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CHARACTERISTICS OF OC/EC IN $PM_{2.5}$ OBSERVED IN A MOUNTAINOUS SUBURB OF MEGACITY CHONGQING

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ABSTRACT

Mass concentrations of organic carbon (OC) and elemental carbon (EC) were measured during winter of 2014 and summer of 2015 in the mountainous Fuling District of megacity Chongqing to understand the Carbon Aerosol in $PM_{2.5}$. Results showed that the average concentration of OC was 8.22 $\mu g \cdot m^{-3}$ in the summer and 30.61$\mu g \cdot m^{-3}$ in the winter. The average concentration of EC was 2.95$\mu g \cdot m^{-3}$ and 12.39$\mu g \cdot m^{-3}$, in summer and winter, respectively. Both OC and EC concentrations in winter were higher than those in summer. The ratios of OC/EC for both seasons exceeded 2 with values of 3.18 and 2.97 in summer and winter, respectively, indicating the existence of the secondary pollution. By the tracer method of EC, the estimated concentration of the secondary organic carbon (SOC) was between 0.96~7.76$\mu g \cdot m^{-3}$ with an average of 3.59 $\mu g \cdot m^{-3}$, and accounted for 45.43%of the total OC in summer. In contrast, the SOC concentration in winter was between -0.04~29.07$\mu g \cdot m^{-3}$ and averaged at 11.16$\mu g \cdot m^{-3}$ and 41.64% of the total OC. The high concentration of SOC in winter is probably due to the cloudy weather in the mountain region, which is not conducive for pollutant spreading and/or diffusion.

KEYWORDS:
Organic carbon, Element carbon, Secondary organic carbon

INTRODUCTION

Many researches have been conducted in-depth on $PM_{2.5}$ and PM10, and have made great achievements in regional pollution characteristics [1-4]. The organic carbon (OC) and elemental carbon (EC) in the atmosphere $PM_{2.5}$ have been studied since the 1970s.At present, the carbonaceous components are considered an important target for understanding atmospheric pollution and climate change [4-6]. Some scholars reported on the natural disasters of flood and drought in the South and North China [7], respectively. The phenomenon associated with the slow warming in China and India in recent decades may be related to the increase of EC concentration in the atmospheric aerosols. It was found that the carbonaceous aerosol concentrations in Europe from the west to the east were lower in winter than in summer, except for the sampling points showing an opposite trend at the western and eastern ends, and that all EC was contributed mainly from the burning of fossil fuels, as well as that OC was principally from the burning of fossil fuels and biomass in winter and the secondary organic carbon (SOC) in summer [8]. Similarly, throughout the United States, EC was mainly from the burning of fossil fuels and OC primarily from the combustion of vegetation, biofuels, and fossil fuel as well as forest fire [9]. It was found that carbonaceous aerosols are the main components of urban atmospheric aerosols, accounting for about 20%~90%. Past studies [10] showed that carbonaceous aerosols in India Kanpur urban area accounted for 50% of the mass concentration of $PM_{2.5}$. The research on carbonaceous components in China started relatively late, and mainly concentrated in large cities in eastern China, only a few in the western part of China. Studies on pollution characteristics of carbonaceous aerosols in Beijing [11, 12], Shanghai [13-15], Tianjin [16], Chongqing [17-19] in China, showed that EC and OC were enriched in fine particles and that their concentration levels were higher in winter than in summer because of the increasing amount of burning coal, lower atmospheric mixing layer, and less atmospheric deposition in winter. Some researchers [20] studied the distribution characteristics of atmospheric $PM_{2.5}$, OC, and EC concentrations in 14 cities, including Beijing, Jinchang, Qingdao, Changchun, Tianjin, Xi'an and Yulin in the northern China and Chongqing, Guangzhou, Hongkong, Hangzhou, Shanghai, Wuhan and Xiamen in southern China. Their results showed that carbonaceous aerosol concentrations were usually higher in the northern cities than in the southern ones, and in the inland cities than in the coastal ones, and that the energy structure and topographic and climatic conditions had important effects on the concentration levels.

This paper analyzes the characteristics of carbonaceous aerosol in the Fuling district, a mountainous suburb of Chongqing, the largest megacity in western China. Fuling district is in the hinterland of the Three Gorges reservoir and has the largest shale

gas field in China. With the extensive construction investment, rapid economic development, and fast-expanding industrial scales, transportations, and populations, the $PM_{2.5}$ concentrations, particularly the contents of OC and EC in $PM_{2.5}$, are largely increasing and have an important impact on local air quality, climate, visibility, and human health. Notably, due to vast undulating hills and little plains and platforms with high static wind rate, which is not conducive for pollutants to spread but extremely easy to cause haze, the investigation on the characteristics of OC and EC in Fuling is of significance for understanding their distribution and transport.

MATERIALS AND METHODS

The sampling site in Fuling is located in the campus of Yangtze Normal University, away from the traffic road about 200m, facing busy city avenue in its east and north sides and surrounding by commercial stores, small restaurant, and residential apartments, and affected by certain pollutant sources of dust and traffic emission, as well as catering and household waste. The particle sampler was placed on the roof platform of the university administrative building (about 30 m high). The sample data were collected for the winter (January 6 to February 5, 2015) and summer seasons (July 1 to July 31, 2015).

The sampling and analysis method of organic carbon and elemental carbon samples was based on the carbon aerosol analyzer of the American Desert Research Institute. Multiple wavelength measurements can be used to estimate the optical absorption properties of black carbon (BC) and brown carbon (BrC) and aerosol. Black carbon absorbs infrared and near-infrared wavelengths, while brown carbon has strong absorption in short-wavelength (<600 nm).

RESULTS AND DISCUSSION

The change of OC concentrations in summer and winter. The observed diurnal variations of OC concentrations during the summer and winter seasons in Fuling are shown in Fig.1. In the summer, due to strong sunshine and high temperatures, as well as more precipitation and good diffusion conditions, the OC concentrations were relatively low. In contrast, the concentrations in the wintertime were much higher because of less sunshine, significantly decreased wind speed, less precipitation, and worse diffusion conditions, coupled with significantly increased coal and gas combustion for heating and vehicle fuel consumption for heavy traffic.

In the summer, the variation range of daily OC concentration in $PM_{2.5}$ was 2.24-16.86μg·m^{-3} with an average of 7.95μg·m^{-3}. The daily OC during 6-12 and 16-21 July 2015 was relatively small, varied from 6.06 to 11.76 μg·m^{-3} and from 8.69 to 11.21 μg·m^{-3} with an average of 9.15μg·m^{-3} and 9.85μg·m^{-3}, respectively. The lowest value of 2.24μg·m^{-3} appeared on 4 July and low values around 1-5 July 2015, mainly influenced by the weather conditions of the sporadic showers and better visibility, which was favorable for the pollutants to dilute and spread easily. The maximum concentrations were observed during 12-16 July 2015, varied from 5.11 to 16.86μg·m^{-3} with an average of 9.88μg·m^{-3} and the highest value of 16.86μg·m^{-3}, mainly due to increasing vehicle emissions in the working days together with stable atmospheric stratification, which was not conducive to the diffusion of pollutants. The daily changes were larger between 21-24 July 2015 in the range of 6.03~13.75μg·m^{-3} and an average of 9.61μg·m^{-3}.

In the wintertime between 6 January and 5 February of 2015, the daily OC concentrations were 5.77~70.59μg·m^{-3}, averaged at 30.61μg·m^{-3}. The OC changes were obvious during 14-26 January 2015 in the range of 30.98~70.59μg·m^{-3} averaged at 48.64μg·m^{-3}, mainly due to increasing human activities on workdays in the overcast winter cloudy sky, which had weak atmospheric transport and made the emitted pollutants not easy to spread. In contrast, the OC variations for the time periods of 6-12 January and of 27 January-5 February 2015 were relatively small, with a range of 12.68~32.22μg·m^{-3} and of 5.77~24.34μg·m^{-3}, respectively. The former period had cloudy weather with a gentle breeze of the Beaufort wind force scale less than 3 (wind speed less than. 3.4-5.4ms^{-1}), which was not conducive for pollutants to diffuse and led to very small OC variation amplitudes, as shown in Fig. 1. In comparison, the later period mainly had sunny weather with significantly reduced human activities during the school winter break and Chinese New holidays, resulting in the reduction of pollutant emissions and relatively small OC concentrations averaged only at 14.09μg·m^{-3}, and the air quality improved significantly. Especially on 27-31 January, the OC concentrations suddenly decreased to the lowest values of the winter times due to light rains washing the pollutants out of the atmosphere.

The change of EC concentrations in summer and winter. The daily EC concentrations in Fuling in the summer and the winter are shown in Fig.2. The variation trends of EC in both seasons were like those of OC, respectively, EC concentrations were relatively low in the summer, but higher in the winter due to the different conditions of weather and pollutants emissions.

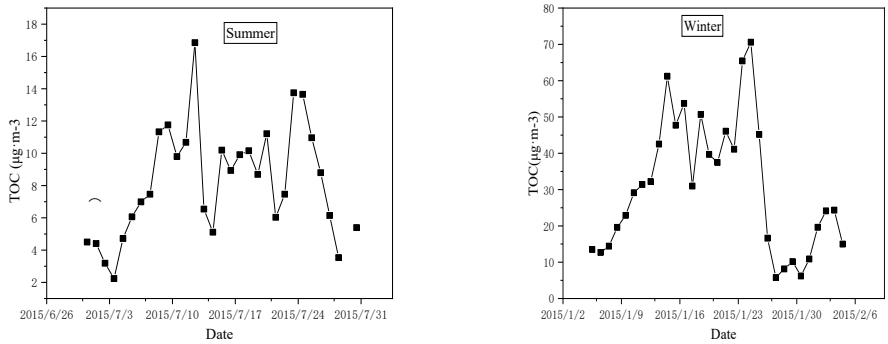

FIGURE 1
Daily change of TOC concentration in summer (left) and winter (right)

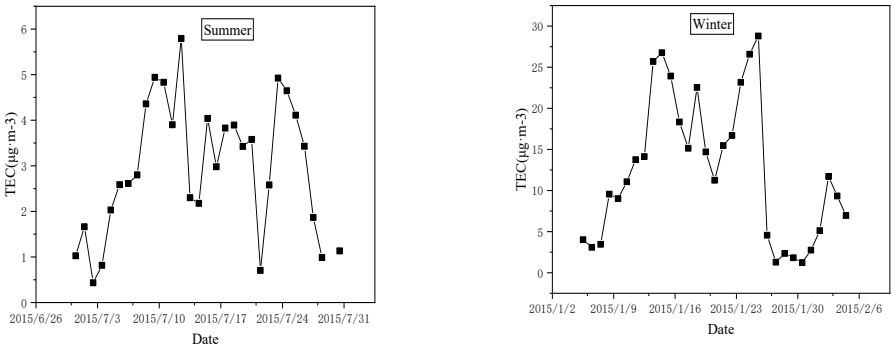

FIGURE 2
Daily change of EC concentration in summer (left) and winter (right)

In the summer, the EC concentrations varied from 0.82 to 5.79 $\mu g \cdot m^{-3}$ with an average of 2.95$\mu g \cdot m^{-3}$. The EC concentrations during 9-13, 16-21, and 24-27 of July 2015 were relatively high, varying from 2.98 to 5.79$\mu g \cdot m^{-3}$ and averaged at 4.18$\mu g \cdot m^{-3}$. The lowest value of 0.82$\mu g \cdot m^{-3}$ was seen on 4 July 2015 due to relatively slow human activity and small traffic volume, coupled to strong precipitation conducive for the pollutants to diffuse. The EC concentrations on 12-14 and 21-24 July 2015 were obviously larger, varied from 0.7 to 5.79$\mu g \cdot m^{-3}$, and averaged at 3.4$\mu g \cdot m^{-3}$. Both EC and OC had the peak values of 5.79$\mu g \cdot m^{-3}$ and 16.86$\mu g \cdot m^{-3}$, respectively, on the same day of 13 July 2015 due to the weather conditions and emission sources. In the winter, the EC concentrations were 1.28~28.80$\mu g \cdot m^{-3}$ with an average of 12.39$\mu g \cdot m^{-3}$. During 14 – 26 January 2015 the changes of EC concentrations were in the range of 11.24~28.80$\mu g \cdot m^{-3}$, more obvious and like those of the OC concentrations. Especially on 24-26 January, the EC concentrations were the highest at 23.16~28.80$\mu g \cdot m^{-3}$ due to increasing human activities and cloudy weather around the Christmas holidays. In contrast, on6-15Januaryof 2015, the daily EC concentrations were 4.01~26.76$\mu g \cdot m^{-3}$ and increased gradually during the time frame when the weather became cloudy and the wind speed decreased below the Beaufort scale 3 in favor of pollutants accumulation. From 28 January to 1 February of 2015, the daily EC concentrations reached their lowest values of 1.23 ~ 2.75 $\mu g \cdot m^{-3}$, due to significantly decreasing human activities during the winter school break.

The changes of OC/EC ratio in summer and winter. The previous research generally shows three sources of carbon particles in the atmosphere: combustion of coal, motor vehicle emissions, and biomass burning. The EC mainly comes from the primary emissions of the pollution sources, but OC also includes the secondary aerosols generated from the primary organic ones through atmospheric chemical reactions [21]. Without considering the secondary contributions to the OC, the ratio of OC/EC can reflect, to a certain extent, the source distributions of carbon components containing the aerosol particles. As shown in Fig. 3, the OC/EC ratios in the two seasons of 2015 in Fuling were more than 2, indicating the existence of the secondary pollutions. (More discussions in Section 3.4). Fig.3 also shows that the values and variability of the OC/EC ratios in the winter were generally greater than those in the summer.

In the summer, the OC/EC values varied largely from 2.03 to 8.57 and averaged at 3.18. In

comparison with the monthly mean value, the OC/EC ratios of 3.29 to 4.76 averaged at 3.88 on 28-31 July 2015 was higher, while the ratios of 2.03~3.13 averaged at 2.65 on 4-21 and 23-27 July was low with a minimum of 2.03 (the OC and EC were 11.76μg·m^{-3}and 4.94μg·m^{-3}, respectively) on 11 July. The large OC/EC fluctuations are probably due to two opposite effects: the strong sunshine and high temperatures in summer are favorable for the production of the secondary aerosols, while frequent precipitation processes and good diffusion conditions can significantly reduce the EC and OC accumulation. In the winter, the OC/EC ratios were relatively stable in the first part of the period, but exhibited large fluctuations of 1.57~5.59 in the second part, with an average of 2.97. During 9-20 and 22-26 January, as well as 3-5 February, the OC/EC ratios were between 1.57 and 2.93 and averaged at 2.31, lower than the monthly mean value. In contrast, on 6-8 January and 27 January-2 February, the larger OC/EC ratios were between 3.36~5.59 and averaged at 4.17. The features are thought to be associated with obviously weak sunshine and worse diffusive condition in winter and overlapped with increasing OC/EC due to a large amount of coal combustion during the winter heating season and reduction of anthropogenic activities around school and spring holidays, which made the OC concentrations to 5.77~19.60μg·m^{-3} while the EC concentrations at 1.28~5.12μg·m^{-3}.

The secondary organic carbon in summer and winter. The secondary organic aerosol (SOA) is referred to as the organic matter produced by the photochemical oxidation of the volatile organic compounds (VOCs) in the atmosphere. SOA is an important part of particulate organic matter. And because of its strong water absorption, optical properties, and chemical reactivity, SOA plays an important role in regional air quality, human health, and global climate change. However, due to the complexity and variety of VOCs, which are the precursors of SOA, as well as very complicated chemical reactions of VOCs in the atmosphere, our understanding of SOA is still very limited, even the estimation of its total volume has great errors. Besides, the generated SOA also participates in other atmospheric chemical processes.

Now, there is no direct analysis method that can distinguish the primary and secondary organic compounds in aerosol [22], but the only indirect method is available. The OC/EC ratio method is simple and straightforward and is commonly used. According to this method, the OC/EC ratio for the primary emissions discharged from pollution sources is relatively stable with some characteristic values determined by the types of pollution sources. The OC/EC ratios exceeding these critical values imply the formation of secondary organic carbon (SOC) [23].

According to this principle, Turpin and other [24] put forward the calculation method of SOC:

$$SOC=TOC - EC \times (OC/EC)_{PRI}, \quad (1)$$

Where TOC represents the total organic carbon (OC/EC), PRI represents the average value of OC/EC source, EC * $(OC/EC)_{PRI}$ indicates an organic carbon content. However, the establishment of $(OC/EC)_{PRI}$ is not easy but requires to grasp the regional emission characteristics of various pollution sources and to consider the impact of the daily change of source emission and seasonal fluctuations as well as meteorological conditions. In view of this, Castro [25] proposed to estimate the SOC according to the lowest value of OC/EC:

$$SOC=TOC - EC \times (OC/EC)_{min}. \quad (2)$$

$(OC/EC)_{min}$ usually appeared in certain weather conditions, such as low temperature, rain, and air mass instability. In these cases, the photochemical activity is low and not conducive to the generation of secondary organic matter, and OC is almost entirely disposable. Chow [26] proposed that OC/EC>2implies the SOC generation. The larger the OC/EC value is, the more serious secondary pollution occurs with SOC of larger fraction and more importance in the carbon components.

Among the sampled 62 days of 2014-2015 winter and summer, the OC/EC values of 60 days were

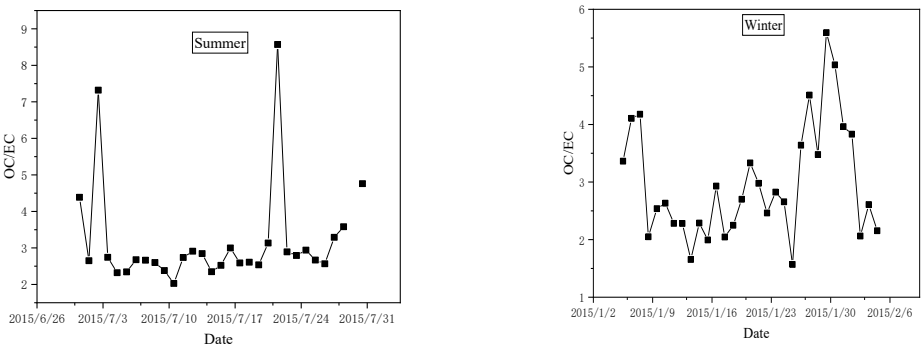

FIGURE 3
Daily change of OC/EC ratio in summer (left) and winter (right)

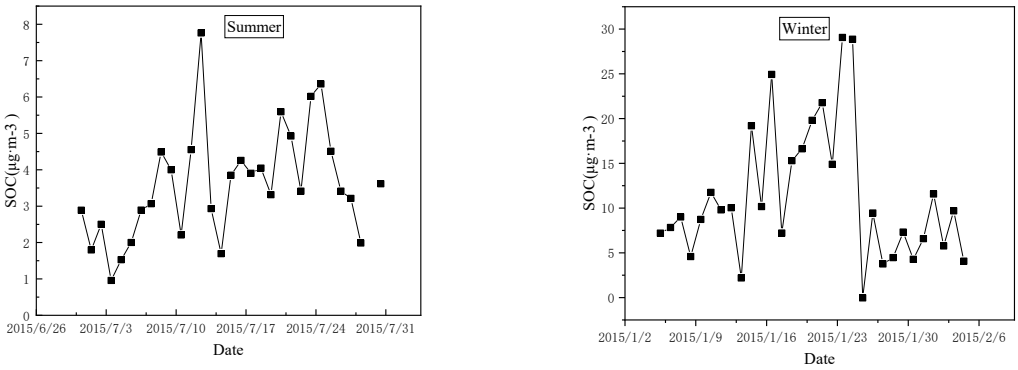

FIGURE 4
Daily change of SOC concentration in summer (left) and winter (right)

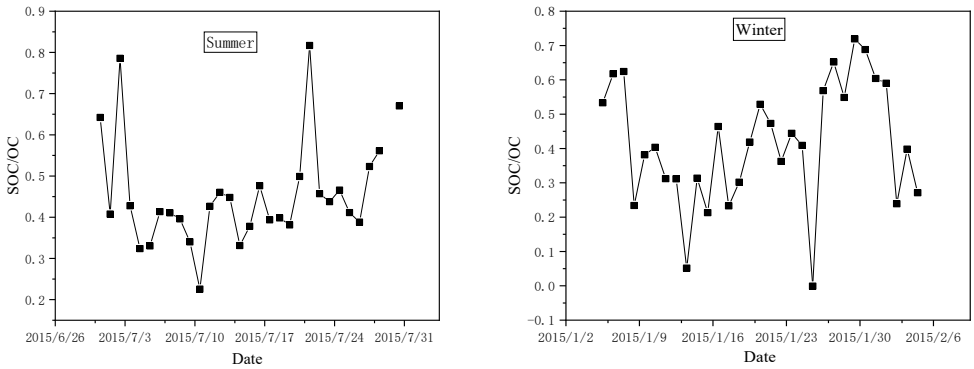

FIGURE 5
The daily change of SOC/OC in summer and winter

greater than 2, accounting for 97% of the total observation days, implying the existence of the secondary organic carbon in the aerosols observed in Fuling. The minimum OC/EC value was 1.57 during the 62 days. Fig.5 shows the daily SOC concentrations in summer and winter calculated by $(OC/EC)_{min}$ =1.57. The SOC concentrations in the winter were seen to be greater than those in the summer.

In the summer, the SOC concentrations were between 0.96 and 7.76μg·m^{-3} with an average of 3.59μg·m^{-3}. On 1-8 July 2015, SOC concentrations remained low, ranged from 0.96 to 3.07μg·m^{-3} and averaged at 2.20μg·m^{-3}. The minimum value 0.96μg·m^{-3} appeared on 4 July, the day with heavy rain and less solar radiation which were not favorable conditions for SOC generation. On 9-17 July, the SOC concentrations changed greatly from 1.69 to 7.76μg·m^{-3} and averaged at 3.97μg·m^{-3}. The maximum value of 7.76μg·m-3 occurred on 13 July. The sunny sky and high temperature with stable atmospheric stratification and low wind speed during this period allowed the primary pollutants accumulated in the atmosphere to generate more SOC.

In the winter, the SOC concentrations were between 0 and 29.07μg·m^{-3} with an average of 11.16μg·m^{-3}. On 19-26 January, the daily SOC concentrations varied significantly, with relatively stable high values of 15.30~21.78μg·m^{-3} and an averaged of 18.38μg·m^{-3} on 19-22 January, and large fluctuations of 0 to 29.07μg·m-3 and an average of 12.72 μg·m-3 on 22-26 January. The highest value of 29.07μg·m^{-3} reached on 24 January, while the lowest value of 0 μg·m^{-3} appeared on 26 January. The high and largely variable SOC concentrations are attributed to the weather conditions and human activities around the school and national holidays, as mentioned in the previous sections. In contrast, the SOC on 6-19 January and 27 January – 5 February were lower with smaller variabilities.

Fig.5 exhibits the day-to-day change of the SOC/OC ratio in the summer and winter. In the summer, the SOC/OC fluctuated from 22.55% to 81.68% with an average of 45.43%. The SOC/OC ratios varied around 40% on most of the days, in particular, below 40% on 4-21 and 23-31 July, and above 40% with a local maximum of 78.55% on 1-4 July. In the winter, the SOC/OC ratios showed large fluctuations from 0% ~ 71.94% with an average of 41.64%. The minimum and maximum occurred on 26 and 30 January, respectively. The features are thought to be determined by different weather conditions and human activities, as discussed in Section 3.3.

CONCLUSION

The conclusions are as follows:

(1) The averaged OC and EC concentrations over the observation periods showed higher values in the winter than in the summer. The mean summer OC and EC are 8.22μg·m^{-3} and 2.95μg·m^{-3}, while the mean winter OC and EC are 30.61μg·m^{-3} and 12.39μg·m^{-3}. The winter values are larger by a factor of ~4.

(2) The average OC/EC ratios are higher in the summer than in the winter, with the values of 3.18 and 2.97 respectively. This was due to the seasonal change of EC (12.39/2.95 = 4.20) that was larger than that of OC (30.61 /8.22=3.72), resulting in a smaller OC/EC ratio in the winter. However, the daily concentrations of OC and EC have similar variation trends, showing a good linear correlation, indicating their sources were common to some extent. The main sources are associated with vehicle and restaurant emissions around the sampling site.

(3) The average SOC concentrations are estimated at 3.59 μg·m^{-3} and 11.16 μg·m^{-3} for the summer and winter, respectively. It accounted for 45.43% and 41.64% of the total OC, indicating very serious secondary organic pollutants in both seasons in Fuling.

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