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Impact of different sources on the oxidative potential of ambient particulate matter PM_{10} in Riyadh, Saudi Arabia: A focus on dust emissions

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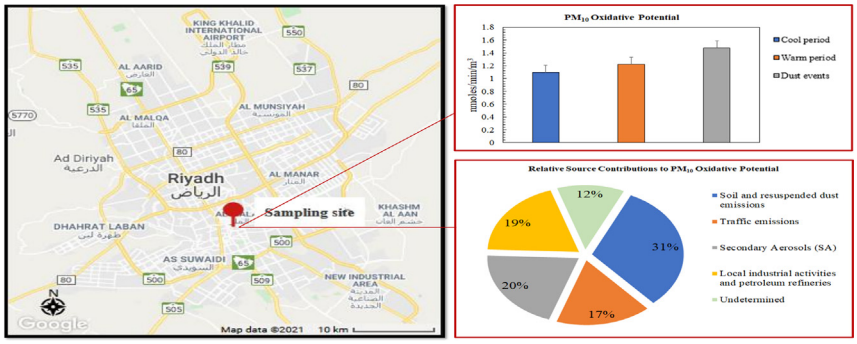
HIGHLIGHTS

- Sources of PM_{10} toxicity in the Riyadh metropolitan area were identified.
- PCA combined with MLR was used to perform oxidative potential source apportionment.
- Soil and resuspended dust emissions is the major contributor to PM_{10} oxidative potential.
- The oxidative potential of total PM_{10} was increased during dust events.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, we employed Principal Component Analysis (PCA) and Multi-Linear Regression (MLR) to identify the most significant sources contributing to the toxicity of PM_{10} in the city center of Riyadh. PM_{10} samples were collected using a medium-volume air sampler during cool (December 2019–March 2020) and warm (May 2020–August 2020) seasons, including dust and non-dust events. The collected filters were analyzed for their chemical components (i.e., water-soluble ions, metals, and trace elements) as well as oxidative potential and elemental and organic carbon (EC/OC) contents. Our measurements revealed comparable extrinsic oxidative potential (P-value = 0.30) during the warm (1.2 ± 0.1 nmol/min-m^3) and cool (1.1 ± 0.1 nmol/min-m^3) periods. Moreover, we observed higher extrinsic oxidative potential of PM_{10} samples collected during dust events (~30% increase) compared to non-dust samples. Our PCA-MLR analysis identified soil and resuspended dust, secondary aerosol (SA), local industrial activities and petroleum refineries, and traffic emissions as the four sources contributing to the ambient PM_{10} oxidative potential in central Riyadh. Soil and resuspended dust were the major source contributing to the oxidative potential of ambient PM_{10}, accounting for 31% of the total oxidative potential. Secondary aerosols (SA) were the next important source of PM_{10} toxicity in the area as they contributed to about 20% of the PM_{10} oxidative potential. Results of this study revealed the major role of soil and resuspended road dust on PM_{10} toxicity and can be helpful in adopting targeted air quality policies to reduce the population exposure to PM_{10}.

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1. Introduction

A growing body of epidemiological and toxicological evidence indicates strong associations between exposure to ambient particulate matter (PM) and adverse effects on human health, including chronic obstructive pulmonary diseases, endothelial dysfunction, cardiovascular illnesses, adverse birth outcomes, and lung cancer (Consonni et al. 2018; Delfino et al. 2005; Du et al. 2016; Fasola et al. 2020; Hyun et al. 2021; Orellano et al. 2020; Sapkota et al. 2012; Vaduganathan et al. 2016). One of the underlying mechanisms for the toxicity of PM is the excessive cellular production of reactive oxygen species (ROS), leading to oxidative stress, inflammation, and consequently produce adverse health outcomes (Delfino et al. 2013; Ghio et al. 2012; Lodovici and Bigagli 2011). As a result, several studies have attempted to develop chemical and biological assays to quantify the airborne particle oxidative potential (Akhtar et al. 2010; Bates et al. 2019). A well-established and widely used assay is the dithiothreitol (DTT) assay, which measures the PM capability to catalyze the transfer of electrons from DTT to oxygen by creating superoxide radicals that can directly be linked to the PM oxidative potential (Borlaza et al. 2018; Chow et al. 2015; Fang et al. 2016).

Ambient PM is a complex mixture of different chemical species that have been associated with distinct health impacts (Amato et al. 2018; Sardar et al. 2005; Watson et al. 1994). Specific PM components including redox active metals (e.g., V, Mn, Ni, and Cu), carbonaceous species (e.g., elemental carbon (EC) and organic carbon (OC)), and polycyclic aromatic hydrocarbons (PAHs) have been consistently linked with the PM oxidative potential (Cheung et al. 2012; Chiara et al. 2018a, 2018b; Daher et al. 2014; Kleinman et al. 2005). These PM species originate from a variety of sources including soil and road dust emissions (Hsu et al. 2016; Jain et al. 2018), road traffic (Kavouras et al. 2001; Ryou et al. 2018; Sardar et al. 2005), biomass burning (Saggu and Mittal 2020; Stracquadanio et al. 2019; Vicente et al. 2021), and atmospheric photochemical processes forming secondary organic aerosols (SOA) (Jain et al. 2018; Rezaei et al. 2018; Ryou et al. 2018). Therefore, it is essential for policy makers to identify these sources and more importantly their contribution to the PM oxidative potential to target the PM emissions with greater toxicity more effectively.

Few studies have investigated the air quality deterioration and negative health endpoints associated with ambient PM in Riyadh, the capital of Saudi Arabia and one of the Middle East's largest metropolitan areas, with approximately 7.3 million residents (Alangari et al. 2015; Nasser et al. 2015; Wahabi et al. 2017). Previous studies in Riyadh have indicated that the ambient PM in the city was heavily impacted by regional natural dust as well as local activities (e.g., traffic and industrial emissions) (Alangari et al. 2015; Alharbi et al., 2015b; El-mubarak et al. 2012; Modaihsh et al. 2015). Alharbi et al. (2015a, 2015b) reported that the average ambient PM_{10} (PM with aerodynamic diameter $<10\,\mu m$) in the metropolitan area of Riyadh exceeded the national standard as well as other recorded PM_{10} values in the region, such as in Tehran, Beirut, Abu Dhabi, and Kuwait. They attributed this increase to the natural dust activities in the area during the summer and vehicular emissions and construction activities during the winter. To the best of our knowledge, no study has provided any insights on the contribution of the PM sources to the toxicological characteristics (i.e., oxidative potential) of PM in the city of Riyadh.

The receptor model is a useful tool that is widely used for identification of sources and quantifications of their contributions to a target variable. Among several receptor models are multivariate factor analysis models, including Principal Component Analysis/Multiple Linear Regression (PCA/MLR), UNMIX, and Positive Matrix Factorization (PMF) (Deng et al., 2018; Hopke et al., 2006; Shi et al., 2014; Wang et al., 2012). Researchers often combine the results of PCA factor analysis with the MLR method to identify the contribution of different resolved factors by PCA to a dependent variable (Chakraborty and Gupta, 2010; Harrison et al., 1996; Shi et al., 2009; Soleimanian et al., 2020; Srivastava et al., 2008; Taghvaee et al., 2019; Verma et al., 2012; Yu et al., 2010). As this model does not require information on source profiles, the source categories and their contributions can be identified according to the PM ambient dataset (Shi et al., 2014; Taghvaee et al., 2019; Zhang et al., 2019). For example, Taghvaee et al. (2019) using PCA/MLR model determined the most significant species contributing to the oxidative potential of $PM_{2.5}$ in Athens, Greece.

This study investigates the chemical and toxicological characterization of ambient PM_{10} during dust and non-dust events in a typical urban area of Riyadh. Ambient PM_{10} samples were collected during a cool period (December–March) and a warm period (May–August), covering dust and non-dust events in the area. Collected PM samples were analyzed for their chemical components, and the PM oxidative potential was determined using the DTT in vitro assay. The Principal Component Analysis (PCA) in combination with Multiple Linear Regression (MLR) were also used to link sources of ambient PM_{10} to the measured oxidative potential.

2. Methodology

2.1. Sampling location and collection period

Weekly time-integrated PM_{10} samples were collected at a residential city park in central Riyadh, Saudi Arabia (24°38′55″N, 46°43′16″E) between December 2019 and August 2020, using a medium volume sampler (model URG3000ABC, URG Corp, Chapel Hill, NC, USA) operating at a flow rate of 8 L/min. To achieve the required mass loading for further chemical analyses, the daily collected samples were composited every three (or four) days. Our selected site is located in a densely populated residential area in the city center of Riyadh, about 2 km away from a major highway (i.e., King Fahad Highway), and in a close proximity (500 m away) of some regular business and auto shops. The site also is about 4 km from the old industrial city. Previous studies in the area have indicated that central Riyadh has a poor air quality as a result of abundant vehicular, household, commercial and industrial activities (Alharbi et al., 2014, 2015a; Bian et al., 2016; El-Mubarak et al., 2014). Fig. 1 shows the map of the investigated area with the location of our sampling site. Additionally, considering that a large number of Riyadh population lives in/near the city center (Alharbi et al., 2015b; Saudi General Authority for Statistics, 2016), it can be argued that our sampling site properly represent the population exposure to major sources of air pollution in the Riyadh metropolitan area. The meteorological parameters (i.e., temperature and relative humidity (RH)) were also obtained from the Royal Riyadh Development Authority station at the same location of our sampling site during the investigated period. The seasonal average values of the abovementioned meteorological parameters during the study period are reported in Table S1. We should note that the sampling campaign coincided with seven dust events, and the filters collected during the time period of these events were investigated separately as will be discussed in Results and discussion section. The dust storm events were forecasted using the Saudi National Center for Meteorology Radar. Samples with average daily concentrations exceeding the 90th percentile were classified as dust samples (Achilleos et al., 2014; Rezaei et al., 2018).

2.2. Gravimetric and chemical analysis

PM_{10} samples on 47 mm quartz (Whatman company, 2.5-μm pore, Marlborough, MA) and teflon (Tisch Scientific, 1-μm pore, North Bend, OH) filters were collected (in parallel) during our sampling campaign. The mass concentration of PM samples was calculated by dividing the collected mass, measured by a microbalance (MT5, Mettler Toledo Inc., Columbus, OH), on filters to the volume of sampled air. The collected PM_{10} mass was determined as the difference between the pre-sampling and post-sampling weight of filters after equilibration under

Fig. 1. Map of the study location in the Riyadh metropolitan area.

stable laboratory conditions (i.e., temperature of 22–24 C and relative humidity of 40–50%).

In addition, the collected PM_{10} samples were evaluated for their content of EC, OC, metals and trace elements, and inorganic ions by the Wisconsin State Lab of Hygiene (WSLH). In summary, the thermo-optical transmittance (TOT) analysis by a model-4- semi-continuous OC/EC field analyzer (Sunset Laboratory Inc., USA) was used to measure the OC/EC content of the PM_{10} samples (Birch and Cary, 2007). Moreover, inductively coupled plasma mass spectroscopy (ICP-MS) analysis and ion chromatography (IC) were employed to measure the metal and trace element components and inorganic ions of PM_{10} samples, respectively (Herner et al., 2006; Karthikeyan and Balasubramanian, 2006).

2.3. Oxidative potential of PM_{10}

The dithiothreitol (DTT) assay was employed to assess the oxidative potential of the collected PM_{10}, as a well-established method in the literature to measure the oxidative potential of PM samples (Calas et al., 2018; Chirizzi et al., 2017; Hu et al., 2008; Molina et al., 2020). For this assay, the linear decay rate of dithiothreitol is used as an index of the oxidative potential of PM. Briefly, the filter-collected PM were stored frozen at −20 °C and then extracted with high-purity water (8.0 mL) with continuous shaking, in the dark, over a 16-h period. PM_{10} extracts were then directly incubated in potassium phosphate (KPO_4) buffer and DTT. The trichloroacetic acid was gradually added to vials of the incubation mixture for stopping the reaction, followed by recording the absorbance at 412 nm (optical density of 2-nitro-5-thiobenzoic acid) and 650 nm (reference wavelength) on an M5e plate reader (Molecular Devices, Sunnydale, CA). The DTT rate of depletion (per units of time) was then determined by converting the recorded absorbance to the remained DTT. Further information regarding the DTT methodology is available in the SI and (Shafer et al. 2016; Cho et al. 2005).

2.4. Source apportionment of the PM_{10} oxidative potential

In this study, the PCA analysis using the Statistical Package for Social Sciences (SPSS) version 25 was applied on the volumetric (i.e., per m^3 of

air) mass concentrations of OC, EC, sulfate, ammonium, and individual metals (e.g., Cu, Zn, Al, Ti and Fe) to identify and estimate the possible source factors that contribute to the PM_{10} mass concentration. In this method, the chemical data was first transformed into a dimensionless standardized form using the following equation:

$$Zij = \frac{C_{ij} - \overline{C}_j}{\sigma_j} \quad (1)$$

where Z_{ij} stands for the dimensionless standardized form of the ith sample and the jth species, C_{ij} is the mass concentration of species j in the ith sample, and $\overline{C}_j$ and σ_j refer to the mean mass concentration and the standard deviation for species j, respectively. The receptor model then mathematically solves the following chemical mass balance equation:

$$Z_{ij} = \sum_{k=1}^{p} g_{ik} h_{kj} \quad (1)$$

where P refers to the resolved factors by the PCA model and g_{ik} and h_{kj} indicate the factor loading and the factor score, respectively. It should be noted that a varimax orthogonal rotation was performed on the resolved factors in order to facilitate the interpretation (Abdi, 2003; Dallarosa et al., 2005). The resolved factors with high eigenvalues compared to the unity were considered to be a significant contributor. Additionally, the Kaiser-Meyer-Olkin (KMO) value was set to 0.5 and above to ensure the PCA procedure's suitability (Thomas et al., 2014). The multi-linear regression (MLR) was then employed between the PCA resolved factor scores (as independent variables) and the extrinsic DTT values (in units of nmol/min. m^3) as dependent variable (Baker, 2003; Zuo et al., 2007). The relative source contribution to the PM_{10} oxidative potential was determined based on the standardized regression coefficients (Beta) and derived R^2 value. In details, the relative source contributions to PM_{10} oxidative potential were calculated by normalizing the derived Beta values.

3. Results and discussion

3.1. PM_{10} mass concentration and chemical composition

3.1.1. PM_{10} mass and carbonaceous species

Fig. 2(a) and Table S2 show the average PM_{10} mass concentrations of collected samples during the warm and cool periods as well as during the dust events. According to the figure, higher concentrations (P-value = 0.01) were observed for PM_{10} in summer (98.7 ± 3.7 μg/m^3) compared to winter season (80.0 ± 6.0 μg/m^3), most likely due to the increase in particle concentration from soil and dust sources during the warm period. The dry atmospheric conditions (i.e., low relative humidity and high temperature (Table S1)) during the warm phase facilitate particle resuspension from the soil and desert areas in/around the city of Riyadh (Alharbi et al., 2015b; Alharbi, 2009; El-Mubarak et al., 2014). It is worth noting that the PM_{10} mass concentrations dramatically increased (up to 218.2 ± 34.8 μg/m^3) during dust events, considerably exceeding the recommended PM_{10} standard (50 μg/m^3) by World Health Organization (WHO) (Lodge, 1988).

The seasonal and dust event average concentrations of carbonaceous compounds including elemental (EC) and organic carbon (OC) are illustrated in Fig. 1(b–c). Based on Fig. 1(b), increased EC mass concentrations were observed in winter season (1.7 ± 0.4) compared to summer season (1.3 ± 0.2). The stable meteorological conditions (i.e., low mixing height) during the cool period limit the horizontal and vertical dispersion of air pollutants, including EC, increasing their concentrations to levels higher than those observed in the warmer season (Kim et al., 2015; Schwartz et al., 2018; Taghvaee et al., 2019). The EC levels significantly decreased (p value < 0.05) during dust events compared to normal days, which can be explained by considerably lower traffic activities, as the major source of EC, during these events in the area.

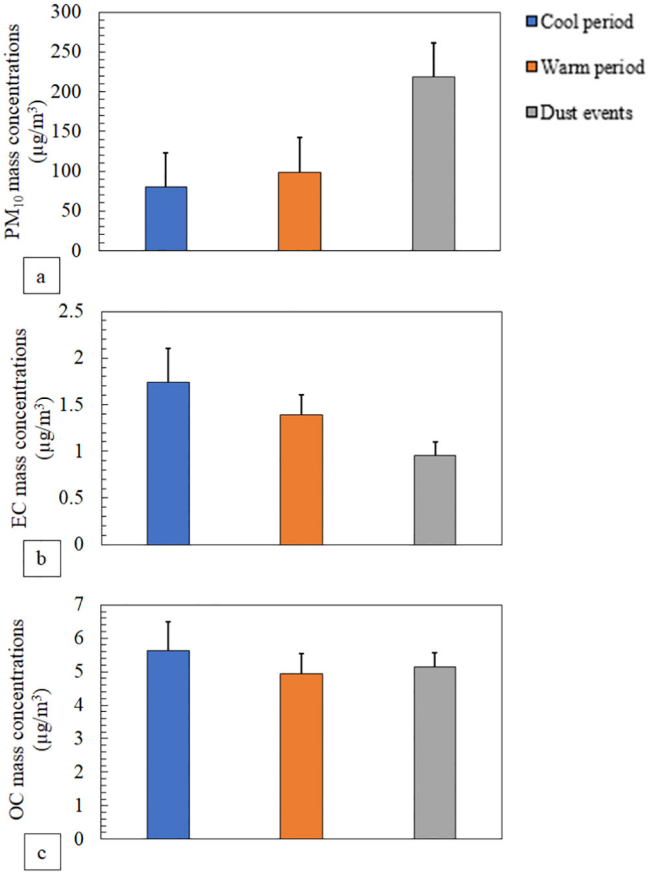

Fig. 2. The seasonal and dust event average concentrations of: a) PM_{10}; b) EC; and c) OC.

Examining the seasonal trend of OC concentrations revealed comparable concentrations (P-values = 0.30) in cool (5.6 ± 0.8 μg/m^3) and warm (4.9 ± 0.6 μg/m^3) period samples, with wintertime values being slightly higher. OC can be originated from both primary and secondary sources (Altuwayjiri et al., 2021; Gianini et al., 2013; Soleimanian et al., 2019a; Von Schneidemesser et al., 2010). The significant contribution of secondary organic aerosols (SOAs) to OC mass concentrations during warm period most likely counterbalances the higher contributions of primary sources (i.e., traffic and industrial activities) due to higher gas-to-particulate partitioning during cool period, leading to comparable OC levels in both periods of the sampling.

3.1.2. Inorganic ions

The average concentrations of sulfate, ammonium, and nitrate, by season, as well as during the dust period, are illustrated in Fig. 3. As shown in the figure, the mass concentration of sulfate was higher in the summer season compared to the cooler period, while the ammonium and nitrate levels were comparable (p value = 0.50 and 0.40, respectively) during these periods. Sulfate is formed as a result of the photochemical oxidation (through gas-phase reactions with the hydroxyl radical (OH)) of sulfur dioxide, emitted by combustion sources with sulfur in the fuel (Fine et al., 2008; Xue et al., 2016). With the higher temperatures during warm season, the degree of solar radiation is enhanced, causing the photochemical reactions to peak and increase the formation rate of sulfate (Na et al., 2004; Seinfeld and Pandis, 2006). Moreover, the inorganic ions (i.e., sulfate, ammonium, and nitrate) levels slightly increased during the dust episodes. This observation is consistent with the results from the previous study by Alharbi

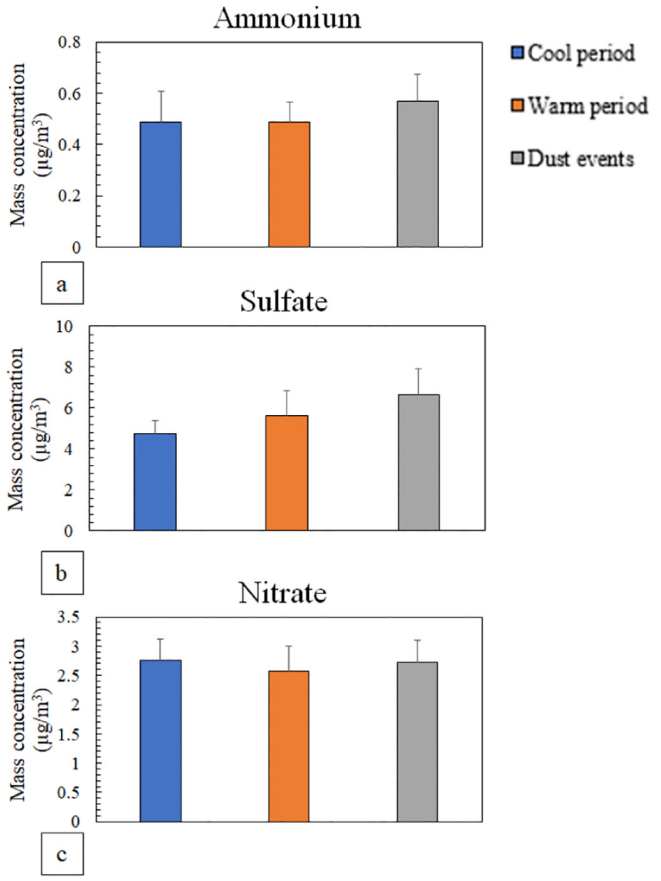

Fig. 3. The seasonal and dust event average concentrations of selected inorganic ions: a) ammonium; b) sulfate; and c) nitrate.

et al. (2015a, 2015b) at the same area, in which the authors reported higher levels of sulfate and ammonium during dust period compared with the normal periods. Increased levels of inorganic ions of secondary origin, such as ammonium nitrate and sulfate, were also observed in previous studies on dust episodes around the globe (Ghosh et al., 2014; Hassan and Khoder, 2017; Javed and Guo, 2021; Naimabadi et al., 2016; Saliba et al., 2014; Stone et al., 2011). For example, a recent study by Javed and Guo (2021) investigated the impact of dust episodes on the chemical characterization of fine and coarse PM in Doha, Qatar, and reported a significant increase in the concentrations of PM chemical components, including inorganic ions, during dust episodes. Additionally, Stone et al. (2011) reported high sulfate enrichment (by a factor of ~2.5) in PM dust samples collected in Gosan, Korea, as opposed to non-dust samples.

3.1.3. Metals and trace elements

Fig. 4 illustrates the concentrations of selected metals (i.e., Al, Ti, Ba, Li, Pb, Fe, Zn, Cu, Ca, Ni, Cr, and K) in PM_{10} samples collected in warm, cool and dust periods. Previous studies have indicated that these metals can be originated from various sources such as soil and road dust, tire and brake wear, and industrial emissions (Almeida et al., 2005, 2006; Harrison et al., 2012; Tian et al., 2016). Overall, higher mass concentrations of redox-active metals, including Al, K, Ti, and Fe as chemical markers of soil and resuspended dust emissions (Almeida et al., 2005; Cardoso et al., 2018), were observed during warm period as well as during dust events compared to the cooler period. This is due mainly to the drier atmospheric conditions (i.e., the lower relative humidity prevailing during these periods) that facilitate the resuspension of soil and desert dust particles (Laidlaw and Filippelli, 2008; Taghvaee et al., 2019). For example, the average levels of Al in dust samples were 7334.8 ± 2214 ng/m^3 which are higher than the observed levels in summer (5404 ± 2500 ng/m^3) and winter (2363 ± 1416 ng/m^3) seasons. Querol et al. (2019) reported that the emissions from large industries (including petrochemical, petroleum, and power plants) located nearby several desert areas globally interact with transported dust particles in the affected region and result in notable increases in the PM metal and element concentrations during dust episodes. Additionally, lower atmospheric boundary layers typically prevailing during these events enhance the accumulation of anthropogenic pollutants, including redox-active metals and elements, during dust storm episodes (Pandol et al., 2014; Querol et al., 2019). Furthermore, lower concentrations were observed for Cu, Zn and Pb, which are tracers of non-tailpipe emissions (e.g., asphalt, brake abrasion and tire wear emissions) (Farahani et al., 2021; Harrison et al., 2012; Soleimanian et al., 2019b; Tecer et al., 2012) during the warm phase of the sampling campaign. The seasonal trends for non-tailpipe tracers in our study are in agreement with the literature (Alharbi et al., 2015b; Galindo et al., 2018; Pekey et al., 2010). Alharbi et al. (2015a, 2015b) investigated the chemical characteristics of PM_{10} in Riyadh and reported similarly higher mass concentrations of non-tailpipe tracers, including Cu and Mo, during the cool period compared to the warm period.

3.2. Oxidative potential of PM_{10}

Fig. 5 shows the extrinsic (per m^3 of air volume) and intrinsic (per PM mass) levels of PM_{10} oxidative potential during the investigated periods. The detailed values during warm, cool, and dust events are also presented in Table S2. Our measurements revealed comparable volumetric oxidative potential (P-value = 0.30) during the warm

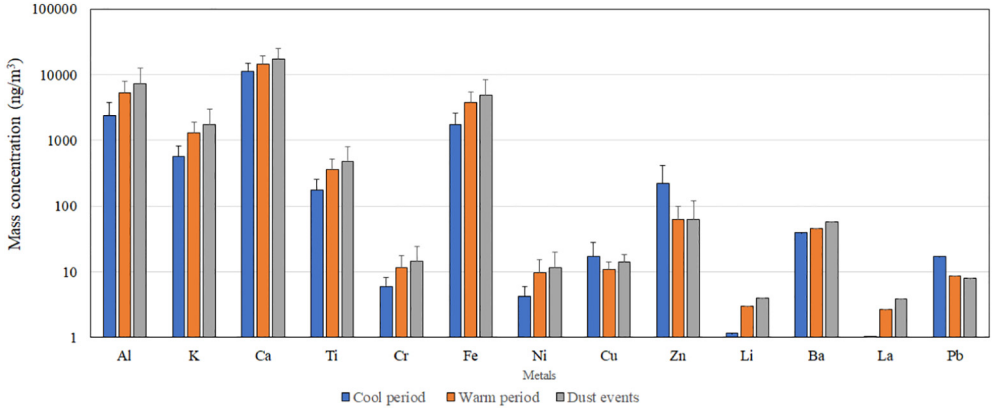

Fig. 4. Average concentrations of metals and trace elements during the investigated periods.

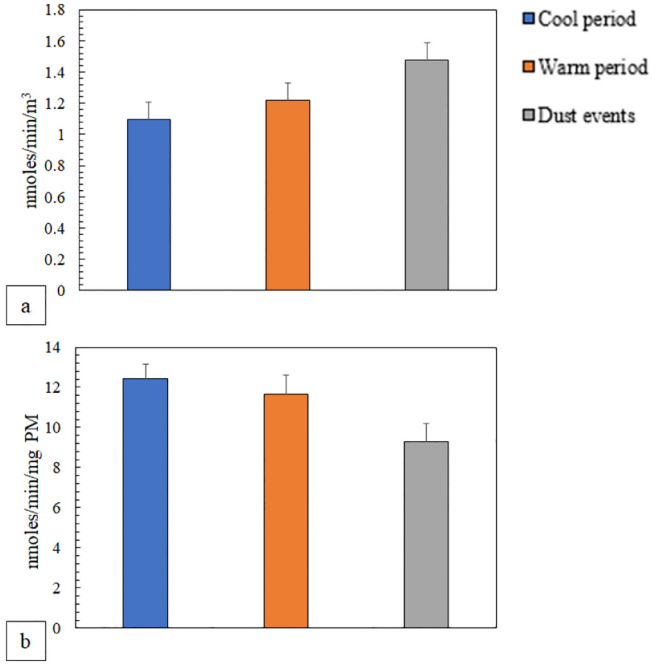

Fig. 5. PM_{10} oxidative potential for cool and warm periods and dust events: a) volume-based, or extrinsic oxidative potential (per m^3 of air); b) mass-normalized, or intrinsic oxidative potential (per PM mass).

(1.2 ± 0.10 nmol/min-m^3) and cool (1.1 ± 0.1 nmol/min-m^3) periods (Fig. 4(a)). The measured summer and winter-time DTT consumption rate is almost within the range of previously reported values in Tehran (1.35 ± 0.37 nmol/min-m^3) (Rezaei et al., 2018), and considerably higher than those reported in Los Angeles (0.35 ± 0.04 nmol/min-m^3) (Shirmohammadi et al., 2016), Atlanta (0.30 ± 0.10 nmol/min-m^3) (Verma et al., 2014) and Athens (0.33 ± 0.20 nmol/min-m^3) (Paraskevopoulou et al., 2019). It is worth noticing that these DTT values are somewhat lower but statistically significant (P-value < 0.05) in comparison to values recorded during dust episodes (1.5 ± 0.2 nmol/min-m^3). In line with our results, Lovett et al. (2018) evaluated the oxidative potential of PM in Beirut during Saharan and Arabian dust events, and revealed an increase in the coarse PM oxidative potential (in units of μg Zymosan/m^3 of air) in dust samples compared to the samples collected during the non-dust events. Interestingly, the investigation of the mass-based oxidative potential of the collected ambient PM_{10} samples revealed that the intrinsic levels of PM_{10} oxidative potential were higher in the cool (12.4 ± 0.7 nmol/min/mgPM) period compared to warm (11.7 ± 1.0 nmol/min/mgPM) and dust (9.3 ± 0.9 nmol/min/mgPM) periods (P-values = 0.27 and 0.01, respectively). Chirizzi et al. (2017a) examined the influence of Saharan dust outbreaks on the oxidative potential of water-soluble fractions of PM_{10} and reported that dust transported from Africa to Lecce has a lower mass normalized DTT in comparison to the average values observed in regular samples. This suggests that the observed higher extrinsic PM_{10} oxidative potential during dust storm events is due to the much higher overall PM mass concentrations, however the predominant PM components during these events may not be as redox-active as species during a regular period. A regression analysis was conducted to distinguish the association of chemical species, such as OC, EC, inorganic ions and water-soluble metals, with the DTT activity (as presented in Table S3). According to our regression analysis results, most notable correlations ($R > 0.70$) were observed between DTT activity and redox active metals in the area. A number of previous studies have also reported correlations between transition metals and DTT activity (Ntziachristos et al., 2007; Verma et al., 2009a, 2009b). These redox active transition metals (e.g., Al, Ti, Cr, Cu and La) in the PM_{10} size range originate from various sources in the area including resuspended dust and soil, vehicular emissions, and local industrial activities. More discussion related to the sources contributing to the PM_{10} toxicity is presented in Section 3.3 of the manuscript.

3.3. Source apportionment of ambient PM_{10} and its associated oxidative potential

3.3.1. Source apportionment of PM_{10} mass concentration using the PCA approach

Table 1 presents the outputs of PCA analysis performed on the weekly time-integrated EC, OC, as well as trace elements and metals concentrations for the whole study period, which resulted in the identification of four factors explaining approximately 91% of total variance in the data. The first factor was identified as soil and resuspended dust emissions due to significant loadings of Fe, Al, K, Li, and Ti as crustal elements. Previous studies have documented that Fe, Al, K, Li and Ti are all well-established chemical markers of soil and resuspended dust emissions in different areas around the globe(Almeida et al., 2005; Karanasiou et al., 2012; Tian et al., 2016). This factor has a significant contribution to total PM_{10} concentrations, accounting for about 40% of total PM_{10} in the city. Although high traffic activities in the area could lead to increase in concentrations of the redox-active metals (e.g., Ti, Al and K), other mechanisms including the dust resuspension can majorly contribute to these emissions in the urban areas. Riyadh located close to Ad-Dhna and Rub'al Khali deserts experiences dry and hot climate along with high wind speeds that facilitate the transport and resuspension of the dust metals to the area (Alharbi et al., 2013; Badarinath et al., 2010; Farahat, 2016; Maghrabi et al., 2011; Modaihsh et al., 2017; Smirnov et al., 2002). Modaihsh et al. (2017) reported that the average annual dust deposition in Riyadh was about 454 tons/km^2 and is significantly higher than the surrounding regional and worldwide areas. Previous studies by Farahani and Arhami (2020) and Givehchi et al. (2013) also highlighted the significant contribution of dust emissions to PM_{10} levels in the Middle Eastern region.

The second factor was characterized by high loadings of EC, Cu and Zn. EC is predominantly emitted from vehicular exhausts and undergoes very limited chemical transformations (Jain et al., 2018). Numerous studies have documented EC as the major tracer of tailpipe emissions (Díaz-Robles et al., 2008; Jain et al., 2018; Yin et al., 2010). Additionally, loading of Cu and Zn in this factor can be attributed to asphalt, brake abrasion and tire wear emissions (Cao et al., 2006; Querol et al., 2008; Srimuruganandam and Shiva Nagendra, 2012). Similar to our study, Jain et al. (2018) employed EC, Cu and Zn as the chemical tracers to

Table 1
Loadings of chemical species in the factors resolved by the principal component analysis (PCA). Loadings >0.7 are bolded.

Species	Soil and resuspended dust emissions	Traffic emissions	Secondary aerosols (SA)	Local industrial activities and petroleum refineries
Ti	**0.979**	−0.019	0.152	0.109
Fe	**0.972**	−0.022	0.154	0.145
Al	**0.971**	−0.111	0.131	0.129
K	**0.965**	−0.030	0.179	0.163
Li	**0.965**	−0.071	0.188	0.126
Cu	−0.220	**0.877**	0.004	−0.095
Zn	−0.342	**0.841**	−0.031	−0.121
EC	0.328	**0.743**	0.256	0.257
OC	0.240	0.636	0.419	0.364
Sulfate	0.096	0.190	**0.911**	0.247
Ammonium	0.310	0.053	**0.903**	0.076
Se	0.105	0.058	0.109	**0.941**
La	0.405	−0.059	0.464	**0.715**
% of variance	40.919	19.230	17.281	13.676
Cumulative %	40.919	60.149	77.430	91.106

resolve the vehicle emissions factor in Delhi, India. Therefore, we labeled this factor as "Traffic emissions". This factor has a moderate contribution (~20%) to total PM_{10} concentrations in the metropolitan area of Riyadh, which is in good agreement with the findings of previous studies in the region (Javed and Guo, 2021; Khodeir et al., 2012; Soleimani and Amini, 2014).

The third factor showed very high levels of sulfate (SO_4^{2-}), and ammonium (NH_4^+) and contributed to approximately 17% of the total PM_{10} concentrations in Riyadh. SO_4^{2-} and NH_4^+ are the constituents of ammonium sulfate and ammonium nitrate produced by gas phase reactions of acidic gaseous precursors (i.e., HNO_3 and H_2SO_4) with ammonia (NH_3). Numerous studies used these species (i.e., SO_4^{2-} and NH_4^+) as indicators of secondary aerosol formations (Jain et al., 2020; Sricharoenvech et al., 2020). Consequently, we selected "secondary aerosol (SA)" as the most suitable title for this factor.

The fourth factor indicated high loadings of lanthanoid (La), and selenium (Se) and contributed to about 14% of total PM_{10} concentrations (Table 1). Previous studies reported Se as a heavy metal used in the electronics, plastic, glass, and paints industry (Risher et al., 1999; Taghvaee et al., 2018). Moreover, loadings of La in this factor can be attributed to local oil-industry emissions (Kulkarni et al., 2006; Moreno et al., 2008). Moreno et al. (2008) used La as a marker to identify the contribution of oil refinery emissions to PM in Puertollano, Spain. Therefore, we believe the most suitable label for this factor is "local industrial activities and petroleum refineries". In line with our results, Bian et al. (2016) showed low to medium contributions of industrial activities including refineries to the average concentration of PM_{10} species (i.e., PAHs) near our sampling site.

3.3.2. Source apportionment of PM_{10} oxidative potential using MLR approach

The MLR analysis was performed based on the PCA resolved factor scores to identify the most significant sources accountable for the PM-induced toxicity (Table 2). As can be seen in the table, "soil and resuspended dust emissions" was the most important source contributing to PM oxidative potential (Beta = 0.65), accounting for 31% of the oxidative potential (Fig. 6). In addition, "SA" and "local industrial activities and petroleum refineries" contributed to 20% (Beta = 0.42) and 19% (Beta = 0.40) of the PM oxidative potential, respectively. Romano et al. (2020) reported significant correlations ($P_{value} < 0.001$) between the oxidative potential of PM_{10} and tracers of SA (e.g., ammonium), further corroborating the findings of this study. Traffic emissions are also no less important in the area as they contribute to about 17% (Beta = 0.35) of PM-induced toxicity. Previous studies also underscored the role of this factor to the overall toxicity of PM in various urban areas (Hu et al., 2008; Pant et al., 2015; Shirmohammadi et al., 2015, 2016; Wang et al., 2020; Weber et al., 2021). For example, Weber et al. (2021) indicated road traffic as a major source contributing to the extrinsic PM_{10} oxidative potential (median 0.36 nmol/min-m^3) across different cities in France.

Table 2
Results of the multiple linear regression (MLR) analysis between PM_{10} oxidative potential (as the dependent variable) and PCA resolved factor scores (as independent variables).

Factors	Unstandardized coefficients (±Std. error)	Standardized coefficients (Beta)	P value	R^2
Constant	1.22 ± 0.03		0.000	0.88
Soil and resuspended dust emissions	0.29 ± 0.04	0.65	0.000	
Traffic emissions	0.15 ± 0.04	0.35	0.002	
Secondary aerosols (SA)	0.18 ± 0.04	0.42	0.000	
Local industrial activities and petroleum refineries	0.18 ± 0.04	0.40	0.000	

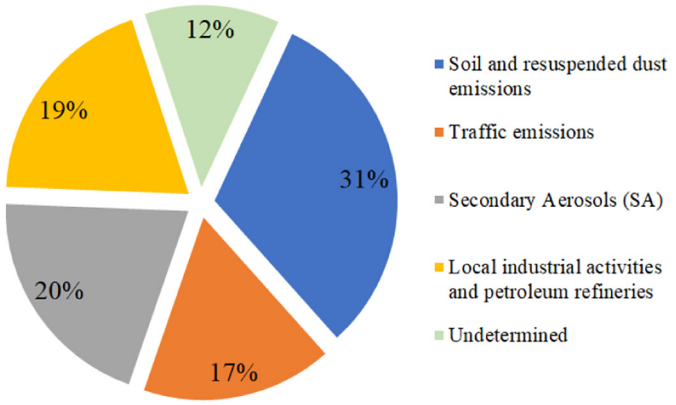

Fig. 6. Relative source contributions to PM_{10} oxidative potential.

4. Summary and conclusions

The main goal of this study was to determine and evaluate the sources of PM_{10} toxicity in the metropolitan area of Riyadh, which is one of the most populous arid areas in the world. Our findings revealed higher PM_{10} mass concentrations in the warm season (98.7 ± 3.7 μg/m^3) compared to the cooler season (80.0 ± 6.0 μg/m^3), with enormous PM_{10} concentrations (as much as 218.2 ± 34.8 μg/m^3) during dust outbreaks. Moreover, most of the redox active metals (e.g., Fe, Al, K, Li and Ti) and inorganic ions (sulfate and ammonium) increased from the cooler to the warm period. We also observed an increase in the DTT levels from the cool period (1.00 ± 0.10 nmol/min-m^3) to warm period (1.20 ± 0.10 nmol/min-m^3) and dust episodes (1.50 ± 0.20 nmol/min-m3). Our statistical analysis (PCA coupled with MLR) indicated soil and resuspended dust emissions, secondary aerosols, local industrial activities and petroleum refineries, and traffic emissions were the four sources of ambient PM-induced toxicity in Riyadh, with corresponding contributions of 31%, 20%, 19%, and 17%, respectively. Our results underscore the significant role of soil and resuspended dust emissions to PM_{10} toxicity in the Riyadh metropolitan area. Therefore, we recommend the application of mitigation strategies, including water sprinkling, tree planting, street cleaning, and dust suppressants, which can effectively reduce the resuspension of dust from loose soil open surfaces in the city of Riyadh and the surrounding regions.

CRediT authorship contribution statement

Abdulmalik Altuwayjiri: Conceptualization, Methodology, Data curation, Writing – original draft. **Milad Pirhadi:** Conceptualization, Methodology, Validation, Visualization, Writing – review & editing. **Mohammed Kalafy:** Data curation, Methodology, Resources, Writing – review & editing. **Badr Alharbi:** Methodology, Data curation, Resources, Writing – review & editing. **Constantinos Sioutas:** Conceptualization, Funding acquisition, Project administration, Resources, Supervision, Writing – review & editing.

Declaration of competing interest

The authors of this paper declare that there is no conflict of interest. I am signing this letter on behalf of the other co-authors of 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.scitotenv.2021.150590.

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