

RECYCLING WASTEWATER SLUDGE FROM MELAMINE
-COATED PAPER FACTORY FOR AGRICULTURAL USE

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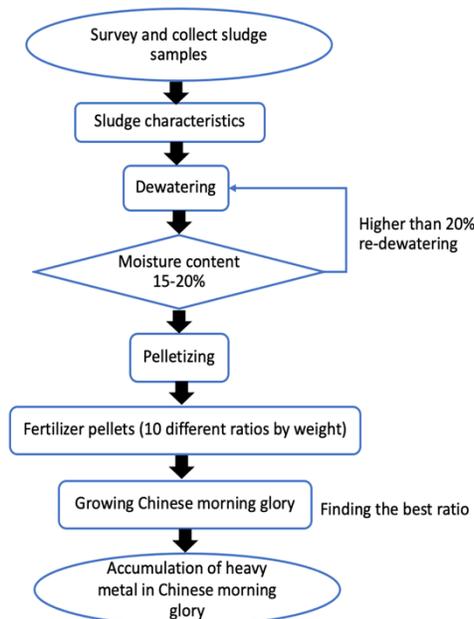
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Graphical abstract



Abstract

This research aims to study the characteristics of adhesive sludge from a melamine-coated paper factory after dewatering and investigate the potential of adhesive sludge for agriculture use as fertilizer pellets. The adhesive sludge was collected from the final sedimentation pond, where the moisture content exceeded 70%. To prepare the sludge for pelletization, it was dewatered using a centrifuge until the moisture content was reduced to 15-20%. The sludge was then evaluated based on pH, organic matter content, Total N, P, K, and heavy metal contents. All parameters met the material standards for fertilizer production set by Department of Agriculture, Thailand [1], indicating the sludge’s potential as a fertilizer or soil conditioner. Fertilizer pellets were produced by mixing the sludge with organic or chemical fertilizers in various ratios (7:3, 6:4, and 5:5 by weight) and tested by cultivating Chinese morning glory. The experiment showed that the optimal ratio for mixing chemical fertilizer with adhesive sludge was 5:5, which have the most nutrients in fertilizer pellets such as Total N P K and Organic matter that produced the highest fresh weight and plant height of Chinese morning glory. Statistical analysis using Completely Randomized Design (CRD) and one-way ANOVA, conducted with SPSS showed no significant difference in the height, leaf size (width and length), root length, trunk circumference, and number of leaves of Chinese morning glory between non-mixed sludge and non-mixed organic fertilizer pellets. These results indicate that organic fertilizer pellets can be substituted with sludge fertilizer pellets. Furthermore, substituting organic fertilizer pellets with sludge-based pellets and reducing chemical fertilizer usage through a 5:5 sludge-to-chemical fertilizer ratio was feasible without compromising plant growth. No significant difference at the 95% confidence level was observed, indicating equivalent performance between non-mixed chemical fertilizer pellets and those mixed with adhesive sludge. This finding suggests that adhesive sludge could mitigate chemical fertilizer dependency, lowering costs and preventing soil degradation. Although formaldehyde and heavy metal contents in Chinese morning glory may be influenced by cultivation methods and environmental factors, the levels remained within the food safety standards established by the Ministry of Public Health’s, Thailand [2]. Despite meeting safety standards, it is recommended to prioritize using sludge-based pellets for cultivating ornamental plants or flowers, as this application reduces concerns regarding potential formaldehyde and heavy metal accumulation in edible crops. These results highlight the feasibility of recycling adhesive sludge as an eco-friendly and cost-effective agricultural resource.

Keywords: Sludge, Dewatering, Pellet, Waste management, Agricultural use, Melamine-coated paper factory, Chinese morning glory

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1.0 INTRODUCTION

Sludge generated from industrial production processes has become a significant challenge for disposal. As the industrial sector rapidly expands to accommodate a growing population,

the result is an increase in industrial waste sludge with relatively low utilization rates compared to the amount produced [3,4]. Landfilling is the most commonly used method for sludge disposal [5]. However, it is costly and releases pollutants into the environment, posing risks to human health [6]. The landfill

process involves two steps: first, the sludge is usually treated by anaerobic decomposition to break it down before hardening, and then it is buried in a containment area lined with materials like high-density polyethylene to prevent leaks. Landfilling requires substantial space, and if the containment is not adequately constructed, toxic substances (organic acids, aldehyde, alcohols) may leach into the environment, causing harm.

Therefore, reducing the amount of sludge that ends up in landfills and discovering new ways to dispose of it without harming the environment are both regarded as effective and sustainable solutions.

A more sustainable approach to managing industrial sludge involves exploring various possibilities that allow for sludge disposal without generating pollution, as well as the potential for transforming sludge into new products [7]. Before sludge can be utilized, it must undergo processing to remove water, reduce pathogens, and minimize odor. Understanding the characteristics and composition of the sludge is crucial for selecting an appropriate treatment method. There are multiple methods for utilizing and disposing of sludge, including converting it into fertilizer pellets, mixing it with concrete to create lightweight construction materials, composting (especially for sludge with high organic content), using sludge as fuel through pyrolysis and gasification, or incinerating it to harness heat energy for other processes. Recycling waste back into the production cycle can also improve the financial performance of the industry [8]. However, depending on the sludge's characteristics and the treatment method used, there may be specific limitations on its application [9].

The melamine-coated paper factory produces melamine-coated paper used for surface laminating on Medium Density Fiberboard (MDF), particle board and other types of plywood board, utilizing advanced production techniques and efficient management system. The factory incurs significant costs to transport and dispose of adhesive sludge, a by-product of the melamine-coated paper production process, to off-site disposal. This adhesive sludge, which is generated during machinery cleaning after changes in adhesive paper formulations, accounts for 66% of the hazardous waste produced by the factory. Currently, the factory seeks to reduce the costs associated with both the transportation and off-site disposal of adhesive sludge. However, there is a lack of sufficient knowledge and techniques for effectively managing and recycling the adhesive sludge for reuse.

Melamine coated paper or Melamine resin is versatile plastic widely used in the manufacture of kitchenware, including spoons, forks, chopsticks, plates, and bowls. It serves as a key material in the production of Formica sheets, melamine laminate flooring, and whiteboards. Additionally, melamine resin is commonly used as a surface coating for particleboards in the furniture industry.

Melamine is combined with formaldehyde to create melamine resin, a robust thermosetting plastic, and melamine foam, a specialized polymer widely employed for cleaning applications [10].

Formaldehyde (HCHO) is a colorless, flammable gas at ambient temperature, characterized by a pungent odor [11]. Its exposure may lead to several adverse health effects. Formaldehyde is commonly found in the resins used in the production of composite wood products, including hardwood

plywood, particleboard, and medium-density fiberboard (MDF) [12].

As an organic compound, formaldehyde is naturally occurring in numerous food items like apples, bananas, pears, spinach, carrots, beef, poultry, fish, and coffee beans, where it is generated as a byproduct of metabolic processes. While food sources contain relatively low levels of formaldehyde, heightened concentrations of this compound can trigger severe health complications. Specifically, formaldehyde can be found in food products at concentrations ranging from 300 – 400 mg/kg in items such as fruits (e.g., pear, apple, green onion), meats, fish, crustaceans, and dried mushrooms. It is produced as a byproduct of metabolic processes. Although food contains relatively low levels of formaldehyde, high concentrations of the compound can cause severe health issues. Frequent exposure to formaldehyde can lead to significant health risks; for instance, a concentration of 50 mg/kg may result in pulmonary reactions, whereas exposure at concentrations of 100 mg/kg is deemed highly toxic and potentially life-threatening [12].

The physical properties of adhesive sludge from melamine coated-paper factory are typically consist of 70% water by weight, much of which can be separated through gravity sedimentation. After undergoing the water separation process, the moisture content should reduce to approximately 20% [13] for further use. The remaining moisture content is a crucial factor in selecting the appropriate sludge disposal technology and determining its potential utilization. For instance, when sludge is intended for use as fuel or in fertilizer production, moisture reduction is essential prior to use. Sludge that has undergone water separation will typically clump into a mass with a texture similar to clay, often referred to as "cake". This cake will harden after further drying or moisture reduction, eventually resembling a fine powder mixed with small lumps. The dewatering process is beneficial as it helps to reduce both transportation and disposal costs. Various techniques are employed to extract water from sludge, including physical methods such as percolation through a sand layer using a sludge drying bed, and mechanical methods such as belt filter press, vacuum filter, and centrifuge.

Previous studies have explored the potential applications of industrial sludge. Among the various methods of sludge management, agriculture is considered the least expensive, as sludge contains essential nutrients for plants, including nitrogen, phosphorus, potassium, and other organic compounds [15-16]. However, before sludge can be used in fertilizer production, it must undergo dewatering [17]. Additionally, there are several important considerations to address, such as the sludge's properties, odor, the hazardous nature of organic materials, salts, and heavy metals present in the sludge [18-19].

From the results of the study evaluating the feasibility of producing organic chemical fertilizers (Organo-Mineral Fertilizers) from dried wastewater sludge and mixed with poultry litter ashes and chemical fertilizers, the goal was to identify a formula suitable for growing corn. The organic matter in the fertilizer was measured to exceed 20%, as the incorporation of poultry manure into the sludge-based fertilizer helped increase the organic content [20]. Additionally, a study on the effects of fertilizer pellets derived from the pulp and paper industry on corn growth and yield found that using a combination of sludge pellets and chemical fertilizers was as effective as using chemical fertilizers alone. This resulted in the largest pod and seed size, as

well as the greatest plant height across all formulations tested [21].

When using sludge to produce fertilizer pellets, the sludge must first be prepared as a raw material by addressing moisture content, particle size, and contamination (Such as dust, stones, sand, and metals). Before the sludge can be utilized for pellet production, contaminants should be removed using sieves, screens, or metal-removal equipment. Additionally, the raw materials must be free from pathogens, insects, and weeds. The carbon-to-nitrogen (C/N) ratio should be increased by mixing the sludge with other organic materials to enhance decomposition efficiency. The sludge is then compressed into molds, and its moisture content must be reduced to 10-20% [22].

The dried sludge is ground into a fine powder. In some cases, when the raw materials are less adhesive, a binder may be needed to increase the bonding strength of the pellets. Commonly used binders include clay and tapioca starch. After the compression process is complete, the pellets must undergo cooling and screening (to remove improperly sized pellets) before packaging and storage. The significance of pelletizing fertilizers lies in their ability to release essential nutrients, such as nitrogen, phosphorus, and potassium, more effectively, making them easier for plants to absorb. Pelletizing also helps slow down nutrient loss and improves soil quality.

Chinese morning glory, also known as Chinese water spinach (*Ipomea aquatica*), is a vegetable widely consumed and cultivated in many Asian countries, particularly in Thailand, China, and Vietnam. It is commonly used in growth studies due to its rapid growth, adaptability, and relevance to environmental and agricultural research. It grows quickly, with a harvest cycle of 20-30 days, making it suitable for experiments requiring fast results. Its ability to thrive in diverse environments, including nutrient-poor or contaminated conditions, allows researchers to evaluate the effects of fertilizers, soil amendments, and water quality. Additionally, Chinese morning glory is known for accumulating heavy metals, making it valuable for studies on phytoremediation and pollution monitoring. As an economically and nutritionally important vegetable in Asia, research on its yield and safety for consumption is highly relevant. Its responsiveness to fertilizers and cost-effectiveness further makes it an ideal model for plant growth experiments [23-25].

Chinese morning glory has been studied for its role in wastewater treatment and agriculture. It effectively removes nutrients from aquaculture wastewater and grows well in sludge-amended media, improving sustainability [23-24]. However, sewage sludge can increase heavy metal accumulation, requiring careful management [25-27]. These findings highlight its potential in phytoremediation and sustainable agriculture.

To better understand the characteristics of adhesive sludge discharged from a melamine-coated paper wastewater treatment plant which generates the largest quantity of sediment, dewatering process was initiated to form this sludge into fertilizer pellets, thereby enhancing the factory's capacity for sustainable waste management [27]. Thailand's climate, characterized by high temperatures and abundant sunlight throughout the year, is ideal for agriculture and helps reduce pathological issues. In southern Thailand, frequent rainfall further supports agricultural activities by easing the burden on farmers. Utilizing sludge from wastewater treatment systems in agriculture is more economically viable than other methods and poses minimal environmental challenges [28].

In addition to maintaining appropriate moisture content throughout the production process, several other factors must be considered when utilizing sludge for pellet fertilizer production. For environmental impact and food safety concerns, it is essential to assess the accumulation of heavy metals in both plants and the surrounding environment. As noted, the adhesive sludge contains melamine resin, formaldehyde, and heavy metals. Melamine resin naturally degrades and is not absorbed by plants, while formaldehyde is a volatile compound that can decompose through natural processes.

To ensure the safe use of sludge as fertilizer, heavy metal concentrations must comply with the standards set by the Department of Agriculture, Thailand. Furthermore, the accumulation of heavy metal and formaldehyde in plants should be monitored to determine compliance with food safety standards, ensuring the safety of agricultural products for human consumption.

2.0 METHODOLOGY

This study explored the production method and quantity of adhesive sludge using a backhoe to dredge the sludge. Adhesive sludge was collected in containers and stored at 5-10°C prior to laboratory analysis. Various characteristics of the sludge were measured before initiating the dewatering process to create fertilizer pellets. The pellets were then tested by growing Chinese morning glory to study its growth, analyze the accumulation of heavy metals in the plants, and analyze formaldehyde content.

2.1 Studying on Dewatering Sludge

2.1.1 Chemical Analysis Of Raw Materials

Experiments were conducted in the chemistry laboratory of the Faculty of Engineering, Prince of Songkla University. The concentrations of total organic carbon, total nitrogen, total phosphorus, total potassium and heavy metals in the samples were determined using ICP-OES analysis. The concentration of formaldehyde was measured using a colorimetric method. Moisture content was determined using the gravimetric method, and pH was measured with a portable pH meter. Each measurement was duplicated.

2.1.2 Dewatering

The adhesive sludge was dewatered using a centrifuge at a speed of 2,000 – 3,000 rpm for 5, 10, 15, 20, 25, 30, 35 and 40 mins to determine the optimal speed for water separation, producing sludge suitable for pelletization (moisture content of 15–20%) [28].

A volume of 8 ml of adhesive sludge sample was placed in a 14 ml polystyrene centrifuge tube and maintained at 30°C. The temperature should not be set too low to prevent the tube from becoming brittle, and both temperature and speed were carefully controlled to preserve the sample's integrity. After centrifugation, the moisture content of the adhesive sludge was measured to ensure it met the required range before proceeding with the pelletizing experiment.

2.2 Studying on pelletizing sludge and potential of fertilizer pellets

2.2.1 Pelletizing

The sludge was mixed with organic fertilizer or chemical fertilizer in different ratios. A total of 10 mixture ratios were designed for comparison, including: N (None Fertilizer), S (Sludge), CF (Chemical fertilizer), OF (Organic fertilizer), SCF1 (Sludge + Chemical fertilizer 7:3 by weight), SCF2 (Sludge + Chemical fertilizer 6:4 by weight), SCF3 (Sludge + Chemical fertilizer 5:5 by weight), SOF1 (Sludge + Organic fertilizer 7:3 by weight), SOF2 (Sludge + Organic fertilizer 6:4 by weight), SOF3 (Sludge + Organic fertilizer 5:5 by weight).

The ratios of 7:3, 6:4 and 5:5 was selected to facilitate experiment and clearly observe differences in outcomes, such as heavy metal accumulation and nutrient content in the pellet fertilizer. This approach helps identify the optimal ratio or inform the design of a more effective blend. Adhesive sludge, chemical fertilizer, and organic fertilizer were ground into a fine powder and thoroughly mixed, with each mixture weighing 3 kg (dry weight). The 15-15-15 (N-P-K) chemical fertilizer formula was chosen for its suitability for vegetable cultivation, while the organic fertilizer met the Department of Agriculture's standard, with an organic matter concentration of at least 20%.

The mixtures were pelletized using a mold with a 10 mm diameter. If the materials were not sufficiently cohesive, a binding agent, such as clay or starch, was added at a concentration of at least 3% to enhance the bonding force between particles and improve pellet strength and durability. After pelletizing process, the pellets were placed in sunlight for at least 6 hours to increase their toughness.

2.2.2 Compare The Potential Of The Different Fertilizer Pellets

The study involved analyzing the density, pH, organic matter and organic carbon, total nitrogen, phosphorus, potassium and moisture content of the pellet fertilizers. Chinese morning glory was planted to assess the effectiveness of each fertilizer formula. Data on the morphological growth of the plants-including the number, width and length of leaves, trunk circumference, and root length-were collected and compared against the control (N; None Fertilizer), CF (Chemical fertilizer), OF (Organic fertilizer) and S (Sludge).

The Chinese morning glory cultivation experiment was designed by adding 6 kg of soil to trays measuring 38x50x20 cm. Forty seeds were sown per tray in a shaded area to allow for the selection of 40 healthy seedlings. After 10 days, the seedlings were relocated to a sunny environment, with watering performed 1-2 times/day. On the 11th day, 120 g of fertilizer pellets were applied to each tray.

Twenty seeds were planted per tray. Data were collected at 21-30 days or when the Chinese morning glory plants reached a height of about 1 foot, with measurements taken three times in total. The plant height was measured every 5 days to compare growth rates. The data were analyzed using a Completely Randomized Design (CRD) and one-way ANOVA with the SPSS program.

2.3 Studying the Concentrations of Heavy Metal in The Fertilizer Pellets

2.3.1 Heavy Metal Analysis

Chinese morning glory plants harvested on the last day were examined for the accumulation of heavy metals. The Chinese morning glory should be washed twice with tap water and once with distilled water and then dried before shredded. The Chinese morning glory is then divided into four parts-trunk, leaves, shoots, and roots-to assess the accumulation of heavy metals in each part. Weigh 1 g of the shredded Chinese morning glory and dry it for 1 hour at 100°C in a hot air oven. The prepared samples are sent to the Standard Inspection and Certification Service Center at the Faculty of Science, Prince of Songkla University for heavy metal analysis.

The digestion process involves placing the shredded Chinese morning glory in a beaker, adding 1 ml of concentrated nitric acid (HNO₃), and leaving it in a fume hood for 24 hours. The solution is then filtered using Whatman filter paper No. 1 and transferred to a 50 ml bottle. The volume is adjusted with 0.01M HNO₃ before the solution is analyzed using an ICP-OES machine.

2.3.2 Formaldehyde Analysis

Fresh Chinese morning glory samples were harvested on the final day, cut, and weighed to 5 g. The samples were soaked in 100 ml of distilled water before being sent for analysis at the Inspection Service Center, Faculty of Science, Prince of Songkla University, using a formaldehyde test kit technique. The experiment focused on measuring formaldehyde accumulation in morning glory grown with only adhesive sludge fertilizer, compared to plants fertilized with the lowest sludge content in the pellets (e.g., a 5:5 ratio of sludge mixed with either chemical or organic fertilizer). The levels of formaldehyde and heavy metals accumulated in Chinese morning glory were evaluated and compared against the Ministry of Public Health's food contamination standards to assess compliance with safety regulations.

3.0 RESULTS AND DISCUSSION

3.1 Quantity and Type of Adhesive Sludge

Adhesive sludge from the wastewater treatment plant of a melamine-coated paper factory is produced at a rate of approximately 30,000 kg/month, averaging 1,000 kg/day or 1 ton/day. The adhesive sludge is classified under the 6-digit waste code 08 04 11 HM, indicating the presence of organic solvents in the sealant or other materials. The HM designation refers to hazardous waste, which can be tested and analyzed to determine its potential harm.

According to the Ministry of Industry's regulations, the prescribed disposal method for adhesive sludge with the code 08 04 11 HM is "042", which involves either incineration or landfill disposal. Currently, the factory transports the sludge to a distant disposal facility, resulting in high disposal costs.

3.2 Characteristics of Adhesive Sludge

The physical characteristics of the sludge are coagulated, with a white/gray color and an unpleasant odor. Chemically, the sludge contains 37.62% organic matter and has a pH value of 8.26, indicating a slightly alkaline nature. Both parameters meet the Department of Agriculture's standards. The sludge was found to contain 21.82% nitrogen, which plants can absorb through their leaves, along with 0.01% phosphorus and 0.04% potassium, though the phosphorus and potassium content remains below the required standards.

Six heavy metals were tested: arsenic, cadmium, copper, chromium, lead, and mercury. Chromium was present at 1.08 mg/kg, copper at 5.98 mg/kg, and lead at 21.4 mg/kg, all of which are well below the Department of Agriculture's safety threshold of 500 mg/kg. Therefore, the heavy metal concentrations in the sludge fall within safe limits for fertilizer production. Additionally, the sludge has a high moisture content of 79.75%, preventing direct extrusion into fertilizer pellets. Consequently, the sludge must undergo a dewatering process to reduce moisture content before use in fertilizer production.

3.3 Sludge Dewatering and Suitable Conditions for Pelletizing

According to previous studies, a moisture content of 15–20% is considered optimal for pelletizing [29]. However, due to the unique characteristics of adhesive sludge, it was observed that after centrifugation at 2,800 rpm for 30 mins (the maximum speed achievable with a laboratory-scale centrifuge), the sludge separated distinctly from the liquid phase. Despite this separation, the remaining sediment retained a moisture content of approximately 53%, making it unsuitable for pelletizing. To overcome this issue, the sludge underwent further treatment in a water bath at 95°C for 3 hours, resulting in a dried sludge with a hard, lump-like appearance. This dried sludge could then be ground into a fine powder with a moisture content of approximately 20%, suitable for pellet production.

3.3.1 Application of Suitable Dewatering Conditions for Industrial Plants.

Studies have shown that the optimal moisture content for adhesive sludge pelletization is between 15–20%. For an industrial plant producing 1 ton of sludge/day, the following dewatering machines are recommended:

- Belt-Filter Press: This system is widely used in large-scale plants for sludge handling. It offers high capacity, low energy consumption, and operates with minimal noise. A belt-filter press with a width of 1 m can dewater sludge at a rate of 90–400 kg/hr, producing sludge with a moisture content of 18–25%, depending on the rolling speed settings.

- Centrifuge: This system uses centrifugal force to separate water from sludge and is commonly applied in industries such as food processing, wastewater treatment, and textile production. While centrifuges can handle large quantities of sludge, their suitability depends on the specific characteristics of the material, and certain operational factors must be carefully considered.

3.4 Pelletizing

The moisture content of the dry sludge powder was 15–20%, allowing it to be pelletized without the need for additional water

or binding agents. Since this sludge is adhesive in nature, it has inherent binding properties, making it possible to form pellets without any additives, as shown in Figure 1. Pelletizing was carried out using a portable compactor with a diameter of 10 mm, as shown in Figure 2.



(a) (b) (c)
Figure 1 Fertilizer Pellets: (a) S (Sludge), (b) SOF3 (Sludge + Organic Fertilizer 5:5 by weight) and (c) SCF3 (Sludge + Chemical Fertilizer 5:5 by weight)

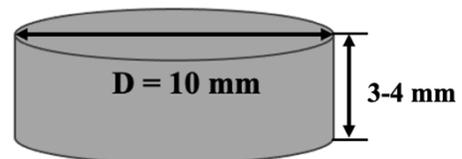


Figure 2 Size of the pelletized fertilizer

3.5 Characteristics of Fertilizer Pellets

After drying the fertilizer pellets in the sun, pH values were found to range between 5.5–8.5, meeting the standards set by the Department of Agriculture. As shown in Table 1, fertilizer pellets made solely from adhesive sludge (S) exhibited the highest pH value at 8.25, classifying them as neutral to slightly alkaline. In contrast, the chemical fertilizer (CF) had the lowest pH value at 6.99. It was observed that blending adhesive sludge with chemical or organic fertilizers in various ratios effectively reduced the sludge's pH, bringing it closer to neutral. This demonstrates a direct correlation between the sludge proportion in the pellet and the resulting pH level.

The pellets were further analyzed for key plant nutrients, including organic matter and organic carbon. The results indicated that the adhesive sludge (S) contained higher levels of organic matter and organic carbon (16.78%) compared to chemical fertilizers (CF) (13.14%) and commercial organic fertilizers (OF) (3.96%). Mixing adhesive sludge with chemical fertilizers proved to be highly effective, as the combination not only increased the organic matter content but also enhanced the N, P, and K levels of the resulting pellets. This blending approach compensates for the low organic content in chemical fertilizers while boosting the nutrient profile of the sludge. Furthermore, the nutrient content of organic fertilizers (OF) was found to be comparable to that of adhesive sludge (S). These findings suggest that adhesive sludge can serve as an effective substitute for conventional organic fertilizers. For targeted plant growth, using a blended fertilizer pellet – containing both adhesive sludge and either chemical or organic fertilizer – is recommended, as it provides more balanced nutrient availability and superior results compared to using either component alone. Additionally, mixing adhesive sludge with chemical fertilizers

reduces the overall chemical fertilizer usage, contributing to more sustainable agricultural practices.

Regarding the main nutrients-total nitrogen, total phosphorus, and total potassium-the fertilizer pellets made from adhesive sludge met all the standards set by the Department of Agriculture, as shown in Table 1.

Table 1 Characteristics of fertilizer pellets

Type	Ratio Sludge+ Fertilizer (by weight)	pH	OM (%)	OC (%)	Total N (%)	Total P (%)	Total K (%)
N	0:10	-	-	-	-	-	-
S	0:10	8.25	16.78	9.75	32.27	0.07	0.01
CF	0:10	6.99	3.96	2.30	12.77	12.86	21.50
OF	0:10	7.83	13.14	7.64	1.10	0.59	0.97
SCF1	7:3	7.37	15.85	10.38	23.11	4.00	7.64
SCF2	6:4	7.41	14.14	8.22	22.72	5.19	7.65
SCF3	5:5	7.42	11.90	6.92	21.59	6.74	8.10
SOF1	7:3	8.20	16.72	9.84	23.92	0.22	0.39
SOF2	6:4	8.16	16.47	9.57	20.55	0.29	0.44
SOF3	5:5	8.13	16.15	9.39	16.53	0.33	0.47

3.6 Cultivation of Chinese Morning Glory

Chinese morning glory was cultivated in trays for approximately 3 weeks before being harvested. Typically, the plants reach a height of about 1 ft by the time of harvest.

3.7 Growth of Chinese Morning Glory Plant

The study on the growth of Chinese morning glory revealed that during the first 5-10 days, the plants exhibited similar average heights, ranging from 5-8 cm. However, after the application of fertilizer on the 11th day, a noticeable increase in growth rate was observed between day 10 and 20. The most significant growth was recorded in the plants treated with SCF3 (Sludge + Chemical fertilizer at a 5:5 ratio by weight). By day 25, these plants exhibited the tallest stems, followed in descending order by those treated with CF (Chemical fertilizer), SOF3 (Sludge + Organic fertilizer at a 5:5 ratio), SOF2 (Sludge + Organic fertilizer at a 6:4 ratio), SCF2 (Sludge + Chemical fertilizer at a 6:4 ratio), SCF1 (Sludge + Chemical fertilizer at a 7:3 ratio), OF (Organic fertilizer), S (Sludge) and SOF1 (Sludge + Organic fertilizer at a 7:3 ratio). The control group (N; None fertilizer) exhibited the least growth, with shortest stems at 25 days. These results are illustrated in Figure 3.

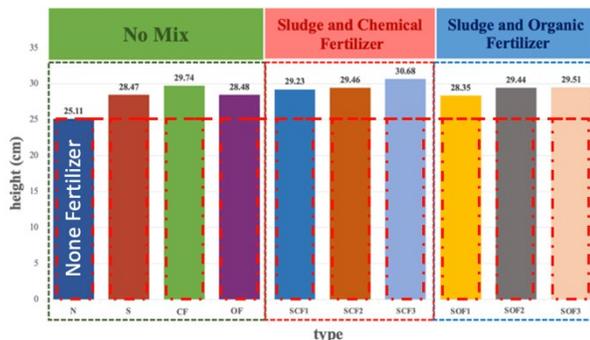


Figure 3 Height of Chinese morning glory plants on Day 25

As show in Figure 4, the fresh weight of Chinese morning glory plants at Day 25 reached its maximum in those treated with SCF3 (Sludge + Chemical fertilizer at a 5:5 ratio by weight). This result is consistent with the height data presented in Figure 3, which demonstrated that plants grown with SCF3 exhibited the highest growth rate. In contrast, the control group (N; None Fertilizer) had the lowest fresh weight. Therefore, it can be concluded that mixing sludge with fertilizer in the SCF3 ratio yields the best agricultural outcomes.

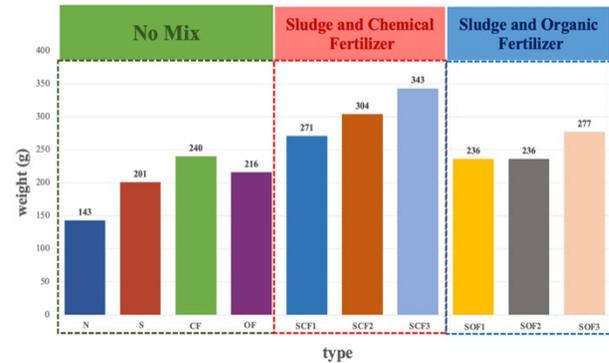


Figure 4 Fresh weight of Chinese morning glory plants on Day 25

3.8 Statistic analysis

The experiment was designed using a Completely Randomized Design (CRD), often referred to as a completely randomized trial strategy. This approach is employed when the experimental units are as uniform or similar as possible. Experimental units were randomly assigned to treatments were specified: N (None Fertilizer), S (Sludge), CF (Chemical fertilizer), OF (Organic fertilizer), SCF1 (Sludge + Chemical fertilizer 7:3 by weight), SCF2 (Sludge + Chemical fertilizer 6:4 by weight), SCF3 (Sludge + Chemical fertilizer 5:5 by weight), SOF1 (Sludge + Organic fertilizer 7:3 by weight), SOF2 (Sludge + Organic fertilizer 6:4 by weight), SOF3 (Sludge + Organic fertilizer 5:5 by weight). The experiment was replicated three times, with data analyzed using Excel and a one-way analysis of variance (ANOVA) conducted in SPSS.

The analysis assumed that the various ratios of fertilizer pellets made from sludge were related to the growth data of Chinese morning glory. The results were used to determine whether the different ratios of fertilizer pellets had a statistically significant effect on plant growth. The findings indicated significant differences in growth rates across treatments at the 0.05 level of significance, corresponding to a 95% confidence level. This demonstrates that the different fertilizer ratios had varying effects on the growth of Chinese morning glory plants. The P-Value (Sig.) was less than 0.05, indicating a significant difference in means between at least one pair of treatments, although it does not specify which pair. The detailed results are presented in Table 2.

Table 2 Results of ANOVA

Parameter	Variance	SS	Df	MS	F	P
Height	Between Groups	60.213	9	6.690	701.052	.000
	Within the Groups	0.191	20	0.010		
	Total	60.404	29			
Leave Width	Between Groups	0.562	9	0.062	3.761	.007
	Within the Groups	0.332	20	0.017		
	Total	0.894	29			
Leave Length	Between Groups	55.577	9	6.175	9.983	.000
	Within the Groups	12.372	20	0.619		
	Total	67.949	29			
Number of Leaves	Between Groups	8.535	9	0.948	5.491	.001
	Within the Groups	3.454	20	0.173		
	Total	11.989	29			
Roots	Between Groups	18.589	9	2.065	18.275	.000
	Within the Groups	2.260	20	0.113		
	Total	20.850	29			
Trunk Circumference	Between Groups	0.966	9	0.107	103.573	.000
	Within the Groups	0.021	20	0.001		
	Total	0.987	29			

*At confident level .05

Based on the experimental results, it was observed that each fertilizer pellet ratio exhibited at least one pair of mean differences, although the specific pairs were initially unknown. Therefore, a pairwise comparison was conducted using the Least Significant Difference (LSD) test, which is the most liberal statistical test for comparing paired means, minimizing the risk of overlooking significant differences. The LSD test results, presented in Table 3, demonstrated that the fertilizer ratio had a significant effect on the height of Chinese morning glory plants at the 0.05 significance level. Notably, the treatments S, OF, and SOF1 did not show statistically significant differences from one another, indicating that plants grown with S (Sludge), OF (Organic fertilizer), or SOF1 (Sludge + Organic fertilizer 7:3 by weight) exhibited similar growth outcomes in terms of height.

These findings suggest that adhesive sludge can effectively replace organic fertilizer or be combined with it to reduce agricultural dependence on organic fertilizers. Among the tested

ratios, SCF3 (Sludge + Chemical fertilizer 5:5 by weight) produced the most favorable results, yielding the tallest plants. This highlights the potential of using sludge in combination with chemical fertilizer to enhance plant growth.

When comparing the average leaf size, no significant differences were observed, as the average leaf width across all fertilizer ratios was very similar. However, the fertilizer ratio that resulted in the greatest average leaf width was OF (Organic Fertilizer). Therefore, mixing adhesive sludge with either organic or chemical fertilizers can help increase the leaf size of Chinese morning glory, likely due to the high nitrogen content in adhesive sludge, which promotes leaf growth.

3.9 Heavy Metal Accumulation in Chinese Morning Glory Plants

3.9.1 Heavy metal

After harvesting the Chinese morning glory plants, heavy metal accumulation was analyzed in various plant parts-trunk, leaves, shoots, and roots (Table 4-7, Figure 5-9). The summarize results are as follow:

- Heavy metal accumulation: The analysis confirmed that the levels of arsenic, cadmium, chromium, lead, copper, and mercury in the plants complied with the Ministry of Public Health's food contaminant standards, indicating they are safe for consumption.

- Copper accumulation: Copper was found to be the most accumulated heavy metal across all fertilizer treatments. The highest copper levels were detected in the trunks and leaves of plants grown without fertilizer. In contrast, copper accumulation decreased when plants were fertilized with sludge (S), suggesting that the soil may naturally contain high copper levels, but the use of sludge, organic, and chemical fertilizers helped reduce copper uptake.

- Arsenic and Mercury: Arsenic was not detected in the trunks or shoots of plants, and mercury was not found in all plant parts across treatments.

- Fertilizer impact: The use of sludge, chemical fertilizers, and organic fertilizers led to copper accumulation in the leaves, though all levels remained within the Ministry of Public Health's safety limits. Notably, combining sludge with fertilizers generally reduces heavy metal accumulation compared to using sludge alone.

In summary, none of the tested fertilizers caused heavy metal concentrations to exceed safety standards, indicating their suitability for agricultural use. However, to prevent potential health impacts, it is recommended to use them for growing ornamental plants and flowers rather than food crops.

Table 3 Results of LSD

Parameter	Type	AVG	N	S	CF	OF	SCF1	SCF2	SCF3	SOF1	SOF2	SOF3
Height	N	25.103	-	3.367*	4.640*	3.377*	4.127*	4.367*	5.577*	3.247*	4.333*	4.450*
	S	28.470		-	1.273*	0.100	0.760*	0.990*	2.210*	0.120	0.967*	1.083*
	CF	29.743			-	1.263*	0.513*	0.283*	0.937*	1.393*	0.307*	0.190*
	OF	28.480				-	0.750*	0.980*	2.200*	0.130	0.957*	1.073*
	SCF1	29.230					-	0.230*	1.450*	0.880*	0.207*	0.323*
	SCF2	29.460						-	1.220*	1.110*	0.233	0.093*
	SCF3	30.680							-	2.330*	1.243*	1.127*
	SOF1	28.350								-	1.087*	1.203*
	SOF2	29.437									-	0.117*
	SOF3	29.553										-
Leave size	N	1.250	-	0.120	0.193	0.460*	0.290*	0.397*	0.390*	0.267*	0.267*	0.410*
	S	1.370		-	0.073	0.340*	0.170	0.277*	0.270*	0.137	0.147	0.290*
	CF	1.443			-	0.267*	0.097	0.203	0.197	0.063	0.073	0.217
	OF	1.710				-	0.170	0.063	0.070	0.203	0.193	0.050
	SCF1	1.540					-	0.107	0.100	0.033	0.023	0.120
	SCF2	1.647						-	0.007	0.140	0.130	0.013
	SCF3	1.640							-	0.1333	0.123	0.020
	SOF1	1.507								-	0.010	0.153
	SOF2	1.517									-	0.143
	SOF3	1.660										-

Table 4 Results of heavy metal accumulation in Chinese morning glory (Leaves)

Type	Heavy Metal (mg/kg)					
	As	Cd	Cr	Cu	Pb	Hg
Std. (mg/kg)	2	0.2	0.3	20	0.3	0.02
N	0.238	0.040	0.175	10.526	0.337	ND
S	0.278	0.040	0.118	6.614	0.229	ND
CF	0.059	0.050	0.104	7.019	0.248	ND
OF	0.079	0.030	0.117	3.845	0.119	ND
SCF1	0.120	0.040	0.169	6.749	0.190	ND
SCF2	0.117	0.040	0.138	5.957	0.220	ND
SCF3	0.159	0.050	0.177	5.635	0.219	ND
SOF1	0.219	0.030	0.199	4.380	0.239	ND
SOF2	0.119	0.039	0.192	4.997	0.125	ND
SOF3	0.030	0.040	0.129	6.241	0.290	ND

Remark: ND = Not detectable

Table 5 Results of heavy metal accumulation in Chinese morning glory (Trunk)

Type	Heavy Metal (mg/kg)					
	As	Cd	Cr	Cu	Pb	Hg
Std. (mg/kg)	2	0.2	0.3	20	0.3	0.02
N	ND	0.050	0.201	7.719	0.090	ND
S	ND	0.050	0.279	7.614	0.120	ND
CF	ND	0.060	0.259	6.520	0.130	ND
OF	ND	0.050	0.298	6.430	0.119	ND
SCF1	ND	0.030	0.295	3.998	0.069	ND
SCF2	ND	0.040	0.119	5.876	0.120	ND
SCF3	ND	0.049	0.127	4.810	0.079	ND
SOF1	ND	0.030	0.178	6.098	0.049	ND
SOF2	ND	0.040	0.138	4.384	0.119	ND
SOF3	ND	0.030	0.278	5.011	0.109	ND

Remark: ND = Not detectable

Table 6 Results of heavy metal accumulation in Chinese morning glory (Shoot)

Type	Heavy Metal (mg/kg)					
	As	Cd	Cr	Cu	Pb	Hg
Std. (mg/kg)	2	0.2	0.3	20	0.3	0.02
N	ND	0.030	0.117	7.894	0.129	ND
S	ND	0.030	0.107	8.705	0.129	ND
CF	ND	0.040	0.186	5.173	0.129	ND
OF	ND	0.030	0.130	5.523	0.140	ND
SCF1	ND	0.030	0.238	5.252	0.099	ND
SCF2	ND	0.030	0.137	5.680	0.139	ND
SCF3	ND	0.030	0.106	6.593	0.139	ND
SOF1	ND	0.030	0.149	7.360	0.130	ND
SOF2	ND	0.030	0.179	5.720	0.140	ND
SOF3	ND	0.030	0.149	6.899	0.179	ND

Remark: ND = Not detectable

Table 7 Results of heavy metal accumulation in Chinese morning glory (Root)

Type	Heavy Metal (mg/kg)					
	As	Cd	Cr	Cu	Pb	Hg
Std. (mg/kg)	2	0.2	0.3	20	0.3	0.02
N	0.294	0.049	0.112	6.721	0.167	ND
S	0.653	0.049	0.184	9.274	0.237	ND
CF	0.357	0.050	0.186	5.935	0.248	ND
OF	0.039	0.049	0.285	6.457	0.187	ND
SCF1	0.268	0.040	0.168	5.198	0.139	ND
SCF2	0.119	0.030	0.135	3.460	0.139	ND
SCF3	0.346	0.040	0.116	3.780	0.208	ND
SOF1	0.079	0.030	0.159	5.157	0.108	ND
SOF2	0.188	0.040	0.176	5.317	0.218	ND
SOF3	0.168	0.039	0.149	5.648	0.237	ND

Remark: ND = Not detectable

The food we consume daily may be contaminated by heavy metals. However, the risk of heavy metal exposure can be mitigated if manufacturers prioritize consumer safety, such as by reducing the use of chemical fertilizers and pesticides in agriculture. In addition, proper management of industrial waste, particularly adhesive sludge from wastewater treatment, is crucial in the production of fertilizer pellets. It is essential to determine whether the heavy metal content in adhesive sludge meets the required standards for use as fertilizer. Measurement protocols must be implemented to prevent any adverse effects on the environment, ensuring that the accumulation of heavy metals in plants complies with the standards set by the Ministry of Public Health.

The experiment demonstrated that the raw material, fertilizer pellets, and the heavy metal accumulation in Chinese morning glory plants all met the established standards. As a result, crops can be safely grown using this fertilizer without leaving harmful residues in the plants or the environment.

3.9.2 Formaldehyde

Formaldehyde levels were analyzed for Chinese morning glory plants grown with S and SCF3 fertilizers. The SCF3 (Sludge + Chemical fertilizer 5:5 by weight) treatment, which demonstrated the best growth performance, was selected for formaldehyde analysis. This treatment used a balanced ratio of sludge and chemical fertilizer, compared with S (Sludge) treatment, which exclusively used highest sludge content. The results, as summarized in Table 8.

According to the European Food Safety Authority (EFSA), exposure to formaldehyde from consuming 1 kg of food/day is estimated to not exceed 100 mg/kg [30]. The table 8 shown that the formaldehyde content in Chinese morning glory from this research (11.114 mg/kg for sludge and 10.701 mg/kg for sludge mixed with chemical fertilizer 5:5) falls within a moderate range compared to values reported in other studies, like the levels found in plums (11.2 mg/kg [31]) and green onions (13.3 mg/kg [32]) but significantly lower than in kohlrabi (31 mg/kg) and bananas (16.3 mg/kg). Conversely, it exceeds levels observed in spinach (3.3 mg/kg [32]), leafy vegetables (5 mg/kg [33]), and non-leafy vegetables (2.5 mg/kg [33]). When compared to rice (10.74 mg/kg [33]), the formaldehyde levels in Chinese morning glory are nearly equivalent, indicating that these values are typical and fall within the range observed in other consumable crops.

Table 8 Results of formaldehyde content

Commodity	Formaldehyde content (mg/kg)	References
Chinese morning glory S (Sludge)	11.114	
Chinese morning glory (Sludge + Chemical fertilizer 5:5)	10.701	
Vegetables	15.1	[12]
Banana	16.3	[36]
Beetroot	35	[31]
Plum	11.2	[31]
Kohlrabi	31	[31]
Potato	19.5	[31]
Pear	60	[37]
Green onion	13.3	[32]
Spinach	3.3	[32]

Shiitake mushroom	100-320	[38]
Rice	10.74	[33]
Leafy vegetables	5	[33]
Non-leafy vegetables	2.5	[33]
Fruits	3.08	[33]

In the production of melamine-coated paper, formaldehyde is present in resin form, which can naturally decompose and cannot be absorbed by plants in this state. However, when adhesive sludge waste from the factory is used to produce fertilizer for plant cultivation, it is essential to verify whether the plants accumulate formaldehyde at levels higher than normal.

Despite the nature presence of formaldehyde in food, there exists a scarcity of research regarding the mechanisms of its formation and concentration levels. Moreover, the addition of formaldehyde to food products has been shown to be detrimental to consumer health [34]. Consequently, organizations such as the US Environmental Protection Agency and the World Health Organization have stipulated tolerable daily intake levels for formaldehyde at 0.2 mg/kg/day [31] and 0.15 mg/kg/day for humans, respectively [35]. Nevertheless, as a precautionary measure to mitigate potential health risks, it is advisable to prioritize their application in the cultivation of ornamental plants and flowers, rather than edible crops.

4.0 CONCLUSION

This study investigates the potential of recycling adhesive sludge from the wastewater treatment system of a melamine-coated paper factory for agricultural use. The findings are as follows:

- The dried adhesive sludge, with a moisture content of 15-20%, was found to be suitable for pelletizing without the need for additional binders.

- The resulting pellets conformed to the Department of Agriculture's standards for all criteria. Adhesive sludge has the ability to be a fertilizer, it can be used to replace chemical and organic fertilizer and mixing adhesive sludge with chemical and organic fertilizer has been shown to raise the value of organic matter and N P K.

- Chinese morning glory plants fertilized with SCF3 (Sludge + Chemical fertilizer 5:5 by weight) exhibited the most favorable growth, both in terms of height and fresh weight. Other formulations, such as OF (Organic Fertilizer) and SCF2 (Sludge + Chemical fertilizer 6:4 by weight), also demonstrated satisfactory performance.

- The levels of heavy metals in the Chinese morning glory were within the safety limits established by the Ministry of Public Health. Copper was the most accumulated metal, particularly in the leaves and roots. Notably, the use of sludge mixed with fertilizers generally resulted in lower heavy metal accumulation compared to the use of sludge alone.

- The formaldehyde content in plants grown with SCF3 (10.701 mg/kg) was lower than in those grown with sludge alone (11.114 mg/kg), and both values were below those typically found in other commonly consumed vegetables and mushroom.

In conclusion, this study demonstrates that adhesive sludge can effectively reduce the reliance on additional fertilizers and can be safely utilized in agriculture. The sludge not only supports plant growth but also meets safety standards for heavy metal and formaldehyde content, making it a suitable option for sustainable agricultural practices. Incorporating sludge into

fertilizer production can provide economic benefits by lowering fertilizer costs and addressing waste disposal challenges. This study indicates that sludge, even when mixed with chemical fertilizers, can be used to cultivate Chinese morning glory without raising significant safety concerns regarding formaldehyde content. However, it is recommended that the adhesive sludge be used for cultivating ornamental and decorative plants, where consumption is not a concern, to mitigate potential risks of formaldehyde and heavy metal accumulation.

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Conflicts of Interest

The author(s) declare(s) that there is no conflict of interest regarding the publication of this paper

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