

# Synthesis and Characterization of a Mixed Nanofertilizer Influencing the Nutrient Use Efficiency, Productivity, and Nutritive Value of Tomato Fruits

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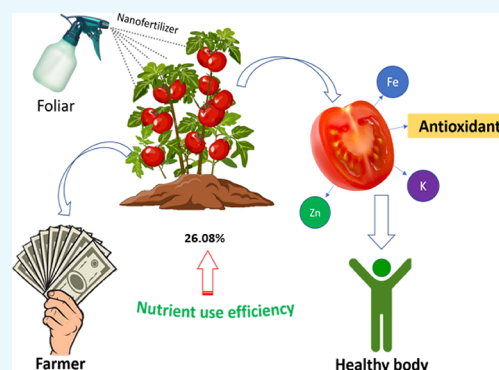


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**ABSTRACT:** Due to the higher potential for enhancing nutrient use efficiency, nanofertilizer (NF) is crucial in sustainable crop production. Thus, foliar-applied mixed nanofertilizer (MNF<sub>f</sub>) and commercial fertilizer (CF) into the soil (CFs) were claimed together ([MNF<sub>f</sub> + CFs]) and comparative nutrient use efficiency (NUE), productivity, and nutritional properties of tomato fruits were investigated. The mixed nanofertilizer (MNF) was prepared in our laboratory and characterized using scanning electron microscopy, X-ray diffraction, and Fourier transform infrared. To avoid the interference of other factors, all the treatments were divided into three groups: (i) blank treatment (no fertilizer), (ii) CF treatment, and (iii) combined [MNF<sub>f</sub> + CFs] treatment. The vegetative growth and qualitative and quantitative attributes of tomatoes were recorded, and the NUE, total production, and benefit–cost ratio (BCR) were also calculated. In addition, comparative nutritional properties for all treatments were analyzed. The plant's height, stem diameter, root length, photosynthetic pigments, leaf minerals, and qualitative traits of tomato fruits were significantly ( $p < 0.05$ ) increased by [MNF<sub>f</sub> + CFs] treatment compared to CFs. The protein, fiber, Fe, Zn, and K contents were significantly ( $p < 0.05$ ) increased by 23.80, 38.10, 44.23, 60.01, and 2.39%, respectively, with the [MNF<sub>f</sub> + CFs] treatment as compared to CFs, while the ash and protein contents were both lower than the untreated tomato. Moreover, [MNF<sub>f</sub> + CFs] treatment has significantly ( $p < 0.05$ ) increased the antioxidant properties. The NUE, total production, and BCR were also increased by 26.08, 26.04, and 25.38%, respectively, with the same treatment. Thus, [MNF<sub>f</sub> + CFs] treatment could be a potential alternative for reducing the excess use of CF.



## 1. INTRODUCTION

Tomato fruit (*Solanum Lycopersicum* L.) is considered as one of the most effective and nutritious foods in the human diet. This fruit is rich in bioactive compounds like vitamins, carotenoids, and phenolic compounds. These compounds have high antioxidant activity and are then beneficial to human health.<sup>1</sup> Aside from its nutritional value, it is the second most commercially consumed vegetable after potatoes.<sup>2</sup> The overall recommended fertilizer for crop production was 184 metric tons in 2015; however, it has been expected to exceed 200 MT by 2020.<sup>3</sup> This was to increase the global agricultural crop production by 70% for supporting the rapid growth of the world population by 2050.<sup>4</sup> Interestingly, the increases in crop yields are not linearly correlated with the increase in nitrogen (N) application rates, which inevitably leads to decrease nutrient use efficiency (NUE) and increase N losses.<sup>5</sup> However, such indiscriminate use of CF frequently fails to hit the target sites of the crop plants. Thus, it reduces NUE resulting from the disproportionate dissolution rate, removal or leaching, and run-off caused by wind, rain, and sunlight.<sup>6</sup> We reported that the problem of run-off and 40–70% of leaching

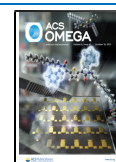
lead to lower NUE that forces farmers for heavy periodic fertilization to maintain high agricultural productivity, causing a huge economic loss.<sup>7</sup> Also, the production process of these CFs involves huge environmental costs in terms of energy (ammonia) and renewable energy (phosphorous and potassium).<sup>3</sup> Therefore, an alternative fertilizer management practice is extremely required for the current agricultural management system that will increase the production as well as nutritional properties.

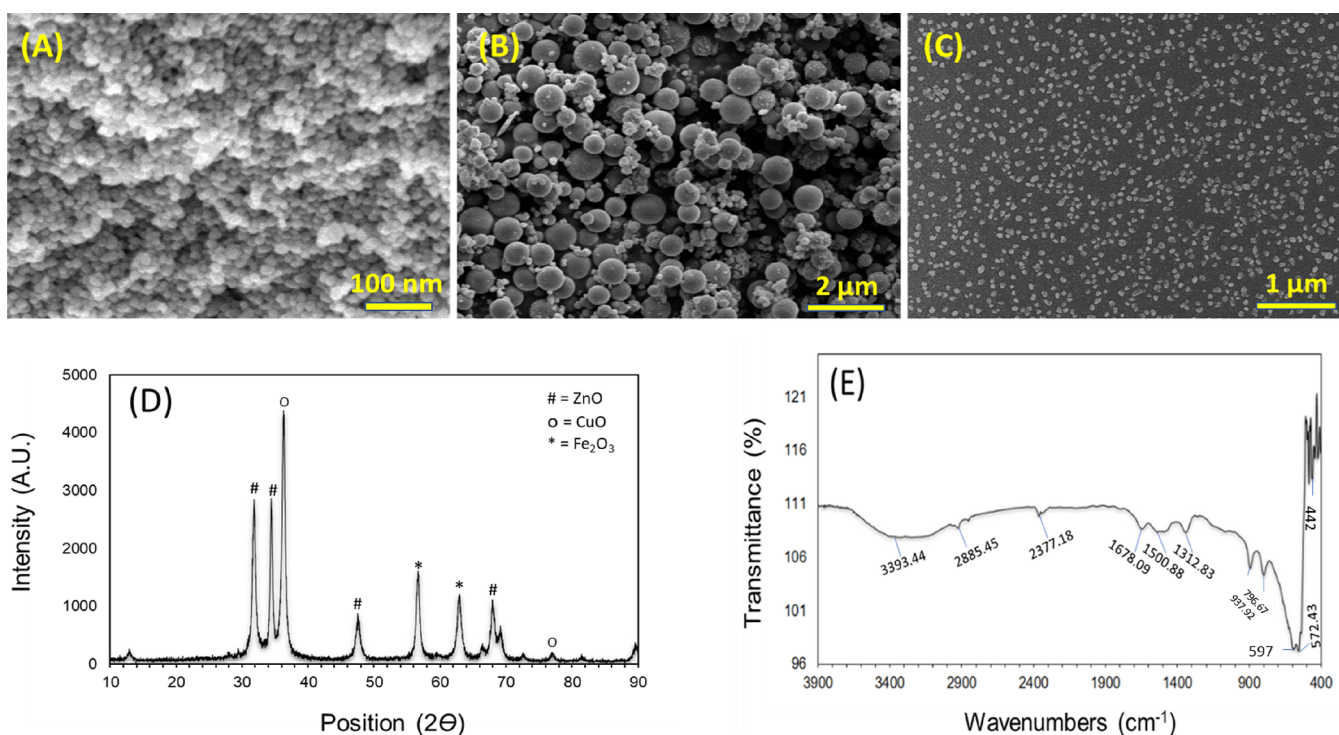
Nanotechnology in modern agriculture is such a promising technology with tremendous potential to resolve nutrient shortages and leaching losses. The literature revealed that the use of NF as the macro and/or micronutrients with different

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**Figure 1.** Structural characteristics of mixed nanofertilizer (MNF). SEM images of as-synthesized (A) FeNPs, (B) ZnNPs, and (C) CuNPs. Intensity peaks of XRD analysis (D) and attenuated total reflectance (ATR)-FTIR spectra (E) of prepared MNF.

application methods demonstrated slow and sustainable nutrient release boosting maximum nutrient utilization with comparatively higher production<sup>7–9</sup> because CF includes only the basic nutrients like N, P (phosphorous), and K (potassium); however, it does not include the other macro- and/or micronutrients. Therefore, the synthesis of NF with different macro/micronutrients based on nanotechnology emphasizes the controlled release and sustainable delivery systems to the plants. For example, eco-compatible cassava starch films as a controlled-release nutrient,<sup>10</sup> chitosan nanoparticle as the sustainable delivery system,<sup>11</sup> and controlled released phosphorous fertilizer based on biological macromolecules<sup>12</sup> were found to be applied as part of nanotechnology in the sustainable agricultural production. As a part of nanotechnology, foliar application of NF as macronutrients, also known as foliar feeding, has gained considerable attention to sustainable agricultural crop production. In this method, the nutrients are absorbed through the leaves of the plant. For example, foliar applications of selenium (Se), silicon (Si), and copper (Cu) nanoparticles (NPs) in bell pepper fruits,<sup>13</sup> nano zinc (nZn) and nano iron (nFe) in *Rosmarinus officinalis* plants,<sup>14</sup> calcium (Ca) fertilizer in a pomegranate tree,<sup>15</sup> Cu NPs (nanoparticles) in a tomato fruit,<sup>16</sup> nCu, nZn, and nMn (nano manganese) in a wheat plant,<sup>17</sup> nZn and nB (nano boron) in a pomegranate fruits,<sup>15</sup> nCeO<sub>2</sub> (nano ceric oxide) and CuO (copper oxide) in a cucumber fruit,<sup>18</sup> and nNPK (nano NPK) in a potato tuber<sup>19</sup> were investigated and positive changes in terms of the respective plant's growth, development, and their bioactive compounds were reported. However, to the best of our knowledge, no study was reported on the combined application of MNF<sub>f</sub> and CFs ([MNF<sub>f</sub> + CFs]) for tomato cultivation.

Therefore, the objective of this current study was to investigate the synergistic effect of [MNF<sub>f</sub> + CFs] on the growth and development, vitamin and mineral contents, and antioxidant properties of tomatoes. Moreover, the effect on NUE, total production, and BCR was also studied. Thus, the current study could offer a potential alternative fertilizer management system that would be economically feasible promoting sustainable agricultural crop production with a higher nutritive value.

## 2. RESULTS AND DISCUSSION

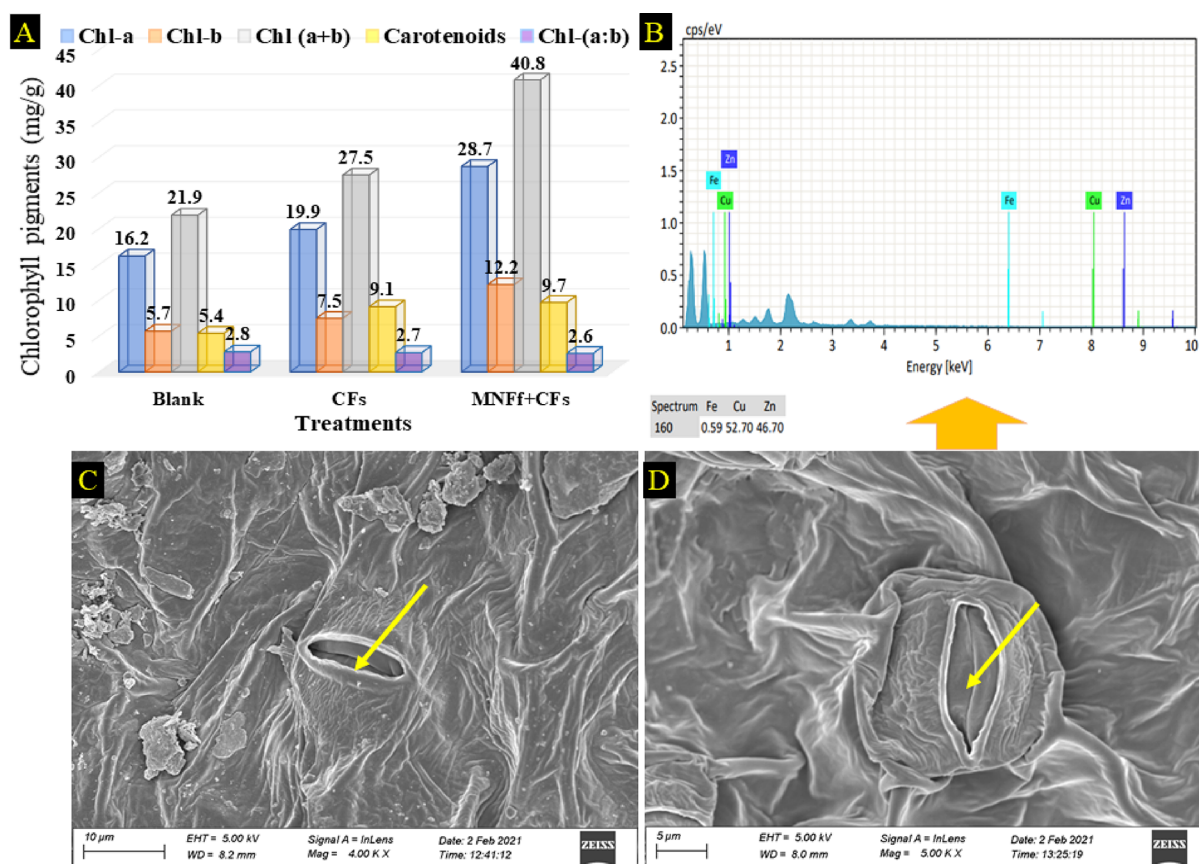
**2.1. Characterization of MNF.** Scanning electron microscopy (SEM) measurement was done to study the size and morphology of synthesized nanoparticles. Figure 1A confirms the shape and size of FeNPs (size 8–18 nm). Meanwhile, nanoball-like ZnNPs (100–300 nm) were observed, as shown in Figure 1B. Furthermore, cubic-structured CuNPs were formed with an average size of 30–60 nm, as can be seen in Figure 1C. Figure 1D shows the X-ray diffraction (XRD) of MNF at which some peaks displayed at 32, 33.6, 36.20, 47.81, 56.90, 63, 68.53, 77, and 81.7°. Thus, the observed pattern from Figure 1D reveals the face-centered cubic structure of the synthesized nanoparticles.<sup>20–23</sup> Figure 1E shows the transmittance of the Fourier transform infrared (FTIR) spectrum of MNF, at which the prominent bands occur at 3393.44 (O–H), 2885.45 (C–H), 2377.18 (C–O), 1678.09 (C=C), 1500.88 (C=O), 1312.83 (C–O), 937.92 (C–N), 796.67 (C–H), 597 (Fe–O), 572.43 (Zn–O), and below 442 (Cu–O) cm<sup>-1</sup>.<sup>24,25</sup> The spinel cubic structure was confirmed from the absorption bands that occur in the range of 400–500 cm<sup>-1</sup>.<sup>26–28</sup>

**2.2. Effects of [MNF<sub>f</sub> + CFs] on Growth, Development, and Yield.** **2.2.1. Effects on Vegetative Growth.** When [MNF<sub>f</sub> + CFs] is applied, the plant height, stem diameter, root

Table 1. Effects of [MNF<sub>f</sub> + CFs] on the Growth and Development of Tomato Plants<sup>a</sup>

treatments	growth measurements (cm)			mass of dried biomass (g/100 g)		
	plant height	stem diameter	root length	leaf	stem	root
blank	90 ± 3.81b	2.9 ± 0.12b	19.2 ± 1.78c	12.7 ± 0.18a	11.12 ± 0.19a	15.45 ± 0.12b
CFs	104 ± 3.63b	3.0 ± 0.16b	23.2 ± 2.21b	11.7 ± 0.13b	10.96 ± 0.28a	16.95 ± 0.13b
[MNF <sub>f</sub> + CFs]	131 ± 5.41a	3.6 ± 0.13a	29.8 ± 2.57a	11.9 ± 0.18b	10.71 ± 0.17a	18.51 ± 0.21a

<sup>a</sup>Values were expressed as means of triplicate samples ± standard deviation ( $n = 3$ ); different letters in the same column are significantly ( $p < 0.05$ ) different. Blank: untreated tomato; CFs: CF applied into soil; [MNF<sub>f</sub> + CFs]: combined application of mixed nanofertilizer (MNF) and CF fertilizer on the leaf and soil, respectively.



**Figure 2.** Chlorophyll pigment contents (A), microscopic view of the stomatal opening on the tomato leaf surface for mixed nanofertilizer uptake at 4000 $\times$  (C) and 5000 $\times$  (D) for magnification with a 10 and 5  $\mu$ m distance, respectively, and apparent concentrations of Fe, Cu, and Zn in the tomato leaf treated with [MNF<sub>f</sub> + CFs], as determined by SEM–EDX (B).

length, and dried root biomass were significantly ( $p < 0.05$ ) increased by 25.9, 20.0, 28.5, and 9.2%, respectively, compared to CFs alone (Table 1 and Figure S1). This indicates that the addition of foliar application of MNF with CFs significantly enhanced the growth and development of tomato plants. This might be due to the foliar application of MNF that properly distributed its Fe, Zn, and Cu on tomato leaves (Figure 2B). A similar agreement was found in the growth and development of hydroponic tomato plants,<sup>29</sup> wheat plants,<sup>30</sup> and pot marigold (*Calendula officinalis*) plants<sup>31</sup> with the foliar application of macronutrients, micronutrients, and nickel (Ni), respectively. Similarly, the chlorophyll pigments in tomato leaves were significantly ( $p < 0.05$ ) increased by the treatment of [MNF<sub>f</sub> + CFs] compared to untreated and CFs. It also could be attributed to the fact that the [MNF<sub>f</sub> + CFs] led to enhance the photosynthetic pigments in tomato leaves over the growing period. Similar results were observed in grapefruit plants

treated with a mixture of Cu and chelated Fe<sup>32</sup> and maize plants treated with foliar application of ZnO NPs.<sup>33</sup>

### 2.2.2. Effects on the Leaf Structure and Mineral Contents.

The surface morphology was studied to investigate the stomatal openings on tomato leaves as well as the Fe, Cu, and Zn distribution with SEM–EDX (ZEISS Gemini 500) and is shown in Figure 2C,D. Results show that the larger stomatal openings are present in the tomato leaves treated with [MNF<sub>f</sub> + CFs], suggesting the easy penetration of MNF (smaller than 100 nm) into the tomato leaves, enhancing the growth and development of tomato plants (Table 1). Moreover, SEM–EDX was performed to confirm the contents of minerals in the tomato leaf. However, the Fe content was extremely lower in the leaf than Cu and Zn (Figure 2B).

**2.2.3. Effects on Tomato Fruits.** The average number of tomatoes per plant, average weight of a single tomato, average size of a fruit, and the average total yield per plant were significantly ( $p < 0.05$ ) increased by 12.1, 10.79, 10.0, and



**Table 2. Effects of [MNF<sub>f</sub> + CFs] on Tomato Fruit Development<sup>a</sup>**

treatments	quantitative attributes			qualitative attributes			
	number of fruits/plants	avg weight (g)/fruit	yield (kg)	fruit shape	size of fruit (cm)	size of seed number	dry weight (%)
blank	11c	97.5 ± 3.3c	5.36 ± 0.05c	oblate	17 ± 0.41c	small	5.7 ± 0.16a
CFs	19b	136.2 ± 4.2b	12.93 ± 0.07b	ellipsoid	20 ± 0.82b	higher	4.0 ± 0.17b
[MNF <sub>f</sub> + CFs]	21.6a	150.9 ± 5.4a	17.05 ± 0.13a	ellipsoid	22 ± 0.24a	intermediate	5.8 ± 0.12a

<sup>a</sup>Values were expressed as means of triplicate samples ± standard deviation ( $n = 3$ ); different letters in the same column are significantly ( $p < 0.05$ ) different. Blank: untreated tomato; CFs: CF applied into the soil; [MNF<sub>f</sub> + CFs]: combined application of mixed nanofertilizer (MNF) and CF fertilizer on the leaf and soil, respectively.

**Table 3. Effects of [MNF<sub>f</sub> + CFs] on Proximate Composition of Tomato Fruits<sup>a</sup>**

treatments	proximate composition of tomato fruits (%)						
	moisture	ash	protein	fiber	fat	carbohydrate	energy(kcal/100 g)
blank	95.2 ± 1.74a	0.43 ± 0.08a	0.95 ± 0.09a	0.66 ± 0.12b	0.23 ± 0.00c	3.31 ± 0.71c	20.2 ± 1.62c
CFs	92.8 ± 1.23bc	0.29 ± 0.02bc	0.63 ± 0.05c	0.63 ± 0.01c	0.69 ± 0.05a	5.56 ± 0.44a	32.9 ± 1.49a
[MNF <sub>f</sub> + CFs]	93.7 ± 1.25b	0.31 ± 0.02b	0.78 ± 0.09b	0.87 ± 0.10a	0.26 ± 0.01b	4.90 ± 0.45ab	26.9 ± 1.28b

<sup>a</sup>Values were expressed as means of triplicate samples ± standard deviation ( $n = 3$ ); different letters in the same column are significantly ( $p < 0.05$ ) different. Blank: untreated tomato; CFs: CF applied into the soil; [MNF<sub>f</sub> + CFs]: combined application of mixed nanofertilizer (MNF) and CF fertilizer on the leaf and soil, respectively.

**Table 4. Effects of [MNF<sub>f</sub> + CFs] on Vitamin and Minerals of Tomato Fruits<sup>a</sup>**

treatments	vitamin A (mg/100 g)	minerals					pH
		Na (mg/100 g)	K (mg/100 g)	Fe (mg/100 g)	Cu (mg/100 g)	Zn (mg/100 g)	
blank	40.5 ± 0.37a	74.2 ± 0.21a	341.3 ± 0.81c	2.3 ± 0.10b	0.018 ± 0.01b	0.071 ± 0.01a	4.9 ± 0.07a
CFs	40.5 ± 0.39a	69.6 ± 0.43c	418.4 ± 0.71a	2.1 ± 0.01b	0.022 ± 0.02a	0.450 ± 0.03b	4.9 ± 0.03a
[MNF <sub>f</sub> + CFs]	40.61 ± 0.31a	71.5 ± 0.43b	428.2 ± 0.97b	3.3 ± 0.25a	0.015 ± 0.01c	0.720 ± 0.01a	4.7 ± 0.02a

<sup>a</sup>Values were expressed as means of triplicate samples ± standard deviation ( $n = 3$ ); different letters in the same column are significantly ( $p < 0.05$ ) different. Blank: untreated tomato; CFs: CF applied into the soil; [MNF<sub>f</sub> + CFs]: combined application of mixed nanofertilizer (MNF) and CF fertilizer on the leaf and soil, respectively.

31.8%, respectively, with [MNF<sub>f</sub> + CFs] treatment over the CFs (Table 2 and Figure S1). This indicates that the foliar application of a small amount of MNF with CFs had a tremendous effect on the production of tomatoes without significantly affecting the input cost (Table 6). It might be due to the balanced and controlled distribution of MNF and CFs leading to higher growth and development of plants (Table 1 and Figure S1), indicating the maximum size of tomato fruits as well as the yield (Table 2 and Figure S1). Moreover, a higher seed number was observed in the tomato fruit treated with CFs, indicating the less tomato flesh or consumable part and that could reduce the quality as well as the market value of tomato fruits. Thus, the application of [MNF<sub>f</sub> + CFs] could be one of the most promising approaches for agricultural practices. It was reported that the numbers of pods and eggplants were significantly ( $p < 0.05$ ) increased when the bean and eggplants were treated with foliar application of ceria (Ce) NPs as well as ZnSO<sub>4</sub>.<sup>34,35</sup>

**2.3. Effects of [MNF<sub>f</sub> + CFs] on Nutritional Properties of Tomato Fruits.** **2.3.1. Effects on Proximate Composition.** The protein and fiber contents were significantly ( $p < 0.05$ ) increased by 23.80 and 38.10%, respectively, with [MNF<sub>f</sub> + CFs] treatment compared to CFs. However, the fat, carbohydrate, and energy were decreased by 62.3, 11.8, and 18.2%, respectively, with the same treatment (Table 3). The moisture, ash, protein, and fiber contents were decreased by 2.5, 32.5, 33.7, and 4.5%, respectively, when the CFs were applied. However, the carbohydrate and energy contents were increased by 67.9 and 62.8%, respectively. This variation might be due to the foliar application of MNF with CFs that could

have enhanced the contents of the proximal constituents. Most interestingly, the fat content of the tomato fruit treated with [MNF<sub>f</sub> + CFs] was significantly decreased by 62.31% compared to the tomato fruit treated with CFs. These proximal differences might be attributed to the differences in plant biosynthesis as well as fruit constituents that might have been affected by the foliar application of MNF. It was reported that, under different treatments of NF, similar ranges of moisture, ash, protein, fiber,<sup>36</sup> fat,<sup>37</sup> carbohydrate, and energy<sup>38</sup> contents were observed in ripe tomato fruits.

**2.3.2. Effects on Vitamins and Mineral Composition.** No treatment had a significant ( $p < 0.05$ ) effect on vitamin A compared with untreated tomato fruits, as shown in Table 4. However, the mineral contents such as Zn, K, and Fe were significantly affected by the foliar application of MNF with CFs, as can be seen in Table 4. The analyzed data showed that the contents of Zn and Fe were increased by 60.01 and 44.23%, respectively, in tomatoes treated with [MNF<sub>f</sub> + CFs] as compared to CFs. However, the contents of Na in tomato fruits treated with CFs and [MNF<sub>f</sub> + CFs] were decreased by 7.71 and 4.05%, respectively, compared to the untreated tomatoes. Moreover, the contents of Cu and Zn were at an acceptable level for human consumption. The overall mineral distribution in tomato fruits indicates the balanced uptake and translocation of MNF by tomato leaves, suggesting higher plant growth and development (Table 1).

**2.3.3. Effects on Antioxidant Composition.** The application of [MNF<sub>f</sub> + CFs] had significant ( $p < 0.05$ ) effects on the antioxidant composition of ripe tomatoes in terms of ascorbic acid (AsA), the total phenolic content (TPC), flavonoid

**Table 5. Effects of [MNF<sub>f</sub> + CFs] on Antioxidant Properties in Tomato Fruits<sup>a</sup>**

treatments	antioxidant compositions				antioxidant activities		
	vitamin C (mg/100 g)	total phenolic contents (mg GAE/100 g)	total flavonoids (mg/100 g)	tannins (mg/100 g)	DPPH (%)	ABTS <sup>++</sup>	IC <sub>50</sub> (% inhibition)
blank	18.91 ± 0.31c	7.40 ± 0.33b	46.21 ± 0.16a	12.23 ± 0.16a	90.82 ± 0.66a	28.11 ± 0.10b	14.14 ± 0.13a
CFs	22.02 ± 0.36a	5.51 ± 0.18c	37.22 ± 0.19c	11.12 ± 0.57b	90.12 ± 0.73a	25.32 ± 0.20c	11.42 ± 0.11b
[MNF <sub>f</sub> + CFs]	19.51 ± 0.42b	9.30 ± 0.20a	43.22 ± 0.17b	12.92 ± 0.74a	86.83 ± 0.6b	35.31 ± 0.23a	10.23 ± 0.14c

<sup>a</sup>Values were expressed as means of triplicate samples ± standard deviation ( $n = 3$ ); different subscript letters in the same column are significantly ( $p < 0.05$ ) different. Blank: untreated tomato; CFs: CF applied into the soil; [MNF<sub>f</sub> + CFs]: combined application of mixed nanofertilizer (MNF) and CF fertilizer on the leaf and soil, respectively.

**Table 6. Effects of [MNF<sub>f</sub> + CFs] on NUE, Total Production, Total Input Cost, Net Income, and BCR<sup>a</sup>**

treatment	total fertilizer (kg/ha)	total production (kg/ha)	total cost (BDT/ha)	net income (BDT/ha)	NUE (kg/ha)	BCR
blank	0	6512.76	455650.06	260510.33	0	0.57
CFs	364.52	15710.81	479951.38	628432.56	43.11	1.30
[MNF <sub>f</sub> + CFs]	364.61	19802.19	486938.03	791737.55	54.31	1.63

<sup>a</sup>Values were expressed as means of triplicate samples. Blank: untreated tomato; CFs: CF applied into the soil; [MNF<sub>f</sub> + CFs]; combined application of mixed nanofertilizer (MNF) and CF fertilizer on the leaf and soil, respectively.

content (FC), and tannin content (TC) over CFs, as shown in Table 5. Results showed that the application of CFs has greatly increased the content of AsA by 16.4% as compared to untreated tomato; however, it was again decreased by 3.24% with the application of [MNF<sub>f</sub> + CFs], although the content of AsA was insignificantly increased by 3.17% in [MNF<sub>f</sub> + CFs]-treated tomato compared to the untreated tomato fruit, as presented in Table 5. This increment of AsA can be attributed to the induction of antioxidant compounds with the foliar application of MNF. Moreover, the contents of TPC, FC, and TC were significantly ( $p < 0.05$ ) increased by 69.11, 16.10, and 16.23%, respectively, in the tomato fruits treated with [MNF<sub>f</sub> + CFs] as compared to CFs. This increase in TPC, FC, and TC might be due to the complementary effect of MNF on the plant metabolism, thus increasing the antioxidant composition as bioactive compounds.<sup>16</sup> Similar findings were reported in a previous study conducted on the foliar application of Cu NPs in the tomato fruit,<sup>39</sup> and the foliar application of Zn and B in pomegranate showed minor changes in total phenols.<sup>15</sup>

**2.3.4. Effects on Antioxidant Activities.** The antioxidant activities of ripe tomato fruits treated with CFs and [MNF<sub>f</sub> + CFs] were investigated and are presented in Table 5. The determination of DPPH scavenging activities (%) showed that the oxidative reaction caused by free radicals was reduced by 4.5%, while the [MNF<sub>f</sub> + CFs] was applied as compared to CFs. This indicates that the antioxidant compounds found in tomatoes treated with [MNF<sub>f</sub> + CFs] possess a strong potential to scavenge the free radicals. Similarly, the ABTS<sup>++</sup> discoloration assay was performed for the determination of antioxidant activities, and the results showed that the tomato fruits treated with [MNF<sub>f</sub> + CFs] were significantly ( $p < 0.05$ ) increased by 25.6 and 39.52% compared with untreated and CF treatment, respectively. IC<sub>50</sub> measures the concentration for 50% antioxidant activity, and the lower IC<sub>50</sub> means the higher antioxidant activities. Consequently, the tomato fruits treated with [MNF<sub>f</sub> + CFs] showed the lowest IC<sub>50</sub> indicating the higher antioxidant activities by 27.70%, while the CF is 19.10% as compared to untreated tomatoes. It suggests that the application of [MNF<sub>f</sub> + CFs] added 8.6% antioxidant activities. This all might be attributed to the addition of foliar application of MNF with CFs that induced more bioactive compounds, causing higher antioxidant activities.<sup>16</sup> The more closely

related findings were observed in mung bean,<sup>40</sup> potato,<sup>41</sup> tomato,<sup>39</sup> and Jalapeno pepper<sup>42</sup> plants treated with the application of different NPs as NF.

**2.4. Effects of [MNF<sub>f</sub> + CFs] on NUE and BCR.** The NUE of tomato plants treated with [MNF<sub>f</sub> + CFs] was significantly ( $p < 0.05$ ) increased by 26.08% as compared to CF treatment (Table 6). This higher NUE might be attributed to the higher uptake of MNF by tomato leaves due to having a larger stomatal opening (Figure 2C,D) influencing the enhancement of plant growth as well as total production. A similar agreement was found by other researchers,<sup>19</sup> and they reported that the foliar application of nanofertilizer in potato leaves has greatly enhanced the NUE. This also indicates that the use of MNF could reduce the excessive use of CF. The economic sustainability and the effectiveness of this [MNF<sub>f</sub> + CFs] treatment for crop production could also be justified in terms of the B:C ratio and net income (Table 6). The tomato plants treated with [MNF<sub>f</sub> + CFs] increased the net income by 25.98%, and the B:C ratio was increased from 1.30 to 1.63 followed by CF-treated tomato plants, suggesting the increased net profit as 2-fold compared to that obtained by CFs (Table 6).

### 3. CONCLUSIONS

The application of [MNF<sub>f</sub> + CFs] had shown an excellent beneficial effect on the growth, development, and nutritional properties of the tomato fruits, indicating the reduction of excessive use of CF that pollutes the environment as well as human health. In general, the growth parameters like plant height, stem diameter, and root length of tomato plants treated with [MNF<sub>f</sub> + CFs] were considerably increased. Similarly, the number of fruits per plant, average individual weight of the fruit, size of the fruit, and the average yield per plant were also significantly increased by 13.7, 10.79, 10.0, and 31.8%, respectively, with [MNF<sub>f</sub> + CFs] treatment. Moreover, the antioxidant activities were significantly augmented by [MNF<sub>f</sub> + CFs] treatment as compared to CFs. In addition, the NUE and BCR also showed a substantial intensification with the inclusion of foliar application of MNF with CFs, suggesting potential fertilizer management practices for tomato production indicating maximum profit without affecting the input

cost. Thus, the application of  $[MNF_f + CF_s]$  could be one of the most potential substitutions of CF in future agriculture enhancing the nutrient use efficiency, total production, and a sustainable environment.

## 4. MATERIALS AND METHODS

**4.1. Experimental Site, Chemicals, and Tomato Variety.** The experiment was conducted from September 2020 to December 2020 in the field just beside the north side of Jashore University of Science and Technology, Bangladesh. The field was at  $23^\circ 10'$  North latitude and  $89^\circ 13'$  East longitude at a height of 9 m above sea level, according to the global positioning system. The chemical analysis of the soil conditions of the cultivated land is reported in Table 7. To

**Table 7. Soil Chemical Properties of the Selected Land for Tomato Cultivation**

soil properties	amounts
electrical conductivity (dS/m)	7.55
pH	0.78
total nitrogen (%)	0.12
exchangeable phosphorous ( $\mu\text{g/g}$ )	155.5
exchangeable potassium (Cmol <sub>k</sub> /g)	0.35
sulfur ( $\mu\text{g/g}$ )	14.75
sodium (Cmol <sub>k</sub> /g)	0.115
organic matter (%)	1.65

prepare the MNF, sodium hydroxide (NaOH), zinc acetate, copper chloride ( $\text{CuCl}_2$ ), and ferrous chloride ( $\text{FeCl}_2$ ) were used. However, potassium sulfate ( $\text{K}_2\text{SO}_4$ ), 98% sulfuric acid ( $\text{H}_2\text{SO}_4$ ), boric acid ( $\text{H}_3\text{BO}_3$ ), petroleum ether, ethyl alcohol ( $\text{C}_2\text{H}_5\text{OH}$ ), and chlorhydric acid solution were used for proximate analysis. Potassium oxide (KOH), xylene ( $\text{C}_8\text{H}_{10}$ ), and nitric acid were used for vitamin and mineral determination. Moreover, absolute methanol (99.9%), 10% sodium carbonate ( $\text{Na}_2\text{CO}_3$ ), Folin–Ciocalteu reagent, sodium nitrate ( $\text{NaNO}_3$ ), aluminum chloride ( $\text{AlCl}_3$ ), gallic acid, tannic acid, DPPH (2,2-diphenyl-1-picrylhydrazyl), potassium sulfate buffer, and ABTS (2,2-azinobis-(3-ethylbenzothiazoline-6-sulfonate) were used for the determination of antioxidant properties. All the chemicals and reagents were purchased of analytical grade from Sigma-Aldrich, China. The CF was purchased from a local market, Jashore, Bangladesh. BARI tomato-5 cultivar was used in this experiment.

**4.2. Synthesis and Characterization of MNF.** To prepare Zn NPs, 10 g of NaOH (1.0 M) was dissolved in 250 mL of ultrapure water and stirred at  $90^\circ\text{C}$ . Then, 17.0358 g of  $\text{ZnCl}_2$  (0.5 M) solution was prepared and kept in a burette. The solution was then dropwise added into the NaOH solution for 26 min and was continuously stirred for 2 h at  $90^\circ\text{C}$ . The obtained solution was kept overnight to be settled down from the precipitate. The collected suspension was washed with absolute ethanol and ultrapure water several times to remove unreacted molecules. Similarly, Fe and Cu NPs were prepared by the chemical reduction method using  $\text{FeCl}_2$  and  $\text{CuCl}_2$ , respectively.<sup>7</sup> However, to confirm the formation of the spinel structure of the as-synthesized nanoparticles, ATR-FTIR analysis was performed in the frequency range of  $400\text{--}4000\text{ cm}^{-1}$ . Moreover, a BRUKER X-ray diffractometer with  $\text{CuK}\alpha$  radiation of a wavelength of  $1.5406\text{ \AA}$  was used to study the crystallinity and phase formation of the MNF sample over the angular range of  $10\text{--}90^\circ$ .

**4.3. Agronomic Practices and Management.** The cultivated land was plowed well to get fine tilth. Then, the 30 day old, healthy, and vigorous tomato seedlings were collected and transplanted into a separated bed with an equal area of  $8.23 \times 10^{-4}$  ha. The plant-to-plant distances within the bed as well as between the beds were 55 and 90 cm, respectively. The bed-to-bed distance was 60 cm. Standard management practices such as intercultural operations like weeding, watering, and insecticide application were done as per traditional methods. The land was divided into three beds: (i) untreated bed (no fertilizer used), (ii) CF-treated bed (CF is applied to soil), and (iii) combined  $[MNF_f + CF_s]$ -treated bed (foliar application of MNF and soil application of CF simultaneously).

**4.3.1. Application of MNF and CFs.** No fertilizer was used in the untreated bed. Only, 20 g of CF (5 g of potash, 5 g of urea, and 10 g of triple superphosphate) was applied into the CF-treated bed. However, prepared 5 mg of MNF was diluted with 50 mL of water in a foliar fertilizer sprayer pot. Then, the solution was properly mixed and sprayed on the tomato leaf surface in the combined  $[MNF_f + CF_s]$ -treated bed. At the same time, 10 g of CF (2.5 g of potash, 2.5 g of urea, and 5 g of triple superphosphate) was also applied to this combined  $[MNF_f + CF_s]$ -treated bed at a 4 in. distance from the plant root by digging the soil 1 in. depth as fertilizer placement methods. However, all fertilizers for all treatments were applied four times of interval in the whole growing period. Initially, after 15 days of transplanting for the first time as well as after the next 15 days for the second time, the third time application was done just before the flowering of tomato plants and the fourth and final application was performed when the tomato fruits were the age of 10 days.

**4.3.2. Vegetative Growth and Biomass Measurements.** Plant growth indicators such as plant height (cm), stem diameter (cm), and root length were recorded. The average number of fruits/plants, average weight/fruit per bed, total yield/plants per bed, and qualitative attributes like fruit shape, size of the fruit, and dry mass of the tomato fruit yield were also recorded. This was performed with a similar method followed by Magwaza et al.<sup>43</sup> To determine the chlorophyll pigment contents, the previous extraction method of Hernández-Hernández et al.<sup>44</sup> was used with slight modifications. Scanning electron microscopy (SEM) with energy-dispersive X-ray analysis (EDX) (SEM–EDX) measurements were carried out using a Quanta 200 FEI instrument equipped with a Quantax EDX detector to study the morphology of leaf surfaces and elemental distribution in the harvested matured tomato leaves according to the previous method of Xiong et al.<sup>45</sup>

**4.3.3. Nutrient Use Efficiency.** The following formula was used to measure NUE, which is the return in the tomato yield per unit of fertilizer nutrient applied<sup>19</sup>

$$\text{nutrient use efficiency (kg/ha)} = \frac{\text{tomato yield (kg/ha)}}{\text{quantity of fertilizer (kg/ha)}} \quad (1)$$

**4.4. Nutritional Analysis of Tomato.** **4.4.1. Proximate Composition, Vitamins, and Mineral Contents.** The major proximate components: moisture, protein, fat, ash, fiber, and carbohydrate were carried out by using AOAC methods (AOAC, 2000).<sup>46</sup> To determine the vitamin A content in ripe tomato, 1 mL of tomato extract and KOH solution were taken



in a test tube and shaken vigorously for 1 min. The absorbance of the extracted sample was measured at 335 nm. Finally, the content of vitamin A was calculated following the formula used by Olufemi Awolu.<sup>47</sup> The Vit-C content was also determined using the previous method of Ochoa-Velasco et al.<sup>36</sup> with slight modifications. The result was expressed as mg of AsA/100 g of the tomato sample. Similarly, the mineral content in tomato fruits was determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES) instruments (Model Trilogy-7).<sup>37</sup> Results were expressed as mg/100 g of samples. Sample measurements were performed in triplicate.

**4.4.2. Determination of Antioxidant Properties.** The total phenolic contents (TPC) and total tannin contents (TTC) were measured using the Folin–Ciocalteu assay as previously followed by Bao et al.<sup>48</sup> and Gaafar et al.,<sup>49</sup> respectively, with slight modifications. The results of TPC and TTC were expressed as mg of gallic acid equivalent (GAE) and mg of tannic acid equivalent (TAE), respectively, per 100 g of the tomato sample. In addition, the total flavonoid content (TFC) was also measured followed by the method previously used by Alenazi et al.<sup>2</sup> with slight modifications. The TFC of the extract (mg QE/100 g) was estimated by comparing their concentration against the standard curve. In addition, the free radical scavenging activity of the tomato solution was evaluated using the DPPH and the antioxidant capacity by the ABTS<sup>++</sup> assay was evaluated according to the previous method,<sup>50</sup> with slight modifications. All samples were determined as triplicate.

**4.5. Benefit–Cost Ratio (BCR) Analysis.** For the BCR analysis, the total income (BDT/ha) and the total cost of production (BDT/ha) were calculated. The following formula was used to calculate the BCR<sup>19</sup> for each treatment.

$$\text{benefit: cost (B: C) ratio} = \frac{\text{total income (BDT/ha)}}{\text{cost of cultivation (BDT/ha)}} \quad (2)$$

**4.6. Statistical Analysis.** The obtained data was statistically analyzed and scientifically presented. The significance of the differences was estimated and compared using the Duncan test at a 5% level of probability ( $p < 0.05$ ). To demonstrate the relationship between experimental variables, a simple linear correlation analysis was performed. However, all statistical analyses were carried out using “SPSS-version 20” a computer software package (2016).

## ■ ASSOCIATED CONTENT

### SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.1c03727>.

Experimental growing periods of tomato plants with matured fruits (see details in the Supporting Information as Figure S1) (PDF)

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## Notes

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