

# Modelling of Disinfection by-products Formation via UV Irradiation of the Water from Tajan River (Source Water for Sari Drinking Water. Iran)

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## A-R-T-I-C-L-E I-N-F-O

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## A-B-S-T-R-A-C-T

**Background & Aims of the Study** Irradiation with ultraviolet light (UV) is used for the disinfection of bacterial contaminants in the production of potable water. The main objective of the study was to investigate and model Disinfection By-Products (DBPs) formation due to the UV Irradiation of the Tajan River water under different Irradiation conditions.

**Materials & Methods:** Water samples were collected throughout September 2011 to August 2013. Transportation of the sample to the laboratory was done on ice in a cooler, and physiochemical analysis was conducted immediately within one day. Dissolved organic carbon (DOC) was determined by a TOC analyzer. Irradiation experiments were conducted in a series of 25 mL glass serum bottles with Teflon septa. The present study adopts an orthogonal design. The design involved irradiation with UV at a UV/DOC ratio of 0.5–3.0 and incubating (headspace-free storage) for 5–25 sec. A 1 mM phosphate buffer maintained the pH at 6, 7, or 8 respectively, and an incubator maintained the temperature (Temp) at 15, 20, or 25 °C respectively. The development of empirical models for DBPs formation used a multivariate regression procedure (stepwise) which applied the SPSS System for Windows (Version 16.0).

**Results:** The results showed that the total DBPs formation ranged between 12.3 and 67.4 mg/l and that control of the levels was primarily due to the reaction time and the dissolved organic carbon level (DOC) in the water.

**Conclusions:** Reaction time and level of DOC concentrations in water exerted a dominant influence on the formation of DBPs during the UV irradiation of water from the Tajan River. The relationships between the measured and predicted values were satisfactory with R<sup>2</sup> values ranging from 0.89 (for Octanal)–0.92 (for Formaldehydes). The DOC level in water is the key factor in controlling DBPs formation.

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## Background

Disinfection, as applied in water and wastewater treatment, is a process by which

pathogenic microorganisms are inactivated to provide public health protection (1). Chlorination has been the dominant method employed for disinfection for almost 100 years;

however, it is no longer the disinfection method automatically chosen for either water or wastewater treatment because of potential problems with disinfection by-products and associated toxicity in treated water (2, 3). Among the alternatives to conventional chlorination, ultraviolet (UV) irradiation is chosen the most frequently (4).

Irradiation with ultraviolet light (UV) is used for the disinfection of bacterial contaminants in the production of potable water, and in the treatment of selected wastewater. It has a lot of advantages including no need for any chemical addition (5), fast contact times typically less than 60 sec (6,7), low capital and operating costs, simple operation with minimal system maintenance (8), and hence greater safety for operators (9). However Ultraviolet disinfection efficiency can be impeded by high levels of particulates and chemicals in water or wastewater, micro-organism aggregation, and the geometry between the UV lamp and surrounding sleeve leading to suboptimal flow paths through the lamp assembly (10,11).

A further disadvantage with UV disinfection is producing DBPs such as Octanal, Pentanal, Aldehydes and Formaldehyde, which have unfavorable effects on human and other animal's health (12,13). Other disadvantage with UV disinfection is that unlike other forms of chemical treatment it does not leave a persistent residual in the treated water, which can lead to problems of regrowth of organisms in some circumstances (14). Therefore, there is a need to gain a better understanding of the effectiveness of using UV for the disinfection of wastewater.

Most process models for UV systems are empirical in nature and do not adequately represent the distribution of UV dose that is actually delivered to micro-organisms in a continuous flow system (15). Using the published data of Nguyen, four established model forms were accessed to account for the combined effect of suspended solids and/or

soluble UV absorbing compounds, and UV dose on the efficacy of disinfection. The four model forms were: a log-linear form, Davey Linear-Arrhenius, Square Root and a general  $n^{\text{th}}$  order Polynomial form that was limited to a third order (16). Criteria for assessment of an adequate predictive mode were established including: accuracy of predicted against observed values, percent variance accounted for, and; appraisal of residuals. The Davey Linear-Arrhenius model was shown to best fit the data for UV disinfection of *Escherichia Coli* (ATCC 25922); followed by the  $n^{\text{th}}$  order Polynomial, log linear and Square Root forms. However, the Davey Linear-Arrhenius form must be used in conjunction with a first order chemical reaction, and was shown to predict poorly at high experimental values of UV dose ( $>40000 \mu\text{Ws cm}^{-2}$ ). Davey Linear-Arrhenius model was not amenable to extrapolation beyond the observed UV dose range.

To overcome the shortcomings of the Davey Linear-Arrhenius model synthesis of two new, non-linear model forms were undertaken. The two models were a modified exponentially damped polynomial (EDP<sub>m</sub>) and a form based on the Weibull probability distribution. The EDP<sub>m</sub> model has three terms: a rate coefficient (K), a damping coefficient: and a breakpoint dose ([dose] B). The rate coefficient governs the initial rate of the onset of tailing. The dumping coefficient controls curvature in the survivor curve. The Weibul model has just two terms: a dimensionless scale parameter ( $\beta_0$ ), and; a shape parameter ( $\beta_1$ ). The scale parameter represents the level of disinfection in the tail of the survivor curve (as  $\log_{10} N/N_0$ ), whilst the shape parameter governs the degree of the curvature of the survivor data (17,18).

**Aims of the study:** In order to control DBPs formation and better monitor DBPs in the drinking water, a series of laboratory Irradiation Experiments were conducted using water from the Tajan River. Key operational parameters relating to irradiation conditions were identified

and multiple regression models for predicting DBPs formation were established.

## Materials & Methods

Water samples were collected throughout September 2011 to August 2013. Transportation of the sample to the laboratory was done on ice in a cooler, and physiochemical analysis was conducted immediately within one day. Dissolved organic carbon (DOC) was determined by a TOC analyzer (Xangfu TOC 5000). Irradiation experiments were conducted in a series of 25 mL glass serum bottles with Teflon septa. The present study adopts an orthogonal design, which allows the variation of only one parameter at a time while other parameters maintain a designated “baseline” condition (Table 1). The design involved irradiation with UV at a UV/DOC ratio of 0.5–3.0 and incubating (headspace-free storage) for 5–25 Sec. A 1 mM phosphate buffer maintained the pH at 6, 7, or 8 respectively, and an incubator maintained the temperature (Temp) at 15, 20, or 25 °C respectively. Different DOC levels ([DOC] mg/l) of 100, 200, 300 mg/l were introduced in the water (Fabbricino & Korshin 2009). The conditions of UV/DOC=1.0 (ambient DOC concentration: 2.02 mg/l), pH=7.5 (ambient level) Temp=21°C (ambient level), were utilized as “baseline” conditions.

These glass bottles were cleaned and dried in a muffle furnace at 400°C for 30 minutes prior to use. The reactions were suspended after designated reaction period by adding 12 mg ammonium chloride following USEPA method 551.1 (19). Phosphate buffer was used to maintain pH within 4.8 to 5.5. Fifty ml from each of the samples was taken for DBPs analysis. The DBPs were extracted from these 50 ml by adding 3 ml MTBE and 20g muffled sodium sulphate (20). A solution of decafluorobiphenyl was used to achieve final surrogate concentration of 10µg/l. The use of

decafluorobiphenyl determined the performance of the analysis. All samples were analyzed within 1 week of collection following USEPA method 551.1 (19). Two micro litres of the DBPs extracts were analyzed using gas chromatography (GC) equipped with a fused silica column (0.25mm×30m) and micro electron capture detector (µ-ECD). The stand-alone methodology to determine DBPs was followed for these analyses. Procedural calibrations were developed using DBPs standards. The NOM was measured as UV<sub>254</sub> absorption capacity at 254 nm wavelengths (UV) using an Ian 235 Spectrophotometer. The TOC and DOC were measured using a Xangfu TOC 5000A analyzer. The CO<sub>2</sub> detector was a linearized non-dispersive infrared detector (NDIR) (21). The organic carbon determination was made by injection mode with two injections: the first for determining total carbon and the second to find inorganic carbon. The organic carbon was then calculated as the difference between total and inorganic carbon. For DOC and UV<sub>254</sub>, the samples were filtered using 0.45 µm membrane filters and then analysed (22). The pH was measured with model 200 Hatch pH Meter, which was calibrated using three point calibration (pH: 4, 7, and 10). The temperature and reaction time were adjusted in the laboratory.

### Parameter Investigation:

Although 10 parameters have been identified in a number of modelling approaches, this paper focuses on the most frequently used parameters: DOC, UV<sub>254</sub>, pH, temperature and reaction time. The development of empirical models for DBPs formation used a multivariate regression procedure (stepwise) which applied the SPSS System for Windows (Version 16). The THM level was designated as the dependent variable (Y), and the parameters for the raw water quality and disinfection conditions (t, UV /DOC, pH, Temperature, [DOC<sup>-</sup>]) were defined as independent variables (X). The regression placed independent

variables into the equation in the order of their partial correlation coefficients (rpt) with the dependent variable, and thus the most important predictive variables Were identified via this process. Empirical equations  $Y=10^{b_0}X_1^{b_1}X_2^{b_2}1/4X_i^{b_i}$  (equivalent to

logarithmic function  $\text{Log}_{10}Y=\text{Log}_{10}b_0+b_1\text{Log}_{10}X_1+b_2\text{Log}_{10}X_2^{1/4}+b_i\text{Log}_{10}X_i$ ) were generated. (Brenner et al 2005).

Table 1) Orthogonal design for chlorination experiments of water from The Tajan River

Treatment	UV/DOC	pH	Temp(°C)	[DOC <sup>-</sup> ](mg/l)	Reaction time (sec)
1	0.5	7	20	8.5	5, 10, 15, 20, 25
2	1	7	20	8.5	5, 10, 15, 20, 25
3	1.5	7	20	8.5	5, 10, 15, 20, 25
4	2	7	20	8.5	5, 10, 15, 20, 25
5	3	7	20	8.5	5, 10, 15, 20, 25
6	1	6	20	8.5	5, 10, 15, 20, 25
7	1	8	20	8.5	5, 10, 15, 20, 25
8	1	7	15	8.5	5, 10, 15, 20, 25
9	1	7	25	8.5	5, 10, 15, 20, 25
10	1	7	20	100	5, 10, 15, 20, 25
11	1	7	20	200	5, 10, 15, 20, 25
12	1	7	20	300	5, 10, 15, 20, 25

## Results

Table 2 shows the data from the irradiation experiments, and shows the DBPs formation curve in Fig 1. Octanal, Pentanal, Aldehydes and Formaldehyde are described in the following equations (1)–(4) as a function of t (5–25 sec), UV/DOC (0.5–3), pH (6–8), Temp (15–25 °C) and [DOC<sup>-</sup>] (8.5–300 µg/l), respectively:

$$\text{Octanal} = 10^{-1.111}(t) 0.178(\text{UV/DOC}) 0.203(\text{pH}) 1.46(\text{Temp}) 0.328 \times ([\text{DOC}^-]) 0.268 \quad (R^2_{\text{adj}}=0.89, p<0.0002, n=72) \quad (\text{Eq.1})$$

$$\text{Pentanal} = 10^{-1.203}(t) 0.197 (\text{pH}) 1.31(\text{Temp}) 0.279 \times ([\text{DOC}^-]) 0.546 \quad (R^2_{\text{adj}}=0.91, p<0.0002, n=72) \quad (\text{Eq.2})$$

$$\text{Formaldehydes} = 10^{-1.278}(t) 0.206 (\text{pH}) 1.0307(\text{Temp}) 0.299 \times ([\text{DOC}^-]) 0.287 \quad (R^2_{\text{adj}}=0.92, p<0.0001, n=72) \quad (\text{Eq.3})$$

$$\text{Aldehydes} = 10^{-1.212}(t) 0.189(\text{UV/DOC}) 0.0242(\text{pH}) 1.324(\text{Temp}) 0.287 \times ([\text{DOC}^-]) 0.246 \quad (R^2_{\text{adj}}=0.91, p<0.0002, n=72) \quad (\text{Eq.4})$$

It shows that the total Octanal formation Eq. (1) positively correlated with all the parameters indicating the raw water quality and irradiation conditions including reaction time, pH, temperature, and DOC concentration. Pentanal and Formaldehydes formation Eq<sub>s</sub> (2) and (3) appeared to have a positive correlation to all the independent irradiation variables but obtained no significant correlations with UV energy (UV/DOC). Finally, the Aldehydes formation Eq. (4) positively correlated to all the independent irradiation variables but negatively correlated to DOC concentration. According to the rpt values for both Octanal and Aldehydes formation models (1, 4) the reaction time (Octanal:  $R^2_{\text{pt}}=0.364$ ; Aldehydes:  $R^2_{\text{pt}}=0.299$ ) and the DOC concentration (Octanal:  $R^2_{\text{pt}}=$

0.357; Aldehydes:  $R^2_{pt}=0.392$ ) exerted the most  $R^2_{pt}= 0.0359$ ; Aldehydes:  $R^2_{pt}=0.0285$ ) and UV/DOC (Octanal:  $R^2_{pt}= 0.0264$ ; Aldehydes:  $R^2_{pt}=0.0484$ ) while the temperature showed the least influence (Octanal:  $R^2_{pt}= 0.0076$ ; Aldehydes:  $R^2_{pt}=0.0132$ ). On the other hand, for UV/DOC independent DBPs formation (Eq<sub>s</sub> (2) and (3)), DOC concentration showed the predominant influence (Pentanal:  $R^2_{pt}= 0.717$ ; Formaldehydes:  $R^2_{pt}=0.483$ ) followed by reaction time (Pentanal:  $R^2_{pt}= 0.65$ ; Formaldehydes:  $R^2_{pt}=0.210$ ).

The relationships between the measured and predicted values were satisfactory (Fig. 2) with  $R^2$  values ranging from 0.89 (for Octanal)–0.94 (for Pentanal), compared with other DBPs

important influence, followed by pH (Octanal: formation models with  $R^2$  values ranging between 0.34 and 0.99 (23, 24). This deemed these models valid to describe the experimental data. Regarding the models for Octanal and Formaldehydes, 78.6–81.8.8% of the predicted values fell within  $\pm 20\%$  of the measured values with small fractions (2.23–4.01%) of extreme values (out of  $\pm 31\%$  of the measured values). The models for Pentanal and Aldehydes showed relatively weak prediction ability as only approximately 59.78–64.6% of the predicted values fell within  $\pm 20\%$  of the measured ones, whereas the extreme predicted values accounted for 10.21–11.11%.

**Table 2) Mean  $\pm$  SD and range of DBPs (mg/l) from the irradiation experiments (reaction time: 5–25 sec)**

Treatment	Octanal	Pentanal	Formaldehydes	Aldehydes
1	7.6 $\pm$ 3.02 (4.58–10.62)	4.52 $\pm$ 1.64 (2.88–6.16)	1.203 $\pm$ 0.21 (0.99–1.41)	3.04 $\pm$ 0.87 (2.17–3.91)
2	8.9 $\pm$ 4.07 (4.83–12.97)	4.32 $\pm$ 1.21 (3.11–5.53)	1.48 $\pm$ 0.61 (0.87–2.09)	4.08 $\pm$ 0.98 (3.1–5.06)
3	11.54 $\pm$ 4.6 (6.94–16.14)	4.42 $\pm$ 1.09 (3.33–5.51)	1.52 $\pm$ 0.97 (0.55–2.49)	4.19 $\pm$ 1.01 (3.18–5.54)
4	10.36 $\pm$ 3.87 (6.49–14.23)	3.59 $\pm$ 1.21 (2.38–4.8)	1.66 $\pm$ 0.56 (1.1–2.22)	4.67 $\pm$ 1.32 (2.35–5.99)
5	14.8 $\pm$ 4.35 (10.45–19.15)	6.24 $\pm$ 1.58 (4.66–7.82)	1.22 $\pm$ 0.44 (0.78–1.66)	4.78 $\pm$ 0.99 (3.79–5.77)
6	8.9 $\pm$ 3.12 (5.78–12.02)	3.89 $\pm$ 1.99 (1.9–5.88)	1.02 $\pm$ 0.34 (0.68–1.36)	3.21 $\pm$ 1.11 (2.1–4.32)
7	14.25 $\pm$ 6.25 (8–20.5)	5.47 $\pm$ 2.21 (3.26–7.68)	0.89 $\pm$ 0.76 (0.13–1.65)	6.32 $\pm$ 2.56 (3.76–8.88)
8	9.24 $\pm$ 3.11 (6.13–12.35)	3.64 $\pm$ 1.23 (2.41–4.87)	0.93 $\pm$ 0.34 (0.59–1.27)	3.34 $\pm$ 0.65 (2.69–3.99)
9	10.49 $\pm$ 5.25 (5.24–15.47)	4.59 $\pm$ 1.12 (3.47–5.71)	0.78 $\pm$ 0.21 (0.57–0.99)	5.15 $\pm$ 1.23 (3.92–6.38)
10	8.02 $\pm$ 3.11 (4.91–11.13)	6.87 $\pm$ 2.4 (4.47–9.27)	1.02 $\pm$ 0.18 (0.84–1.2)	6.89 $\pm$ 2.2 (4.67–9.09)
11	6.2 $\pm$ 1.21 (4.99–7.41)	7.97 $\pm$ 4.21 (3.79–12.18)	1.58 $\pm$ 0.32 (1.26–1.9)	9.25 $\pm$ 4.4 (4.85–13.65)
12	6.8 $\pm$ 1.25 (5.55–8.05)	9.12 $\pm$ 3.11 (6.01–12.23)	2.01 $\pm$ 1.74 (0.27–3.75)	10.23 $\pm$ 6.01 (4.22–13.24)

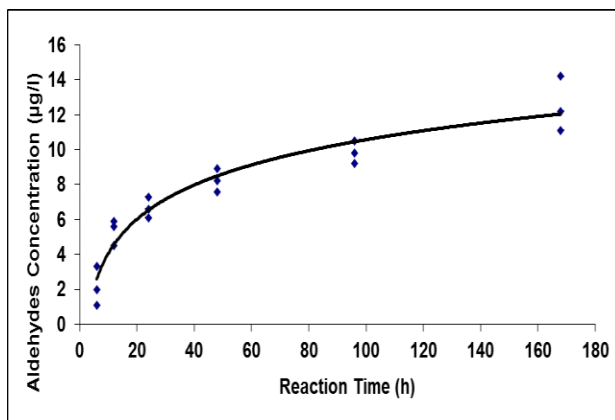


Figure 1) Aldehydes formation curves (temp: 15–25 °C; pH: 6–8; DOC: 8.5–300 µg/l; UV /DOC: 0.5–3): Aldehydes =  $10.1t^{0.242}$  ( $R^2=0.91$ ;  $p=0.0002$ ).

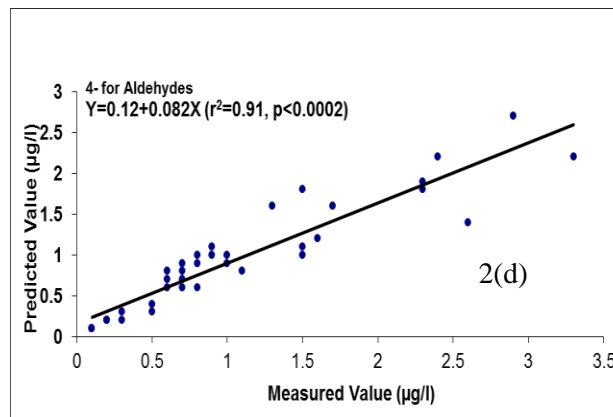
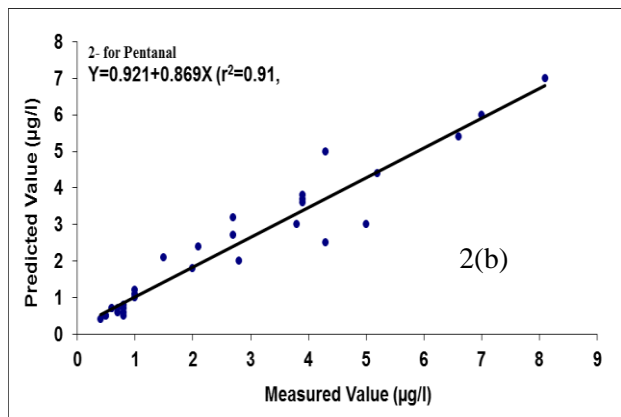
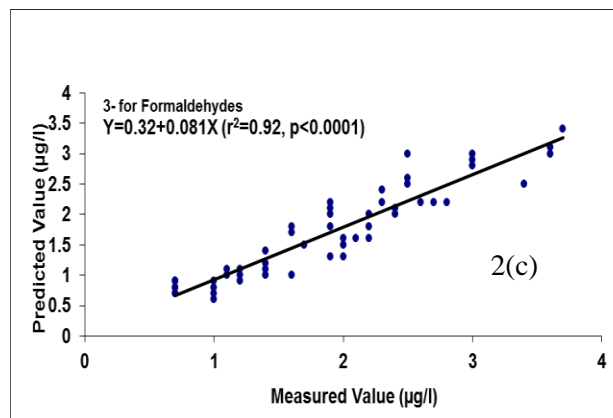
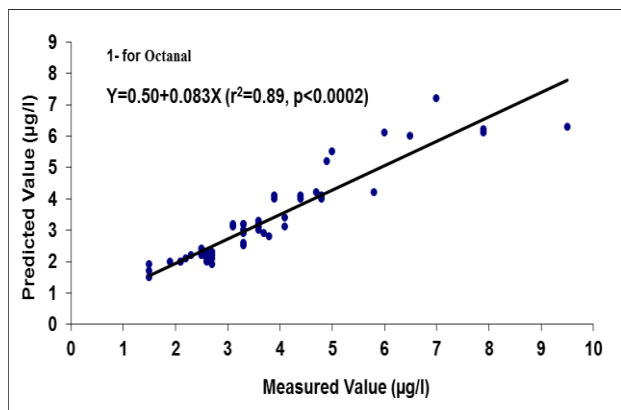


Figure 2) Regressions of predicted values from multiple regressions empirical models (a–d) on the measured values (99% confidence intervals)

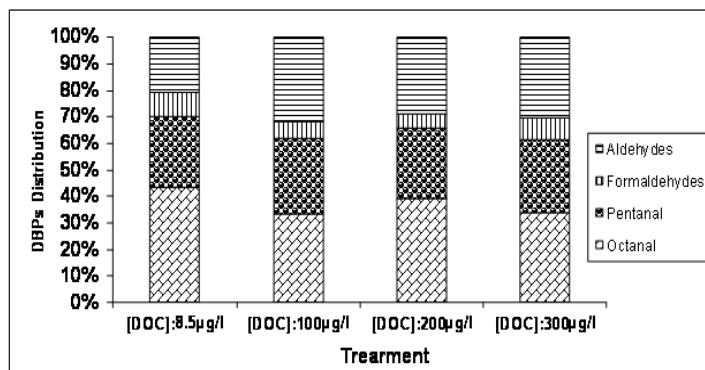


Figure 3) Effects of DOC levels on DBPs distribution

## Discussion

In general, the effects of the raw water quality and irradiation conditions were expected. Higher temperatures may promote irradiation reaction rates and longer reaction time may lead to complete and higher production of DBPs (25). Generally, high DOC concentration in water provided higher inorganic precursors in the water, although additionally, types of organic material in water such as Ketones and Aldehydes an important role in determining DBPs formation (26,27). Fig. 3 shows the changes of speciation of DBPs with increasing levels of [DOC] (8.5–300 µg/l). It appeared that with the elevation of DOC level, increased levels of DBPs were observed. The result was in agreement with research by Jin et al (2013) who indicated that the peak concentration of Aldehydes occurred at DOC concentrations of 300–500 µg/l.

Reaction time and level of DOC concentrations in water exerted a dominant influence on the formation of DBPs during the irradiation of water from the Tajan River. The relationships between the measured and predicted values were satisfactory with  $R^2$  values ranging from 0.89 (for Octanal)–0.92 (for Formaldehydes). The DOC level in water is the key factor in controlling DBPs formation.

With knowledge of the DBPs productivity of the water, this model offers a simple and straightforward tool which can be readily applied to WTWs and distribution systems alike to provide an initial assessment of the risks of total DBPs formation at different sites, and to identify sites and systems at risk of compliance failure. Relying only on the measurement of analytically undemanding parameters, under appropriate circumstances this model offers advantages of simplicity and cost-effectiveness over other, more complex models.

## Footnotes

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### Conflict of Interest:

The authors declare no conflict of interest.

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