

# Effect of soil composition in copper (II), lead (II), and zinc (II) ion adsorption capacity

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## ABSTRACT

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Received: 7 December 2022

Revised: 11 May 2023

Accepted: 22 May 2023

Published: 24 July 2023

**Citation:**

Tan, W., Surugau, N., Bono, A., Bahrur, M. H. V., Idris, R., Ali, S. A. M., Tair, R., and Rahim, S. A. (2023). Effect of soil composition in copper (II), lead (II), and zinc (II) ion adsorption capacity. *Science, Engineering and Health Studies*, 17, 23020001.

The ability of soil to bind with heavy metals is related to soil adsorption capacity. Different soil compositions have different adsorption capabilities for heavy metal ions. In this study, the excess adsorption isotherm of binary adsorption by individual heavy metals ( $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$ ) onto different clay–sand ratios was investigated. Five different ratios were examined (1:0, 9:1, 7:3, 1:1, and 0:1). The excess adsorption isotherm was measured using a typical batch adsorption approach, and the experimental data were interpreted using the pseudo-ideal monolayer adsorption model. The adsorption capacities ranged from  $0.003$  to  $1.83 \times 10^{-3}$  mg/g ( $\text{Cu}^{2+}$ ),  $0.007$  to  $1.62 \times 10^{-2}$  mg/g ( $\text{Pb}^{2+}$ ), and  $2.0$  to  $1.71 \times 10^{-5}$  mg/g ( $\text{Zn}^{2+}$ ). The results revealed that increasing the clay composition in the admixture increased the selectivity of  $\text{Pb}^{2+}$  more than  $\text{Cu}^{2+}$  and  $\text{Zn}^{2+}$ . The sequence of adsorption capacity in the non-competitive adsorption of different compositions of clay and sand showed the following heavy metal adsorption pattern:  $\text{Pb}^{2+} > \text{Cu}^{2+} > \text{Zn}^{2+}$ .

**Keywords:** excess adsorption isotherm; pseudo-ideal monolayer adsorption model;  $\text{Pb}^{2+}$ ;  $\text{Cu}^{2+}$ ;  $\text{Zn}^{2+}$

## 1. INTRODUCTION

The anthropogenic sources of heavy metals are primarily from industrial, pharmaceutical, mining, and smelting discharge; domestic disposal; and improper management of industrial waste or agricultural activities (Bagul et al., 2015; Sankhla et al., 2016; Yahya et al., 2018). These heavy metals can be disposed of through treatment, filtration, or immediate release into rivers, oceans, lakes, or land. Some industries haphazardly release heavy metals into the environment due to a lack of adequately equipped plants, improper environmental management, and operations outside of appropriate standards, particularly in developing countries (Anyanwu et al., 2018). The lack of

environmental technologies and regional law enforcement in developing nations also contributes to the increased rate of heavy metal contamination in the environment (Hu et al., 2014; Ding, 2019). As heavy metals are non-biodegradable, non-thermdegradable, persistent, and cannot be chemically degraded (Oluyemi and Olabanji, 2011; Taghipour et al., 2013), they tend to accumulate in very small amounts and remain in soil or water. An increase in heavy metal concentration accumulation over time will significantly affect environmental quality.

In nature, heavy metals exist in several complex or compound forms, including oxides, phosphates, sulfates, hydroxides, sulfides, nitrates, silicates, and organic matter (Masindi and Muedi, 2018). These also posess various

oxidation states. Consequently, oxidation properties also affect the behavior of heavy metals. Likewise, the oxidation state of numerous heavy metals in the environment, particularly in soil components, determines their toxicity. The level of oxidation state can affect the determination of heavy metals in both natural and artificial geochemical barriers. In contaminated soils, the oxidation state of heavy metals, such as arsenic, selenium, chromium, and antimony increases (Vodyanitskii, 2013). Heavy metals can become hazardous to the environment and other forms of life when their oxidation state increases or decreases due to their ability to form free radicals. This causes DNA damage, alters sulfhydryl homeostasis, and leads to lipid peroxidation. For instance, chromium (III) is an essential trace element for human health, whereas chromium (VI) is carcinogenic and the most dangerous form of chromium. Meanwhile, due to its high solubility, arsenic (III) is more hazardous than arsenic (V) at a lower oxidation state. Moreover, it has been discovered that the mobility of heavy metals, particularly in soil, is closely related to their oxidation levels. The mobility of arsenic, chromium, and vanadium is greater in lower oxidation states, whereas antimony's mobility improves in higher oxidation states.

In general, soil is composed of different compositions, minerals, and particles, which result in soil properties that vary from one location to another. Common features of soil include clay, sand, and silt. Diverse soil constituents possess a variety of binding capacities with various contaminant forms that can be bound to the soil (Bhakta and Munekage, 2013). Soil has evolved into a natural adsorbent capable of accumulating contaminants like heavy metals from natural and anthropogenic sources. Heavy metals in soil might dissolve in the soil solution or occupy exchange sites in inorganic soil constituents. They may also be adsorbed in inorganic soil constituents, precipitated as pure or mixed solids, present in the structure of secondary minerals, or present in the structure of primary minerals (Kome et al., 2019; Draszawka-Bolzan, 2017). Among these interactions, adsorption plays the most significant role in the accumulation of heavy metals in soils. Nonetheless, it is necessary to comprehend the role of soil in determining whether it is a sink or source of heavy metals because each type of soil possesses different compartments that influence its adsorption capacity and retention time to retain heavy metals in soil. Even a small amount of clay can be separated into numerous types, including kaolinite, bentonite, chlorite, glauconite, and vermiculite (Adeyemo et al., 2017). This study examined the adsorption of selected heavy metals onto various soil components and determined their adsorption capacities. Due to the wide variety of soil types, the scope of this investigation was limited to sand and clay. The sand was river sand, while the clay was kaolin. In addition, adsorption through a composition containing varying proportions and types of clay or sand has not been performed or reported.

In previous studies, the measurement and models of conventional adsorption isotherm were directly interpreted from measured excess adsorption isotherm with the assumption that only single adsorption was involved in a binary system (Müllerová et al., 2003; Jiang et al., 2010; Budianta, 2011; Ghasemi-Fasaei et al., 2012; Arabzai and Honma, 2014). Only the solute component was involved in the adsorption system, whereas the components in the solvent (e.g., distilled water) were ignored or assumed to play no part in the adsorption process. Compared to the

concentration of solute in the bulk solution, this assumption referred to a very dilute solute. Most of the literature was reported without further examination based on such assumptions. Contrary to the excess adsorption isotherm, which assumes that all system components participate in adsorption, this assumption is less accurate. In this study, the pseudo-ideal monolayer adsorption model was utilized for the analysis and interpretation of the adsorption isotherm, as well as to measure the excess isotherm. As all components dissolved in the solution participate in the adsorption process, this model describes binary and multiple adsorption. There is no presumption that only solute components participate in adsorption while solvent components do not (Bono et al., 2008; Bono et al., 2014; Bono et al., 2016).

## 2. MATERIALS AND METHODS

### 2.1 Materials

The chemicals used in this study were copper nitrate trihydrate ( $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ ), lead (II) nitrate ( $\text{Pb}(\text{NO}_3)_2$ ), and zinc (II) nitrate hexahydrate ( $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ). All were purchased from Sigma-Aldrich. The kaolin clay ( $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ ) was purchased from R&M Chemicals, and the river sand from a local sand supplier.

### 2.2 Adsorption study

The experimental measurement of excess adsorption isotherms with some modifications has been explained in detail by Tan et al. (2018, 2021). Individual adsorbate solutions ( $\text{Cu}^{2+}/\text{Pb}^{2+}/\text{Zn}^{2+}$ ) with concentrations ranging from 10 to 1,000 mg/kg were prepared. The weight ratio between the adsorbent and the adsorbate solution was fixed at 1:50. The soil's clay-sand ratio was examined at 1:0, 9:1, 7:3, 1:1, and 0:1. All samples were prepared in triplicate. Each sample was placed in a 250 mL sealed conical flask in an incubator shaker for 24 h at 30 °C and at 100 rpm to achieve equilibrium. The samples were then centrifuged, and the concentrations of the solutions before and after adsorption were determined using inductively coupled plasma-optical emission spectrometry.

### 2.3 Pseudo-ideal monolayer adsorption model

In accordance with Bono et al. (2014), six equations were used to apply the pseudo-ideal monolayer adsorption model. Equation 1, was used to determine the numerical value of excess adsorption isotherm of adsorbate:

$$\Gamma_1 = \frac{W_0(x_0 - x_1)}{W_s} \quad (1)$$

Where  $x_0$  is the initial concentration of adsorbate,  $x_1$  is the concentration of adsorbate in equilibrium,  $W_0$  is the weight of solution, and  $W_s$  is the weight of adsorbent.

Equation 2 was applied to calculate the conformity of pseudo-ideal:

$$\frac{x_1 x_2}{\Gamma_1} = \frac{1}{N_s} \left[ x_1 + \frac{1}{K-1} \right] \quad (2)$$

Where  $K$  is the equilibrium constant,  $N_s$  is the total mole of adsorbed phase (adsorption saturation value), and  $x_2$  is the equilibrium concentration.

Equations 3 and 4 determined the concentration of components 1 and 2 of adsorbed phase (mole fraction),

while Equations 5 and 6 identified the number of moles of components 1 and 2 of adsorbed phase (mol/g):

$$x_1^s = \frac{Kx_1}{1+(K-1)x_1} \quad (3)$$

$$x_2^s = 1 - x_1^s \quad (4)$$

$$n_1^s = N_s \frac{Kx_1}{1+(K-1)x_1} \quad (5)$$

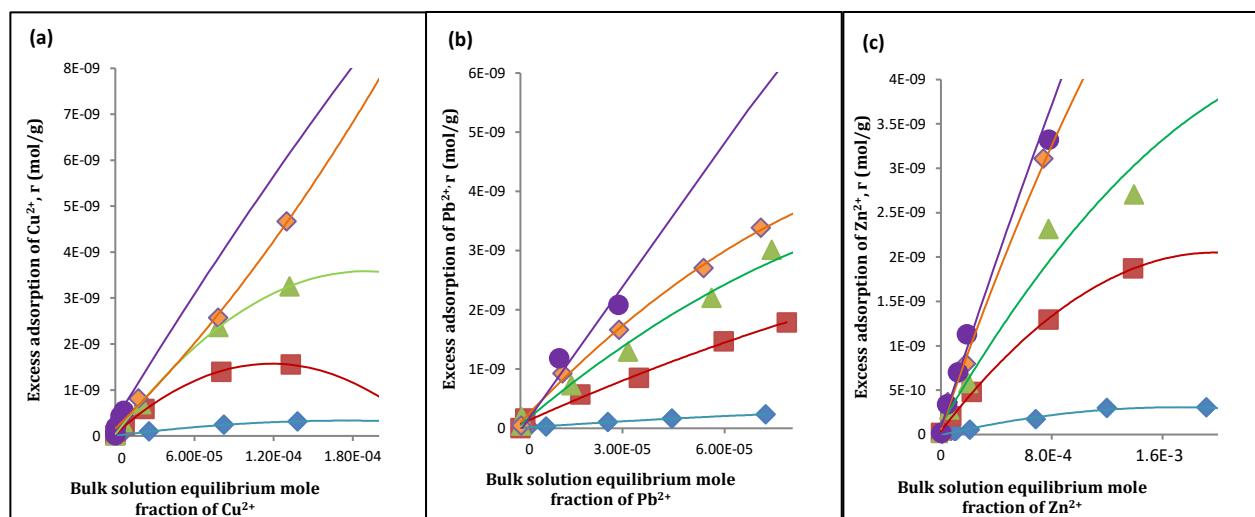
$$n_2^s = N_s - n_1^s \quad (6)$$

Where  $x_1^s$  and  $x_2^s$  are the respective concentrations of components 1 and 2 in the adsorbed phase, and  $n_1^s$  and  $n_2^s$  are the respective moles of components 1 and 2 adsorbed.

### 3. RESULTS AND DISCUSSION

Figure 1 shows the excess adsorption isotherm of binary adsorption of individual  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  ions at different clay–sand ratios. The pseudo-ideal monolayer adsorption model revealed that the excess isotherm obtained from all individual adsorptions of  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  exhibited the same U-shaped pattern, either monolayer type or Langmuir monolayer type (Kippling, 1965). U-shaped isotherms are more favorable for one component according to composite

isotherm (monolayer type) classification. In this case, it refers to the solute, which is the heavy metal ion used. An S-shaped isotherm is preferred for different particular components at different particular concentrations (Song et al., 2013). Therefore, there may be a solute component preference at low solute concentrations and a solvent component preference at high solute concentrations (i.e., low solvent concentration). Consequently, only  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  were adsorbed during each adsorption. The excess adsorption curve formed earlier as the sand composition increased (Figure 1). The formation of the curve indicates a decreasing rate of adsorption and selectivity for the heavy metal involved. As the number of adsorption sites decreased, the adsorption surface period became saturated, and the curve quickly formed. Due to the larger surface area of the clay, the increase in clay composition delayed the formation of the excess adsorption curve (related to the adsorption rate). Excess adsorption of heavy metals on 100% clay was higher than for other mixed compositions. Based on these non-competitive excess adsorption isotherms, the clay–sand ratio in soil and sand has a major impact on the number of adsorption sites on the surface area and the adsorption capacity of metal ions.



**Figure 1.** Excess adsorption isotherm of heavy metal ions (a)  $\text{Cu}^{2+}$ , (b)  $\text{Pb}^{2+}$ , and (c)  $\text{Zn}^{2+}$  onto different compositions of clay and sand at 30 °C. (Note:  $\blacklozenge$  sand,  $\blacksquare$  1:1 (clay:sand),  $\blacktriangle$  7:3 (clay:sand),  $\blacklozenge$  9:1 (clay:sand), and  $\bullet$  clay)

The equilibrium constant ( $K$ ) and adsorption capacity ( $N_s$ ) yielded from the slope and intercept of the plot for different compositions of clay and sand were obtained using linear regression of the pseudo-ideal monolayer adsorption model (Equation 2). All data of individual adsorption of  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  onto mixed compositions of clay and sand satisfactorily obtained a Langmuir relationship, and the correlation coefficient ( $R^2$ ) was shown to have a strong or moderate linear relationship (Table 1).  $R^2$  for  $\text{Cu}^{2+}$  adsorption was above 0.82, in all ratios except 9:1, which was 0.70. The  $R^2$  for  $\text{Pb}^{2+}$  adsorption was above 0.76, except in 100% sand, which showed a moderate linear relationship of 0.69.  $\text{Zn}^{2+}$  showed a strong positive linear relationship with an  $R^2$  above 0.74 for all respective compositions. This confirms that the individual non-competitive adsorption of  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  onto different compositions of clay and sand are of monolayer type and conform the pseudo-ideal monolayer adsorption model. The obtained

adsorption capacity values were negligible because all the aqueous speciation in the solution was considered when calculating the concentration. Heavy metal ions, anions, and water molecules were included in calculating the concentration and the mole fraction involved in the adsorption system (Equation 2). Meanwhile, in the conventional adsorption isotherm, only the solute component is assumed to be involved in adsorption. Other present speciation and water molecules from aqueous solution are ignored. The assumption might be valid for a very dilute aqueous solution, or a very low solubility solute in the solution, but it is not applicable in a completely miscible solution. Thus, it is reasonable to include all of the aqueous speciation and water molecules when calculating the concentration in the adsorption system.

The capacity to absorb heavy metals in soil decreased as sand content increased or clay decreased. As the clay–sand ratio changed from 9:1 to 1:1, the adsorption

capacity for  $\text{Cu}^{2+}$  decreased from  $4.4 \times 10^{-4}$  to  $2.1 \times 10^{-4}$  mg/g. In contrast, the  $\text{Pb}^{2+}$  adsorption capacity increased significantly as the ratio decreased from  $1.02 \times 10^{-3}$  mg/g (9:1), to  $9.32 \times 10^{-4}$  mg/g (7:3) and  $5.28 \times 10^{-4}$  mg/g (1:1). However, the  $\text{Zn}^{2+}$  adsorption capacity continued to increase as the ratio increased from  $1.58 \times 10^{-4}$  mg/g (1:1), to  $3.48 \times 10^{-4}$  mg/g (7:3) and  $7.09 \times 10^{-4}$  mg/g (9:1). The difference in adsorption capacity is proportional to the surface area and cation exchangeable adsorbent.

Clay has a specific surface area of  $16.60 \text{ m}^2/\text{g}$  and a pore volume of  $0.10 \text{ cm}^3/\text{g}$ , while sand has a specific surface area of  $0.57 \text{ m}^2/\text{g}$  and a pore volume of  $0.003 \text{ cm}^3/\text{g}$  (Mustapha et al., 2019). These results demonstrate that the number of adsorption sites on the surface of clay is significantly greater than that of sand. Thus, adsorption capacity in soil increases with a higher clay–sand ratio. In addition, cation exchange occurs only on the surface of clay.

**Table 1.** Equilibrium constant ( $K$ ) and adsorption capacity ( $N_s$ ) of different proportions of clay and sand toward the heavy metal ions  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$

Adsorbent Ratio (%)		$K \text{ g/mol}$	$N_s \text{ mg/g}$	$R^2$
	Clay	Sand		
$\text{Cu}^{2+}$	0	100	11,210	0.00003
	50	50	26,065	0.00021
	70	30	10,829	0.00033
	90	10	10,767	0.00044
	100	0	4,113	0.00183
$\text{Pb}^{2+}$	0	100	20,100	0.00007
	50	50	21,700	0.00053
	70	30	22,900	0.00093
	90	10	23,600	0.00102
	100	0	1,110	0.01620
$\text{Zn}^{2+}$	0	100	11,000	0.000025
	50	50	16,600	0.000158
	70	30	10,100	0.000348
	90	10	6,880	0.000709
	100	0	2,490	0.001710

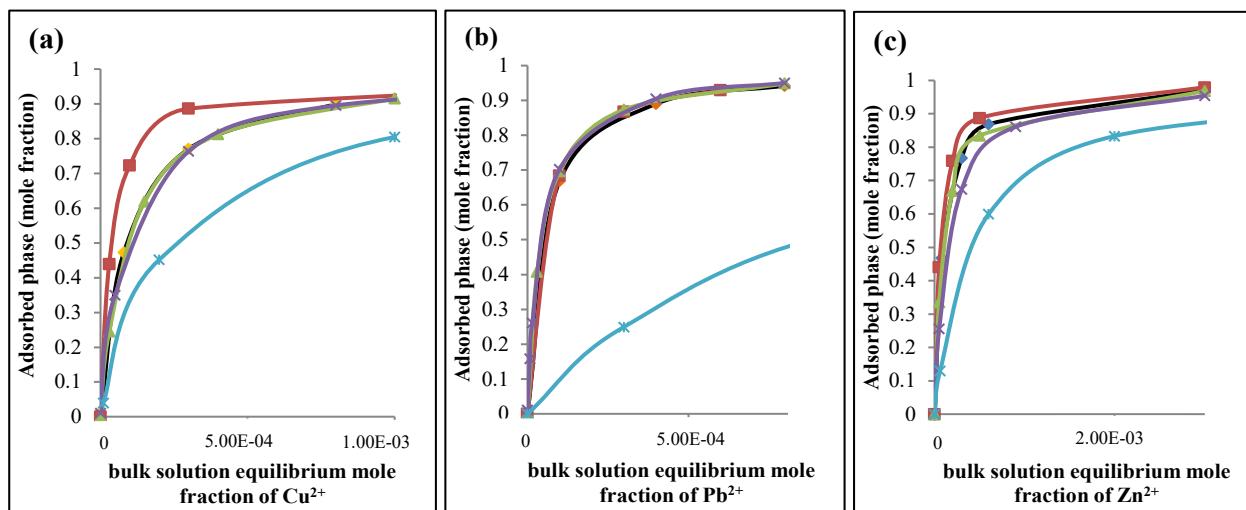
Due to its smaller hydrated ionic size,  $\text{Pb}^{2+}$  has a greater adsorption capacity than  $\text{Cu}^{2+}$  and  $\text{Zn}^{2+}$ , making it more absorbable by clay–sand mixtures. Heavy metal ions are known to become hydrated in aqueous solutions.  $\text{Pb}^{2+}$  has a radius of  $4.01 \text{ \AA}$ , while  $\text{Cu}^{2+}$  and  $\text{Zn}^{2+}$  have radii of  $4.19 \text{ \AA}$  and  $4.01 \text{ \AA}$ , respectively. Consequently, selecting the surface-active sites (hydroxyl group) and the pore size of admixtures with heavy metal ions depends on their hydrated ion radii (Al-Essa and Khalili, 2018). In addition, the ability of  $\text{Pb}^{2+}$  and  $\text{Cu}^{2+}$  surface species to form hydrolyzed complexes on surface-active admixtures increases their adsorption capacity, whereas  $\text{Zn}^{2+}$  only interacts with the adsorption sites of admixtures as divalent ions (Srivastava et al., 2005). Based on the results obtained, the ascending series of non-competitive adsorption onto various mixed clay and sand compositions can be demonstrated as  $\text{Pb}^{2+} > \text{Cu}^{2+} > \text{Zn}^{2+}$ .

Figure 2 depicts the concentrations of  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  in the adsorbed phase. The selectivity toward  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  was exceptionally high at relatively low solution concentrations. At low concentrations, heavy metals were predominantly adsorbed onto the adsorption sites of clay and sand, whereas at higher concentrations, clay and sand lost their ability to bind to heavy metals. The degree of selectivity of  $\text{Cu}^{2+}$  and  $\text{Zn}^{2+}$  was greatest at a 1:1 clay–sand composition ratio. The

degree of selectivity of  $\text{Pb}^{2+}$  was not markedly different. However, as the composition ratio of clay increased, selectivity gradually decreased. Clay had the lowest selectivity of the three heavy metals. The large surface area increased the number of adsorption sites and decreased the selectivity of the adsorbent. Consequently, the large surface area increased the equilibrium mole fraction during the adsorption phase.

#### 4. CONCLUSION

Pseudo-ideal monolayer adsorption theory is applicable in the adsorption of heavy metals  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  onto mixed compositions of clay and sand. The performance of high and moderately high  $R^2$  in the linear form indicated that the obtained data were satisfactorily created a Langmuir relationship. Adsorption capacity increased as the component of clay increased in the clay–sand admixture. The difference in adsorption capacity was related to the higher surface area and cation-exchangeable clay. This study's descending adsorption capacity of heavy metals in different clay and sand compositions is  $\text{Pb}^{2+} > \text{Cu}^{2+} > \text{Zn}^{2+}$ .



**Figure 2.** Selectivity of metal ions (a)  $\text{Cu}^{2+}$ , (b)  $\text{Pb}^{2+}$ , and (c)  $\text{Zn}^{2+}$  onto different compositions of clay-to-sand ratio at 30 °C  
Note:  $\diamond$  sand,  $\blacksquare$  1:1 (clay:sand),  $\blacktriangle$  7:3 (clay:sand),  $\times$  9:1 (clay:sand), and  $\ast$  clay

## ACKNOWLEDGMENT

The authors would like to acknowledge the financial support of Universiti Malaysia Sabah under the Skim Penyelidikan Bidang Keutamaan grant (SBK0486-2021).

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