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Multi-pollutant air pollution and associated health risks in China from 2014 to 2020

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HIGHLIGHTS

- High multi-pollutant air pollution (MPAP) days were distributed over North China.
- $PM_{2.5}$-PM_{10} co-pollution accounted for 80–100% of MPAP days in winter.
- MPAP days declined due to decreases in $PM_{2.5}$-PM_{10} and $PM_{2.5}$-O_3 co-pollution days.
- 89% (20%) of population were exposed to unhealthy (very unhealthy) air in winter.
- Health risks in five provinces except Shanxi experienced remarkable declines.

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ABSTRACT

We characterized air pollution over China during 2014–2020, with a novel focus on multi-pollutant air pollution (MPAP) for the first time (defined as co-pollution of two or more contaminants) which was quite common in China but has not been evaluated, and assessed associated health risks of air pollution exposure which specially took into account the effects of exposure to multi-pollutant mixture, by introducing a health-risk based and multi-pollutant air quality index (HAQI). High values of MPAP days were mostly distributed over North China with a 7-year average of 51.9 MPAP days. The severest MPAP occurred in winter with $PM_{2.5}$-PM_{10} co-pollution accounting for 80–100% of MPAP days for six provinces of North China. One interesting phenomenon was that MPAP days exhibited almost overall declines (average trend of -11.9 days yr^{-1}) over North China during 2014–2020 due to decreases in $PM_{2.5}$-PM_{10} and $PM_{2.5}$-O_3 co-pollution days, while single-pollutant air pollution (SPAP) days showed almost overall rising trends with an average trend of $+8.0$ days yr^{-1} which was attributed to rapid increases in O_3 pollution days. The HAQI also exhibited high values over North China with a 7-year average HAQI value of 102.4. Health risks were the greatest in winter with 89.4% of population exposed to "unhealthy" air (HAQI >100) and 20.1% of population suffering from "very unhealthy" (HAQI >200) air over North China. Thanks to the mitigation of particulate pollution (shown as decreases in $PM_{2.5}$ pollution days, $PM_{2.5}$-PM_{10} co-pollution days, and $PM_{2.5}$-O_3 co-pollution days), health risks in five provinces of North China except Shanxi experienced remarkable declines with HAQI trends of -3.1 $yr^{-1}\sim -9.7$ yr^{-1} during 2014–2020. Unrelieved particulate pollution and deteriorating O_3 pollution, however, jointly led to rising health risks for Shanxi. These findings are expected to provide a scientific cognition of complex air pollution and associated health impacts.

1. Introduction

Air pollution in China has received intensive attention from the general public and the authorities concerned since 2011. The new 'Chinese Ambient Air Quality Standards' (CAAQS) released in 2012 sets six criteria pollutants including sulfur dioxide (SO_2), nitrogen dioxide (NO_2), carbon monoxide (CO), ozone (O_3), fine particulate matter ($PM_{2.5}$), and inhalable particulate matter (PM_{10}). Benefiting from 'Air

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Pollution Prevention and Control Action Plan' (Action Plan) promulgated in 2013 and 'Three-year Action Plan to Win the Blue Sky Defense War' (Blue Sky Defense War) issued in 2018, air quality in China has been greatly improved (Zhang et al., 2019; Gao et al., 2020a; Tao et al., 2020; Han et al., 2021). From 2015 to 2019, the annual mean SO_2, CO, NO_2, $PM_{2.5}$, and PM_{10} concentrations averaged over China decreased by 51.2%, 25.3%, 10.6%, 27.9%, and 23.8% (Guo et al., 2020). However, current air pollution in China is still serious, with 61.9% ($PM_{2.5}$) and 38.7% (PM_{10}) of 168 key cities exceeding CAAQS Grade II in 2020 (Ministry of Ecology and Environment of the People's Republic of China, 2021). During a widespread haze episode in January 2017, the hourly SO_2 concentration reached as high as 1300 μg/m^3 in Linfen, a city in Fen-Wei Plain (Shen et al., 2020). For NO_2, only 75.7% of 362 cities exhibited negative changes from 2015 to 2019 (Guo et al., 2020). Deteriorating O_3 pollution is arousing unprecedented concerns (Ma et al., 2016; Zhu et al., 2017; Cheng et al., 2018; Li et al., 2019, 2020; Gao et al., 2020b; Gu et al., 2020a, 2020b; Liu and Wang, 2020; Dang et al., 2021). Observed trends in annual average MDA8 O_3 concentrations during 2014–2018 were +4.2, +3.9, and +6.6 μg m^{-3} yr^{-1} for North China Plain, Yangtze River Delta, and Fen-Wei Plain, respectively (Chen et al., 2020).

Actually, the atmosphere we live by is not only exposed to a single contaminant, but also confronted with a complex mix of multiple pollutants. Although some previous studies have investigated the correlations between air pollutants (Zhu et al., 2019, 2021; Chu et al., 2020; Tian et al., 2020), less attention was paid to simultaneous pollution of two or more contaminants. Wang et al. (2019) found O_3 and $PM_{2.5}$ pollution simultaneously occurred on 8 days during June 2015 in Jinan, with a maximum 8-h-averaged O_3 concentration of 255 μg m^{-3} and a maximum daily averaged $PM_{2.5}$ concentration of 111 μg m^{-3}. Dai et al. (2021) reported that the days with co-pollution of $PM_{2.5}$ and O_3 were frequently observed in the Yangtze River Delta, which reached 54 days in Shanghai and 71 days in Jiangsu during 2013–2019. Co-occurrence of $PM_{2.5}$ and O_3 pollution was also reported in other regions of China (Tang et al., 2018; Miao et al., 2021). However, there are very few studies focusing on other types of multi-pollutant air pollution except $PM_{2.5}$-O_3 co-pollution. Only two recent studies examined multi-contaminant air pollution in China which incorporated five criteria pollutants except CO (Han et al., 2018, 2020). Based on the World Health Organization (WHO) guideline limits, high daily mixture levels of $PM_{2.5}$ and PM_{10}, $PM_{2.5}$ and SO_2, and PM_{10} and SO_2 occurred on over 146 days in 145 cities in 2014 (Han et al., 2018).

Air pollution caused 4.9 million deaths worldwide and 1.2 million deaths in China in 2017 based on analysis for the Global Burden of Disease Study 2017 (Stanaway et al., 2018; Yin et al., 2020). The Air Quality Index (AQI) is used by authorities to release real-time air quality information to the general public, from which certain behaviors are advised to respond to health risks of air pollution. Currently, the most commonly used index is single-contaminant-oriented index, which was firstly introduced and implemented by the United State Environmental Protection Agency and applied in other countries (e.g., mainland China and South Korea) with different breakpoints. The index takes the maximum sub-AQI as the overall AQI after calculating every sub-AQI based on the concentration of every criteria pollutant. Therefore, the single-contaminant-oriented AQI ignores the combined health effects of exposure to multiple contaminants. Correspondingly, multi-contaminant-oriented index, namely aggregate air quality index (AAQI) was proposed and developed (Swamee and Tyagi, 1999; Kyrkilis et al., 2007). However, although AAQI takes into account the exposure to multi-pollutant mixture, it does not explicitly incorporate the established exposure-response relationships of contaminants. In recent years, risk-based and multi-pollutant air quality health indexes (AQHI or HAQI) have been introduced and developed to consider such exposure-response characteristics (Stieb et al., 2008; Wong et al., 2013; Hu et al., 2015; Du et al., 2020; Tan et al., 2021). Currently, AQHI (or HAQI) is adopted in Canada and Hong Kong, China.

As mentioned above, China has entered the stage of complex air pollution. However, very few studies have characterized multi-contaminant air pollution over China. What's more, the lack of investigation on multi-pollutant air pollution has also hindered further multi-contaminant health analysis. The current AQI used in mainland China may underestimate the health risks of simultaneous exposure to multi-pollutant mixture. Although existing studies have developed risk-based and multi-pollutant air quality indexes, health risk assessment of multi-contaminant mixture at a national scale for recent years using these indices remains scarce. This paper aims to: (1) obtain the spatial-temporal characteristics of air pollution over China with a novel focus on multi-pollutant air pollution, based on the concentrations of all the six criteria pollutants measured at all monitoring sites for years 2014–2020, and (2) assess the health risks of air pollution exposure (taking into account the exposure to multi-pollutant mixture), by using a health-risk based and multi-pollutant air quality index (HAQI). To the best of our knowledge, this is the first paper showing the spatial-temporal variations in multi-pollutant air pollution and associated health risks of exposure to multi-contaminant mixture at a national scale in recent years, based on observed concentrations of all the six criteria pollutants over China. The obtained knowledge is expected to provide a scientific cognition of complex air pollution and associated health impacts over China.

2. Materials and methods

2.1. Air quality data

Ministry of Ecology and Environment of the People's Republic of China has begun to release to the public the real-time monitoring concentrations of six criteria pollutants (SO_2, NO_2, CO, O_3, $PM_{2.5}$, and PM_{10}) at a national scale since 2013. Due to the unavailability of partial data in 2013, we obtain the observed hourly concentrations for years 2014–2020 at http://106.37.208.233:20035/. The monitoring network covers 1701 sites across China in 2020. For each site, the negative or missing values are removed; those sites with less than 80% valid data are abandoned. Following the CAAQS, we also calculate 24-h (or 8-h) average concentrations for exceedance judgment. The data validity treatment is conducted as follows: the 24-h average concentrations (for SO_2, NO_2, CO, $PM_{2.5}$, and PM_{10}) are calculated when there are valid data for at least 20 h during that day; the 8-h average concentrations (for O_3) are calculated when there are valid data for more than 6 h for every 8 h. Because high values of air pollution days and associated health risks are mostly distributed over North China (See Sections 3.1.1 & 3.2.1), the seasonality and trend analyses are focused on North China which in this paper includes Beijing, Tianjin, Hebei, Shanxi, Shandong, and Henan.

2.2. Multi-pollutant air pollution (MPAP)

The CAAQS Grade II sets limits on 24-h average concentrations for SO_2 (150 μg m^{-3}), NO_2 (80 μg m^{-3}), CO (4 mg m^{-3}), $PM_{2.5}$ (75 μg m^{-3}), and PM_{10} (150 μg m^{-3}), and maximum daily 8-h average (MDA8) O_3 concentration (160 μg m^{-3}) or maximum daily 1-h average (MDA1) O_3 concentration (200 μg m^{-3}). An air pollution (AP) day is defined as a day with at least one pollutant exceeding the CAAQS Grade II limits. A single-pollutant air pollution (SPAP) day is defined as a day with only one pollutant exceeding the CAAQS Grade II limit. A multi-pollutant air pollution (MPAP) day is defined as a day with two or more pollutants simultaneously exceeding the CAAQS Grade II limits. In this study, MPAP is also denoted as multi-contaminant air pollution, multi-pollutant mixture, complex air pollution, a complex mix of multiple pollutants, simultaneous pollution, co-pollution, etc.

2.3. Health-risk based and multi-pollutant air quality index (HAQI)

Currently, the AQI released to the general public is a single-contaminant-oriented index in mainland China, which takes the maximum Individual AQI (IAQI) as the overall AQI after calculating every IAQI based on the concentration of every criteria pollutant. The IAQI of 100 corresponds to the CAAQS Grade II limit of corresponding pollutant concentration. Therefore, a day with AQI larger than 100 indicates an AP day. As mentioned in Introduction, the single-contaminant-oriented AQI may underestimate the combined health risks of simultaneous exposure to multi-pollutant mixture. A health-risk based and multi-pollutant air quality index (HAQI) can address this issue by taking into account the exposure to multi-contaminant mixture and explicitly incorporating the established exposure-response relationships of pollutants.

HAQI is calculated as follows: firstly, the relative risk (RR_i) of pollutant i, calculated from concentration-response function, is estimated using a general form (Shang et al., 2013; Hu et al., 2015; WHO, 2016):

$$RR_i = exp\left[\beta_i\left(C_i - C_{i,0}\right)\right],\ C_i > C_{i,0} \tag{1}$$

where β_i is the exposure-response coefficient of air pollutant i; C_i is the observed concentration of air pollutant i; $C_{i,0}$ is the threshold concentration of air pollutant i. The β_i values for pollutants are taken from Shang et al. (2013). The threshold concentration of air pollution is a baseline or reference exposure against which the health impacts of air pollution are calculated. This level of air pollution may be defined differently in different health risk assessments. It may be defined as the national air quality standard, the WHO air quality guideline (AQG) level, the natural level, or the lowest level observed in epidemiological studies (WHO, 2016). This study focuses on air pollution and health risks in China, therefore the CAAQS Grade II limits are chosen as the $C_{i,0}$ values, based on the assumption that air pollution causes no health risks if concentrations of contaminants are lower than CAAQS Grade II limits which indicates that $RR_i = 1$ when $C_i \le C_{i,0}$.

The total excess risk (ER_{total}) of all pollutants is calculated as the sum of excess risk of every pollutant (ER_i), which is defined as ($RR_i -$ 1) (Cairncross et al., 2007). ER_{total} takes the following form:

$$ER_{total} = \sum_{i=1}^{n} ER_i = \sum_{i=1}^{n} (RR_i - 1) \tag{2}$$

Here, the equivalent concentration of pollutant i (C_i^*) is introduced and defined as the concentration at which ER of pollutant i is equal to ER_{total} (Hu et al., 2015), as shown in the following equation:

$$RR^* = ER_{total} + 1 = exp\left[\beta_i\left(C_i^* - C_{i,0}\right)\right] \tag{3}$$

where RR^* is the relative risk of pollutant i calculated by using the equivalent concentration C_i^*. Therefore, C_i^* can be shown as:

$$C_i^* = \ln(RR^*)/\beta_i \quad + \quad C_{i,0} \tag{4}$$

As we can see, RR^* represents the total risks of multiple exposures; therefore, the equivalent concentration (C_i^*) actually incorporates the combined health risks from all contaminants. After obtaining C_i^* values for all criteria pollutants, we can calculate HAQI similarly to AQI by using C_i^* values instead of actual concentrations C_i (Hu et al., 2015). The calculation procedures for AQI can be obtained in 'Technical Regulation on Ambient Air Quality Index' (Ministry of Environmental Protection of the People's Republic of China, 2012). HAQI has been successfully applied in air quality studies over China (Hu et al., 2015; Shen et al., 2017, 2020).

3. Results and discussion

3.1. Spatiotemporal characteristics of multi-pollutant air pollution

3.1.1. Spatial distributions of MPAP days

Fig. 1(a–c) show the spatial distributions of AP days, SPAP days, and MPAP days over China averaged during 2014–2020. The AP days exhibited high values over North China (including Beijing, Tianjin, Hebei, Shanxi, Shandong, and Henan), especially southern Hebei, northern Henan, and western Shandong with AP days exceeding 160. The 7-year average number of AP days was calculated to be 32.4 days for China and 117.7 days for North China. Besides North China, Xinjiang located in northwestern China was another hotspot with high AP days, which was attributed to the source region of dust (Luo et al., 2020). Generally, air pollution in northern China was more severe than that in southern China due to excessive emissions as well as unique topographic and climate features (He et al., 2017; Fan et al., 2020; Kuerban et al., 2020; Tian et al., 2020). Song et al. (2017) reported that the annual concentrations of six criteria pollutants in northern China were 5.9–96.7% higher than those in southern China during 2014–2016. The SPAP days and MPAP days exhibited the similar spatial distributions to AP days; high values of SPAP days and MPAP days were mostly distributed over North China. The 7-year average number of SPAP (MPAP) days was calculated to be 24.1 (8.3) days for China and 65.9 (51.9) days for North China.

We further pay our attention to MPAP. The proportions of MPAP days to AP days are shown in Fig. 1(d). Similarly, the high ratios were also distributed over North China; the MPAP days accounted for 25.6% (for China) and 44.1% (for North China) of AP days, indicating that the health risks reflected by current AQI were underestimated on nearly a quarter (for China) and half (for North China) of air pollution days.

3.1.2. Seasonal variations in MPAP days

Fig. 2 shows the seasonal variations in MPAP days and pollution type for six provinces in North China which suffered from the severest MPAP. In this study, the primary pollutants (or pollution type) of multi-pollutant air pollution are defined as the two pollutants with the maximum and the second maximum Individual AQI. The severest MPAP occurred in winter; the MPAP days in a month reached 4.4, 8.5, 12.5, 9.8, 9.2, and 13.7 days, respectively, for Beijing, Tianjin, Hebei, Shandong, Shanxi, and Henan in winter. The primary pollution type was $PM_{2.5}$-PM_{10} co-pollution in winter, which occurred on 100% (for Hebei, Shandong, and Henan), 95% (for Shanxi), and 80% (for Beijing and Tianjin) of MPAP days. Particulate matter was the greatest threat to air quality in winter during the past decade; the combination of fine particulate and coarse particulate was the primary MPAP type in winter for North China. For Shanxi, the $PM_{2.5}$-SO_2 co-occurrence accounted for 5% of MPAP days in winter. Shanxi is a province well-known for the largest coal reserves in China; high SO_2 emissions from coal-fired power plants lead to SO_2 pollution in Shanxi (Song and Yang, 2014). Shen et al. (2020) also reported the highest SO_2 concentrations in Shanxi over China by analyzing SO_2 measurements in 2018. The $PM_{2.5}$-NO_2 co-pollution occurred on approximately 15% of MPAP days in Beijing and Tianjin. NO_2 is considered as an indicator of combustion sources, such as motor vehicle emissions in urban areas; high vehicle ownership contributes to NO_2 pollution in Beijing and Tianjin (Zheng et al., 2019; Shen et al., 2021).

There was almost no MPAP in summer except Beijing which suffered from mild MPAP with 2.6 days in a month. The $PM_{2.5}$-O_3 co-pollution accounted for nearly 100% of MPAP days in summer for Beijing. The complex air pollution caused by $PM_{2.5}$ and O_3 has been an emerging problem that threatens public health in Chinese megacities during recent years (Song et al., 2017; Tang et al., 2018; Wang et al., 2019; Dai et al., 2021; Miao et al., 2021; Yang et al., 2021). It is noted that $PM_{2.5}$ concentrations over China exhibit a remarkable seasonal variability with the highest in winter and the lowest in summer; wintertime high

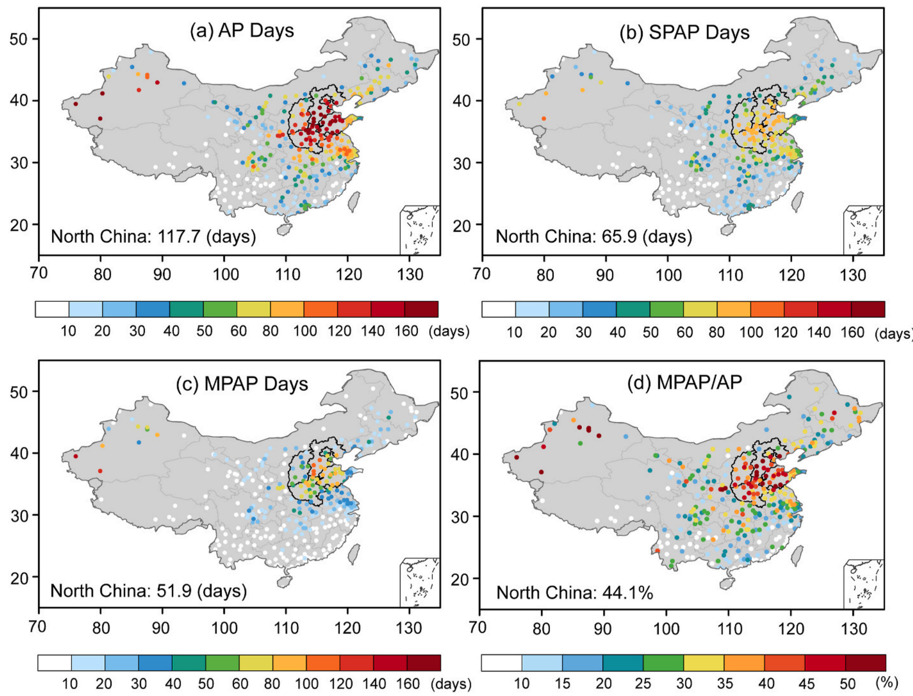

Fig. 1. Spatial distributions of (a) air pollution (AP) days, (b) single-pollutant air pollution (SPAP) days, (c) multi-pollutant air pollution (MPAP) days, and (d) proportions of MPAP days to AP days over China averaged during 2014–2020. The pollution days (and proportion) for North China are shown at the lower left corner of each panel.

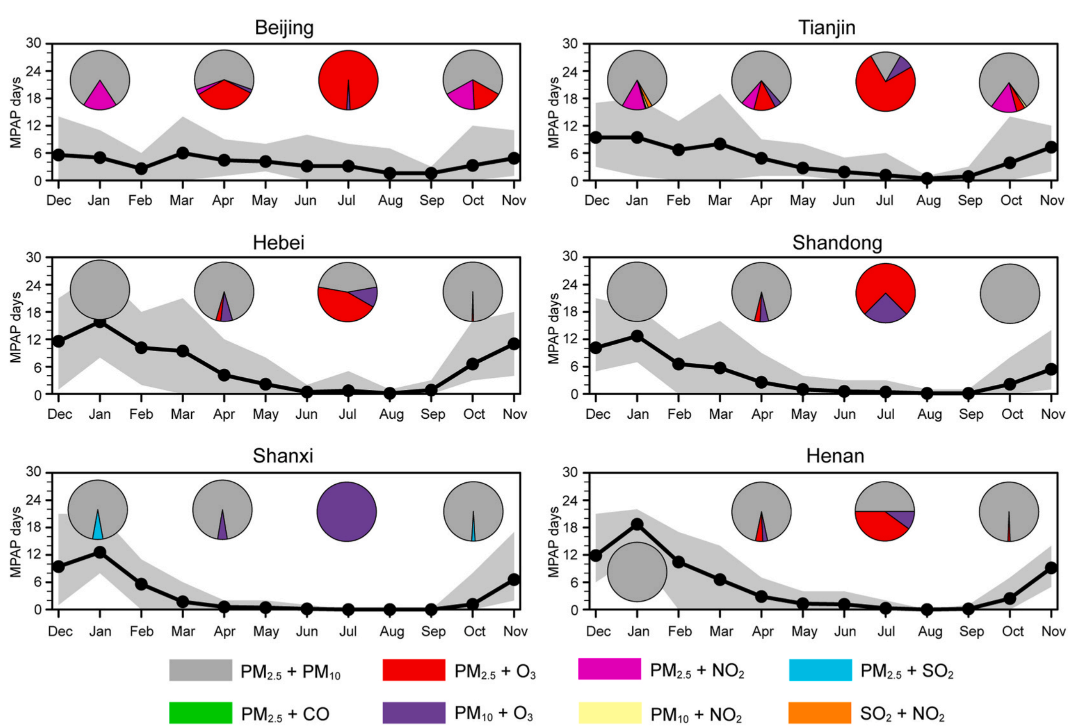

Fig. 2. Seasonal variations in MPAP days for six provinces in North China. Black lines represent 7-year average monthly MPAP days and grey shadings represent the range of MPAP days during the seven years 2014–2020. Pie charts exhibit the proportions of MPAP pollution type during each season.

levels are associated with enhanced anthropogenic emissions (e.g., fossil fuel combustion and biomass burning for residential heating) and unfavorable meteorological conditions (e.g., low boundary layer height) (Zhang and Cao, 2015; Fan et al., 2020; Shen et al., 2020). Except the elevated PM_{10} owing to dust events in spring in some western and northern cities, PM_{10} shows a similar seasonal variation as $PM_{2.5}$ in most cities of China (Wang et al., 2014). Therefore, there are few $PM_{2.5}$-PM_{10} MPAP days in summer. During spring and autumn, the $PM_{2.5}$-PM_{10} co-pollution occurred on more than 90% of MPAP days in Hebei, Shandong, Shanxi, and Henan. For Beijing and Tianjin, the $PM_{2.5}$-PM_{10} co-occurrence accounted for 60–80% of MPAP in spring and autumn, followed by $PM_{2.5}$-O_3 co-pollution and $PM_{2.5}$-NO_2 co-pollution.

3.1.3. Seven-year trends in MPAP days

The seven-year trends in AP, SPAP, and MPAP days during 2014–2020 are displayed in Fig. 3. We use linear regression analysis, one of the conventional methods of analyzing trends in environmental data, to calculate the trends in pollution days (and HAQI index in Section 3.2.3). The regression coefficient (i.e., slope) output by the linear fitting model is regarded as the trend (Nunifu and Fu, 2019). The AP days generally exhibited downward trends in Central and East China and Northeast China, and small changes in other regions of China. Interestingly, significant decreases of AP days were found in five provinces of North China except Shanxi while remarkable increases occurred in Shanxi. On average, the trend was calculated to be -3.9 days yr^{-1} for North China. Further investigation on trends in SPAP and MPAP days accounted for the distinct changes of AP days between Shanxi and other five provinces. The SPAP days showed almost overall rising trends in all provinces of North China with an average trend of $+8.0$ days yr^{-1} for North China. The MPAP days, by contrast, exhibited almost overall declines in all provinces of North China with an average trend of -11.9 days yr^{-1}; it is noted that, however, Shanxi suffered from much weaker decreases (-2.6 days yr^{-1}) compared with other five provinces of North China. The significant declining increasing trends in MPAP days overpowered the increasing trends in SPAP days, leading to the decreases of AP days for the five provinces of North China except Shanxi; the weak decreasing trends in MPAP days were reversed by the remarkable rising trends in SPAP days, resulting in the increases of AP days for Shanxi.

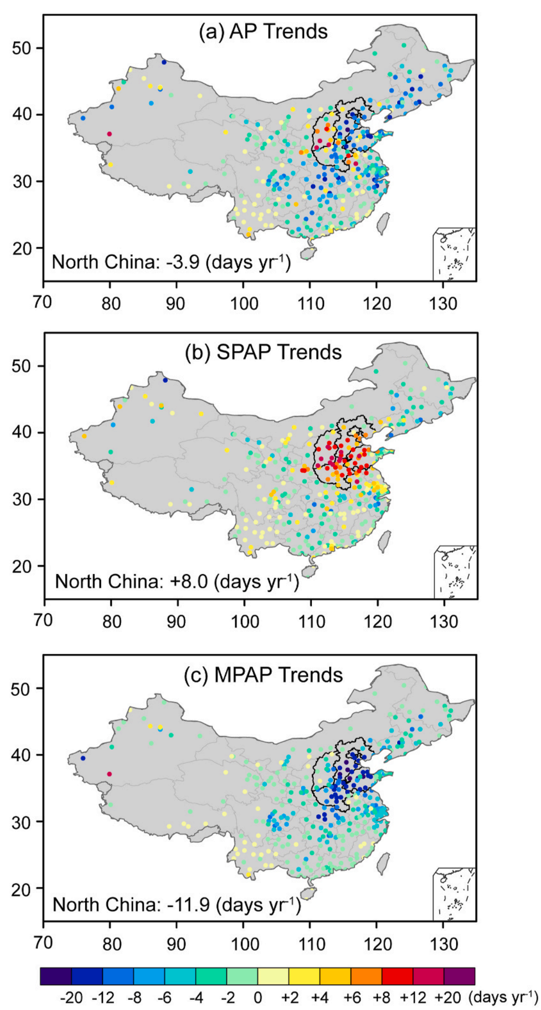

Fig. 3. Seven-year trends in (a) AP, (b) SPAP, and (c) MPAP days over China during 2014–2020. The trends for North China are shown at the lower left corner of each panel.

To reveal the reasons for the inverse trends in SPAP days and MPAP days, we further examine the yearly evolutions of SPAP (MPAP) days and corresponding pollution type for six provinces of North China from 2014 to 2020 in Fig. 4 (Fig. 5). The rapid increases in O_3 pollution days contributed to the rising trends in SPAP days for all the six provinces of North China. The trends in O_3 pollution days (days with only O_3 exceeding the limit) were $+7.1$ ~ $+12.7$ days yr^{-1} for the six provinces. For Shanxi, the unrelieved particulate pollution, in addition to deteriorating O_3 pollution, intensified SPAP rises. The rapid decreases in $PM_{2.5}$-PM_{10} co-pollution days made the largest contributions to MPAP declines for Beijing, Tianjin, Hebei, Shandong, and Henan. Benefiting from $PM_{2.5}$ pollution mitigation, the decrease in $PM_{2.5}$-O_3 co-pollution became the second contributor to MPAP decline for Beijing in spite of worsening O_3 pollution. It is noted that $PM_{2.5}$-NO_2 co-pollution was emerging as the primary MPAP type in Tianjin, which accounted for half of MPAP days in 2020. For Shanxi, the $PM_{2.5}$-PM_{10} co-pollution was not drastically alleviated from 2014 to 2020, which led to weaker MPAP decreases.

3.2. Health risks of multi-pollutant air pollution

3.2.1. Spatial distributions of HAQI

The concentrations among different pollutants vary by orders of magnitude; their health impacts are also different. Therefore, a dimensionless index called Air Quality Index (AQI), in place of concentration, is used to indicate air pollution levels and corresponding health risks to the Chinese public. Based on the AQI value, air quality is divided into six health risk levels and certain behaviors are advised to respond to health risks. Air quality on a given day is defined as "unhealthy" when AQI is greater than 100. However, as mentioned above, single-contaminant-oriented AQI may underrate the health risks associated with simultaneous exposure to multi-pollutant mixture. A health-risk based and multi-pollutant air quality index (HAQI) has shown improvements over the current AQI in evaluating the health risks of exposure to simultaneous pollution (Hu et al., 2015; Shen et al., 2017).

The spatial distributions of AQI, HAQI, and their difference (HAQI minus AQI) over China averaged during 2014–2020 are displayed in Fig. 6. Both AQI and HAQI exhibited high values over North China; compared with AQI, HAQI showed increases across China except the south. The 7-year average AQI (HAQI) was calculated to be 95.2 (102.4) for North China. Because HAQI was higher than AQI on MPAP days, the increases from AQI to HAQI were mostly distributed over North China which suffered from the severest MPAP (Fig. 1(c)). The health risk category changed from "healthy" ($95.2 < 100$) to "unhealthy" ($102.4 > 100$) for North China when the combined health effects of exposure to multiple contaminants were considered. Although the average increment was only 7.2 from AQI to HAQI for North China, the largest increases reached 20–30 in southern Hebei, where the maximum MPAP days occurred.

3.2.2. Seasonal variations in HAQI

The monthly variations in HAQI for North China are shown in Fig. 7 (a). Generally, the HAQI values were in an order of winter>spring>autumn>summer. The 7-year average monthly HAQI exceeded 100 in December, January, February, March, June, and November, indicating that the air was "unhealthy" for the public in half of months during a year over North China. The greatest health risk was in winter with an average HAQI of 140. This was in agreement with Hu et al. (2015) which reported the maximum HAQI value in winter based on air pollution data during March 2013 to February 2014.

For further investigation on population exposure, Fig. 7(b) depicts the cumulative distributions of population-weighted seven-year average HAQI values for North China during four seasons. The health risks were significantly greater in winter than those in other three seasons; 89.4% of population were exposed to "unhealthy" air (HAQI >100) and 20.1% of population were even exposed to "very unhealthy" (HAQI >200) air during winter. The largest health risk of air pollution in winter was

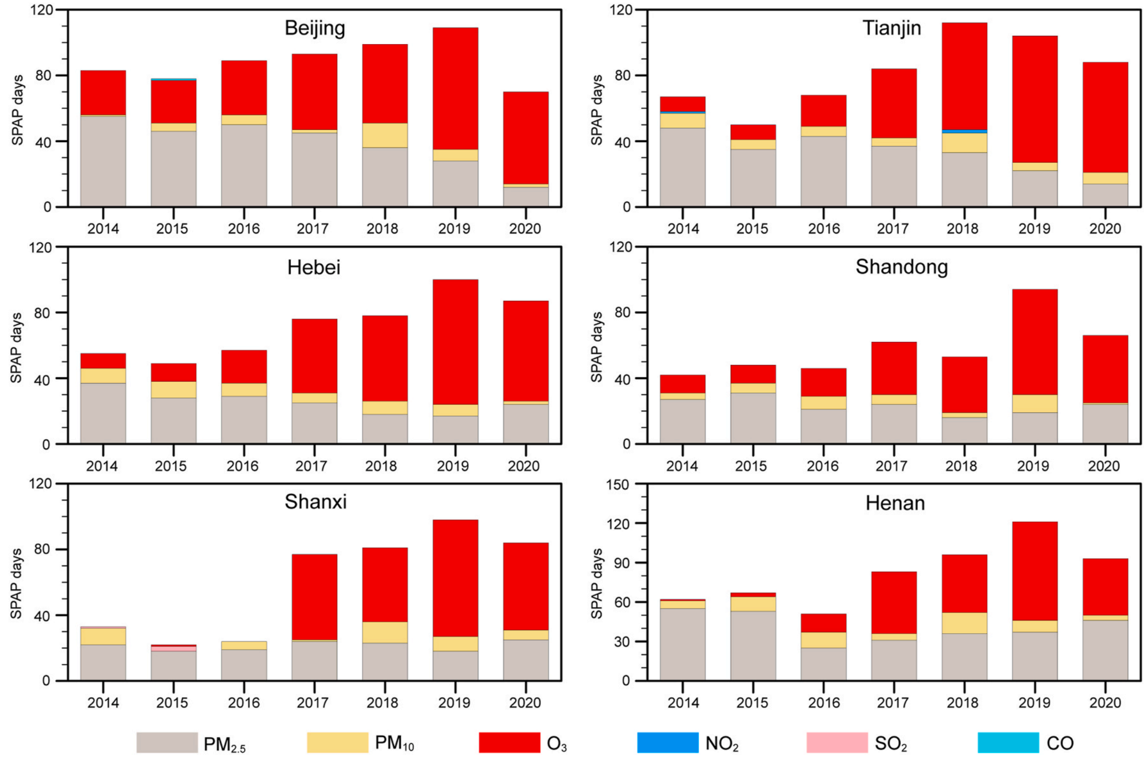

Fig. 4. Yearly evolutions of SPAP days and pollution type for six provinces of North China from 2014 to 2020.

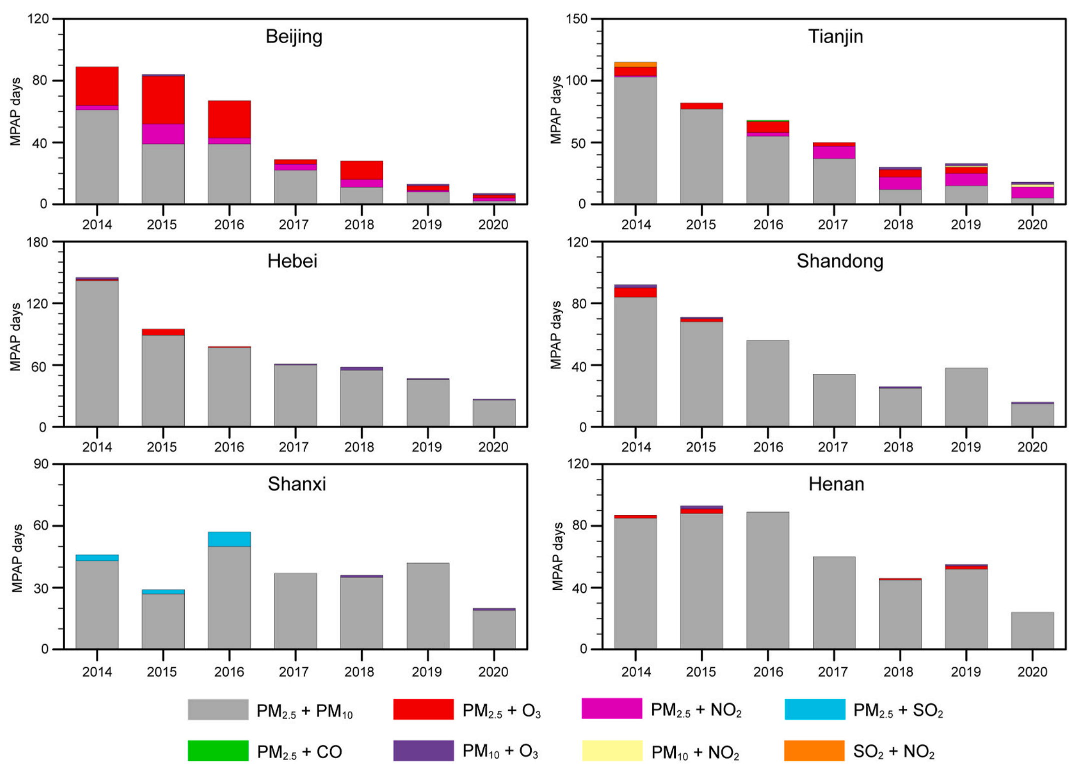

Fig. 5. Yearly evolutions of MPAP days and pollution type for six provinces of North China from 2014 to 2020.

followed by moderate health risk in spring with 62.1% of population living in "unhealthy" air. The health risks were comparatively lower in autumn and summer; nearly half population, however, still suffered from "unhealthy" air even during the two seasons.

3.2.3. *Seven-year trends in HAQI*

Profiting from a series of air pollution control plans, the $PM_{2.5}$, PM_{10}, SO_2, and CO air quality have been significantly improved and NO_2 levels have also been gradually falling while O_3 air quality has been deteriorating since 2013 over most regions of China. Different and even inverse

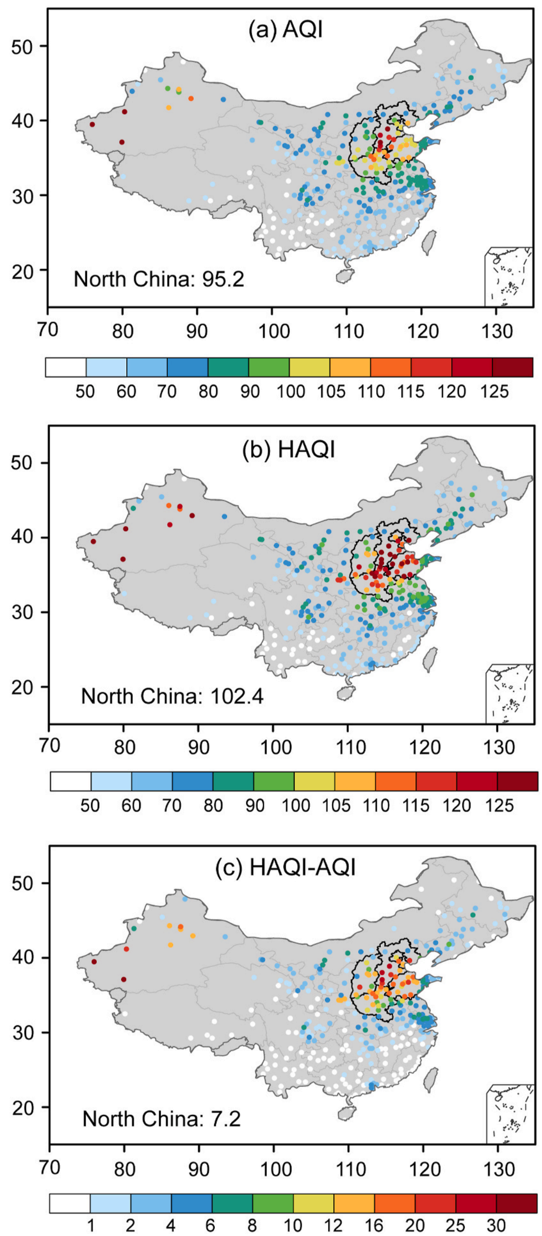

Fig. 6. Spatial distributions of (a) AQI, (b) HAQI, and (c) HAQI-AQI over China averaged during 2014–2020. The indexes for North China are shown at the lower left corner of each panel.

changes in concentrations of different pollutants lead to the difficulty in assessing the trends in health risks of exposure to a complex mix of multiple pollutants.

Here we show in Fig. 8 the yearly evolutions and trends in HAQI, a multi-contaminant-oriented index, to evaluate the changes in health risks of exposure to complex air pollution. Negative HAQI trends occurred in Central and East China and Northeast China, and weaker trends were found in other regions of China (Fig. 8(a)). The most significant downward trends were distributed in five provinces of North China except Shanxi. On average, the HAQI trend was calculated to be -3.5 yr^{-1} for North China. We further explore the yearly evolutions of HAQI for the six provinces of North China in Fig. 8(b). The HAQI in five provinces except Shanxi experienced remarkable declines during 2014–2020 with trends of -3.1 yr^{-1}~ -9.7 yr^{-1} while the trend in Shanxi was calculated to be $+2.1$ yr^{-1}. That is to say, the health risks of air pollution exposure have been shrinking in the five provinces during the past seven years when the impact of exposure to multi-pollutant

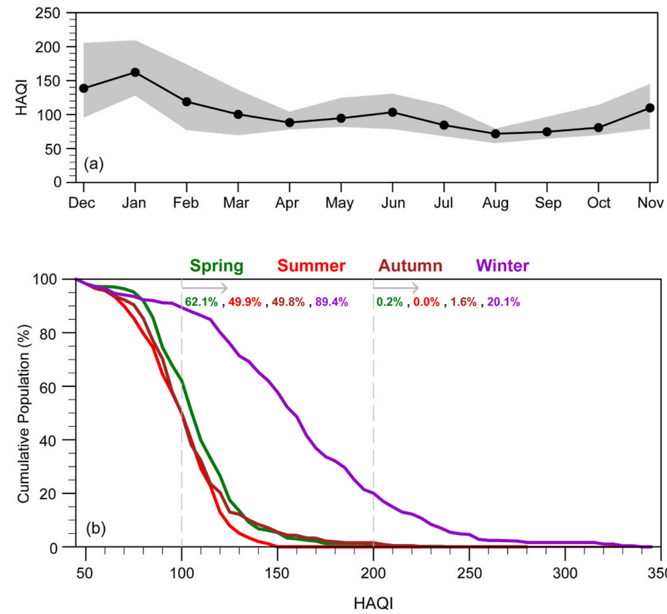

Fig. 7. (a) Monthly variations in HAQI for North China. Black line represents 7-year average monthly HAQI and grey shading represents the range of HAQI during the seven years 2014–2020. (b) Cumulative distributions of population-weighted seven-year average HAQI values for North China during four seasons. The population exposed to “unhealthy” (HAQI >100) and “very unhealthy” (HAQI >200) air in each season are also listed in colored values.

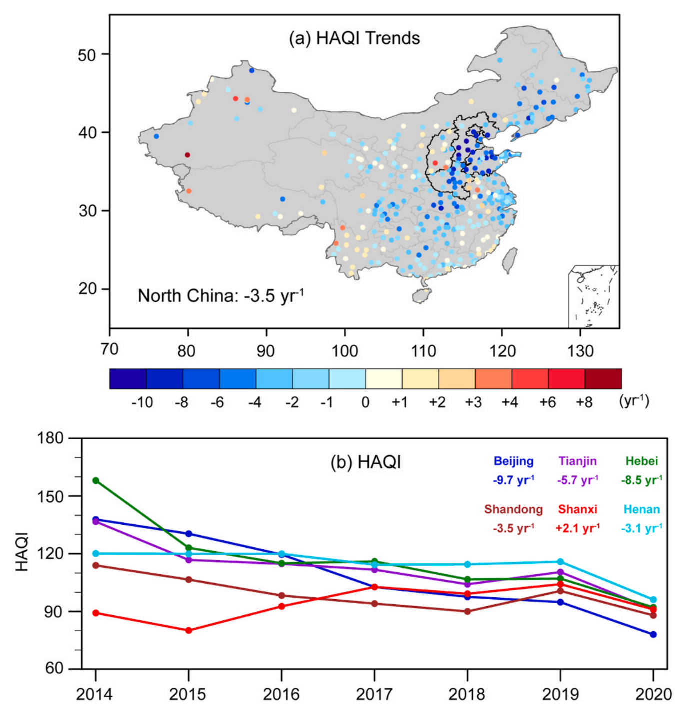

Fig. 8. (a) Seven-year trends in HAQI over China during 2014–2020. The HAQI trend for North China is shown at the lower left corner. (b) Yearly evolutions of HAQI for six provinces in North China from 2014 to 2020. The HAQI trends for each province are listed at the top right corner. Note that the HAQI trend for North China calculated based on average concentrations of North China is not exactly the mean value of HAQI trends for six provinces calculated based on average concentrations of each province.

mixture was considered. The mitigation of particulate pollution was the greatest contributor to the declines of health risks for the five provinces of North China since $PM_{2.5}$ pollution days, $PM_{2.5}$-PM_{10} co-pollution days, and $PM_{2.5}$-O_3 co-pollution days exhibited significant decreases (Figs. 4 and 5). However, for Shanxi, the unrelieved particulate pollution and deteriorating O_3 pollution jointly resulted in rising health risks.

We also present yearly evolutions of excess risk from six criteria pollutants in Fig. S1 (in Supplementary Material). Similar to HAQI, the total excess risk exhibited significant decreases in five provinces except Shanxi and a slight increase in Shanxi during 2014–2020. For the five provinces except Shanxi, the percentage contributions of particulates ($PM_{2.5}$ plus PM_{10}) to total excess risk fell from 75%–99% in 2014 to 17%–76% in 2020, while those of O_3 rose from 0%–12% in 2014 to 24%–81% in 2020. The contributions of the other three pollutants (i.e., NO_2, SO_2, CO) were much smaller. For Shanxi, the percentage contribution of particulates to total excess risk showed a slight change but that of O_3 rose from 0% in 2014 to 40% in 2020. This, again, revealed that the declines of health risks in the five provinces except Shanxi during 2014–2020 were attributed to the mitigation of particulate pollution while the unrelieved particulate pollution and deteriorating O_3 pollution jointly contributed to rising health risks for Shanxi.

4. Conclusions

By analyzing concentrations of all six criteria pollutants (SO_2, NO_2, CO, O_3, $PM_{2.5}$, and PM_{10}) measured at all monitoring sites of China during 2014–2020, we investigated the spatial-temporal characteristics of air pollution over China with a novel focus on multi-pollutant air pollution (MPAP) which was quite common in China but has not been systematically evaluated. By introducing a health-risk based and multi-pollutant air quality index (HAQI) which shows improvements over the current AQI used in mainland China in evaluating health risks of exposure to multi-pollutant mixture, we also assessed the health risks of air pollution over China during 2014–2020.

The MPAP days accounted for 25.6% and 44.1% of air pollution (AP) days for the whole China and North China respectively during 2014–2020, indicating that the health risks reflected by existing AQI were underestimated on nearly a quarter (for China) and half (for North China) of air pollution days. High values of MPAP days were mostly distributed over North China; the 7-year average number of MPAP days was calculated to be 51.9 days for North China.

The severest MPAP occurred in winter with the MPAP days reaching 4.4–13.7 days in a month for the six provinces of North China. The primary pollution type was $PM_{2.5}$-PM_{10} co-pollution in winter, occurring on 100% (for Hebei, Shandong, and Henan), 95% (for Shanxi), and 80% (for Beijing and Tianjin) of MPAP days. For Shanxi, the $PM_{2.5}$-SO_2 co-occurrence accounted for 5% of MPAP days in winter; for Beijing and Tianjin, the $PM_{2.5}$-NO_2 co-pollution occurred on approximately 15% of MPAP days. In summer, only Beijing suffered from mild MPAP which was almost entirely $PM_{2.5}$-O_3 co-pollution. During spring and autumn, the $PM_{2.5}$-PM_{10} co-pollution occurred on more than 90% of MPAP days in Hebei, Shandong, Shanxi, and Henan; for Beijing and Tianjin, the $PM_{2.5}$-PM_{10} co-occurrence accounted for 60–80% of MPAP, followed by $PM_{2.5}$-O_3 and $PM_{2.5}$-NO_2 co-pollution.

The single-pollutant air pollution (SPAP) days showed almost overall rising trends during 2014–2020 in all provinces of North China with an average trend of $+8.0$ days yr^{-1}, which could be attributed to the rapid increases in O_3 pollution days. On the contrary, the MPAP days exhibited almost overall declines in all provinces of North China with an average trend of -11.9 days yr^{-1}, resulting from the decreases in $PM_{2.5}$-PM_{10} and $PM_{2.5}$-O_3 co-pollution days. It was noted that the $PM_{2.5}$-PM_{10} co-pollution was not drastically alleviated from 2014 to 2020 for Shanxi, leading to weaker MPAP decreases in Shanxi than those in other five provinces. As a result, significant decreases of AP days were found in five provinces of North China except Shanxi while remarkable increases of AP days occurred in Shanxi.

We further examined the health risks of air pollution reflected by HAQI featuring the consideration of exposure to multi-pollutant mixture. The HAQI exhibited high values over North China with a 7-year average HAQI value of 102.4. It was noted that, when the combined health effects of exposure to multiple contaminants were considered, the health risk category changed from "healthy" (AQI: $95.2 < 100$) to "unhealthy" (HAQI: $102.4 > 100$) for North China. What's more, the air was "unhealthy" for the public in half of months during a year over North China. Generally, the HAQI values were in an order of winter>spring>autumn>summer for North China; 89.4% of population were exposed to "unhealthy" air (HAQI >100) and 20.1% of population were even exposed to "very unhealthy" (HAQI >200) air during winter; 62.1% of population were living in "unhealthy" air in spring; nearly half population suffered from "unhealthy" air in autumn and summer. During 2014–2020, the HAQI in five provinces except Shanxi experienced remarkable declines with trends of -3.1 $yr^{-1}\sim -9.7$ yr^{-1} which could be attributed to the mitigation of particulate pollution. On the contrary, the trend of HAQI in Shanxi was calculated to be $+2.1$ yr^{-1}; the unrelieved particulate pollution and deteriorating O_3 pollution together resulted in rising health risks for Shanxi.

There are some uncertainties in our study that need to be addressed in future studies. Firstly, the CAAQS Grade II limits are chosen as the threshold concentrations in this study during the HAQI calculation procedure. However, comparative studies using different threshold concentrations reported large variations among estimated health impacts (Gao et al., 2015; Hu et al., 2015; Liu et al., 2018), highlighting the research need of the threshold values of air pollutants in future studies. Besides, it is noted that the total excess risk of all pollutants is calculated as the sum of excess risk of every pollutant during the HAQI calculation procedure. The assumption that the health effects of individual pollutants are additive ignores the fact that there are indeed non-additive interactions (e.g., synergism or antagonism) among pollutants in reality. Although several methods (e.g., source identification method, indicator approach, statistical interaction approach) have been developed to assess the health risks of exposure to multi-pollutant mixture during the past years, the interaction mechanism among pollutants remains unclear and health risk assessment of multiple exposures is full of challenges (Dominici et al., 2010; Hu et al., 2015). Nonetheless, the spatiotemporal characteristics of health risks of multi-pollutant exposure reflected by HAQI in this paper still hold true. Additional research is needed to improve HAQI by sorting out the associations between HAQI and observed health impacts, in order to describe the health risks of multi-contaminant exposure more accurately in the future.

CRediT authorship contribution statement

Jia Zhu: Investigation, Resources, Visualization, Formal analysis, Writing – original draft, Funding acquisition. **Lei Chen:** Conceptualization, Software, Validation, Writing – review & editing, Supervision, Funding acquisition. **Hong Liao:** Writing – review & editing, Funding acquisition.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2021.118829.

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