Air Quality, Atmosphere & Health (2022) 15:209–220
https://doi.org/10.1007/s11869-021-01097-5

# Seasonal abundances of primary and secondary carbonaceous aerosols at a high-altitude station in the Western Ghat Mountains, India

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Received: 11 March 2021 / Accepted: 13 September 2021 / Published online: 22 September 2021


**Abstract**
The combustion-related primary and secondary carbon particles have gained more importance in the recent past due to their radiative and chemical properties. The present paper deals with the results obtained from observations on Organic Carbon (OC) and Elemental Carbon (EC) during 2019–20 over a high-altitude location, Mahabaleshwar in the Western Ghats in peninsular India. The data is classified into summer (March to May 2019) and winter (December 2019 to February 2020). Mean OC mass was more during summer ($13.6 \pm 4.4$ μg / $m^3$) than winter ($11.1 \pm 3.2$ μg / $m^3$). However, mean EC mass was more in winter ($3.6 \pm 1.1$ μg / $m^3$) than in summer ($2.2 \pm 1.1$ μg / $m^3$). The mean annual OC/EC ratio was $7.4 \pm 4.4$ in summer and $3.1 \pm 0.7$ in winter, suggesting more presence of secondary organic carbon (SOC) during summer. Estimated SOC formed about 62% of OC in summer whereas in winter both SOC and primary organic carbon (POC) formed 50% each of OC. Together both POC and EC contributed to 46% of the total carbon (TC) in summer and 62% in winter indicating more primary fraction during winter. The effective carbon ratio (ECR) was 1.6 and 0.7 during summer and winter respectively, indicating the dominance of scattering-type secondary carbonaceous aerosols in summer. Cluster and concentrated weighted trajectory (CWT) analysis indicated high concentrations of OC and EC in continental originated trajectories. However, a high OC/EC ratio was observed for air masses arriving from long-distance sources.

**Keywords** Organic and elemental carbon · Summer and winter · High-altitude mountainous site · Effective carbon ratio · Long-range transport

## Introduction

Carbonaceous aerosols contribute a major fraction of both coarse (particles less than or equal to 10-μm size, i.e. $PM_{10}$) and fine (particles less than or equal to 2.5-μm size, i.e. $PM_{2.5}$) atmospheric particles, comprising about 10–43% and 21–78% of their respective masses (Putaud et al. 2004; Fuzzi et al. 2006). Especially, the $PM_{2.5}$ particles are responsible for several phenomena like human health problems (Pope et al 2002; Bell et al. 2007), agricultural crop production, chemical composition of the atmosphere, and air pollution–related problems such as visibility reduction (Ramanathan et al 2001; Li and Bai 2009) and radiation balance of the Earth (Novakov et al. 2005; Bond et al 2013). Overall, the carbonaceous aerosols are mainly composed of organic carbon (OC) as a major component, contributing up to 90% of total carbon and a relatively low contribution of about 10% from elemental carbon (EC). The primary organic carbon (POC) occurs as particulate organic matter (POM) derived directly from combustion activities whereas the secondary organic carbon (SOC) is formed due to the oxidation of gas phase precursors of volatile organic species or through the ageing process of POC, in the atmosphere. Both OC and EC differ in their optical and chemical properties. OC mostly scatters the solar radiation while EC is an efficient absorber (Ackerman et al. 2000, Jacobson 2001). OC comprises a large variety of organic compounds (aliphatic,

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aromatic compounds and acids). It is reported that some of the OC compounds act as cloud condensation nuclei (CCN) and thereby show indirect climatic effects by affecting the cloud microphysical properties and precipitation mechanism (Kulmala et al. 1996).

The major anthropogenic sources of OC in the atmosphere are vehicular exhausts and combustion of fossil fuel and biomass burning, whereas EC mainly originating from incomplete combustion processes exists as a basic constituent of soot particles that is highly refractive and is chemically inert and highly absorbing type. The relative abundances of EC and OC, i.e. OC/EC ratio, are critical in assessing the impact of carbonaceous species on the climate forcing (Novakov et al. 2005). Due to their crucial role in the Earth's radiation budget, air quality, atmospheric chemistry, and human health, it becomes very important to measure and study the physical as well as chemical characteristics of both OC and EC. So far, several studies on this aspect have been reported from different Indian locations highlighting the dominance of both OC and EC in the winter season and comparatively lower concentrations in the summer and monsoon seasons (Safai et al 2014; Gawhane et al 2017; Kumar and Yadav 2016; Ram et al 2008; Pachauri et al 2013; Sandeep et al 2020). Overall, the number of studies on the characterisation of carbonaceous aerosols is reported more from the Indo Gangetic Plain (IGP) than other parts of the Indian region. In fact, although the studies on OC and EC aerosols have gathered momentum over the past one decade, still such studies from the high-altitude and rural locations, especially from the Western Ghats region, are very few (Rengarajan et al. 2007; Ram et al. 2008; Ram and Sarin 2010, 2011; Satsangi et al. 2012; Safai et al 2013; Panicker et al. 2015; Ali et al. 2016). The present study conducted at a high-altitude site in the mountainous ranges of the Western Ghats in Peninsular India during 2019–2020 covering summer and winter seasons is an attempt to fill this gap to some extent.

## Sampling location, meteorology, and methodology

### Sampling location and meteorology

Measurements on OC and EC were undertaken at the High Altitude Cloud Physics Laboratory (HACPL) at Mahabaleshwar (17.9217° N, 73.6556° E). HACPL is situated at the height of ~ 1378 m above mean sea level. The station height remains above the mixed layer height (MLH) throughout the year (MLH varies from 60 to 1293.6 m). It is situated at the hill top of Western Ghats mountain ranges and it is surrounded by dense forest. Also, being one of the famous tourist places in India, there are several hotels, shops, and residential houses. The site experiences local emissions from the vehicular exhaust from tourist activities, biomass burning for domestic and agricultural purposes, and also from the long range transport from distant sources. As such, the combination of complex sources (both natural and anthropogenic) which are active during different times of the year (Buchunde et al. 2019; Mukherjee et al. 2018) and regional photochemistry fabricates the site as an ideal testbed for studying the aerosol formation, their ageing, and dispersal/deposition processes. The present study utilises the data from March to May 2019 and December 2019 to February 2020 to represent the summer and winter seasons, respectively. This classification of seasons has been derived from the prevailing changes in the general meteorology over this region.

Figure 1 depicts the mean hourly variation of major meteorological parameters (wind speed, temperature, relative humidity, and boundary layer height) over the observational location during the summer and winter seasons. The seasonal variation over Mahabaleshwar showed distinct variation with the mean temperature varying from 19 °C during summer to 26 °C during winter. Relative humidity also showed distinct seasonal behaviour with a mean of 49% in summer and 75% in winter. Wind speeds were moderate to low during both summer and winter (~ 2 to 3 m/s). Wind direction was dominantly south-easterly during winter whereas, during summer, it was mainly south-westerly. The boundary layer height was less in winter with maxima of 1393 m whereas in summer it reached up to 2150 m. Mahabaleshwar receives heavy rainfall (~ 5426 ± 1177 mm) during the SW monsoon period of June to September (Kothawale et. al. 2016).

### Methodology

Observations on OC and EC were carried out using a semi-continuous OC-EC analyser of Sunset Laboratory, USA (Model 4G). The mass concentration of OC and EC was analysed at an hourly interval using the National Institute of Occupational Safety and Health (NIOSH) protocol based on thermal optical transmittance (TOT) (Birch and Cary 1996). Aerosols < 2.5-µm size were only sampled using an inlet fitted with $PM_{2.5}$ cut-off cyclone. Also, the sample was passed through a parallel plate organic denuder to reduce the effects of vapour phase organic adsorption to clean the quartz filter. The analysis was performed in 2 stages: an aliquot of sample filter (area 1.5$cm^2$) was stepwise heated in a furnace at 820 °C, was then cooled to 550 °C, and then it was again stepwise heated to 870 °C in an oxidising atmosphere (98% He and 2% $O_2$). During each temperature step, evolved carbon is oxidised to $CO_2$ and then reduced to methane using a catalyst and was detected by a non-dispersive infra-red detector (NDIR). A calibration was performed at the end of each analysis by introducing a known amount of methane gas into the oven and measuring its constant response.

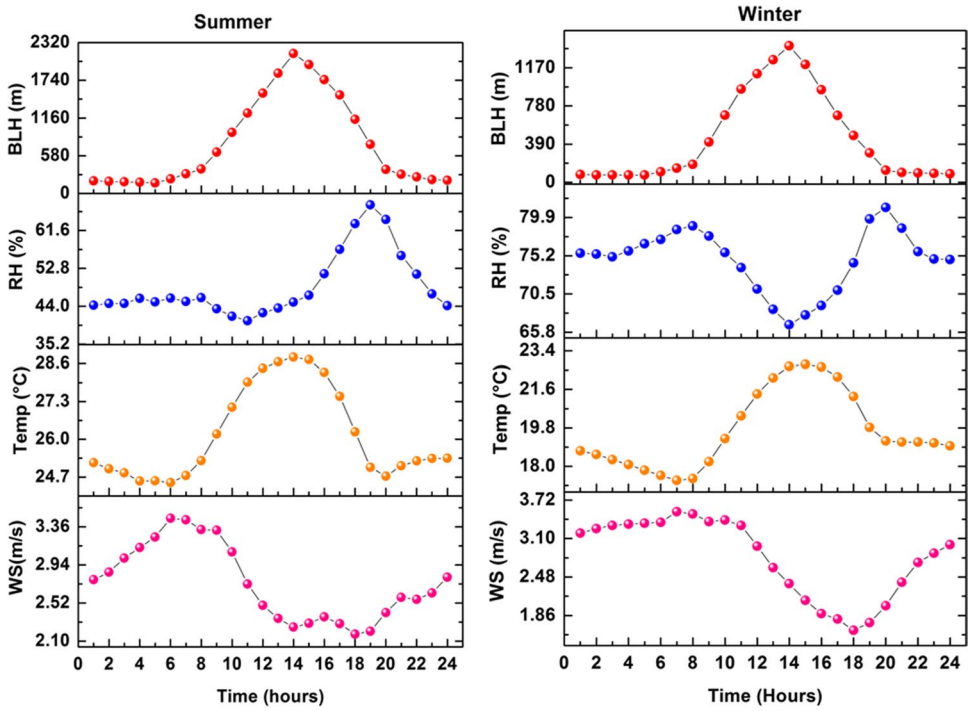


**Fig. 1** Mean hourly variation of wind speed (WS), temperature (T), relative humidity (RH), and boundary layer height (BLH) during the summer and winter

Standardisation of the instrument is carried out by using the sucrose solution (3.2 mg/ml). The analyser was calibrated frequently for quality control purposes by using a blank punch of pre-heated quartz fibre filter and standard sucrose solution. More details about the technique used, uncertainties, and calibration and correction factor are discussed in detail elsewhere (Safai et al. 2013, 2014).

An automatic weather station (AWS) was used to monitor the meteorological conditions prevailing over the observational site such as wind direction (WD), wind speed (WS), temperature (T), and relative humidity (RH) at the time interval of 1 min. Data was averaged to a time resolution of 1 h to compare with the data on OC and EC. The hourly variation of the boundary layer height (BLH) was computed from the National Oceanic and Atmospheric Administration Hybrid Single Particle Lagrangian Integrated Trajectories (NOAA-HYSPLIT) model (http://ready.arl.noaa.gov/HYSPLIT.php).

In order to unveil the role of long-range transport influencing the carbonaceous aerosol concentration over Mahabaleshwar, cluster and concentration weighted trajectory (CWT) analyses were performed. In short, CWT analysis estimates the concentration weighted trajectories for each grid on a geographical scale based on (i) the number of trajectories passing through the grid and (ii) the residence time of each trajectory in that particular grid by evenly distributing concentrations along the trajectories. On the other hand, cluster analysis was performed to minimise the variability between the trajectories within a cluster and maximise the variability among different clusters (Vinoj et al. 2010; Mukherjee et al. 2018, Buchunde et. al. 2019). The optimum number of back trajectory clusters was chosen based on the total spatial variance (TSV). Zefir, an Igor-based tool (Petit et. al. 2017) was utilised here for performing back trajectory cluster and CWT analysis. Hourly 5-day air mass back trajectory data utilised in the present study was obtained from HYSPLIT (Draxler and Rolph 2003) during winter and pre-monsoon seasons. The final arriving point of the air mass back trajectories was 100 m above the ground level.

Beta Attenuation Monitor (Model BAM-1020) was used to measure the $PM_{2.5}$ particles at 1-h interval and with the sampling flow rate of 16.7 L/min. In this sampler, at the beginning of each sample hour, a small C (carbon-14) source emits a constant source of high-energy electrons (known as beta rays) through a spot of clean filter tape. These beta rays are detected and counted by a sensitive scintillation detector to determine a zero reading. The BAM 1020 then advances this spot of tape to the sample nozzle, where a vacuum pump pulls a measured and controlled amount of ambient air through the filter tape, loading it with ambient dust. At the end of the sample hour, this dust spot is placed back between the beta source and the detector, thereby causing attenuation of the beta ray signal which is used to determine the mass of the particulate matter on the filter tape. This mass is used to calculate the volumetric concentration of particulate matter in ambient air. Simultaneous measurements of OC, EC, and $PM_{2.5}$ particles are used to compute the mass fractions of OC and EC to $PM_{2.5}$ for each observation.

## Results and discussion

### Temporal variation of OC, EC, and TC over Mahabaleshwar

#### Seasonal variation

As can be seen from Fig. 2, OC showed more concentration during summer ($13.6 \pm 4.4$ μg/m$^3$) than during winter ($11.0 \pm 3.2$ μg/m$^3$), whereas the mean concentration of EC was more in winter ($3.6 \pm 1.1$ μg/m$^3$) than in summer ($2.2 \pm 1.1$ μg/m$^3$). However, as the overall mean concentration of total carbon (TC) was formed majorly by OC (86% in summer and 75% in winter), the mean TC concentration was more in summer ($15.8 \pm 5.1$ μg/m$^3$) than in winter ($14.7 \pm 4.0$ μg/m$^3$). The EC/TC was 0.14 and 0.25 respectively for summer and winter. This feature indicates more absorbing carbonaceous aerosols (EC) during the winter season at Mahabaleshwar. This observation is also corroborated by the simultaneous observations on black carbon (BC) aerosols at the same location. The mean BC mass concentration was more during winter ($2.21 \pm 1.003$ μg/m$^3$) than during summer ($1.00 \pm 0.53$ μg/m$^3$). Similar results have been reported earlier for the BC concentration at the same location by Raju et al (2020). One of the possible reasons for the observed enhancement in OC and OC/EC ratios during the summer season could be more photochemical production of OC through oxidation of certain volatile organic compounds during this period (mainly assisted by more ambient temperature). Mukherjee et al (2018) have reported the formation of secondary organic aerosols (SOA) at this location during summer (MAM) through photochemical oxidation of isoprene-derived biogenic precursors. However, further studies are required to confirm this assumption. Table 1 gives information on OC and EC concentrations at different high-altitude and other locations over India. As seen from Table 1, the concentrations of both OC and EC observed at Mahabaleshwar during summer and winter were comparatively less than those reported at different urban/semi-urban locations (Pune, Delhi, Agra, Jabalpur, Udaipur, Hyderabad, Srinagar and Kangra) while they were more than those reported at other high-altitude locations like Nainital and Mt. Abu. Table 2 shows the statistical details (mean and standard deviation) of carbonaceous aerosols (OC, EC, TC, POC, SOC) and related ratio values (OC/EC, EC/TC, ECR, POC + EC/TC) during summer and winter at Mahabaleshwar.

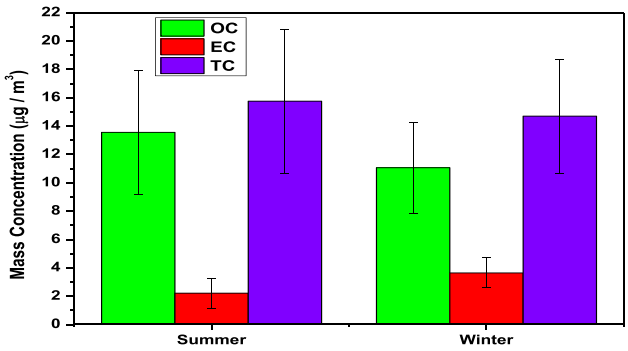


**Fig. 2** Mean seasonal variation of OC, EC, and TC during summer and winter

#### Diurnal variation

Figure 3 shows the mean hourly variation of OC and EC over Mahabaleshwar during summer and winter. OC showed

**Table 1** Comparison of EC and OC mass concentration from different locations in India

| Location | Altitude (mAMSL) | Period | OC (μg/m$^3$) | | EC (μg/m$^3$) | | Reference |
|---|---|---|---|---|---|---|---|
| | | | Summer | Winter | Summer | Winter | |
| Mahabaleshwar | 1353 | Mar 2019 – Feb 2020 | 13.56 | 10.99 | 2.22 | 3.64 | Present study |
| Pune | 559 | April 2015-Apr 2016 | 12.7 | 13.5 | 3.45 | 5.06 | Gawhane et al. (2017) |
| Delhi | 218 | 2011–2012 | 19.6 | 24.9 | 5.4 | 22.2 | Kumar and Yadav (2016) |
| Jabalpur | 412 | Jan 2011-Nov 2012 | 20 | 27 | 5 | 7 | Panicker et al. (2015) |
| Udaipur | 423 | Jan2011-Sep 2011 | 15 | 25 | 3 | 10 | Panicker et al (2015) |
| Hyderabad | 542 | 2010–2011 | 15.2 | 19.7 | 9.25 | 9.9 | Ali et al. (2016) |
| Nainital (Manora Peak) | 1950 | Apr 2005-Mar 2006 | 8.6 | 11 | 0.9 | 1.5 | Ram et al. (2008) |
| Mt. Abu | 1680 | May 2005-Feb 2006 | 3.5 | 3.6 | 0.1 | 0.8 | Ram et al. (2008) |
| Agra | 169 | May 2010-Apr 2011 | 20.3 | 36.1 | 2.9 | 5.0 | Pachauri et al. (2013) |
| Srinagar | 560 | Jan 2017-Dec 2017 | 15.6 | 18.21 | 5.74 | 6.24 | Sandeep et al. (2020) |
| Kangra | 600 | Jan 2012 to March 2013 | 11.9 | 18.26 | 3.24 | 7.97 | Kumar and Attri (2016) |

**Table 2** Statistical details of carbonaceous aerosols over Mahabaleshwar during summer and winter seasons of 2019–2020

| | OC | EC | OC/EC | TC | POC | SOC | EC/TC | ECR | (POC + EC)/TC |
|---|---|---|---|---|---|---|---|---|---|
| Summer mean | 13.56 | 2.21 | 7.43 | 15.76 | 5.08 | 8.46 | 0.14 | 1.56 | 0.46 |
| SD | 4.36 | 1.07 | 4.37 | 5.07 | 2.46 | 3.47 | 0.05 | 1.32 | 0.16 |
| Winter mean | 11.06 | 3.64 | 3.10 | 14.71 | 5.44 | 5.63 | 0.25 | 0.65 | 0.62 |
| SD | 3.22 | 1.06 | 0.67 | 4.02 | 1.57 | 2.43 | 0.04 | 0.27 | 0.10 |

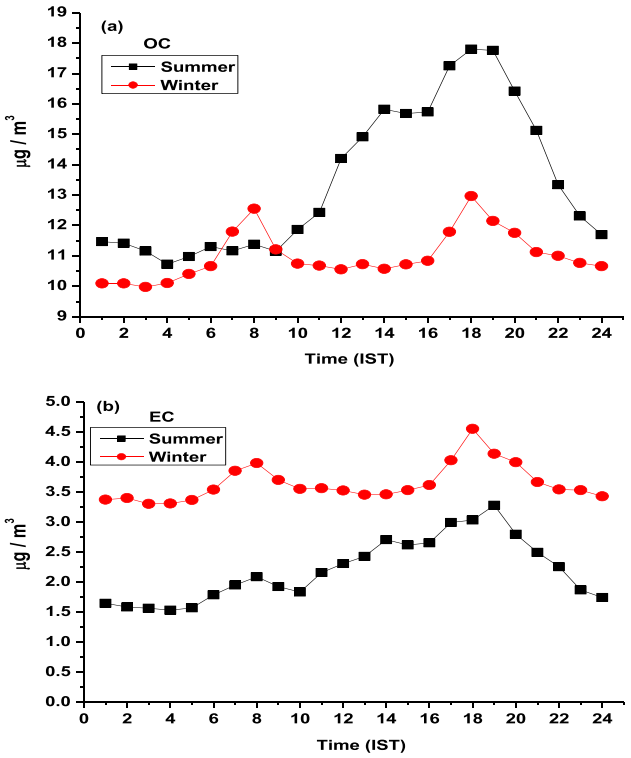


**Fig. 3** Mean hourly variation of **a** OC and **b** EC during summer and winter

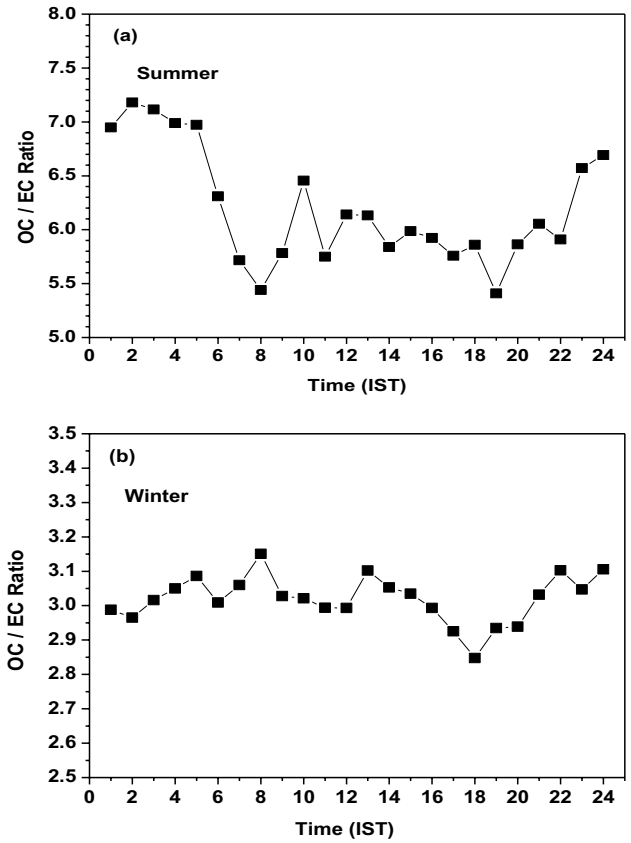


**Fig. 4** Mean hourly variation of OC/EC ratio during **a** summer and **b** winter

more concentrations at all the hours during summer as compared with those in winter, except during morning hours (07.00 IST to 09.00 IST). Especially, the difference was significantly high during noon (12.00 IST) to night hours (22.00 IST). This infers more incursion of biogenically produced mainly SOC during noon through night hours in the summer season. This assumption is supported from the hourly variation of SOC (Fig. 4) during summer and winter as shown in the "Estimation of primary and secondary organic carbon and effective carbon ratio" section wherein the computation of SOC and its hourly variation is discussed. Contrary to this feature, EC showed more concentration during all the hours in winter than those in summer. There was a bimodal distribution of both OC and EC during winter with a peak at 08.00 h IST in the morning and 18 h IST in the evening indicating towards the occurrences of local burning activities especially for domestic purpose in that period. During summer, the OC and EC concentrations started increasing from around 10.00 h IST in the morning and reached the maximum at around 18.00–19.00 h IST in the evening. The low concentrations of both OC and EC were observed during midnight to early morning hours (01.00 to 05.00 IST) in both summer and winter seasons. However, in winter, another trough was observed from 10.00 to 16.00 h IST for both OC and EC. The observed different patterns of variation of both OC and EC in summer than in winter can be attributed to the changes in dynamics of the local boundary layer as well as varying human activities such as vehicular traffic intensity, burning of fossil fuels from household residential, and agricultural and commercial activities in the local surroundings. Babu and Moorthy (2002) had suggested that the ABL evolves after sunrise, the strengthened thermals lift and eventually break the nighttime inversion causing

the aerosols in the residual layer to mix with those near to the surface, leading to a sharp increase in the near-surface concentrations during morning hours; an effect known as fumigation (Stull 1999). Safai et al. (2004) have reported similar bimodal behaviour of absorbing black carbon aerosols at an urban site, Pune, with higher concentrations during the morning and evening hours when the traffic density was more inside the city.

## Mass fraction of OC and EC to $PM_{2.5}$ in summer and winter

The mass concentration of $PM_{2.5}$ was simultaneously measured over the sampling site at Mahabaleshwar. Mass concentration of OC contributed about 42% and 79% of $PM_{2.5}$ during winter and summer seasons respectively, whereas EC contributed about 13% and 12% of $PM_{2.5}$ during winter and summer respectively. Overall, the mass concentration of TC formed about 55% and 90% of $PM_{2.5}$ during the winter and summer seasons, respectively. This infers that a significant fraction of $PM_{2.5}$ is formed by carbonaceous aerosols, especially more during the summer season. It is stated that the carbonaceous aerosols, comprising of organic and elemental carbon (OC and EC), are ubiquitous in the Earth's atmosphere and contribute significantly (about 21 to 78%) to the fine mode particulate matter (Putaud et al 2004; Yu et al 2004). Kanakidou et al. (2005) have reported that these aerosols account for about 90% of the total aerosol load over tropical forest areas. Mahabaleshwar being a tropical high-altitude station, surrounded by forest and vegetation, has a substantial abundance of carbonaceous aerosols, especially significant in the summer season. Biogenic production of secondary organic particles enhanced by more photochemical processing during summer season generation due to high temperatures in summer is attributed to this feature. However, in other urban, semi-urban locations in India, the contribution of carbonaceous particles to total aerosol load in fine size is comparatively less. Panicker et. al. (2015) have reported the contribution of TC to $PM_{2.5}$ in the range of 31–75% over Jabalpur and 30–83% over Udaipur, two urban cities in northwestern India. Gawhane et al (2017) have reported about 30% contribution from OC and 9% from EC to $PM_{2.5}$ at Pune, a rapidly emerging mega city in India. They have also reported a maximum (12%) fraction of EC to $PM_{2.5}$ during the winter season. Srinivas and Sarin (2014) have reported that OC and EC account for 33.7 and 5.2% of $PM_{2.5}$ mass, respectively, at Kharagpur, an urban site located at the eastern end of the IGP. Recently, Sandeep et al. (2020) have observed about 24 to 32% of $PM_{2.5}$ formed from carbonaceous (OC and EC) aerosols at Srinagar, a semi-urban site located near the foothills of Western Himalayan mountainous ranges.

## OC/EC ratio

Various researchers have assigned characteristic OC/EC ratios to the different emission sources. The OC/EC ratios above 2 indicate the production of secondary organic aerosols. Therefore, these ratios are generally used to indicate the presence of primary as well as secondary organic aerosols (Chow et al. 1996). This ratio has been used as a measure for differentiating two major sources of carbonaceous aerosols, i.e. fossil fuel combustion and biomass burning. Saarikoski et al. (2008) reported an OC/EC ratio of 6.6 for biomass burning and 0.71 for vehicular emissions. OC/EC ratio of 7.7 corresponds to biomass burning (Zhang et al. 2007; Feng et al. 2009). The presence of an OC/EC ratio below 4 shows the dominance of fossil fuel burning sources, mainly vehicular emissions. To understand the source characteristics of OC and EC, OC/EC ratio is estimated for both summer and winter seasons. The average OC/EC ratio at Mahabaleshwar for the summer season was found to be $7.4 \pm 4.4$ and that in the winter was $3.1 \pm 0.7$ (Fig. 4). There was more variation in this ratio during summer (2–30) than during winter (1.5–10) indicating divergent sources of emission of OC and EC in the summer. The higher OC/EC ratio during summer is attributed to the predominance of organic carbon mainly derived from biogenic sources that generate mainly the secondary organic carbon (SOC) and fossil fuel/biomass burning which forms mainly primary organic carbon (POC) and partially some SOC too along with POC. Correlation analysis between OC and EC components is used to have an elementary idea of the source characteristics as well as to interpret emission and transformation characteristics of carbonaceous aerosols (Cao et al. 2004; Ram and Sarin 2010a, b; Satsangi et al. 2012). Both OC and EC were well correlated during summer ($r = 0.60$, $p < 0.0001$) and winter ($r = 0.69$, $p < 0.0001$) which indicates to the common source mechanism, i.e. influence of the local combustion activity either from fossil fuel or from biomass burning. At Mahabaleshwar, it is observed that the common source of atmospheric pollutants irrespective of seasons are the local activities, such as vehicular emissions, domestic burning, and heating as there is no major industrial activity around the observational site. Also, during the summer season, winds blow from a westerly direction that brings in the residual of forest fire emissions which also could be the possible source for pollutants over the observational site (Leena et. al. 2017).

## Estimation of primary and secondary organic carbon and effective carbon ratio

An important factor influencing OC/EC ratio and hence source characteristic is the formation of SOC due to oxidation and gas to particle partitioning of low volatile organic compounds (VOCs). At Mahabaleshwar, the OC/EC ratio

ranged from a minimum value of 2 to a maximum of 30 in summer whereas in winter, it ranged from 1.5 to 10. Thus, apart from the contribution of diverse emission sources for POC to the total OC, there is the presence of a significant amount of SOC, especially during the hot summer season at the study location which could be attributed to biogenic sources from surrounding vegetation. So far, the direct measurement of SOC and POC is not possible as they are being originated from various physical and chemical transformation processes. As such, they are estimated by (i) EC-tracer method, i.e. computation of the primary organic carbon using minimum OC/EC ratio method where EC is considered as a tracer and then subtracting it from total organic carbon (Castro et. al. 1999, Turpin and Huntzicker 1995) and (ii) by summing up the concentration of all such oxidation products found in aerosols (Schauer et. al. 1996). However, the EC-tracer method is widely used by several researchers (Ram et al 2008; Safai et al 2014; Ali et al., 2015; Pachauri et al 2013). In the present study, the concentration of SOC has been estimated using the EC-tracer method as follows:

The primary organic carbon (POC) is estimated by equation.

$[POC] = [OC/EC]_{min}$ X [EC].

Then, the SOC is estimated by subtracting the POC from total organic carbon as:

$[SOC] = [OC] - [POC]$.

This method assumes that samples that have a low OC/EC ratio entirely contain primary carbonaceous compounds only which is supposed to be formed through direct emissions from fossil fuel/biomass combustion processes. The limitations of this method are mentioned by Castro et al (1999).

Millet et al. (2005) proposed another technique known as the minimum R-squared (MRS) method for the estimation of $[OC/EC]_{min}$ and POC and SOC values. In this method, the $[OC/EC]_{min}\left(\frac{OC}{EC}\right)_{primary}$ is determined from the minimum coefficient of determination ($R^2$) of the linear regression between the SOC and EC obtained from the entire range of OC/EC values. The use of this method is reported in many studies (Wu and Yu 2016; Kaskaoutis et al 2020). The SOC values are initially estimated for each OC/EC value, and the $R^2$ value is calculated for each estimated SOC and the corresponding EC. The OC/EC ratio used for the SOC determination in which the least $R^2$ value obtained is then considered as the $[OC/EC]_{min}$ value. Apart from the EC-tracer method, we have also attempted the MRS method for computation of $[OC/EC]_{min}$ and thereby the POC and SOC. However, the $[OC/EC]_{min}$ value obtained by this technique was found to be 2.4 and 2.0 in summer and winter, respectively, whereas the $[OC/EC]_{min}$ ratio observed by EC-tracer method at Mahabaleshwar was 2.3 and 1.5 during the summer and winter seasons, respectively. Thus, there was no difference between the $[OC/EC]_{min}$ values obtained by employing these two methods, except for that in winter. The mean concentrations of POC and SOC obtained from EC-tracer method were $5.1 \pm 2.5$ μg/m$^3$ and $5.4 \pm 1.6$ μg/m$^3$, respectively, in summer and winter. With MRS method, the mean concentrations of POC and SOC were obtained as $5.3 \pm 2.6$ μg/m$^3$ and $7.5 \pm 2.1$ μg/m$^3$, respectively, during summer and winter. To validate the values of POC obtained by these two methods, we have compared them with the simultaneously measured mass concentrations of black carbon (BC) aerosols by using Aethalometer (AE-33) at the sampling location. BC aerosols are reported to be indicative of primary carbon particles that are originated from the incomplete combustion of fossil fuel and/or biofuel/biomass (Bond et al 2013). A good correlation was observed between BC and POC obtained by both the EC-tracer and MRS methods ($r = 0.62$ in summer and 0.50 in winter with $p < 0.0001$ for both the methods). However, the standard error was found to be slightly more in the MRS method (0.08) than for the EC-tracer method (0.05). Therefore, we have used the POC and SOC values derived from EC-tracer method for the further discussion.

Higher $[OC/EC]_{min}$ ratio in summer is attributed to the more OC mass abundance. The seasonal variation of SOC and POC over Mahabaleshwar showed that the percentage contribution of SOC in OC was 62% in summer and that of POC was 38%. During the winter, both SOC and POC formed almost equal (50% each) of OC. This feature again depicts the important role of photochemical oxidation of certain biogenic volatile organic carbon compounds that led to the enhancement of SOC in summer, whereas concentration of POC was significant in winter due to its formation from direct emissions from fossil fuel (vehicular exhaust) and biomass (domestic/agricultural burning) combustion sources which was also assisted by the prevailing low mixing heights especially during morning and evening hours. High temperature and less humidity are favourable for the gas to particle conversion of volatile organic compounds to produce more amount of secondary organic carbon in the summer season (Komppula et al. 2009; Ali et al 2016). Higher contribution of SOC to OC during the summer season is also reported by several studies (Castro et al. 1999; Gu et al. 2010; Khan et al. 2010). However, some studies have reported more SOC contribution to OC in winter (Dan et al. 2004; Li and Bai 2009; Pachauri et al 2013).

The hourly variation of SOC and POC followed the identical pattern as that of OC in both the summer and winter seasons (Fig. 5a, b). However, the variation of SOC showed more resemblance with that of OC in both seasons (especially in the summer). SOC was well correlated with OC in summer ($r = 0.82$, $p < 0.0001$) than POC ($r = 0.60$, $p < 0.0001$). Similarly in winter also, SOC showed better correlation with OC ($r = 0.88$, $p < 0.0001$) than POC ($r = 0.69$, $p < 0.0001$). Therefore, it can be emphasised

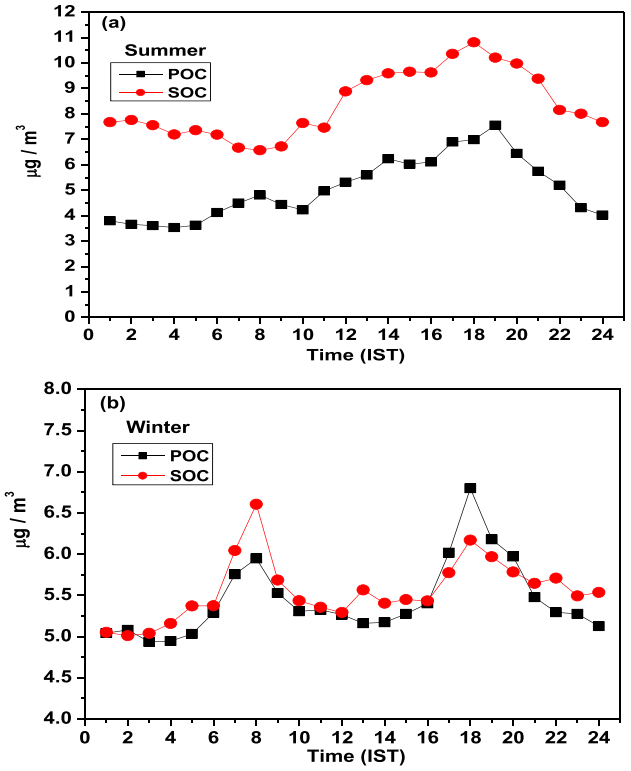


**Fig. 5** Mean hourly variation of POC and SOC during **a** summer and **b** winter

that the formation of OC showed more influence of local anthropogenic as well as biogenic sources during summer and in winter, apart from these sources; the contribution of possible transport from distant sources also added to it since the air mass during the winter season is mostly of the inland origin. It is also inferred from the high association among POC and SOC during the winter season (correlation between SOC and POC was 0.27, $p < 0.0001$) as compared to the summer season (correlation between SOC and POC was 0.03, $p = 0.37$) which may be the result of a major contribution from long-/short-range transport during the winter season. POC showed a much better correlation with EC in summer and winter ($r \geq 0.95$, $p < 0.0001$) indicating the clear influence of direct emission of primary organic aerosols mainly from combustion processes involved in vehicular as well as domestic and agricultural activities over the sampling location. The overall importance of primary and secondary carbonaceous aerosols can be judged by estimation of (POC + EC)/TC ratio which gives a fraction of primary carbonaceous aerosols in total carbonaceous aerosols. This ratio in the summer and winter seasons was 0.46 and 0.62, respectively, which shows the dominance of secondary carbonaceous aerosols in summer and primary carbonaceous aerosols in winter over Mahabaleshwar.

Even though the OC/EC ratio has been extensively used in climate models, it has certain limitations in terms of proper source assignment to both OC and EC as well as for the assessment of radiative impact of these carbonaceous aerosols (Novakov et al 2005; Safai et al 2014). OC comprises both primary (POC) and secondary (SOC) aerosols each having different source mechanisms as well as different ways to interact with solar radiation. It is generally observed that POC and EC originate from fossil-fuel burning from vehicular exhausts, biomass burning from agriculture and domestic cooking and have absorbing nature which leads to warming effect; whereas SOC mainly originates from oxidation of various types of volatile organic compounds and have scattering properties leading to the cooling effect. Therefore, a new term has been coined, i.e. effective carbon ratio (ECR) which is computed as the ratio SOC/(POC + EC). The higher value of ECR indicates low concentrations of POC and EC that could lead to the reduction in atmospheric warming effect of combustion-related carbonaceous aerosols (Safai et al. 2014). The ECR value at Mahabaleshwar was found to be $1.6 \pm 1.3$ in summer and $0.7 \pm 0.3$ in winter. This feature infers more abundance of absorbing carbonaceous aerosols during winter than in summer. This speculation is corroborated by the observed high concentrations of POC, EC, and BC in winter than those in summer.

## Long-range transport of OC and EC: CWT and Cluster analysis

The CWT analysis is generally utilised to ascertain the potential source regions responsible for the observed concentrations at the receptor site and associated concentration fields around the observation site, whereas cluster analysis reveals the regional transport pathways and quantifies the possible contribution of source regions to the observed airborne particle concentration by grouping the back trajectories of similar history. The detailed procedure for the cluster analysis and CWT analysis is given elsewhere (Cape et al. 2000; Jeong et al. 2011; Kabashnikov et al. 2011; Mukherjee et al. 2018; Buchunde et al. 2019).

Figure 6 depicts the results of cluster analysis during summer (a) and winter (b) seasons. During the summer season, > 84% of the trajectories are either originated from the sea or have travelled for the maximum period over the Arabian Sea (Fig. 6a). Cluster 1 is originated over Pakistan and travelled through Gujarat before reaching the observational site. The mean OC mass concentration associated with all the 3 clusters were observed to be equivalent (13.31–13.74 $\mu gm^{-3}$) as all the clusters are travelling through a similar continental region before reaching the observational site. Interestingly, the difference in mean EC concentration among the clusters is appreciable (range is 3.03–1.96 $\mu gm^{-3}$). The variability in EC loading between

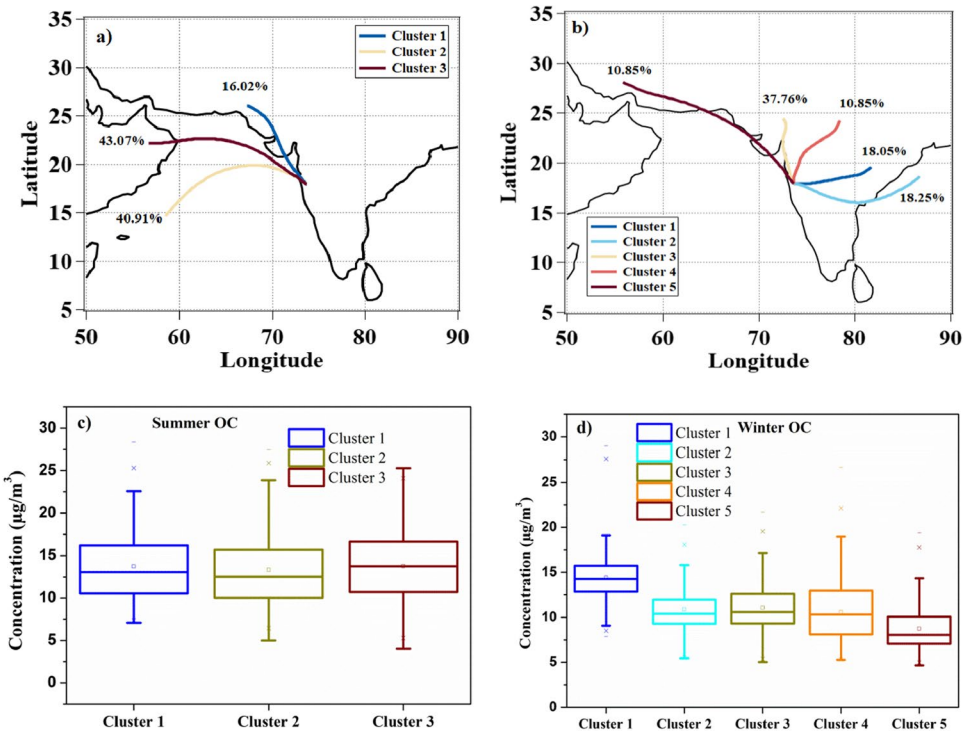


**Fig. 6** Cluster analysis during **a** summer and **b** winter and cluster-associated OC concentration (box and whisker plot) during **c** summer and **d** winter at Mahabaleshwar. The upper level of the box represents the 75th percentile and the lower level represents 25th percentile. The middle line of the box shows the median (50th percentile) and the star represents the mean of the variable. The box represents the inter quartile range of the variable. The upper and lowermost level of the line from the outside of the box defines maximum (95th percentile) and minimum (5th percentile) of the variable. Data points which lie outside of the box and whisker are defined as outliers and/or single data point

cluster 1 and clusters 2 and 3 may be attributed to the origin and pathway of air masses. As mentioned earlier, cluster 1 is of continental origin and travelled mostly through continental region before reaching Mahabaleshwar which might have carried higher air mass load as compared to clusters 2 and 3 which are of oceanic origin (or travelled more over the sea). Since cluster 3 is originated in the Middle East and covered a long distance before reaching the observational site, the OC/EC ratio was estimated to be higher than other clusters indicating less influence of EC from the long-distance source. Five clusters were prevailing during the winter season (Fig. 6b) and about 82% of the back trajectories originated from the continental region, whereas only 18% of the back trajectories originated from the sea region (both Arabian Sea and Bay of Bengal). Cluster 1 which originated in Chhattisgarh and travelled through Telengana before reaching the observational site accounted for about 18% of the total back trajectories and associated with the highest OC ($14.38 \pm 2.82$ $\mu gm^{-3}$) and EC ($4.34 \pm 0.91$ $\mu gm^{-3}$) concentration. Clusters 3 and 4 form air masses are coming from north/northwest of the site, i.e. from Mumbai and central Indian region, and showed second highest concentrations of OC and EC. Cluster 5, a fast-moving cluster originated from the Middle East region, contributed about 11% of total back trajectories and was associated with the minimum OC ($8.96 \pm 3.07$ $\mu gm^{-3}$) and EC ($2.67 \pm 1.07$ $\mu gm^{-3}$) concentrations. Interestingly, the OC/EC ratio was estimated to be highest ($3.39 \pm 0.38$) for cluster 5 that showed the impact of distant sources which is indicative of the fact that the air masses are getting aged by travelling long distance before reaching the observational site.

Figure 7 describes the CWT analysis for OC and EC during the winter and summer seasons. As can be seen in the figure, the majority of the OC and EC concentrations during winter are contributed from the adjacent Mumbai (which is a metropolitan city) and the Telengana, Andhra Pradesh. During the summer season, Mumbai and Gujarat majorly contributed to the EC mass loading. The CWT analysis for OC reveals that OC is majorly contributed by the Mumbai region. Also, OC was contributed by the long-range transport from the Middle East regions. The analysis further emphasises that apart from the local man-made as well as natural sources, the possible regional transport from distant sources may significantly modulate the carbonaceous aerosol loading over the Mahabaleshwar region. Similar results for this location have been earlier reported by Mukherjee et al (2018), Yang et al (2019), and Meena et al (2021).

## Conclusions

A year-long study on temporal variations of organic and elemental carbon aerosols and their possible regional transport from distant sources revealed that:

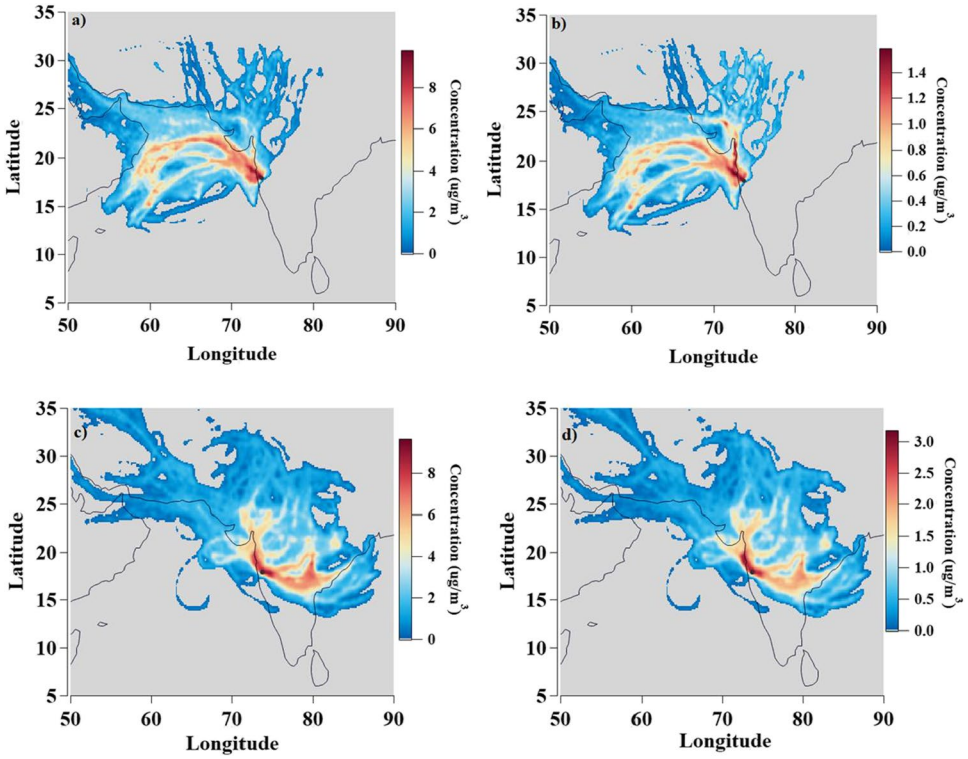


**Fig. 7** CWT analysis for OC and EC during **a**, **b** summer and **c**, **d** winter

1. Higher mass concentration of OC ($13.6 \pm 4.4$ μg/m$^3$) during summer and EC during winter ($3.6 \pm 1.1$ μg/m$^3$). OC/EC ratio more than doubled in summer than in winter. Secondary organic carbon formed a major fraction of OC during summer (> 60%) while in winter, both SOC and POC shared equal fractions of OC. Higher OC/EC ratio during summer ($7.4 \pm 4.4$) than during winter ($3.1 \pm 0.7$) also inferred towards the more presence of secondary carbon aerosols in summer.
2. EC contributed about 14% to total carbon in summer and 25% in winter. Primary carbon aerosols (POC and EC) together contributed 46% and 62% of total carbon in summer and winter, respectively, indicating the dominance of primary carbon aerosols in winter.
3. The high effective carbon ratio in summer (1.6) than in winter (0.7) showed the dominance of scattering type secondary carbonaceous aerosols in summer which are attributed to be from the local biogenic sources.
4. As observed from the cluster and concentrated weighted trajectory (CWT) analysis, apart from local anthropogenic and natural sources, the possible long-range transport from distant sources could be responsible for the observed concentrations of OC and EC at Mahabaleshwar indicated high concentrations of OC and EC in continental-originated trajectories. However, high OC/EC ratio was observed for air masses arriving from long-distance sources.

**Acknowledgements** The authors are thankful to the Director, IITM, for the encouragement and support to undertake this work. Also, the authors are thankful to scientific staff and engineers at the HACPL, Mahabaleshwar, for their help during the observations.

**Author contribution** P.S. Buchunde: conceptualization, data curation, formal analysis, writing—original draft. P.D. Safai: data curation, writing—review and editing. S Mukherjee: formal analysis, writing and editing. M.P. Raju: investigation, data curation. G.S. Meena: formal analysis. S. M. Sonbawne: formal analysis. K.K. Dani: resources, supervision. G. Pandithurai: project administration, supervision, writing—review and editing.

**Data availability** The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

## Declarations

**Competing interests** The authors declare no competing interests.

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