

# Photocatalytic removal of xylene as a pollutant in the air using ZnO-activated carbon, TiO<sub>2</sub>-activated carbon, and TiO<sub>2</sub>/ZnO-activated carbon nanocomposites

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## Abstract

**Background:** Today, advances in different areas of science and technology along with their application in industries have led to an increase in dangerous pollutants which can resist biodegradation. Volatile organic compounds (VOCs) are regarded as important factors of air pollution in closed environments. Xylene is one of these compounds which is produced in mass quantities and widely used in industries, therefore, the removal of this compound is necessary. One of the available technologies for removing this compound is photocatalytic degradation. The present study aimed to determine the efficiency of photocatalytic removal of xylene as a pollutant in air using TiO<sub>2</sub>-ZnO nanoparticles and TiO<sub>2</sub>-ZnO composite coated on activated carbon under ultraviolet radiation.

**Methods:** In this experimental study, after coating of the nanoparticles on activated carbon, the produced catalysts with a specific surface area were characterized using Brunauer-Emmett-Teller (BET) surface area and porosity analysis, scanning electron microscope (SEM), and the type and percentage of the main elements present in the bed were determined using energy dispersive x-ray spectroscopy (EDS). The tests were carried out at laboratory scale and ambient temperature. In order to produce polluted air containing 100 ppm xylene vapor at a specific flow rate and concentration, a dynamic concentrator system was used. The removal of xylene was investigated under continuous flow mode.

**Results:** The results of the specific surface area using BET analysis and SEM images showed that nanoparticles were well coated on activated carbon. According to the results of the photocatalytic removal, the efficiencies of photocatalytic removal of xylene by AC/ZnO 5%, AC/TiO<sub>2</sub> 15%, and AC/3TiO<sub>2</sub>/1ZnO were 80.1, 89, and 95.1%, respectively.

**Conclusion:** According to the results, the use of ZnO-TiO<sub>2</sub> nanocomposite on activated carbon can be an appropriate method for the photocatalytic removal of xylene from polluted air.

**Keywords:** Xylene, Nanocomposites, Titanium dioxide, Zinc oxide, Air pollution

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## Introduction

Volatile organic compounds (VOCs) are among those pollutants which have attracted the attention of researchers. In the atmosphere, over 500 organic compounds have been identified (1). Due to high vapor pressure (above 0.1 mm Hg), they evaporate easily and enter the atmosphere. These compounds can spread across a wide area, contaminating water, dust, and air (2,3). With the rapid growth of industrial activities across the world, especially in developing countries, the emission of such pollutants in the environment has increased and created some problems in limiting the emission of industrial pollutants within the

national borders of countries (4). The above-mentioned compounds pose a threat to not only the environment, but also to human health, even at low concentrations (5). They lead to the outbreak of problems, such as burning eyes, liver damage, mutagenesis, and cancer (6).

One of these VOCs is xylene with the formula C<sub>8</sub>H<sub>10</sub>, a sweet-tasting, highly flammable and colorless aromatic hydrocarbon. Xylene is thought to be involved in creating disorders in the central nervous system and according to the NIOSH classification, it falls into the category of group E, which are suspected of carcinogenicity. Its threshold limit value is 100 ppm (7,8). Xylene available



in the air around cities and industrial areas is a result of using fossil fuels, industrial processes, and traffic. The problems occurring as a result of being exposed to these compounds have attracted the attention of researchers to this dangerous substance (9).

Due to the effect of VOCs on humans and the environment, developing strategies for reducing organic vapors is a very important issue (10). One of the most important and common methods used to control VOCs is photocatalytic degradation (11,12). In the photocatalytic degradation of VOCs, semiconductors such as zinc oxide are used for carrying the photon of oxidation process in order to convert VOCs to water and carbon dioxide (13-15). Due to its low cost and safety, this method is appropriate for removing organic compounds at low concentrations (16). The coating of photocatalysts on an absorbent enhances the removal efficiency of pollutants to a great extent (17,18). Activated carbon is one of various absorbers which is widely employed in industries to remove VOCs from polluted air due to its ease of use, low operational cost, and the possibility of efficient recycling of VOCs. The main features of activated carbon are high strength, high mechanical strength, good absorption, high purity, smooth surface, and low pressure drop. This type of carbon is widely used for removing VOCs like hydrocarbon, solvents, toxic, and organic gases (19,20).

The most common photocatalysts are  $\text{TiO}_2$ ,  $\text{WO}_3$ , and  $\text{ZnO}$  (21-23).  $\text{TiO}_2$  particles with high energy photons elevate electrons from valence band to conduction band, resulting in a gap of electron holes (13,24-26). Since zinc oxide has a band gap energy almost similar to that of titanium dioxide (3.2 eV), it is expected to have a photocatalytic capacity similar to  $\text{TiO}_2$  (27,28). Some studies have shown that although  $\text{TiO}_2$  has a variety of environmental applications,  $\text{ZnO}$  can be a suitable replacement for  $\text{TiO}_2$  (29-31).

Ultraviolet lamps are commonly used for removing pollutants, because with producing electron pairs and holes on nanoparticles, these lamps increase their power of oxidation. The process of UV/nanoparticles photocatalysis is based on the optical stimulation of a semiconductor (nanoparticles) due to the absorption of electromagnetic radiation (32,33). According to Tanha et al, with increasing the intensity of UV radiation under similar condition, the degradation efficiency of toluene decreased (34).

Using  $\text{ZnO}$  and  $\text{TiO}_2$  catalysts coated on the activated carbon as an absorbent and  $\text{AC}/\text{TiO}_2/\text{ZnO}$  nanocomposite, this research attempted to use the most appropriate catalyst for removing xylene vapors. Therefore, the present study aimed to examine the efficiency of  $\text{TiO}_2$  and  $\text{ZnO}$  photocatalysts coated on activated carbon in removing xylene vapors from polluted air by ultraviolet radiation.

## Materials and Methods

This experimental study was conducted at laboratory scale. At first,  $\text{ZnO}$  nanoparticles (Merck Company),

$\text{TiO}_2$  (Merck Company), activated carbon (Merck Company),  $\text{ZnO}$  nanoparticles catalysts coated on activated carbon ( $\text{ZnO-AC}$ ),  $\text{TiO}_2$  nanoparticles catalysts coated on activated carbon ( $\text{TiO}_2\text{-AC}$ ) and  $\text{ZnO}/\text{TiO}_2$  nanocomposite coated on activated carbon ( $\text{AC}/\text{TiO}_2/\text{ZnO}$ ) were synthesized. Then, their physical features, surface morphology, and capability for the degradation of xylene (86%, Merck Company) in the gas-phase under ultraviolet radiation were assessed.

### Nanoparticles coated on activated carbon

In this study, 4 wt% of each  $\text{TiO}_2$  (5, 10, 15, and 20%) and  $\text{ZnO}$  (3, 5, 10, and 15%) nanoparticles separately and 3 wt% of the  $\text{ZnO}/\text{TiO}_2$  nanocomposite were coated on activated carbon ( $\text{AC}/1\text{TiO}_2/3\text{ZnO}$ ,  $\text{AC}/3\text{TiO}_2/1\text{ZnO}$ ,  $\text{AC}/1\text{TiO}_2/1\text{ZnO}$ ). To prepare 15 wt% of  $\text{TiO}_2$ , for example, a 50 cc Erlenmeyer flask was filled with 0.15 g of  $\text{TiO}_2$  and Double-distilled water was added to it. Then, it was placed in an ultrasonic device, and then, on a 250 rpm shaker {1}. Afterwards, 1 g of activated carbon was added and the mixture was placed in the ultrasonic device again and on the 250 rpm shaker {2}. Then, the materials were passed through a filter paper and the filter paper was placed in an oven at a temperature of  $37^\circ\text{C}$ . Later, the materials were poured in a crucible and dried in a furnace at  $300^\circ\text{C}$  for 2.5 hours so that the process of coating was properly conducted (35). In order to prepare the appropriate amount for every concentration, this process was repeated several times. In order to prepare other ratios, this process was also carried out with different amounts of nanoparticles.

{1} x grams of Nanoparticle+30 cc twice-distilled water → 30 minutes to ultrasonic → 30 minutes to Shaker 250 rpm  
{2} 1 + x gram of AC → 30 minutes to ultrasonic → 2 hours on the 250 rpm shaker

### Determining the features of the photocatalyst

In order to determine nitrogen absorption-desorption isotherms, surface area and pores size, a nitrogen-flow surface area analyzer (Quantachrome Chem BET) was used. By measuring nitrogen injected into the specimen, this method measures the specific absorption area. In order to determine morphology and estimate the size of particles, a 15-kV accelerating voltage scanning electron microscope (TESCAN Mira3 FESEM) was used. And in order to determine the elements present on the bed, a 15-kV accelerating voltage energy dispersive x-ray spectroscopy (EDS) Model Ametek device at different magnifications was used.

### Photocatalytic reactor system

The photocatalytic degradation system of xylene for the experiment was designed at continuous dynamic flow. The photo-reactor was formed of a cylinder chamber, which was 12 m in length and 4 cm in diameter, made of quartz wall (Figure 1). The air inlet was located at the upper

part of photo-reactor and the air outlet at the lower part. A thin-layer porous bed with a uniform distribution of nanoparticles was placed between the inlet and the outlet. In the middle of the cylinder, an 8-Watt ray generator (UV-C lamp) (Philips Company) was placed. Around the reactor, three 6-Watt UV lamps were placed. In order to prevent energy loss, when lamps were on, an aluminum layer was used around them to restore the wasted energy to the system.

### Photocatalytic removal

When the necessary xylene concentration was prepared and the total gas flow was kept fixed, the xylene vapor with a fixed flow rate of 1 L/min entered the reactor. In this process, the inlet and outlet xylene concentrations in the reactor were assessed in darkness. When the inlet and outlet concentrations were equal, there was a dynamic balance. The lamps were turned on immediately and there was a reduction in the outlet concentration. In this interval, the data for experiment were collected. Xylene vapors were measured using a direct-reading device (PhoCheck Tiger model 5000, England) based on a photoionization detector (PID). All specifications of PhoCheck Tiger are against isobutylene calibration via calibration kit accessory at 23°C, 90% RH and detect 1 ppb to 3000 ppm. The rate of removal efficiency was calculated using the equation below:

$$X = (C_i - C_o / C_i) \times 100 \quad (1)$$

where  $X$  is the efficiency of photocatalytic degradation of xylene (%), and  $C_i$  and  $C_o$  are the inlet and outlet xylene concentration, respectively ( $\text{mg}/\text{m}^3$ ). In order to reach more accurate results, each experiment was repeated at least three times and the mean of measurements was used after being recorded in the preliminary tables. In order to analyze tables and draw charts, Excel 2013 was used.

## Results

### Features of photocatalysts

In this study, the specific surface area of Activated Carbon (AC), AC/TiO<sub>2</sub> 15%, AC/ZnO 5%, and AC/3TiO<sub>2</sub>/1ZnO were 920.51, 796.39, 844.92, and 810.64  $\text{m}^2/\text{g}^{-1}$ , respectively. The reduction in the surface was due to the coating of nanoparticles on the bed and blocking a part of its pores. In order to determine the type and percentage of the main elements present in the bed, EDS analysis was used (Figure 2). The mean elemental percentages are shown in Figure 2.

The images obtained from SEM are shown in Figure 3. They show that nanoparticles are sitting on the pores and voids of the activated carbon in masses. The uneven surface of the bed and holes can be clearly seen in the figures. The activated carbon surface absorbent pores blocked by nanoparticles will lead to a reduction in the specific area and the absorption capacity of the combined catalyst.

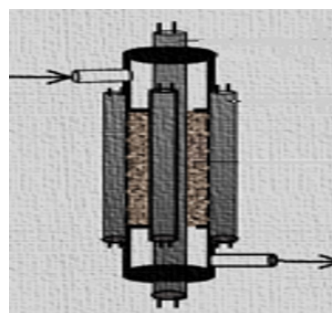


Figure 1. The photo-reactor and UV lamps.

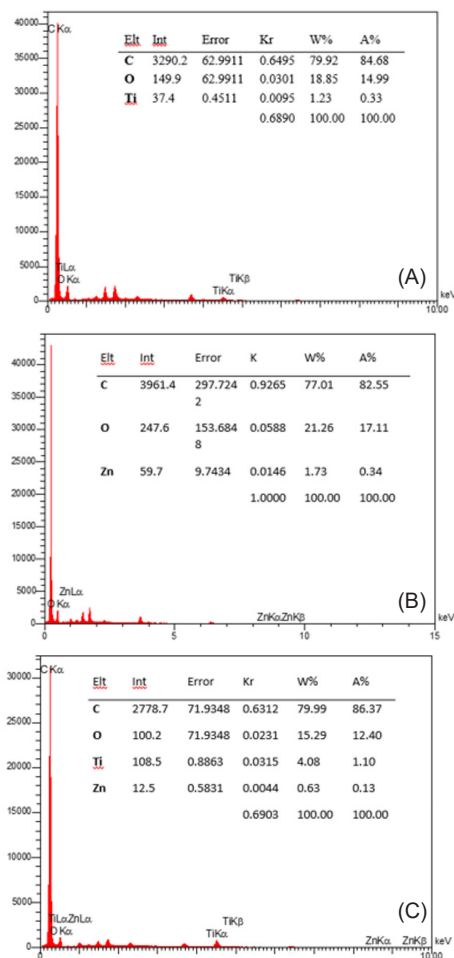
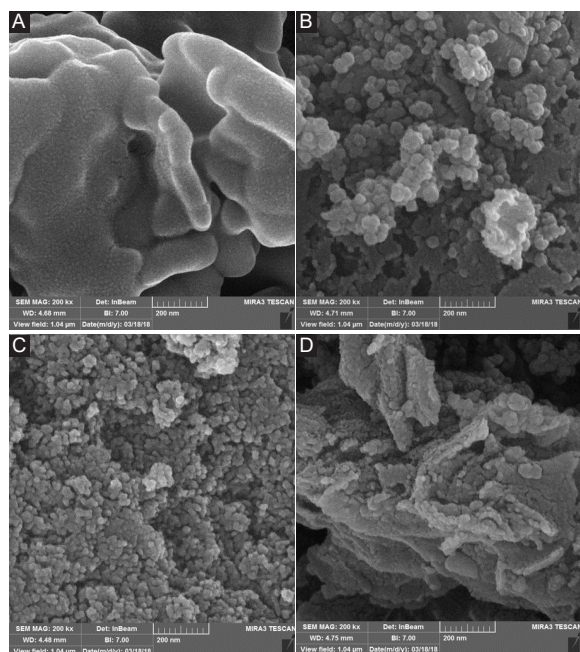


Figure 2. Results of the EDS analysis. 9A) Activated carbon impregnated with TiO<sub>2</sub> nanoparticles, (B) Activated carbon impregnated with ZnO nanoparticles, and (C) Activated carbon impregnated with TiO<sub>2</sub> and ZnO nanoparticles.

### Removal efficiency of xylene by photocatalysts

In order to determine the efficiency of photocatalysts, at each stage, 1 g of a photocatalyst was placed in the reactor under UV, and polluted air containing 100 ppm of xylene with a fixed flow rate of 1 L/min entered the reactor (36). The results of xylene degradation by AC/TiO<sub>2</sub>, AC/ZnO, and AC/TiO<sub>2</sub>/ZnO photocatalysts are shown in Figure 4. As shown in this figure, 15 and 5 wt% of TiO<sub>2</sub> and ZnO nanoparticles and the ratios of 3TiO<sub>2</sub> and 1ZnO in the composite have respectively the highest removal efficiency



**Figure 3.** FESEM images. A) AC, B) AC/TiO<sub>2</sub> 15%, C) AC/ZnO 5%, and D) AC/3TiO<sub>2</sub>/1ZnO.

of xylene from polluted air.

### Discussion

In this study, the mean temperature and ambient humidity were fixed and equal to  $23 \pm 1^\circ\text{C}$  and  $35 \pm 1\%$ , respectively, and polluted air contained 100 ppm of xylene (36). Previous studies have introduced ambient temperature as the best temperature for photocatalytic removal, which is regarded as an advantage over other methods. In a study by Rangkooy et al, the photocatalytic removal of formaldehyde from air was investigated and humidity of 35% was reported as the best humidity for the photocatalytic activity of bone char ZnO (37).

The results of this study showed that the specific surface areas of AC/TiO<sub>2</sub> 15% were smaller than that of the bed and the other catalysts, respectively, because in AC/TiO<sub>2</sub> 15%, due to high rate of nanoparticles, a higher number of pores and voids of AC were occupied. According to the results of Brunauer-Emmett-Teller (BET) analysis, the specific surface area of the activated carbon was  $920.51 \text{ m}^2/\text{g}^{-1}$ . A large specific surface area results in a better distribution of metal oxide nanoparticles, which in turn, will have an increasing impact on the performance of the catalyst (38). The results related to the microscopic images showed that due to the coating of the nanoparticles on AC, the nanoparticles not only accumulate on the surface of the bed but also they occupy the pores and voids present in the structure. Consistent with the results of the present study, Sobana and Swaminathan (39), Pulido Melián et al (40), and Rangkooy et al (37), in their studies on the combined effect of ZnO nanoparticles on bone char in the photocatalytic removal of formaldehyde have reported that ZnO nanoparticles reduce the specific surface area of

bone char.

In this study Or the removal efficiencies of ZnO nanoparticles coated in 3, 5, 10, and 15 wt% on activated carbon, were 59.4%, 80.1%, 60.1%, and 58.5%, respectively, and the optimal percentage belonged to AC/ZnO 5%, which is consistent with the results of the study of Rangkooy et al (41).

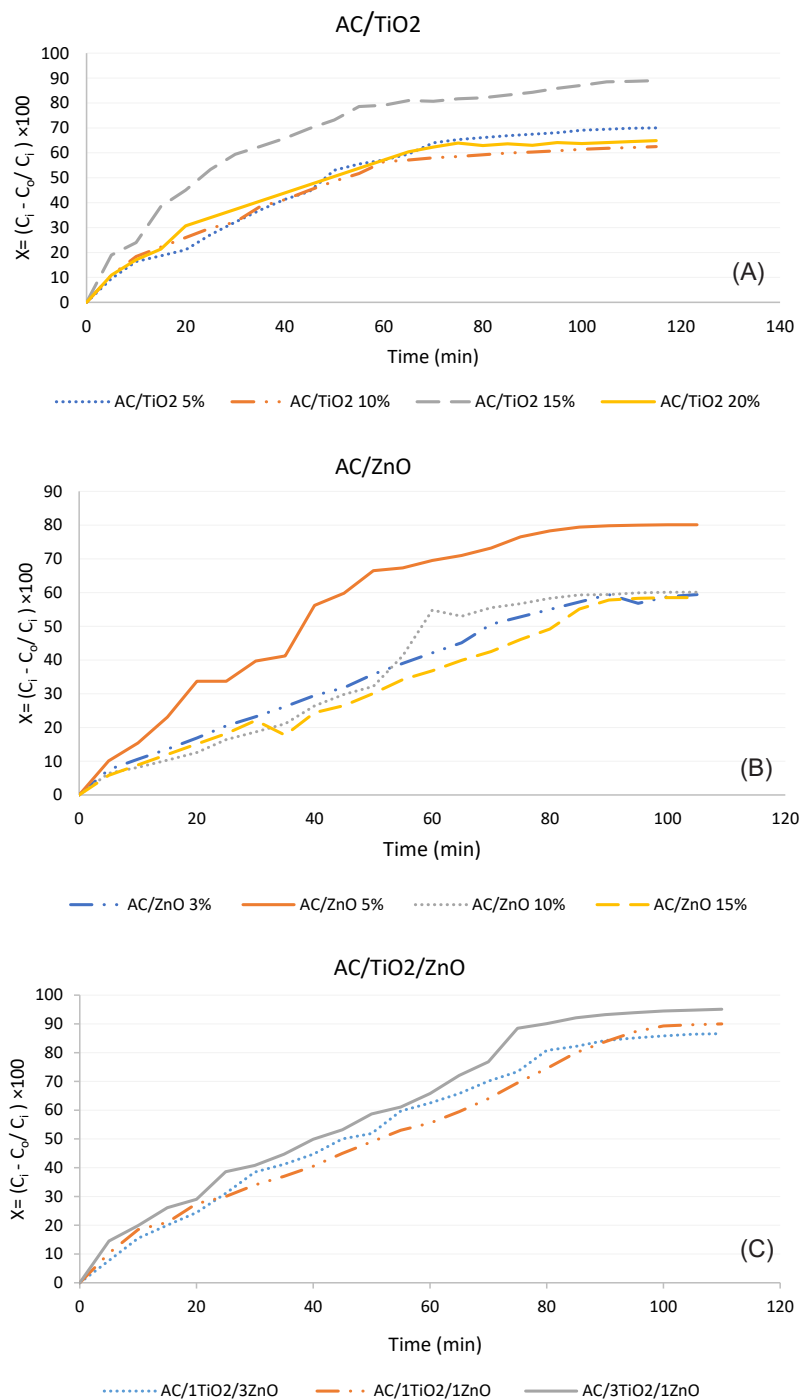
According to the findings of this study, the best ratio of nanoparticles with regard to the photocatalytic removal of xylene from polluted air were AC/TiO<sub>2</sub> 15%, AC/ZnO 5%, and AC/3TiO<sub>2</sub>/1ZnO, which were equal to 89%, 80.1%, and 95.1%, respectively. The photocatalytic removal efficiency of AC/3TiO<sub>2</sub>/ZnO with the ratios of 3:1, 1:1, and 1:3 in TiO<sub>2</sub> and ZnO were 95.5%, 90%, and 86.6%, respectively, and that of AC/TiO<sub>2</sub> 5% and AC/ZnO 15% were 70% and 58.5%, respectively. The results showed that in the simultaneous presence of TiO<sub>2</sub> and ZnO particles, the photocatalytic removal efficiency of xylene increased. Previous studies confirm this effect on other pollutants using different beds. Hadjltaief et al conducted a study on the photocatalytic degradation of methyl green dye in aqueous solution over natural clay-supported ZnO–TiO<sub>2</sub> catalysts. They found that photodegradation efficiency for ZnO–TiO<sub>2</sub>/clay was higher than that for the TiO<sub>2</sub>/clay catalyst, clearly pointing to a promoting effect of ZnO (42). In another study by Nezamzadeh-Ejehieh and Khorsandi, ZnO and clinoptilolite zeolite were used for the photocatalytic removal of phenylhydrazine from the aqueous solution. They found that the photocatalytic removal efficiencies of phenylhydrazine using ZnO/nano-clinoptilolite zeolite, ZnO, and TiO<sub>2</sub> were 69, 37, and 45%, respectively (17). These studies have been carried out in aqueous environment, and no independent effect of the presence of ZnO nanoparticles has been reported.

Abedi et al examined the catalytic effect of TiO<sub>2</sub>/GAC, ZnO/GAC, and TiO<sub>2</sub>-ZnO/GAC combined with non-thermal plasma (NTP) for the decomposition of chlorinated volatile organic compounds (CVOCs). The results revealed that the chloroform removal efficiency of the catalysts was in the order TiO<sub>2</sub>-ZnO/GAC  $\cong$  TiO<sub>2</sub>/GAC > ZnO/GAC, while in regard to the removal of chlorobenzene, the order changed to TiO<sub>2</sub>-ZnO/GAC > ZnO/GAC > TiO<sub>2</sub>/GAC. It was revealed that the use of ZnO-TiO<sub>2</sub> nanocomposites had a better effect on the removal of the pollutant (43).

Habib et al, also reported that ZnO-TiO<sub>2</sub> nanocomposite with the ZnO/TiO<sub>2</sub> molar ratios of 3:1 and 1:1 were efficient photocatalysts. They concluded that nanocomposites could be used as the photocatalyst for treating dye-containing wastewaters (44). This study was conducted in aqueous environment by a thermal method.

### Conclusion

According to the results, using TiO<sub>2</sub>-ZnO nanocomposite coated on activated carbon can be an appropriate method for the photocatalytic removal of xylene from polluted



**Figure 4.** Removal efficiency of xylene by: A) AC/TiO<sub>2</sub>, B) AC/ZnO, and C) AC/TiO<sub>2</sub>/ZnO.

air. Using photocatalytic methods with no high energy consumption at ambient temperature, pollutants can be removed from polluted air. In this case, a lower level of energy is consumed and the reaction temperature does not rise, and as a result, harmful by-products do not enter ambient air. This feature of the photocatalytic removal method indicates its advantage over other methods for removing pollutants.

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#### Ethical issues

The authors have thoroughly observed ethical issues and no data from the study has been or will be published separately elsewhere.

#### Competing interests

The authors declare that they have no competing interests.

### Authors' contributions

All authors participated in the data collection, analysis, and interpretation. All authors critically reviewed, refined, and approved the manuscript.

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