

Environmental state of soils and plants in the area of exposure to emissions of fluorides of the cryolite plant and agrochemical methods of protection from effects of pollution

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Abstract— According to field surveys of arable soils in the zone of anthropogenic emissions of the cryolite plant, the levels of soil contamination with fluorine, depending on the distance to the source, were determined and the correlation of agrochemical soil indicators with the content of fluorine in them was identified. Soil 20 km away from cryolite plant, can be considered clean (fluoride content of 2.6 mg/kg). Evaluation of the productivity of plants at the sampling points demonstrated that the content of water-soluble fluorine in the soil above the MPC leads to a decrease in crop yields. In micro plot and small experiments, the effect of fluorine pollution on the productivity of plants was studied, the protective shields from fluoride contamination were tested. It was found that enhanced mineral nutrition contributes to the active plants resistance to oppression caused by fluoride pollution. Breakdowns in the anatomical structure and formation of generative cells under the influence of toxic effects of fluorine were revealed. The nature of chromosomal rearrangements after fluoride pollution resembles the effect of physical and chemical mutagens on plants.

Keywords— *fluoride pollution, soils, plant productivity, protective suspensions, anatomical structure, cytomorphological analysis, meiosis, chromosomal rearrangements.*

I. FLUORIDE POLLUTION - SOURCE AND EXTENT

Fluorine by weight occupies the thirteenth place among the elements that make up the earth's crust, and nineteenth among the elements that make up the soil. The presence of fluorine in plants was defined by Müller in 1845, but only in 1913 Gauthier and Klausman detected it in all of the 64 plant species they studied [1].

Even though, the physiological need for fluorine in plant metabolism has not yet been proven, the participation of fluorine in the synthesis of humus has been noted (i.e. the need for it for the life of specific soil microorganisms) [2].

The technogenic accumulation of fluorine in the environment is the greatest risk for humans, animals and plants [3,4,5]. Among the existing atmospheric pollutants, fluorine is the third in volume and hazard after sulphur and nitrogen oxides.

The most intensive fluorine pollution occurs because of the anthropogenic emissions in the production of aluminum,

phosphorus fertilizers, glass, ceramics, as well as the use of fertilizers containing residual fluorine. The fluoride contamination rate and the negative effects on the environment around the aluminum smelters are inversely proportional to the distance from the source of pollution. The most severe harm was observed within a radius of 2 km from the emission source [6,7,8,9,10]. Also, the systematic long-term application of superphosphate, amorphous, or a one-time application of fertilizers containing 500 kg or more P₂O₅ per hectare, reduces yield and leads to the accumulation of fluorine in the biomass [11].

II. FLUORINE IN SOIL

The major part of fluorides which get into the soil, goes into an insoluble or hardly soluble condition and only a small amount remains in a mobile bonding. The amount and intensity of this transformation depends on many factors, but primarily on the chemical composition of the soil.

The accumulation of fluorine in the soil leads to the decrease of the redox potential and of the amount of cation exchange, an increase in the solid residual from the aqueous extract. The mobility of iron, aluminum, manganese increases, the soil dispersion increases, the availability of nutrients to plants decreases, the total number of microorganisms and enzymatic activity decreases [12].

III. FLUORINE IN PLANTS

Under normal conditions, the fluorine content in various plant organs does not exceed 5 mg / kg: in cultivated plants mostly in the green mass, in wood plants - in the branches and bark [14,15]. Resistant to fluorine plants include buckwheat, potatoes, millet, sugar beet, canola, roses, asters, cherries. Corn, barley, wheat, peas, clover, spinach react negatively to the increase of the soil fluorine content [16].

Atmospheric air pollution with fluorine leads to the assimilation apparatus disfunction in all plants, reduces the resistance of plants to adverse conditions [17]. Fluorine is an inhibitor of lactic acid formation, in its presence the content of citric, succinic, fumaric, malic, oxalic acids increases, RNA activity decreases, cell division and elongation slow down, phytin decay decreases [18]. When plants are poisoned with fluoride, necrosis of the leaves is noted, they

become dense and covered with a whitish-gray bloom, deformation and premature abscission of the fruit is being marked, yield is significantly reduced. Reduced yields can be additionally caused by a violation of root nutrition.

IV. FLUORIDE POLLUTION IN THE IMPACT ZONE OF THE CRYOLITE PLANT

The nature of changes in the content of fluorine in the soil was studied at arable soils located in the vicinity of the Polevskoy cryolite plant (located near Yekaterinburg).

The samples for agrochemical soil analysis were taken from the depth of the arable layer. The content of water-soluble fluorine exceeds maximum permissible concentration (MPC) [13] in almost all samples (Table 1). It indicates a significant contamination of arable land with fluorine.

TABLE I. THE WATER-SOLUBLE FLUORINE CONTENT IN THE ARABLE SOIL LAYER, DEPENDING ON THE DISTANCE FROM THE SOURCE OF POLLUTION (AVERAGE FOR 2 YEARS, MG / KG)

Observation point number	Direction from emission source	Distance from emission source, km	Fluorine content
1	North-East	3	4.1
2	North-East	5	10.0
3	North-East	10	5.7
4	East	7	7.0
5	East	9	4.1
6	South	5	4.3
7	South	7	4.5
8	South	10	4.0
9	South	10	2.6
MPC			3.0

In our study, the maximum content of water-soluble fluorine in the arable soil layer was determined in the North-East direction at a distance of 5 km and in the East direction (prevailing in wind rose) at a distance of 7 km from the plant. In the southern direction, the soil is less polluted, fluorine content at a distance of up to 10 km is 4.0-4.5 mg/kg.

The record of plant productivity at the sampling points showed that the content of water-soluble fluorine in the soil above the MPC leads to a decrease in crop yields. Yield loss, expressed in fodder units (Y), depending on the content of water-soluble fluorine (x) in the soil is described by the function:

$$Y = 0.22x^2 - 0.34x + 1.9 \quad (1)$$

The dependence of the water-soluble fluorine content from the soil agrochemical characteristics

The agrochemical analysis of soil samples taken in the area of fluoride pollution showed that there is a correlation between the content of water-soluble fluorine and chemical indicators of the soil (table 2). In the table: H – hydrolytic acidity; S – the sum of exchange cations, r – the coefficient of correlation.

TABLE II. AGROCHEMICAL PARAMETERS OF SOIL AND ACTIVE FLUORINE

Fluoride, mg/kg	Humus, %	pH _{KCl}	mmol/100 g of soil		mg/kg of soil		
			H	S	N	P ₂ O ₅	K ₂ O
2.0	4.8	4.9	9.0	23.1	200	67	195
2.5	3.4	4.6	7.0	17.7	150	94	178
4.5	3.4	5.1	9.2	12.2	180	59	87
5.0	5.1	4.8	8.9	26.0	210	67	178
5.0	3.4	5.3	4.6	34.1	205	54	160
5.5	2.7	6.0	1.8	29.9	124	98	224
6.0	4.7	5.3	6.3	24.4	184	45	178
8.0	6.4	6.4	2.0	41.7	210	170	249
8.5	4.3	4.7	9.1	17.2	180	37	128
r	0.33	0.39	-0.39	0.37	0.16	0.09	-0.08

A positive correlation is noted between the content of water-soluble fluorine, pH_{KCl}, the sum of the cation exchange, as well as humus (r = 0.33-0.39).

There is no close relationship between the content of fluorine and hydrolytic acidity (inverse dependence), as well as the content of nutrients (nitrogen, phosphorus, potassium).

Thus, our data confirm the conclusions about the positive correlation between the content of fluorine and organic matter, the content of fluorine and acidity, expressed by a number of authors [12,15].

V. THE FLUORINE CONTENT IN THE PLANT BIOMASS IN THE CONTAMINATED ZONE

The analysis of plants for the content of fluorine in the technogenic contamination zone showed that the accumulation of fluorine can be higher than 5mg/kg of dry matter even within a radius of 20 km from the source of pollution (table 3).

The table data demonstrate, that the content of water-soluble fluorine in plants depends not only on the distance from the source of pollution, but also on agricultural crop species.

The maximum amount of the detected fluorine was in the sunflower leaves, the necrotic damage was visible [19].

TABLE III. FLUORINE CONTENT IN PLANT BIOMASS IN THE TECHNOGENIC POLLUTION ZONE

Distance from the source of pollution, km (the wind rose)	Crops	Fluoride content in mg/kg of dry matter
3	Annual grass	17
5	Cabbage	5
5	Potato	6
7	Carrot	8
7	Sunflower	11
9	Turnips	6
10	Sunflower	14
20	Winter rye	5
25	Barley	4

VI. NEUTRALIZATION OF FLUORIDE HARMFUL EFFECTS ON YIELD

At the Ural State Agrarian University, the series of experiments were conducted with barley, which was subjected to aerosol contamination with hydrogen fluoride. The experiments have shown that mineral fertilizers

significantly increase crop irrespective of the degree of contamination by fluorides (Table 4).

TABLE IV. THE EFFECT OF FERTILIZERS ON THE YIELD OF BARLEY DURING THE TREATMENT OF PLANTS WITH HYDROGEN FLUORIDE (G OF DRY MATTER /POT)

Background nutrition	HF concentration, %	Total biomass	Grain	The grain: straw ratio
0	0.00	3.5	1.8	1:0.96
	0.05	2.8	1.4	1:0.98
	0.10	2.3	1.0	1:1.26
	0.50	2.2	0.8	1:1.28
NPK	0.00	6.3	3.4	1:0.85
	0.05	6.2	3.4	1:0.79
	0.10	6.3	3.2	1:0.95
	0.50	4.1	2.9	1:0.92

The experiment included ammonium nitrate, double superphosphate, potassium chloride at the rate of 100 mg N, P₂O₅, K₂O per 1 kg of soil. The enhanced mineral nutrition contributes to the active resistance of plants to oppression caused by fluoride pollution. In the test without fertilizers, the yield decreased more intensively the higher the concentration of fluorine in the aerosol was. The amount of straw in the total biomass at these variants increased.

VII. THE EFFECT OF THE PROTECTIVE SUSPENSION ON THE BARLEY CROP'S

The choice of the protective suspension, which is applied by spraying before the most important phases of development, was based on the assumption that the chemically active forms of fluoride compounds, getting to the plants, will be fully or partially converted into insoluble compounds when interacting with the protective background substance. To do this, we chose slaked lime Ca(OH)₂ and silica gel (SiO₂*2H₂O) 4% concentration with the addition of soap (40 g per 10 l) to ensure adhesion. The protective suspension was applied to the plants at the beginning of the tillering phase (Z21-Z23).

The results of the micro-field experiment with barley indicate that the protective suspension applied to the plants does not reduce the yield of barley when spraying with silica gel suspension. Spraying with slaked lime solution reduces the yield of grain (table 5).

TABLE V. THE EFFECT OF PROTECTIVE SUSPENSION ON THE YIELD OF BARLEY (G OF DRY MATTER /POT)

Protective suspension	HF concentration, %	Total biomass	Grain	The grain: straw ratio
0	0.00	5.9	3.19	1:0.85
	0.05	5.9	3.24	1:0.79
	0.10	5.8	3.31	1:0.75
	0.50	4.1	2.37	1:0.73
Ca(OH) ₂	0.00	5.3	2.70	1:0.96
	0.05	4.1	2.16	1:0.90
	0.10	4.4	2.38	1:0.85
	0.50	5.0	2.76	1:0.81
SiO ₂ *2H ₂ O	0.00	6.3	3.15	1:1.00
	0.05	5.5	2.52	1:0.95
	0.10	4.0	2.59	1:0.89
	0.50	5.0	2.75	1:0.82

When plants were exposed to low concentrations of fluoride aerosols (0.05-0.1% HF), both calcium and silica suspensions did not have a protective effect, on the contrary, chemicals reduced the accumulation of total biomass and

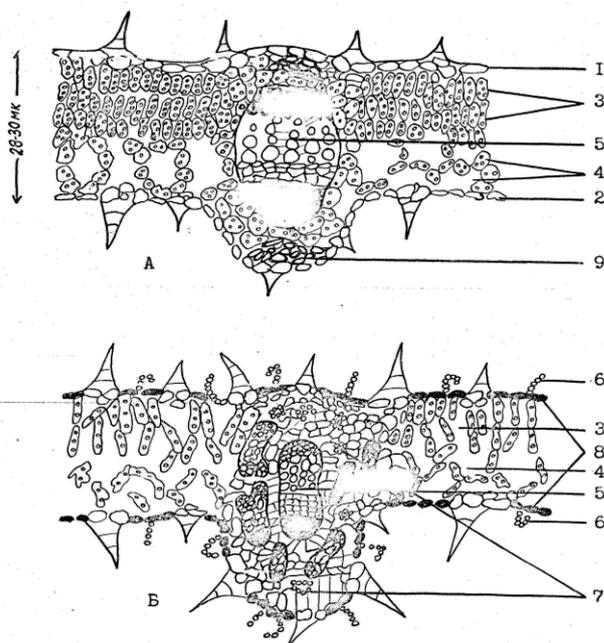
grain yield from the vegetative pot. The protective properties of suspensions were registered only after spraying the plants by the aerosols with a high fluorine content (0.5% HF).

VIII. EFFECT OF FLUORIDE CONTAMINATION ON THE SUNFLOWER LEAF MORPHOLOGICAL STRUCTURE

The study of the sunflower leaf anatomical structure under the microscope showed that the areolar parenchyma cells are the first to undergo necrosis. The palisade parenchyma cells are destroyed, they lose their vertical position, and the intercellular cells appear in the columnar tissue (Fig. 1). The chloroplasts lose chlorophyll and become red-yellow. There are regular rows of small cells such as "assimilants" among the fuzzes on the epidermis.

In the central vein, 3-5 collateral bundles (in the lower leaves 2-3 and up to 10-15 in the upper ones) are formed instead of one.

The cytoplasm stains in dark tones in the cells of the epidermis. Perhaps, fluorine is captured by the cytoplasm of the epidermis, but diffusing into other cells (located below) does not occur.



1 - the upper epidermis with fuzzes; 2 - the lower epidermis; 3 - columnar tissue; 4 - spongy cell tissue; 5 - vascular tissue; 6 - «assimilators»; 7 - dividing cells; 8 - painted cells of epidermis; 9 - collenchyma

Fig. 1. The anatomical structure of the sunflower leaf from the area without (A) and with (B) fluoride pollution

IX. CYTOMORPHOLOGICAL ANALYSIS OF BARLEY PREPARATIONS BASED ON THE MODEL EXPERIMENTS

Cytomorphological analysis of barley preparations showed that the fertilization stimulates the development of spikelet tubercles under the fluoride pollution. Without fertilizer the length of the tubercle reaches 0.8-1.0 cm (archesporial cells did not start to divide), whereas the plants from the NPK background are in the fourth stage of organogenesis. The value of their cones reaches 1.2-1.5 cm, the stamens are originated in a flower, meiosis starts in the cells of archesporium.

The barley meiosis consists of the reductional and equational stages, each of which includes a number of phases that modify with violations caused by fluoride pollution.

The analyzed plants were in metaphase 1 and anaphase 1 of the reductional stage and metaphase 2 and anaphase 2 of the equational stage.

The anomalies of different kinds were noted in all phases of the division (Fig.2). Thus, there are the pictures of cytomixis (A); the release of chromosomes and their fragments beyond the spindle into the cytoplasm in metaphases 1 and 2 (B); premature divergence of bivalents and chromosomes to the poles (C). Some chromosomes linger at the cell equator, lag in the divergence to the poles, the chromatin bridges are formed between the daughter cells in anaphases 1 and 2. Chromosomes can be detected outside the spindle, forming a micronucleus (D).

All these serious deviations lead to the appearance of the broken tetrads and spores with abnormalities in the number of chromosomes (aneuploidy).

Sometimes the pentads and hexads are formed instead of tetrads at the end of meiosis. This occurs when the cytoplasm, which is isolated around the micronucleus, forms the additional cells (E).

Different concentrations of fluorine and protective suspensions affect the type and number of aberrations (Tables 6 and 7).

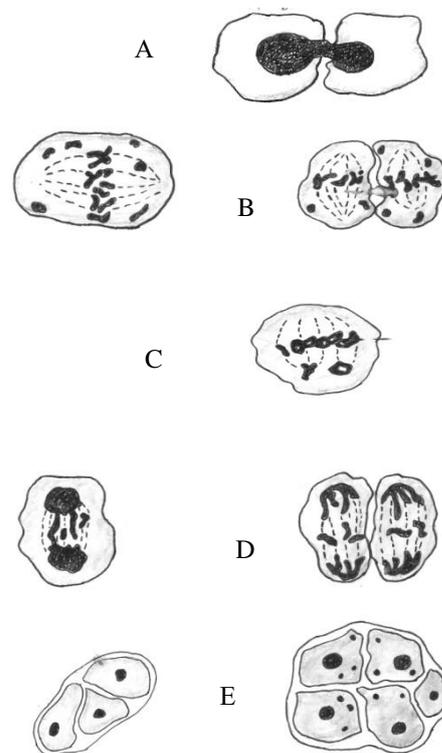


Fig. 2. Chromosomal abnormalities in the phases of meiosis in the formation of barley pollen

TABLE VI. THE CONDITION OF BARLEY DIVIDING CELLS IN THE STAGE OF ANAPHASE I

Fertilizer	Protective suspension	HF concentration, %	The cells without breaks, %	The cells with chromosome abnormalities					
				Bridges	Fragments	Lagging	Accelerating	Multipolarity	Total
0	0	0	98.9	0.1	0.3	0.6	-	0.1	1.1
		0.05	81.8	2.1	4.3	5.4	4.8	1.6	18.2
		0.1	84.5	1.8	3.2	4.9	5.1	0.5	15.5
		0.5	92.5	0.9	1.1	2.8	2.6	0.1	7.5
N ₆₀ P ₆₀ K ₆₀	0	0	98.9	0.2	0.2	0.6	-	0.1	1.1
		0.05	81.9	2.8	4.2	4.6	3.9	2.6	18.1
		0.1	83.2	3.1	3.8	3.2	4.9	1.8	16.8
		0.5	94.8	1.4	0.6	1.8	1.4	-	5.2
N ₆₀ P ₆₀ K ₆₀	Ca(OH) ₂	0	99.0	0.1	0.3	0.5	-	0.1	1.0
		0.05	85.9	2.4	3.4	3.6	2.9	1.8	14.1
		0.1	86.2	2.8	2.9	2.8	3.1	2.2	13.8
		0.5	95.9	0.6	1.1	0.5	0.8	1.1	4.1
N ₆₀ P ₆₀ K ₆₀	SiO ₂ *2H ₂ O	0	98.9	0.1	0.2	0.5	-	0.1	1.1
		0.05	89.2	1.8	2.2	3.4	1.8	1.6	10.8
		0.1	87.6	1.4	2.8	3.2	3.2	1.9	12.4
		0.5	95.0	0.9	1.6	2.7	-	0.8	5.0

It should be noted that the lower the hydrogen fluoride concentration, the higher the percentage of cells had the chromosomes behavioral. Protective suspensions of Ca(OH)₂ and SiO₂ have slightly reduced aberrations, but have increased the percentage of breaks in tetrads substantially at the end of anaphase 2. Thus, there was an appearance of monads, triads, abnormal tetrads, pentads, hexads, poliads.

The cytotomy brakes were noticed after the contamination with 0.5% HF: tetrads of microspores are formed in the simultaneous type (the successive type is normal).

TABLE VII. THE CONDITION OF BARLEY DIVIDING CELLS IN THE STAGE OF ANAPHASE 2

Fertilizer	Protective suspension	HF concentration, %	The cells without breaks, %	The cells with chromosome's abnormalities				
				Bridges	Fragments	Asynchrony in divergence	Breaks in tetrads	Total
0	0	0	91.4	1.1	0.2	1.1	6.2	8.6
		0.05	66.3	12.4	4.1	8.9	8.3	33.7
		0.1	67.8	12.1	4.9	7.6	7.6	32.2
		0.5	77.0	9.4	2.3	6.2	5.1	23.0
NPK ₍₆₀₎	0	0	91.4	1.1	0.2	1.1	6.2	8.6
		0.05	61.0	10.6	4.9	14.1	9.4	39.0
		0.1	59.6	11.2	4.4	13.4	11.4	40.4
		0.5	68.5	9.6	2.2	12.8	6.9	31.5
NPK ₍₆₀₎	Ca(OH) ₂	0	91.4	1.1	0.2	1.1	6.2	8.6
		0.05	61.0	10.1	5.7	12.8	10.4	39.0
		0.1	62.4	11.2	4.9	11.4	10.1	33.6
		0.5	70.9	8.4	4.1	10.8	5.8	29.1
NPK ₍₆₀₎	SiO ₂ *2H ₂ O	0	91.4	1.1	0.2	1.1	6.2	8.6
		0.05	65.8	11.3	3.2	10.6	9.1	34.2
		0.1	67.2	10.8	2.8	11.1	8.0	32.7
		0.5	74.2	9.6	2.0	9.9	4.2	25.7

X. THE POLLEN QUALITY IN CASE OF FLUORIDE POLLUTION

The pollen size and fertility can serve as a criterion of plants' adaptation to environmental conditions and may change under the influence of extreme factors.

When determining the degree of pollen fertility it is important to determine the size of pollen grains (large, small or normal), as their viability varies.

The spollen grains are regarded as sterile if there are cells which are empty or have small protoplast amount, cells with plasma-lysed content, without the starch, cells with pycnosis and structureless cytoplasm. The effect of fluoride contamination on the quality of barley pollen is shown in table 8.

TABLE VIII. EFFECT OF FLUORIDE POLLUTION ON POLLEN QUALITY

Fertilizer	Protective suspension	HF concentration, %	Pollen size, mkm	Fertility, %
0	0	0	2.20	95
		0.05	1.80	35
		0.1	1.60	30
		0.5	1.15	21
N ₆₀ P ₆₀ K ₆₀	0	0	2.50	95
		0.05	1.66	45
		0.1	1.65	40
		0.5	1.20	25
N ₆₀ P ₆₀ K ₆₀	Ca(OH) ₂	0	2.29	92
		0.05	1.87	87
		0.1	1.89	78
		0.5	1.69	56
N ₆₀ P ₆₀ K ₆₀	SiO ₂ *2H ₂ O	0	2.10	90
		0.05	1.76	51
		0.1	1.65	60
		0.5	1.58	40

With the increase of fluoride contamination the pollen sizes decrease, the growth processes are inhibited, the vegetation cone' differentiation is delayed, the percentage of sterile pollen increases.

The minimum amount and the lowest fertility of pollen was noted in the plants subjected to aerosol treatment with a concentration of 0.5% HF.

XI. CONCLUSIONS

- At a distance of up to 10 km along the wind rose from the cryolite plant, soil contamination with fluorides exceeds the maximum permissible concentration by 1.5-2 times.
- The content of fluoride in plants in the 20-kilometer zone of fluoride pollution exceeds 5 mg/kg.
- A positive correlation between the content of water-soluble fluorine, pHKCl, the sum of the cation exchange and humus was noted.
- Fluoride contamination leads to serious violations in the morphological structure of the sunflower leaf.
- Mineral fertilizers significantly increase yields regardless of the degree of contamination with fluorides.
- Protective properties of calcium and silicon suspensions appeared only when sprayed on plant with a high concentration of fluorine (0.5% HF).

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