_f) of cities in China was 3.0 per year, with a baseline of 56.8 μg/m^3, while for North American cities it was 2.5 per year and 8.3 μg/m^3, respectively and for European cities, 2.8 per year and 15.0 μg/m^3.

5.1. Trend in severe air pollution events

Fig. 2 shows the annual trend in the overall severity of $PM_{2.5}$ pollution events for the 5-year period investigated. Although a 5% significance level is more commonly used, we opted to use 10% because the slopes were calculated for 5-yearly values and only a few cities satisfy the 5% criterion. A similar approach was taken in other environmental science studies (e.g. (Do et al., 2017)). The trend was negative (e.g. 10% significance level that its value was less than zero, similarly hereinafter) for Haerbin, Haikou, Kitchener, Montreal, Nanjing, Macau, Hangzhou, Philadelphia, Helsinki, and Wuhan (listed in sequence of increasing significance, similarly hereinafter), which means that the overall severity of $PM_{2.5}$ pollution events in these nine cities, relative to their respective baselines, experienced statistically significant decreases over the 5-year period at the 10% significance level. Sasolburg,

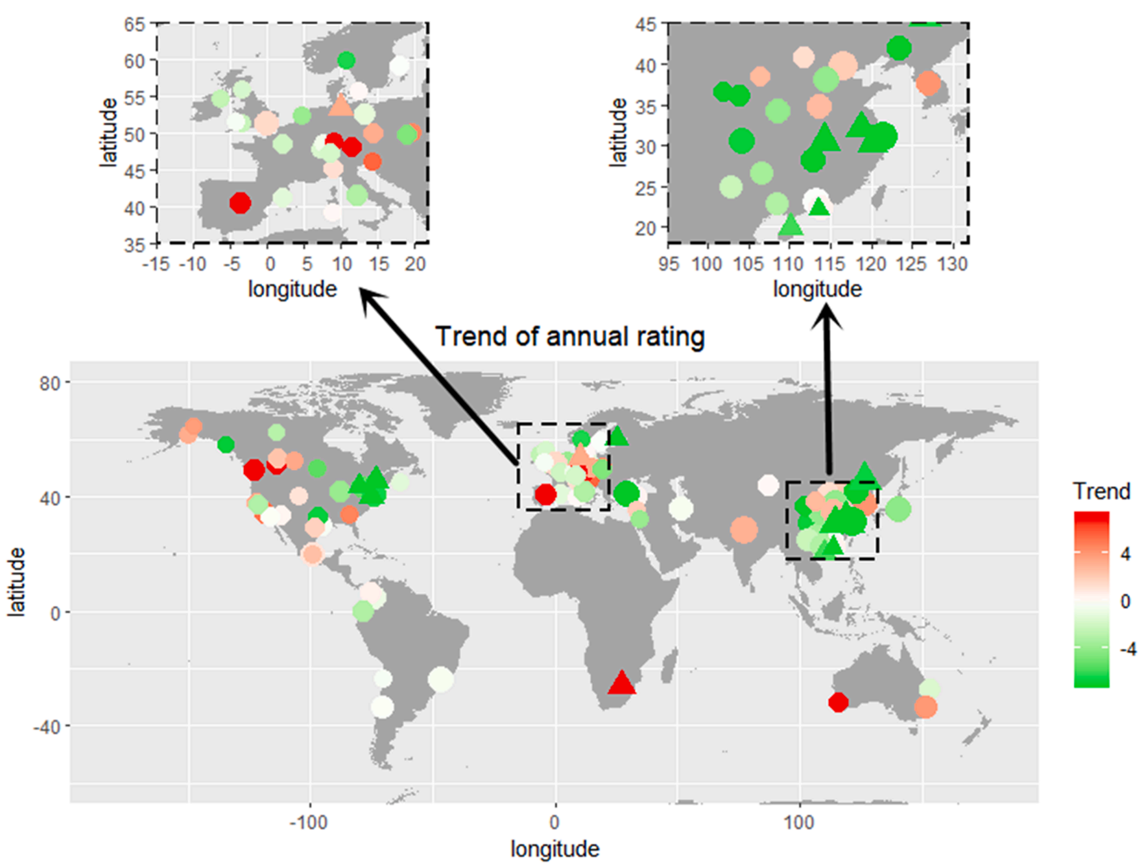

Fig. 2. Linear regression slopes ($S_{PE,norm}$; unit: per year; a higher value means worsening severity) of the normalized rating of the severity of pollution events ($R_{PE,norm}$ range: 0–100; no unit) for all the cities. Specific values of $S_{PE,norm}$ can be found in Table S3.1. Triangles indicate cities in which the corresponding slopes are different from zero at a 10% significance level.

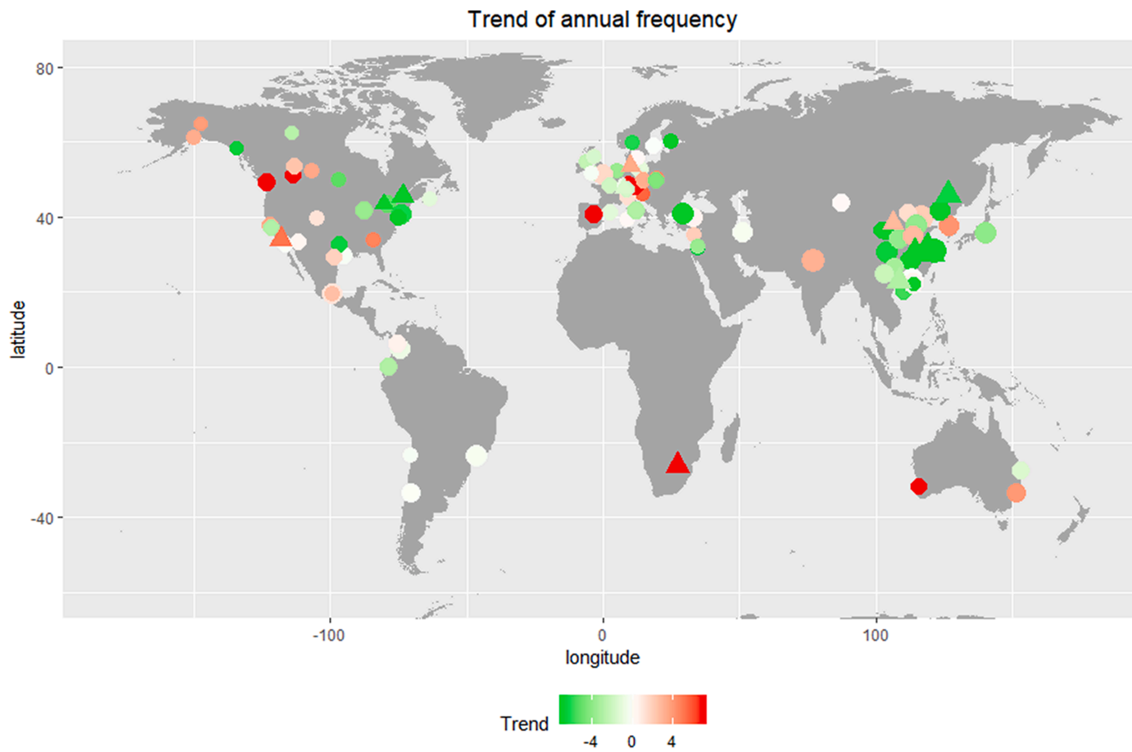

Fig. 3. Linear regression slopes for the normalized annual frequency of severe air pollution events ($I_{f,norm}$; range: 0–100; no unit) for all the cities (e.g. $S_{f,norm}$; unit: per year; a higher value means the annual frequency of pollution events is increasing faster). Specific values of $S_{f,norm}$ can be found in Table S3.1. Triangles indicate cities in which the corresponding slopes are different from zero at a 10% significance level.

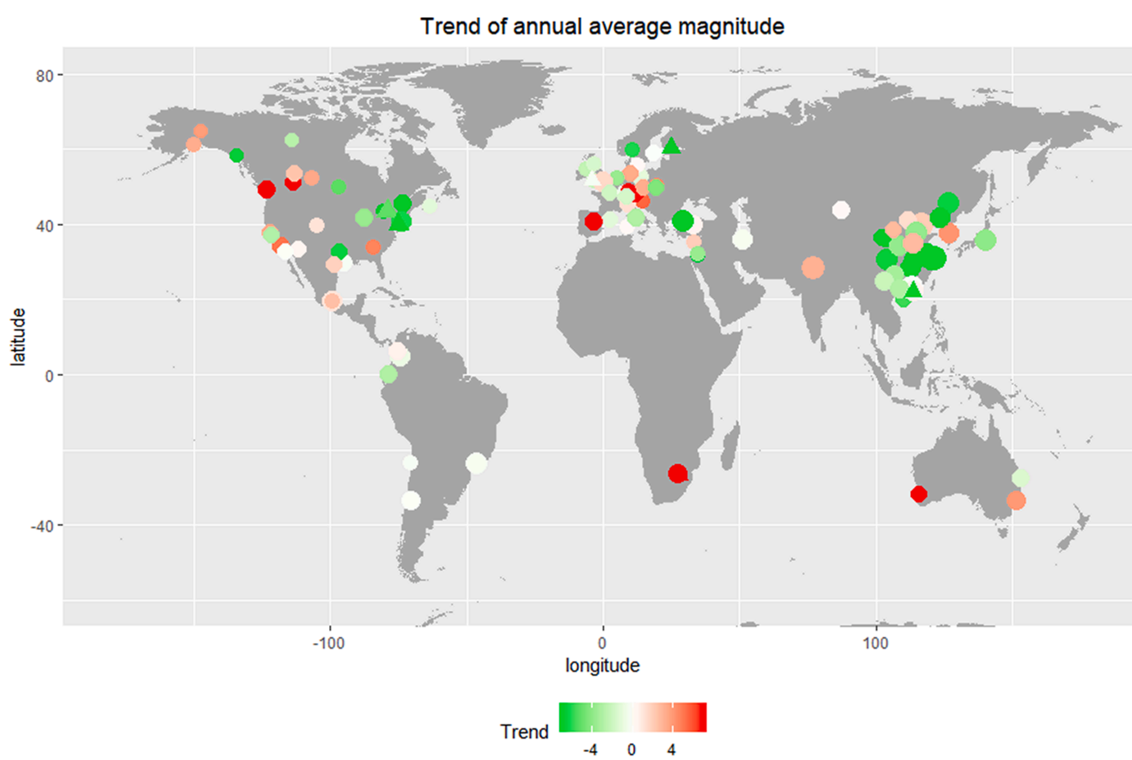

Fig. 4. Linear regression slopes ($S_{m,norm}$; unit: per year; a higher value means the annual average magnitude of pollution events is increasing faster) of the normalized annual magnitude of severe air pollution events ($I_{m,norm}$; range: 0–100, no unit) for all the cities. Specific values of $S_{m,norm}$ can be found in Table S3.1. Triangles indicate cities in which the corresponding slopes are different from zero at a 10% significance level.

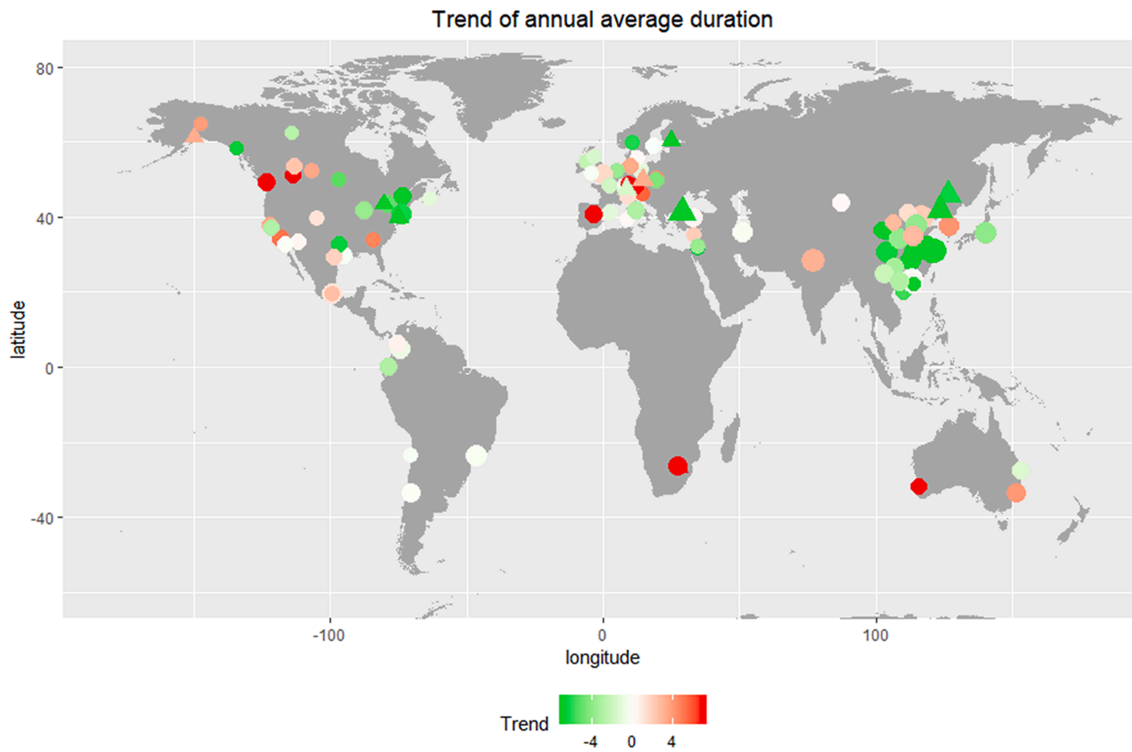

Fig. 5. Linear regression slopes ($S_{d,norm}$; unit: per year; a higher value means the annual average duration of pollution events is increasing faster) for the normalized annual duration of severe air pollution events ($I_{d,norm}$; range: 0–100; no unit) for all the cities. Specific values of $S_{d,norm}$ can be found in Table S4. Triangles indicate cities in which the corresponding slopes are different from zero at a 10% significance level.

Johannesburg, and Hamburg are the cities that experienced a statistically significant increase in $PM_{2.5}$ pollution events over this period at the 10% significance level. The aforementioned cities are shown as triangles in Fig. 2 while the other cities are shown as circles.

Figs. 3–5 show the results of linear regression slopes for the normalized annual frequency, normalized annual average pollution

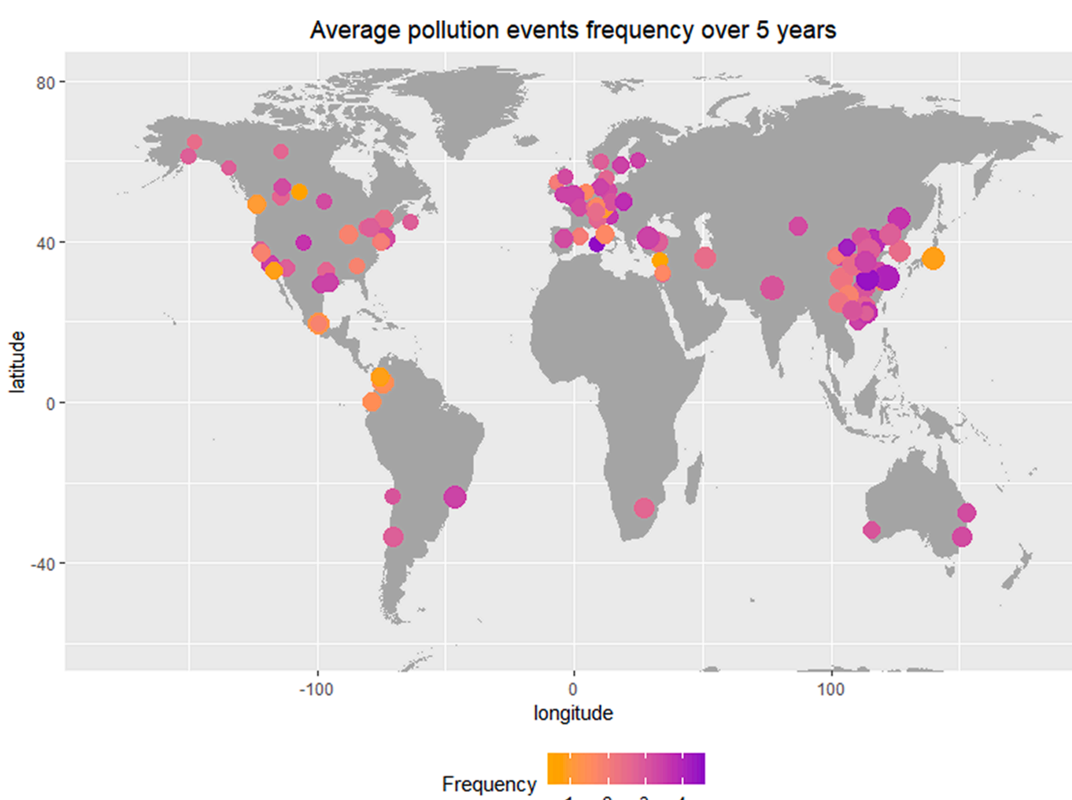

Fig. 6. Average frequency of $PM_{2.5}$ pollution events (counts per year) over the 5-year period for all the cities.

magnitude, and the normalized annual average pollution event duration ($I_{f,norm}$, $I_{m,norm}$ and $I_{d,norm}$) of all the cities, while Fig. 6 shows the average frequency of $PM_{2.5}$ pollution events over the 5-year period for all the cities. Comparing the average frequency of $PM_{2.5}$ events over the 5-year period (Fig. 6), and the trend in annual frequency (Fig. 3), an observation can be made that while the former presents an overall overview for the period under analysis for each of the cities, the latter provides better insight into the changes occurring. For example, while in the eastern part of China the overall frequency is high for many of the cities (Fig. 6), the annual frequency is decreasing for most of these cities (Fig. 3). However, the opposite is true for the Australian cities, which are characterized by an overall high frequency (Fig. 6), with the annual frequency increasing (Fig. 3). Another observation that can be made from Figs. 2–5 is that the frequency, magnitude, and duration tend to accompany each other.

When a significance level of 10% is selected, Hangzhou, Kitchener, Haerbin, Nanning, Nanjing, Mulhouse, Montreal, and Wuhan experienced significant decreases in event frequency ($I_{f,norm}$) from 2013 to 2017, while Sasolburg, Johannesburg, Yinchuan, Los Angeles, and Hamburg experienced significant increases. Philadelphia, Helsinki, Macau, Swansea, and Toronto experienced significant decreases in the relative magnitude of events ($I_{m,norm}$), while Sasolburg experienced significant increases. Haerbin, Philadelphia, Istanbul, Helsinki, Shenyang, and Kitchener experienced significant decreases in event duration ($I_{d,norm}$), while Zurich, Prague, Sasolburg, and Anchorage experienced significant increases. It can be concluded that in different cities, different aspects of the severity are more significant (frequency, magnitude, or duration), and these aspects would have a higher impact on the overall rating of the severity of pollution events for the cities.

Considering all the cities, I_m and I_d are in positive correlation with each other (Pearson's r is about 0.37, Kendall's tau is about 0.36), while I_f shows only a weak correlation with the other two indices (Pearson's r is about −0.028 and −0.077). Similar to the approach adopted by Shaker (Shaker, 2018), we applied principal component analysis (PCA) to identify the direction constituted by the linear combination of I_m and I_d and where most of the variance in I_m and I_d concentrate. The identified direction explains 68% of the total variance.

5.2. Relationships between the trends in baseline air pollution and the trends in severe air pollution events

The trends in baseline $PM_{2.5}$ pollution for all the cities are listed in Table S3.1. Based on a comparison of the trend of the normalized rating of the severity of pollution ($R_{PE,norm}$), and the long-term trend in baseline air pollution for every city during 2013–2017, we found that 30 cities belonged to Group 1 (positive $R_{PE,norm}$ trend with air pollution decreasing but the event rate on the rise); 6 cities belonged to Group 2 (negative $R_{PE,norm}$ with air pollution increasing, but the event rate decreasing); 10 cities belonged to Group 3 (positive $R_{PE,}$ with both air pollution and severe air event rate increasing); and 54 cities belonged to Group 4 (negative $R_{PE,norm,}$ with both air pollution and event rates decreasing). The cities in each group are listed in Table S3.2.

(ii) *Results: Qualitative analysis*

We conducted qualitative analysis by considering the outcomes of the quantitative analysis, and then attempting to contextualize and explain (qualitatively) the trends in the selected cities included in the quantitative analysis. This involved gaining an understanding of the mitigation measures taken in these cities (based on the insights provided by the co-authors), and/or an understanding of the specific meteorological events of relevance. This was done with a view to answering the four specific questions set out below.

5.3. What were the measures being taken to mitigate baseline air pollution in cities?

It was not possible within the scope of this work to quantitatively analyse each individual city to determine the reasons behind the trends observed. Measures taken to successfully reduce baseline air pollution in the cities from Group 4 (where the severity of pollution events also decreased) varied considerably between cities. In particular, a comprehensive set of measures was implemented in China (Zhang et al., 2019) and, as a result, most (but not all) of the Chinese cities are in Group 4. In the Canadian city of Toronto for example, the phase-out of coal-based electricity generation in 2005–2013 led to an overall improvement of air quality that may also have contributed to weaker events in 2013–2017 (Jeong et al., 2013). In New York State, on the other hand, changes in air quality occurred from 2008 to 2013 as a result of implementation of regulations on fuel quality, and vehicular and power plant emissions. Such changes continued to occur after 2013 because of several other reasons including the economic instability of the 2008–2013 period with the recession of 2008–2009, the drastic drop in the price of natural gas relative to coal, the increased contributions of gas direct injection engines to the car fleet, and the reformulation of gasoline (Zhang et al., 2018a). In Stockholm, a congestion charge was implemented in 2007 as a tax levied on most vehicles entering and exiting central Stockholm for the purpose of reducing traffic congestion and improving the air quality. In 2016, congestion taxes were increased in the inner-city parts of Stockholm. Since 2010, driving with studded tyres is no longer allowed in certain areas of Stockholm. In addition, various initiatives were undertaken to bind and remove settled dust on the roads. However, in countries from Groups 1 and 3, while measures were taken to mitigate air pollution, they were not successful.

5.4. What were the reasons for severe air pollution events in cities?

Although the basic mechanisms involved in the formation of severe air pollution events are well understood (as schematically presented in Fig. 1), on a local or regional scale the hypotheses about what triggers the events are not always fully proven or supported. However, in many places there is a good understanding of the key drivers behind these events, and examples of these (as related to the mechanisms listed above), are set out below.

2. *Occurrence of meteorological conditions leading to efficient formation and rapid growth (vapor condensation) of secondary pollutants.* Secondary pollutants, such as secondary organic aerosols (SOAs) and O_3, which are formed in the air by some of the source-emitted precursors undergoing a series of physico-chemical processes, are present during severe air pollution events. Even during the COVID-19 lockdown, in some areas $PM_{2.5}$ pollution episodes took place because the atmospheric conditions were favourable for the formation of secondary PM (vapor condensation) from gaseous precursors (Huang et al., 2021). As reported above, most of the $PM_{2.5}$ mass in the urban background is attributable to secondary PM formation. High insolation and high temperatures are favourable to the formation of O_3 as it is a product of photochemical reactions that are further enhanced under the vertical recirculation of air masses or stagnant conditions (Brines et al., 2015; Millán et al., 2000; Saiz-Lopez et al., 2017). The occurrence of secondary PM includes nucleation and subsequent growth processes (Ehn et al., 2014) that are more efficient in conditions of high insolation (Kulmala et al., 2014), low temperature and low relative humidity (Cheng et al., 2019; Hamed et al., 2011). Stronger solar radiation produces more free radicals to efficiently oxidize precursors, accelerating O_3 formation and the generation of large amounts of PM (contributing mainly to particle number, not mass concentration). O_3 generally has a higher production rate at a higher temperature, while secondary PM is formed more easily at a lower temperature that promotes higher supersaturation and

therefore a higher nucleation rate. Relative humidity is a critical factor in the development of secondary PM. In addition to hygroscopic growth, a rapid increase of sulphates and secondary organic aerosols has been observed at high relative humidity owing to aqueous-phase chemistry in severe air pollution episodes in China (Brines et al., 2015; Hama et al., 2020; Hamed et al., 2011; Huang et al., 2021). *A rapid increase in the intensity of source operation and accidental emissions*, resulting in a significant increase of source-emitted pollution in the vicinity of the source, and at distance due to its transport, as a one-off, seasonal, or cyclic event. Examples include PM emissions from (i) a dust source (desert) (Al-Dabbous and Kumar, 2014), the co-transport of pollutants with PM dust, the bioaerosol load, the interaction (e.g. condensation or adsorption) of desert dust with local gaseous pollutants; and the local accumulation of pollutants due to the decrease of the boundary layer height during intense dust episodes (Dominguez-Rodriguez et al., 2020; Querol et al., 2019; Sakhamuri and Cummings, 2019); (ii) wildfires (one-off events) (e.g. a megafire in Chile in 2017 (de la Barrera et al., 2018); (iii) agricultural waste burning – practiced for example during winter upwind of Delhi (Hama et al., 2020) and causing severe episodes of pollution in the city (Kumar et al., 2015), and occurring in China in June, September and October, and in Brazil all year round; (iv) massive application of natural fertilizers with high emissions of ammonia (NH_3); and (vi) biogenic emissions occurring during particular times of the day or season (Guenther et al., 1996; Li et al., 2017); (vii) substantial increase in residential coal (or wood) combustion for heating during cold seasons, such as during the historic 1952 London fog (Bell and Davis, 2001; Davis, 2002).

3. *Occurrence of meteorological conditions favourable for stagnant air, trapping and accumulating pollution.* Conditions that favour stability generally include a shallow planetary boundary layer, weak surface winds, and descending air flows. Unfavourable meteorological conditions have been linked to orographic forcing in many places, such as in the region of the north China plain where a unique basin terrain effect and a 'harbors' effect of the leeside slope of the Tibetan Plateau often results in stagnation development and pollution accumulation leading to large-scale episodic pollution (Long et al., 2016; XU et al., 2015). Furthermore, climate change (e.g. global warming, drought, El Niño–Southern Oscillation, Atlantic meridional overturning circulation) may affect meteorological conditions by influencing large-scale circulations (e.g. East Asian winter monsoons, prevailing north-westerly winds) leading to deterioration in the city's ventilation conditions (Cai et al., 2017; Westervelt et al., 2016; Wu et al., 2018; Zhang et al., 2018b).
4. *Synergetic effects of emissions and atmospheric processes* (An et al., 2019). Adverse meteorological conditions can limit the dispersion of primary pollutants and precursors, leading to a rapid increase in secondary pollutants. The increase in oxidants (e.g. O_3, NO_3 and OH) elevates the atmospheric oxidative capacity and accelerates the formation of other secondary pollutants. High concentrations of PM, on the other hand, enhance the air stability by aerosol–radiation interaction, which cools the surface and warms the air, leading to temperature inversion, decreased planetary boundary layer height and accumulated water vapor (Peng et al., 2016; Stocker et al., 2013). The interaction of the planetary boundary layer, moisture and PM forms a positive feedback cycle to trap PM near the surface (Zhang et al., 2018b). Furthermore, atmospheric transport of pollutants can play a contributing role. For example, even though forests could be far away from urban centres, the city's anthropogenic VOCs that reach the forest would result in more secondary organic aerosol than that formed from biogenic sources (Shrivastava et al., 2019). Therefore, mixing of urban air with emissions from biogenic sources can exacerbate the problem and cause a continental event. A few examples of synergist events follow.

In Spain, five atmospheric basins (those of Madrid, north Barcelona, Tarragona, Valencia-Castelló, Puertollano and Guadalquivir River) recorded very intensive summer O_3 episodes (Querol et al., 2016). These pollution episodes were caused by high emissions of anthropogenic and biogenic pollutants (NOx and biogenic and anthropogenic VOCs), coupled with the vertical recirculation of air masses produced by the interaction of sea and mountain breeze circulations with a complex orography (Millán, 2014; Querol et al., 2017). On the other hand, the high vehicle density in Madrid, coupled with a dense urban structure, high dieselization of the fleets and the well-known 'dieselgate', as well as the development of intensive anticyclonic scenarios in a continental basin characterized by a high mountain chain in its northern side, very often yields very high NO autumn-winter episodes with extremely high NO_2 episodes (Borge et al., 2018). A more detailed discussion of the causes and abatement measures taken in Spanish cities to prevent severe air pollution events is presented in Supplement 4. In London, in contrast, while there is some indication that the frequency of severe $PM_{2.5}$ episodes is falling, and where such episodes typically occur in March, April, September and December, a mix of meteorological conditions is suggested as a cause (Beddows et al., 2015). It has also been demonstrated that all severe episodes involved a large regional component (broadly represented by Outer London concentrations), with an additional lesser increment added by local sources (Inner London), under similar meteorological conditions – a continental air mass that gathered particles and gases as it passed over urban and industrial regions of Continental Europe transporting polluted air over the UK via light easterly winds. In general, these episodes are dominated by secondary particles, and those in March and April are particularly high in ammonium nitrate and coincide with periods of heavy agricultural crop and land spraying in northern Europe (Graham et al., 2020). By contrast, in Colombia (Guevara Luna et al., 2018; Hernandez et al., 2019; Mendez-Espinosa et al., 2019), $PM_{2.5}$ pollution episodes occur in the dry period, from January to March, and are regional in nature, and are not restricted to individual cites (Guevara Luna et al., 2018; Hernandez et al., 2019; Mendez-Espinosa et al., 2019). The cause of the events was the reduction of mixing height during the dry period coupled with a substantial increase in wildfires in Northern South America.

5.5. What leads to the reduction in severe air pollution events?

Two main approaches are considered to be the key to reducing/eliminating severe air pollution events:

1. *Long-term measures towards reducing baseline air pollution.* In the cities belonging to Group 4, the decrease in baseline $PM_{2.5}$ pollution was accompanied by a decrease in severe pollution events, strongly implying that measures aimed at improving the former are also efficient in mitigating the latter. While we cannot conclusively prove this for each individual city, the fundamental principle that a reduction in emissions at any point of time is the most obvious precaution against severe air pollution events is the most likely reason why Group 4 is the largest group (less baseline pollution, less severe pollution).
2. *Short-term measures towards lessening the severity of the events*. The key premise here is the prediction of an event based on meteorological conditions conducive to transporting emissions to a particular location/region, trapping pollution, and/or conditions under which secondary pollutants are formed (usually in conjunction with trapping and/or transport). For example, while Santiago (Group 2) has experienced a clear decline in $PM_{2.5}$ concentrations over the last 20 or 30 years (Barraza et al., 2017; Gallardo et al., 2018), baseline air pollution is on the rise, explained in part by an increase in the vehicular fleet and a decline in the use of public transportation (Gallardo et al., 2018). However, the number of episodes decreased in 2017, which has been attributed to the introduction of a more efficient forecasting system (Saide et al., 2011, 2016) that started in full in 2016. The effectiveness of mitigation measures to curb

extreme pollution in Santiago has been addressed in the scientific literature, and in particular the application of a state-of-the-art model to compare the efficacy of vehicular restrictions and banning of residential combustion (Mazzeo et al., 2018). Other authors have compared days with similar atmospheric conditions for which contingency measures were applied or not (Mullins and Bharadwaj, 2015; Troncoso et al., 2012). Saide et al. (2011) (Saide et al., 2011, 2016) used atmospheric modeling to show that episodes are the result of multiple days of accumulated emissions and that same-day emissions often contribute only a small fraction of the observed pollution during an episode.

5.6. What led to the increase in severe air pollution events?

1. *Insufficient reduction in baseline pollution at the urban scale*, which can be claimed to be the case for many of the cities in Group 1.
2. *Insufficient reduction in baseline pollution at the regional scale.* A number of $PM_{2.5}$ and/or O_3 pollution episodes in urban areas have been attributed to the direct or indirect impact of regional emissions. In particular, agricultural ammonia is responsible for increased $PM_{2.5}$ levels in urban environments (Giannakis et al., 2019) and for a relevant attributable premature mortality (Lelieveld et al., 2015). However, most of the O_3 pollution episodes affecting urban areas are highly influenced by regional and long-range transport of pollution.
3. *Climate change has affected the increase in episodic events of pollution emissions*. Globally, climate change resulting from anthropogenic emissions has an impact on meteorological conditions on a local scale and, in turn, on local pollution events (Hou and Wu, 2016; Westervelt et al., 2016; Zhang et al., 2017). For example, the mechanisms through which drought or heatwaves resulting from climate change may influence extreme $PM_{2.5}$ events include dust storms and bushfire smoke (Doherty et al., 2017; Jones and Fleck, 2020). Generally, $PM_{2.5}$ mass is more likely contributed to by combustion than by mechanical processes such as windblown dust, so it is possible that fires during the early drought periods may be the main driver of $PM_{2.5}$ increases; whereas in later drought periods there is less vegetation to burn, and dust storms are more likely (but possibly contributing less $PM_{2.5}$). For example, an increase in the severity of events in Sydney was linked to increased dryness across the State of New South Wales, leading to an increase in fire intensity (Johnston et al., 2011), with a similar situation occurring in the Canadian west coast cities of Edmonton and Vancouver (Canada, 2020; Wang et al., 2017b).

6. Conclusion

Health, environmental, economic and social risks of air pollution are well understood and therefore countries around the world are taking steps to counteract it, in the first place by mitigating emissions. Therefore, there is an expectation that overall baseline air pollution is decreasing as well as the frequency, magnitude and duration of severe air pollution events. Our results show that indeed this is the case in 54 out of the 100 cities investigated (cities belonging to Group 4). However, this is not the case in 46 cities. In general, the situation is improving in Chinese cities, deteriorating in Australian cities, not changing in South American cities, and varying across European cities. In the US, the east coast is improving while the west coast is becoming worse.

Our results are indicative because the trends were not statistically significant for many of the cities; nevertheless, our findings provide a global overview for the first time. Longer observation times of the trends investigated are needed to determine the impact of regulations and the influence of changing climate and thus meteorological conditions (both usually require observation for more than 5 years). Further in-depth analysis of all the drivers in each individual city is also required. The length of time required to acquire sufficient observations to forecast trends is uncertain considering not only the variation in meteorology, but also natural and anthropogenic changes (e.g. the introduction of new regulations). Although we did not identify or model all such factors, as this would itself be a large task, our observational results remain informative of current trends. Nonetheless, repeating our analyses based on 10 years of data (after 2023), and possibly extending it by using monitoring data from additional locations or/and remote sensing data, would provide a much-improved insight.

Air pollution monitoring should be extended, because at present the data for some of the world's most heavily polluted cities are not available. This applies in particular to African cities and cities in India other than Delhi. The analysis should also be extended to geographic and political regions, rather than being focused only on cities.

Despite these limitations, there are several important findings from this study that point to actions that must be taken to reduce the burden of severe air pollution events.

Firstly, our results highlight that reduction of baseline air pollution on a long-term city and country basis will eliminate severe air pollution caused by anthropogenic sources. Therefore, reducing baseline air pollution should be the prime focus of national and regional pollution mitigation strategies.

Secondly, we acknowledge that this will not be sufficient to completely eliminate the "plague" of severe air pollution, as those events that are influenced by climate change and therefore change in local meteorology (fires or desert storms) are expected not only to continue but to worsen in severity, as the example of the 2019/2020 bushfires in Australia demonstrates (Yu et al., 2020). We should now face the reality that significant reductions in anthropogenic emissions that contribute to climate change from all sources are essential to reverse the escalating trend of severe air pollution events.

Declaration of Competing Interest

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

Acknowledgment

The authors would like to acknowledge the support of the: Australia-China Centre on Air Quality Science and Management (ACC-AQSM) to undertake and conduct this study; Department of Water and Environmental Regulation (DWER) in Western Australia for access to the data; Spanish Ministry of Science and Innovation and FEDER funds under the project HOUSE (CGL2016-78594-R), and by the Generalitat de Catalunya (AGAUR 2017 SGR41); Climate and Resilient Research (FONDAP 15110009), and the PAPILA (Prediction of Air Pollution in Latin America and the Caribbean) project (ID: 777544, H2020-EU.1.3.3.); ASAP-Delhi project (An Integrated Study of Air Pollutant Sources in the Delhi National Capital Region) funded by the Natural Environmental Research Council under the grant number NE/P016510/1; Stockholm Air and Noise Analysis at Stockholm City Environmental Department for the provision of the data; National Key R&D Program of China via grant No. 2017YFC0212001; National Air Pollution Surveillance (NAPS) program in Canada for data on $PM_{2.5}$.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2021.106732.

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