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The Supply of Macro- and Microelements to Cotton Plants at Different Distances from a Fertilizer Production Factory

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Abstract: Environmental pollution from industrial factories via air deposition is an urgent problem worldwide. Phosphate fertilizers, derived from rock phosphate, are characterized by the presence of potentially toxic elements, such as Zn, Co, Pb, Ni, Cr, Mn, Fe, and Cu, which are dispersed in the form of solid dust-like materials from the pipes of the factory. This study aimed to investigate the effects of airborne industrial emissions on the chemical and biochemical compositions of cotton grown in the immediate vicinity of a fertilizer factory in Uzbekistan. The composition of airborne dust deposited on the plants, the chemical composition of the cotton leaves before and after washing, as well as that of above- and below-ground plant organs, and their protein contents were determined. The concentrations of macro- and microelements in the leaves and roots were determined using an atomic absorption spectrophotometer. The fluorine contents in the leaves and in the roots were determined using a fluorine-selective electrode. The radius of dispersion of industrial emissions in the air was best described by measuring the fluorine contents in washed and unwashed cotton leaves. The relationships among P, K, Mg, Ca, S, F, and Mn in plant roots and leaves as a function of distance from the pollutant source were analyzed. Based on the fluorine contents in washed and unwashed cotton leaves, the two following zones of technogenic pollution were distinguished: the zone < 5 km from the factory, with high technogenic pollution, and the zone > 5 km from the factory, with moderate technogenic pollution. It was found that the resistance of cotton to air pollution from industrial emissions is determined by the ability of cotton plants to neutralize toxic compounds by increasing the influx of alkaline earth metals into the affected tissues. This study showed the possibility of growing cotton at a distance of >5 km from the fertilizer factory. It is strongly recommended to analyze the chemical composition of plants located in a highly polluted zone only after the dust particles have been washed off of the plant's surface. Despite the resilience of cotton to industrial pollution, the monitoring of areas identified as pollution zones is recommended.

Keywords: cotton; industrial pollution; fluorine; sulfur; plant tissues; microelements; alkali earth metals

1. Introduction

Air pollution of potentially toxic elements (PTEs) from industrial emissions is a serious environmental problem [1–4]. The range of PTE transmission is determined by various factors, among which climatic conditions (e.g., temperature, humidity, and wind speed), the composition of emissions (the ratio of solid, liquid, and gaseous components), the particle size, the height of pipes, the affected terrain, vegetation cover, etc. should be emphasized [5–7]. Of all emission components, gaseous particles spread further than solid particles. The problem of air pollution from industrial emissions is exacerbated in arid regions [1,5,8]. The pollution level peaks with increasing distance from the emission source and then gradually decreases [9,10]. Dry deposition accounts for up to 50% of pollutants in the atmosphere [11], or 63% of PM10 removal [12].

Some microelements are essential for plant metabolism in low concentrations. However, they become toxic to living organisms when their concentration exceeds a certain threshold [13]. Many studies have shown that the effect of pollutants on plants depends on their concentrations in the atmosphere and the duration of exposure [14–24]. It is generally recognized that the damage first manifests itself at the biochemical level (impairment of photosynthesis, respiration, biosynthesis of lipids, proteins, etc.) and then spreads to the ultrastructural (disorganization of cell membranes) and cellular (destruction of the nucleus, cell walls, and mesophyll) levels. A variety of effects at the biochemical and physiological levels caused by volatile toxins lead to the development of visible symptoms of damage (chlorosis and necrosis of leaf tissue), slowed plant growth, and reduced crop yields.

Although numerous studies have been published, to date, on the effects of man-made air pollution on soils, plants, water, the environment, and human and animal health, there are few studies on the effects of industrial emissions on the growth and development of cotton plants. Cotton (*Gossypium hirsutum* L.) is a natural plant fiber of great economic importance that is grown in more than 50 countries [25]. Its production provides incomes for more than 250 million people worldwide; 87% of the world's cotton-growing areas are located in developing countries [26]. In addition, around 65% of conventional cotton products enter the food chain through edible oils or indirectly, through the consumption of meat and milk from animals fed on cotton meal and ginning by-products [27]. Although cotton is considered to be one of the most polluting crops, due to the use of agrochemicals and water consumption [28–30], it is also very sensitive to air pollution. The cotton plants grown in smog free, carbon-filtered air produced 20–30% more raw cotton compared to similar cotton grown in unfiltered air [31].

Cotton is an important crop in Uzbekistan, accounting for 17% of the country's exports in 2006. With an annual cotton production of about 1 million tonnes of fiber (4–5% of world production) and exports of 700,000–800,000 tonnes (10% of world exports) [32], cotton has been cultivated in Uzbekistan for more than 2500 years.

There are numerous studies on cotton cultivation in terms of its environmental impact (the use of pesticides, water consumption, and soil degradation), agronomic approaches, the phenological and physiological aspects of cotton growth and development, cotton cultivation during climate change, sustainable cotton production and organic cotton, the effects of air pollution on cotton fabric, cotton breeding and genetics, cotton yield and quality, the utilization of cotton, the cotton market, etc. [26,29,33–37]. However, no published studies were found on the effects of technogenic air pollution on the biochemical composition of cotton grown in the immediate vicinity of a nitrogen–phosphate fertilizer factory. Phosphate fertilizers, produced from rock phosphate, are characterized by the presence of a large number of impurities, containing Zn, Co, Cd, Pb, Ni, Cr, Mn, Fe, and Cu, which are stirred up as parts of solid powdery materials from the factory pipes.

The aim of this study was to investigate the chemical and biochemical compositions of cotton at different levels of pollution from a mineral fertilizer production factory in Uzbekistan. The research objectives were as follows:

- To investigate the composition of technogenic dust from the pipes of the factory;
- To determine the ash contents in washed and in unwashed cotton leaves;

- To determine the chemical compositions of the above- and below-ground organs of cotton at different distances from the pollutant source, and to determine the protein content in the leaves of the plants;
- To develop empirical models describing the relationships among the concentrations of P, K, Mg, Ca, S, Mn, and F in the roots and leaves of cotton plants;
- To identify zones of technogenic stress.

2. Materials and Methods

2.1. Site Description

The factory for the production of ammophos (39°39′3″ N and 66°49′42″ E), with annual amounts of gas and dust emissions over 10 thousand tons, is located in a dry subtropic zone. There are three workshops for the production of concentrated sulfuric acid, two workshops for the production of extractive phosphoric acid, and three workshops for the production of ammophos on the industrial site. The height of the pipes in the production hall for sulfuric acid is 180 m, while the height of those in the production halls for ammophos is 120 m. The composition of the precipitates is determined by the technological cycle and consists of sulfuric acid, sulfurous anhydride, fluorine compounds, and solid pulverized materials.

The area of active cotton production borders the factory. The cotton was sown on 12–15 April. The harvest was carried out during the budding phase. The cotton variety used was Tashkent 6. The amount of precipitation in the study year was 330 mm/year, well below the long-term average (March–May, 46% of the annual amount; July–September, 2% of the total amount; October–November, 12% of the annual amount). The cotton-growing season (mid-April to October) is characterized by extreme drought, with rainfall between 0.3 and 5 mm per month. Such an amount is not able to have a significant effect on the plants. Selective measurements for fluorine and sulfur contents, carried out during the period with the highest precipitation (November–March), have shown that their concentrations can reach the following levels: F, 0.41 mg/L; SO_3, 64 mg/L. During the cotton-growing season, 7 irrigations were carried out. Cotton was irrigated in furrows according to a 2-4-1 scheme (i.e., 2 irrigations before flowering, 4 irrigations during flowering, and 1 irrigation during full maturity). The recommended single-irrigation rate for soils with heavy granulometric composition is 1100–1200 m^3 per hectare. The degree of cotton canopy closure by July is 40–93%, depending on the cultivation technique [33].

The soils in the study areas are old irrigated soils, or Irragric Anthrosols, according to World Reference Base classification [38]. The soils are characterized by a similar chemical and granulometric composition at each sampling point. The gross chemical composition is as follows: SiO_2, 52–54.1%; Fe_2O_3, 3.4–3.8%; Al_2O_3, 11.6–12.3%; and K_2O, 2.3–2.9%. The granulometric composition is heavy loam. The contents of the fractions are <0.001 mm: 17.5–19.2% and <0.01 mm: 48.4–50.4%. The reaction of the soil is alkaline, with pH (H_2O) 7.7–8.2. The agrotechnical measures applied for cotton cultivation are similar.

2.2. Sampling

Sampling of technogenic dust was carried out directly from the pipes for the production of ammophos and the extraction of phosphoric acid in ashless filters. Leaves and roots of cotton plants were sampled at key sites of 50 × 50 m, on typical relief elements at different distances (0.6–20 km) from the factory, in the direction of the prevailing winds (northeast), at the budding stage, and over the entire height of the stems. The average sample comprised 90 plants from each array. The exact geographical location of each sampling point is shown in Table 1.

2.3. Analyses

The acidity of the soil (pHKCl) was determined potentiometrically after the soil was shaken in a 1N KCl solution. The particle size distribution was determined using the density sedimentation method [39]. Both washed and unwashed leaves were incinerated in a muffle furnace at 500 °C for 8 h. The resulting ash was dissolved in nitric acid to determine the contents of macro- and microelements. The concentrations of metals in the leaves and roots were determined using an atomic absorption spectrophotometer (Perkin Elmer, Shelton, CT, USA). The fluorine contents in the leaves and in the roots were determined using a fluoroselective electrode (Thermo scientific, Dublin, Ireland).

Table 1. Locations of the sampling points at eight different distances from the fertilizer factory.

Distance from the Factory, km	Latitude	Longitude
0.6	39°40′24.1″	66°48′32.7″
3.0	39°41′31.6″	66°47′43.7″
5.0	39°42′20.3″	66°46′48.5″
7.5	39°43′30.5″	66°45′55.9″
10.0	39°44′28.0″	66°44′34.4″
12.5	39°45′38.0″	66°43′44.0″
15.0	39°46′40.7″	66°42′35.3″
20.0	39°48′55.9″	66°40′34.2″

The levels of dust content on the cotton leaf surfaces were determined through the difference between the ash contents of the unwashed and the washed leaves. The dust deposited on the leaf surface was collected by washing 100 g of fresh leaves in 100 mL of distilled water. The pH value was determined in the filtrate. The completeness of the washing was checked by washing the leaves with a large volume of deionized water. The filters were boiled in a mixture of nitric and hydrochloric acids. The dust washed off of the leaves was decomposed in a similar way.

2.4. Data Analyses

Data processing was performed according to [40]. When assessing the statistical significance of the relationships between biogenic elements and protein, the threshold value was chosen as p-value = 0.05, with the number of observations as $n = 7$. For the washed and unwashed leaf treatments, the initial data were first logarithmized, and then an empirical model, based on a second-degree polynomial, was created for the second zone (from a distance of 5 km). An exact empirical model was then constructed for the first zone, also based on a second-degree polynomial (with three unknown parameters), so that the constructed models for the first and second zones matched at the boundary (at the 5 km point).

3. Results

3.1. Compositions of Cotton Leaves and Roots

Quantitatively, the dust content can be characterized by the difference between the ash contents of unwashed and washed leaves. When unwashed leaves were incinerated, the ash yield was 1.1–4.3 g/100 g dry weight higher (5.3–25.6%) than that for the washed leaves (Table 2). The maximum contamination of the leaves was found in the 5 km zone. The phosphorus concentration in the leaves after washing was 3.45–4.8 times lower (Table 3) than that without washing. The nature of the variability of phosphorus content in plant organs was generally the same (Table 4). Empirical models for the phosphorus contents in roots and leaves are shown in Table 5. The phosphorus contents in the plant organs decreased slightly with increasing distance from the factory.

Table 2. Ash content of plant leaves, g/100 g air-dried weight.

Distance from the Factory, km	Ash Content of Unwashed Leaves	Ash Content of Washed Leaves	Difference	
			g	%
0.6	21.1 a	19.1 a	2.0	9.5
3	21.4 a	17.1 a	4.3	20.1
5	23.5 a	21.2 ab	2.3	9.8

Table 2. *Cont.*

Distance from the Factory, km	Ash Content of Unwashed Leaves	Ash Content of Washed Leaves	Difference	
			g	%
10	22.6 a	21.0 ab	1.6	7.1
15	21.7 a	20.6 a	1.1	5.1
20	20.9 a	19.2 a	1.7	8.1

The different letters signify statistically significant differences within a column, at $p < 0.05$.

Table 3. The chemical compositions of cotton leaves, %.

Distance from the Factory, km	Mg	P	S	K	Ca	Protein
0.6	1.08 0.66 a	0.57 0.12 a	1.37 1.30 a	1.37 1.10 a	ND 5.65 a	10.6 a
3	0.67 a	0.11 a	1.29 a	1.40 a	6.05 a	10.3 a
7.5	0.61 a	0.08 b	1.04 b	1.51 ba	5.21 ab	12.0 a
10	0.59 0.60 a	0.37 0.11 a	0.94 1.16 b	1.83 1.63 b	ND 5.02 b	12.9 ab
12.5	0.53 b	0.16 a	0.91 b	1.56 ba	5.01 b	12.9 ab
15	0.46 0.59 ab	0.40 0.12 a	1.00 1.20 ab	1.53 1.68 b	ND 5.07 b	14.1 b
20	0.49 b	0.09 b	0.84 b	1.68 b	4.52 b	14.2 b
Control	0.48 b	0.08 b	0.83 b	1.61 b	4.69 b	

Note: for the distances 0.6, 10, and 15 km, the number above the line is the concentration of the element in unwashed leaves; that below the line is the concentration in washed leaves; in all other treatments, the determination was carried out only in washed leaves. The different letters signify statistically significant differences within a column, at $p < 0.05$. ND—not determined.

Table 4. The chemical compositions of cotton roots, %.

Distance from the Factory, km	Mg	P	S	K	Ca
0.6	0.46 a	0.18 a	0.09 a	1.64 a	0.77 a
3	0.40 a	0.16 a	0.11 a	1.58 a	1.15 b
7.5	0.31 b	0.17 a	0.11 a	1.41 b	0.58 a
10	0.30 b	0.19 a	0.10 a	1.45 b	0.68 a
12.5	0.33 b	0.18 a	0.11 a	1.42 b	0.69 a
15	0.37 ab	0.16 a	0.10 a	1.45 b	0.71 a
20	0.28 b	0.11 b	0.06 ba	1.39 b	0.51 a
Control	0.27 b	0.13 b	0.10 a	1.30 b	0.60 a

Note: The different letters signify statistically significant differences within a column, at $p < 0.05$.

Table 5. Empirical models of the dependence of plant composition on the distance from a pollutant source.

Model	*p*-Value	R^2	v
(1) P roots $y_1 = 0.19 - 0.0024 \cdot x$	0.14	0.38	−0.0024
(2) P leaves $y_2 = 0.114 - 0.00015 \cdot x$	0.9	0.0014	−0.00015
(3) P roots/leaves $y_3 = 0.112 + 0.46 \cdot y_{2.2}$	0.31	0.2	0.46
(4) F roots $\ln(y_1) = 3.877 - 0.95 \cdot x + 0.103 \cdot x^2 - 0.0044 \cdot x^3 + 0.0000645 \cdot x^4$	0.008	0.945	–
(8) Mn leaves $y_8 = \exp(4.86 - 0.466 \cdot x + 0.075 \cdot x^2 - 0.0045 \cdot x^3 + 0.000091 \cdot x^4)$	0.02	0.9999	–
(9) S roots $y_9 = 0.11 - 0.0014 \cdot x$	0.23	0.27	−0.0014

Table 5. *Cont.*

Model	p-Value	R^2	v
(10) S leaves $y_{10} = 1.31 - 0.02 \cdot x$	0.04	0.58	−0.02
(11) S roots/leaves $y_{11} = 0.05 + 0.04 \cdot y_{3.2}$	0.36	0.16	0.04
(12) Mg roots $y_{12} = 0.42 - 0.007 \cdot x$	0.053	0.558	−0.007
(13) Mg leaves $y_{13} = 0.68 - 0.0088 \cdot x$	0.0038	0.837	−0.0088
(14) Mg roots/leaves $y_{14} = -0.085 + 0.73 \cdot y_{1.2}$	0.05	0.559	0.73
(15) Ca roots $y_{15} = 0.92 - 0.019 \cdot x$	0.12	0.41	−0.019
(16) Ca leaves $y_{16} = 5.86 - 0.066 \cdot x$	0.0056	0.81	−0.66
(17) Ca roots/leaves $y_{17} = -1.2 + 0.369 \cdot y_{5.2}$	0.007	0.79	0.369
(18) K roots $y_{18} = 1.59 - 0.012 \cdot x$	0.017	0.71	−0.012
(19) K leaves $y_{19} = 1.25 + 0.027 \cdot x$	0.01	0.76	0.027
(20) K roots/leaves $y_{20} = 2.09 - 0.41 \cdot y_{4.2}$	0.008	0.781	−0.41
(21) leave protein $y_{21} = 10.27 + 0.22 \cdot x$	0.0006	0.92	0.22

3.2. Contents of Fluorine in Cotton Leaves and in Roots

The concentrations of fluorine in the roots and in the washed and unwashed leaves of the cotton are shown in Table 6. Most of the fluorine was easily removed by washing the leaf surface. The degree of contamination of the plant with fluorine, in general, naturally decreased with distance from the source of pollution.

Table 6. Fluorine concentrations in the roots and in washed and unwashed leaves of cotton, mg/kg air-dried weight of the plants.

Distance from the Factory, km	Roots	Leaves	
		Without Washing	With Washing
0.6	26.5 a	70.0 a	17.5 a
3	7.0 b	300.0 b	72.0 b
5	3.5 c	50.0 a	12.0 a
7.5	2.4 c	40.0 a	12.0 a
10	2.25 c	23.0 ac	7.0 ac
12.5	2.65 c	20.0 c	5.2 c
15	5.6 bc	21.0 c	4.8 c
20	3.6 c	1805 d	4.2 c
25	4.2 bc	12.2 c	4.1 c

Note: different letters signify statistically significant differences within a column, at $p < 0.05$.

The dependence of the fluorine content in the roots on the distance from the factory was not significantly linear (Table 5). There was a rapid decrease in the fluorine content, after which the fluorine content stabilized at a distance of 5 km. There was a very strong correlation between the fluorine contents in unwashed and washed leaves (r = 0.999).

However, there was no significant relationship between the fluorine contents in the roots and the washed leaves (r = 0.178). The dependence of the fluorine content in the roots was statistically significant at a very high level (Model (4), Table 5).

3.3. Microelements

The contents of microelements in the dust deposited on the leaf surfaces are shown in Figure 1. The concentrations of microelements in solid particles on the leaf surfaces are given in Table 7. The concentrations of microelements in the washed cotton leaves and their concentrations in the technogenic dust differed greatly (Figure 1; Table 7). The concentrations of most microelements in the tissues of the washed leaves did not differ significantly at different distances from the pollution source (except for Mn) (Table 7). However, the contents of microelements in the leaves were significantly lower than those in the technogenic dust washed from these plants. The sulfur concentrations in the individual organs of the cotton are given in Table 4. Plants within 3 km of the factory experienced maximum inhibition. Visual observations of the plants showed signs of chronic damage to the cotton by sulfur dioxide. The sulfur content in the cotton leaves exceeded its content in the roots by 8.5–15.2 times. Empirical models for the sulfur contents in roots and leaves are given in Table 5. Model (9), which describes the change in the sulfur content in the plant roots depending on the distance from the pollutant, is not statistically significant.

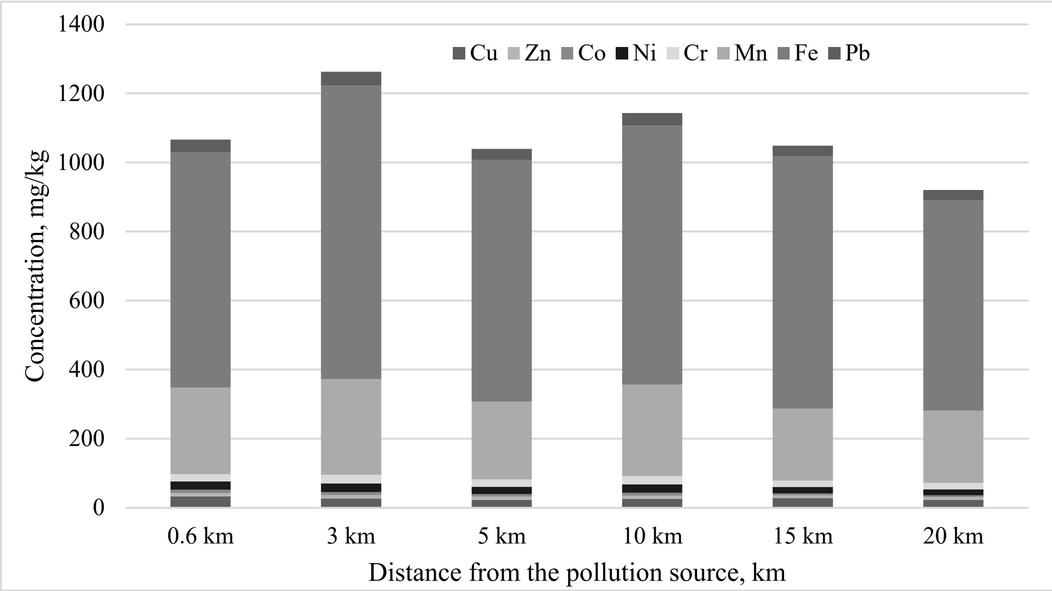

Figure 1. The relative distribution of potentially toxic microelements in washed cotton leaves at different distances from the factory, mg/kg dry weight of leaves.

Table 7. The contents of potentially toxic microelements in washed cotton leaves at different distances from the factory, mg/kg of dry weight of plants.

Element	Distance from the Factory, km					
	0.6	**3**	**5**	**10**	**15**	**20**
Zn	22.0 a	16.2 a	16.2 a	15.0 a	21.8 a	21.8 a
Co	2.0 a	2.0 a	2.0 a	1.8 a	1.9 a	1.8 a
Pb	10.2 a	10.5 a	10.0 a	10.5 a	11.0 a	10.2 a
Ni	3.7 a	3.6 a	3.5 a	3.6 a	3.7 a	3.3 a
Cr	1.3 a	1.2 a	1.5 a	1.6 a	1.4 a	1.2 a
Mn	101.0 a	56.0 b	50.0 b	61.0 b	63.5 b	57.5 b
Fe	135.0 b	140.0 b	165.0 a	150.0 ab	140.0 b	100.0 c
Cu	8.4 a	5.4 b	5.2 b	5.8 b	7.4 ab	7.4 ab

Note: different letters signify statistically significant differences within a row, at $p < 0.05$.

Furthermore, there were no significant changes, on average, over the entire measurement interval. On the contrary, Empirical Model (10), describing the change in the concentration of sulfur in the cotton leaves with distance from the factory, is statistically significant (Table 5). No significant relationship was found between the sulfur contents in the roots and the leaves (Model (11)).

3.4. Alkali and Alkaline Earth Metals

The concentrations of Mg, Ca, and K in cotton roots and leaves changed with distance from the pollution source. (Tables 3 and 4). Plants grown in the immediate vicinity of the factory showed the highest Mg accumulation. With increasing distance from the emission source, the Mg concentrations in the individual plant organs decreased. The Mg content in the roots of plants grown directly on the factory site was 0.46%, and at a distance of 20 km, it was 0.28%; i.e., the concentration decreased 1.64-fold. In the leaves, content fluctuations of 0.66 to 0.49% were found. The difference was 1.35-fold.

The statistical significance of the model describing the relationship between the Mg content in the roots (Model (12)) (Table 5) and the distance from the pollution source is high, and in the leaves (Model (13)), it is very high. There is also a strong statistically significant relationship between the concentrations of Mg in the aboveground and belowground organs of the plants (Model (14)).

The Ca contents in the aboveground and belowground organs of cotton are also very sensitive to the location of the pollution source. The nature of the variability of the Ca contents in the roots and leaves of cotton are the same. The closer to the factory, the higher the Ca concentrations in the studied plant organs (Tables 3 and 4).

Depending on the distance from the factory, Empirical Models (15) and (16) (Table 5) of the Ca contents in the individual plant organs are extremely comparable. In addition, there is a high statistically significant correlation (Model (17), Table 5) between the Ca contents of cotton roots and leaves. The highest concentration of K in the cotton root system was found in the immediate vicinity of the factory. At the same time, the leaves selected at the same location had a very low K content. The K concentration in the leaves increased, while the concentration in the roots decreased, with increasing distance from the pollution source. Empirical models with very high statistical significance relate the potassium contents in the roots (Model (18)) and the leaves (Model (19)) to the distance from the factory. The K contents in the roots and leaves were significantly negatively correlated ($r = -0.88$). There was also a strong negative correlation between the K and Ca concentrations in the leaves ($r = -0.73$), and between K and Mg concentrations ($r = -0.7$).

The suppression of protein synthesis was observed in all plants in the zone of technogenic impact (Table 3). The lowest protein content was observed in cotton plants grown at a distance < 3 km from the pollutant, at 10.3–10.6%. This corresponds to about 70% of the protein content in the control plants. At a distance of 7.5–15 km from the pollutant, the decrease in protein content was 15–20%. At a distance of 15–20 km, the effects of factory emissions on the biochemical processes in the plants weakened. The decrease in protein content in leaves was 6–7% of the control level. Empirical Model (21), which describes the protein content of cotton as a function of the distance from the pollutant, has a very high statistical significance (Table 5). There is a statistically significant correlation between the potassium and protein contents in the leaves ($r = 0.86$).

3.5. Cotton Yield as Affected by the Distance from the Pollutant

Visual observations showed signs of chronic damage to cotton by sulfur dioxide, with maximum inhibition of plants within a radius of 3 km from the factory. The data from the biometric observations are shown in Table 8. The increase in yield was only evident at a distance of 5 km from the emission source.

Table 8. Average plant height and yield of raw cotton at different distances from the factory.

Distance from the Factory, km	Average Plant Height, cm	Productivity, t/ha
0.6	63.7	2.0
3	64.8	2.1
5	70.1	2.8
10	79.3	2.6
15	89.1	2.6
20	75.5	2.7

4. Discussion

4.1. Contents of Phosphorus in Cotton Leaves and in Roots

The technogenic dust on leaf surfaces was represented by particles of ammophos pulp, with varying degrees of dispersion. Particles of phosphate raw materials dominated the emissions from the phosphoric acid production shops. The deeply dissected leaf blades of cotton plants hold dusty sediment well. The little precipitation from May to October virtually eliminates the possibility of technogenic particles washing off of the leaf surfaces. In addition, a significant decrease in P concentration on the leaves after washing indicates the aerial dispersion of phosphorus-containing particles, which are fixed on the leaf blades (Table 3). Although there was a weak correlation between the P content in the roots and the distance from the factory, a decreasing trend was observed with the distance from the pollution source (Tables 4 and 5).

The negative effect of technogenic dust on cotton plants is associated with a reduction in the influx of solar energy to the photosynthetic cells, the clogging stomata, and the chemical processes caused by acidic dust components (the pH of ammophos pulp is 3.85), as well as the transfer of fluorine in the composition of technogenic dust directly into the leaf tissue [41].

4.2. Contents of Fluorine in Cotton Leaves and in Roots

Technogenic dust from the factory contains fluorine and heavy metal impurities. The content of fluorine in the ammophos pulp is 3.7%, and in the phosphate rock, it is 2.1%, indicating that fluorine was deposited on the cotton leaves as part of the technogenic dust. The measurements of fluorine contents in washed and unwashed cotton leaves proved to be the most informative for determining the radius of technogenic impact, and allowed us to distinguish the two following zones of technogenic impact on the F contents in cotton leaves and roots: the first zone is located at a distance < 5 km; the second is located at a distance > 5 km from the factory (Table 6).

Data processing revealed strong correlations between the fluorine contents of the washed and unwashed leaves. However, the lack of a significant correlation in the fluorine contents between roots and washed leaves could mean that fluorine is mainly concentrated in the organs in direct contact with its source, as follows: in the leaves, when it originates from the technogenic dust deposited on the leaf surface; in the roots, when it penetrates from the soil through the root system.

The aerial deposition on the leaf surface did not take place as passive "storage". There was a relationship between the content of fluorine on the leaves, in the composition of technogenic dust, and its concentration in leaf tissues. On average, $\frac{1}{4}$ of fluorine in the unwashed leaves was concentrated in the leaf tissues. Some of this amount can pass into leaf mesophyll from technogenic dust [21]. The bioaccumulation of fluoride in different organs of cotton varies, depending on its transfer from the soil solution to the roots and its translocation from the root to the shoot [16,18,21,24].

In our experiments, the pH values of the washings ranged from 9.15 to 9.25. Generally, in neutral and alkaline pH values, fluoride is readily bound to the soil surface and is not available to plants [42]. However, contamination with fluoride causes a decrease in chlorophyll, thus affecting the germination, growth, and physiological parameters of cotton [41].

4.3. Microelements

The compositions of the solid particles on leaf surfaces showed that the concentrations of metals were not high, and they decrease as follows: Fe > Mn > Pb > Cu > Ni > Cr > Zn > Co (Table 6). For most metals, there is no correlation between the content in plants and the distance from the factory. To determine the degree of metal toxicity for cotton, we used the scale of normal concentrations of metals in plants given in [13]. A comparison of our data with the scale showed that the concentrations of metals are within the range of natural variation in uncontaminated plants and pose no risk to the growth and development of cotton. The exception is manganese, the content of which, in the leaves of plants selected in the immediate vicinity of the factory (3 km), was 1.6–2.0 times higher than in remote areas. Increasing the influx of manganese in cotton leaves, which participates in the photosynthesis processes under conditions of strong technogenic stress, is aimed at maintaining the required amount of its physiologically active compounds in functional cell structures, the number of which decreases when the affected areas of the leaf surface die off [43]. The results of the contents of metals in the plant tissues did not allow us to draw an unambiguous conclusion about the presence or absence of foliar uptake of metals in cotton. However, even in the presence of such mechanisms, the role of foliar absorption is hardly significant, which is associated with the anatomical structure of cotton leaves (due to a well-developed cuticular layer) [44], and the lack of precipitation during the cotton-growing season.

4.3.1. Manganese

Manganese is an essential microelement for cotton. Foliar exposure of Mn nanoparticles can improve Mn contents in the shoots and the grain [45]. It can be assumed that the increase in the influx of Mn, which is involved in the processes of photosynthesis under strong technogenic impact, is aimed at maintaining the required amount of its physiologically active compounds in functional cellular structures, the number of which decreases when the affected areas of the leaf surface die off. The dependence of the Mn content in leaves on the distance from the emission source is essentially non-linear and is precisely described using Empirical Model (8) (Table 5). The statistical significance and coefficient of determination of the model are very high. Similarly, increases in the contents of manganese in the leaves of trees and in those of shrubs in technogenic zones were also found [14].

4.3.2. Sulfur

Sulfur is the most important pollutant emitted from the factory. Because sulfur dioxide is twice as heavy as air, it is concentrated in places of accumulation in industrial factories. Sulfur and sulfur-containing compounds act as signaling molecules in stress management, as well as during normal metabolic processes [46].

The content of sulfur in leaves exceeded its concentration in the roots by 8.5–15.2 times, indicating an aerial pathway of cotton pollution, in which gaseous sulfur compounds (SO_2 and H_2S) penetrate through the stomata and are adsorbed into leaf tissues [47]. Empirical data processing confirms the aerial route of sulfur entry into the leaves, as well as its deposition in those organs that are in direct contact with the source of the pollution.

According to [20], the entry of sulfur compounds through the stomata proceeds according to a barrier-free type, and the degree of accumulation depends on the concentration in the air. Higher plants have the ability to maintain intracellular pH. The role of these buffering mechanisms in the removal of H^+ ions produced following SO_2 uptake has not been studied to date [48]. Important roles in the absorption of sulfur dioxide are played by humidity, wind speed, light, and air temperature. With the distance from the source of emissions, the pollution of cotton leaves with sulfur decreased.

4.4. Alkali and Alkaline Earth Metals

Undoubtedly, the contents of magnesium in the roots and in the leaves of cotton depend very much on the distance from the factory. This is supported by the high degree of significance of the model describing the relationship between the accumulations of Ca and Mg in roots and leaves and the distance from the pollution source. Both cations showed similar patterns of accumulation in plants' above- and belowground tissues, depending on the distance from the pollution source. The opposite pattern of accumulation of K in the roots and leaves implies that the technogenic impact causes a violation in the supply of K to the photosynthetic organs. Apparently, this influence is indirect. A strong negative relationship between concentrations of K and Ca in the leaves assumes that the active transport of alkaline earth metals into the affected leaves causes a decrease in the influx of K. Cotton is a crop sensitive to potassium deficit [49], but different organs have different requirements for K, in the following order: fruits > stems > roots > leaves [50]. Each ton of raw cotton, uptakes 40–60 kg of potassium [51]. Potassium deficiency in plants affects carbohydrate metabolism [52], delays protein synthesis, and inhibits the formation of complex sugars [53].

It is considered that the damage to plants by acid gases is closely related to their saturation with free calcium, magnesium, potassium, and sodium cations. Plants that have adapted their phylogenesis to growing in alkaline soils are potentially the most gas-resistant [22,44]. The increased consumption of Ca and Mg by cotton in the zone of industrial airborne emissions is associated with the important role of alkaline earth metals in the detoxification of the SO_2 and H_2S transformation products, as well as other gases [20,48]. In response to the influx of SO_2 and H_2S in sub-lethal doses, the influx of mineral cations into the leaves increases, and their absorption from the soil increases as well. There are statistically significant correlations between Mg and S ($r = 0.93$) and Ca and S ($r = 0.82$) in cotton leaves. This implies that the gas resistance of the cotton is based on its ability to activate its metabolic processes. This allows for increasing the concentrations of necessary metabolites in tissues in response to unfavorable growth conditions. Sulfur and calcium play similar biochemical roles, since they simultaneously regulate photosynthetic activity in plants [54]. Ref. [55] showed that the heavy metal uptakes in leaves and roots were higher for sulfur applied than for non-sulfur applied treatments, while those in grains, husks, and stalks were lower.

4.5. Cotton Yield Affected by the Distance from the Pollutant

Various effects at the biochemical and physiological levels, caused by volatile toxic substances, lead to the appearance of visible symptoms of damage (e.g., chlorosis and necrosis of leaf tissues), slowed growth, and reduced crop yields. The biometric measurements indicated that factory emissions negatively affected plant height and productivity. The decrease in K content in leaves in the zone of high technogenic impact can explain the decrease in cotton productivity, identified within a radius of 3 km from the source of pollution.

Despite the negative impacts of emissions, cotton has been shown to be highly resistant to pollution, due to its anatomical and biological characteristics [20,43], such as a deeply penetrating root system, covering a large volume of soil, a lignified stem, a dense cuticular layer on the leaves, and certain protective mechanisms. This should include the regulatory function of absorption, which allows for increasing the concentrations of necessary metabolites in tissues in response to unfavorable growth conditions. These characteristics of cotton make it possible to grow cotton near the fertilizer factory (3 km from the pollutant).

4.6. Impact Zones of the Factory as an Emission Source of Pollutants

Usually, the zone of technogenic pollution extends largely from the factory in the direction of the prevailing winds [10,15]. With this work having been carried out in the direction of intense pollution, the distances characterize the maximum lengths of the corresponding zones. The fluorine contents of washed and unwashed cotton leaves allowed us to distinguish the two following zones of technogenic impact: the first zone was located at a distance < 5 km; the second was at a distance > 5 km from the factory.

4.6.1. Zone of Strong Technogenic Impact (0–3 km from the Factory)

Cotton grown in this zone was characterized by high dust contents on the leaf surfaces. The total amounts of fluorine in the leaves and on the leaf blades reached 300 mg/kg. Visually, there were signs of chronic sulfur dioxide damage to the cotton, and of apical and marginal necrosis on individual plants. There was a suppression of growth processes, protein synthesis (protein losses in leaves were 30% of the control level), and a decrease in yield. The negative impact of technogenic dust deposited on plants is associated with a reduction in the flow of solar energy to photosynthetic cells, the blockage of the stomata, and the chemical processes caused by acidic components of the dust. However, the transfer of chemical elements from the surface into the leaf tissue is unlikely, partly due to the lack of precipitation during the growing season, and partly due to a dense cuticular layer protecting the leaf surface. For this study, the effect of sulfur dioxide on cotton was the most harmful. This is due to its high toxicity, its systematic release into the atmosphere, and its ability to penetrate leaf tissue.

4.6.2. Zone of Moderate Technogenic Impact (5–15 km from the Factory)

In this zone, the dust contents of plants were much lower, and the fluorine content of the unwashed leaves did not exceed 50 mg/kg dry weight. Leaf damage caused by sulfur dioxide was recorded as separate spots. The suppression of protein synthesis in the leaves was less pronounced, and the losses were 15–20% of the control level. The yield of raw cotton was at the control level.

4.6.3. Zone of Low Technogenic Impact (15–20 km from the Factory)

It was difficult to establish the boundaries of this zone, since the smallest particles of dust and gases are transported beyond 25 km. However, their effect on cotton is insignificant, although fluorine was present in the composition of dust on leaf blades (up to 20 mg/kg). Visual signs of leaf damage from sulfur dioxide were rare and were only established after careful examination. Protein losses in the leaves were 6–7% of the control level.

5. Conclusions

This study allowed us to make the following conclusions:

- In the impact zone of the factory, the chemical and biochemical compositions of cotton plants change;
- The fluorine concentrations made it possible to precisely delimit the radius of the most intense atmospheric dispersion of emissions from the pipes of the fertilizer factory. Two impact zones were identified in the study area: <5 km and >5 km from the pollution source. These zones correspond to a strong and a moderate technogenic impact;
- The physiological mechanisms of cotton make it possible to control the negative impacts of technogenic emissions by regulating the metabolic processes in its tissues. It is possible to grow cotton at a distance > 5 km from a nitrogen–phosphate fertilizer factory;
- Considering the results obtained, it is recommended that the analysis of the chemical compositions of plants located in an impact zone be carried out only after removing dust particles from their surface;
- The recommendation is to continuously monitor the concentrations of microelements in agricultural crops in the area most vulnerable to technogenic pollution;
- The results of this study can be applied to other cotton-growing areas with similar climatic and soil conditions.

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