$PM_{2.5}$ chemical composition and geographical origin of air masses in Cape Town, South Africa

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Abstract

$PM_{2.5}$ in the indoor and outdoor environment has been linked in epidemiology studies to the symptoms, hospital admissions and development of numerous health outcomes including death. The study was conducted during April 2017 and April 2018. $PM_{2.5}$ samples were collected over 24 h and every third day. The mean $PM_{2.5}$ level was 13.4 μg m^{-3} (range: 1.17–39.1 μg m^{-3}). $PM_{2.5}$ levels exceeded the daily World Health Organization air quality guideline (25 μg m^{-3}) on 14 occasions. The mean soot level was 1.38 $m^{-1} \times 10^{-5}$ (range: 0 to 5.38 $m^{-1} \times 10^{-5}$). Cl^-, NO_3^-, SO_4^{2-}, Al, Ca, Fe, Mg, Na and Zn were detected in the $PM_{2.5}$ samples. The geographical origin of air masses that passed Cape Town was estimated using the Hybrid Single Particle Lagrangian Integrated Trajectory software. Four air masses were identified in the cluster analysis: Atlantic-Ocean-WSW, Atlantic-Ocean-SW, Atlantic-Ocean-SSW and Indian-Ocean. The population of Cape Town may experience various health outcomes from the outdoor exposure to $PM_{2.5}$ and the chemical composition of $PM_{2.5}$.

Keywords: $PM_{2.5}$; Soot; Health effects; Chemical composition; HYSPLIT; South Africa

Introduction

Various indoor and outdoor air pollutants are linked in epidemiology studies to the symptoms, hospital admissions and development of numerous health outcomes such as asthma (Fan et al. 2016), cardiovascular disease (Cesaroni et al. 2014; Wang et al. 2015; Münzel et al. 2018), skin diseases (Balmes 2019), birth outcomes (Li et al. 2019), sperm quality (Lafuente et al. 2016), type 1 and 2 diabetes (Ritz et al. 2019; Howard 2019), lung cancer (Lipfert and Wyzga 2019, Hamra et al. 2014)—even with the spread and increase of Covid-19's morbidity and mortality (Bilal et al. 2020; Comunian et al. 2020; Rodríguez-Urrego and Rodríguez-Urrego 2020) and other disease mortality (Fajersztajn et al. 2017; Liu et al. 2019; Orellano et al. 2020). Most of these epidemiology studies were conducted in the developed world and very few in Africa (Ostro et al. 2018; Rodríguez-Urrego and Rodríguez-Urrego 2020, Orellano et al. 2020; Liu et al. 2019; Ofori et al. 2020, Katoto et al. 2019; Wichmann and Voyi 2012; Coker and Kizito 2018; Lokotola et al. 2020).

An epidemiology study in Cape Town, South Africa, reported that exposure to outdoor particulate matter smaller or equal to 10 μm in aerodynamic diameter (PM_{10}), nitrogen dioxide (NO_2), sulphur dioxide (SO_2) levels in Cape Town during 2001–2006 posed a much

higher risk to die from cardiovascular and respiratory diseases than reported in developed countries, even though the outdoor levels were on average similar to those in European cities (Wichmann and Voyi 2012).

Another study in Cape Town observed that a 10 μg m^{-3} increase in PM_{10}, NO_2, SO_2 led to increases in 2.4% (1.3%, 3.5%), 5.1% (3.0%, 7.2%) and 5.1% (3.0%, 7. 2%) in cardiovascular disease hospital admissions during 2011–2016, respectively (Lokotola et al. 2020). These adverse effects were stronger on days warmer than 20.3 °C.

Wichmann and Voyi (2012) and Lokotola et al. (2020) did not investigate particulate matter smaller or equal to 2.5 μm in aerodynamic diameter ($PM_{2.5}$) as this air pollutant is not currently monitored in Cape Town. The daily and yearly $PM_{2.5}$ South African national ambient air quality standard came into effect on June 29, 2012 (Department of Environmental Affairs 2012). It is widely accepted that $PM_{2.5}$ is more hazardous to human health than PM_{10}, as it can penetrate deeper into the respiratory tract than PM_{10}, penetrate the lung barrier and enter the blood system (World Health Organization 2013).

Very few studies in Africa reported on $PM_{2.5}$ levels (Maenhaut et al. 1996; Petkova et al. 2013; Gaita et al. 2014; Snider et al. 2016; Fayiga et al. 2018; Tshehla and Djolov 2018; Kalisa et al. 2019; deSouza 2020). No study ever reported on $PM_{2.5}$ levels in Cape Town nor investigated its seasonal and weekly variation, chemical composition and possible source region contributions by using backward trajectory cluster analysis. The objectives of this study were to address these research gaps.

Materials and methods

Study setting

The sampling site was at an urban background, located on the roof of a house in the suburb of Kraaifontein, Cape Town (3 m above ground level, 27 km ENE of the city centre and 1 km from a busy freeway) (Fig. 1 and Fig. S1). This site was a favourable choice for sampling due to the assurance of safety and a continuous supply of electricity. The University of the Western Cape campus was not selected as sampling site due to its close proximity to Cape Town International Airport.

Cape Town occupies the south-western most point of the Western Cape Province. Topographically, the city is situated 10–150 m above sea level and has many peaks exceeding 300 m, the most well-known being Table Mountain (1000–1100 m above sea level).

Cape Town has a Mediterranean-style climate that is influenced by both the warm Agulhas current and the cold Benguela current (Western Cape Air Quality Management Plan 2016; Beck et al. 2018; World Meteorological Organization 2020). It is the second-most populous city in South Africa; 3.7 million residents during the latest census of 2011 (Western Cape Air Quality Management Plan 2016).

Fig. 1. Study sampling site and ambient air quality monitoring stations used for correlation analyses. Somerset West (1), City Hall (2), Goodwood (3), Wallacedene (4), Tableview (5), Kraaifontein study sampling site (6)

$PM_{2.5}$ sampling, gravimetric and chemical analyses

24-h ambient $PM_{2.5}$ filter samples were collected manually every third day for a period of 1 year (April 18, 2017, to April 16, 2018) using GilAir-5 personal air samplers (Sensidyne, Schauenburg Electronic Technologies Group, Mulheim-Ruhr, Germany) on 37 mm PTFE membrane filters (Zefon International, Florida, USA). The flow rate was 4 l/min, similar to other studies (Gaita et al. 2014; Novela et al. 2019; Adeyemi 2020). Sampling started 9 a.m. and ended 9 a.m. the next day. The start and stop times were selected for practical reasons as

starting and stopping a sample at midnight was not practical. $PM_{2.5}$ was sampled on the same days and times in Pretoria and Thohoyandou, South Africa (Novela 2019; Adeyemi 2020). A large multi-country study started and ended sampling at 9 a.m. to reduce loss of semi-volatile components (Snider et al. 2016). Snider et al. (2016) described that the loss rates of NH_4NO_3 during passive air flow were less than during active air flow. The sampling protocol of Snider et al. (2016) was designed to actively sample for one diurnal cycle and to avoid daytime sampling after collecting night time $PM_{2.5}$.

Other studies used pumps that were similar to the GilAir-5 for ambient air monitoring (Novela 2019; Adeyemi 2020; Mwase 2020). Mwase (2020) indicated that there was a good correlation between $PM_{2.5}$ levels measured on filter samples using a GilAir-5 pump and those obtained using a continuous real-time instrument in Pretoria, South Africa during May 2018 to May 2019 (Fig. S2).

In total, 146 24-h $PM_{2.5}$ filter samples (including 25 duplicate samples) were collected on 121 days during the 1-year study period. In addition to the 24-h $PM_{2.5}$ filter samples, four composite samples (two weekdays and two weekend days) were collected over four consecutive weeks (9 am to 9 am at intervals of 7 days) during September 2017 and January 2018 to determine the anion and elemental composition of $PM_{2.5}$. Total exposure time for each composite sample was 96 h.

The 24-h $PM_{2.5}$ filter samples were weighed at the School of Health Systems and Public Health (SHSPH), University of Pretoria (UP) in batches of 20 before and after sampling. An ultra-micro-balance (Mettler-Toledo XP6) was used under climate-controlled conditions (temperature: 20.1–22.0 °C, relative humidity: 43–54%) (Fig. S3). These batches were hand delivered and kept dry from direct sunlight and at room temperature during transit.

The 96-h $PM_{2.5}$ filter samples were weighed at the Department of Chemical Sciences, University of the Western Cape (UWC) with a microbalance (Mettler-Toledo ML 204) before and after sampling.

The 24-h and 96-h $PM_{2.5}$ filter samples were stored in a refrigerator at 4 °C. After gravimetric analysis, reflectance measurements were performed using an EEL43 reflectometer (Diffusion Systems Ltd. EEL model 43 D) at the SHSPH, UP. Measuring light absorption or reflectance of PM collected on filters is an alternative method to determine elemental carbon, a marker for particles produced by incomplete combustion; also referred to as soot (RUPIOH 2002).

The results were transformed into absorption coefficient (hereafter 'soot'). The following equation was used, as done in other studies (RUPIOH 2002; Cesaroni et al. 2014):

$$a = (A/2V) \times \ln(R_0/R_f) \quad (1)$$

where a is the absorption coefficient ($m^{-1} \times 10^{-5}$), V the sampled volume (m^3), R_0 the reflection of a primary control filter (%), R_f the reflection sampled filter (%) and A the loaded filter area (m^2). The average reflectance of the primary control filter was 100.075 (SD 0.1).

Soot levels ($m^{-1} \times 10^{-5}$) of the 96-h $PM_{2.5}$ filter samples were converted to equivalent black carbon (eBC) levels ($\mu g\ m^{-3}$) in order to calculate the percentage mass in comparison with $PM_{2.5}$ mass. The following equation was used, as done in other studies (Davy et al. 2017):

$$\mathrm{eBC}\ \left(\mu g\ m^{-3}\right) = \frac{A \times 10^6}{2 \times V \times \sigma_{ATN}} \times \ln\left(\frac{R_0}{R_s}\right) \times \left[1 + k \times \ln\left(\frac{R_0}{R_s}\right)\right] \quad (2)$$

where A is the loaded filter area (8.55×10^{-4} m^2), V the sampled volume (m^3), σ_{ATN} is black carbon extinction coefficient (19.5 m^2 g^{-1} for PTFE filters), k is the loading correction factor (0.3 for PTFE filters) and R is reflectance.

Elemental levels of the 96-h $PM_{2.5}$ samples were determined at UWC using a Varian 710-ES Inductively Coupled Plasma-Optical Emission Spectrometer (Supplementary text S1). Al, Ca, Fe, Mg, Na and Zn levels were above the LOD (0.001 μg m^{-3}), whilst As, Be, Cd, Cr, Co, Cu, Pb and Li levels were below the LOD.

Anion levels of the 96-h $PM_{2.5}$ filter samples were determined using a Dionex ICS-1600 Ion Chromatograph at UWC (Supplementary text S2). Bromide, fluoride, nitrite and phosphate levels were below the LOD. Chloride (Cl^-), nitrate (NO_3^-) and sulphate (SO_4^{2-}) were detected in the four samples and had LOD of 0.4 μg m^{-3}, 0.1 μg m^{-3} and 0.2 μg m^{-3}, respectively.

Geographical origin of air masses

The geographical origin of air masses that passed Cape Town was applied as surrogates for long-range transported air pollution from distant sources and its composition, as done in other studies (Kim et al. 2014; Saikat et al. 2015; Schwarz et al. 2016; Molnár et al. 2017; Tshehla and Djolov 2018; Novela 2019; Adeyemi 2020).

For each day in the 1-year study period, 72-h backward trajectories were produced using the Hybrid Single Particle Lagrangian Integrated Trajectory software (HYSPLIT) (the latest version, July 2018, was downloaded at www.ready. noaa.gov/HYSPLIT.php).

The HYSPLIT software downloaded and executed by the NCEP/NCAR (National Centers for Environmental Prediction/National Centre for Atmospheric Research) Global Reanalysis Meteorological Data at the web server of the National Oceanic and Atmospheric Administration Air Resources Laboratory (NOAA ARL). An analysis field (resolution 2.5° × 2.5° and 17 vertical levels) was provided every 6 h (0:00, 6:00, 12:00, 18:00) for a 72-h backward trajectory and the wind field was interpolated linearly between each analysis, as done in other studies (Wichmann et al. 2014; Molnár et al. 2017; Novela 2019; Adeyemi 2020).

Since a single backward trajectory has a large uncertainty and is of limited significance, an ensemble of trajectories with 500 m starting height and a fixed offset grid factor of 250 m was used in this study (i.e. 250 m and 750 m also used) as done in other studies (Wichmann et al. 2014; Molnár et al. 2017, Novela 2019, Adeyemi 2020).

A total of 4380 72-h backward trajectories were generated and applied in the cluster analysis with the HYSPLIT software. Four geographical origins of air masses were identified in the cluster analysis (Fig. 2, Fig. S4 and Fig. S5): Atlantic-Ocean-WSW, Indian-Ocean, Atlantic-Ocean-SW and Atlantic-Ocean-SSW. Each day in the 1-year study period was allocated to one of these geographical origins of air masses as done in other studies (Wichmann et al. 2014; Molnár et al. 2017, Novela 2019, Adeyemi 2020).

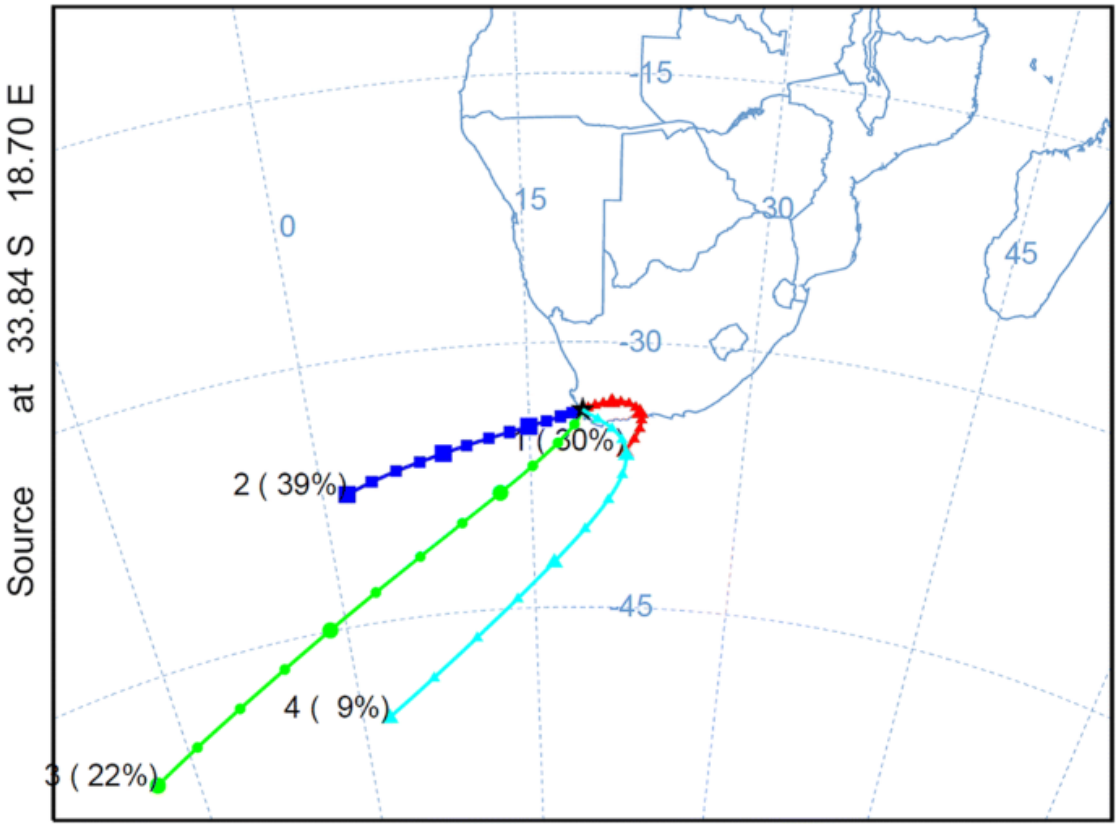

Fig. 2. Four geographical origins of air masses during April 2017 to April 2018 in Cape Town, South Africa

Other air pollution data and meteorological data

Additionally, existing air pollution data were obtained from the City of Cape Town after the $PM_{2.5}$ sampling campaign had ended. The hourly data included daily levels of PM_{10}, NO_2, SO_2 and ground-level ozone (O_3) measured at six ambient air quality monitoring (AAQM) stations during the 1-year study period. The National Environmental Management: Air Quality Act (NEMAQA) (Act 39 of 2004) requires the monitoring of criteria air pollutants in South Africa (Department of Environmental Affairs 2005). The NEMAQA currently enforces NAAQS for $PM_{2.5}$, PM_{10}, NO_2, SO_2, carbon monoxide, ground-level O_3, lead and benzene (Department of Environmental Affairs 2005, 2012). However, as mentioned before, $PM_{2.5}$ is not currently measured at any AAQM station in the City of Cape Town.

Figure 1 indicates the location of the six AAQM stations in the city (Supplementary text S3 and S4; Table S7). The AAQM stations at Wallacedene, Goodwood, Tableview, City Hall and Somerset-West are within a 30 km radius of the $PM_{2.5}$ sampling site in Kraaifontein. The PM_{10}, NO_2, SO_2, O_3 levels measured at these five AAQM stations were used to indicate potential local sources of ambient PM within a 30 km radius of the $PM_{2.5}$ sampling site. The Atlantis AAQM station is situated 37 km from the $PM_{2.5}$ sampling site and 100 km from the town of Saldanha, which has ore refineries. The Atlantis AAQM station was used to indicate long-range transported $PM_{2.5}$ that arrived at the $PM_{2.5}$ sampling site.

These six AAQM stations continuously assess real-time levels of the criteria air pollutants using equivalent methods of the United States Environmental Protection Agency and in

accordance with ISO 17025 guidelines (Department of Environmental Affairs 2005; Western Cape Air Quality Management Plan 2016).

Hourly temperature (°C), relative humidity (%), wind speed (m s^{-1}) and precipitation (mm) data were obtained from the South African Weather Service for the 1-year study period.

The daily averages of PM_{10}, NO_2, SO_2, ground-level O_3 and meteorological variables were calculated from 9 a.m. to 9 a.m. on days when $PM_{2.5}$ was sampled. The daily averages were based on at least 18 hourly values. If more than six hourly values were missing during the 24-h period, then the daily average was set as missing.

Statistical analyses

Statistical analyses were performed with SAS version 9.3. Descriptive statistics were reported for the 121 observations of the 24-h $PM_{2.5}$, soot, PM_{10}, NO_2, SO_2, O_3 and meteorological variables as well as the four composite samples (96-h anion and elemental composition of $PM_{2.5}$).

According to the Shapiro-Wilk's test, in general, the 121 observations of 24-h air pollution and meteorological variables did not have normal Gaussian distributions and non-parametric tests were applied. Spearman rank-ordered correlation analyses were applied to investigate the correlation between the 24-h air pollution and meteorological variables for the 1-year study period and by seasons.

Seasons were defined as: autumn (18/4/2017 to 31/5/2017 and 1/3/2018 to 16/4/2018), winter (1/6/2017 to 31/8/2017), spring (1/9/2017 to 30/11/2017) and summer (1/12/2017 to 28/2/2018).

Kruskal–Wallis tests were conducted to determine whether the 24-h median air pollution levels differed significantly between seasons and between the four geographical origins of air masses. Wilcoxon's rank-sum test was applied to test whether 24-h median air pollution levels differed significantly between weekdays and weekends.

Results and discussion

24-h $PM_{2.5}$, soot and meteorological conditions

Descriptive statistics are presented in Table 1. Time-series graphs of $PM_{2.5}$, soot and the meteorological conditions are presented in Fig. 3. The mean temperature was 17.7 °C and ranged from 9.2 to 25.3 °C. Wind speed ranged from 1.0 to 8.1 m s^{-1} and relative humidity 37.3 to 90.7%. Figure S6 indicates the frequency of wind speed by seasons and direction. There was a severe drought in Cape Town during the sampling period and the maximum precipitation recorded was 11.4 mm (Fig. S7).

Table 1. Descriptive statistics of 24-h $PM_{2.5}$, soot and meteorological conditions on 121 days during April 18, 2017, to April 16, 2018, in Kraaifontein, Cape Town, South Africa

Variable	*N*	Minimum	Mean	Median	Maximum	Std dev
All year						
$PM_{2.5}$ (μg m^{-3})	121	1.2	13.3	10.9	39.1	8.1
Soot ($m^{-1} \times 10^{-5}$)	121	0	1.368	0.937	5.390	1.233
Temperature (°C)	120	9.2	17.7	18.0	25.3	3.7
Relative humidity (%)	120	37.3	67.9	68.0	90.7	9.7
Wind speed (m s^{-1})	114	1.0	3.7	3.4	8.1	1.7
Precipitation (mm)	121	0.0	0.6	0.0	11.4	1.9
Autumn						
$PM_{2.5}$ (μg m^{-3})	31	1.9	11.0	9.8	21.7	5.2
Soot ($m^{-1} \times 10^{-5}$)	31	0.200	1.685	1.493	4.269	1.114
Winter						
$PM_{2.5}$ (μg m^{-3})	30	3.2	15.9	13.2	39.1	10.4
Soot ($m^{-1} \times 10^{-5}$)	30	0	2.265	2.008	5.390	1.610
Spring						
$PM_{2.5}$ (μg m^{-3})	30	1.2	17.1	18.0	32.7	8.7
Soot ($m^{-1} \times 10^{-5}$)	30	0.118	0.918	0.837	3.933	0.732
Summer						
$PM_{2.5}$ (μg m^{-3})	30	2.0	9.0	8.9	17.3	3.7
Soot ($m^{-1} \times 10^{-5}$)	30	0.121	0.594	0.547	1.465	0.331

No missing values for $PM_{2.5}$ or soot. 1 and 7 missing values for relative humidity and wind speed, respectively

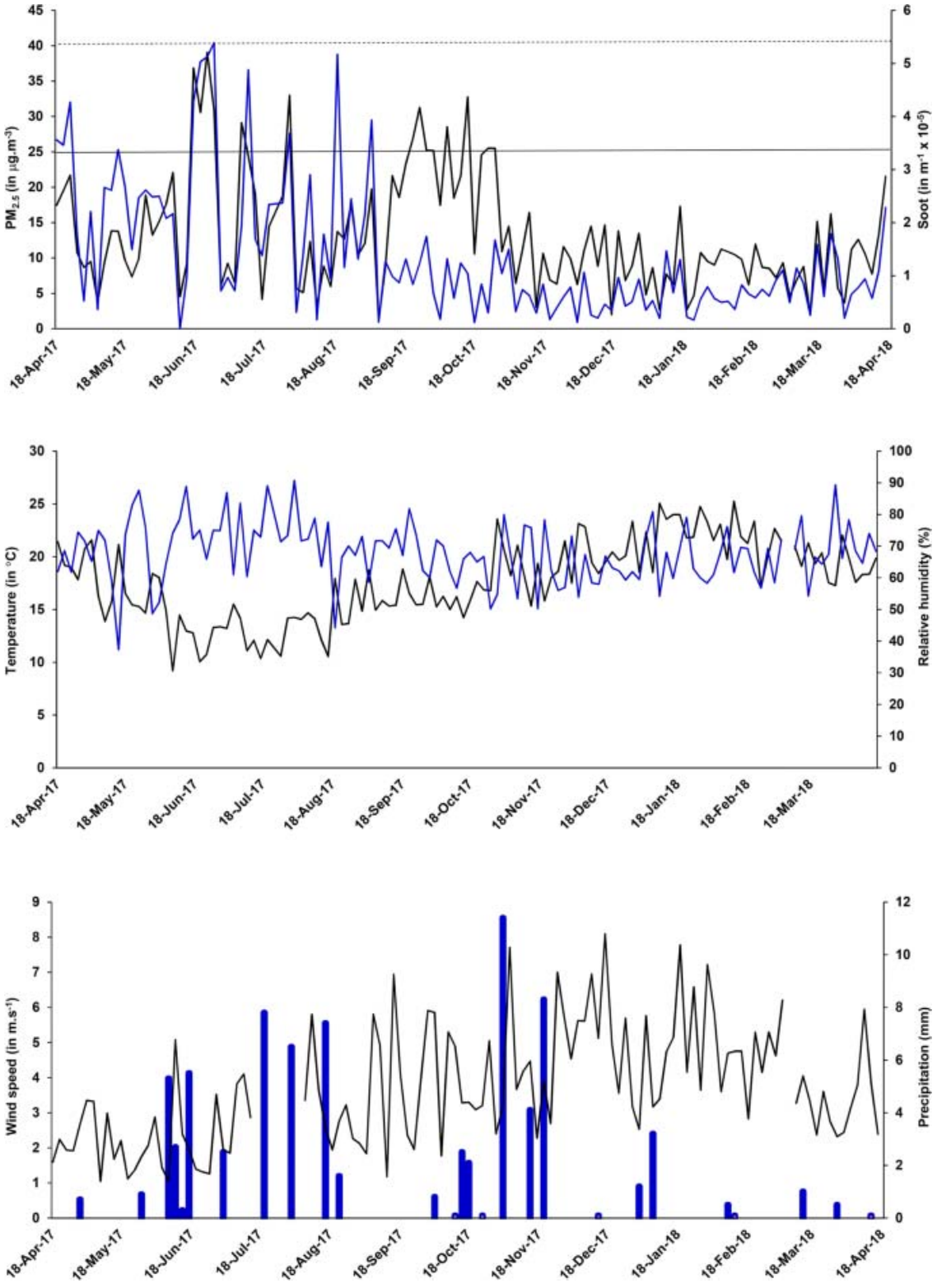

Fig. 3. Time-series graphs of $PM_{2.5}$, soot and meteorological conditions on 121 days during April 18, 2017, to April 16, 2018, in Cape Town, South Africa

The mean $PM_{2.5}$ level for the 1-year study period was 13.3 μg m^{-3}, which was below the yearly South African National Ambient Air Quality Standard (SA NAAQS) (20 μg m^{-3}) (Department of Environmental Affairs 2005), but exceeded the yearly WHO air quality guideline (10 μg m^{-3}) (World Health Organization 2005). Median $PM_{2.5}$ levels in winter (13.2 μg m^{-3}) and spring (18.0 μg m^{-3}) were significantly higher ($p < 0.05$) than in autumn (9.8 μg m^{-3}) and summer (8.9 μg m^{-3}). The daily WHO air quality guideline (25 μg m^{-3}) was exceeded on 5 and 9 days during winter and spring, respectively. The daily SA NAAQS (40 μg m^{-3}) was never exceeded. These exceedances indicate that the population of Cape Town may experience various health outcomes due to outdoor $PM_{2.5}$ exposure, as indicated previously.

The median $PM_{2.5}$ level on 38 weekend/public holidays (14.1 μg m^{-3}) was significantly higher ($p < 0.05$) than that on 83 weekdays (9.9 μg m^{-3}). A possible reason may be that neighbours close by performed metal works over weekends or due to the South African tradition of barbequing on weekends; the samples were collected in a residential area. However, median soot levels on weekdays and weekends did not differ significantly, see below.

The mean $PM_{2.5}$ level in this study was lower than the mean in 499 cities of 24 countries (37.5 μg m^{-3}) (Liu et al. 2019). In 2012, the yearly average $PM_{2.5}$ levels in South Africa ranged between 4.9 and 43.3 μg m^{-3}, and for all 21 AAQM stations the annual average was 24.1 μg m^{-3} (Altieri and Keen 2019). The maximum $PM_{2.5}$ level was 39.1 μg m^{-3}, which is generally lower than those reported in cities from other African countries (deSouza 2020; Kalisa et al. 2019; Fayiga et al. 2018; Gaita et al. 2014; Petkova et al. 2013). The mean and maximum $PM_{2.5}$ levels were higher than those reported in rural areas of South Africa (Tshehla and Djolov 2018; Novela 2019; Novela et al. 2019), but lower than those reported in Pretoria, located about 1600 km north of Cape Town (Morakinyo et al. 2019; Adeyemi 2020; Mwase 2020) or the South African towns located in the Vaal Triangle and Highveld air pollution priority areas (Olutola et al. 2019).

The mean soot level was 1.368 $m^{-1} \times 10^{-5}$ (Table 1). Median soot levels in autumn (1.493 $m^{-1} \times 10^{-5}$) and winter (2.008 $m^{-1} \times 10^{-5}$) were significantly higher ($p < 0.05$) compared with those in spring (0.837 $m^{-1} \times 10^{-5}$) and summer (0.547 $m^{-1} \times 10^{-5}$). Soot levels had a consistent downward trend during June 2017 to December 2018 (Fig. 3). Autumn and winter had the largest variations in soot levels. Median soot levels on weekdays (0.916 $m^{-1} \times 10^{-5}$) and weekends (1.096 $m^{-1} \times 10^{-5}$) ($p > 0.05$) did not differ significantly. There is no SA NAAQS or WHO air quality guidelines for soot.

Very few studies in Africa reported soot levels. The mean soot levels in Pretoria were higher and varied between 2.3 $m^{-1} \times 10^{-5}$ during April 2017 to April 2018 and 0.02 $m^{-1} \times 10^{-5}$ during May 2018 to Apr 2019 (Adeyemi 2020, Mwase 2020). The mean soot levels in Thohoyandou, a town located in a rural area about 2000 km north of Cape Town, were lower: 0.69 $m^{-1} \times 10^{-}5$ during April 2017 to April 2018 (Novela 2019). Luben et al. (2017) concluded in a review that atmospheric black or elemental carbon is a risk factor for hospital admissions and mortality. Cesaroni et al. (2014) focused on air pollution exposure over many years and reported a 10% increase in acute myocardial infarction or other acute and sub-acute forms of ischemic heart disease per unit $m^{-1} \times 10^{-5}$ increase in soot levels in five European countries.

96-h $PM_{2.5}$, anion and elemental composition levels

The 96-h $PM_{2.5}$ mass ranged between 300 and 400 µg (Table 2). The 96-h $PM_{2.5}$ level was higher on Saturdays (17.4 µg m^{-3}) than on Thursdays (13.0 µg m^{-3}) in September 2017 (spring). In contrast, the 96-h $PM_{2.5}$ level was lower on Saturdays (13.0 µg m^{-3}) than on Tuesdays (17.4 µg m^{-3}) in January 2018 (summer). The meteorological conditions were similar during the 24-h and 96-h sampling.

Table 2. Anion and elemental composition levels (in µg and µg m^{-3}) of the four 96-h $PM_{2.5}$ samples collected on 22 days in September 2017 and January 2018 in Kraaifontein, Cape Town, South Africa

	September 2017						January 2018					
	Weekday ($n = 1$)			Weekend ($n = 1$)			Weekday ($n = 1$)			Weekend ($n = 1$)		
	µg m^{-3}	µg	% of mass	µg m^{-3}	µg	% of mass	µg m^{-3}	µg	% of mass	µg m^{-3}	µg	% of mass
$PM_{2.5}$	13.0	300.0	100.0	17.4	400.0	100.0	17.4	400.0	100.0	13.0	300.0	100.0
Anions	4.9	114.0	38.0	5.3	123.0	30.7	7.1	164.0	41.0	7.0	161.0	53.7
Cl^-	3.5	79.8	26.6	3.7	85.4	21.4	4.0	92.9	23.2	4.0	91.5	30.5
NO_3^-	0.6	14.6	4.9	0.6	14.3	3.6	1.6	35.8	9.0	1.1	25.5	8.5
SO_4^{2-}	0.8	19.5	6.5	1.0	23.1	5.8	1.5	35.1	8.8	1.9	44.2	14.7
Elements	4.7	109.0	36.3	7.9	181.0	45.2	8.8	202.0	50.5	3.6	82.3	27.4
Al	ND	ND	-	0.1	2.7	0.7	ND	ND	-	0.1	1.2	0.4
Ca	ND	ND	-	0.2	4.2	1.1	1.9	43.6	10.9	1.0	23.2	7.7
Fe	ND	ND	-	1.3	29.4	7.4	ND	ND	-	1.2	28.2	9.4
Mg	ND	ND	-	ND	ND	-	0.2	5.7	1.4	0.1	2.4	0.8
Na	4.7	109.0	36.3	6.3	144.0	36.0	6.5	150.0	37.5	1.1	25.2	8.3
Zn	ND	ND	-	0.03	0.6	0.2	0.1	3.0	0.8	0.1	2.1	0.7
Inorganic carbon	1.8	42.6	14.2	2.1	48.5	12.1	1.2	27.9	7.0	1.4	32.5	10.8
Undetermined	1.5	34.5	11.5	2.1	47.7	11.9	0.3	6.3	1.6	1.0	24.0	8.0

ND not detected

The largest fraction of $PM_{2.5}$ was due to anionic and metallic species ranging from 31 to 54% of mass and 27–51% of mass, respectively (Table 2 and Fig. S8). Na and Cl^- were the most abundant constituents of $PM_{2.5}$. The Atlantic Ocean (20 km west from the study site) and Indian Ocean (25 km south from the study site) are the most probable sources of Na and Cl^- in the $PM_{2.5}$ samples. Several epidemiology studies that investigated acute health outcomes, such as hospital admissions, published since 2005 have included a source category for sea salt. Sea salt has not been associated with adverse health outcomes (World Health Organization 2013).

A study from Pretoria, located inland about 600 km from the Indian Ocean, sampled $PM_{2.5}$ and reported lower mean 24-h Cl^- levels during spring and summer, 19.3 ng m^{-3} and 16.8 ng m^{-3}, respectively (Adeyemi 2020). A study from Nairobi, Kenya, located inland about 500 km from the Indian Ocean, sampled $PM_{2.5}$ and reported lower mean Cl^- (Gaita et al. 2014).

Mg may also originate from sea spray, although it was only detected in the January 2018 samples. Ca was detected in three of the four samples. Ca content can be attributed to either sea sand, soil or mineral dust due to traffic or wind. The mean 24-h Ca levels were much lower in Pretoria, namely 95.4 ng m^{-3} and 87.8 ng m^{-3} during spring and summer, respectively. Mean Ca levels (0.31 µg m^{-3}) were lower in Nairobi, Kenya (Gaita et al. 2014).

Tshehla and Djolov (2018) reported $PM_{2.5}$ Ca levels in a rural area in Limpopo province, South Africa, that ranged from 0.3 to 9.9 μg m^{-3}.

Samples collected in Saturdays during spring and summer contained Al (0.4–0.7% of mass) and Fe (7.4–9.4% of mass); these elements were not detected in samples collected on Tuesdays and Thursdays. A possible reason for Fe detected in weekend samples is that two neighbours, approximately 20–50 m from the study site, had performed metal works over weekends.

The mean 24-h Fe levels measured at a background site in Pretoria were lower, namely 0.355 μg m^{-3} and 0.237 μg m^{-3}, during spring and summer respectively and 20.9 ng m^{-3} and 9.9 ng m^{-3} for Zn (Adeyemi 2020). A study from Nairobi, Kenya, reported lower mean Fe and Zn levels, namely 0.53 μg m^{-3} and 0.91 μg m^{-3}, respectively (Gaita et al. 2014). Tshehla and Djolov (2018) reported Fe levels that ranged from 0.1 to 1.4 μg m^{-3}. A large European cohort epidemiology study reported much lower Fe and Zn levels (Beelen et al. 2015). Even at such lower Fe and Zn levels, an increase of 3% in natural-cause mortality was observed per 500 ng m^{-3} increase in Fe or per 20 ng m^{-3} increase in Zn (Beelen et al. 2015). The observed Fe and Zn levels, if assumed to be experienced citywide in Cape Town, may thus pose a significant risk to human health.

Zn:Al ratios indicate the relative contribution of local road dust to $PM_{2.5}$ levels (Snider et al. 2016). Al is predominantly from natural sources (Snider et al. 2016), whilst Zn is mainly from tire wear, but also from solid waste or biomass burning and industrial emissions. Snider et al. (2016) reported Zn:Al ratios that ranged from 0.13 to 3.74 with a mean of 0.73. The mean ratio in this study was 2.17, which is higher than of the Pretoria study site (0.86) reported by Snider et al. (2016).

SO_4^{2-} and NO_3^- were present in all samples and indicated combustion sources in the area, which may include traffic and biomass fuel burning. SO_4^{2-} in $PM_{2.5}$ leads to a substantial increase in the bioavailable metals and soot (World Health Organization 2013). A review concluded that natural-cause mortality increased by 15% and 17% per 1 μg m^{-3} increase in SO_4^{2-} and NO_3^-, respectively (Atkinson et al. 2015). The observed SO_4^{2-} and NO_3^- levels, if assumed to be representative of levels in Cape Town, may thus pose a significant risk to human health.

The inorganic carbon fraction ranged from 7.0–14.2% of mass and the level was higher on Saturdays than on Tuesdays and Thursdays, which may be due to the South African tradition of barbequing on weekends.

Another sizeable part of $PM_{2.5}$ mass was the 'undetermined' portion that consisted most probably of ammonium (as NH_4Cl), volatile organic carbons, polyaromatic hydrocarbons, peroxyacyl nitrates and water (Kreidenweis et al. 2008; Widziewicz-Rzońca and Tytła 2020).

24-h $PM_{2.5}$ and soot levels by geographical origin of air masses

Four geographical origins of air masses were identified: Atlantic-Ocean-SW, Atlantic-Ocean-SSW, Atlantic-Ocean-WSW and Indian-Ocean (Fig. 2). Air masses emanating from the Atlantic Ocean dominated in winter (87%), summer (73%), spring (77%) and the entire year (71%) (Table S7). In autumn, 51% of the air masses emanated from the Indian Ocean, with 29% in the entire study period.

The median 24-h $PM_{2.5}$ and soot levels did not differ significantly by the geographical origin of air masses ($p > 0.05$) (Table 3).

Table 3. Descriptive statistics of 24-h $PM_{2.5}$ and soot levels on 121 days during April 18, 2017, to April 16, 2018, in Kraaifontein, Cape Town, South Africa, by geographical origin of air masses

Variable	*N*	Minimum	Mean	Median	Maximum	Std dev
Indian-Ocean						
$PM_{2.5}$ ($\mu g\ m^{-3}$)	35	2.7	13.1	11.2	30.9	6.6
Soot ($m^{-1} \times 10^{-5}$)	35	0.121	1.592	0.916	5.390	1.462
Atl-Ocean-WSW						
$PM_{2.5}$ ($\mu g\ m^{-3}$)	48	1.2	12.7	9.9	36.8	8.6
Soot ($m^{-1} \times 10^{-5}$)	48	0.118	1.172	0.848	4.289	0.999
Atl-Ocean-SW						
$PM_{2.5}$ ($\mu g\ m^{-3}$)	29	2.0	13.8	11.6	39.1	9.1
Soot ($m^{-1} \times 10^{-5}$)	29	0.000	1.393	1.157	5.122	1.219
Atl-Ocean-SSW						
$PM_{2.5}$ ($\mu g\ m^{-3}$)	9	3.0	15.2	13.8	31.3	8.5
Soot ($m^{-1} \times 10^{-5}$)	9	0.201	1.465	1.061	4.881	1.479

The median temperature and relative humidity levels differed significantly by the geographical origin of air masses ($p < 0.05$) (Table S8), with the highest temperature (19.7 °C) observed when the air mass originated in the warm Indian Ocean and the lowest relative humidity (58.8%) observed when the air mass originated in the cold Atlantic Ocean (Atl-Ocean-SSW). Wind speed and precipitation levels were not influenced by the geographical origin of air masses ($p > 0.05$).

Correlation between 24-h air pollutant levels and meteorological conditions

The correlation between $PM_{2.5}$ and soot levels was positive (0.596) (Fig. S9) and the strongest during winter (0.818), followed by autumn (0.750), spring (0.546) and summer (0.427) (Table S9). All these correlations were significant.

PM_{10}, NO_2, SO_2 and O_3 levels varied by the hour of the day and indicated possible sources (Figs. S10 to S13; Supplementary text S4). The descriptive statistics are indicated in Table S10 and discussed in Supplementary text S4.

$PM_{2.5}$ and soot levels measured at the Kraaifontein sampling site had positive and significant correlations ($p < 0.0001$) with PM_{10} levels measured 3 km away at the Wallacedene AAQM station (Table 4). This may indicate that the PM_{10} sources indicated by the Wallacedene AAQM station (Supplementary text S4), namely traffic and biomass burning for space heating and barbequing, may influence the $PM_{2.5}$ levels measured at the Kraaifontein sampling site. The correlation between $PM_{2.5}$ and PM_{10} (0.481) was weaker than the correlation observed in 652 cities in 24 countries (0.78) (Liu et al. 2019).

Table 4. Correlation between $PM_{2.5}$, soot, PM_{10}, NO_2, SO_2, O_3 levels on 121 days during April 18, 2017, to April 16, 2018, in Cape Town, South Africa

	$PM_{2.5}$	Soot
Wallacedene		
PM_{10}	0.481*	0.614*
NO_2	0.638*	0.221
SO_2	0.308*	0.276*
O_3	−0.114	−0.485*
Goodwood		
NO_2	0.415*	0.380**
SO_2	0.597*	0.681*
O_3	−0.214	−0.140
Tableview		
NO_2	0.438*	0.726*
SO_2	0.241*	0.440*
City Hall		
NO_2	0.345*	0.690*
SO_2	0.051	0.334**
Somerset West		
SO_2	0.223*	0.231*
Atlantis		
SO_2	0.198*	0.359*
O_3	0.228*	0.067

*$p = 0.05$

*$p = 0.05$

For number of missing values, refer to Table S10

$PM_{2.5}$ levels had positive and significant correlations with NO_2 levels measured at the Wallacedene, Goodwood, Tableview (18 km away) and City Hall (27 km away) AAQM stations, with the strongest correlations (0.638) between NO_2 levels measured at the Wallacedene AAQM station (Table 4). NO_2 is a precursor for ions of water-soluble inorganic salts that can partition to the particulate-phase; hence, traffic in the vicinity of these four AAQM stations (Supplementary text S4) may contribute to the $PM_{2.5}$ levels measured at the Kraaifontein sampling site. The strength of the correlations with Goodwood, Tableview and City Hall AAQM stations did not follow a clear trend according to distance from the Kraaifontein sampling site. Liu et al. (2019) reported a correlation of 0.48 between $PM_{2.5}$ and NO_2 in 652 cities in 24 countries.

As with $PM_{2.5}$, soot levels had positive and mostly significant correlations with NO_2 levels with the strongest correlation (0.726) between NO_2 levels measured at the Tableview AAQM station (Table 4). As with $PM_{2.5}$, the strength of the correlations with Goodwood, Tableview

and City Hall AAQM stations did not follow a clear trend according to distance from the Kraaifontein sampling site.

$PM_{2.5}$ levels had positive and mostly significant correlations with SO_2 levels measured at the six AAQM stations, with the strongest correlation (0.597) at the Goodwood AAQM station and the weakest (0.198) at the Atlantis AAQM station (37 km away) (Table 4). As with NO_2, SO_2 is a precursor for ions of water-soluble inorganic salts that can partition to the particulate-phase; hence, the SO_2 sources indicated by these six AAQM stations (Supplementary text S4), namely traffic and an oil refinery, may lead to higher $PM_{2.5}$ levels measured at the Kraaifontein sampling site. The lack of significant polluters to the north of the study site and poor correlation with SO_2 levels in the town of Atlantis indicated that air mass transport of airborne particulate matter from the north was minimal. The strength of the correlations did not follow a clear trend according to distance from the Kraaifontein sampling site. Liu et al. (2019) reported a correlation of 0.40 between $PM_{2.5}$ and SO_2 in 652 cities in 24 countries.

As with $PM_{2.5}$, soot levels had positive correlations with SO_2 levels measured at the six AAQM stations. The strongest correlation (0.681) was between SO_2 levels measured at the Goodwood AAQM station and the weakest (0.231) at the Somerset West AAQM station (Table 4). As with $PM_{2.5}$, the strength of the correlations did not follow a clear trend according to distance from the Kraaifontein sampling site.

$PM_{2.5}$ and soot levels had negative and mostly insignificant correlations with ground-level O_3 levels measured at Wallacedene and the Goodwood AAQM stations, but a positive and significant correlation with ground-level O_3 levels measured at the Atlantis AAQM station (Table 4). Liu et al. (2019) reported a correlation of 0.22 between $PM_{2.5}$ and O_3 in 652 cities in 24 countries.

Meteorological conditions can diffuse, dilute and accumulate air pollution. The results are presented in Tables S9 and S11 and discussed in Supplementary text S5.

Conclusions

The mean $PM_{2.5}$ level for the 1-year study period was 13.3 μg m^{-3}, which exceeded the yearly WHO air quality guideline (10 μg m^{-3}). Median $PM_{2.5}$ levels in winter and spring were significantly higher than in autumn and summer These exceedances indicate that the population of Cape Town may experience various health outcomes due to outdoor $PM_{2.5}$ exposure.

Median soot levels in autumn and winter were significantly higher compared with those in spring and summer. The largest fraction of $PM_{2.5}$ was due to anionic and metallic species. The inorganic carbon fraction ranged from 7.0–14.2% of mass. Another sizeable part of $PM_{2.5}$ mass was the “undetermined” portion.

Four geographical origins of air masses were identified and those emanating from the Atlantic Ocean dominated most of the year. In autumn, the air masses emanated from the Indian Ocean were more prevalent. The $PM_{2.5}$ and soot levels did not differ significantly by the geographical origin of air masses though.

It was observed that air pollution sources indicated by the AAQM stations, namely traffic, biomass burning for space heating and barbequing and an oil refinery, may lead to higher $PM_{2.5}$ levels measured at the Kraaifontein sampling site.

Recommendations are that all the $PM_{2.5}$ samples are analysed for chemical composition to perform source apportionment and health risk assessment studies.

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Data availability

Upon request, but then, the authors want to be co-authors on the other study's manuscript.

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