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ORIGINAL ARTICLE

Adsorption of gaseous naphthalene on carbonaceous sorbent: perspective of affecting factors

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Abstract

The innovative and sustainable adsorption technologies for gas phase naphthalene (GPNAP) have attracted attention as a successful method. To remove polycyclic aromatic hydrocarbons effectively, biomass-based activated carbon was produced by chemical activation. The adsorption behavior of naphthalene onto produced activated carbon was also investigated by the BET-specific surface areas, total pore volumes, and average pore sizes. The resulting DCAC800 has successfully prepared a very high surface area (607.5 m^2/g) by the weight ratio of 1:3 as the H_2SO_4 activation agent and the pyrolysis of the resultant material at 800 °C under an N_2 atmosphere. It has been determined that when the humidity in the environment is increased from 30 to 90% RH, it causes a loss of efficiency between 64–71% for DCAC500, DCAC600 DCAC700 DCAC800, and DCAC900. The adsorption experiments at 30% RH showed that the DCAC800 had a strong adsorption affinity to naphthalene and the maximum adsorption amount was shown to reach up to 296 mg/g in a fixed-bed reactor.

Keywords Adsorption sites · Air pollutant · Biomass-based activated carbon · Prevention strategy

1 Introduction

The diversity of chemical usage, the unconscious consumption of domestic resources, and the uncontrolled pollution of natural resources can cause unpredictable consequences aspects of typical global industrialization and urbanization [1, 2]. As a direct result of the rapid development of industrial activities, chemicals released or escaping from various sources can turn out to be destructive to the environment [3]. Polycyclic aromatic hydrocarbons (PAHs) have taken their place among the environmental pollutants in terms of the prevalence of their usage areas [4]. Due to its harmful ecological and human health effects, it has been recognized as a priority pollutant worldwide according to the European Commission, the International Agency for Research on Cancer (IACR), the United States Environmental Protection Agency (USEPA) and has attracted worldwide attention among researchers-academics. Excessive PAHs have exacerbated environmental pollutants released into the environment as mutagenic, carcinogenic, and teratogenic effect [5] due to worsened the air with the incomplete combustion of wood, coal, and oil-based fuels [6, 7]. In terms of its prevalence, naphthalene ($C_{10}H_8$) is the leading pollutant among PAHs in the chemical industry. Found naturally in coal tar, it is a highly toxic and long-lasting environmental pollutant used as a preservative for wood and a moth repellent [8]. Indeed, the primary sources of naphthalene, which has typical gas components showing toxicological effects on environmental biosystems, include motor vehicles [9], airplanes [10], fossil fuel combustion [11], herbicides [12], rubber materials [13] and insect repellents [14], gasoline combustion [15], synthesis of compounds and resins [16], cooking [13], tobacco smoke [17], leakage during petrol transportation and filling [18], industrial plants [19], forest fires as well as its widespread distribution in air of the enclosed spaces [20]. Naphthalene is highly found indoors between 3 and 26 µg/m^3 in the gas form [21], damages the liver and nervous system, and may cause cancer due to prolonged exposure when long-term exposure and accumulated in a particular concentration in the human body [22]. Therefore, controlling naphthalene emissions has become an urgent solution to reduce air pollution and related human health risks [23].

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The innovative and sustainable biotechnologies for the removal of gas phase naphthalene (GPNAP) have attracted attention as a successful method such as non-thermal plasma [24], wet purification [25], ion exchange [26], adsorption [27] used worldwide due to their wide range of applications and benefits. Ensuring the removal of waste and unwanted gas streams captured under optimum conditions, and minimized their current concentration in the environment [28]. The selection of the gas to be removed, the appropriate design and operation of the competitive technology, and the content of organic and inorganic pollutants contained in the gas are essential conditions for efficient removal [29]. Overall, gas adsorption is preferred because of its effectiveness, simplicity, high efficiency, and low energy consumption to remove gas-phase environmental pollutants [30]. The traditional adsorbent widely used in adsorption is carbon-based activated carbon, especially for benzene ring-containing compounds [31, 32]. Several raw materials have been used to synthesize activated carbon, such as different vegetal species [33], raw Ashitaba waste [34], and agricultural and forestry biomass [35]. Various carbonization methods have been applied to synthesize high surface area activated carbon from other wastes [36]. For the recovery and recycling of biomass in the form of waste, awareness of adsorbent production has been created in the sustainable environmental management plan with excellent adsorption performance. At the same time, efforts have been made to produce activated carbon, waste management with an environmentally friendly approach to biomass, and mitigate climate change to remove wastes of various amounts and forms from landfills. In particular, the utilization of herbaceous and agricultural biomass waste has become a good precursor for the preparation of activated carbons.

2 Materials and methods

2.1 Preparation of activated carbons

Datisca cannabina residues (DCR) were dried and ground to 0.5 mm particle size. The activation process involved mixing the initial material and the sulfuric acid (H_2SO_4) as activating agent in a weight ratio of (DCR to reagent) 1:3. The mixing was performed at 80 °C under stirring for 2 h and then waited at room temperature for 24 h. After mixing, the solid was subjected to drying at 105 °C overnight. The chemical-loaded sample was then carbonized in an N_2 atmosphere (50 mL/min). Carbonization was carried out by heating the sample at 10 °C min^{-1} from room temperature up to 500–900 °C and then heated at this temperature for 120 min. After cooling under N_2, the carbonized product was washed to remove the residual chemical until pH neutral. The final products (DCACs) were then dried at 105 °C and labeled as DCAC500, DCAC600, DCAC700, DCAC800, and DCAC900.

The proximate analysis of DCR and DCACs was carried out using ASTM D 3172–3175 methods. Textural properties of the DCR and DCACs were measured using the elemental analyzer (Eurovector, EA3000-Single) for C, H, N, and O content. The morphologies of the DCACs were obtained utilizing SEM (scanning electron microscopy) brand by FEI Quanta FEG 250 as 15 kV and 1000–10,000 x.

2.2 Evaluation of adsorption performance

The experimental naphthalene vapor adsorption is mainly composed of a gas-phase naphthalene generation unit in an Agilent 7890 A GC system equipped with a flame ionization detector (FID) with an Agilent HP-5 capillary column (30 m length × 0.25 mm i.d. × 0.25 µm film thickness). The gas-phase naphthalene is formed by the sublimation of crystals in a quartz tube, and the amount of naphthalene was about 50 mg in the U-tube by changes in the temperature of the water bath (10 ~ 100 °C). The carrier gas (high-purity nitrogen) carried gas-phase naphthalene into the adsorption test system as initial gaseous naphthalene (300 mg/m^3) and 30% and 90% relative humidity at 25 °C and atmospheric pressure with a gas flow of 60 mL/min^{-1} was fed into a fixed-bed reactor (an inner diameter of 8 mm and a length of 235 mm) within 0.1 g DCACs. Prior to the adsorption measurements, DCACs were treated at 135 °C for 2 h to remove the physically adsorbed water molecules and other impurities. Naphthalene concentration was detected by the FID method and then the pipeline is purged, gas stream carrying gas-phase naphthalene is passed through the adsorption bypass. When the FID detection value became stable, naphthalene gas was passed into the adsorption bed for adsorption experiments, and the experimental values were recorded. The adsorption temperature was set as 135 ± 0.2 °C and the adsorption column was a quartz glass tube. The adsorption column was placed in a chromatograph furnace. A blank test verified the adsorption capacity of inert quartz sand to naphthalene was found to be negligible. To avoid naphthalene condensing in the pipeline, the pipeline was wrapped with heating tape between the naphthalene generating device's outlet and the chromatograph's inlet. The adsorption capacity is calculated by the following formula using Eq. 1:

$$q_{(mg/g)} = \left(\frac{FxC_0x10^{-9}}{W}\right)\left[\left(\frac{C_i}{C_0}xt_s\right) - \left(\int_0^{t_s}\frac{C_i}{C_0}d_t\right)\right] \tag{1}$$

This equation shows that q is the saturated styrene sorption capacity (mg/g), and F is the airflow rate (mL/min). The amount of SMACs (g) represents W; C_0 and C_i are styrene inlet and outlet concentration (ppm), and t_s is saturation time (min), respectively.

3 Results

3.1 Characterization studies of the DCACs

Porous structure and carbon content play a significant role in the adsorption capacity of DCACs. The apparent surface area values (S_{BET}), and pore volumes (V_T, $V_{MİC}$, and V_{MESO}) of DCACs by H_2SO_4 impregnation are given in Table 1. Thermo-chemical decomposition of the lignocellulosic components of the DCR occurs under the combined effect of H_2SO_4 dehydration and heat treatment. The H_2SO_4 (while acting as a dehydrating agent) alters the chemistry of pyrolytic processes so that the tar formation is restricted to a minimum, and opens up a porous structure. As temperature increases, the S_{BET} also increases as more non-carbon contents are removed. This is seen by the rising temperature (500 to 800 °C) in S_{BET} value for DCACs from 226.3 to 607.5 m^2/g excluding at 900 °C due to deteriorating porous structure. Maximum values of $V_{MİC}$ were obtained with DCAC800 as 0.497 cm^3/g.

The proximate analysis of moisture content, volatile matter, fixed carbon, and ash contents, all of which are parameters often employed to classify and evaluate the suitability of the biomass materials such as DCR for DCACs synthesis and yield values in Fig. 1. The percentage results of moisture content, volatile matter, ash content, and fixed carbon for DCR were 8.73, 70.48, 1.88, and 18.91%. Fixed carbon is the most important parameter of the proximate analysis which increased from 18.91 to 74.6% although volatile matter decreased from 70.48 to 15.9% by carbonization processes.

Figure 2 demonstrates the SEM images of DCR, DCAC500, DCAC600, DCAC700, DCAC800, and DCAC900 prepared under optimized conditions. The micrographs of DCR have no cavities, cracks, or crevices, although the surface of the DCAC500, DCAC600, and DCAC700 have rough and porous forms of several dimensions. The excavations were emergent during carbonization due to the activation of H_2SO_4. In addition, during the calcination operation at 800 and 900° C, the release of large volatile molecules from the surface left behind the porous, rough, and ruptured surface of DCAC800, and DCAC900. Thus, sights affirm that the structure of the DCAC800 and DCAC900 would be nanoparticles agglomerated and stuck to each other. A similar SEM scene at high temperatures was also observed for carbon-based adsorbent acquired from agricultural waste [37].

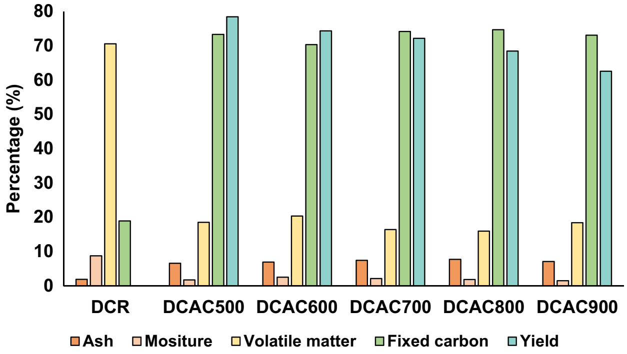

Fig. 1 Proximate and ultimate analysis of DCR and DCACs

3.2 Naphthalene adsorption experiments

In experiments performed after naphthalene sublimation, the effects of initial concentration amount and residence time were investigated in Fig. 3. By keeping the amount of adsorbent used constant and increasing the humidity conditions of the environment, 30 and 90%RH, the filling of the pores of the adsorbents accelerated. The amounts adsorbed above 30% RH, DCAC500, DCAC600 DCAC700 DCAC800, and DCAC900 shown in Fig. 3(a) were 226, 242, 267, 296, and 279 mg/g, respectively. Although the adsorbents were directly dependent on the micropore and surface area of the adsorbents, they were equally effective under ambient conditions. The trend slope shown in Fig. 3(b) is that by increasing the ambient humidity to 90% RH, the amounts adsorbed on DCAC500, DCAC600

Table 1 Structural parameters of DCACs

Samples	Surface area (S_{BET} m^2/g)	Total pore volume (V_T cm^3/g)	Micropores volume ($V_{MİC}$ cm^3/g)	Mesopores volume (V_{MESO} cm^3/g)	Average pore diameter (Dp nm)
DCAC500	226.3	0.190	0.165	0.025	1.67
DCAC600	423.8	0.278	0.220	0.058	1.88
DCAC700	571.6	0.459	0.364	0.095	2.04
DCAC800	607.5	0.553	0.497	0.056	2.18
DCAC900	583.4	0.550	0.486	0.064	1.94

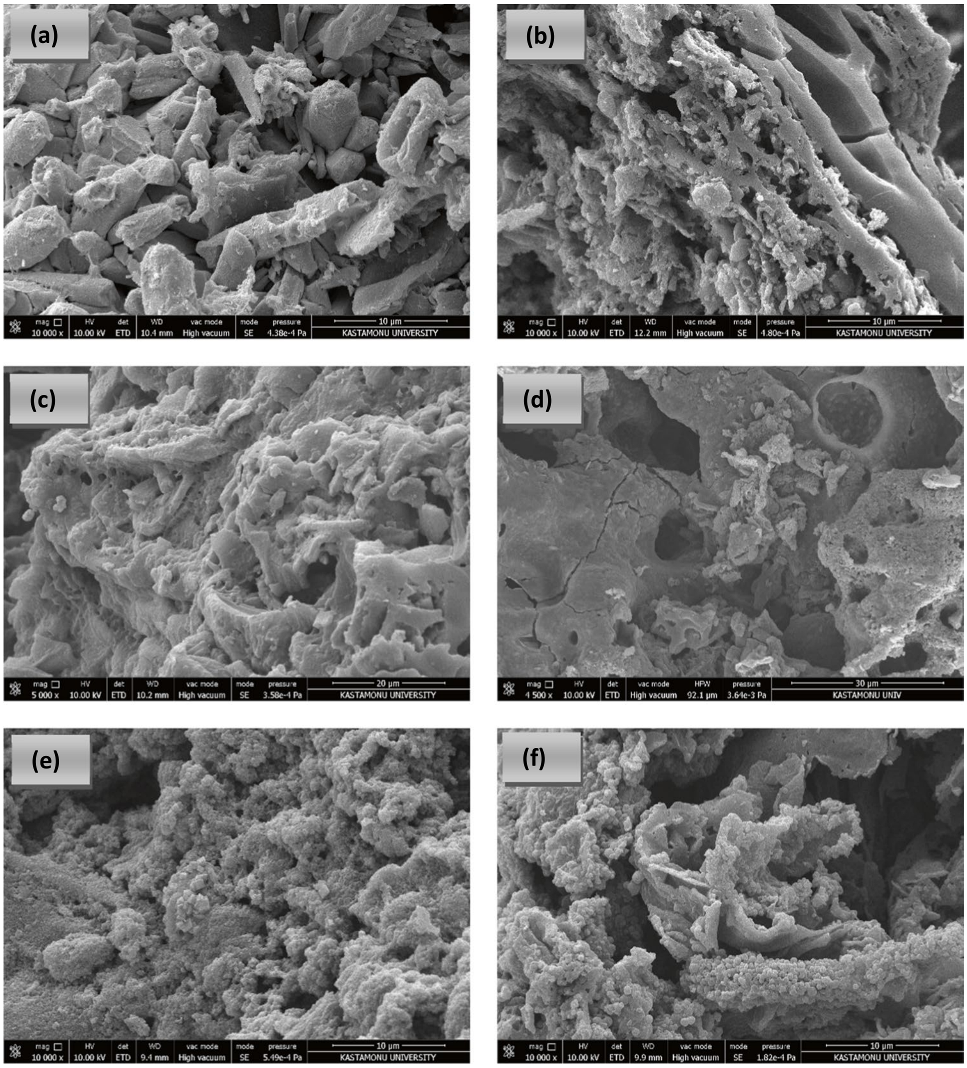

Fig. 2 SEM micrographs of **a** DCR, **b** DCAC500, **c** DCAC600, **d** DCAC700, **e** DCAC800, and **f** DCAC900

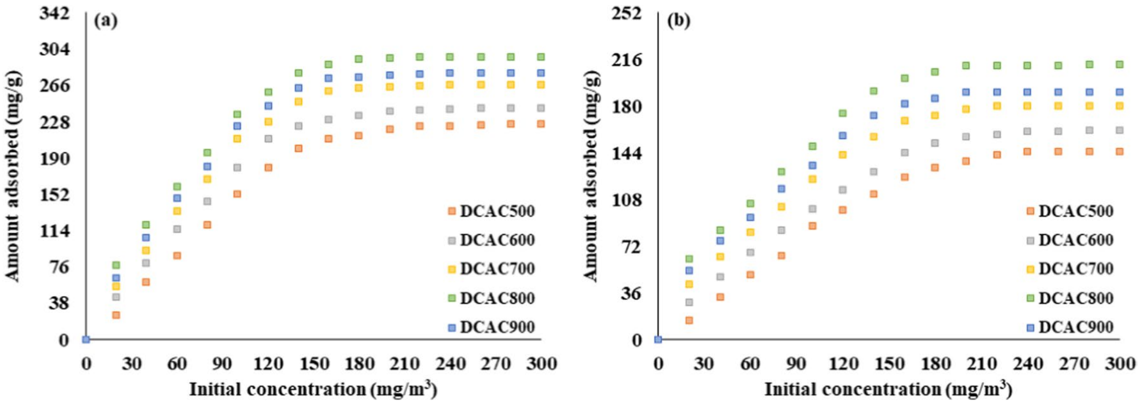

Fig. 3 The amount of DCACs adsorbed depends on initial naphthalene concentration with **a** 30RH% and **b** 90RH%

DCAC700 DCAC800, and DCAC900 were found to be 145, 161, 180, 212, 191 mg/g, respectively. Increasing moisture content causes a loss of efficiency between 64 and 71% in naphthalene adsorbing capacity. The main reason for this is that the pores are quickly filled with water vapor and cannot contain naphthalene molecules.

It is seen that the residence time of the adsorbents increases the holding capacity of the molecules, but the increase occurs until the equilibrium conditions. Figure 4(a) In the experiments where the moisture content is 30RH% and the waiting time is 100 min, there is a difference in naphthalene adsorption amounts according to the pore volumes of different adsorbents. In the equilibrium condition, the amount adsorbed at 30–40 min of the residence time was 220, 242, 266, 295, and 276 mg/g for DCAC500, DCAC600 DCAC700 DCAC800, and DCAC900, respectively. However, as a result of the losses in adhesion with the increase in moisture content in Fig. 4(b), they were 144, 160, 178, 208, and 192 mg/g, respectively. Net losses in efficiency for DCAC500, DCAC600 DCAC700 DCAC800, and DCAC900 after 100 min were found to be between 64 and 70%.

4 Discussion

As an environmental pollutant, naphthalene (NAP) is one of the simplest and the most stable chemical property of PAHs, it has toxic properties on humans depending on its concentration. Therefore, the yields obtained under different conditions by adsorption, which is an effective removal method, were compared with the existing literature. Although NAP removal is partially successful in different methods used, it is seen that it is not as effective and sustainable as adsorption [38]. There are main differences in the adsorbents used. It is clearly understood that it is not an environmentally friendly and sustainable method, whether it is modified or supplemented with chemicals [39]. In this context, the production and regeneration of preferred environmentally friendly porous carbon materials that are produced using agricultural and forestry wastes and that support biomass energy conversion has become a very popular topic in air pollution control. Biomass-based adsorbents have played an active role in the removal of gases commonly found in indoor air such as NAP, especially volatile organic compounds. Although there are differences in its effectiveness on molecules, especially according to the type of raw material, production stage, activation, and the environment in which it is used, the efficiency has been determined higher than other adsorbents. In recent years, the liquid phase of naphthalene removal has been reported in different studies and some useful information has been obtained within various adsorbents; however, fewer investigations have been conducted on gas-phase NAP adsorption due to extremely difficult to stabilize optimal conditions. Wang et al. [40] showed that NAP adsorption onto OMC-B0-90 derived from phenol–formaldehyde resin. Initial gas NAP rate and residence time were fixed at 3.32 cm/s and 0.72 s at a temperature of 150 °C. In equilibrium adsorption, OMC-B10 showed high adsorption capacity with 302.2 mg/g although its mesopore volume was the lowest. Even though it does not seem to have a logical explanation, micropores are the ones that take part in the actual adsorption. Hu et al. [41] investigated the adsorption of polychlorinated naphthalenes (PCNs) onto Amberlite XAD-2 resin in 27 stack gas from nine incinerating plants. They concluded that PCNs in the air were easily useful for controlling adsorbed with suitable adsorbent. Goriaux et al. [42] reported that XAD-4 resin was performed for the GPNAP under continuous ozone flow. Their research indicated that the adsorption capacity was found to be 10.1 ± 0.4 10^{-19} cm^3/molecule s^{-1}. Hübner et al. [43] tried the capture of GPNAP with char particles from woody biomass as feedstock. Experiments conditions were prepared with 10^{15} molecules/cm^3 for the initial GPNAP which tried to compare the specific energy deposition (SED) in the gas volume. Their findings show a remarkable decrease in the naphthalene concentration with increasing SED. Di Gregorio et al. [44] examined the adsorptive removal of naphthalene using three commercially activated carbons (AC1, AC2, and AC3). For each adsorbent, the removal efficiency increased with temperature from 42 to 83.3% and AC3 was found with the highest value because of its highest S_{BET}. Another study reported that naphthalene adsorption onto several mesoporous materials (10–30 mg) existed strong and homogeneous Nap adsorption [45]. The removal performance of GPNAP and its related adsorbents is that most of the conditions utilized for GPNAP adsorption are most

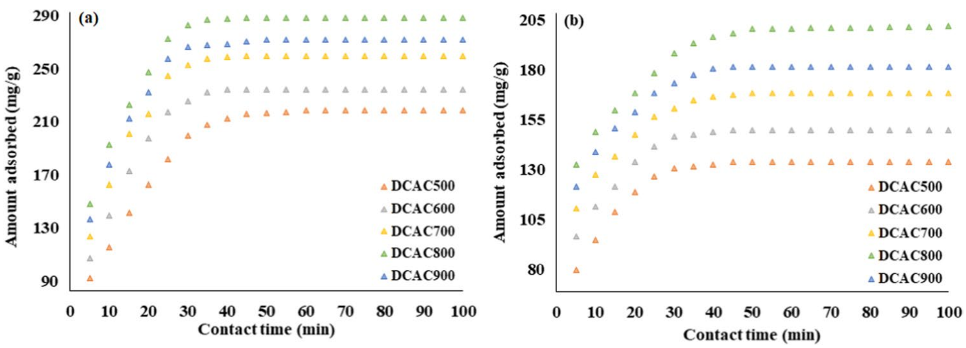

Fig. 4 The amount of DCACs adsorbed depends on contact time with **a** 30RH% and **b** 90RH%

likely linked to the loading surfactants, contact time, and pore structure. According to the results in this work as well as previous studies, the amount of the desorbed naphthalene drastically depends strongly on the S_{BET} and $V_{MİC}$ of the adsorbents by Liu et al. [46] and Teoh et al. [47]. Overall, the adsorption process is reversible and can be depicted as physical adsorption which is mainly affected by the functional groups on the surface of the adsorbent [48]. Therefore, the adsorption capacity of the adsorbent can be improved by adjusting the pore structure of the adsorbent or adding more functional groups to the surface of the adsorbent.

5 Conclusion

NAP is largely emitted to indoor areas and can pose a severe risk to human health, thus, the sorption of NAP onto carbon-based adsorbent has drawn continuous attention. However, gaseous NAP has been rarely studied even though it may be more harmful than the liquid phase. This study focused on the sorption of gaseous NAP onto DCAC800 and the effect of ambient conditions on the adsorption mechanism. It was found that DCAC800 had a significantly higher sorption capacity than DCAC derivatives due to its higher property of S_{BET} and $V_{MİC}$. Although there are some uncertainties in the results, such as the lack of qualitative and quantitative analyses of the secondary product of NAP in the adsorption mechanism. Increasing moisture content causes a loss of efficiency between 64–71% in naphthalene adsorbing capacity. The main reason for this is that the pores are quickly filled with water vapor and cannot contain naphthalene molecules. Future studies could be conducted concerning its regeneration cycle to better understand the reusability for a sustainable environment. Furthermore, the DCAC800 resultantly leads to other PAH derivative adsorptions. A future study could be considered in the sorption of PAH derivatives onto DCAC800.

Author contribution Kaan Isinkaralar: Investigation, Data curation, Original draft preparation, Writing- Reviewing and Editing. Investigation, Writing- Original draft preparation.

Data availability The data that support the findings of this study are available from the corresponding author, upon reasonable request.

Declarations

Ethics approval Not applicable.

Competing interest The author declares no competing interests.

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