**Supplementary materials**

Section 1: Adsorption isotherm and kinetic models

The Langmuir adsorption isotherm model (Equation 2, [1]) was used to study fluoride removal and to describe the amount of fluoride adsorbed per unit weight of COMA at equilibrium:

$$\frac{1}{Q_e} = \frac{1}{Q_m} + \frac{1}{Q_m K_L C_e} \quad (1)$$

where $C_e$ is the equilibrium concentration of fluoride (mg/L), $Q_e$ is the amount of fluoride adsorbed per gram of COMA at equilibrium (mg/g), $Q_m$ is maximum monolayer coverage capacity (mg/g), $K_L$ is the Langmuir isotherm constant (L/mg), $R_L$ is a dimensionless constant separation factor (Equation 3):

$$R_L = \frac{1}{[\,1 + (1 + K_L C_o)]} \quad (2)$$

where $C_o$ is the initial fluoride level (mg/L), and $K_L$ is the Langmuir isotherm constant. $R_L$ value describes the adsorption mechanisms, which is unfavorable ($R_L > 1$), linear ($R_L = 1$), favorable ($0 < R_L > 1$), or irreversible ($R_L = 0$) [2].

Freundlich model was applied for fluoride removal to ascertain the adsorption characteristics (Equation 4, [3]):

$$\log\left(Q_e\right) = \log(K_f) + \frac{1}{n}\log(C_e) \quad (3)$$

where $K_f$ is the Freundlich isotherm constant (mg/g), $n$ is the adsorption intensity, $C_e$ is the equilibrium concentration of fluoride (mg/L), and $Q_e$ is the amount of fluoride adsorbed per gram of COMA at equilibrium (mg/g). The Freundlich constant ($n$) indicates the adsorption mechanism and when $2 < n < 10$, adsorption is favorable, between $1 < n < 2$ moderately difficult and $n < 1$ poor [4].

Brunauer-Emmett-Teller (BET) adsorption isotherm model (Equation 5, [5]) was used to postulate that there is adsorption in the form of multilayers and that there is no interaction among adsorbed ions:

$$\left[\frac{C_e}{(C_s - C_e) \times Q_e}\right] = \left(\frac{K_B - 1}{K_B \times Q_o}\right) \times \frac{C_e}{C_s} + \frac{1}{(K_B Q_o)} \quad (4)$$

where $C_e$ is equilibrium concentration (mg/L), $C_s$ is fluoride monolayer saturation concentration (mg/L), $Q_o$ is the maximum adsorption capacity of fluoride for forming single layers per gram of COMA (mg/g), and $K_B$ is BET adsorption isotherm, which relates to the energy of surface interaction (L/mg).

The data were fitted to Dubinin-Radushkevich isotherm, which is applied to express the adsorption mechanism with a Gaussian energy distribution onto a heterogeneous surface distinguishing between chemisorption and physisorption (Equation 6, [6]):

$$\ln Q_e = \ln Q_d - B_D \varepsilon^2 \quad (5)$$

where $Q_e$ is the amount of fluoride adsorbed per gram of COMA at equilibrium (mg/g), $Q_d$ is the theoretical isotherm saturation capacity (mg/g), $B_D$ is the Dubinin-Radushkevich isotherm constant ($mol^2/kJ^2$), $\varepsilon$ is the potential energy (Equation 7), and $E$ is Dubinin-Radushkevich isotherm constant (kJ/mol) (Equation 8):

$$\mathcal{E} = RT\,ln\,[1 + (\frac{1}{C_e})] \quad (6)$$

$$E = \frac{1}{\sqrt{2B_D}} \quad (7)$$

where physisorption is likely if $E$ < 8 kJ/mol, chemisorption is likely if 8 < $E$ < 16 kJ/mol and through ion exchange if $E$ > 16 kJ/mol.

The data were fitted to Temkin adsorption isotherm, which contains a factor that explicitly considers the COMA-fluoride interactions (Equations 9 and 10, [7]):

$$Q_e = B\,ln\,C_e + B\,ln\,A_T \quad (8)$$

$$\mathrm{B} = \left(\frac{RT}{b}\right) \quad (9)$$

where $A_T$ is the Temkin isotherm equilibrium binding constant (L/g), $b$ is the Temkin isotherm constant, $R$ is universal gas constant (8.314 J/mol/K), $T$ is the Temperature (K), and $B$ is a constant related to the heat of sorption (J/mol).

Kinetics of fluoride adsorption were analysed using Lagergren's pseudo-first-order kinetic model (Equation 11, [8]) and pseudo-second-order kinetic model (Equation 12, [9]). These two models identify kinetics of fluoride adsorption processes to be fitted either to chemisorption or physisorption:

$$log\,(Q_e - Q_t) = log\,Q_e - \left(\frac{K_1}{2.303}\right)\,t \quad (10)$$

$$\frac{t}{Q_t} = \frac{1}{\left[K_2\,(Q_e)^2\right]} + \left(\frac{1}{Q_e}\right)\,t \quad (11)$$

where $Q_e$ is amount of fluoride adsorbed per gram of COMA at equilibrium (mg/g), $Q_t$ is the amount of fluoride adsorbed per gram of COMA at time $t$ in minutes (mg/g), $K_1$ is rate constant ($min^{-1}$) for pseudo-first order kinetics, and $K_2$ is rate constant (mg/g/min) for pseudo-second-order kinetics.

Section 2:

**Table 1.** Adsorption isotherm parameters for fluoride adsorption on COMA

| ***Isotherm models*** | ***Parameters*** | ***Fluoride*** |
|---|---|---|
| *Langmuir* | $Q_m$ mg $g^{-1}$ | *17.82* |
| | $K_L$ L $mg^{-1}$ | *0.85* |
| | $R^2$ | *0.99* |
| *Freundlich* | $K_f$ L $mg^{-1}$ | *0.13* |
| | $1/n$ | *0.93* |
| | $R^2$ | *0.86* |
| *BET* | $Q_o$ mg $g^{-1}$ | *0.44* |
| | $K_b$ L $mg^{-1}$ | *864.9* |
| | $R^2$ | *0.89* |
| *Temkin* | $A_T$ L $mg^{-1}$ | *0.30* |
| | $B$ J $mol^{-1}$ | *9.23* |
| | $R^2$ | *0.80* |
| *Dubinin-Radushkevich* | $Q_e$ mg $g^{-1}$ | *1.00* |
| | $E$ KJ $mol^{-1}$ | *1.45* |
| | $B_D$ $mol^2$ $KJ^{-2}$ | *0.24* |
| | $R^2$ | *0.81* |

Section 3:

**Table 2.** Equilibrium constants and thermodynamic parameters for kinetics models of fluoride adsorption studies

| ***Pseudo-first order*** | | | | | ***Pseudo-second order*** | | | |
|---|---|---|---|---|---|---|---|---|
| *Fluoride* | $Q_{e(exp)}$ | $Q_{e(Cal)}$ | $K_1$ | | $Q_{e(Exp)}$ | $Q_{e(Cal)}$ | $K_2$ | |
| *mg* $L^{-1}$ | *mg* $g^{-1}$ | *mg* $g^{-1}$ | $min^{-1}$ | $R^2$ | *mg* $g^{-1}$ | *mg* $g^{-1}$ | *g* $min^{-1}$ $mg^{-1}$ | $R^2$ |
| *5* | *4.50* | *0.48* | *0.003* | *0.80* | *2.57* | *2.98* | *0.09* | *0.97* |
| *10* | *4.90* | *0.43* | *0.016* | *0.90* | *4.60* | *4.83* | *0.34* | *0.99* |
| *15* | *7.00* | *0.26* | *0.017* | *0.80* | *6.82* | *6.96* | *0.66* | *1.00* |
| *20* | *10.30* | *0.12* | *0.030* | *0.65* | *10.18* | *10.44* | *0.23* | *0.99* |
| *25* | *12.40* | *2.50* | *0.002* | *0.07* | *10.09* | *10.09* | *0.14* | *0.99* |
| *30* | *15.80* | *3.53* | *0.001* | *0.90* | *12.35* | *12.53* | *0.21* | *1.00* |
| *35* | *20.20* | *4.07* | *0.001* | *0.65* | *16.31* | *16.64* | *0.21* | *1.00* |
| *40* | *20.20* | *4.66* | *0.013* | *0.77* | *19.71* | *20.04* | *0.31* | *1.00* |

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