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RESEARCH PAPER

Vertical profile of organic and elemental carbon in sediments of Songkhla Lake, Thailand

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Abstract
In this study, a historical record of atmospheric deposition in the sediment cores from Songkhla Lake, the second largest lake in Southeast Asia, located in the southern part of Thailand is reported. It is well known that lake sediments, including spheroidal carbonaceous particles generated by both anthropogenic and natural emissions, contain records of lake, catchment, and atmospheric deposition histories. Vertical profiles of these carbonaceous particles can be used to investigate enormously influential disturbances, particularly those triggered by extreme paleo events, over large spatial areas. In this study, organic carbon/elemental carbon (OC/EC) ratios displayed unusually high values of 3.07 and 4.02 for depths 240 and 340 mm, respectively. Previous studies have attributed remarkably high values of OC/EC ratios to both biomass burnings and volcanic eruptions. Although anthropogenic emissions (e.g. fossil fuel combustions) can be responsible for relatively high levels of contamination, as expected, the existence of relatively low OC/EC ratios (i.e. 1.43 ± 0.30) for all sediment samples (except those collected at 240 and 340 mm depths) suggests a tropical background of these particles.

Keywords Organic carbon · Elemental carbon · Lake sediment · Songkhla lake

Introduction

Numerous studies have extensively investigated the physicochemical properties of organic carbon (OC) and elemental carbon (EC) in aerosols (Huang et al. 2013; Li et al. 2006; Pongpiachan et al. 2013, 2014a, b; Srivastava et al. 2014; Zhang et al. 2009, 2011), soils (Lal 2006;

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Möller et al. 2005; Raich and Schlesinger 1992), and sediments (Gacia et al. 2003; Hung et al. 2006; McCourt et al. 1996), especially in Asian countries over the past few years. Although previous reports have highlighted the importance of anthropogenic emissions of carbonaceous aerosols in different environmental compartments (Chen et al. 2005; Ito and Penner 2005), the influences of biomass burning, forest fires, and volcanic eruptions also play a major role in governing OC and EC contents (Birch and Cary 1996; Cachier et al. 1989; Ito and Penner 2005; Martinsson et al. 2009; Szidat et al. 2006). Recent studies suggest that there has been a tendency toward enhanced summer floods in south China, increased droughts in north China, and moderate cooling in China and India, despite the ambient air warming trends in other parts of the world (Menon et al. 2002; Ramanathan and Carmichael 2008; Yihui et al. 2007). Since heat-absorbing carbonaceous aerosols increase the air temperature and influence both regional atmospheric stability and vertical movements, the investigation of OC/EC ratios can assist the understanding of regional-scale circulation and hydrologic cycles with significant regional climate impacts for many reasons.

Firstly, OC/EC ratios have been used as chemical tracers for characterizing emission sources from vehicle exhausts. According to a busy roadway tunnel experiment in central Lisbon, OC/EC ratios in aerosol components were in the range of 0.3–0.4 (Pio et al. 2011). Similar OC/EC ratios were detected at the roadside in Birmingham, UK (Pio et al. 2011). The average OC/EC ratio in PM_{10} (i.e. particulate matter less than 10 microns), collected from seven air quality observatory sites in heavily polluted roadsides of Bangkok, was 0.99 ± 0.63, indicating that traffic emissions were responsible for relatively low OC/EC ratios (Pongpiachan et al. 2014b). Secondly, biomass and agricultural waste combustions play a crucial role in elevating the OC/EC ratios, as discussed in numerous studies (Cao et al. 2007; Gonçalves et al. 2011; Pongpiachan et al. 2009). For instance, the generated smoke aerosols were characterized by relatively high OC/EC ratios detected from controlled field burning of rice straw (10) and wood combustions (7.8) (Engling et al. 2009; Ram and Sarin 2010). Since OC/EC and char/soot ratios provide valuable insights for source identifications, many scientific reports have focussed on the chemical characterisation of carbonaceous compounds, particularly in lake sediments, which is exceedingly advantageous for deciphering historical trends related to biomass burnings/forest fires (Cong et al. 2013; Han et al. 2011). During El Niño-Southern Oscillation (ENSO) years, severe droughts provoke forest leaf-shedding and greater flammability, and thus forests become vulnerable to fire. Since numerous studies underline the strong correlation between the frequency of forest fires and ENSO (Nepstad et al. 1999; Schoennagel et al. 2005; Siegert et al. 2001), it appears reasonable to apply OC/EC ratios for reconstructing historical trends of forest fires in Southeast Asian regions.

A previous study also highlighted the importance of dissolved organic carbon (DOC) (i.e. water soluble organic carbon in aquatic ecosystems) as a key indicator for predicting and understanding the response of lake ecosystems to multiple threats such as acid rain, toxic heavy metals and hazardous persistent organic pollutants, enhancement in UV radiation, and climate change (Williamson et al. 1999). As a consequence of 20 years of global warming, drought and enhanced biomass burnings between 1970 and 1990 appear to be responsible for a DOC reduction of 15–25% in lakes of North-western Ontario, Canada (Schindler et al. 1997). It is also crucial to note that DOC is deeply connected with microbial metabolism, light climate, acidity, and primary production in lakes (Sobek et al. 2007). While the altitude, mean annual runoff, and precipitation were negatively correlated with lake DOC, the conductivity, soil carbon density, and soil C:N ratio were positively associated with lake DOC (Sobek et al. 2007). It has been suggested that increasing trends in DOC in the surface waters of glaciated landscapes across eastern North America and northern and central Europe between 1990 and 2004 can be briefly described by an elementary simulation based solely on variations in atmospheric deposition chemistry and catchment acid-sensitivity (Monteith et al. 2007). A similar rising trend of DOC contents in streams and lakes of the UK within a range of 8–42 years was also detected with the average annual enhancement in DOC content of 0.17 mg C l^{-1} $year^{-1}$(Worrall et al. 2004). Overall, it appears reasonable to assume that carbonaceous aerosols are deeply connected with DOC contents in lakes and reservoirs, underlining the impacts of particulate OC–EC on numerous stressors in aquatic ecosystems.

Despite a large number of research studies focusing on the computation of emission factors of carbonaceous particles released from different fuels and vehicle types (Alves et al. 2015; Shen et al. 2014; Wei et al. 2014), little is known about their past records in tropical sediments. To the best of our knowledge, there is no information available on the vertical profile of OC/EC ratio distributions in the lake sediments of Thailand. Overall, the main objectives of this study were to (1) generate novel insights into the nature of complex climate systems in Southeast Asian countries with some assistance from OC/EC ratio data; (2) obtain a vertical profile of total carbon (TC), OC, and EC for the Songkhla Lake sediments; and (3) quantify the OC/EC ratios and compare their values with previous combustion source studies.

Materials and methods

Study site

The Thale Noi Lake (TNL, located at 7° 46′00″N 100° 09′11″E), which is the largest lagoon lake in Thailand, is a protected freshwater wetland situated in Phatthalung Province and covers an area of over 460 km^2. TNL became regionally acknowledged as an ecosystem dynamic hotspot in 1975 when the Ministry of National Resources and Environment, in conjunction with the International Union for Conservation of Nature (IUCN), declared it a Protected Area Category III (Natural Monuments). TNL can be further separated into four subareas, namely Melaleuca forests (170 km^2), rice paddies (153 km^2), swamp (109 km^2), and open water (28 km^2). It is also important to highlight that TNL is positioned in the northern part of Thale Luang, Thale Sap, and Songkhla Lake. The area around the lake consists of farmland, forests, and swamps. There is no main river flowing through this area, but sediment loads from many small man-made canals as well as run-off water from the high steep mountains is observed (VKI 1997). The sediment core samples of TNL were collected from three sites (Fig. S1 and Table S1) and Fig. S2 shows the sampling method and collected sediment cores.

Sediment collection

Three uninterrupted sediment cores were obtained from the northern, central, and southern parts of the TNL in August 2017 when the water level ranged between 150 and 170 cm (see Figs. S1–S2). A gravity corer was lowered from a speedboat equipped with a transparent PVC plastic tube 12 cm in diameter 1.2 m in length. All materials used for core sectioning were washed carefully with detergent and water, and rinsed successively with methanol and dichloromethane prior to removing the frozen core from the freezer. The putty knife and spatulas were properly cleaned with tap water, methanol, and then dichloromethane. For handling and analysis, we strictly followed the standard operating procedure for the USGS Reston, Virginia Environmental Organic Geochemistry Laboratory Appendix 3 (https://water.usgs.gov/nrp/biogeochemical-processes-in-groundwater/forms/SOP_LMWOA_05272015_FINAL_Website.pdf).

The retrieved sediment columns were kept vertical during transport to the laboratory to avoid disturbance or damage to sediments. In this study, no physical evidence of bioturbation was detected in the retrieved sediment columns, indicating biological limitations in the TNL. For this study, as displayed in Fig. S1, only the No. 1 sediment core was selected for chemical analysis of OC/EC ratios. The core (580 mm) was precisely subdivided into a series of slices at 20-mm intervals. The 29 sediment sections (i.e. 580/20) were subsequently freeze-dried in order to remove the water content without greatly altering the physicochemical properties of the lake sediment. The samples were then passed through a 0.15-mm mesh sieve and kept in a refrigerator at − 20 °C in labelled zip lock bags for further OC/EC ratio analysis.

Analysis of OC and EC

The dried sediment samples were ground and homogenised with an agate mortar and sieved through a 200-mesh sieve. The sample pre-treatment procedure has been clearly described in previous studies (Han et al. 2007a, 2007b) and will not be discussed here (see Table S2 for more details). Entire sediment samples were quantitatively identified employing a DRI Model 2001 Thermal/Optical Carbon Analyser (Desert Research Institute, Division of Atmospheric Sciences 2215 Raggio Parkway Reno, NV 89,506) (Chow et al. 1993, 2001). The analytical instrument depends on the oxidation of OC and EC components under various heating conditions. Its operation depends on the fact that OC can be evaporated from the filter in a non-oxidising helium (He) atmosphere while EC has to be ignited by an oxidiser. The degree of decomposition brought about by high temperature can be calculated by repeatedly observing the filter reflectance and/or transmittance throughout an analysis cycle. The reflectance and transmittance, mainly affected by the existence of light absorbing EC, reduces as pyrolysis occurs and enhances as light-absorbing carbon is liberated over the subsequent process of the determination. By observing the amount of light transmitted by a sample (i.e. transmittance) and the amount of light that reflects from the surface of a sample (i.e. reflectance), the EC peak area is theoretically positively correlated with pyrolysed OC, which can be precisely converted to the OC fraction. The computation for the charring conversion of OC to EC is important for eliminating the bias in the detection of carbon components (Johnson et al. 1981). The charring corrections of thermal optical reflectance (TOR) and thermal optical transmittance (TOT) are not essentially identical, owing to charring of organic vapours adsorbed within the quartz fibre filter (Chow et al. 2004; Chen et al. 2004). All samples were analysed by a DRI Model 2001 Thermal/Optical Carbon Analyser (Atmoslytic Inc. Calabasas, CA). The operation of the DRI Model 2001 Thermal/Optical Carbon Analyser is based on the preferential oxidation of OC compounds and EC at different temperatures. Its function relies on the fact that organic compounds can be volatilised from the sample deposit in a non-oxidising He atmosphere while EC must be combusted by an oxidiser.

Probability distribution function (PDF) of carbonaceous sediments

The PDF was applied to TC, OC, and EC of sediments collected at the TNL. Normally, a PDF is an equation that explains the relative probability of a random parameter to take a given value. The probability for the random parameter to fall within a specific area is given by the Gaussian distribution, which can be described as follows:

$$y = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(\frac{-(x-\mu)^2}{2\sigma^2}\right), \quad (1)$$

where y, σ, σ^2, μ and x symbolise PDF, standard deviation, variance, arithmetic mean, and contents of carbonaceous compositions in lake sediments, respectively. In addition, Statistical Program for Social Sciences (SPSS) version 13 was used for simple linear regression analysis (SLRA), analysis of variance (ANOVA), and Pearson correlation analysis.

Estimation of secondary organic carbon (SOC) in PM_{10} collected at Hat-Yai City

Since numerous constraints can cause comparatively high OC/EC ratios in atmospheric deposits, it is important to perform further evaluation of SOC, which is generally related to the atmospheric long-range transportation (ALRT) process (Wang et al. 2012; Zhou et al. 2012). Secondary Organic Carbons (SOCs) are carbonaceous pollutants released from both natural and anthropogenic sources. SOCs are formed through a complex interaction of photo-oxidation, aqueous phase reaction, biogenic volatile organic compounds (BVOCs) from forests, plants, vehicles or imperfect combustions from industrial activities, and other particulate pollutants (Bessagnet et al. 2008; Claeys et al. 2004; Sartelet et al. 2018; Zhang et al. 2018). It is well known that SOCs can play a major role in governing gas-particle partitioning of persistent organic pollutants (POPs), which has been detected to cause lung cancers, respiratory problems and other adverse health impacts (Cocker et al. 2001; Odum et al. 1996; Pongpiachan et al. 2009, 2013). In this study, particulate carbonaceous contents were cited from a previous study that collected PM_{10} at two air quality observatory stations, namely Novotel Centara Hat-Yai Hotel (7°00′20.65″N 100°28′15.65″E) and Lee Gardens Plaza Hotel (7°00′21.39″N 100°28′15.94″E), which were situated at the centre of Hat-Yai City, Songkla Province (Pongpiachan et al. 2014a). The computation of SOC was conducted by applying the protocol proposed by Na et al. (2004). This method is based on the hypothesis that atmospheric deposits possessing the smallest OC/EC ratios constitute essentially primary carbonaceous compositions (Castro et al. 1999). For the atmospheric deposits observed at Hat-Yai city, the arithmetic mean of the three lowest OC/EC ratios was 8.47 and hence, these could be employed for the calculation of the SOC. It is also crucial to highlight that the three lowest OC/EC ratios were assumed to have solely primary OC and the impacts of small proportions of SOC was ignored. The content of SOC was estimated as follows:

$$OC_{sec} = OC_{tot} - EC \times (OC/EC)_{primary}, \quad (2)$$

where OC_{sec}, OC_{tot}, and $(OC/EC)_{primary}$ are SOC, TOC, and the arithmetic mean of the three lowest OC/EC ratios, respectively.

Results and discussion

Statistical descriptions of TC, OC, EC, and SOC detected during the sampling interval in the TNL are shown in Table 1. The arithmetic mean contents of TC, OC, EC, and SOC ranged from 178 to 1136 mg g^{-1}, 142 to 636 mg g^{-1}, 35 to 555 mg g^{-1}, and 27 to 520 mg g^{-1}, respectively. For percent contributions relative to TC mass, OC varied from 51 to 80% with an arithmetic mean of 60 ± 6.9%, whilst EC ranged from 20 to 49% with an arithmetic mean of 40 ± 6.9%. SOC differed from 15 to 46% contributing on average 37 ± 7.3%. In addition, the ANOVA test revealed a statistical difference between the mean values of OC (396 ± 126 mg g^{-1}) and EC (284 ± 125 mg g^{-1}). These results indicate that OC is the major chemical composition of the TC mass concentration. It is worth mentioning that three main features for the vertical profile of OC/EC ratio in the sediment core were observed. Firstly, three maximum peaks of TC were observed at sediment layer depths of 160–180 mm, 260–300 mm, and 500–520 mm, which were in good agreement with those of OC (see Fig. 1). Secondly, two maximum peaks of EC and SOC were detected at the same sediment layer depths of 480–500 mm and 500–520 mm. Thirdly, two

Table 1 Statistical descriptions of TC, OC, EC, SOC, and OC/EC ratios in sediment samples collected from the TNL

	TC [mg g^{-1}]	OC [mg g^{-1}]	EC [mg g^{-1}]	SOC [mg g^{-1}]	OC/EC
Average	679	396	284	260	1.59
SD	245	126	125	118	0.65
Min	178	142	35	27	1.02
Max	1136	636	555	520	4.02

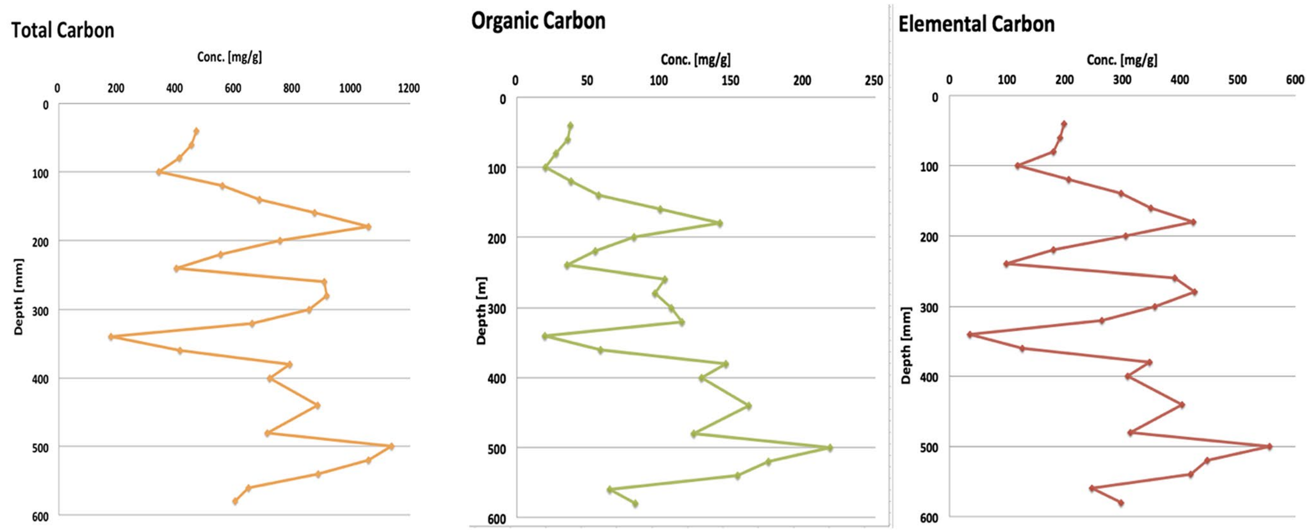

Fig. 1 Vertical profile of TC, OC, and EC in sediments collected from the TNL

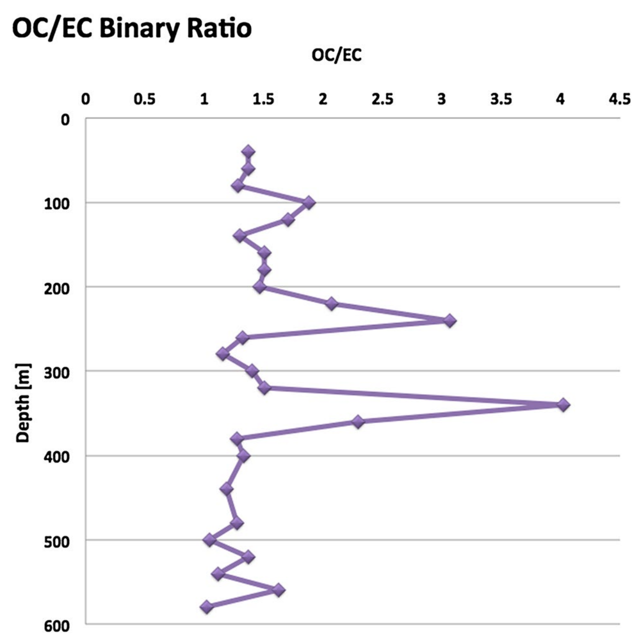

Fig. 2 Vertical profile of OC/EC ratios in sediments collected from the TNL

maximum peaks of OC/EC ratios were measured at sediment layer depths of 320–340 mm and 220–240 mm (Fig. 2).

OC/EC ratios and estimation of SOC

In this study, TC, OC and EC contents are assumed to be random parameters and remain unchanged during early diagenesis in sediments for numerous reasons. Diagenesis is the alteration of deposits or existing sedimentary rocks into various other sedimentary rocks (i.e. lithification), at temperatures and pressures less than that necessary for the creation of metamorphic rocks (Berner 1980). Since there are no hydrothermal vents or hot springs in the study sites, it appears reasonable to think that diagenesis can take place at Songkla Lake sediments. However, it is crucial to note that diagenesis excludes surface alteration and metamorphism. In other words, diagenesis does not include any changes from physical, chemical and biological weathering. As a consequence, it seems rational to conclude that diagenesis plays a minor role in governing the physicochemical properties of Songkla lake deposits, and thus TC, OC and EC concentrations should theoretically remain unchanged at the early stage of diagenesis in lacustrine deposits. One of the major factors associated with variations of carbonaceous contents in sediments is simply the chemical compositions of TC, OC, and EC contents originally contained in aerosols prior to its wet and/or dry deposition into Songkla Lake. In spite of the possible impacts of atmospheric depositions, biota living in the lake and in its watershed can be considered to be the crucial sources of the organic compounds initially contributed to the lake system. Microbial reprocessing in the middle of sinking and early sedimentation noticeably reduces the total amount of organic matter while substituting much of the primary organic matter with secondary organic matter (Meyers and Ishiwatari 1993). Therefore, much of the organic matter content of lacustrine sediments is the product of this microbiological decomposition. Numerous carbonaceous compounds of lake sediments still preserve information about their sources, and this helps us better understand regional paleolimnological conditions.

Over the last few years, OC/EC ratios have been comprehensively employed for interpreting the photo-oxidation processes of carbonaceous composition, formation of secondary organic aerosols (SOAs), and quantification of its potential sources (Gray et al. 1986; Turpin and Huntzicker 1995; Strader et al. 1999). In this study, as displayed in Table 1 and Fig. 3, OC/EC ratios ranged from 1.02 to 4.02 with an arithmetic mean of 1.59 ± 0.65. In order to categorise any plausible contributors of carbonaceous compositions, the average OC/EC ratio in the sediment core of the TNL was compared with a previous study on emission sources of carbonaceous aerosols as illustrated in Fig. 3 (Pongpiachan et al. 2013). It should be noted that the average OC/EC ratio of the TNL sediments was similar to those of Rubber factory and Traffic intersection (see Table 2 and Fig. 3), but lower than those of $PM_{2.5}$ collected at Chaumont, Switzerland (2.8), Guangzhou, China (2.8 ± 2.8), and Xi'an, China (2.9 ± 2.7) (Cao et al. 2003, 2005; Hueglin et al. 2005). The comparatively low average OC/EC ratio observed in the sediment core of the TNL reflects the impacts of agricultural waste burnings coupled with local vehicle releases in this area.

However, it is crucial to note that two maximum peaks of OC/EC ratios, detected at 320–340 mm and 220–240 mm sediment layers, were 4.02 and 3.07, respectively. The relatively high OC/EC ratios measured at these two peaks could be attributed to numerous causes. First, earlier investigations underline the significance of the generation of SOC via ALRT (Wang et al. 2012; Zhou et al. 2012). For example, the considerably high $PM_{2.5}$-bound OC/EC ratios (range 1.6–10.4; average 5.2 ± 1.8) detected at Mount Heng, China, were attributed to in-cloud SOA creation coupled with ALRT (Zhou et al. 2012). Second, both heterogeneous and homogeneous photochemical reactions of carbonaceous particles enhance dramatically during spring and summer, which were responsible for the comparatively high OC/EC ratios observed in the North China Plain (Wang et al. 2012). Since the TNL is located adjacent to the equator, it is reasonable to assume that the seasonal effect over the fluctuations of OC/EC ratios is of minor importance. Third, the unusually high OC/EC ratios can be ascribed to an extremely low EC value during the observation period. Unfortunately, this interpretation cannot be used to explain the relatively low OC/EC ratios (i.e. comparatively high EC) found in other sediment layers.

Further investigations on the formation of SOC were conducted by applying the Eq. 2. OC_{sec} concentrations and the percentage contributions of PM_{10} collected at Hat-Yai City, the nearest city to the sediment sampling sites: the values recorded were 3.96 ± 2.18 μg m^{-3} and 63 ± 25%, respectively. This arithmetic mean percentage contribution is considerably higher than that of Kaohsiung (40.0%, Lin and Tai 2001) and almost 3.7 times higher than that of Birmingham, United Kingdom (17%, Castro et al. 1999). This underlines

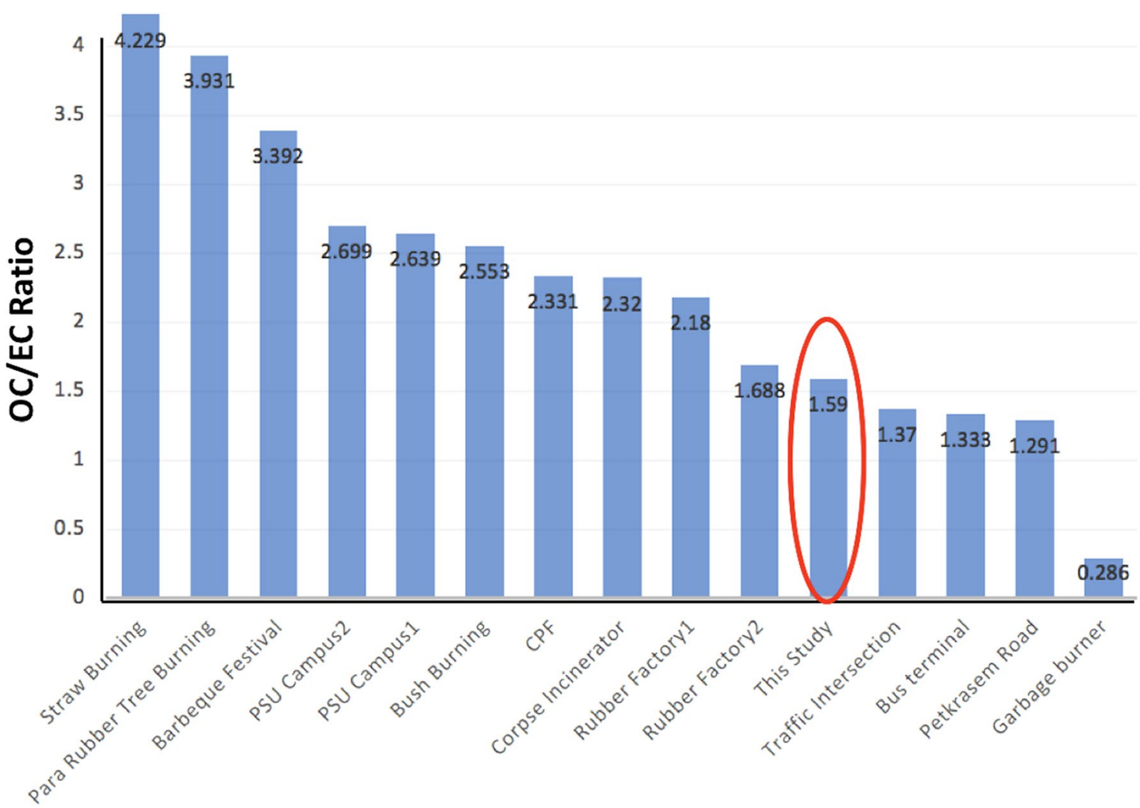

Fig. 3 OC/EC ratios in sediments collected from the TNL in comparison with those of various emission sources

Table 2 Statistical descriptions of TC, OC, EC, and OC/EC ratios in PM_{10} samples collected from various emission sources (Pongpiachan et al. 2013)

	PM_{10} [μg m^{-3}]	TC [μg m^{-3}]	OC [μg m^{-3}]	EC [μg m^{-3}]	OC/EC
PSU campus1	35.7 ± 10.3	6.671 ± 8.391	4.838 ± 5.650	1.833 ± 0.856	2.639
PSU campus2	27.9 ± 8.7	4.897 ± 3.640	3.573 ± 1.937	1.324 ± 0.673	2.699
Traffic intersection	46.9 ± 30.6	14.831 ± 37.439	8.572 ± 10.572	6.259 ± 13.786	1.370
Corpse incinerator	35.9 ± 28.7	7.485 ± 15.925	5.230 ± 5.854	2.254 ± 4.079	2.320
CPF	24.5 ± 5.4	7.385 ± 15.428	5.168 ± 4.448	2.217 ± 4.221	2.331
Songkla Lake1	13.8 ± 2.9	4.296 ± 2.000	3.063 ± 1.426	1.232 ± 0.268	2.486
Songkla Lake2	11.6 ± 3.7	0.977 ± 1.849	0.757 ± 0.397	0.221 ± 0.401	3.425
Rubber factory1	34.4 ± 8.6	15.829 ± 31.658	10.852 ± 17.158	4.977 ± 6.096	2.180
Rubber factory2	36.7 ± 15.7	11.022 ± 24.784	6.922 ± 10.449	4.100 ± 6.833	1.688
Bus terminal	42.8 ± 24.9	14.112 ± 29.391	8.062 ± 8.154	6.050 ± 11.015	1.333
Garbage burner	86.6 ± 65.4	24.449 ± 64.879	5.427 ± 11.266	19.023 ± 31.442	0.286
Barbeque festival	30.2 ± 14.1	6.280 ± 8.799	4.850 ± 2.611	1.430 ± 1.849	3.392
Petkrasem Road	25.1 ± 9.2	9.650 ± 22.896	5.438 ± 8.693	4.212 ± 7.385	1.291
Kor Hong Hill	9.6 ± 4.0	1.850 ± 1.224	1.352 ± 0.894	0.499 ± 0.055	2.709
Straw burning	217.8 ± 96.1	80.362 ± 91.936	64.992 ± 51.974	15.370 ± 12.574	4.229
Bush burning	25.5 ± 2.3	11.106 ± 10.069	7.980 ± 2.234	3.126 ± 2.696	2.553
Pará rubber tree burning	83.7 ± 23.2	48.390 ± 66.486	38.578 ± 35.154	9.812 ± 10.090	3.931

Prince of Songkla University (PSU): the sampling station is positioned at approximately 3 m above the basement of the Faculty of Environmental Management, Prince of Songkla University. This location is about 3 km away from the city centre and thus can be acknowledged as an urban residential zone

Traffic intersection (TI): this monitoring site is situated at the traffic intersection adjacent to Tesco Lotus department store in Hat-Yai City. TI can be considered as a representative of vehicular exhausts

Corpse incinerator (CI): the monitoring site is located at crematory of Kor-Hong Buddhist monastery adjacent to PSU and approximately 1.5 km away from TI. CI can be considered as a representative of both timber and tyre combustions

Charoen Phokphand factory (CPF): CPF is located inside the fish-can producing factory owned by Charoen Phokphand group. This site can be considered as a representative of crude oil combustion

Songkhla Lake (SL): SL is positioned at the coastal area of Songkhla Lake and is roughly 13 km away from Hat-Yai City. SL is also located approximately 14 km away from the western side of the Gulf of Thailand. This sampling site can be acknowledged as a rural background monitoring site

Rubber sheet manufacturing factory 1 (RMF1): RMF1 can be considered as a mixture of Pará rubber tree combustion coupled with emissions of latex fragments and sulphuric acid particles. This sampling station is situated at Tumbol Tungwan, Hat-Yai District, Songkla Province

Rubber sheet manufacturing factory 2 (RMF2): RMF2 can be considered as a mixture of Pará rubber tree combustion coupled with emissions of latex fragments and sulphuric acid particles. This sampling stations is situated at Tumbol Tachang, Banglum District, Songkla Province

Bus terminal (BT): this monitoring site is positioned at a bus terminal about 1.4 km away from PSU. BT was carefully chosen as a representative of diesel emissions since most of the buses are diesel-fuelled

Waste incinerator (WI): WI is a facility which is a part of the municipality of Hat-Yai City. This sampling station can be acknowledged as a mixture of solid wastes and diesel oil combustions

Barbeque festival (BF): BF is positioned at the centre of PSU campus on the rooftop of the Faculty of Natural Resources. PM_{10} samples were collected during the barbeque festival which is an annual tradition normally occurring in the second week of August. BF can be acknowledged as a representative of charcoal combustion

Petkrasem Road (PR): PR is situated close to Petkrasem Road at the city centre of Hat-Yai. This site can be considered as the most congested area of Songkla Province. As a consequence, PR can be considered as a representative of a mixture of diesel and benzene combustions

Kor-Hong Hill (KHH): KHH is located at the top of Kor-Hong hill with a height of 356 m. KHH can be regarded as a mixture of anthropogenic emissions from Hat-Yai City

Rice straw burning (RSB): RSB can be regarded as a representative of rice straw combustion. This sampling site is positioned at a rice paddy field in Satingpra District, Songkla Province

Biomass burning (BB): BB can be considered as a mixture of agricultural waste burnings at the planting areas of Namon District, Songkhla Province

Pará rubber tree burning (PTB): PTB can be regarded as an emission source of Pará rubber tree combustion the considerable impacts of atmospheric depositions from Hat-Yai City as the potential mechanism responsible for relatively high OC/EC ratios observed in the sediment layers at 320–340 mm and 220–240 mm depths. In spite of the overwhelming impacts from Hat-Yai City, it is also crucial to underline other plausible influences such as local biomass burnings and ALRT as alternative causes for comparatively high OC/EC ratios detected in the lake deposits. A previous study detected the radioactivity of isotope ^{137}Cs using a gamma-ray spectrometer in the Songkhla Lake sediments (Chittrakarn et al. 1996). Results of the analysis for ^{137}Cs in all 20 sediment cores show that the average sedimentation rate in Songkhla Lake, determined from each core, ranged from 0.0 to 8.7 mm year^{-1} with an arithmetic mean of 5.4 ± 0.2 mm year^{-1}. By using this sedimentation rate, the age of the two maximum peaks of OC/EC ratios could be indirectly quantified as 59–63 years and 41–44 years for the sediment layers at 320–340 mm and 220–240 mm depths, respectively.

As described in OC/EC ratios and estimation of SOC, the relatively high OC/EC ratios observed for the sediment layer depths of 320–340 mm and 220–240 mm are probably related to high OC rather than low EC values. There are three possible causes for this, i.e. human activities (e.g. traffic releases and factory emissions), biogenic emissions (e.g. forest fires, agricultural waste burnings, and plant wax), and ALRT of carbonaceous particles from outside of the TNL. The relative contributions of ALRT and local biogenic emissions can be evaluated by applying the SLRA for OC and EC concentrations to the lake sediments. If a larger part of OC in the atmospheric deposits of the TNL sediments were governed by local biomass burnings, the *R*-value of OC and EC should be low since EC is principally emitted from vehicular exhausts. In contrast, if *R*-values of OC and EC are high, it appears reasonable to assume that both were released instantly from a single source, namely traffic emissions (Chen et al. 2012).

SLRA

In order to test this hypothesis and to evaluate the influence of ALRT on maximum peaks of OC/EC ratios observed in the TNL, linear regression between OC and EC concentrations in each sediment layer was performed. As indicated in Figs. 4, 5 and 6, the SLRA of TC vs OC, TC vs EC and OC vs EC were plotted along with their respective slopes and intercepts. Generally, a comparatively high *R*-value ($R = 0.81$) coupled with a lower *p* value ($p < 0.0001$) was detected in the SLRA of OC vs EC in all sediment layers, indicating a single dominant contributor (plausibly traffic-associated emissions). In contrast, the lowest *R*-value ($R = 0.79$, $p < 0.0001$) was observed in the SLRA of TC vs

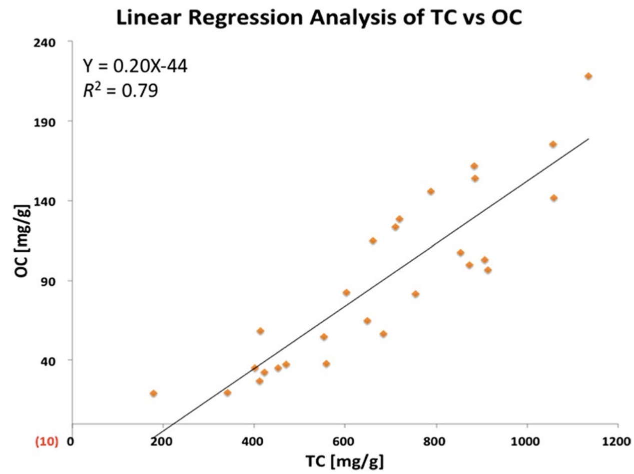

Fig. 4 Linear regression analysis of TC vs OC in sediments collected from the TNL

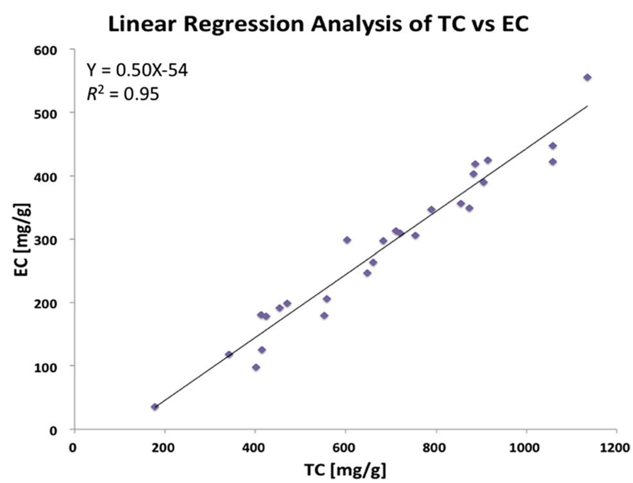

Fig. 5 Linear regression analysis of TC vs EC in sediments collected from the TNL

OC, while the highest *R*-value ($R = 0.95$, $p < 0.0001$) was detected in the SLRA of TC vs EC. These findings highlight the importance of vehicular exhausts, which are predominantly connected with EC emissions, as one of the main contributors of carbonaceous particles in the TNL sediments. These results also indicate that the unusually high OC/EC ratios measured at sediment layer depths of 320–340 mm and 220–240 mm are probably associated with non-traffic emissions, namely ALRT, rather than the impacts of local biomass burnings.

PDF

The PDF was applied to all TC, OC, and EC contents at 29 sediment sections as explained in Study site. PDF is a

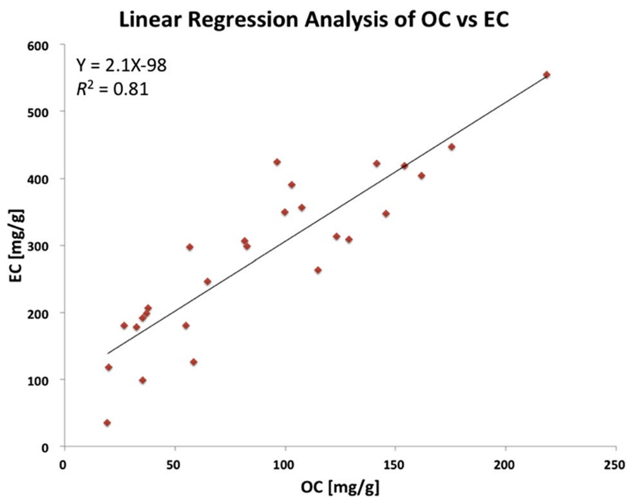

Fig. 6 Linear regression analysis of OC vs EC in sediments collected from the TNL

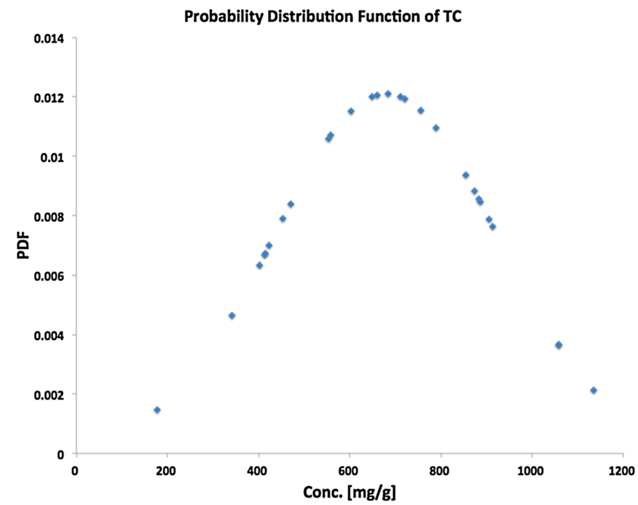

Fig. 7 PDF of TC in sediments collected from the TNL

function that describes the relative probability for a random parameter to assume a given value. The probability for the random variable to fall within a particular region is given by the Gaussian distribution as explained by Eq. 1. The PDF will give an idea of the vertical spatial distribution pattern of target compounds. It is important to underline that "vertical spatial distribution" indicates the distribution of target compounds along with numerous sediment depths not in the sense of distribution among several sampling sites. For instance, if PDF is normally distributed, this indicates a homogeneous distribution of chemical substances in Songkla Lake sediments. In the case of right skewness, this implies comparatively high inputs of chemical species in the study area. In contrast, if the PDF is skewed to the left, this underlines a potential mechanism of decay (e.g. microbiodegradation) of chemical species at the observation site. As shown in Figs. 7, 8 and 9, symmetrical bell-shaped curves were observed for all carbonaceous compositions. Since the detected values of the PDF are more concentrated in the middle than in the tails, it seems rational to attribute it to moderately homogeneous spatial distribution of carbonaceous compositions in the background lake sediments that were less likely to be influenced by extreme events (e.g. forest fires and volcanic eruptions). Previous studies have highlighted the significance of large-scale forest fires and volcanic eruptions on fluctuations of carbonaceous compositions in atmospheric deposits (Bhugwant et al. 2000; Cachier et al. 1989; Lavoué et al. 2000; Martinsson et al. 2009; Pio et al. 2008). A previous study suggested that volcanic eruptions (Piton de la Fournaise, 2632 m above sea level) do not emit EC directly, and during the air quality observation period from 10 to 28 March 1998, no significant vegetation fires ignited by the lava were observed (Bhugwant et al. 2000). An exceedingly high OC/EC ratio of 5.08 was also observed during the intense forest fire episode that occurred during the summer of 2003 in the Aveiro region, Portugal (Pio et al. 2008). Since no asymmetrical distribution curves for the carbonaceous compositions were observed in the sediment cores of the TNL, it appears reasonable to assume that atmospheric deposits in the TNL were mainly released from a single dominant source, probably vehicular exhaust. Nevertheless, it is safe to mention that the exceedingly high OC/EC ratios detected at the sediment layer depths of 320–340 mm and 220–240 mm are plausibly related to some extreme episodes such as forest fires and/or volcanic eruptions.

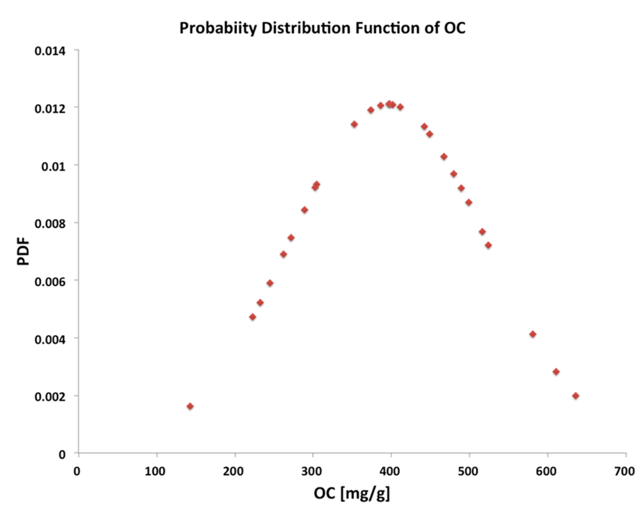

Fig. 8 PDF of OC in sediments collected from the TNL

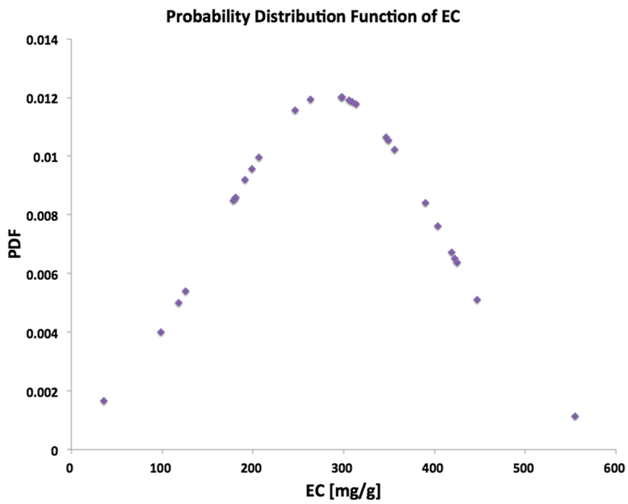

Fig. 9 PDF of EC in sediments collected from the TNL

Conclusions

The analyses of OC/EC ratios, OC_{soc}, SLRA, and PDF reveal that traffic emissions are the most influential factor controlling the atmospheric deposits of carbonaceous compositions observed in the TNL core sediments. Although vehicular exhausts play an important role in governing carbonaceous compositions of most sedimentary samples, multiple types of extreme events, including the ALRT of forest fire particulate matter and smoke from volcanic eruptions, seem to be the principal contributors detected at 320–340 mm and 220–240 mm sediment layers. By applying a sedimentation rate of 5.4 ± 0.2 mm year^{-1}, the age of the two extreme events could be roughly estimated as 59–63 years and 41–44 years, respectively. Irrespective of some degree of uncertainty resulting from sediment age dating techniques, the overwhelming contribution of traffic releases to carbonaceous deposits is unquestionably evident. These findings also provide evidence for considerable concerns regarding ecotoxicology and environmental safety of communities surrounding the Songkhla Lake, and thus are likely to encourage policy makers to develop realistic plans for the reduction of traffic related pollutants, especially under the context of sustainable development.

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