




## Article

# Effect of Silicon Nanoparticles on Tomato Plants Exposed to Two Forms of Inorganic Arsenic

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**Abstract:** In the environment arsenic (As) can be found mainly as arsenite (As<sup>III</sup>) and arsenate (As<sup>V</sup>), which are highly toxic and threaten food security. Currently, there is great attention on the effects of silicon dioxide nanoparticles (SiO<sub>2</sub> NPs) on plant development, and their ability to restrict As uptake. The results show that the two forms of As negatively impacted aerial dry biomass and fruit yield. Silicon content is lower in roots than in leaves. It is observed that As<sup>III</sup> is the form that accumulates the most in the root; in addition, the SiO<sub>2</sub> NPs reduce the translocation of As<sup>V</sup>. The data show that As<sup>III</sup> induced a negative effect on the uptake of Ca, P, Mg, and Cu, while SiO<sub>2</sub> NPs enhances the accumulation of Fe and Zn when exposed to As<sup>III</sup>. The two forms of As do not impact chlorophyll content but increases when interacting with SiO<sub>2</sub> NPs. Antioxidant enzymes APX, CAT, and SOD are higher in roots than in leaves. Phenols, flavonoids, and glutathione increased when SiO<sub>2</sub> NPs interacted with As<sup>III</sup> in roots. H<sub>2</sub>O<sub>2</sub> increases in roots and leaves by exposure to As<sup>V</sup> and As<sup>III</sup>, and its interactions with SiO<sub>2</sub> NPs, while in the fruit, H<sub>2</sub>O<sub>2</sub> production decreases. As for the total antioxidant capacity ABTS is observed to increase by As<sup>III</sup> + SiO<sub>2</sub> NPs only in roots. The bioactive compounds of the tomato fruits are modified by the treatments and the addition of SiO<sub>2</sub> NPs alone increase in lycopene content. Therefore, our results reveal the negative impacts of As<sup>III</sup>, and that SiO<sub>2</sub> NPs can at least partially mitigate As toxicity and reduce As<sup>V</sup> translocation in tomatoes.

**Keywords:** antioxidant defense system; arsenate; arsenite; nanotechnology



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## 1. Introduction

Arsenic (As) is a highly toxic chemical element that has been related to different negative effects on human health, such as bladder, kidneys, lung, and skin cancer, cardiovascular disease, and diabetes [1]. In addition, it is omnipresent in the lithosphere and can contaminate all water sources [2]. Additionally, in turn, As can be transferred in the food chain and distributed to entire populations of plants and animals, representing an enormous risk to human health [3].

The main forms of inorganic As in nature are found in two states of chemical oxidation, pentavalent arsenate (As<sup>V</sup>) and trivalent arsenite (As<sup>III</sup>) [4]. The oxidation state directly impacts toxicity, since As<sup>III</sup> can generally be 100 times more harmful than As<sup>V</sup> in most plant species [5]. The toxicity of As is because it alters the physiological, biochemical, metabolomic, and genomic processes of plants, resulting in a considerable decrease in their productivity [6]. Particularly, in northeast Mexico, As is found in high concentrations in water irrigated for agricultural purposes (8.684 mg L<sup>-1</sup>), and As concentrations exceed the 0.4 mg L<sup>-1</sup> established by the regulations for irrigation water in Mexico [7], which can induce potential negative effects on crop production and human health [8].

Tomato (*Solanum lycopersicum* L.) is consumed all over the world and is one of the most important crops, around 182.3 million tons of tomato fruits are produced. This crop is sensitive to stress induced by As negatively affecting plant growth and metabolism [9,10]. Interestingly, the concentration of As in the root and aerial parts of the tomato crop is dependent on the concentration in the irrigation water, thus being able to accumulate in the fruit and therefore be a risk for human consumption [11,12].

Silicon (Si) is found in soils naturally, with about 28% of the soils ranging from 0.52% to 47% since most of the rocks and minerals contain Si [12,13]. Si accumulates in plant tissue as hydrated amorphous silica polymer, in concentrations ranging between 0.1 and 10% of dry weight [14]. This element has the capacity to alleviate the harmful impact of heavy metals on plants [15]. It has the ability to reduce the deterioration of the cellular ultrastructure of roots and leaves of plants stressed by toxic metals [16].

Nanotechnology in agriculture is mainly focused on the ecological production of nanomaterials (NMs) for agricultural use, nanofertilizers, nanoencapsulates, nanofilms, and nanoparticles (NPs) [17]. NPs and NMs in low concentrations can favor plant growth; therefore, they can be considered biostimulants [18]. In addition, NPs have the potential as a protective agent for the alleviation of As toxicity in plants [19]. However, these nanometric materials can have different formulations and therefore different mechanisms of action, resulting in different responses in plants [20].

SiO<sub>2</sub> NPs have been shown to enhance antioxidant enzymatic activity and photosynthetic activity in *Aurundianria pigmaea* plants under metal stress [21]. Moreover, improve the development of *Triticum aestivum* L. plants and reduce the accumulation of cadmium in grains [22]. Even SiO<sub>2</sub> NPs increase phenolic compounds and essential oils of *Satureja hortensis* L. under cadmium stress [23]. In addition, SiO<sub>2</sub> NPs induce K<sup>+</sup> uptake, modulate Na<sup>+</sup> content, and decrease the negative impact on the cell wall of *Musa acuminata* “Gran Nain” plants in abiotic stress conditions [24]. Even so, they help induce root endodermal silicification and regulate cellular water balance [25]. As for the As-SiO<sub>2</sub> NPs interaction, a decrease in the absorption of As in *Oryza sativa* has been reported, and it can also stabilize cell integrity [26]. Interestingly, SiO<sub>2</sub> NPs can reduce the translocation of As<sup>V</sup> to the shoots of tomato plants and maintain the photosynthetic balance [27].

Therefore, the present research aimed to evaluate the impact of SiO<sub>2</sub> NPs, As<sup>III</sup>, and As<sup>V</sup> on tomato plant growth and fruit quality, as well as to determine the concentrations of Si, As, and nutrients in the plant tissue to know the translocation of these elements in the tissues.

## 2. Materials and Methods

### 2.1. Crop Development

The experiment was established in a polycarbonate greenhouse of Autonomous Agrarian University Antonio Narro. Tomato seed indeterminate growth and type saladete of the hybrid var. “El Cid F1” was used in this research. The seeds were germinated and subsequently transplanted into black bags with holes, filled with a mixture of perlite-peat moss as growth substrate. To make the correct use of fertilizers more efficient to provide adequate plant nutrition, we use the Steiner solution [28]. The pH of the solution was adjusted to 6.5 and the electrical conductivity (EC) reached 2.1 dS m<sup>-1</sup>. The concentration applied was 25% for the vegetative stage, 50% for flowering, 75% for fruit set, and 100% for fruit harvest.

A drip irrigation system was used, with four irrigations per day at different times. The amount of water contained in the Steiner solution applied was different for each phenological stage, applying about 2.5 L per plant per day in the stages of highest consumption.

### 2.2. Treatments

The following two forms of arsenic were supplemented: arsenate (Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O) and arsenite (NaAsO<sub>2</sub>), through the irrigation system containing the Steiner nutrient solution, at a constant amount of 3.2 mg L<sup>-1</sup> of As, to simulate contaminated water. SiO<sub>2</sub>

NPs ( $1000 \text{ mg L}^{-1}$ ) were applied directly to the growth substrate, and 10 mL of NPs solution was applied per plant every 21 days (6 applications in total). NPs solution was vortex before application to disperse NPs.  $\text{SiO}_2$  NPs were purchased at SkySpring Nanomaterials (Houston, TX, USA) with the following characteristics: spherical shape, size (10–20 nm), and surface area of  $160 \text{ m}^2 \text{ g}^{-1}$ .

The As concentration was chosen from a previous study where different concentrations of As in irrigation water were tested (0.2, 0.4, 0.8, 1.6, and  $3.2 \text{ mg L}^{-1}$  of  $\text{As}^{\text{V}}$ ) [27]. In addition, the chosen dose is considered contaminated irrigation water for agricultural use according to the Mexican legislation as follows: NOM-001-ECOL-1996 [29].

The concentration of NPs was chosen from a previous study with application of  $\text{SiO}_2$  NPs at concentrations between 250 and  $1000 \text{ mg L}^{-1}$  in tomato plants [27]. In addition, concentration selection in the above study was based on a study with applications of 100, 500, and  $2000 \text{ mg L}^{-1}$  of  $\text{SiO}_2$  NPs in cotton plants [30].

The experimental design was completely random, with the following six treatments: (1) Control; (2)  $1000 \text{ mg L}^{-1}$   $\text{SiO}_2$  NPs; (3)  $3.2 \text{ mg L}^{-1}$  de  $\text{As}^{\text{V}}$ ; (4)  $3.2 \text{ mg L}^{-1}$  de  $\text{As}^{\text{III}}$ ; (5)  $3.2 \text{ mg L}^{-1}$  de  $\text{As}^{\text{V}}$  +  $1000 \text{ mg L}^{-1}$   $\text{SiO}_2$  NPs; (6)  $3.2 \text{ mg L}^{-1}$  de  $\text{As}^{\text{III}}$  +  $1000 \text{ mg L}^{-1}$   $\text{SiO}_2$  NPs.

### 2.3. Agronomic Variables and Plant Material Sampling

At five months after transplantation the plants were evaluated the variables yield, to determine the vigor of the tomato plants. In addition, root and leaf samples were collected and the dry biomass of both shoots and roots was determined.

The samples dried in drying oven for 72 h at  $80 \text{ }^\circ\text{C}$ , then the dried tissue was ground with a mortar and pestle until you obtain a fine powder, it was used for analysis of minerals nutrients, and As. For biochemical analysis, five samples of roots and leaves were collected for each treatment (randomly). Roots were washed with water until all residues were removed and air dried inside the greenhouse. Both the leaves and the root were stored in a freezer at  $-20 \text{ }^\circ\text{C}$ . The samples were lyophilized at  $-80 \text{ }^\circ\text{C}$  for 72 h in a lyophilizer model D401 (Yamato Scientific Co., Ltd., Santa Clara, CA, USA). Completely lyophilized tissue samples were ground in a mortar to powder.

### 2.4. Arsenic, Silicon and Macro-Micronutrient

The concentrations of As, Si, Ca, K, P, Mg, S, Zn, Fe, and Cu, in roots, leaves, and fruit were determined. Subsequently, acid digestion was performed using concentrated  $\text{HNO}_3$ , and 0.2 g of the dry sample was taken. The final solution was filtered on Whatman filter paper (No. 42). The determination was made on an ICP-plasma emission spectrophotometer (Model 7400, Termo Jarrel Ash).

### 2.5. Biochemical Analysis

All biochemical analysis readings were performed on a spectrophotometer (UV-Vis-model UNICO UV2150, Dayton, NJ, USA).

#### 2.5.1. Chlorophyll Content

Chlorophylls in leaves were determined by the technique described in [31], using 0.1 g of fine lyophilized tissue. For this, a mixture of hexane-acetone solution (ratio 3:2) was made and 20 mL was added to the lyophilized tissue. Subsequently, it was centrifuged, and an aliquot of the supernatant was collected for reading in the spectrophotometer at two absorbances (between 645 and 663 nm). The absorbances used in Equations (1)–(3), and the values were expressed as  $\text{mg } 100 \text{ g}^{-1}$  dry weight.

$$\text{Chlorophyll } a = 0.999 \times \text{Abs}^{(663)} - 0.0989 \times \text{Abs}^{(645)} \quad (1)$$

$$\text{Chlorophyll } b = 0.328 \times \text{Abs}^{(663)} + 1.77 \times \text{Abs}^{(645)} \quad (2)$$

$$\text{Total Chlorophyll} = \text{Chlorophyll } a + \text{Chlorophyll } b \quad (3)$$

### 2.5.2. Antioxidant Enzymes

For antioxidant enzyme activity determinations, 20 mg of polyvinylpyrrolidone and 200 mg of lyophilized tissue (leaves and roots) were weighed. Subsequently, a 0.1 M phosphate buffer was prepared at a pH of 7, and 1.5 mL of this solution was added, and the mixture was then centrifuged for 10 min at  $16,128 \times g$  at  $4^\circ\text{C}$ . Finally, the supernatant was filtered through a nylon membrane and used in the determinations of antioxidant enzymes, ABTS, and glutathione.

Glutathione peroxidase activity (GPX) was determined using glutathione as substrate according to the technique of [32]. Catalase enzyme activity (CAT) was based on the quantification of the rate of  $\text{H}_2\text{O}_2$  reduction according to the technique described by [33]. Ascorbate peroxidase enzyme activity (APX) was based on the quantification of the rate of ascorbate oxidation according to the technique described by [34], and the SOD Cayman 706002<sup>®</sup> kit was used to determine the superoxide dismutase enzyme activity (SOD).

### 2.5.3. Non-Enzymatic Compounds

Vitamin C was performed by colorimetry using 2,6 dichlorophenol according to [35]. The determination of glutathione (GSH) was performed colorimetrically by reaction with 5,5-dithio-bis-2 nitrobenzoic acid, as described in [32]. Total flavonoids were determined with the technique described by [36]. Total phenols were quantified using the Folin-Ciocalteu assay, according to the technique described [37]. Lyophilized tissue was used for all determinations.

### 2.5.4. $\text{H}_2\text{O}_2$ Concentration and ABTS Radical

Hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) content was determined by the technique reported in [38]. Although some simple modifications were made as described in [39].

The ABTS radical was used to determine antioxidant capacity using the technique of [40]. For this, the lipophilic compounds were extracted with hexane:acetone solution, and the hydrophilic compounds using a phosphate buffer solution. For both  $\text{H}_2\text{O}_2$  and ABTS we use lyophilized tissue.

## 2.6. Fruit Quality and Antioxidants

At the time of harvest, the tomato fruits were selected without damage, homogeneous size, and intense red color [41]. The quality of the tomato fruit is determined by the following variables: fruit firmness, total soluble solids, and hydrogen potential, as described in [42]. While Vitamin C, flavonoids, phenols, and glutathione are described in Section 2.5.3.

For the determination of the lycopene and  $\beta$ -carotene the procedure was followed as described in Section 2.5.1. according to [31]. In addition, four absorbances (453, 505, 645, 663) are used to calculate the concentration of these antioxidants as shown in Equations (4) and (5).

$$\text{Lycopene} = -0.0458 \times \text{Abs}^{(663)} + 0.204 \times \text{Abs}^{(645)} + 0.372 \times \text{Abs}^{(505)} - 0.0806 \times \text{Abs}^{(453)} \quad (4)$$

$$\beta\text{-carotene} = 0.216 \times \text{Abs}^{(663)} - 1.22 \times \text{Abs}^{(645)} - 0.304 \times \text{Abs}^{(505)} + 0.452 \times \text{Abs}^{(453)} \quad (5)$$

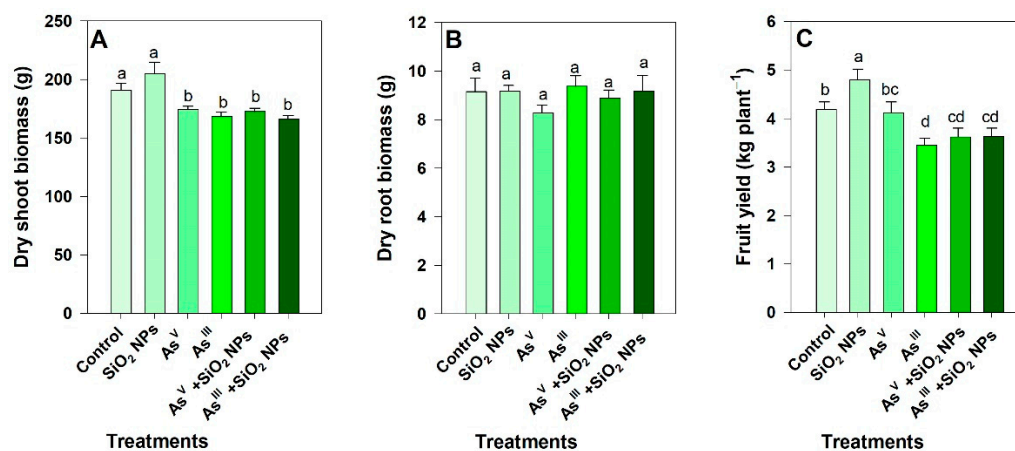
### 2.7. Statistical Analysis

A one-way analysis of variance was performed for all the variables evaluated, and to compare the means the Fisher's Least Significant Difference test was applied ( $p \leq 0.05$ ), and data normality was evaluated by Shapiro-Wilks method. All procedures were performed with the statistical program Infostat (V2018) ([www.infostat.com.ar](http://www.infostat.com.ar), accessed on 26 September 2022).

### 3. Results

#### 3.1. Effect of SiO<sub>2</sub> NPs and Arsenic on Tomato Plants

The application of As<sup>V</sup>, As<sup>III</sup>, and SiO<sub>2</sub> NPs had significant effects on plant vigor variables of tomato plants (Figure 1). The dry shoot biomass decreased by 8.53%, 11.57%, 9.49%, and 12.75% when As<sup>V</sup>, As<sup>III</sup>, and their interactions with SiO<sub>2</sub> NPs were applied, respectively (Figure 1A). The dry root biomass did not show any statistical differences between the treatments evaluated (Figure 1B). The fruit yield was increased by 14.55% by the single application of SiO<sub>2</sub> NPs compared to the control. As for the As<sup>V</sup>, it did not show statistical differences with respect to the control; however, when it interacted with SiO<sub>2</sub> NPs, it reduced the fruit yield by 13.60%. The application of As<sup>III</sup> presented the lowest fruit yield, being 17.66% less than the control, but the interaction with SiO<sub>2</sub> NPs only reduced it by 13.36% (Figure 1C).



**Figure 1.** Effect of two forms of inorganic As (As<sup>V</sup>-As<sup>III</sup>) and SiO<sub>2</sub> NPs on the growth and yield of tomato plants. (A) Dry shoot biomass, (B) dry root biomass, and (C) fruit yield. Plants treated with 3.2 mg L<sup>-1</sup> of As<sup>V</sup> (Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O) and As<sup>III</sup> (NaAsO<sub>2</sub>) via irrigation through the nutrient solution and 1000 mg L<sup>-1</sup> of SiO<sub>2</sub> NPs via substrate. Different letters per bar indicate significant differences according to the Fisher's least significant difference test ( $p \leq 0.05$ ).  $n = 5 \pm$  standard error.

#### 3.2. Silicon and Arsenic Concentration, and Translocation Factor

Si concentration did not show statistical differences in root tissue (Figure 2A). In the leaves, the treatment with As<sup>III</sup> decreased the Si concentration by 54.67% with respect to the control, while the rest of the treatments were statistically equal (Figure 2B). Si concentration in fruits increased 52.9% and 61.39% due to As<sup>V</sup> exposure and As<sup>III</sup> + SiO<sub>2</sub> NPs interaction, respectively (Figure 2C).

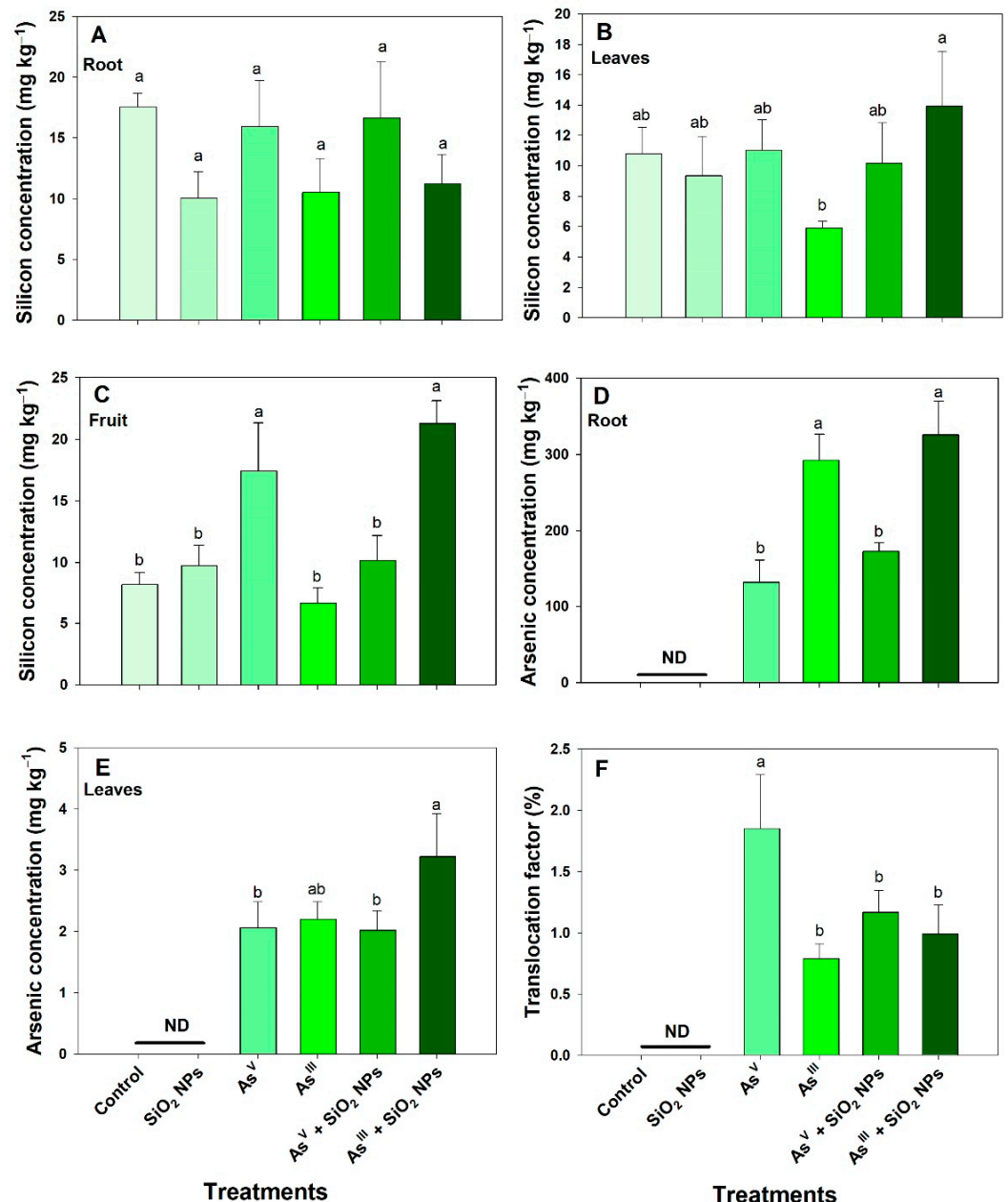
The concentration of As in roots showed a greater accumulation when exposed to As<sup>III</sup> and its interaction with the SiO<sub>2</sub> NPs (Figure 2D). While the concentration of As in leaves showed an increase in the treatment of As<sup>III</sup> + SiO<sub>2</sub> NPs, while the rest of the treatments were statistically equal (Figure 2E). On the other hand, As was not detected in ripe tomato fruits.

The treatment with As<sup>V</sup> showed a higher translocation factor, but when the SiO<sub>2</sub> NPs were applied, 37.3% of As was reduced to the aerial part. While As<sup>III</sup> did not show a statistical difference in the translocation factor when the SiO<sub>2</sub> NPs were applied (Figure 2F).

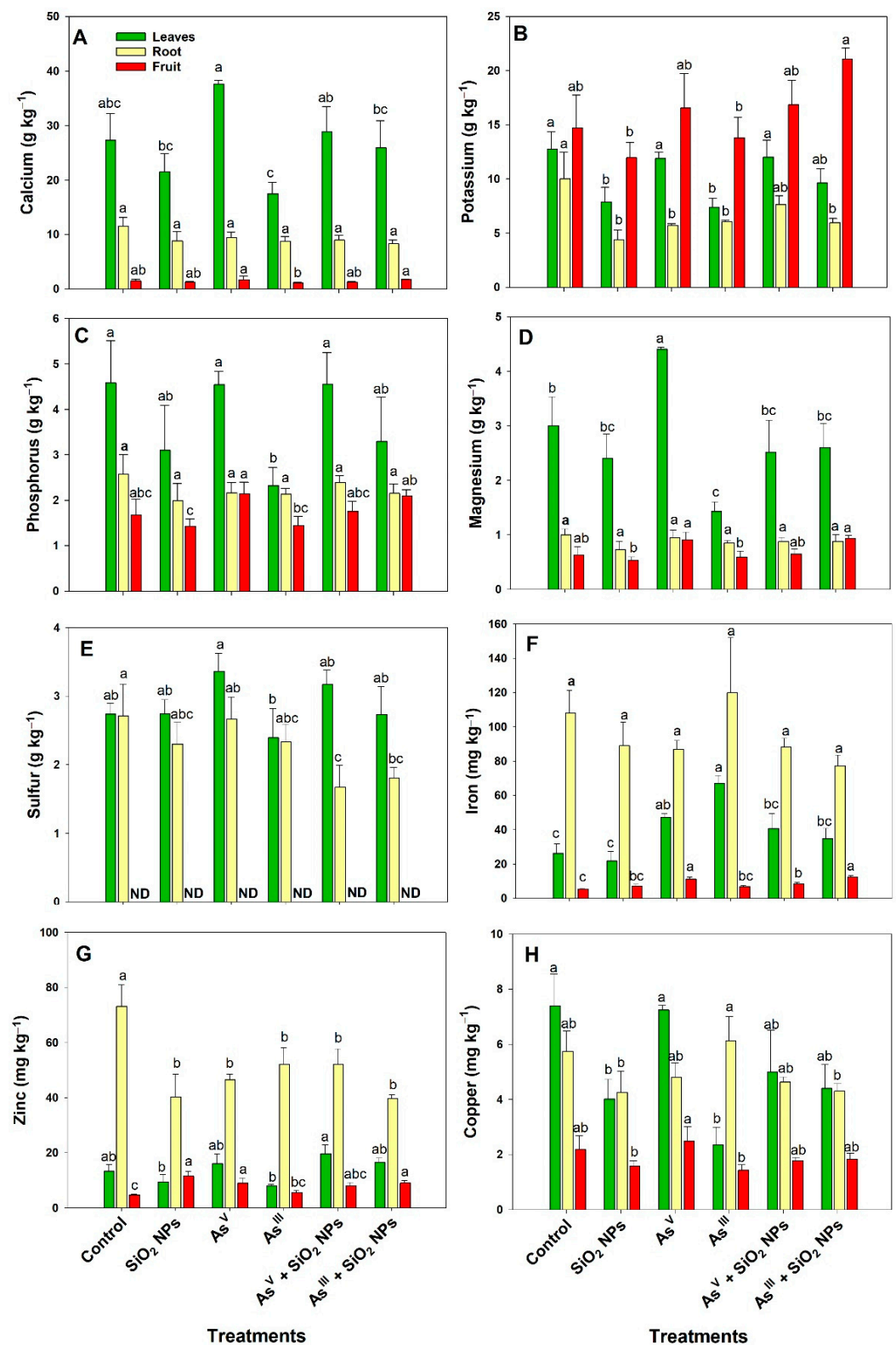
#### 3.3. Concentration of Macro and Micronutrients in Tissues

As<sup>III</sup> decreased 43.7% and 38.31% of the concentration of Ca in leaves and fruits, respectively; however, in roots there were no differences (Figure 3A). The concentration of K showed differences in the three plant organs. The K in leaves decreased by 38.48% and 42.22% in the SiO<sub>2</sub> NPs and As<sup>III</sup> treatments, respectively. The concentration of K in roots was lower in all treatments compared to the control. While in fruits, the As<sup>III</sup> + SiO<sub>2</sub> NPs treatment increased it by 43.04% (Figure 3B). The concentration of P in leaves decreased

50.43% with the application of  $\text{As}^{\text{III}}$ , while the rest of the treatments showed no statistical differences. In roots and fruits, the concentration of P did not show statistical differences (Figure 3C). The concentration of Mg in the leaves increased 46.66% with exposure to  $\text{As}^{\text{V}}$ ; however, when exposed to  $\text{As}^{\text{III}}$ , it decreased by 47.33%. In roots and fruits, Mg did not have statistical differences between treatments compared to the control (Figure 3D). The concentration of S in leaves did not show statistical differences between treatments evaluated, while in roots a decrease of 38.37–33% was observed in the  $\text{As}^{\text{V}}$  +  $\text{SiO}_2$  NPs and  $\text{As}^{\text{III}}$  +  $\text{SiO}_2$  NPs treatments, respectively; in fruits, S was not detected (Figure 3E).



**Figure 2.** Silicon concentration in (A) roots, (B) leaves, and (C) fruits. Arsenic concentration in (D) roots, (E) leaves, and (F) arsenic translocation factor. Plants treated with  $3.2 \text{ mg L}^{-1}$  of  $\text{As}^{\text{V}}$  ( $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ ) and  $\text{As}^{\text{III}}$  ( $\text{NaAsO}_2$ ) via irrigation through the nutrient solution and  $1000 \text{ mg L}^{-1}$  of  $\text{SiO}_2$  NPs via substrate. ND: no detected. Lower-case letters different per bar indicate statistical differences according to the Fisher's least significant difference test ( $p \leq 0.05$ ).  $n = 5 \pm$  standard error.



**Figure 3.** Macronutrients (A–E) and micronutrients (F–H) in leaves, roots, and fruits of tomato plants exposed to As<sup>V</sup> and As<sup>III</sup> in irrigation water and SiO<sub>2</sub> NPs via substrate. Plants treated with 3.2 mg L<sup>-1</sup> of As<sup>V</sup> (Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O) and As<sup>III</sup> (NaAsO<sub>2</sub>) via irrigation through the nutrient solution and 1000 mg L<sup>-1</sup> of SiO<sub>2</sub> NPs via substrate. ND: no detected. Lower-case letters different per bar indicate statistical differences according to the Fisher’s least significant difference test ( $p \leq 0.05$ ).  $n = 5 \pm$  standard error.

The concentration of Fe in leaves increased by 79.64% and 155.6% due to the application of As<sup>V</sup> and As<sup>III</sup>, respectively, while in roots there were no differences between treatments. The concentration of Fe in fruits increased by 105.34% by the application of As<sup>V</sup> alone, while the As<sup>V</sup> + SiO<sub>2</sub> NPs and As<sup>III</sup> + SiO<sub>2</sub> NPs treatments increased the accumulation of Fe by 55.43% and 127.99%, respectively (Figure 3F). The concentration of Zn in leaves did not present statistical differences in the treatments evaluated; while in roots, all treatments decreased compared to the control. In fruits, the SiO<sub>2</sub> NPs, As<sup>V</sup> and As<sup>III</sup> + SiO<sub>2</sub> NPs treatments increased Zn by 151.6%, 96.07% and 96.04% respectively (Figure 3G).

The concentration of Cu in leaves decreased 45.60% due to the application of SiO<sub>2</sub> NPs and 214.46% due to exposure to As<sup>III</sup>. The application of SiO<sub>2</sub> NPs alone decreased the Cu concentration in roots by 25.95%, and 27.98% in fruits (Figure 3H).

### 3.4. Impact on Chlorophyll Content

Chlorophylls (*a*, *b*, and total) in tomato plants exposed to As and SiO<sub>2</sub> NPs showed statistical differences between treatments evaluated (Figure 4). Chlorophyll *a* content increased by 13.02% and 12.52% in treatments that were exposed to As<sup>V</sup> and As<sup>III</sup> in interaction with SiO<sub>2</sub> NPs compared to As alone (Figure 4A). Chlorophyll *b* increased by 25.17% when As<sup>V</sup> + SiO<sub>2</sub> NPs were applied, while As<sup>III</sup> + SiO<sub>2</sub> NPs increased by 19.17%. The rest of the treatments did not show statistical differences with respect to the control (Figure 4B). SiO<sub>2</sub> NPs, As<sup>V</sup>, and As<sup>III</sup> when applied alone did not impact the total chlorophyll content; however, when plants were exposed to As<sup>V</sup> and As<sup>III</sup> in interaction with SiO<sub>2</sub> NPs, there was an increase of 18.60% and 19.26% respectively (Figure 4C).

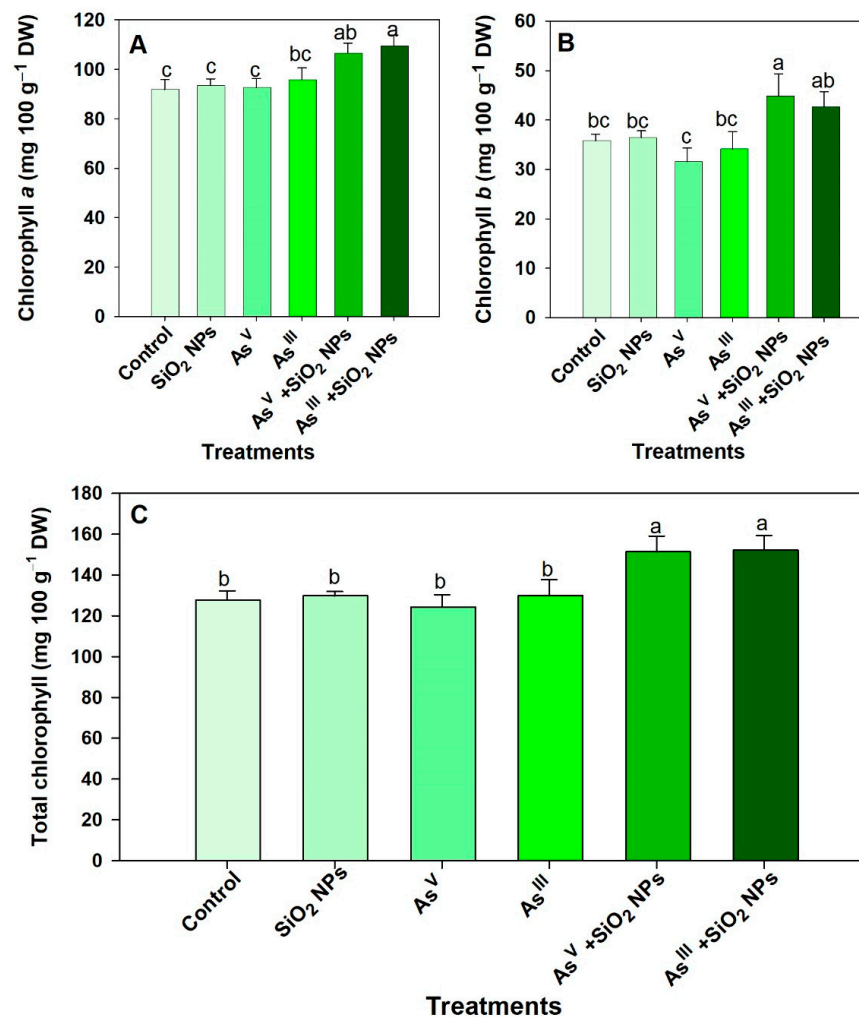


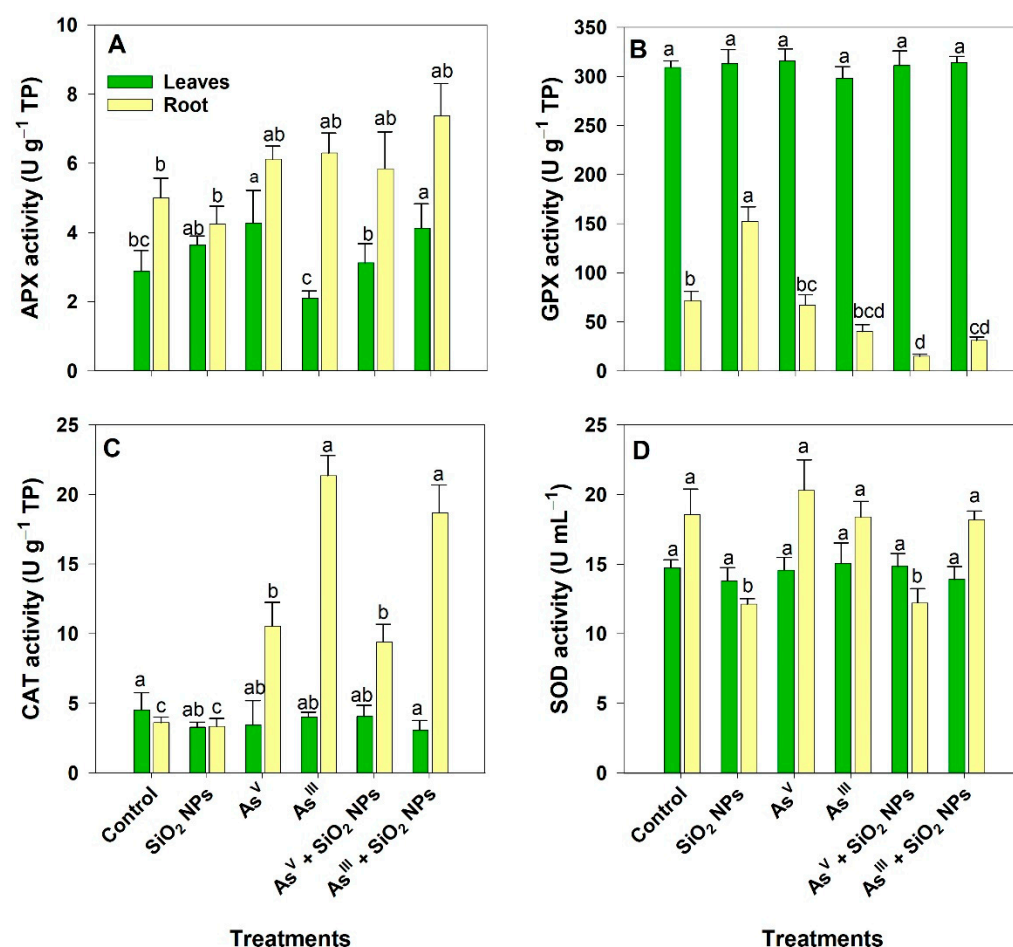
Figure 4. Chlorophylls content in leaves of tomato plants. (A) chlorophyll *a*, (B) chlorophyll *b*, and



(C) total chlorophyll increase when plants are exposed to the combination of As<sup>V</sup>-As<sup>III</sup> with SiO<sub>2</sub> NPs. Plants treated with 3.2 mg L<sup>-1</sup> of As<sup>V</sup> (Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O) and As<sup>III</sup> (NaAsO<sub>2</sub>) via irrigation through the nutrient solution and 1000 mg L<sup>-1</sup> of SiO<sub>2</sub> NPs via substrate. Lower-case letters different per bar indicate statistical differences according to the Fisher's least significant difference test ( $p \leq 0.05$ ).  $n = 5 \pm$  standard error.

### 3.5. Antioxidant Enzymes

Antioxidant enzymatic activities of both roots and leaves were modified when exposed to As and SiO<sub>2</sub> NPs (Figure 5). APX activity in the leaves increased 48.9% due to the effect of As<sup>V</sup>, while As<sup>III</sup> had a negative effect on APX activity, decreasing up to 26.98%. However, the addition of SiO<sub>2</sub> NPs increased APX activity by 42.90%, reversing the negative impact of As<sup>III</sup>. In the root, an increase was observed due to the application of As alone, and in interaction with the SiO<sub>2</sub> NPs, the interaction of As<sup>III</sup> + SiO<sub>2</sub> NPs increased by 47.6%; however, all treatments were statistically equal to the control (Figure 5A).



**Figure 5.** Antioxidant enzyme activity in the roots and leaves of tomato plants. (A) APX: ascorbate peroxidase, (B) GPX: glutathione peroxidase, (C) CAT: catalase, and (D) SOD: superoxide dismutase. Plants treated with 3.2 mg L<sup>-1</sup> of As<sup>V</sup> (Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O) and As<sup>III</sup> (NaAsO<sub>2</sub>) via irrigation through the nutrient solution and 1000 mg L<sup>-1</sup> of SiO<sub>2</sub> NPs via substrate. Lower-case letters different per bar indicate statistical differences according to the Fisher's least significant difference test ( $p \leq 0.05$ ).  $n = 5 \pm$  standard error.

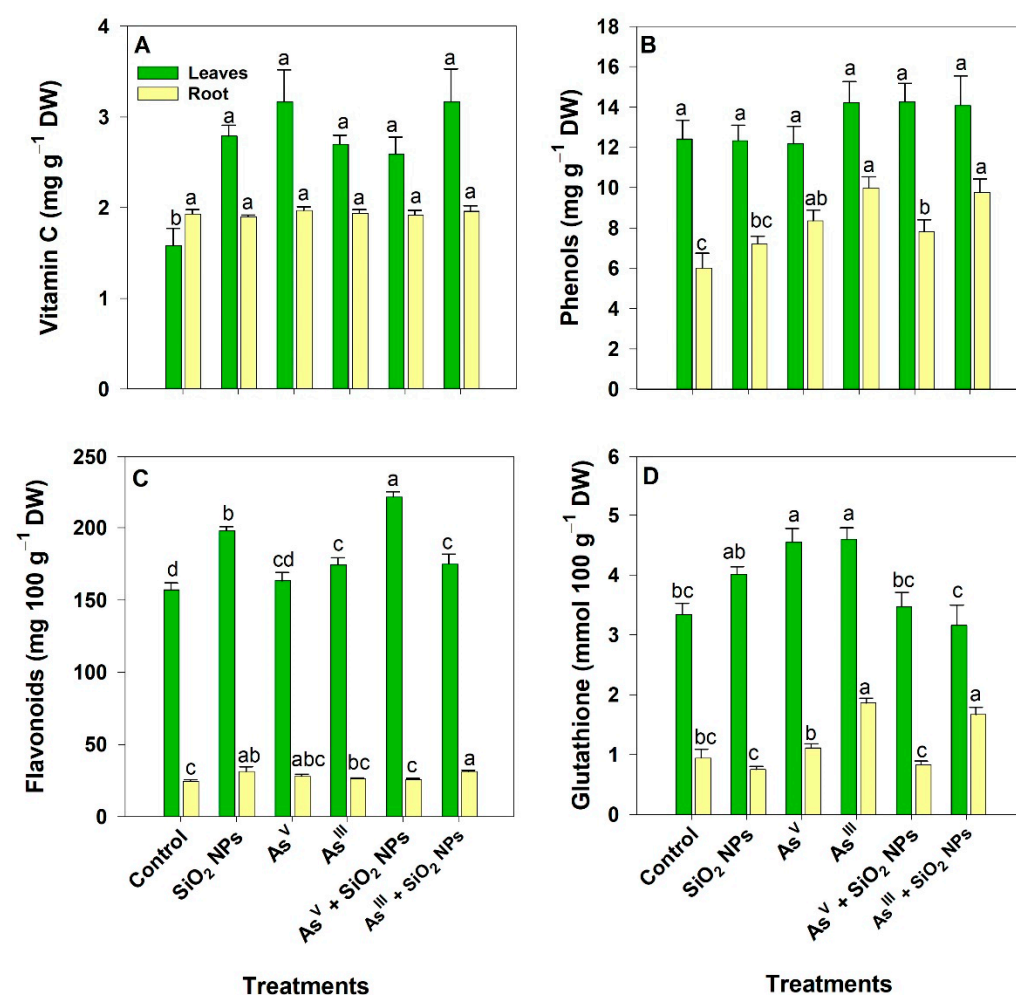
GPX enzymatic activity was different in roots than in leaves. In leaves, there were no statistical differences between treatments. In the root, the GPX activity increased by 112.80% by the application of SiO<sub>2</sub> NPs, while in interaction with As<sup>V</sup> and As<sup>III</sup> it decreased by 79.04% and 70.30%, respectively (Figure 5B).

The CAT activity did not show statistical differences by application of the treatments. In the CAT activity in roots,  $As^V$  induced an increase of 191%, and when it interacted with  $SiO_2$  NPs, it increased by 160.38%. The presence of  $As^{III}$  seems to have a greater effect on CAT activity. It was increased by up to 491%, while the  $As^{III} + SiO_2$  NPs interaction increased by 417.7% (Figure 5C).

The SOD enzymatic activity in leaves did not present statistical differences between treatments. In the root, the  $SiO_2$  NPs treatment decreased SOD activity by 34.67%, while the  $As^V + SiO_2$  NPs interaction reduced it by 34.19%; the remaining treatments were statistically the same as the control (Figure 5D).

### 3.6. Non-Enzymatic Antioxidants

The vitamin C content is generally higher in the leaves than in the roots, with statistical differences only in the leaves where vitamin C increased in all treatments with respect to control in a range of 63.92–100.63% (Figure 6A).



**Figure 6.** Non-enzymatic antioxidant compounds in tomato roots and leaves exposed to two forms of inorganic As and  $SiO_2$  NPs. (A) Vitamin C, (B) phenols, (C) flavonoids, and (D) glutathione. Plants treated with  $3.2 \text{ mg L}^{-1}$  of  $As^V$  ( $Na_2HAsO_4 \cdot 7H_2O$ ) and  $As^{III}$  ( $NaAsO_2$ ) via irrigation through the nutrient solution and  $1000 \text{ mg L}^{-1}$  of  $SiO_2$  NPs via substrate. Lower-case different letters per bar indicate statistical differences according to the Fisher's least significant difference test ( $p \leq 0.05$ ).  $n = 5 \pm$  standard error.

The concentration of phenols in the leaves did not present statistical differences between the treatments. While phenols increased in roots (39.16% and 30.14%) with the application of  $As^V$  and in interaction with  $SiO_2$  NPs, respectively. While  $As^{III}$  increased

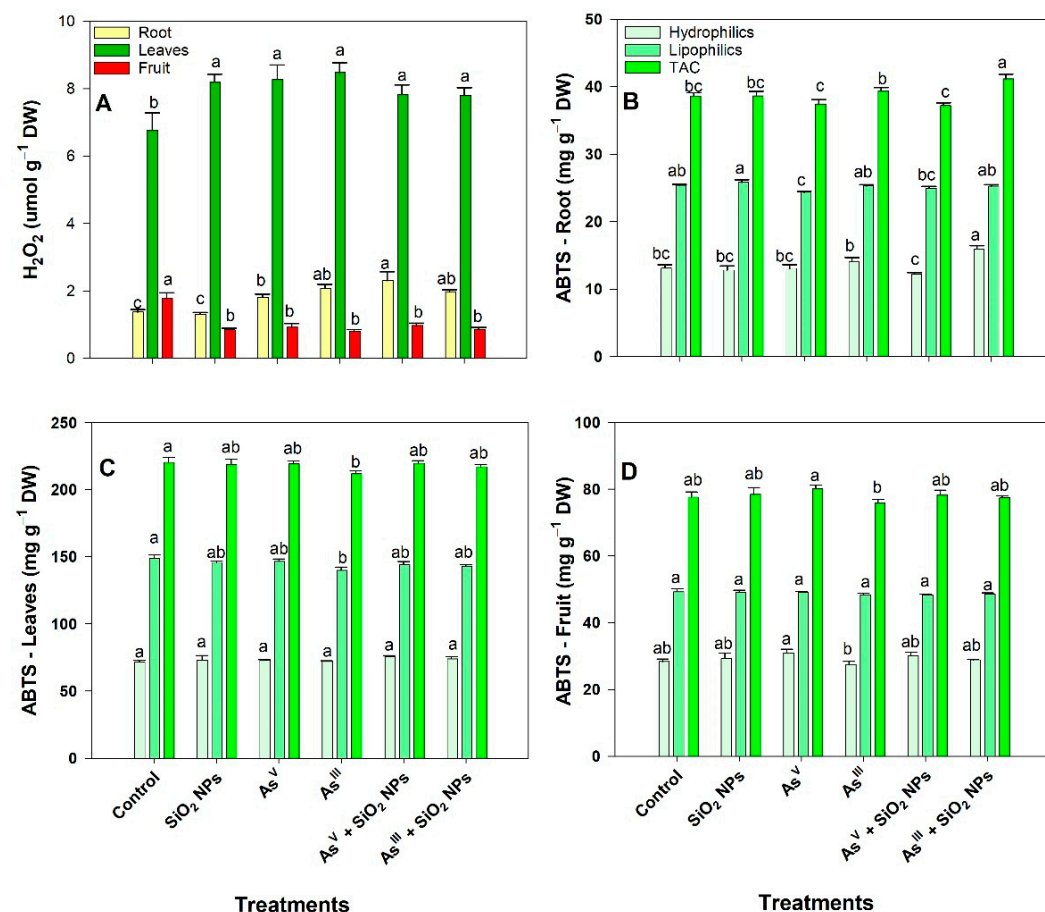
the phenol content by 58.16%, and the  $\text{As}^{\text{III}} + \text{SiO}_2$  NPs interaction induced an increase of 62.66% (Figure 6B).

Flavonoids in roots and leaves behave differently; in the leaves, a higher concentration of flavonoids was observed. The application of  $\text{SiO}_2$  NPs increased the flavonoid content by 26.1%,  $\text{As}^{\text{III}}$  increased by 11%, and the  $\text{As}^{\text{V}} + \text{SiO}_2$  NPs and  $\text{As}^{\text{III}} + \text{SiO}_2$  NPs interactions increased by 41.35% and 11.37%, respectively. In the root, the  $\text{SiO}_2$  NPs increased the flavonoid content by 27.62%, while the  $\text{As}^{\text{III}} + \text{SiO}_2$  NPs interaction induced an increase of 11.37% (Figure 6C).

The GSH content in the leaves increased by 36.22% and 37.72% due to exposure to  $\text{As}^{\text{V}}$  and  $\text{As}^{\text{III}}$ , respectively; the other treatments were statistically equal to the control. In the roots,  $\text{As}^{\text{III}}$  induced an increase of GSH of 94.68%, and in interaction with  $\text{SiO}_2$  NPs it increased by 77.65%. The other treatments were statistically equal to the control (Figure 6D).

### 3.7. Hydrogen Peroxide and Antioxidant Capacity

The level of  $\text{H}_2\text{O}_2$  in the leaves increased with all treatments in a range from 15.2 to 25.65%. In the root,  $\text{H}_2\text{O}_2$  increased by 31.15% and 50% due to exposure to  $\text{As}^{\text{V}}$  and  $\text{As}^{\text{III}}$ , respectively. While the interaction of  $\text{As}^{\text{V}}$  and  $\text{As}^{\text{III}}$  with  $\text{SiO}_2$  NPs increased the  $\text{H}_2\text{O}_2$  level by 67.39% and 43.47%, respectively. In the fruits, all the treatments decreased the level of  $\text{H}_2\text{O}_2$ , in a range of 44.9–54.49% (Figure 7A).



**Figure 7.** Hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) and ABTS antioxidant capacity in roots, leaves, and fruits of tomato plants. (A)  $\text{H}_2\text{O}_2$  levels, (B) antioxidant capacity ABTS in roots, (C) antioxidant capacity ABTS in leaves, and (D) antioxidant capacity ABTS in fruits. Plants treated with  $3.2 \text{ mg L}^{-1}$  of  $\text{As}^{\text{V}}$  ( $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ ) and  $\text{As}^{\text{III}}$  ( $\text{NaAsO}_2$ ) via irrigation through the nutrient solution and  $1000 \text{ mg L}^{-1}$  of  $\text{SiO}_2$  NPs via substrate. Lower-case letters different per bar indicate statistical differences according to the Fisher's least significant difference test ( $p \leq 0.05$ ).  $n = 5 \pm$  standard error.

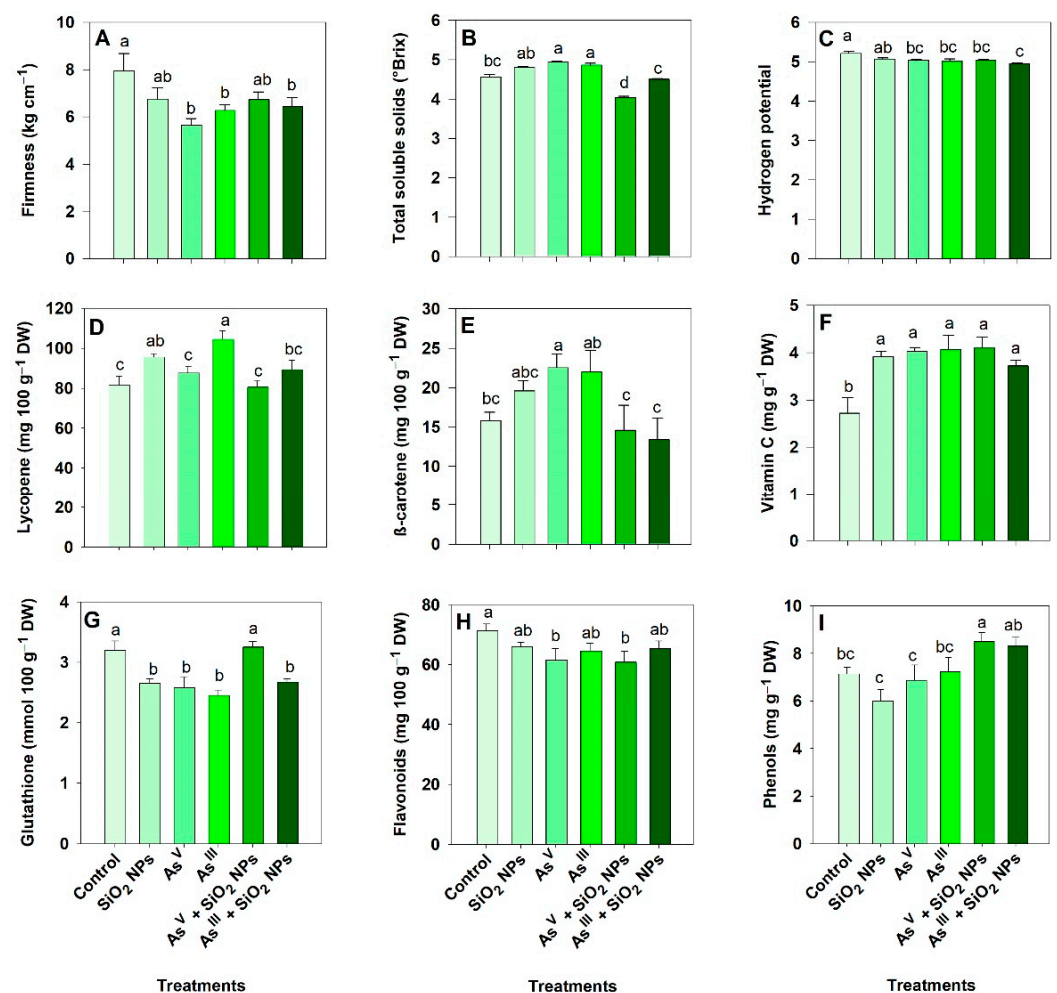
ABTS antioxidant capacity in roots had statistical differences between treatments. In hydrophilic compounds, the interaction of As<sup>III</sup> with SiO<sub>2</sub> NPs increased by 21.24%. In the lipophilic compounds, As<sup>V</sup> decreased the antioxidant capacity by 4.13%. The total antioxidant capacity was higher (6.6%) in the As<sup>III</sup> + SiO<sub>2</sub> NPs treatment, in relation to control (Figure 7B).

The antioxidant capacity in leaves did not present a statistical difference between treatments in hydrophilic compounds. In the lipophilic compounds, the As<sup>III</sup> treatment decreased the antioxidant capacity by 6.36%. Total antioxidant capacity was increased by 3.8% with As<sup>III</sup> treatment (Figure 7C).

The antioxidant capacity in fruits did not show any statistical differences in lipophilic compounds. While the antioxidant capacity of hydrophilic compounds was the only difference between the two forms of arsenic, being higher with As<sup>V</sup> (Figure 7D).

### 3.8. Effect of As and SiO<sub>2</sub> NPs on the Quality of Tomato Fruits

Fruit firmness decreased by 28.93% and 21.13% in the treatments with As<sup>V</sup> and As<sup>III</sup> application, respectively, while the As<sup>III</sup> + SiO<sub>2</sub> NPs interaction decreased by 18.99% in relation to the control (Figure 8A).



**Figure 8.** Quality of tomato fruits exposed to two forms of inorganic As and SiO<sub>2</sub> NPs, (A) Firmness, (B) total soluble solids, (C) hydrogen potential, (D) lycopene content, (E) β-carotene content, (F) vitamin C, (G) glutathione, (H) flavonoids, and (I) phenols. Plants treated with 3.2 mg L<sup>-1</sup> of As<sup>V</sup> (Na<sub>2</sub>HAsO<sub>4</sub>·7H<sub>2</sub>O) and As<sup>III</sup> (NaAsO<sub>2</sub>) via irrigation through the nutrient solution and 1000 mg L<sup>-1</sup> of SiO<sub>2</sub> NPs via substrate. Lower-case letters different per bar indicate statistical differences according to the Fisher's least significant difference test ( $p \leq 0.05$ ).  $n = 5 \pm$  standard error.

Total soluble solids increased when plants were exposed to As<sup>V</sup> (8.33%) and As<sup>III</sup> (6.57%); however, when As<sup>V</sup> interacted with SiO<sub>2</sub> NPs, they decreased by 11.40% (Figure 8B).

The pH of ripe fruits was not modified by the application of SiO<sub>2</sub> NPs, however, the exposure to the two forms of As and its interaction with the SiO<sub>2</sub> NPs decreased the pH in a range of 3.45–4.99% (Figure 8C).

The main antioxidant lycopene in ripe tomato fruit increased by 17.17% by the application of SiO<sub>2</sub> NPs, while As<sup>III</sup> induced an increase of 28.12% (Figure 8D).

β-carotene increased with As<sup>V</sup> by 42.44% (Figure 8E). Vitamin C increased with all applied treatments in a range of 36.8–49.63% in relation to the control (Figure 8F).

With the exception of the As<sup>V</sup> + SiO<sub>2</sub> NPs treatment, the rest decreased the glutathione content in the fruits in a range of 13.70–23.43% (Figure 8G).

Flavonoids were decreased by exposure to As<sup>V</sup> alone or in combination with SiO<sub>2</sub> NPs by 13.70 and 14.74%, respectively (Figure 8H).

Phenol content increased with the As<sup>V</sup> + SiO<sub>2</sub> NPs treatment by 19.12%, while the As<sup>III</sup> + SiO<sub>2</sub> NPs interaction induced an increase of 16.38%, although it was not different from the control (Figure 8I).

## 4. Discussion

### 4.1. Impact of As and SiO<sub>2</sub> NPs on Biomass and Fruit Yield

In this study, both As<sup>V</sup> and As<sup>III</sup> negatively impacted shoot dry biomass, and As<sup>III</sup> reduced fruit yield. The literature reports a negative effect on plant development by exposure to arsenic, and our study showed that reported trend. For example, negative effects on biomass due to the presence of As in the soil have been reported in tomato, rice, and parsley plants [9,43]. Moreover, As<sup>III</sup> causes a reduction of the dry biomass of rice plants by up to 50% when they are grown in a nutrient solution with a concentration of 4.0 mg L<sup>-1</sup> of this metalloid [43]. Although it has been reported that As<sup>III</sup> is much more toxic than As<sup>V</sup>, a serious affectation of growth was observed in the *Atriplex atacamensis* plant, an effect not observed in As<sup>V</sup> treatment [44]. Therefore, the exposition of plants to As causes physiological alterations in plants and interrupts the accumulation of biomass due to its high toxicity [45].

The negative impact is due to arsenic contamination in any of its two forms (As<sup>V</sup> and As<sup>III</sup>) can negatively interfere with plant metabolism, increasing the formation of reactive oxygen species (ROS) [46]. Furthermore, As<sup>V</sup> disrupts cellular metabolism because it replaces phosphorus in respiration processes, instead of generating adenosine triphosphate, it generates adenosine diphosphate-arsenate [47]. While the interaction of As<sup>III</sup> with the sulfhydryl groups of enzymes and the formation of ROS, severely alters plant metabolism, DNA structure, and lipid-protein metabolism [48].

In our study, SiO<sub>2</sub> NPs alone increased fruit yield. When the NPs come in contact with the root or leaves, they are absorbed and can be transported, generating a stimulating effect on plants subjected to abiotic stress [49]. This may be due to the promotion of enzymatic activity induced by the NPs, which intervene in the metabolism and acquisition of nutrients, and improve the opening of stomata, which results in an increase in gas exchange and CO<sub>2</sub> assimilation [50]. They also improve hormone content in plants, such as indoleacetic acid, gibberellins, and cytokinins, and can positively regulate gene transcription [51,52].

The application of Si NPs to plants subjected to heavy metal stress conditions can improve the development of plants [53,54]. Although in our study, SiO<sub>2</sub> NPs could not reverse the negative effect on shoot biomass and yield of tomato plants in the presence of As, and also did not cause increased toxicity when interacting with As. In this regard, the effect of Si NPs on plant stress is related to the physical and chemical properties of the Si NPs, the soil, and the type of stress [55].

### 4.2. Concentration of Si, As and Nutrients

In this study, the application of As<sup>III</sup> in the irrigation water did not impact root Si uptake but decreased the content of silicon in the leaves of tomato plants. This impact

can be attributed to As<sup>III</sup> uptake by roots, which is mediated by Si transporters [56]. For example, (*Lsi1*, *Lsi2*, *Lsi3*, and *Lsi6*), and also the anatomy of the different roots, show a variable absorption of Si between plants [57]. It has also been shown that Si accumulates mainly in plant shoots and has a lower content in the roots [58]. While a higher applied amount of Si linearly increases the content of this element in the different organs [59].

Interestingly, in our study, the application of SiO<sub>2</sub> NPs to As<sup>III</sup> stressed plants induced a higher Si content in leaves. This is due to the form of Si (ionic or NPs) translating into a different concentration, being higher when it is applied in the form of NPs [60]. In this regard, Wang et al. [61] reported that the SiO<sub>2</sub> NPs in rice seedlings induced a greater accumulation of Si when the seedlings developed in the presence of Cd and As. The impact was dependent on the concentration of NPs (between 150, 500 and 2000 mg kg<sup>-1</sup>) and the conditions of irrigation.

In our study, both forms of As induced As accumulation in the roots and leaves of tomato plants, but in the tomato fruits, it was not detected. The entry of As into tomato plants is due to its similarity with P. Therefore, As can enter through aquaglyceroporins and P nutrient uptake pathways, e.g., phosphate permeases [62]. In addition, the increase in As in the water used to irrigate agricultural plants results in a greater accumulation of As in the roots and shoots [63]. Various studies have shown that tomato plants accumulate most of the As in the roots and a very low concentration is translocated to the aerial parts, which means that the consumption of the human population presents a lower risk [8,64–66]. Interestingly, tomato plants reduce As transport to the aerial parts and increase As accumulation in the roots, as a strategy to tolerate this metalloid [67].

In our research, the application of SiO<sub>2</sub> NPs reduces the translocation of As<sup>V</sup> to the aerial parts but has no impact on the translocation of As<sup>III</sup>. It has been shown that the addition of SiO<sub>2</sub> NPs to the roots of tomato plants irrigated with As<sup>V</sup> helps to restrict the translocation of As to the aerial parts [27]. Even the application of chitosan-Si NPs promotes arsenic fixation in the leaf cell wall and reduces the As concentration in rice grain by 61.2% [68]. In this regard, [69] informed that Si can chelate, compartmentalize, and coprecipitate toxic metals, and can regulate antioxidants. In addition, Si NPs decrease oxidative stress and increase the potential of the photosynthetic system in metal-stressed plants [22]. Even the application of SiO<sub>2</sub> NPs can enhance gene expression of *OsNIP1;1* and *OsNIP3;3* and can significantly reduce the entry of As into the cells [26]. All of these processes can improve plant health and can restrict metal translocation. On the other hand, a reduction of As<sup>III</sup> translocation in rice plants with silicon application is commonly reported, but it has also been shown that Si application does not affect As<sup>III</sup> concentration in roots and does not affect DMA uptake and accumulation in roots and shoots [56]. Therefore, further studies should be conducted on tomato plants to understand more about this Si-As<sup>III</sup> interaction process because in our study, the application of SiO<sub>2</sub> NPs did not influence As<sup>III</sup> translocation.

In this study, the uptake and accumulation of most essential nutrients in tomato plants were negatively impacted by the application of the two forms of As in irrigation water. This is because As can alter metabolic processes and can compete directly with nutrients, influencing the uptake and distribution of nutrients in plants [70]. Reduced Ca, P, Mg, and N uptake have been reported in different tissues of perennial ryegrass plants subjected to an As stress condition [71]. In *Cajanus cajan* L. plants, lower nutrient uptake was observed when they were exposed to As<sup>III</sup> compared to As<sup>V</sup> [72].

Furthermore, findings of current research revealed that the addition of SiO<sub>2</sub> NPs in tomato plants subjected to arsenic stress conditions on the absorption of mineral nutrients showed the following three impacts: (1) no effect, (2) negative effect, and (3) positive effect at least on Ca and Cu in fruits. In this context, it has been shown that SiO<sub>2</sub> NPs increased the content of nutrients such as Mg, P, and S in *Lilium* plants, improving the quality of their flowers and shelf life [73]. In *Cucumis sativus* plants, Si NPs increased K uptake in different plant organs [74]. In tomato plants, SiO<sub>2</sub> NPs have a positive impact on the uptake and concentration of Fe in leaves, and Cu and Zn in roots even the uptake of macronutrients

in the roots and leaves also increases even when tomato plants are subjected to As<sup>V</sup> stress conditions [75]. These observed effects are due to the fact that SiO<sub>2</sub> NPs can modify nutrient uptake due to the roots can excrete organic acids and therefore can improve the absorption of mineral nutrients [76].

Moreover, the negative impact of high concentrations of SiO<sub>2</sub> NPs (500 and 2000 mg L<sup>-1</sup>) on the content of mineral nutrients, such as Cu and Mg, has been reported in cotton plants [30]. The addition of SiO<sub>2</sub> NPs in our study reduced S and Zn uptake in roots, but the rest of the nutrients were not impacted. Probably there was a regulation in the homeostasis of essential nutrients to tolerate the toxic effect of As.

#### 4.3. Impact of As and SiO<sub>2</sub> NPs on Plant Photosynthetic Pigments

In this study, the two forms of As did not affect chlorophyll content. A negative effect is commonly reported due to the fact that the As is toxic to organisms in the ecosystem, including plants. The degree of toxicity of As species follows the following order: (As<sup>III</sup>) > (As<sup>V</sup>) > (MMA: monomethylarsonous acid) > (DMA: dimethylarsinic acid) [77]. Nevertheless, our results are not the only ones where a null impact of As on plants is reported. For example, a high tolerance to As<sup>V</sup> has been reported in some plants such as Taray (*Tamaris gallica*), without showing a significant variation in chlorophyll synthesis [78]. Even the electron transport rate of the photosystems (PSII and PSI) of rice seedlings exposed to As<sup>V</sup> does not show significant effects [79]. As<sup>III</sup> in *Vitis vinifera* plants first reduces photosynthesis and, over time, stimulates it [80].

The processes involved in not generating a negative impact on chlorophylls or plant development may be due to As<sup>V</sup> being rapidly reduced to As<sup>III</sup> in plants. The efflux of As<sup>III</sup> to the external environment is a form to minimize the effect of As toxicity on plants [81]. In addition, As<sup>III</sup> forms complexes with phytochelatins (PC) within root cells and subsequently accumulates As<sup>III</sup>-PC complexes in vacuoles via *OsABCC1* transporters as a detoxification strategy [82].

In our research, the addition of SiO<sub>2</sub> NPs to plants subjected to high As concentration induced an increase in chlorophyll content. As<sup>III</sup>-Fe<sub>3</sub>O<sub>4</sub> NPs interaction has been reported to improve chlorophyll and carotenoid content in *Brassica juncea* plants in relation to exposure to As applied alone [83]. Under chromium exposure, Si NPs increase chlorophyll levels in *Pisum sativum* plants [53]. Even in banana plants (*Musa acuminata* 'Grand Nain') under abiotic stress, the addition of SiO<sub>2</sub> NPs improves the chlorophyll content [24].

The positive impact of the application of the NPs on photosynthetic pigments may be due to NPs having photocatalytic properties that allow them to induce a rapid oxidation-reduction reaction, which leads to charge transfer with the light-harvesting complexes that are linked to the photosystems (PSI and PSII) [84]. In this regard, the addition of Si NPs shows a maximized enhancement of PSII and of the electron transport rate in *Cymbopogon flexuosus* (Steud.), and the leaves appear more proficient at capturing and quenching energized photons [85]. Even the chlorophylls in the photosynthetic reaction center can bind to NPs, and due to the plasmon resonance of NPs, they can produce more excited electrons, thus forming a new hybrid system [25]. In addition, Si NPs initiate a cascade of morphophysiological adjustments that improve plant physiology by regulating the expression of many photosynthetic genes and proteins together with photosystem I (PSI) and PSII assemblies [86]. Therefore, it is possible that SiO<sub>2</sub> NPs may have modulated the expression level of photosynthesis-related genes in tomato plants.

#### 4.4. Impact of As and SiO<sub>2</sub> NPs on Antioxidant System

In our study, APX, CAT, and SOD enzymatic activities in roots were higher compared to activity in leaves. Similarly, a study with *Centella asiatica* plants subjected to Pb stress conditions showed higher activities of GPX, APX, and CAT in the roots than in the leaves [87]. The difference in enzyme activities between the leaf and the root may be due to the fact that the contaminant comes into natural contact with the radical system, and this system activates a rapid response as protection against oxidative stress [88].

Furthermore, in this study, enzymatic activity shows different responses due to exposure to the two forms of As in both roots and leaves. For example, APX enzymatic activity in leaves was the only one that increased by As exposure, while in roots, APX and CAT activity showed an increase. In this context, Duquesnoy et al. [89] reported that in *Zea mays* plants exposed to As<sup>V</sup> and As<sup>III</sup>, CAT activity increased in both leaves and roots. While SOD activity was reduced in *Hydrilla verticillata* by exposure to high doses of As<sup>V</sup> and As<sup>III</sup> [90]. Although in our study, the two forms of As did not influence the SOD activity in the two plant organs evaluated. As generates oxidative stresses in plants in the following two ways: (1) binding with a sulfhydryl group deactivates antioxidants, and (2) producing ROS in the oxidation-reduction process of their chemical forms [91]. The plant rapidly activates its antioxidant defense system with which it can counteract the oxidative stress generated by this metalloid [46].

In our study, SiO<sub>2</sub> NPs applied to As-stressed tomato plants showed an increase in APX and CAT activity, while GPX and SOD reduced their activity. In this context, Tripathi et al. [92] showed a significant enhancement of APX activity in *Zea mays* plants by the addition of Si NPs that had been inhibited by As. In addition, the application of ZnO NPs increased CAT activity in *Glycine max* plants supplemented with As<sup>III</sup> [6]. While the As<sup>V</sup>-Fe<sub>2</sub>O<sub>3</sub> NPs interaction in *Vigna radiata* plant roots decreased the activity of this enzyme compared to the addition of only As [93]. Interestingly, the high concentration of As in the irrigation water and its interaction with high concentrations of Si NPs may induce additional stress to tomato plants, which may be reflected in decreases in SOD and GPX activity [27]. Regarding the reduction of SOD activity, it may be due to the fact that this enzyme actively participates in the elimination of free radicals produced by stress in plants and is considered the first line of defense [94]. While GPX activity reduction may be due to an inhibition of glutathione production in the leaves, as shown in Figure 6D, since this compound is part of the substrate of this enzyme.

Furthermore, the effect of NPs on the antioxidant system of plants can be diverse. The interactions of the surface charges of the NPs cause the non-specific interaction of the cell membrane receptors and cell wall, which can lead to the following two responses: toxicity or biostimulation [95]. When biostimulation occurs, plants can accumulate higher concentrations of antioxidant compounds due to exposure to NPs. This increase will be reflected in the fruits as a redox state that NPs induce in response to stress [96].

As for non-enzymatic antioxidants such as flavonoids, glutathione, carotenoids, phenols, and vitamin C, act naturally against stress in plants [97]. In our study, non-enzymatic antioxidant compounds were lower in the roots than in the leaves. In addition, GSH, vitamin C, and flavonoids were increased by exposure to both forms of As in leaves, and phenols in roots were increased by As exposure. In this context, GSH is a relevant compound in heavy metal tolerance since it is the main substrate of non-protein thiols in most plant cells and can detoxify metalloids by phytochelatin in response to As toxicity [98]. In addition, As forms a complex with GSH and can be sequestered in the vacuole [99]. Therefore, the As deposited in the vacuole is probably inert. Likewise, it has been reported that in hyperaccumulator plants, arsenic exposure induces a higher flavonoid content [100]. As for as phenols, exposure of *Ulmus laevis* Pall to different chemical forms of As accumulation increases the accumulation of this important antioxidant [101]. This is because phenols act as protectants in response to high doses of toxic metals or metalloids [102]. In contrast to our results, it has been reported that in *Oryza sativa* seedlings the ascorbic acid content decreased by 33% and 51% with the exposition to 0.5 and 1 mM As<sup>V</sup> respectively [103].

In this study, the addition of SiO<sub>2</sub> NPs in the absence of both forms of arsenic increased vitamin C and flavonoid content. While when SiO<sub>2</sub> NPs are applied to tomato plants stressed with As<sup>V</sup>, only flavonoid content in leaves increases and glutathione content in roots decreases. The other compounds were not affected and in interaction with As<sup>III</sup> showed no effect. An increase in the content of phenols and flavonoids due to exposure to NPs has been reported in plants with abiotic stress [104]. Possibly, this increase is related to an upregulation of phenylpropanoid pathway genes [105]. Moreover, in lettuce seedlings



with exposure to NPs, it has been observed that an increase in glutathione and ascorbic acid, possibly as a result of an alteration in the ascorbate-glutathione (AsA-GSH) pathway [106]. Even so, the application of NPs shows a positive regulation in the expression of genes related to glutathione biosynthesis, glutathione reductase, and glutathione S-transferase in *Arabidopsis thaliana* [107]. The positive impact of the Si NPs on the antioxidant system may be due to the fact that the Si allows a lower entry of the metal into the plants [108]. In addition, SiO<sub>2</sub> NPs can maintain cell integrity by decreasing cellular ROS levels, increasing cell wall thickness, and increasing the proportion of As in pectin [26].

In this study, the total ABTS antioxidant capacity in roots showed an increase due to the interaction of As<sup>III</sup> + SiO<sub>2</sub> NPs, in leaves, it decreased due to exposure to As<sup>III</sup>, and in fruits, a lower capacity of As<sup>III</sup> was observed compared to As<sup>V</sup>. Antioxidant capacity is widely used as a parameter to characterize the ability to scavenge or neutralize free radicals [109]. Exposure to high doses of As<sup>V</sup> and SiO<sub>2</sub> NPs has been reported to increase antioxidant capacity in both hydrophilic and lipophilic compounds in tomato fruits and also H<sub>2</sub>O<sub>2</sub> levels [39]. NPs entering the plant can induce the generation of ROS because they cause an imbalance at the molecular, biochemical, and physiological levels that produces oxidative stress [110]. However, ROS induced by NPs can act as signalers in plants and may be able to alter secondary metabolism through their transcriptional reprogramming, causing a series of positive responses [111].

In our study, H<sub>2</sub>O<sub>2</sub> production increased in leaves in all treatments with respect to the control and decreased in fruits. In roots, the two forms of As applied alone and in interaction with SiO<sub>2</sub> NPs increased H<sub>2</sub>O<sub>2</sub> production. Contrary to our results, Tripathi et al. [92] reported that Si NPs can reduce H<sub>2</sub>O<sub>2</sub> production in *Oryza sativa* leaves exposed to As<sup>V</sup>. However, H<sub>2</sub>O<sub>2</sub> is also an important signaler in biological pathways, so that slight accumulations of H<sub>2</sub>O<sub>2</sub> can stimulate photosynthesis [112], and in our study, chlorophylls were stimulated as shown in Figure 4.

On the other hand, in fruits, H<sub>2</sub>O<sub>2</sub> decreased. This result was similar to a previous study, in which the concentration of 1000 mg L<sup>-1</sup> SiO<sub>2</sub> NPs decreased the production of H<sub>2</sub>O<sub>2</sub> in tomato fruits [39]. The decrease in H<sub>2</sub>O<sub>2</sub> in tomato fruits is probably due to a higher concentration of some antioxidant compounds in the fruit, as shown in Figure 7, and these compounds could counteract this free radical. In addition, both the natural ripening process, arsenic stress, and environmental changes can modify the production of ROS in tomato fruit.

#### 4.5. Impact of As and SiO<sub>2</sub> NPs on the Quality of Ripe Tomato Fruits

The antioxidant content of plants can be modified by both the stage of fruit development and environmental restrictions. The stage of fruit maturity at harvest is an essential factor in determining antioxidant levels in tomatoes [113]. In our study, both forms of arsenic decreased the firmness, glutathione, and hydrogen potential of tomato fruits. However, both As<sup>V</sup> and As<sup>III</sup> increased the content of total soluble solids, β-carotene, and vitamin C. As exposure can modify the nutraceutical quality, e.g., GSH, β-carotene, lycopene, phenols, and H<sub>2</sub>O<sub>2</sub> in tomato fruits of different cultivars [11], as a response to oxidative stress generated by As in the plant [114]. This is because As can accumulate and be distributed to tomato fruits, and the degree of entry of As into the fruit varies from one cultivar to another [115]. Even As causes ROS accumulation in tomato fruits, accumulation of proteins related to the response to oxidative stress and other proteins related to ubiquitination [116]. Alterations in quality attributes of tomato fruit due to the effect of a heavy metal are related to the expression of genes associated with sucrose biosynthesis (*SLSuSys*, *SILin5*, *SILin6*, *SILin7*) and carotenoids (*SIZDS*, *SICRTISO*, *SibLCY*) [117].

In this study, the application of SiO<sub>2</sub> NPs in the absence of As increases lycopene and vitamin C content. The increase in the concentrations of bioactive compounds by exposure to NPs will be reflected in the fruit as a redox state that NPs induce in response to oxidative stress [96]. Moreover, the NPs improve the quality of the fruit since they can significantly alter the proteins related to ascorbate and glutathione, exhibiting a greater amount of

antioxidants in the fruits [118]. Furthermore, the addition of NPs to tomato plants can increase fruit quality through transcriptional upregulation of the carotene isomerase gene (*CRTISO*), a gene involved in carotenoid biosynthesis [119]. While the nutritional quality of the fruits exposed to NPs can be negatively affected; however, this may depend on different factors such as type, shape, size, and applied concentration of NPs, as well as the crop [120].

Furthermore, in this study, the  $\text{As}^{\text{V}}$  +  $\text{SiO}_2$  NPs interaction showed a decrease in total soluble solids, hydrogen potential, and flavonoids, but increased phenols and vitamin C in ripe tomato fruits. While the  $\text{As}^{\text{III}}$  +  $\text{SiO}_2$  NPs interaction decreased glutathione content, firmness, and hydrogen potential, this interaction only increased vitamin C content. Interestingly, it has been shown that the  $\text{SiO}_2$  NPs under abiotic stress conditions improve the yield and quality of mango fruits [121]. Even plants growing under different arsenic concentrations and  $\text{SiO}_2$  NPs are applied, which have a positive effect on the modification of the generation processes of antioxidants in tomato fruit [39]. This may be due to the fact that Si improves the antioxidant compounds of fruits and improves commercial qualities such as firmness, this last attribute indicates that Si may have a key role in stabilizing the cell wall, protecting it from degrading enzymes such as polygalacturonase,  $\beta$ -galactosidase, and pectin methyl esterase [122].

In tomato fruits, there is still a lack of knowledge about redox metabolism. Future studies could generate a better understanding because there is a lack of information on the impact of nanoparticles on the nutraceutical quality of ripe fruits, as generally only an increase or decrease is reported for different doses and types of NPs. There is even a lack of information on the impact of As on fruit quality, as there are few studies that address this problematic on the quality of fruit that is ultimately consumed by humans.

## 5. Conclusions

This research shows that  $\text{As}^{\text{III}}$  has a greater negative impact than  $\text{As}^{\text{V}}$ . Both forms of As reduced shoot dry biomass production and firmness, and glutathione content in ripe fruit. However,  $\text{As}^{\text{III}}$  reduced fruit yield and decreased the concentration of Si, Mg, P, K, and Cu in the leaves of tomato plants.

The application of  $\text{SiO}_2$  NPs increased the content of chlorophyll. Moreover, it improves nutrient uptake and antioxidant content when plants were exposed to  $\text{As}^{\text{III}}$  and reduces  $\text{As}^{\text{V}}$  translocation, resulting in an adaptation of tomato plants to stress caused by this metalloid. Therefore, the use of  $\text{SiO}_2$  NPs is possible as an alternative to improve the productivity of tomato plants when the waters contain the presence of As and they are used for agricultural purposes.

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